COMMUNAUTE FRANÇAISE DE BELGIQUE
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HYDRAULIC AND REMOVAL EFFICIENCIES OF HORIZONTAL FLOW TREATMENT WETLANDS

Nathalie FONDER

Dissertation originale présentée en vue de l'obtention du grade de docteur en sciences agronomiques et ingénierie biologique

Promoteur: Prof. D. Xanthoulis

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Fonder Nathalie. (2010). Hydraulic and removal efficiencies of horizontal flow treatment wetlands
(PhD thesis in English). Gembloux, Belgium
ULG, Gembloux Agro-Bio Tech
191p., 18 tabl., 66 fig.

Summary: The hydraulic and removal efficiencies of a Horizontal Flow Treatment Wetlands (HF TW) were investigated through an internal three dimensional grid of sampling ports. Tracer tests and regular monitoring of water quality parameters were performed. Results demonstrated that the HF TW has generally good hydraulic and volumetric efficiencies, with relatively low dead zones. The application of models developed by chemical engineering provided the number of tanks in series and the calculated detention times which were input as parameters in the multi flow with dispersion hydraulic model. This second model identified that water fluxes were not homogeneous with depth inside the TW and 60% of the flow was along the bottom layer of the bed. It also indicated the water flow velocities, which were faster on the bottom of the bed, and the axial dispersion, which was higher where flow velocity was lower. The reviewed inflow rate distribution allowed review for all layers of the nominal detention time and of the hydraulic indexes, which are developed by the chemical engineering theory, and based on the incorrect assumption of homogeneous systems. The *P-k-C** degradation model was applied in order to define degradation *k*-rate values of BOD and COD and the frequency distribution profiles were developed. The degradation rate coefficients for BOD ranked from less than 10 m/yr to more than 300 m/yr. Significant higher degradation rates were observed for all the bottom layers and for the closest sampling line from the inlet. The results of COD were similar to those observed for BOD. Finally, the specific pollutants of nitrogen and phosphorus were analysed for total nitrogen (TN) and total phosphorus (TP). The global trend of the TN degradation coefficient values was a slow and regular decrease over length, having systematically higher degradation coefficients for the bottom layers. The saturation of the media sites for sorption capacity of TP was demonstrated being in progress.

Fonder Nathalie. (2010). Efficacités hydraulique et épuratoire du filtre planté à écoulement horizontal (thèse de doctorat en anglais). ULG, Gembloux Agro-Bio Tech, 191p., 18 tabl., 66 fig.

Résumé : Les efficacités hydraulique et épuratoire du filtre planté à écoulement horizontal ont été mesurées grâce à un dispositif de prélèvement tridimensionnel dans un filtre en fonctionnement. Des essais de traçage ont été réalisés ainsi que des analyses régulières de la qualité de l'eau en tous les points d'échantillonnage. Les résultats ont démontré que le filtre présente des efficacités hydraulique et volumétrique généralement bonnes, avec relativement peu de zones mortes. L'application des modèles mathématiques issus du génie chimique ont permis la détermination du nombre *N* de réservoirs en série et de calculer les temps de séjour ; ceux-ci ont ensuite été introduit dans le modèle hydraulique de dispersion multi-couches. Ce second modèle a identifié que les flux en eau n'étaient pas homogènes avec la profondeur à l'intérieur du système et que 60 % de l'écoulement se concentraient dans la couche de fond du filtre. Il a également fournit les vitesses d'écoulement par couche, qui se présentent comme plus rapides dans le fond du filtre, et la dispersion axiale, dont la valeur augmente en corrélation avec une diminution de la vitesse. La distribution du débit par horizon a pu être revue comme non homogène avec la profondeur et a permis de recalculer les index hydrauliques et les temps de séjour par horizon en évitant l'hypothèse erronée des modèles du génie chimique de l'homogénéité du système. Le modèle de dégradation des polluants *P-k-C** a été appliqué pour définir les valeurs des coefficients de dégradation de la DBO, la DCO et l'azote total. Les profils de distribution de fréquence ont été dressés. Les différences de coefficients de dégradation de la DBO sont très hautement significatives avec la profondeur et la longueur, en ayant des valeurs plus importantes pour la couche de fond et pour la première ligne de prélèvement la plus proche de la zone de distribution de l'effluent dans le filtre. Les résultats obtenus pour la DCO ont été semblables. Les coefficients de dégradation de l'azote total ont montré une décroissance lente et régulière avec la longueur, et des valeurs systématiquement plus importantes avec la profondeur. Finalement, il a été démontré que les sites d'adsorption du phosphore total sont en cours de saturation.

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As I finalise this work, it is a pleasure to me to be able to thank some of people who have taken an active part in this final achievement.

I want to thank first my advisor and employer, Professor Dimitri Xanthoulis. He has always encouraged me to lead this task and has given me the opportunity to combine it with my employment contract so I could earn a living while pursuing a PhD. He has always provided me all the tools, whichever they were, to lead this long term research till its end and given me the freedom to develop a subject which was in the frame of our activities but was also a personal interest of mine. Finally, his infallible trust and belief in me have been a helpful support in my personal growth and development since I work for him and with him in a collaborative partnership.

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LIST OF ACRONYMS

BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
CW	Constructed Wetland(s)
CST(R)	Complete Stirred Tank (Reactor)
DF TW	Down Flow Treatment Wetland
DF-EF TW	Ephemerally Flooded Down Flow Treatment Wetland
DM	Dispersion Model
DPF	Dispersed Plug Flow
DTD	Detention Time Distribution
ET(M)	Evapotranspiration (Maximum)
FaD TW	Fill and Drain Treatment Wetland
FEM TW	Floating Emergent Macrophyte Treatment Wetland
FFM TW	Free-Floating Macrophyte Treatment Wetland
GAMMADIS	Gamma Distribution
HLR	Hydraulic Loading Rate
HF TW	Horizontal Flow Treatment Wetland(s)
HRT	Hydraulic Retention Time
IN	Inlet
L1	Line 1
L2	Line 2
L3	Line 3
LRI	Inlet Loading Rate
LSQS	Least SQuarred Method
MFDM	Multi Flow with Dispersion Model
MM	Method of Moments
NTIS	Number of Tank in Series
NVSS	Non Volatile Suspended Solids
OLR	Organic Loading Rate
OUT	Outlet
Pe	Peclet number
PE	Per Equivalent inhabitant
PF(R)	Plug Flow (Reactor)
PLR	Phosphorus Loading Rate
PTIS	Apparent Number of Tank In Series
RDT	Residence Distribution Time
SF TW	Surface Flow Treatment Wetland
SSQE	Sum of SQuarred Errors
TBC	Tracer Breakthrough Curve
TIS	Tank in Series
TN	Total Nitrogen
TP	Total Phosphorus
TSS	Total Suspended Solids
TW(s)	Treatment Wetland(s)
VSS	Volatile Suspended Solids

OUTLINE

OUTLINE

THE MAIN QUESTION

"The water environment is/was in a state of decline and the Water Framework Directive edited by Europe intends to recover the good water quality for our surface bodies and to protect groundwater. For many years our rivers, lakes, coasts and wetlands have been used as natural sinks - a repository for sewage, slurry, and industrial effluents – in fact almost anything which was either too difficult or too expensive to get rid of in any other way. Water quality is an issue for even the most progressive countries and how to deal with the polluting effect of growth and economic development is an on-going problem. Constructed wetlands offer solutions to many water quality issues. They are not simply another alternative to an existing technology. Industrial wastewaters, surface water run-off pollution, Sustainable Urban Drainage Systems (SUDs), Combined Sewer Overflow (CSO), domestic and agricultural effluents, amongst others, can all be treated using constructed wetlands" (Constructed Wetland Association, 2010).

Treatment wetlands are built, studied and monitored since the 1970's, but they always have been investigated based on IN- and OUT- data sets. The general rule of the thumb exists to design treatment wetlands for secondary or tertiary domestic wastewater treatment adapted to northern Europe climate. But once, designers want to develop the technology for the treatment of other type or composition of wastewaters, to remove specific pollutants, to adapt design to other climates, etc. the necessity to fully understand processes occurring within the wetland beds emerges without clear answers.

This was the start of the main question of this work:

"What is occurring, how and where within a horizontal flow treatment wetland system?"

THE INVESTIGATION PROCESS

The treatment wetland field is an amazingly interesting subject as it relies on water chemistry, engineering design and hydraulics, botany and zoology (with microbial fauna aspects) which are amongst major background fields of any agronomist. Additionally, daily challenges of engineers are to solve problems, find solutions and practical issues. In the treatment wetland field, the agronomy engineering background provides the ability to connect and link the various topics involved, to understand and to combine all relevant studies and information in order to provide practical solutions and results.
Usual studies are investigating part of the above question, through compartmented studies led in depth. The chosen route in this work is to focus on linking specific 'vertical' studies into a 'horizontal' understanding process, based on experiments, existing theories, models, and any relevant references.

With the start of the works, a preliminary work was to propose a harmonised nomenclature and classification of treatment wetlands, as the current terminology uses are rather non systematic and often confusing. Chapter I opens the thesis providing the proposed terminology and overlook of existing TW technologies.

In following chapters, and in order to answer the main question, the investigation method developed was a three dimensional grid or matrix of sampling points within the "black box" of an onsite and operational Horizontal Flow Treatment Wetland (HF TW), which was monitored over almost three years in a row, in parallel to experiments under greenhouse on small scale pilot systems. According to references, only a few isolated studies with punctual sampling within the TW beds were performed what has justified the systematic and scientific original aspects of the study.

Chapter II answers the sub-question which was: "how are pollutants carried within the bed?" This sub-question translated in hydrological terms was: "what is the water flow path within the bed?" The hydraulic behaviour and water flow paths existing and carrying pollutants through the bed was investigated through onsite three dimensional tracer test experiments, replicated twice. Their results and conclusions are provided in Chapter II.

The discharge of treated wastewater into the environment is regulated in the Walloon region according to the European Directive 91/271/CEE on urban wastewater treatment by water quality standard based on Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD) and Total Suspended Solids (TSS). Chapter III investigates their removal processes and location within the onsite HF TW, through the three dimensional monitoring matrix and sampling campaign over the years of experimentation.

Finally, the specific pollutants of Nitrogen and Phosphorus are regulated according to the same European Directive 91/271/CEE on urban wastewater treatment, translated in the Walloon regulation, for treatment plants connecting more than 10.000 inhabitants. Small scale systems, as the one monitored in the frame of this work, do not have to respect any regulation on nitrogen and phosphorus for wastewater discharge in the environment. But the two specific pollutants are investigated in Chapter IV as they are a major concern for agronomist either for their nutrient value for crop production or harmful effects on the environment. Additional microbial analysis provides some indication of the bacteria presence and location in the HF TW.

All those aspects ('vertical studies') could have been investigated more in depth (e.g. for the composition of the biofilm, the nitrogen forms and budget, etc.) and many other additional aspects could have been developed (e.g. vegetative aspects, hydraulic conductivity and clogging problems, etc.), but all works have to be soon or late defined into their frame, stopped and concluded. All those potential works are an invitation to any future PhD studies to be continued.

INNOVATIVE ASPECTS DEVELOPED BY THE PHD

The following points developed by this PhD are innovative, based on existing and published literature:

- To propose an harmonised nomenclature and terminology for TW,
- To monitor points inside the TW, over length and also depth (tri-dimensional monitoring matrix) for onsite and operational system 3 years in a row,
- To replicate tracer tests experiments, for both onsite and pilot HF TW systems,
- To apply mathematical models issued from hydraulics instead of those issued from chemical engineering, usually applied in the TW field,
- To link microbial aspects and pollutants removal to the hydraulic pattern of the HF TW.

CHAPTER I

Treatment Wetland Systems

I.CHAPTER I : TREATMENT WETLAND SYSTEMS

I.1 INTRODUCTION

With the start of this investigation work on treatment wetlands, I was struck by the fact that the literature is riddled with a wide ranging and disconnected terminology. Although some systematic approaches for classifying and naming treatment wetland types currently exist, numerous names are used, almost interchangeably to describe any given wetland variant, even if the physical design and operational characteristics are essentially the same. In some cases, the same name has been used for systems with very different design configurations. The applied terminology often varies by region, culture, discipline-base or the author's desire to give the impression that their design is new or innovative. This diversity in nomenclature within the treatment wetland field leads to confusion, inefficiency and the general impression that the wetland industry is lacking in organisation and professionalism, particularly to those new-comers or observers from other disciplines. Furthermore, there is a need for greater consistency in the fundamental design and operational information that is provided when reporting or publishing data from treatment wetlands.

Thanks to the partnership of a co-author, I wrote a paper proposing a review and clarification of the current terminology used to define and describe constructed treatment wetlands; we proposed a standardised classification system that provides a framework for nomenclature in this field; and we summarised the important necessary details that should be provided when reporting about a treatment wetland system or study.

The intention was not to provide a definitive nomenclature or to replace pre-existing terminology with a new system, but rather to make an initial step towards providing a structured foundation for classifying and naming different treatment wetland design alternatives. It is hoped that this can provide a platform for standardising the terminology that is used in our field. It is acknowledged that there will be existing and emerging applications that do not neatly fit the classification system given. In many cases, authors will prefer a different name for their system; other, more common, names may be more appealing, user-friendly or politically attractive depending on the context or target audience.

This introduction chapter presents the systematic nomenclature issued from this paper edited by Springer (Fonder and Headley, 2010) to present wetlands in a general frame, and the system investigated for this thesis. It defines wetlands; the specificities of treatment wetlands (TW) are listed and the nomenclature to identify the variant types is presented. Standard types of treatment wetlands and their variants are briefly described. The TW site in operation investigated by the study is described as well as the small scale pilots used for related studies.

I.2 GENERAL REMARKS ON WETLAND SYSTEMS

I.2.1 General definition

Wetlands can be defined as areas of land where the water table is at or near the surface for at least part of the year and are characterised by the presence of vegetation types and soil characteristics that have developed in response to the wet and saturated conditions (Kadlec and Wallace, 2009; Mitsch and Gosselink, 2007).

Wetlands can be first split into the two major types of Natural and Constructed wetlands. Natural Wetlands are only defined here as those wetland areas that exist in the landscape due to natural processes rather than having been created either directly or indirectly as a result of anthropogenic influences. The classification or terminology for natural wetlands already exists (see related references, e.g. Wetzel, 2001; Mitsch and Gosselink, 2007), and is not

presented here, and this chapter focuses on terminology related to constructed treatment wetlands.
Constructed wetlands are artificially created ecosystems that would not otherwise exist without significant human intervention, such as earthworks or hydrologic manipulation. They are generally designed to mimic many of the conditions and/or processes that occur in natural wetlands (Vymazal and Kropfelova, 2008).

I.2.2 Purpose of Constructed Wetlands

Constructed wetlands can be split into three categories according to their purpose.

- Restored Wetlands: areas which were formerly natural wetlands that were lost or heavily degraded in the past and which, through human intervention, now support a near-natural wetland ecosystem.
- Created Wetlands: non-wetland areas which have been converted to a wetland ecosystem by civil engineering works.
- Treatment Wetlands: artificially created wetland systems designed to provide a specific water treatment function.

I.2.3 Treatment Wetlands

Treatment wetlands are human-made systems designed to enhance and optimise certain physical and/or biogeochemical processes that occur in natural wetland ecosystems for the primary purpose of removing contaminants from polluted waters. As TWs can be constructed in a variety of hydrologic modes (Kadlec and Wallace, 2009), and numerous design variations have been developed for such a large array of pollutant removal mechanisms, there are a large number of design variants currently in use. Correspondingly, the commonly used terminology describing different TW systems is increasing and the need for a standardised set of terminology to clearly identify and define the most commonly used types in a technically accurate and consistent manner was proposed by Fonder and Headley (2010). This terminology is briefly presented here below.

The first important question is: what constitutes a Treatment Wetland? Treatment Wetland designs range from those having standing water to those with no obvious surface water, and systems with vegetation types ranging from small free-floating species like duckweed, to forested swamps consisting of large trees like Cypress or Melaleuca species. Despite this diversity, three characteristics can be identified which are common to all TWs:

1. The presence of macrophytic vegetation that typically occur in natural wetlands;
2. The existence of water-logged or saturated substrate conditions for at least part of the time; and
3. The inflow of contaminated waters with constituents that are to be removed.

The first criterion excludes ponds and lagoons that consist primarily of microscopic algae (microphytes) without higher plants (macrophytes). However, it is acknowledged that ponds and TWs are closely related technologies often used in combination. One exception to this requirement for the presence of wetland plants is within the context of research, where "unplanted" versions are sometimes included to distinguish the effect of the plants in the system. Outside of this context, treatment units that do not include wetland vegetation, such as gravel or sand filters, should not be classified as a TW.

There may be some contention as to whether or not free-draining systems such as down flow wetlands satisfy the second inherent requirement of a TW (the existence of water-logged

conditions for at least part of the time). However, even intermittently loaded down flow systems that are intended to operate in a primarily unsaturated mode, still experience ephemeral (periodic), localised water-logging of the substrate (e.g. during a loading event). Hence, such systems are considered to satisfy the requirement of partial water-logging.
The general structure of the hierarchy in presenting TW is illustrated by figure 1.

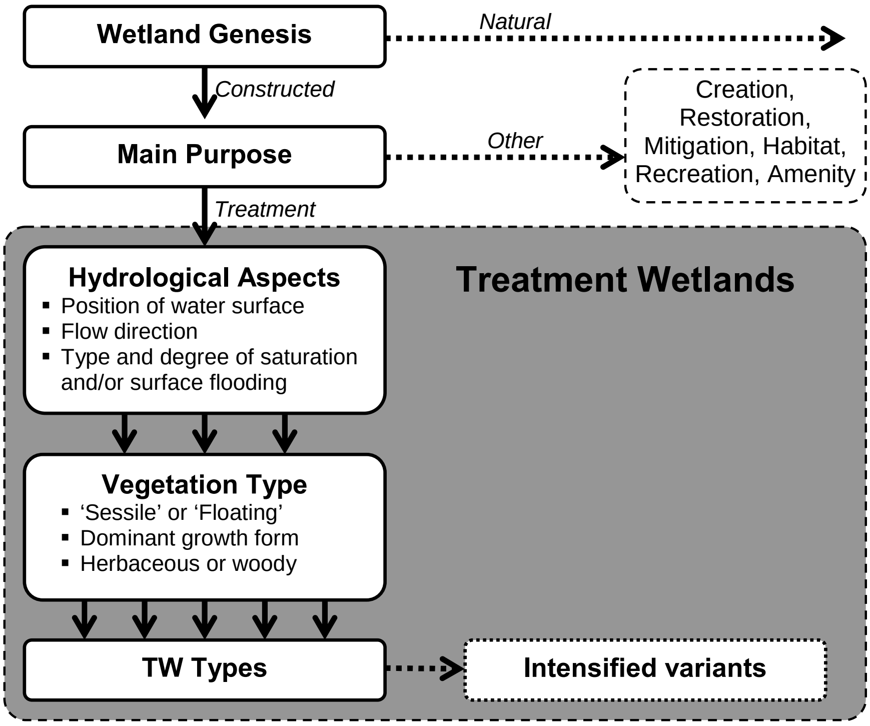


Figure 1 : The general structure of the classification hierarchy used to define treatment wetlands and their different types.

I.3 CLASSIFICATION OF TREATMENT WETLANDS

The various TW designs that exist can be categorised based on two main physical attributes:

a) Hydrology; and
b) Vegetation characteristics.

These form the basis for the proposed hierarchical classification system which identifies TWs based on the six specific traits presented in Table 1. To classify a given TW unit using the hierarchy, the various physical traits are considered in a sequential order starting with the water position, followed by the flow direction, saturation degree, type of surface flooding and finally the vegetation sessility and growth form. Each step of the hierarchy gives rise to different classes of TW design.

The different traits considered in the classification hierarchy distinguish between key physical and operational aspects of the design which lead to fundamental differences between the various TW units in terms of their structure and functioning. Consequently, these traits have a major influence over the treatment conditions that develop within the TW unit and in determining its suitability for a particular application. The technical nomenclature used to identify different TW units reflects this classification based on physical attributes.

Table 1 : Traits used to define the different classes of treatment wetland within the classification hierarchy.

Physical Attribute	Specific Trait	Description	Defined Classes for each trait	Sub-class
Hydrology	a. Water position	Position of water surface relative to soil or substrate	Surface Flow[a]	-
			Subsurface Flow[b]	-
	b. Flow direction	Predominant direction of flow through system	Horizontal	-
			Vertical[c]	Down
				Up
				Mixed
	c. Saturation of media[c]	Degree of saturation in media-based systems	Free-draining	-
			Intermittent	-
			Constant	-
	d. Surface flooding[c]	Type of surface flooding in media-based systems	None	-
			Ephemeral	-
			Permanent	-
Vegetation	a. Sessility[d]	Location of the roots: attached in the benthic sediments or floating	Sessile (benthic bound)	-
			Floating	-
	b. Growth Form	Dominant growth form of the vegetation in relation to the water	Emergent	Herbaceous
				Woody
			Submerged[d]	-
			Floating leaved[d]	-
			Free-floating[d]	-

[a] majority of flow through a column of water overlying a benthic substrate.
[b] majority of flow through a porous media.
[c] only relevant to Subsurface Flow systems (by virtue of design, all Surface Flow TWs have horizontal flow, are constantly saturated and with a permanently inundated substrate).
[d] only relevant to Surface Flow systems (subsurface flow excludes submerged or floating plants).

I.3.1 Water Position

Whether the TW has Surface Flow or Subsurface Flow has a major impact on the physicochemical conditions that develop within the treatment reactor and the types of vegetation that can be grown in the wetland. Surface flow TWs are defined as aquatic systems in which the majority of flow occurs through a water column overlying a benthic substrate. This is in contrast to subsurface flow TWs where the majority of flow is through a porous media. The term Surface Flow is selected rather than the other commonly used term, Free Water Surface, for this category of TWs because it is a clear antonym for subsurface; the alternate type of flow position. The term "free water surface" is a somewhat more ambiguous combination of words, the meaning of which is not inherently clear, especially for people not familiar with the technology.

I.3.2 Flow Direction

By virtue of the design, essentially all surface flow TWs have flow in a predominantly horizontal direction (inlet and outlet horizontally opposed). In contrast, subsurface flow TW units have been designed with a range of different flow directions, with horizontal and vertical down-flow being the most common to date. Systems with up flow also exist along with an increasing number where the flow alternates between up and down flow (mixed). With regards to nomenclature, a distinction based on flow direction is therefore not necessary for surface flow systems (they all have “horizontal flow”). Similarly, the inclusion of a term to identify the flow direction (e.g. horizontal flow or down flow) should inherently imply that a system has subsurface flow, therefore rendering the term “subsurface” redundant in the nomenclature.

I.3.3 Media Saturation

This is a trait of particular relevance to units with subsurface vertical flow and refers to the saturation or moisture status of the media, which varies depending on the specific design. Systems with an outlet structure designed to hold water in the bed and maintain the media in a continuously saturated state are classed as having “constant” media saturation. By virtue of the design, all up flow systems have a constantly saturated media. Although down flow TWs can also be designed to maintain a constant saturation level, they most commonly operate in a free-draining mode, with a permanently open outlet at the bottom of the bed. Thus, such systems only experience rare and localised saturation of the media, such as during a loading event or within zones with large accumulations of water-retentive organic matter. In between constantly saturated and free-draining systems lie those TWs defined as having intermittent saturation, which refers to the situation where the media saturation varies periodically (e.g. TWs with alternating filling and draining cycles) or seasonally (e.g. TWs designed to evapotranspire all of the wastewater on an annual basis).
Surface flow TWs are invariably always designed to operate with a constantly saturated soil substrate, except if they are allowed to dry out for maintenance purposes.

I.3.4 Surface Flooding

Surface flooding refers to the degree to which the surface of the bed is flooded or inundated. It is an operational hydrologic trait only relevant to the classification of systems with subsurface vertical flow. By definition, surface flow TWs have a permanently flooded surface. In contrast, subsurface flow TWs with horizontal flow are virtually always designed with the intention of avoiding surface flooding.
Surface flooding in vertical flow units can be ephemeral (flooded for only a short period, often as a result of flood-loading), permanent (surface of the media is continuously flooded during normal operation) or non-flooded (entirely subsurface). Subsurface flow systems classed as having a permanently flooded media surface (e.g. some up flow TWs) are not considered to fall into the category of surface flow TWs because the majority of the flow still occurs through a porous media, as defined previously by the attribute: water position.

I.3.5 Vegetation Sessility

This is the first distinction made between TW units in the classification scheme based on vegetative traits. Sessile is a term used in the field of limnology to refer to vegetation that is anchored to the benthic environment, as opposed to floating. This trait is only relevant to TWs with surface flow because subsurface flow through a porous media precludes floating plants.

I.3.6 Vegetative Growth Form

The growth form of the dominant plants in the wetland is a major trait for distinguishing between the different surface flow TW designs. The vegetation growth forms used in this classification are based on those defined by Brix and Schierup (1989) and are divided into emergent, submerged, floating leaved and free-floating plants. Submerged and floating leaved plants come under the grouping of sessile vegetation. Emergent macrophytes typically grow in a sessile form. However, they can also grow on a buoyant mat that floats on the water surface which gives rise to a different type of TW design.

I.3.7 Emergent Vegetation Variants

The majority of emergent wetland plants are herbaceous (e.g. reeds, rushes and sedges). That is, they lack lignified tissues in their above-ground parts and consequently die down to ground level at the end of growing season. Due to their common use in TWs, herbaceous macrophytes are therefore the default type of emergent vegetation within the classification system. However, some TWs are dominated by woody emergent macrophytes (trees and shrubs) which tolerate water-logged conditions (e.g. species of Cypress and Melaleuca). Because of the differences in growth cycles, shading, physical structure and element turn-over that can develop, TWs dominated by woody emergent vegetation form their own design variants within the classification system.

I.3.8 The Treatment Wetland Classification Tree

Applying the proposed classification system to the diverse range of TW designs in current use leads to the classification tree presented in Figure 2. The standard types and their design variants are keyed-out at the base of the tree along with the proposed nomenclature and abbreviations. For the sake of clarity, redundant traits that do not play a role in identifying the different TW designs are omitted from the tree diagram.
The classification tree highlights that the subsurface flow wetland group is a diverse class containing a broad range of designs differentiated essentially by hydrologic characteristics. In contrast, TWs with surface flow are classified based on vegetation characteristics.

Considering the current status of TW applications around the world and the fundamental design differences between the TW units identified using the classification system, seven ‘Standard Types’ of TW are distinguished. These are denoted by a number above the nomenclature boxes in Figure 2.

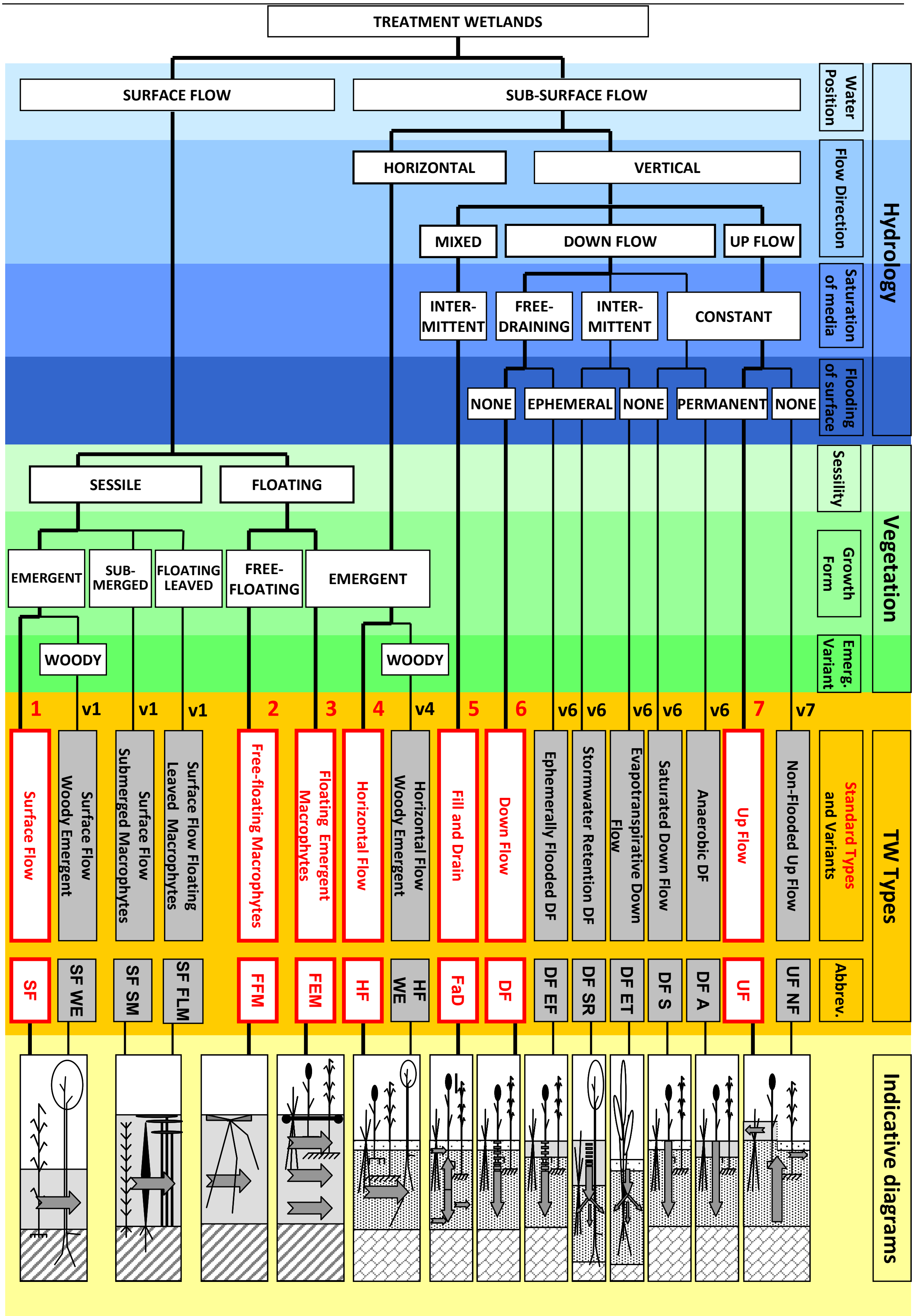


Figure 2 : TW classification tree

The three standard types with surface flow are:

1. Surface Flow (SF) TW, dominated by emergent herbaceous macrophytes.
2. Free-Floating Macrophyte (FFM) TW containing free-floating vascular aquatic plants growing on the water surface.
3. Floating Emergent Macrophyte (FEM) TW with emergent macrophytes growing on a buoyant structure.

The four standard types with subsurface flow are:

4. Horizontal Flow (HF) TW.
5. Fill and Drain (FaD) TW in which the flow direction is mixed, normally alternating between up and down flow.
6. Down Flow (DF) TW, which is free-draining and without surface flooding.
7. Up Flow (UP) TW with a flooded surface.

There are numerous design variants identified in Figure 2, which are considered modified versions of these more commonly applied standard types (e.g. based on atypical vegetation types, saturation degree or flow direction).

An additional sub-set of variants are identified based on specific applications within the Ephemerally Flooded Down Flow (DF-EF) group of TWs as shown by figure 3.

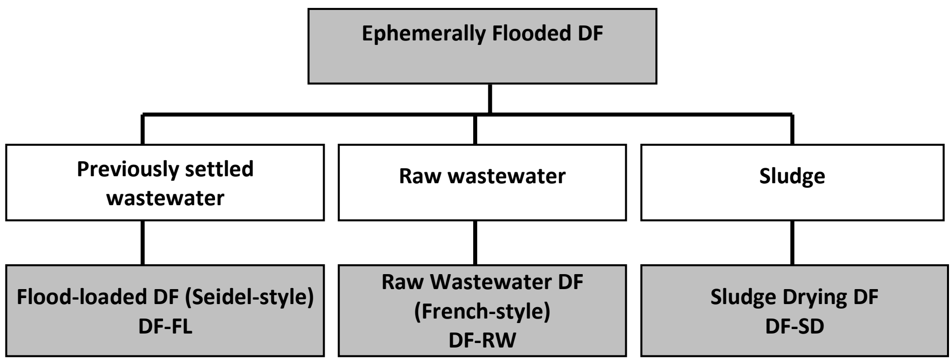


Figure 3 : The sub-classification of the Ephemerally Flooded Down Flow TW variants based on specific influent type.

I.3.9 Intensified Treatment Wetland sub-variants

One special category of amended TW variants are 'Intensified' wetlands, which includes the increasing number of unit designs which are being developed to increase the efficiency and/or overcome process limitations (such as oxygen availability) of conventional zero- or low-energy input, passive wetland designs. After a given TW unit has been classified based on hydrologic and vegetation traits presented in Table 1, it can be further defined using the cross-cutting attribute of 'Intensification' as outlined in Table 2. In principle, intensification represents a range of design or operational modifications that can be applied to virtually any of the TW types that have been classified in Figure 2.

Table 2 : Intensification inputs to enhance the performance of standard treatment wetland units.

Type of intensified input	Intensification Class	Example
Energetic	Aeration	Aerated subsurface flow TWs
	Pumping [a]	Fill and Drain TWs with reciprocation
Physicochemical	Sorptive media	Expanded clay, zeolites, bauxsol, chitinous material
	Chemical dosing	Alum, ferric chloride, oxidising agents
Operational	Frequent plant harvesting	Duckweed systems
	Cyclical resting	Routine alternation between multiple TW units in parallel
	Recirculation of flow [a]	DF TW with recirculation

[a] Energetic intensification via pumping is accompanied by modification of the physical design of the unit, whereas Operational intensification via flow recirculation (also involving pumping) does not necessitate a change in the physical design of the TW unit.

Intensification is generally achieved through increased inputs of electricity (e.g. aeration or recirculation pumping), physicochemical amendments (e.g. dosing with coagulants or flocculants, or use of highly sorptive media), or added operational effort or complexity (e.g. regular plant harvesting or cyclical resting). The use of a pump to move water uphill due to site constraints, or to load water into the wetland in a conventional manner (e.g. pulse loading of a IDF TW) is not considered to be a form on intensification. System designs incorporating specialised or synthesised media, such as those with exceptionally high sorption capacities, are considered to be intensified. The substrates have naturally or have been heavily processed and received relatively intensive energy and/or chemical inputs during the manufacturing process, to lead to an increased treatment efficiency of the TW for certain target contaminants. Intensification is an attribute that cuts across the final level of the classification hierarchy, in most cases representing a modification to one of the standard or variant types of TW identified in Figure 2 and giving rise to an additional sub-set of TW variants.
The types of intensification described thus far can be considered Unit-based forms of intensification because they relate specifically to the design or operation of individual TW units. Another form of intensification can be identified as System-based Intensification, which relates to the way in which different TW units can be coupled together in various ways to form a treatment train with the aim of optimising the treatment efficiency of the overall system. Such systems are commonly referred to as “hybrid” or “integrated” systems.

I.4 THE HORIZONTAL FLOW SYSTEMS, SITE OF NASSOGNE AND PILOTS UNDER GREENHOUSE

I.4.1 General definition

The system investigated within this study belongs to the second major group of TWs, which are those with subsurface flow, the first major group being with surface flow. The majority of flow occurs through a porous media within which most of the treatment processes take place. A main key difference between these systems and those with surface flow is that subsurface flow systems are media-based systems. In some cases there may be ephemeral or permanent flooding of the surface of the media, but this is primarily for the purpose of influent distribution or effluent collection; the majority of treatment still occurs within the porous

media. Subsurface flow systems are sub-classified based on flow direction into those with a horizontal flow path and those where the flow is in a vertical direction.
The operational site of Nassogne is classified as horizontal flow (HF), representing those systems where the inlet and outlet are horizontally opposed. The HF are typically comprised of lined gravel, sand or soil based beds planted with sessile emergent vegetation. The wastewater flows through the rhizoshpere of the plants. Such systems generally have smaller surface areas (<0.5 ha) and higher hydraulic loading rates than SF TW (Xanthoulis *et al*, 2008).

I.4.2 Horizontal Flow TW Standard Type

The standard and most commonly applied type TW with subsurface flow in a horizontal direction contains herbaceous emergent macrophytes as is termed the Horizontal Flow (HF) TW (unit 4 in Figure 2). The presence of herbaceous emergent macrophytes is assumed to be the default condition for the HF TW standard type. The most common species of herbaceous emergent macrophyte used in Europe is the Common Reed (*Phragmites australis*), while a range of other species are often used elsewhere, particularly those from the genera *Schoenoplectus*, *Cyperus*, *Typha*, *Baumea* and *Juncus*.

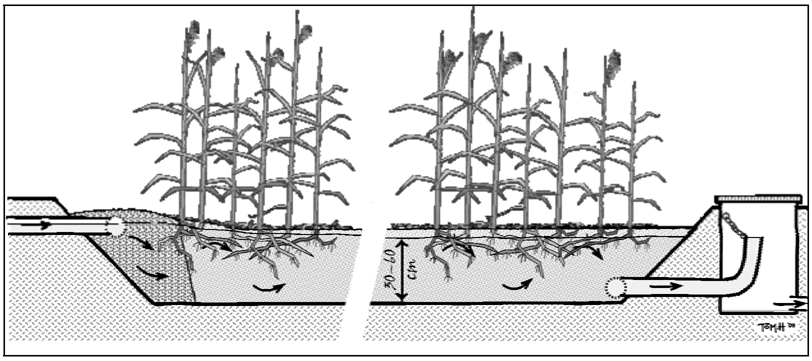


Figure 4 : Horizontal Flow (HF) TW, featuring herbaceous emergent macrophytes.

I.4.3 Common Applications of HF

Horizontal Flow TWs are typically used to treat primary or secondary treated sewage prior to either soil dispersal or surface water discharge. In Europe, the systems tend to provide secondary treatment for village-sized communities of up to about 2,000 population equivalents. In North America, they tend to provide tertiary treatment for larger populations. However, there are many other applications for specialty wastewaters from industry, and acid mine drainage. In general, HF TWs have been utilized for smaller flow rates than SF TWs, mainly because of cost and hydraulic limitations associated with flow through the porous media. These systems are capable of operation under colder conditions than SF TW, because of the ability to insulate the top surface and the thermal buffering provided by the substrate.

I.4.4 Variant of HF

There is one identified variant of the HF TW which is different because of the vegetation type used (variant v4 on figure 2). This variant uses woody rather than herbaceous emergent vegetation, and is termed the Horizontal Flow Woody Emergent (HF WE) TW. Examples of HF WE TWs include systems in Australia used for on-site wastewater treatment and planted with Melaleuca species and systems planted with willows in Europe.

I.4.5 Intensification of HF

The current intensification processes to boost the efficiency of HF TWs in done by means of aeration lines on the bottom of the bed (Intensified aerated HF TW), or specific substrates to enhance reactions like chemical precipitation, pH adjustment, or adsorption.

I.4.6 Le Château du Bois, the HF site in operation of Nassogne

The station investigated was the station of Nassogne, *Chateau du Bois* (Castle of the Wood). The *Chateau du Bois* is also called the Pavillon Bonaparte (Bonaparte's Pavilion). Built and then occupied from 1873 to 1876 by the nephew of Napoleon 1st, it is located at 1,5 km east from Nassogne, on the right of the current National road N889, leading to the Barrière de Champlon in Belgium. Its position is 50°07'22,67'' latitude N and 5°22'19,50'' longitude E.
This old monument is isolated in a wood, and surrounded by more than one hectare of meadows. Occupied for a long time by a large family, it was then taken back by the municipality which completely restored it. A tourist place as rural guest house is developed with 4 rooms for guests, plus one for the caretaker, organised on two floors; a restaurant for 40 people is open on the first floor.
According to municipality studies, the daily average population is 20 PE. The wastewater treatment system has been designed in accordance with this data.
The rural guest house is located in an individual treatment zone (which means that no sewage network will ever collect its wastewater and must treat wastewater on its own before discharge into the environment). The topography with a natural slope and large area available has lead the municipality to order the non profit association Epuvaleau a study to design the treatment system with the planted filters technology and to direct its building and the construction site.
Epuvaleau has designed the EPUVAL 20 G system and all requested documents has been produced for May 2004; the system has been in operation since March 2005.

EPUVAL design for 20 PE , in general
In order to reduce suspended solids and to liquefy wastes, the planted filter EPUVAL 20 G is preceded by a septic tank, the classic model with two chambers, of 12 m³ as minimum volume (600 L/PE). It has a vent hole to evacuate fermentation gases, act as deodorizer and pressure release for the system, and limit the degradation of concrete and steel. This vent hole is connected to a 100 m pipe tube, tied with one of the building wall, leading gases to at least 1 meter higher than the highest window of the building.
The septic tank must be emptied every two years. It is expected to remove 60 % of suspended solids and 30 to 50 % of the BOD. It captures solid materials by flotation and sedimentation, liquefies and partially ferments organic matter. It is located at the highest possible place, to avoid large earthworks and allow gravity flow towards the planted filter located downstream.
A grease remover of 800 litres capacity separates greasy water coming out from the kitchen of the restaurant. Downstream this grease remover, a water trap is placed to exclude any odour upraise towards the kitchen.
As much as possible, the septic tank and planted filter are located away from deciduous trees. The filter is preferably located in a sunny zone. Rainwater is diverted and does neither enter the septic tank, nor the planted bed. It is collected in a separated tank with the aim of reuse for domestic uses such as restrooms, washing machines, etc.
The grease removal system and septic tank are buried at the level of the court yard ("terrasse"). Wastewater flows by gravity from the septic tank to the planted filter through a chamber. The high slope induces some waterfalls from pipes to chamber, what generate beneficial aeration. The lay out of the field is in the humid zone, next to the existing ditch. The longitudinal axis is the natural field slope.

The elements are drawn in figure 5.

EPUVAL filter's cells
The designs constraints were the following:
- All wastewater volumes pass through the filter without overflow or plants flooding
- The filter must remain efficient with foreseeable variation in term of load and flow
- The filter must be able to be totally drained or submerged (regulated level systems by pipes at the entry and at the exit)
- The lay out must be appropriate to the local natural site

The planted filters have a total area of 112 m². According to the gravel media size (3-8 mm) available on the Walloon market and inserted in the calculations of RH Kadlec & RL Knight (1996), 2 cells on parallel separated by a small wall were installed each of which is 4,2 m wide, 13,4 m long and 0,8 m deep. All walls are vertical. Bottoms were levelled and covered by a mix of sand and cement in case of rocky soils; the slope is regular and of 0,4% from inlet to outlet.
An HPDE pond liner (> 1,5 mm, and guarantied for UV and weathering) and a liner against rodents cover bottom and walls of the two cells. Cells were filled 60 cm high with stones of 4-12 cm in the distribution and collection zones over 30 cm, followed for the distribution zone and preceded for the collection zone with large gravels of 10-20 mm over 10 cm and washed pea gravels of 3-8 mm (d_{60} = 5,5 mm (3 to 8 mm) and less than 3% of sand) in the treatment zone. Six pipes per cell (110 mm in diameter) distribute inlet flow; they are below the surface to avoid odours and are 15 cm up from the bottom. The exit drains are 2 cm up from the bottom of the cell and have vacuum breakers (L pipes raising 10 cm up from filters surface) to allow air entry and cleaning maintenance operations. Treated wastewater going out from drains are lifted in a rotating pipe, which regulates the water level in the filter, with a maximum height of 54 cm from the bottom of the filter or 6 cm below the surface.
The station is surrounded by a heavy duty fence, able to avoid boars' invasion in the cells.

Photos of the systems are shown in figure 6; a cross section of the system is shown in figure 7, and a more detailed site plan is shown in figure 8.

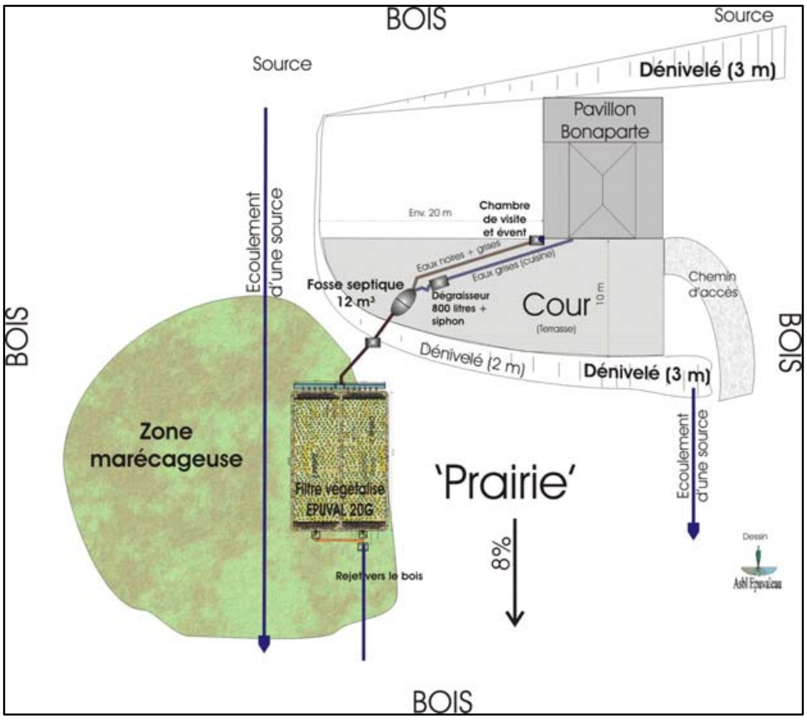


Figure 5 : Schematic lay out of the wastewater treatment system

Figure 6 : Le Château du Bois, Nassogne, over the seasons

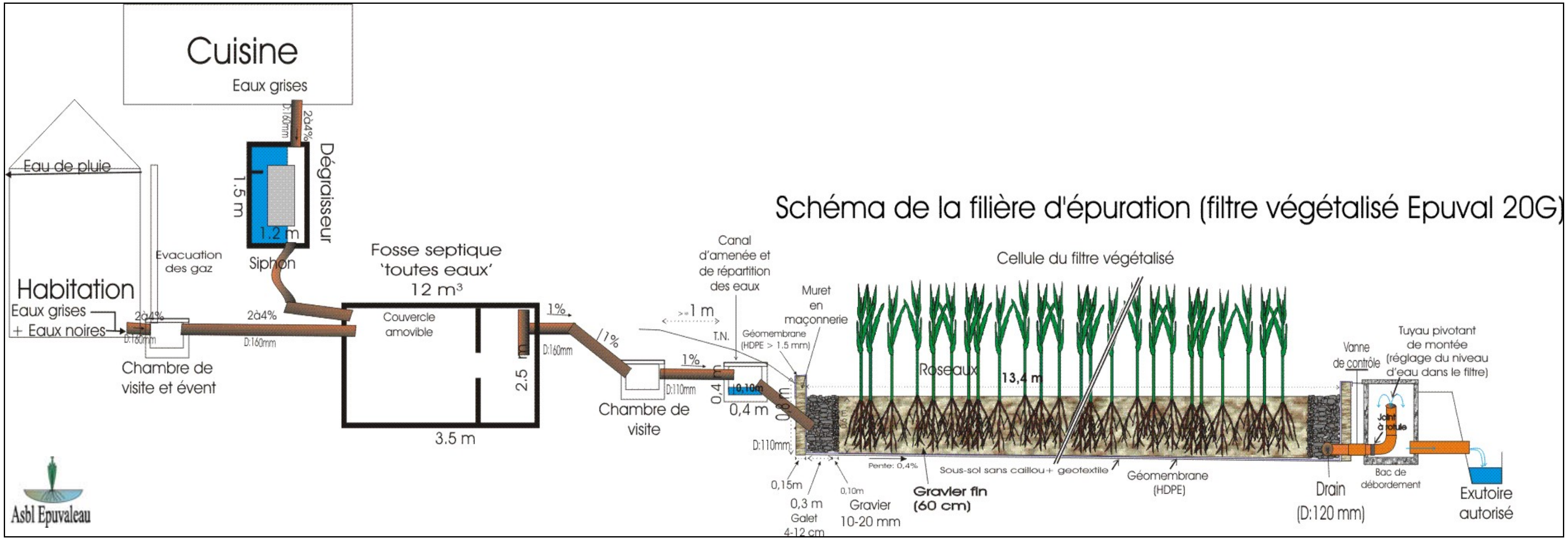


Figure 7 : longitudinal transect of the wastewater treatment system of the Chateau du Bois, Nassogne

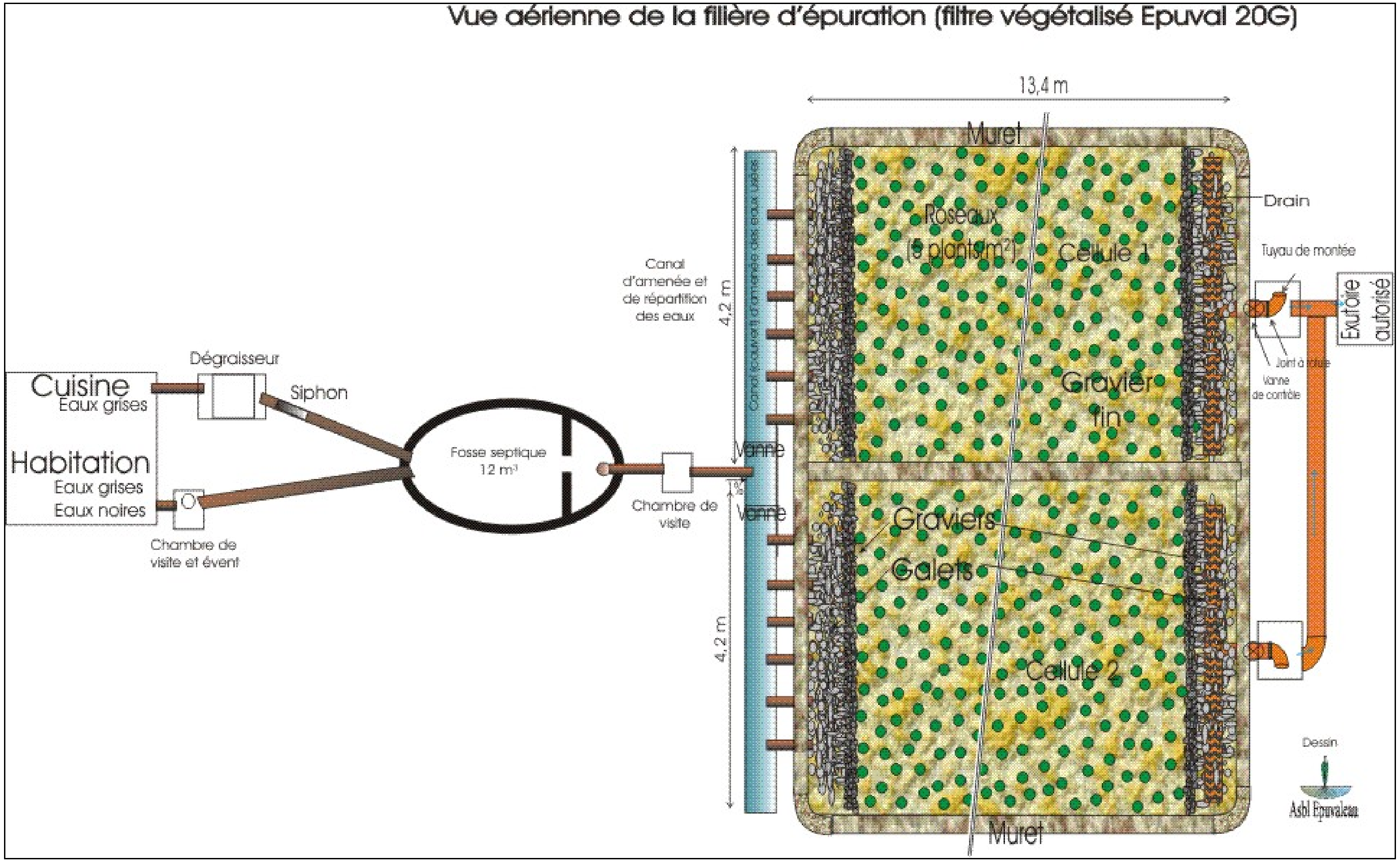


Figure 8 : Aerial schematic view of the wastewater treatment system of the Chateau du Bois, Nassogne

I.4.7 Pilots under the greenhouse

Small scale replications of the Epuval system were built as pilots under a greenhouse. Several studies were done in addition and parallel to the monitoring of the operational site at Nassogne in order to increase the understanding of the HF system developed at full scale.
The four pilot wetland cells were a kind of trench installed under the greenhouse. Trenches have a metal frame with glass sides and bottom (previously used for irrigation in open channel flow tests). The dimensions are : L: 2,8 m, W: 0,3 m, D:0,3 m . They were first covered with a black tarp, and then filled with pea and non-limestone gravels. At the inlet and outlet, the gravel diameter is 10-20 mm, inside the bed the gravel diameter is at first 3-8 mm for all four pilots. The total volume is 0,25 m³ (250 L), water flows horizontally 3 cm under the surface, the apparent net volume is 40 % (or 80 litres). Wastewater is collected at the inlet of the station of Gembloux city, and stored in 4 tanks of 200 litres each.
For the first experiments, 3 pilots were planted with *Phragmites sp.*,1 pilot is running without reeds. According to primary observations, gravels were sifted on October 2006 according increasing granulometry. The media (washed pea gravels) were separated into (3,5-5) mm (5-8 mm), (8-10 mm) and (10-12,5 mm) – using ASTM (American Society for Testing and Materials Standards) sieves from the Hydraulics department. The four pilots each had increasing gravel media sizes, which were not planted to facilitate tracer experiments. These tracer experiments were replicated. After the trace studies, pilots were planted with *Phragmites sp.* Figure 9 provides photos of the pilot units and associated granulometries.

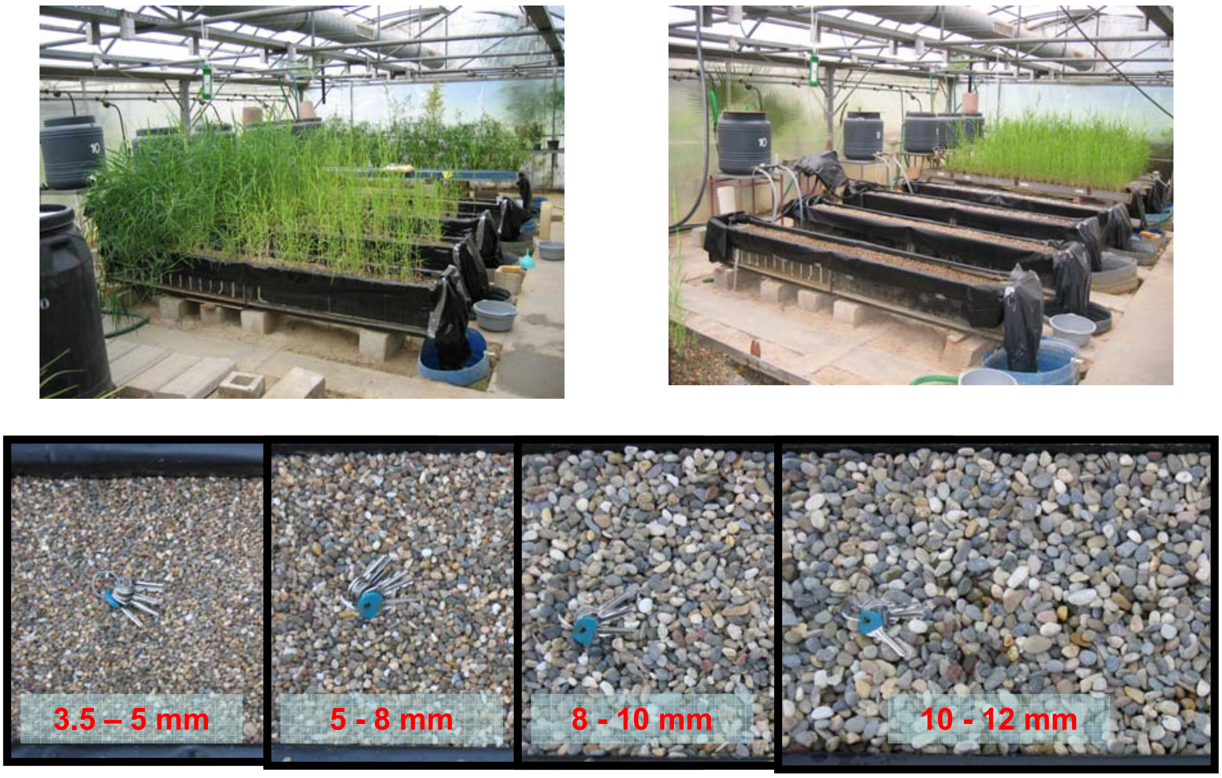


Figure 9 : Photos of the four pilots under greenhouse (planted and unplanted) and the four gravels size within pilots.

I.5 THE VERTICAL FLOW STANDARD TYPES AND THEIR VARIANTS

I.5.1 Vertical flow systems

Vertical Subsurface Flow TWs generally consist of a bed of porous media (sand or gravel) through which the water moves in a vertical direction. In general, this group of TWs incorporates a diverse range of physical designs with respect to hydrologic characteristics. At the level of Flow Direction in the classification hierarchy (Table 1, Figure 2), TWs with vertical flow are sub-divided into systems with downward flow, upward flow, and a combination of up and down flow ("mixed flow") which leads to the three standard types defined within this group of systems. The vegetation type is always emergent and does not play a role in the classification or naming of these systems.

I.5.2 Vertical standard Types: Down Flow, Up Flow, and Fill and Drain TWs

The most common type of vertically flowing TW is the Down Flow (DF) TW (unit 6 in Figure 2) which is free-draining (open outlet at the base of the bed) and remains unsaturated for most of the time. A network of pipes with multiple emitters is used to distribute the flow across the surface of the bed in a way that avoids surface flooding. The vegetation typically consists of herbaceous emergent macrophytes. Inlets are located vertically above the outlets. The bottom-most layer of media usually consists of coarse media with a network of perforated drainage pipes (Cooper *et al.*, 1996), which are sometimes ventilated to the atmosphere to promote passive aeration of the substrate. Often the bed is planted with common reed (*Phragmites australis*), but a range of other emergent macrophytes are also used, as for HF TWs. Influent distribution pipes may be located above the system, or, in cold climates, buried within the granular media bed or under a layer of insulating mulch.

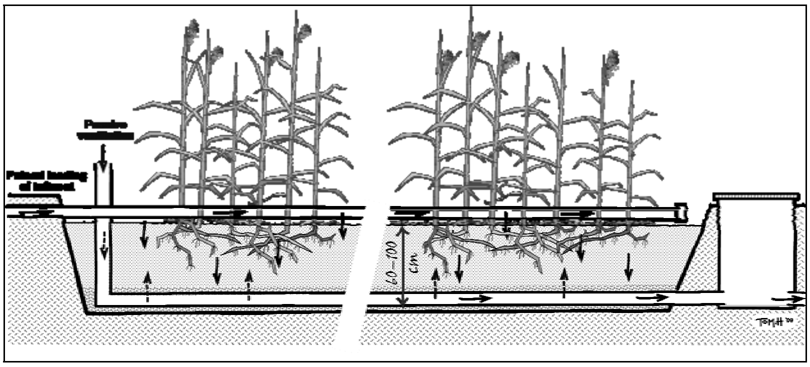


Figure 10: Down Flow (DF) TW; free-draining and with herbaceous emergent macrophytes.

The second vertical standard type of system with vertical subsurface flow is the Up Flow (UF) TW which features a constantly saturated media which is permanently flooded over the surface (unit 7 in Figure 2). The term 'saturated' is redundant because the upward flow configuration dictates that the wetland must have a saturated bed. Wastewater is introduced at the bottom of the media bed via a series of distribution pipes and moves slowly upwards to the substrate surface. For practical reasons of conveying the effluent to the outlet, these systems have a flooded surface. However, these systems are classified as having subsurface flow because the majority of important treatment processes are intended to occur within the saturated bed of media. These systems are sometimes referred to as Anaerobic Beds.

The third vertical standard type of system within this category is the Fill and Drain (FaD) TW which is classified as having a mixed flow direction (Unit 5 in Figure 2). Flow typically alternates between upward and downward flow, although it can sometimes be closer to diagonal flow depending on the relative location of the inlet and outlet. The media in these systems has an intermittent saturation level as it alternates between being saturated and unsaturated as a result of the filling and draining sequences. Normally the upper surface of the media is not flooded. These systems have been investigated for many years at mesocosm and pilot scale and their application at full scale is increasing due to the relatively high rates of oxygen transfer. Fill and Drain TWs can also provide the conditions necessary for complete nitrogen removal within the one reactor, with ammonia adsorption on the media during the filling stage, nitrification under aerobic conditions while the bed is drained, and denitrification with anaerobic condition and carbon source provided by the second filling sequence (Austin, 2006). Several Fill and Drain beds are typically incorporated in series.

Common names for this design include Tidal Flow and Fill and Draw wetlands. However, the term 'tidal' can give the incorrect impressions that these systems may only have two fill and drain cycles per day, may be somehow connected to the lunar cycle, or may be associated with marine or brackish water.

I.5.3 Common Applications of Vertical systems

Down Flow TWs are very similar in design to intermittent sand filters (Liénard *et al.*, 2001), which are widely used throughout the USA, Australia and New Zealand for decentralised wastewater treatment, except that DF TWs are planted with wetland vegetation. Down Flow TWs are used in many European countries particularly for achieving secondary treatment of primary settled sewage. Due to their higher oxygen transfer rates, DF TWs are becoming more common where discharge regulations require removal of ammonium. They are also a common design choice for effluents with a high carbonaceous or nitrogenous oxygen demand, such as landfill leachates and agricultural wastewaters.

Up Flow TWs are commonly applied where anaerobic treatment conditions are required, such as in some mining or industrial applications. They may be less prone to clogging than HF TWs because the influent solids and organic load is distributed over a much wider surface area.

Fill and Drain TWs are being increasingly applied for wastewaters with a high oxygen demand or where removal of total nitrogen is required. They often tend to have a smaller foot-print than other TW alternatives which makes them potentially suitable for arid regions where water loss via evapotranspiration can be a limitation.

I.5.4 Variants to Vertical systems

Down Flow TW Variants

Several variants of the DF TW exist based on the degree of media saturation and the occurrence of surface flooding (unit v6 in Figure 2). The first variant includes a group of designs classed as Ephemerally Flooded DF (DF EF) TWs (Figure 3). Like the standard DF TW, they are free-draining, but they are operated with ephemeral flooding of the media surface as a means of achieving distribution of the influent of the surface of the bed. This group contains three main sub-variants based on the type of inflow they receive. The first variant is similar to the standard Down Flow TW, except that distribution of the influent over the surface is achieved via flood-loading from point discharges, rather than the use of a network of distribution pipes with multiple emitters which maintain the influent essentially below the media surface. Hence, this design variant is termed the *Flood-loaded Down Flow TW* (DF FL) TW. The distribution of the influent relies on the use of a hydraulically restrictive surface layer of media (usually sand). During loading events, the hydraulic conductivity of this surface layer is exceeded and the surface floods. The Flood-loaded DF design is typically used for treatment of wastewater that received at least primary settlement.

It was initially developed by Käthe Seidel in Germany during the 1970s and is still employed today in parts of Europe, particularly the UK (Kadlec and Wallace, 2009).
The second ephemerally flooded down flow sub-variant differs from the flood-loaded DF TW only by the fact it receives unsettled raw wastewater; hence the name *Raw Wastewater Down Flow TW* (DF RW) TW. This is a design variant developed and applied in France for the decentralised treatment of sewage. It is effectively another development of the original Seidel-style DF-FL TW approach. In order to manage the organic sludge layer that accumulates on the surface of the DF-RW beds and maintain permeability, systems employing this design typically have multiple DF-RW beds in parallel with a rotationally rested operation.
The final sub-variant of the DF-EF TW is also defined based on the waste type it receives; namely sludge. The *Sludge-drying Down Flow* (DF SD) TW is periodically flood-loaded with sludge for the purpose of dewatering and mineralisation. The vegetation, soil, sun, and gravity separate solids and liquids from the sludge. The solid fraction of the sludge stays on the surface of the bed while some of the water percolates down through a sand and gravel drainage layer. Substantial dewatering of the sludge occurs via the high evapotranspiration rates of the wetland plants. After each load, a dewatering period is allowed before a new layer of sludge is flood-loaded on top of the dewatered sludge. The plants progressively grow upwards as the stabilised sludge is gradually accumulated in the system. The roots provide drainage channels through the accumulated sludge and also contribute organic matter and enhance the physical structure of the dewatered sludge, thereby facilitating composting and stabilisation processes. This process continues until the bed is filled with dewatered, stabilised sludge (to a depth in the order of one metre) and has to be emptied (after about 10 years). The drainage water receives some treatment as it percolates through the layers of stabilised sludge, sand and gravel and is normally returned to the wastewater treatment plant for further treatment (Nielsen, 2003). They are commonly named Sludge Drying Reed Beds, which can cause confusion with HF TWs also commonly called Reed Beds in the UK. Both common terms are referring to two very different design configurations, and are therefore avoided when describing TW systems.

Another variant of the DF TW identified in Figure 2 is the <u>DF Stormwater Retention</u> (DF SR) TW, commonly referred to as a Bio-retention, Bio-filtration or Rain Garden systems within the stormwater management industry. These systems are typically bunded in order to accumulate runoff during rainfall events which then slowly percolates downwards through the porous substrate of sand or gravel. Hence they are classed as having an intermittent saturation level and ephemeral surface flooding. Woody emergent vegetation is often used in these systems because it tends to be more tolerant of the dry conditions that can develop within the filter between rainfall events.

The <u>Evapotranspirative DF</u> (DF ET) TW variant refers specifically to a design developed in Denmark which uses fast growing woody vegetation with a high evapotranspiration (ET) rate, such as willows, with the aim of evapotranspiring all of the wastewater on an annual basis. Primary treated wastewater is typically introduced near the upper surface of the substrate, giving rise to what could be considered a downward flow path. However, the actual flow direction is somewhat ambiguous because the systems do not have a through-flow of water and the efflux is to the atmosphere. They are classed as having an intermittent saturation level because the water level within the media varies seasonally depending on the balance between inflow, rainfall and evapotranspiration. Down Flow ET TWs are commonly referred to as Zero-discharge Willow Systems. They are typically deeper (approximately 2 m) than other TWs with subsurface flow, in order to provide storage capacity to accumulate water during periods of wet weather and low evapotranspiration.

The forth main variant of DF TW is the Saturated Down Flow (DF S) TW. The main difference to the Standard DF TW lies in the fact that the water level in the bed is maintained slightly below the upper surface of the media resulting in a "constantly saturated" bed (rather than free-draining). Consequently, these systems are very similar to HF TWs with regards to the biogeochemical conditions that develop within the wetland. However, because the influent is distributed across the entire upper surface of the wetland, the influent loading rate of organics and solids in the cross-sectional plain perpendicular to the flow direction are greatly reduced when compared to HF TWs. This means that the substrate clogging potential is substantially reduced compared to a HF TW receiving the same influent loading rate.

The final variant within the DF TW group is the Anaerobic DF (DF A) TW, which is defined as having constantly saturated media and a permanently flooded surface. Although they have a permanently flooded media surface, they are still classed within the subsurface flow group of TWs because the majority of flow and important treatment processes occur within the porous media. These systems are similar in application to Up Flow TWs and are often used to promote anaerobic treatment processes for mining and industrial applications.

Up Flow TW Variants
There is one main design variant for the Up Flow TW group. This is the Non-flooded Up Flow (UP NF) TW design which is identical to the standard UP TW except that the outlet collection pipes are configured in a way to avoid flooding of the upper surface of the media. Maintaining entirely subsurface flow in the up flow configuration can be challenging in terms of design and operation. Hence, most examples of this variant have been only at mesocosm or pilot scale.

Fill and Drain TW Variants
The application at full-scale of this design type is still in its infancy. A range of different design variations based on flow direction have been trialled at mesocosm or pilot-scale, but no consistent variants are currently obvious.

I.5.5 Intensification of vertical systems

Down Flow TWs are often intensified operationally by the inclusion of recirculation of a portion of the effluent back to the pre-treatment stage in order to enhance treatment stability or achieve denitrification. Such systems are called Recirculating DF TWs.
Intensification of the Saturated Down Flow (DF S), Anaerobic DF (DF A) and Up Flow (UF) TWs has been used for treatment of mine waters by incorporation of selected media, such as a mixture of compost and limestone, as the substrate in order to promote anaerobic conditions and the production of alkalinity. In this case, the intensification refers to the use of specific substrates to enhance the treatment efficiency. Such a system is commonly termed an anaerobic wetland or alkalinity producing system (Kadlec and Wallace, 2009). These systems are often constructed without wetland vegetation to avoid plant mediated oxygen transfer to the substrate, in which case they would not be classified as a TW.

Another common form of intensification of the Saturated Down Flow (DF S) TW is to actively pump small bubbles of air into the saturated substrate via a network of aeration lines installed at the base of the bed in order to overcome oxygen transfer limitations. These systems are termed Aerated DF-S TWs, which is abbreviated to A-DF-S TW. Such A-DF-S TWs are often used for treatment of waters with a particularly high oxygen demand, either due to high concentrations of organic compounds (e.g. airport de-icing runoff) or total Kjeldahl nitrogen (e.g. landfill leachates) (Wallace *et al.,* 2006). Such systems are sometimes further intensified through the use of pumping to recycle a portion of the effluent back through the aerated bed. In keeping with the nomenclature protocol, these systems would be termed Recycling Aerated Saturated Down Flow TWs (R-A-DF-S TWs).

An intensified version of the Fill and Drain (FaD) TW is what is commonly termed a Reciprocating Wetland, or if aligned with the proposed nomenclature: a Reciprocating FaD (R-FaD) TW. Intensification is achieved by repeatedly transfer the water in a Fill and Drain fashion between two partnered beds using pumps. A reciprocation cycle typically involves the pumping of the majority of water from one bed to an adjacent drained bed, followed by a rest period. After the rest period, the majority of water is pumped from the full bed back to the original bed which has been resting in a drained state. The number of reciprocation cycles per day is largely dependent on the oxygen demand of the wastewater and is commonly in the order of one complete cycle every one or two hours. Hence, these systems are intensified by the elevated input of energy for pumping of the water.

I.6 THE SURFACE FLOW STANDARD TYPES AND THEIR VARIANTS

I.6.1 Systems with Surface Flow

Systems where the majority of flow occurs through a water column above the surface of the benthic substrate are classified as Surface Flow TWs. These TWs are similar to many natural wetlands in that they contain areas of open water, floating vegetation, and emergent plants, either by design or as an unavoidable consequence of the design configuration.

I.6.2 Surface Flow Treatment Wetlands with Sessile Vegetation

This group of TWs includes all of those which have surface flow and contain macrophytes which have their roots bedded and anchored in the benthic substrate (sessile). Within this grouping are TWs that can be further classified based on the growth form of the dominant vegetation, with the major distinction being between those with emergent, submerged or floating leaved macrophytes.
This group is opposed to the other group where the vegetation is floating rather than sessile. The floating vegetation is distinguished as free-floating or as growing on a floating mat.

I.6.3 Surface Flow TW Standard Type

The most common type of TW with surface flow has water flowing above the surface of a permanently saturated soil substrate in a horizontal direction, with the predominant vegetation type being sessile herbaceous emergent macrophytes as depicted in Figure 11 (unit 1 in Figure 2). This represents one of the most widespread and commonly used TW designs worldwide. Hence, it is designated as the central Standard Type for this group of systems and named it Surface Flow (SF) TW for this design. Note that redundant terms have been excluded from the nomenclature. Because all surface flow systems have flow in a predominantly horizontal direction, the term “horizontal” is not necessary. The presence of sessile emergent macrophytes is assumed to be the default condition.

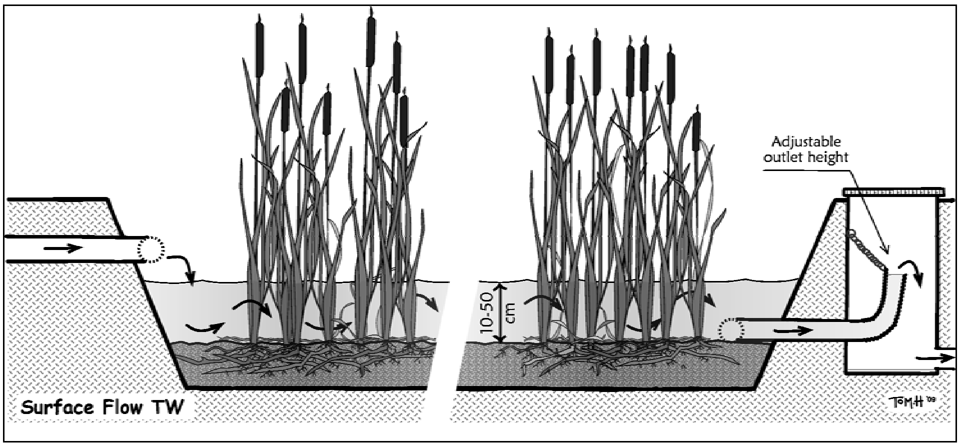


Figure 11: Surface Flow (SF) TW

I.6.4 SF common Applications

The most common application for SF TWs is to polish effluents from secondary treatment processes, such as natural systems (lagoons) or conventional systems (trickling filters and activated sludge). They are suitable in all climates, including very cold regions. Surface Flow TWs are a popular choice for the treatment of urban, agricultural, and industrial stormwaters because of their ability to deal with large variations in inflow rates and moderate, temporary changes in water levels. They are a frequent choice for treatment of mine waters, groundwater remediation and leachate. Surface Flow TWs are also typically the preferred design variant for large-scale applications (greater than one hectare) because at such scales the sand or gravel substrate used in TWs with subsurface flow becomes prohibitive due to cost and hydraulic limitations.
Significant ancillary benefits in the form of recreation and wildlife habitat can be provided by SF TWs. Operating costs of SF with emergent macrophytes are typically quite low and are usually capital cost-competitive with alternative technologies (Kadlec and Wallace, 2009).

I.6.5 SF Variants

All *variants* of the standard SF TW differ based on their vegetation growth form (units designated v1 in Figure 2).
Variant 1a has woody, rather than herbaceous, emergent vegetation and is defined as a Woody Emergent Surface Flow (WE SF) TW. This variant is typically limited to very large applications, often for tertiary polishing of effluents. They typically resemble natural wooded wetlands such as swamps, and are often associated with zones dominated by other vegetation types. Examples exist in the southern states of the USA such as Florida and Louisiana (dominated by Cypress species) and on the east coast of Australia (dominated by Melaleuca species).
Variant 1b is the submerged macrophyte surface flow (SF SM) TW which contains predominantly submerged macrophytic vegetation. These macrophytes have their photosynthetic tissue entirely submerged and typically only grow well in oxygenated water with good clarity. Thus, such systems cannot be used in wastewater with a high content of readily-biodegradable organic matter or high turbidity (Brix, 1993). They are often used for treating storm water where the submerged leaves and associated biofilms provide an effective filter for removing fine suspended sediments.
Variant 1c has sessile floating leaved vegetation and is termed the Floating Leaved Macrophyte Surface Flow (SF FLM) TW. Floating-leaved macrophytes include plant species which are rooted in the benthic substrate with their leaves floating on the water surface, such

as water lilies and some Potamogeton species. To date, the full scale application of SF FLM TWs is rare (Vymazal, 2009). They can have ornamental values when flowering water lily species are used. Floating leaved macrophytes are often used in combination with other emergent and submerged macrophytes (Kadlec and Wallace, 2009) either by accident or to create a more diverse ecosystem. Some floating leaved species have the ability to remove nutrients from the water column which can be beneficial in some cases (Greenway, 2003).

I.6.6 SF Intensification

Surface Flow TWs are intensified in rare cases where effluent is recirculated to the front of the system or where aeration is included at the inlet or in deepened zones which exclude macrophytes. Intensification of SF TWs has been attempted in the form of harvesting farms where the macrophytes are grown with the goal of biomass production (e.g. food for animals, energy biomass or production of ornamental flowers).

I.6.7 Surface Flow Treatment Wetlands with Floating Vegetation

A second sub-set of TWs with surface flow is identified as those where the vegetation is floating rather than sessile. These systems are classified as having surface flow, primarily in a horizontal direction, and remain permanently saturated. The buoyant vegetation means that these systems have more flexibility with regards to water depth as they are not limited by the flooding tolerance of the plants (as with sessile emergent macrophytes) or light penetration through the water column (as with submerged macrophytes).
These systems are further sub-classified into two main types based on whether they contain free-floating plants or emergent macrophytes growing on a floating mat.

I.6.8 Free Floating Macrophyte and Floating Emergent Macrophyte TWs Standard Types

The first Standard Type has free-floating aquatic vegetation (unit 2 in Figure 2) and is named the Free-Floating Macrophyte (FFM) TW (Figure 12).

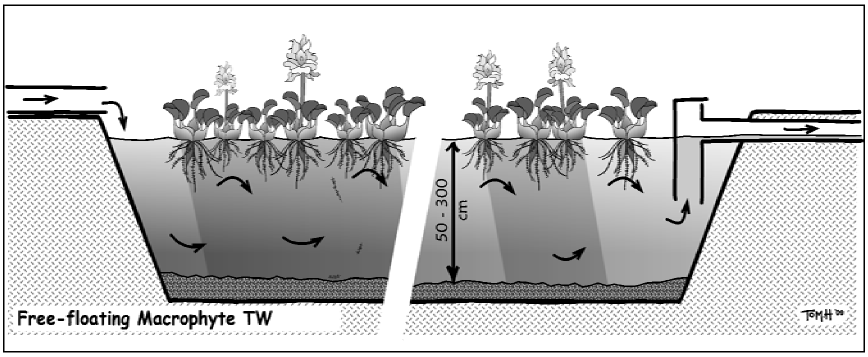


Figure 12 : Free-Floating Macrophyte (FFM) TW.

The second Standard Type of TW with surface flow and floating vegetation contain emergent macrophytes (normally sessile) growing on a mat or raft floating on the surface of a pond (unit 3 in Figure 2) and is depicted by Figure 13. It is referred to here as the Floating Emergent Macrophyte (FEM) TW. Water is treated as it moves through the floating mat and root-biofilm network that hangs in the water column beneath the mat. They are a relatively new and emerging TW type. They have a predominantly horizontal flow direction, with the inlet and outlet horizontally separated, and combined with the capability to tolerate significant vertical water level variations due to the buoyant structure which holds the emergent macrophytes which would not otherwise survive such water level fluctuations.

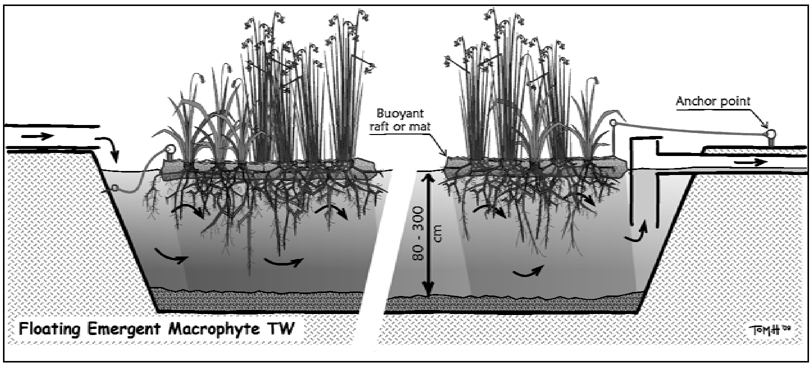


Figure 13 : Floating Emergent Macrophyte (FEM) TW.

The majority of flow in both of these TW units occurs above the surface of the soil substrate. They could therefore be classified as variants of the standard SF TW type. However, due to the unique interaction between the water and the floating plants and the specificity of their application, they are classified as a standard TW types. Hence the term “Surface Flow” has been dropped from the nomenclature and abbreviations for these two designs. In any case, the presence of surface water is an inherent requirement of floating vegetation.

I.6.9 FEM – FFM common applications

Free-floating Macrophyte TWs are most commonly used for the treatment of municipal or industrial wastewaters. Some large-scale systems exist in the USA which have been designed specifically to facilitate regular harvesting of the rapidly growing biomass. There are also numerous examples of their use in tropical countries where suitable species occur naturally and productivity is particularly high. The uptake of this type of TW unit is hampered in many countries by the fact that many of the commonly used free-floating macrophyte species typically present significant weed problems outside of their natural range. Their free-floating nature can make them difficult to contain and easily transportable by waterfowl or flooding.

Floating-Emergent Macrophyte TWs are particularly suitable for drainage and runoff applications that are characterised by fluctuating flow regimes, such as treatment of urban stormwater, combined sewer overflows and agricultural drainage and runoff. This is due to their ability to tolerate variable and relatively deep water levels. They can represent a relatively easy and inexpensive retro-fit option for upgrading existing aquatic systems, such as urban waterways and detention ponds. Because the plants are grown on a buoyant mat, they can be implemented in situations that would otherwise preclude the use of wetland plants due to excessive water depths or unsuitable substrate for planting, such as concrete lined urban canals. Another potential application is the upgrading of existing waste stabilisation ponds, where the root-biofilm network provides additional surface area for attached-growth processes, filtering of particulates. The shade provided by the floating mat can also be of benefit in preventing the excessive growth of phytoplanktonic algae. They have also been used for the remediation of eutrophic lakes and reservoirs.

Currently, no known variants of the FFM TW or FEM TW designs have been classified.

I.6.10 FEM – FFM Intensification

A common form of intensification of FFM TWs is undertaken by the inclusion of regular vegetation harvesting in order to keep the floating plants in an active growth phase and optimise biomass production and uptake and removal of nutrients or other elements. This often requires specially developed harvesting machinery and adapted basin configurations.

Floating Emergent Macrophyte TWs have been intensified through the inclusion of aeration, inclusion of media with high sorption capacity within the floating mat or pumping of water from below to above the floating mat in order to enhance contact between water and the mat-root system. Systems are being developed in China for improving the water quality in polluted canals using commonly eaten plants such as Water Spinach and Watercress with the dual aims of enhancing the harvesting of nutrients while providing an incentive for local people to implement and maintain the FEM TW systems.

CHAPTER II

Hydrology and Hydraulics in Horizontal Flow Treatment Wetlands systems

II. CHAPTER II : HYDROLOGY AND HYDRAULICS IN HORIZONTAL FLOW TREATMENT WETLANDS SYSTEMS.

II.1 INTRODUCTION

Even if treatment wetlands have existed for decades, they are gaining renewal attention as they are adapted systems that qualify as “green technologies” as well as being cost efficient in comparison with many other wastewater treatments.
The challenge in TWs lies in the interconnected processes occurring within the system, being physical, chemical and biological, to achieve the expected pollutant removal.
A principal controlling factor is water movement patterns in the wetlands. Mixing and flow of the various parcels of water and the time that each one resides in a TW determines the extent of reaction for the pollutants of concern (Werner and Kadlec, 2000). This chapter is devoted to the understanding and description of the water movement within the full scale horizontal flow system of Nassogne. Additional studies on small scale pilots compare the general hydraulic behaviour between small scale and full size, but also assess the influence of the gravel media size.
The theoretical background on current knowledge related to hydrology and hydraulics, tracer testing and modelling in TW is presented as an indroduction. Secondly, the experimental method is described and thirdly, results are presented and discussed.

II.2 THEORETICAL BACKGROUND

II.2.1 Water Budget

Water enters wetlands via stream flow, runoff, groundwater discharge and precipitation. Wetlands are losing water via stream flow, seepage and groundwater recharge, bank loses and evapotranspiration, as shown by Figure 14.
In the specific case of treatment wetlands, they receive a continuous or intermittent source of wastewater; they have the same inputs of influent flow and precipitation but often without stream flow or groundwater discharge, and do not trap any catchment runoff. Inflows can be variable and the steadiness of the inflow will drive the ecosystem towards specific ecological conditions, based on microorganisms and macrophytes.
Regarding outflows, treatment wetlands in the Walloon region of Belgium must be watertight and do not allow any loss by seepage, groundwater recharge or bank loss (rules defined in the Water Code). As no stream inflow enters the treatment wetland, no stream outflow is leaving the system either. They are losing water only by the treated effluent outflow and evapotransipiration (ET). ET is the total amount of water contained in soil under liquid phase which is transferred to the atmosphere under the gaseous phase by soil evaporation and plant transpiration. ET depends on atmospheric, weather parameters (solar radiation, air temperature and humidity, wind) and plants parameters (FAO, 1998). ET presents thus the same diurnal and seasonal cycles as those factors. ET can be an important source of water loss on periodic basis and reach 4 to 6 mm/day in the temperate cold climate of Belgium (IRM, online).

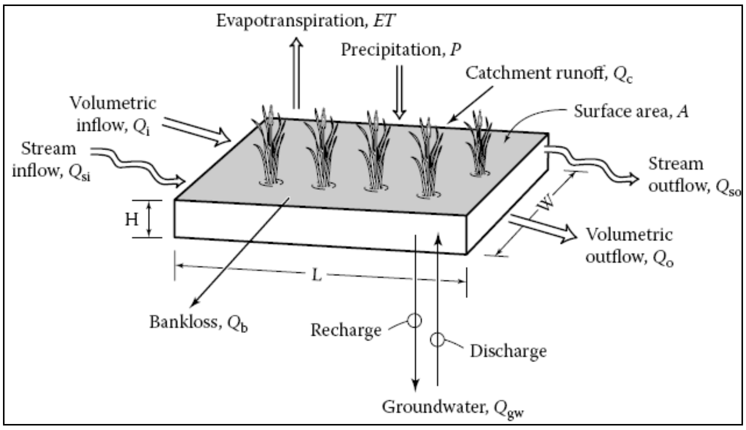


Figure 14: Wetlands water budget (Kadlec and Wallace, 2009).

The current TW investigated offers large variations in the storage in response to the high variability in the inflows. As the TW has outlet water level control structures, the variations in water level are limited. Periods of dry-out or full saturation of the beds can nevertheless occur and they have strong implications for the structure of the ecosystem (plants and microorganisms) but also on water flow patterns which occur through both saturated or non saturated media.
As TWs have the purpose to remove pollutants, the treatment efficiency will highly depend on the duration of the wastewater / biota interactions. This is defined by the concept of detention time, borrowed from other conventional wastewater treatment processes. Other descriptors are also commonly used to define TW hydrology. Those which were investigated and which will be discussed are presented below. Literature terminology on hydrologic variables varies according to the background of authors, which can be from chemical engineering, or hydraulics.

II.2.2 Hydraulics

The hydraulic loading rate (HLR, expressed in m/d) refers to the equivalent water volume of a given flow applied per unit area of wetland and per unit of time:

$$HLR = q = \frac{Q}{A} \qquad \text{(Eq. 1)}$$

where,

q = Hydraulic Loading Rate, m/d
Q = inlet flow rate, m³/d
A = wetland area, m²

It does not imply the physical distribution of water uniformly over the wetland surface (Kadlec and Knight, 1996).

The nominal wetland volume is that enclosed volume multiplied by the porosity of the media.

$$V_n = \varepsilon (LWh)_n \qquad \text{(Eq. 2)}$$

Where, ε = bed media porosity, dimensionless
L = wetland length, m
W = wetland width, m

h	= water depth, m
LWh_n	= nominal wetland volume, m^3

The actual wetland detention time (τ) is defined as the wetland water volume involved in the flow divided by the volumetric water flow:

$$\tau = \frac{V_{active}}{Q} \qquad \text{(Eq. 3)}$$

Where,

τ	=	detention time, days
V_{active}	=	volume of wetland containing water in active flow, m^3
Q	=	flow rate, m^3/d

As it is often more convenient to work with nominal parameters, the nominal detention time (τ_n, in days) sometimes called theoretical mean residence time is defined as follows (Eq.4). For a horizontal flow wetland, the nominal detention time is defined as the volume enclosed by multiplied by the porosity of the clean (unclogged) bed media divided by the volumetric water flow.
A very common designation for nominal detention time is HRT (Hydraulic Retention Time).

$$\tau_n = \frac{V_n}{Q} = \frac{\varepsilon(LWh)_n}{Q} \qquad \text{(Eq. 4)}$$

where:

τ_n	= nominal hydraulic detention time, days
ε	= bed media porosity, dimensionless
L	= wetland length, m
W	= wetland width, m
h	= water depth, m
LWh_n	= nominal wetland volume, m^3
Q	= flow rate, m^3/d

Dimensionless time θ, can be used instead of the nominal hydraulic detention time, τ_n, when comparing tracer response curves monitored over various times, where:

$$\theta = \frac{t}{\tau_n} \qquad \text{(Eq. 5)}$$

Where:

θ	= dimensionless time, unitless
t	= elapsed time, days
τ_n	= nominal hydraulic detention time, days

There are uncertainties related to measurements to obtain Equation 4, in the case of horizontal flow wetlands.

- The first one is related to the measurement of porosity; the porosity of gravel media can be measured at starting but with operational wetlands, roots block some fraction of the pore space, added to mineral and organic deposits.
- The second ambiguity is coming from the flow rate and defining whether it is the inflow rate, whether the outflow rate or an average.

- The third approximate is linked to the active flow as there are stagnant pockets and $A_{active} \leq A$. The gross areal efficiency η is defined as $\eta = A_{active} / A$.
- The last problem is issued from the mean water depth *h*, which is known at the start of the TW but changes as mineral, sand, litter and other deposits occur, added to clogged zones which add to the nominal volume uncertainty .

The volumetric efficiency descriptor lumps empirically those effects and reflects effective volume within a wetland compared to presumed nominal conditions.

$$e_v = \frac{V_{active}}{V_{nominal}} = \frac{\eta(\varepsilon V_{bed})}{V_{nominal}} \qquad \text{(Eq. 6)}$$

Where,
e_v = volumetric efficiency, dimensionless
V_{active} = active wetland volume, m^3
$V_{nominal}$ = nominal wetland volume, m^3
ε = bed media porosity, dimensionless
η = gross areal efficiency, dimensionless
$h_{nominal}$ = nominal water depth, m

In the present specific case of horizontal flow TW, beds are often small enough to exclude significant errors and $V_{bed} / V_{nominal}$ is expected to be close to unity.

Out of equations Eq.3, Eq.4 and Eq.6, it is then clear that

$$\tau = e_v \tau_n \qquad \text{(Eq. 7)}$$

Nominal detention time and hydraulic loading rate (HLR) relationship is given by Eq.8. HLR is inversely proportional to nominal detention time for a given depth. As nominal detention time is meaningful of contact duration, HLR does similarly.

$$q = \frac{Q_i}{LW} = \frac{\varepsilon h}{\tau_n} \qquad \text{(Eq. 8)}$$

Where,
q = hydraulic loading rate, m/d
Q_i = inlet flow rate, m^3/d
L = wetland length, m
W = wetland width, m,
ε = bare media porosity, dimensionless
h = water depth, m
τ_n = nominal hydraulic detention time, days

The actual or interstitial velocity (V, in m/d) of water trough the wetland is defined as :

$$v = \frac{Q}{\varepsilon A_c} \qquad \text{(Eq. 9)}$$

where,
v = actual water velocity, m/d
Q = flow rate, m^3/d

εA_c = interstitial area perpendicular to the flow, m²

The superficial velocity (u, in m/d) is defined as follows :

$$u = \frac{Q}{A_c} \quad \text{(Eq. 10)}$$

where,

u = superficial water velocity, m/d
Q = flow rate, m^3/d
A_c = wetland cross-sectional area, m²

The relationship between the actual and the superficial velocity is easily established

$$u = \varepsilon v \quad \text{(Eq. 11)}$$

This equation demonstrates that in the case of HF wetland systems, where porosity of the bed media is in the average around (0.35 – 0. 4) induces a large difference between superficial and actual water velocities. In SF wetlands systems, porosity is nearly unity (typically around 0.95), so there is not much difference between *u* and *v* (Kadlec and Wallace, 2009).

The water flow through a planted bed of porous media is depending on the fundamental relations between head loss and flow rate. The description of flow through a porous media under saturated conditions is given by Darcy's law. As wastewater enters a HF TW, its velocity is reduced as it flows through the bed media. The majority of the wetland bed is occupied by rooting medium (sand or gravel) which occupies 58% to 65 % of the total volume (U.S. EPA, 1988). As a result, the flow is forced through interstitial spaces in the bed media and the flow rate per unit of cross sectional area is lowered mainly due to frictional losses that occur during subsurface flow (Wallace and Knight, 2006).

Darcy's law is given by Equation:

$$Q = k\, A_c S_w \quad \text{(Eq. 12)}$$

Where,
Q = Average flow through the wetlands, m^3/d
k = hydraulic conductivity, m/d
A_c = wetland cross-sectional area, m²
S_w = slope of the hydraulic gradeline, m/m

At the initial stage of operation, the hydraulic conductivity is simply a function of the size of the selected medium. With time, the microbial biofilm (biomat) developed around each particle of the medium entraps organic and inorganic solids, which reduces the hydraulic conductivity of the medium. As the biomat formation is not the same along the bed, being more developed at the inlet due to high organic loading, hydraulic conductivity is also varying according to the location of the bed. The extent and rate of biomat formation is depending on two factors: (i) the organic loading rate versus the cross sectional area of the bed and, (ii) the size of the bed medium (Wallace and Knight, 2006). This explains why in HF beds, the distribution zones of inlets are usually filled with coarser gravels than the core of the bed. (The problems related to biomat formation are briefly described in the clogging processes within the next chapter but is not part of the main aspects investigated by this work.)

The slope of the water (S_w), or hydraulic gradient is governed by the hydraulic conductivity (k_s) of the bed and the flow velocity (Equation 11).

The one dimensional version of Darcy's law, (restricted to laminar regime) is expressed as follows:

$$u = -k \frac{dH}{dx} \quad \text{(Eq. 13)}$$

Where, u = superficial water velocity, m/d
k = hydraulic conductivity, m/d
H = elevation of the water surface, m

The broader version adding the turbulent regime is:

$$-\frac{dH}{dx} = \frac{1}{k} u + \omega u^2 \quad \text{(Eq. 14)}$$

Where, ω = turbulent factor, d^2/m^2

The turbulent contribution is negligible when the Reynolds number is less than 1.00, and may be ignored with small error at Reynolds numbers up to 10.

The particle Reynolds number is defined as:

$$\mathrm{Re} = \frac{D\rho u}{(1-\varepsilon)\mu} \quad \text{(Eq. 15)}$$

Where, D = particle diameter, m
ρ = density of the water, kg/m^3
μ = viscosity of water, kg/m.d

Sand is typically in the laminar range, and rock media are often in transition region between laminar and turbulent, with significant contributions from the turbulent term. When velocity is beyond laminar range, the effective hydraulic conductivity will depend on the velocity (Kadlec and Wallace, 2009).

These developments make the assumption that the wetlands are in steady state conditions and it is presumed that the porous medium is isotropic. Both assumptions have been demonstrated as not true (Kadlec and Wallace, 2009) as will be demonstrated by the experimental results presented in this thesis.

II.2.3 Tracer testing and modelling

In order to understand the flow process and quantify its parameters, tracer techniques are commonly used in applied hydrology:
"In hydrology, tracer tests are used to address issues like surface water/groundwater interactions, paleohydrology, water movement in very low permeable rocks, calibrating and validating numerical flow and transport models and evaluating vulnerability of water resources. The empirical observation of flow and transport processes with tracers and the

theoretical formulation of flow and transport processes depend on each other and have resulted in a beneficial coevolution of both approaches if adequately combined. Tracers provide empirical data of real and often unexpected flow patterns, whereas models provide tools for flow and transport predictions” (Leibundgut *et al.*, 2009).
Inert tracer tests are used to gain information about internal hydraulics in the treatment wetland field by engineers and wetland scientists, as it provides a mean of tracking the movement of water through a wetland (Kadlec and Wallace, 2009). The theory and practice in the treatment wetland field have been first investigated by US authors, where the majority of wastewater treatment plants were treatment wetlands with surface flow systems or stabilisation ponds. Therefore, the developed theory was mainly based on chemical engineering, considering the pond or wetland as a chemical reactor with laminar flow wetlands. The developed theories were later adapted to European subsurface horizontal flow. It is now questionable whether chemical engineering or hydraulics of solute transport through porous media theories are the most adapted and relevant for subsurface horizontal gravel beds. This work screens both theories and will demonstrate their respective advantages and drawbacks, as well as the distinct information they both provide.

Finding the parameters from the tracer experiment is only possible if an adequate mathematical model is used, meaning that the model is based on the proper concept of tracer transport and its behaviour in the system.

Some definitions are given below from Maloszewski and Zuber (1982) and the application of some of the mathematical models is developed.

- A *Conceptual model* is a qualitative description of a system and its representative factors (e.g. geometry, hydraulic connections, parameters, initial and boundary conditions) related to the intended use of the model. In practice, the conceptual model demonstrates the general concept of water circulation in the system.

- A *Mathematical model* is a mathematical description of a conceptual model, representing a hydrological, physical and/or hydro-chemical system, using functions designed to help in understanding and predicting the behaviour of the system under specified conditions. In tracer hydrology the mathematical model represents the solution to the mathematical equation(s) describing water and tracer transport for given boundary conditions.

- A *Model calibration* is a process in which the mathematical model assumptions and parameters are varied to fit the model to the experimental data. Calibration can be carried out by a trial-and-error procedure, or by an automatic fit based on an objective function. The calibration of the model to experimental data solves the inverse problem by finding the right values for system parameters.

- A *Model validation* is a process of obtaining assurance that a model is a correct representation of the process or system for which it is intended. Ideally, validation is obtained when the parameters derived from the model agree with independently measured parameters (e.g. porosity) as described above.

The tracer method is usually applied to a system that is poorly known. As a consequence the mathematical model required to determine the system parameters must be as simple as possible. Mathematical modelling of experimental data in tracer hydrology can be separated into two different approaches, depending on the tracer method considered.

The two approaches are i) Inverse modelling of information provided by tracers and
ii) Mathematical modelling based on the transport equation.

Tracers are applied in hydrological systems mainly: (i) for the quantitative determination of flow properties (e.g. water velocity, hydraulic conductivity, dispersivities, porosities, transit time, volume of water, flow rates); and (ii) for the calibration or validation of numerical flow and transport models (Maloszewski and Zuber, 1992).
The estimation of parameter values from tracer experiments is only possible if an adequate mathematical model is used: the selected model must reflect the tracer transport and tracer behaviour in the system being studied (Leibundgut et al, 2009).

The removal efficiency of a pollutant within a wetland is depending on the contact duration between wastewater and the biota. The Residence Time Distribution function (RTD), also called Detention Time Distribution (DTD), is a basic characteristic of wetland hydraulic properties as it describe the whole spectrum of time lengths that wastewater spend in the wetland and the time during which it interacts with the biota. Effective RTD are provided by combined use of tracer techniques and mathematical modelling. The RTD function is represented by the Tracer Breakthrough Curve (TBC) measured at the exit of the wetland for an instantaneous injection of tracer into the inlet of the wetland. Models used to describe RTD functions of constructed wetlands are frequently based on concepts developed in chemical engineering (Kadlec, 1994, King et al, 1997, Werner and Kadlec, 2000) to characterize flow patterns in chemical reactors (Levenspiel, 1972). In those approaches, CWs are considered as having different flow regimes: Plug Flow (PF), Dispersed Plug Flow (DPF), Complete stirred Reactors (CSTR) or a simplified version of some of them, e.g. Tank In Series (TIS). Those models are frequently applied in TW field as they were developed at first for stabilization ponds and Surface Flow systems, where it is relevant to consider the ponds as a reactors and the flow within them as laminar. The DPF do not consider the flow as laminar but focuses on intermixing induced by axial dispersion. Those models have then been adapted for horizontal subsurface flow systems having a granular, soil or sand medium.

One of the original aspects of this work is to investigate models developed in groundwater hydrology to describe flow though porous media without stagnant water "which are in principle as well relevant as models from chemical engineering to describe the subsurface flow through gravel beds" (Malosweski and Zuber, 1982). The models developed are Dispersion Model (DM) and the Multi Flow Dispersion Model (MFDM). Dispersed Plug Flow developed by chemical engineering and Dispersion Model developed in hydraulics are at the end similar and consider fluctuations due to different flow velocities and due to molecular and turbulent diffusion.

Regardless of the model used, its parameters are estimated by fitting the theoretical RTD appropriate for the given model to the experimental TBC obtained by the tracer test.
All modelling approaches here below described tracer transport are one-dimension advective-dispersive equations. This is the case as well in chemical engineering approach as for groundwater flows. Significant mathematical differences between models are issued from various formulations of boundary and initial conditions.

In the frame of this work, two tracer studies were replicated on the operational site and three tracer tests were replicated on small scale pilots under greenhouse for comparison and scale up. The aim is to define hydrological parameters of our systems in order to understand the flow pattern within the systems but also adjust future design parameters.

The methodology first to investigate conceptual models, which are:

(1) Plug Flow Reactor – PFR,
(2) Completed Stirred Tank Reactor – CSTR,
(3) Tank In Series - TIS
(4) Dispersion model - DM, also called Dispersed Plug Flow- DPF and
(5) Multi Flow Dispersion Model – MFDM,

They will explain the principal modelled patterns of flow in the wetland.
Secondly, the mathematical equations of some of the models (TIS, DM, MFDM) are presented as well as their solutions.
Thirdly, the most relevant models (TIS, DM, MFDM) are calibrated by the Moment method and the Least Square iteration methods with the application of experimental data to find the system parameter values which will be discussed.
Finally, the validation of the mathematical models is discussed.

The two first parts of presenting the conceptual and mathematical models transport equations are presented here within the theoretical background section; the two other parts of the work are presented within the results and discussion sections.

II.2.4 Conceptual - mathematical models and their transport equations

Chemical engineering approach makes the assumption to investigate flow patterns within reactors with laminar flows while groundwater hydrologists consider working with solute transport through porous media under turbulent flows.
The conceptual models of Plug Flow, Dispersed Plug Flow, Complete Stirred Tank Reactor and Tank in Series are issued from chemical engineering theories. They are describing laminar flows except dispersed plug flow which considers some turbulence.
The two conceptual models of Dispersion Model and Multi Flow Dispersion Models are mathematically describing turbulent flows and are based on hydrology theories.
They are all briefly explained here below in order to compare final values found for hydrological parameters.

1. Ideal Flow Reactors : Plug Flow and Complete Stirred Tank Reactor

In laminar flow, two different ideal approaches (PFR / CSTR) are frequently used in chemical engineering to describe flow patterns in a system. These approaches are based on ideal hydraulic behaviours but are rarely observed in reality. However, they are extremely useful because they are the starting point to model non ideal flows. PFR and CSTR reactors behave very differently in response to the input of a conservative tracer.

In a **plug flow reactor** (PFR), fluid particles pass through the tank and are discharged in the same sequence in which they entered. The PF reactor represents a situation where there is no internal mixing within the reactor; water parcels move in unison from the inlet to the outlet.
All particles remain in the tank for a time equal to $\tau_{n,}$ the theoretical detention time.
This type of flow is approximated in long narrow tanks in which longitudinal dispersion is minimal or absent (Metcalf and Eddy, 1991). An ideal PFR assumes a square wave of tracer transferred without change from the inlet to the outlet (Edeline, 1998).
To discuss tracer behaviour, it is useful to review the concepts of hydraulic detention time. A spike input of tracer entering a PF reactor will move through the system with zero mixing. As a result, the tracer spike will exit the reactor unchanged at τ_n ($\theta = 1$) (Figure 15).

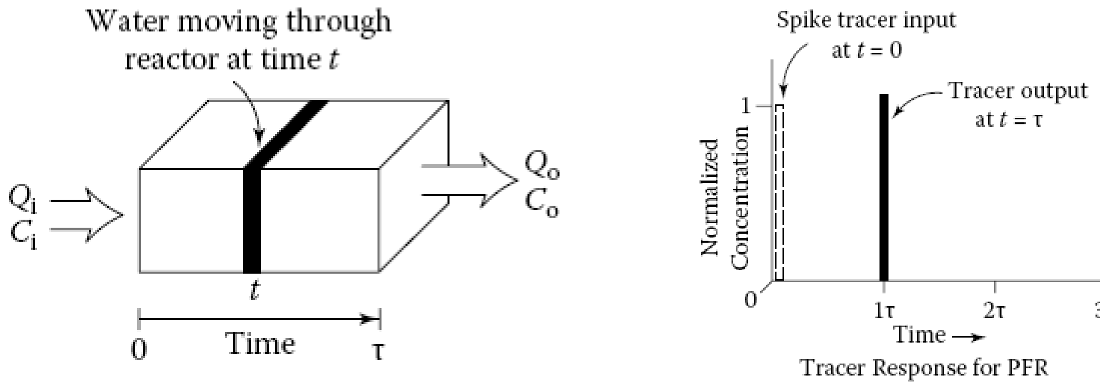


Figure 15 : Tracer response for PFR (Kadlec and Wallace, 2009)

In a **complete stirred tank reactor** (CSTR), particles enter the system and are instantaneously dispersed in the entire volume of the tank. The CSTR represents the ideal of perfect mixing; water entering the system is instantaneously mixed throughout the reactor. Particles leave the tank in proportion to their statistical population (Metcalf and Eddy, 1991). In a CSTR reactor, the tracer impulse is instantaneously and uniformly distributed among the tank contents (Levenspiel, 1972). As flow continues to enter the tank, tracer-contaminated water is displaced, resulting in declining tracer output curve with a long tail (Figure 16).

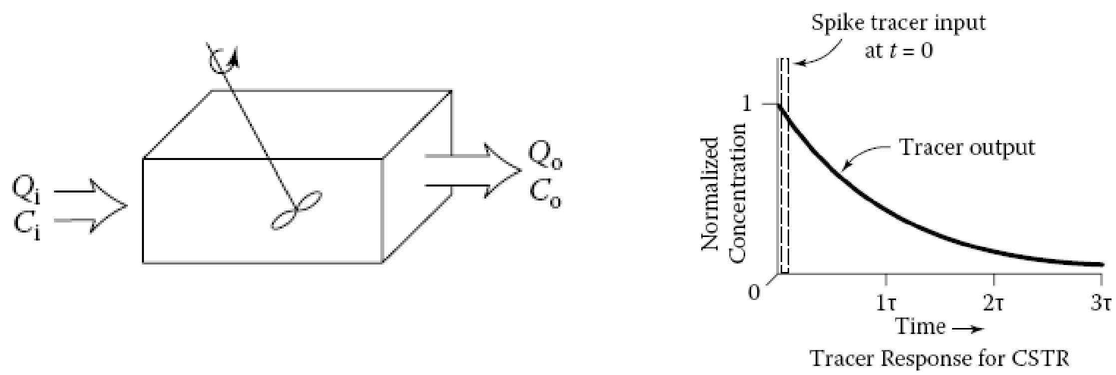


Figure 16 : Tracer Breakthrough curve (TBC) for CSTR (Kadlec and Wallace, 2009)

2. Non Ideal Flow Reactors: Tank In Series

Kadlec, 1999 demonstrated with previous well documented works that the flow patterns through treatment wetland systems are non-ideal and do not conform to either the PFR or CSTR ideals. In subsurface wetland systems, dispersion and mixing occurs within the bed as water flows between the gravel particles or sand grains, and roots may create preferential flow paths near the bottom of the wetland cell (Liehr *et al*., 2000). These combined phenomena produce a distribution of transit times for water parcels. The combined effect of these processes can be demonstrated by passing an inert tracer through the wetland. An impulse of tracer, added across the flow width, moves with water through the wetland as a spreading cloud. Many treatment wetlands have been tracer tested, and all exhibit a significant departure from plug flow (Kadlec, 1993; Stairs and Moore, 1994; King *et al*., 1997). "Real-world" flow patterns, can be approximated using a variety of different flow models. The simplest, and most widely used, is to assume that the wetland can be represented as a series of CSTRs as shown by Figure 17.

The series of completely stirred reactor, **tank in series, TIS** is used to model the flow regime that exists between the hydraulic flow patterns of CSTR and PF reactors. The TIS model is defined as a number (*N*) of equally-sized, perfectly mixed reactors (R_1, R_2, .. R_N) arranged in series. The number of tanks can be any integral number between 1 and ∞. The response of this series of tanks is calculated from the dynamic tracer mass balance equations for the reactors.

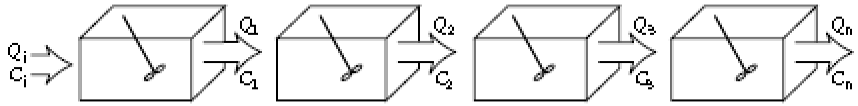


Figure 17 : Equally sized tanks in series (Kadlec and Wallace, 2009)

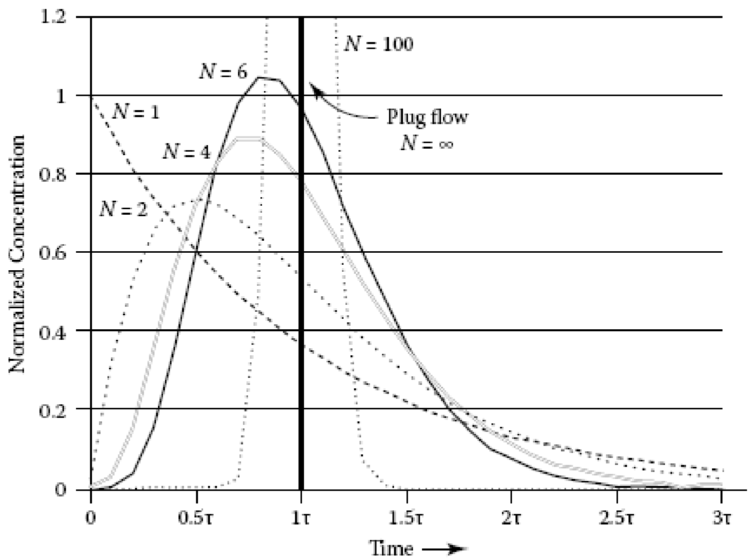


Figure 18 : TBC for TIS (Kadlec and Wallace, 2009)

From Figure 18, it is shown that the shape of the curve is sensitive to change as *N* is small. Nominal detention time Eq. 4 is not necessary indicative of the actual detention time because it assumes that the entire volume of water in the wetland is involved in the flow. In the case of preferential flow pathways and dead zones, this assumption may lead to an erroneous estimation of the actual detention time (Kadlec and Knight, 1996).

Residence times in reactors are studied by injecting a tracer according to a specific program, and by recording tracer concentration at the outlet. Recorded data provide a residence time distribution (RTD), or also Detention Time Distribution (DTD) (Edeline, 1998). The RTD represents the time various fractions of fluid (water in the case of wetland) spent in the reactor; hence, it is the contact time distribution for the system (Kadlec and Knight, 1996).
The RTD is a probability density function for residence times in the wetland. This time function is characterized by several F(t)Δt fractions which stays in the wetland for a length of time equal to t + Δt. For an impulse of tracer (injection: t=0), this density function is:

$$f(t) = \frac{QC(t)}{\int_0^\infty QC(t)dt} \qquad \text{(Eq. 16)}$$

where,

f = RTD function, d^{-1}
C(t) = exit tracer concentration, g/m^3
t = time, day

The numerator is the mass flow of tracer in the wetland at a time t after injection. The denominator represents the total mass of tracer injected.
If the flow rate is constant, it simplifies to:

$$f(t) = \frac{QC(t)}{Q\int_0^\infty C(t)dt} = \frac{C(t)}{\int_0^\infty C(t)dt} \quad \text{(Eq. 17)}$$

The TIS model is defined as a number (*N*) of equally sized, perfectly mixed tank in series. The number of tank can be any integral number between 1 and ∞. If the number of tank in series is N = 1, the reactor behaves as a CST reactor, and if N = ∞, as a PF reactor. Since all the units are of equal volume, the tracer detention time of the entire system is $\tau = N\tau_1$. If a unit impulse of concentration is fed to the series of tanks as a feed concentration condition, the resulting effluent concentration from the N^{th} tank is the tracer concentration response according to the model. Thus Levenspiel (1972) demonstrates that the RTD curve (or TBC) for the TIS model can be represented by:

$$C(t) = \frac{N^N t^{N-1}}{\tau^N (N-1)!} \exp\left[\frac{-Nt}{\tau}\right] \quad \text{(Eq. 18)}$$

In the case of TIS model, the **gamma function** and distribution can be applied and is a more accurate function to define the number of tanks in series, *N* adjusted with τ for the time *t.* It is important to note that gamma distribution describes TIS mixing but the inverse is not true; the gamma distribution of detention time does not imply the existence of turbulent mixing. Indeed, a gamma distribution may also arise from totally unmixed, separated travel paths with different velocities (Kadlec, 2000) what is described by multi flow dispersion model -MFDM.

Because most of wetlands operate as a few number of TIS, it is advantageous to change *N* from a discrete integer variable to a non integer (continuous) one. This enables to use fractional values of *N*, increasing the flexibility to fit data sets in models.

The TIS model is a Gamma distribution *f(t)* of detention time defined as :

$$f(t) = \frac{N^N t^{N-1}}{\tau^N \Gamma(N)} \exp\left(\frac{-Nt}{\tau}\right) \quad \text{(Eq. 19)}$$

Where the Gamma function Γ*(N)* is defined by :

$$\Gamma(N) = \int_0^\infty \exp(-x)\, x^{N-1} dx \quad \text{(Eq. 20)}$$

Both the gamma distribution and Gamma function are now available as computer spreadsheet tools (e.g. GAMMADIS and LNGAMMA in Excel™) and it returns values of *f* for the time *t* and the parameters *N* and τ.

3. Non Ideal Flow Reactors: Dispersion Model (DM)

The conceptual model is based on dispersion theory under turbulent flow, developed in chemical engineering under the name of **Dispersed Plug Flow** and in hydraulics as **Dispersion Model**.

It is another model of non ideal flow reactor, which uses a dispersion process superimposed on a plug flow model (Figure 19). Mixing is presumed to follow a convective diffusion equation. The dispersion coefficient describes eddy transport of water elements both upstream and downstream. If the dispersion is infinite, the system will behave as a CSTR. While if the dispersion equals to zero, then the system will behave as an ideal PFR.

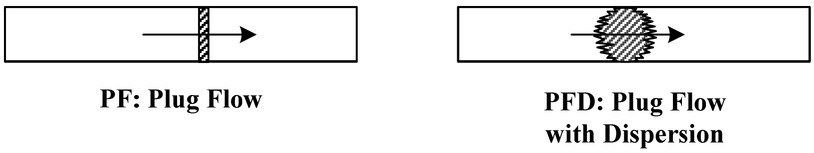


Figure 19 : Comparison of PF and DPF reactors

The theory here presented is issued from the French reference book in hydraulics which is “l’approche mathématique du mécanisme de la dispersion des matières solubles en écoulement turbulent” from Corlier et al, 1972. In chemical engineering, the author of reference is Levenspiel who published on the same year (1972) his book entitled “Chemical Reaction Engineering”.

When a tracer is injected into a turbulent flow, it is carried by convection but on the same time, the initial outline of the zone including the tracer is distorted and the tracer is scattered in all directions. The dispersion process is explained as the result of the combined action of diffusion, caused by turbulent and molecular transfers, and velocities variations across the section. Taylor on 1953 studied the effect of the double action on longitudinal dispersion. His first works were related to dispersion for laminar flows in cylinder pipes. Taylor developed on 1953 a similar theory adapted to turbulent flows. Elder on 1959 adapted the theory from Taylor to flows in open channels, with infinite width. The longitudinal dispersion coefficient (D_L) is directly issued from those theories.

“The dispersion model (DM) considers the medium as homogenous. The flow is characterized by a specific volume rate, mean transit time of water and dispersivity (or dispersion parameters). When the tracer is injected into the water flowing throughout the whole cross sectional section of the column perpendicular to the flow, here taken as the cross sectional area of the HF bed, transverse dispersion can be neglected in both the *y* and *z* directions. If the x-axis is taken along the flow direction, then the concentration gradients in the y and z directions both equal zero. :

$$\frac{\partial C}{\partial y} = \frac{\partial C}{\partial z} = 0 \qquad \text{(Eq. 21)}$$

The 1-D transport equation for molecular diffusion in the *x* direction is given by Fick’s law and the differential equation is:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \qquad \text{(Eq. 22)}$$

Where, D = coefficient of molecular diffusion, and is a parameter which only characterises the process
C = concentration of tracer in the water,
x = distance from inlet injection point,

In an analogous manner, it is considered to describe the contribution to back mixing of fluid flowing in the x direction by a similar form:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \quad \text{(Eq. 23)}$$

Where, D = Longitudinal or axial dispersion coefficient.
(D from equation 22 and D from equation 23 do not express the same coefficient)

It uniquely characterises the degree of back mixing during the flow. Longitudinal or axial is used to distinguish mixing in the direction of the flow from mixing in the lateral or radial direction, which is not the primary concern" (Corlier et al, 1972).

Different PF models with dispersion can be obtained by varying the boundary conditions for solving the advection–diffusion equation of the tracer in one dimension (Levenspiel, 1972):

$$D\frac{\partial^2 C}{\partial x^2} - \frac{\partial (uC)}{\partial x} = \frac{\partial C}{\partial t} \quad \text{(Eq. 24)}$$

where u = actual water velocity, m/d
D = dispersion coefficient, m^2/d
x = distance from inlet toward outlet, m

The boundary conditions are the flow condition at the injection and measurement points; they influence the shape of the obtained curve. Figure 20 sketches two of the many possible boundary conditions for a flow vessel.

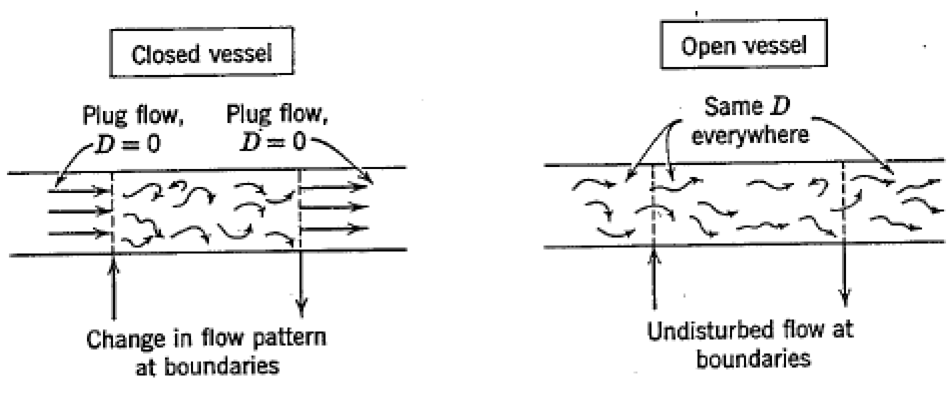


Figure 20 : Closed an opened flow vessels (Levenspiel, 1972)

For low dispersion numbers ($D < 0.01$) the analytical solution of Eq. 24 is not greatly affected by boundary conditions, and it predicts a symmetrical distribution of the tracer response at

any one time, equivalent to a normal probability density distribution function. However, TW with horizontal flow systems have a significant degree of deviation from the PF model ($D > 0.01$) as the tracer response is clearly asymmetrical (Chazarenc et al., 2003). Within this context the appropriate boundary conditions are those of closed–closed vessels and there are no analytical solutions for Eq. 24. In this case the curve can be constructed by numerical methods.
"For tracer experiment performed in a column, when the tracer is injected instantaneously and initial and boundary conditions as follow:

$$\begin{aligned} &C(x=0,t) = \frac{M}{A}\delta(t) \\ &C(x,t=0) = 0 \\ &\lim_{x\to\infty} C(x,t) = 0 \end{aligned} \qquad \text{(Eq. 25)}$$

Where, M = mass of the tracer injected,
A = cross sectional area of injection,
x = location point where the tracer is observed.

The solution to the 1-D transport equation is given by:

$$C(x,t) = \frac{M}{A\sqrt{4\pi D_L t}} \exp\left[-\frac{(x-vt)^2}{4\,D_L t}\right] \qquad \text{(Eq. 26)}$$

This equation has 2 parameters, D_L and v, the values of which can be calculated from experimental data obtained from the outflow" (Leibundgut *et al*, 2009).

This represents a convenient and commonly used experimental device from which it is not difficult to obtain the RDT curve (Levenspiel, 1972). For this reason in environmental problems, although a wide entrance and exit conditions are encountered, most are usually considered approximately open–open (Tchobanoglous *et al*., 2003).

If $D_L = 0$ the system is a PFR,
If D_L has a very high value, the flow tends to equal the flow of a CSTR.

The DPF model may be characterized by only one dimensionless number which is called the Peclet number (*Pe*) and which corresponds to the inverse of a dimensionless dispersion coefficient (*D*):

$$Pe = \frac{vL}{D_L} = \frac{1}{D} \qquad \text{(Eq. 27)}$$

Where, v = interstitial water velocity [m/d]
L = length [m]
D_L = axial dispersion [m²/d]

When analysing RTDs, the dimensionless variance allows us to compare DPF and TIS models. A simple solution approach may also be used to correlate N and Pe (Thonart, 2006):

$$Pe = 2(N-1) \quad \text{for Pe} \le 30 \qquad \text{(Eq. 28)}$$

$$Pe = 2N \quad \text{for Pe} > 30 \qquad \text{(Eq. 29)}$$

The main problem with the application of DM to wetlands is related to the assumptions implicit in the model. The condition of an intermediate amount of axial dispersion is nominally taken to be $D/uL > 0.025$ (Levenspiel, 1972) which correspond to about 20 TIS (Kadlec and Wallace, 2009). On average, surface flow systems are not within the acceptable mixing range, but some subsurface horizontal flow can be within this range. This as literature reports values of N= 4.1 ±0.4 (mean ± SE) for SF and N= 11.0 ± 1.2 for SF systems (Kadlec and Wallace, 2009).

4. Non Ideal Flow Reactors: Multi Flow Dispersion Model (MFDM)

The mathematical description of the multi flow dispersion model was once applied for TW by Maloszewski *et al.*, 2006, considering a multi layered media. The theory presented here below is issued from Leibundgut *et al.*, 2006 and Maloszewski *et al.*, 2006. This model assumes that the tracer transport through the cell can be approximated by a combination of 1-D dispersion equations. They are here corresponding to our variable depths. Each flow path is characterised by a specific volume rate, mean transit time and dispersivity (dispersion parameter). It is assumed that the volume of immobile water within the wetland cell is insignificant; that there are no interactions between flow paths and that the mass of tracer is injected is divided into several flow paths proportionally to the volumetric flow rate. The transport of the ideal tracer along the *i*th flow path is depicted by Figures 21 and 22 and mathematically described by the 1-D equation 30.

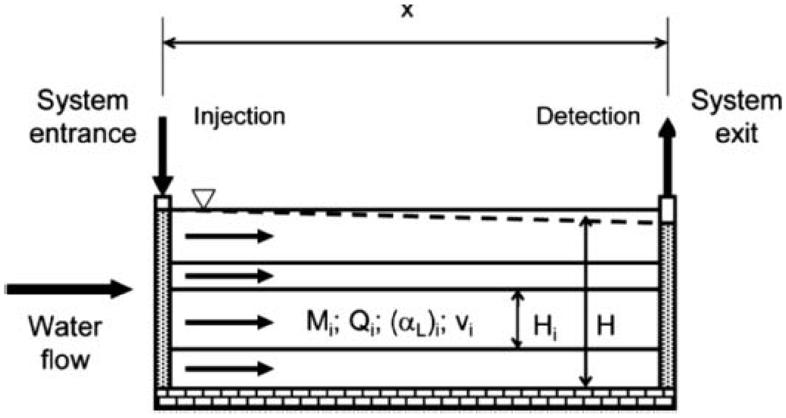


Figure 21: Schematic presentation of tracer transport in a multi flow dispersion model (MFDM) (Leibundgut *et al*, 2009)

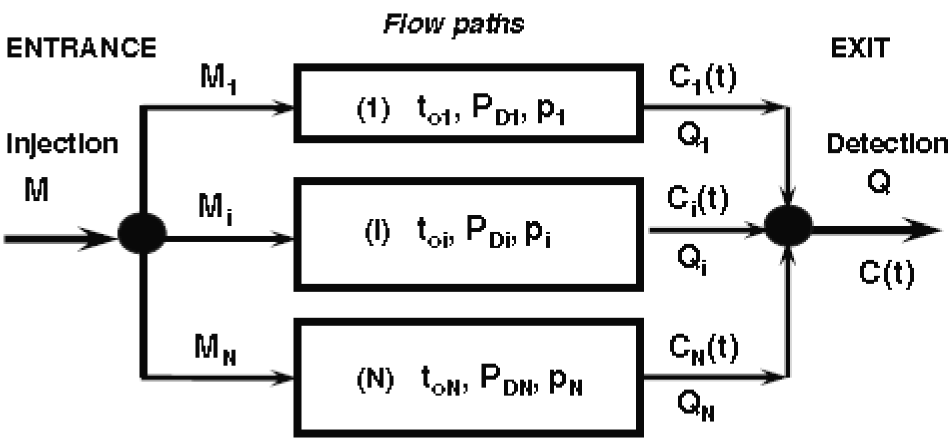


Figure 22 : Conceptual presentation of the multi flow dispersion model (MFDM)

$$\alpha_{Li} v_i \frac{\partial^2 C_i}{\partial x^2} + v_i \frac{\partial C_i}{\partial x} = \frac{\partial C_i}{\partial t} \quad \text{(Eq. 30)}$$

Where, $C_{(i)}t$ = concentration of tracer in the effluent of the *i*th path, mg/l
α_{Li} = longitudinal dispersivity, m
v_i = mean water velocity for the *i*th path, m/d
x = length of the wetland cell, or flow path, m
t = time after injection, d

for instantaneous injection of tracer, the solution is given by :

$$C_i(t) = \frac{M_i}{Q_i t_{0i} \sqrt{4\pi (P_D)_i (t/t_{0i})^3}} \exp\left[-\frac{(1 - t/t_{0i})^2}{4 (P_D)_i (t/t_{0i})} \right] \quad \text{(Eq. 31)}$$

Where, Q_i = volumetric flow rate, m^3/d along the *i*th flow paths,
M_i = mass of the tracer transported along the *i*th flow paths,

$$(P_D)_i = \alpha_{Li} / x_i \quad \text{(Eq. 32)}$$

Is the dispersion parameter, where x_i is the length of the *i*th flow path.

$$t_{0i} = \frac{x_i}{v_i} = \frac{(V_m)_i}{Q_i} \quad \text{(Eq. 33)}$$

Is the mean transit time of the volume of mobile water in the *i*th flow path.

The model assumes that the whole injected tracer mass, *M*, is divided into *N* portions, which enter the *N* flow paths proportional to the volumetric flow rates Q_i :

$$M_i / Q_i = M / Q \quad \text{(Eq. 34)}$$

Where, Q = total flow rate into the system, that is the sum of partial flow rates,

$$Q = \sum_{i=1}^{N} Q_i \quad \text{(Eq. 35)}$$

The outflow tracer concentration C(t) is the flux weighted mean concentration for all flow paths observed in the outflow at the outlet of the system:

$$C(t) = \sum_{i=1}^{N} p_i C_i(t) \quad \text{(Eq. 36)}$$

$$p_i = Q_i / Q = M_i / M = R_i / R = \frac{\int_0^\infty C_i(t)dt}{\int_0^\infty C(t)dt} \quad \text{(Eq. 37)}$$

Where, p_i = fraction of water flux in the *i*th path

R_i = partial tracer recovery observed in the outflow of the *i*th path,
R = total tracer recovery measured in the outflow

The equations Eq. 36 combined with Eq.31 and Eq. 37 are called the multi flow dispersion model, MFDM. The fitting parameters of MFDM are:

- T_{0i}, the mean transit time,
- $(P_D)_I$, the dispersion parameter,
- p_i, the fraction of water flux for each flow path *(i),*
- N, the total number of flow paths.

Due to the high number of unknowns, the calibration of the model with experimental data cannot be done in a straightforward way. The fitting procedure is performed stepwise by fitting one by one the partial TBC's and by subtracting fitted (partial) curves from the total TBC beginning with the earliest peak. The number of flow path N, is then found automatically. After determining all fitting parameters the partial volumetric flow rate (Q_i) and the volume of water in each flow path $(V_m)_i$ can be calculated from Eq. 33 and Eq.37 as : $Q_i = p_i * Q$ and $(V_m)_i = Q_i * t_{0i}$, assuming that the whole water is mobile.
Then the total volume of water in the system is given by :

$$V_m = \sum_{i=1}^{n} V(m)_i \qquad \text{(Eq. 38)}$$

The MFDM assumes that the whole water in the system is mobile, so the total volume of subsurface water (V) is equal to V_m and the water content (ε) in % can be estimated form Eq.38 and the volume of the wetland filtering medium (V_w) as follows:

$$\varepsilon = \frac{V}{V_w} 100\% \qquad \text{(Eq. 39)}$$

And the mean transit time of water through the whole system is given by:

$$t_0 = \frac{V_m}{Q} = \sum_{i=1}^{N} p_i t_{0i} \qquad \text{(Eq. 40)}$$

For the multi layered porous medium, assuming that porosities in each layer are similar and equal to n, and the hydraulic gradient is known, the hydraulic conductivity for each layer (k_i) and the weight mean ($\bar{k}$) can be simply calculated by applying Darcy's law:

$$\bar{k} = \frac{x^2 n}{(\Delta H) t_{0i}} \qquad \text{(Eq. 41)}$$

$$k_i = \frac{x^2 n}{(\Delta H) t_{0i}} \qquad \text{(Eq. 42)}$$

Where, $\Delta H / x$ = hydraulic gradient between the injection and detection wells.

Finally, knowing the hydraulic conductivity for each layer (k_i) the mean thickness of the layer (Hi) can be estimated, combining Eq.42 and Eq. 37:

$$H_i = \frac{p_i H \overline{k}}{k} = p_i H \frac{t_{0i}}{t_0} \qquad \text{(Eq. 43)}$$

Summarising, in the multi porous layered medium, the tracer experiment enables the estimation of transport parameters and hydraulic properties of the individual layers. However, the relative position of the layers in the system remains unknown.
When the system has a homogenous medium and parameters of each flow paths are the same, the system can be approximated by one flow path, and the solution of equation 31 is directly applicable without indexing (i) what corresponds to the dispersion model (DM).

In our case, the dispersion model (DM) is applied to exit tracer data from outlet. DM model provides us mean parameters for dispersion and flow velocity.
Tracer tests made on the 3 depths along length allow us to apply the MFDM inside the black box of the HF bed. The number of layer (N) is assumed to be the three monitored layers and their mean transit time is measured, what reduces the fitting parameters of the model from 4 to 2. This allows the retrofitting of data into the model to identify the dispersion parameters and individual flow velocities with more accuracy than the general ones found by the simplified dispersion model.

II.2.5 Estimation of the transport parameters

The two presented methods of parameters estimation are: the method of moments (MM) and the Least Squared Method (LSQM).

Method of Moments (MM)
The method of moments is a parameter estimation method which was based on computational tools available when the method was developed more than 35 years ago.
It computes the first and second moments of the experimental outlet concentration distribution via numerical integration, and is related to tracer detention time and number of TIS.

RTD is a distribution and it is possible to calculate the different central moments which define the key parameters that are actual detention time and dispersion of a pulse due to mixing (Edeline, 1998; Kadlec and Knight, 1996). The *nth* moment is defined as follow:

$$M_n = \int_0^\infty t^n F(t)\,dt \qquad \text{(Eq. 44)}$$

The zeroth moment corresponds to the sum of all F(t)Δt fractions which, by definition, is equal to unity.
However, if the denominator is assumed to be the mass of tracer injected and the numerator, the outlet mass flow of tracer, the zero moment becomes a mass balance check on recovery of tracer:

$$\text{Recovery}\,(\%) = \frac{\int_0^\infty QC(t)\,dt}{\text{mass of tracer injected}} \times 100\% \qquad \text{(Eq. 45)}$$

The recovery of tracer must be as close as possible to 100%. A recovery which is inferior to 100% may indicate that the tracer adsorbed, was degraded or leaked during passage through the wetland. A low recovery may also mean that recordings were prematurely stopped.

The *first moment* represents the average time spent by a particle in the wetland which is nothing else than the actual tracer residence time (τ, in days). This value defines the centroid of the exit tracer concentration distribution:

$$M_1 = \int_0^\infty tF(t)dt = \tau \qquad \text{(Eq. 46)}$$

The dimensionless time variable is defined as the actual time divided by the tracer detention time :

$$\theta = t / \tau \qquad \text{(Eq. 47)}$$

The *second moment* corresponds to σ^2 (in days^2) which is the variance of the distribution and is calculated from the actual residence time. It characterizes the spread of the tracer response curve about the mean of the distribution. Thus, t^2 must be replaced by $(t-\tau)^2$ to characterize the dispersion of the tracer response curve about the mean of the distribution (Edeline, 1998; Kadlec and Knight, 1996):

$$M_2 = \int_0^\infty (t-\tau)^2 F(t)dt = \sigma^2 \qquad \text{(Eq. 48)}$$

This measure of dispersion may be rendered dimensionless by dividing by τ^2 (Kadlec and Knight, 1996). Actually, the ratio σ/τ defines the coefficient of variation (CV) which is a comparative dimensionless measure of the dispersion of probability distributions (Edeline, 1998). The variance of the RTD is created by mixing of water during passage, or equivalently by a distribution of velocities of passage. This can be lateral, longitudinal or vertical mixing. The variance-to-square-mean ratio is called the dimensionless variance (σ_θ^2) of the tracer pulse:

$$\sigma_\theta{}^2 = \frac{\sigma^2}{\tau^2} \qquad \text{(Eq. 49)}$$

This new parameter (σ_θ^2) has an important relationship with the number (N) of equivalent tanks in series in a TIS model and with the Peclet number (Pe) of a Dispersed Plug Flow (DPF) model (Eq 50). For purposes of parameterisation, it is noted that for the TIS model or gamma RTD distributions:

$$\sigma_\theta^2 = \frac{1}{N} = \frac{\tau - \tau_{peak}}{\tau} \qquad \text{(Eq. 50)}$$

A serious failing of MM is that it emphasised the tail much more than the central peak central area portion.

Least Squares Method (LSQM)
The least squares method estimates the transport parameters by fitting the appropriate theoretical solution to experimental data concentration measurements using a trial and error procedure. The fitting procedure assumes that the solution to the inverse problem, the best fit, is obtained when the values of parameters yield the minimum value of the sum of the squared difference between the theoretical and observed concentrations.
The method is also named Sum of the Squared Errors, SSQE.
This method can be performed on PC using e.g. the SOLVER routine application of Excel™.
The gamma distribution of the TIS model (Eq.19) can be simultaneously solved for parameters N and τ, which minimize the difference between the observed and the predicted RTD.
In the case of Dispersion model (DM), the parameters which are simultaneously adjusted are the P_D, dispersion parameter and τ, and in the multi flow dispersion model, the adjusted parameters will be T_{0i}, the mean transit time, $(P_D)_I$, the dispersion parameter, p_i, the fraction of water flux for each flow path *(i),* and N, the total number of flow paths.

II.2.6 Choice of tracer

The water movement within the horizontal flow beds is investigated by tracer studies. The appropriate choice of the tracer is an important factor in order to achieve good and meaningful results.
The following theoretical tracer description is taken out of the "tracers in hydrology" book, written by Leibundgut *et al.*, 2009.
"Tracers are first divided into two groups : natural or environmental tracers and artificial tracers. *Environmental tracers* are defined as inherent components of the water cycle. Sometimes, accidental injections can be used for hydrological studies. Global input functions have been created as the side effects of industrial or military activities (CFCs, 85Krypton or bomb-tritium).
Environmental tracers are:

- stable isotopes : Deuterium (^2H), Oxygen-18 (^{18}O), Carbon-13 (^{13}C), Nitrogen-15 (^{15}N,) Sulphur-34 (^{34}S),
- radioactives isotopes : Tritium (^3H) (and Helium-3 (^3He), Carbon-14 (^{14}C), Argon-39 (^{39}Ar), Krypton-85 (^{85}Kr), Radon-222 (^{222}Rn), Silicium-32 (^{32}Si), Chlorine-36 (^{36}Cl),
- noble gases,
- geochemical compounds (silicates, heavy metals, chloride) and,
- the regular physio-chemical parameters Electrical conductivity and Temperature.
- They also include the pollution tracers (i.e. nitrate, boron, phosphate, CFCs).

Artificial tracers are defined by their active injection into the hydrologic system in the context of an experiment. Artificial tracers are substances that offer additional information of value in the investigation of hydrological systems and subsystems. The scales of application are limited in both time and space. In general, artificial tracers are used in systems which have a residence time smaller than one year. Typical fields of applications are: the detection of hydrological connections, flow paths and flow directions in catchments and aquifers, delineation of catchments and aquifers (qualitative), determination of flow velocities and further aquifer flow parameters based on the tracer breakthrough curves, hydrodynamic dispersion, runoff separation, residence time, infiltration and runoff generation processes, convection diffusion processes in surface water, simulation of contaminant transport and discharge measurement applying dilution methods.

Artificial tracers are:

- soluted tracers, as fluorescent tracers Naphtionate, Pyranine, Uranine, Eosine, Rhodamine,
  non fluorescent dyes (e.g. brillant blue)
  Salts Sodium/Potassium Chloride, Sodium/Potassium Bromide, Lithium chloride, Potasium iodide, Sodium borate (Borax)
  Fluorobenzoic acids
  Deuterated Water (^2H)
  Radionucleides (e.g. Tritium, Bromide-82, ^{82}Br)
- dissolved gases tracers (Helium, Neon, Krypton, Sulfur Hexafluoride, SF_6) or
- particulates tracers – drifting particles : Lycopodium spores, viruses, bacteria, phages, DNA, phytoplankton, Synthetic microspheres.

All tracers carry discernable and preferably unique information. These two properties – carrying information that can be identified most effectively at low concentrations – categorize substances as trace elements.

Each tracer application is individual and many things have to be taken into consideration. In addition to the most important property which is conservativity, there are certain basic requirements that a tracer should meet. The requirements of solute tracers in general and of artificial tracers in particular, are listed in table 3.

Table 3 : Required properties of artificial tracers in general (Leibundgut *et al*, 2009).

Properties to be considered	Requirement of an ideal (conservative) tracer
1. Conservativity	High
2. Solubility in water	High
3. Fluorescence intensity	High
4. Detection limit	Low
5.pH dependencies	Low
6. Temperature dependencies	Low
7. Photolytic stability	High
8. Sorption processes	Negligible
9. Chemical and biological stability	High
10. Toxicity and related environmental effects	None or minimal
11. Costs and other aspects	Low or moderate

Conservativity: the objective of the application of tracers in hydrology is the investigation of water in all its various guises, behaviours and characteristics within the different media and substrates represented in the water cycle. Consequently, conservative tracers representing the flow of water are required. Conservativity exists if the tracer is physico-biochemically stable (nonreactive in natural water) and not sorptive.

The *solubility* of tracers in water is a crucial requirement of tracers used to investigate water flows in the hydrological cycle as the tracer should be as close to the characteristics of water as possible. Their solubility depends on both the temperature and the pH of the water.

Fluorescence is a luminescence that occurs where energy is supplied by electromagnetic radiation. The substances used for tracing purposes are situated within the small range of visible light between the higher ultraviolet and the infrared wavelengths (ca. 350–750 nm). The maxima of the spectra of fluorescent tracers are characteristic and constant. Having relatively small fluorescence ranges, tracers are well suited for application as hydrological tracers. The fluorescence intensity of a trace substance depends on its physical properties, namely quantum yield, extinction coefficient and tracer concentration.

The dependence of fluorescence not only upon pH, temperature and light, but also upon chemical and microbiological effects (metabolism), can result in the degradation of the fluorescence in water samples and in natural water bodies. Consequently, when performing experiments using fluorescent tracer careful consideration of these aspects is required in both the planning of these experiments and in the analysis.

The *detection limit* is clearly not the same for the different measurement devices. When analysing surface water samples, the background is relevant. The higher the backgrounds signal the lower the detection limit.

Variations in the pH value of the traced water have a twofold impact on fluorescent tracers: (i) on the analysis and (ii) on the degree of sorption affinity of the tracer.

The *sorption* affinity of the tracers also depends on the pH value. As was mentioned above, changing pH values have the potential to change the electrical charge of the molecule. This leads to the sorption effect of pH dependence. The higher the pH, the lower is the sorption.
Fluorescent tracers are mostly organic molecules with various functional groups attached to the molecular kernel. In addition, the functional groups protonate and deprotonate depending on pH, thereby changing the net charge of the molecule (Flury and Wai, 2003). Consequently, certain tracers tend to be absorbed in different proportions within acidic media and substrates. In the case of experiments to be carried out in such media, it is necessary to consider carefully whether it is possible to use fluorescent tracers or not.

Photolytic stability: unlike pH dependence, exposure to light has an irreversible effect on fluorescence. Therefore, the use of fluorescence in the study of surface waters is feasible only to a limited extent. In soil and groundwater experiments photolytic decay is usually not a problem.

Sorption behaviour is the most important property relevant to the use of artificial tracers generally, and fluorescent tracers in particular. Sorption is a crucial process in the performance of experiments in the saturated and unsaturated zones. A first indication of the sorption behaviour of a tracer is provided by the solubility. The higher the solubility of a tracer substance, the lower is its sorption.

Chemical stability: dye tracers may be readily quenched and/or decomposed as a consequence of oxidation and other chemical changes. However, oxidative processes affect the dye tracers to different degrees. Whereas the Rhodamines are more resistant, other dyes will be irreversibly quenched.

Environmental effects: each injection of artificial tracer in a hydrological system is in a sense a contamination of the water body in question. However, carefully planned and correctly prepared tracer experiments generally involve only minimal quantities of tracer substances in the range of grams, or kilograms at most. Therefore, the 'contamination' is usually tolerable. When preparing a tracer experiment it is vitally important that national regulations pertaining to tracer experiments are consulted. In our case of interesting tracers for wetlands, the Rhodamine group is suspected to be toxic.

Of *practical interest* is the price of tracer substances, which is not negligible. The other intesresting aspect is to be relatively easy and inexpensive to analyse (Taylor *et al*., 1990, Whitmer *et al*., 2000).

Salt as and fluorescent tracers are popular among tracer hydrologists because of their relatively easy handling, the seemingly simple analysis, the high sensitivity of the analysis, the low detection limit and, consequently, the small quantity of tracer needed in field experiments. Fluorescent tracers are also attractive because of the linearity of the calibration curve in the measuring scale, and their toxicity levels are very low compared to other tracer substances; some are entirely nontoxic. Fluorescence tracers are the most important and most often applied, followed by the salt and the advanced tracers. Drifting particles are tracers of a different physiochemical origin, and are used to assess special problems, such as the filtration capacity of unsaturated zones. For studies in constructed wetlands, salt and fluorescent tracers are the most convenient and widespread ones used (Schmid *et al*., 2004). Other specific fields of application of salt tracers are tracer tests in the saturated and the unsaturated zone (Singha and Gorelick, 2005).

They are both briefly presented here below.
Dyes have advantages of low detection limits, zero natural background, and low relative cost. However, they are susceptible to both photodegradation and biodegradation (Dierberg and DeBusk, 2005). One of the most suitable dyes is rhodamine WT, because it exhibits the fewest matrix artefacts. Dissolved solids have no effect below about 600 mg/L and pH has no effect above pH=6, although the level of fluorescence is temperature sensitive, changing about 2% per degree Celsius (Smart, Laidlaw 1977). Rhodamine WT is, however susceptible to biodegradation, photolysis and adsorption onto organic solids, detritus and some pIastics (Smart and Laidlaw 1977; Lin *et al*., 2003; Dierburg and DeBusk, 2005). The use of a sorbing tracer can distort the tracer response curve and lead to errors in calculating hydraulic characteristics. An irreversibly sorbing tracer like rhodamine WT may cause the peak time to be shorter than it really is, while a reversibly sorbing tracer will cause a flattening of the RTD and an unrepresentative extension of the tail. Due to the above characteristics, it is recommended that rhodamine WT is only applied at moderate to high initial concentrations, in short term tests (nHRT less than about one week) and within environments that are not highly organic. Samples should be collected in glass bottles and kept in the dark prior to analysis to prevent photo-degradation (Headley and Kadlec, 2007).

Salts are inorganic compounds, which break up into cations and anions when dissolved in water. Ionic compounds in the solid salt form have a high melting point, a brittle consistency and are highly soluble in polar solvents. In solution, and when melted, their electrical conductivity is high due to the mobile ions. Their volatility is low due to strong binding energy (ionic bridges) in the ionic grid. The factors favouring the use of salt tracers are easy handling, availability and the potential for continuous recording. There are also, however, several attributes complicating the application of salt tracers. When using salts as tracers one is confronted with the following potential issues: high natural background and pollution, high detection limits, large injection masses, transport problems, relatively laborious analyses and the problem of sorption and ion exchange.

In natural hydrological systems Br^- usually occurs at a very low background concentration, usually below the detection limit. For this reason, and also because of its high solubility (ca. 850 g/l at 10 ◦C), it is easier to handle in the field than other salts. However, bromide also requires a high injection mass, due to its lower detention sensitivity. Bromide is assumed to be chemically, biologically and photolytically stable, and due to its negative charge sorption is very low in mineral soils, so it can act mostly as an ideal, or nearly ideal, conservative tracer" (Leibundgut *et al*., 2009).

In the treatment wetland field, bromide and lithium are the most extensively used ionic tracers, mainly due to their relatively low cost and ease of analysis. They are typically added as solutions of sodium or potassium bromide, or lithium chloride and have yielded reliable results in numerous wetland studies (Bowmer, 1987; Netter, 1994; Tanner and Sukias, 1997; King *et al*., 1997; Drizo *et al.*, 1999; Rash and Liehr, 1999; Grismer *et al*., 2001; Lin *et al*. 2003; Garcia *et al*., 2004a; Schmidt *et al*., 2004). Bromide may be present in natural waters at concentrations well above detection limits. Although relatively inexpensive, very large quantities of ionic tracers can be required to achieve a significant peak and detection above background levels in large wetland systems, thereby making them most suitable for small to moderate sized wetlands. Bromide is typically analysed through ion chromatography, although less reliable portable probes are available and can be used in the field (Headley and Kadlec, 2007).

II.3 EXPERIMENTAL METHOD

II.3.1 Material and methods – site in operation of Nassogne experiments

The general features of the horizontal flow system of Nassogne are described in Chapter I.
For the tracer tests, one cell was isolated from the system (including in total 2 cells in parallel), while the wastewater inflow from the guesthouse was diverted to the second cell. The cell isolated for the tracer test was loaded with clear water for 3 days before starting the experiment. The clear water was pumped from a natural well located 200 m away from the HF TW.
For the experiment, the expected inflow was 3m^3/day of clear water, measured by a tipping bucket equipped with a mechanical counter device. The outflow was measured by a floating pump equipped with a flow meter. The velocity and concentration of the tracer inside the TW was followed by a network of 9 sampling ports, set at 3 m, 6 and 11 meters from inlet and sampling at 15, 35 and 55 cm depth, all replicated as two transects down the length of the bed. Transect at 3 m, 6 m and 11 m from inlet are respectively named Line 1 (L1), Line 2 (L2) and Line 3 (L3). The collecting systems are small piezometers (permanently installed): these consist of small PVC pipes, such as those used for sanitary conduits, of 2 to 3 cm of diameter. One tip section is perforated with holes on its periphery and closed with an end plug, driven into the gravels of the HF TW. The outlet pipe is the tenth followed point. Sampling was done by automated samplers every hour and they mixed corresponding sample points on the two transects within one sample. They have an automated purging system before collecting samples.
The tracer used was Potassium Bromide (KBr) with a concentration of 1g Br-/L to avoid tracer stratification. 200 g of Bromide were injected into the 8 inlet distribution pipes through 6 jerry cans of 33 Litres each. The jerry cans were filled with water from the well one day before the test and let onsite to avoid temperature stratification when the tracer was injected. The injection lasted 15 min and is considered as single-shot injection as the injection period was less than 2% of the total residence time. Samples were taken before starting the experiment to assess the background conductivity. During the tests, the HF TW was covered with tarps to minimize any rainwater intrusion into the system.

The theoretical detention time was expected to be 4 days based on a targeted inflow of 3000 L/day, however the influent tipping bucket measured an average inflow of 2300 L/day. Problems were also encountered with the automated samplers from the first line (L1) at 55 cm depth and at the outlet.
It was thus decided to replicate the test a second time. The delivery inflow reached 2690 L/day and as it was recommend to monitor such experiments for five time the theoretical HRT, the test was performed over 20 days. Unfortunately, the automated sampler from line 1 at 55 cm depth completely collapsed and gave no valid data.

Test 1 was performed from 25 February till 7 March 2009 and test 2 was performed from 9 to 29 March 2009.

Another original aspects of this work as it is the first time amongst any known literature that such large scale tests, on an operational site, was ever performed within a HF TW system and replicated twice in a row.

Figure 23 is a schematic of the sampling layout for the tests and Figure 24 presents some photos of the field tracer experiment.

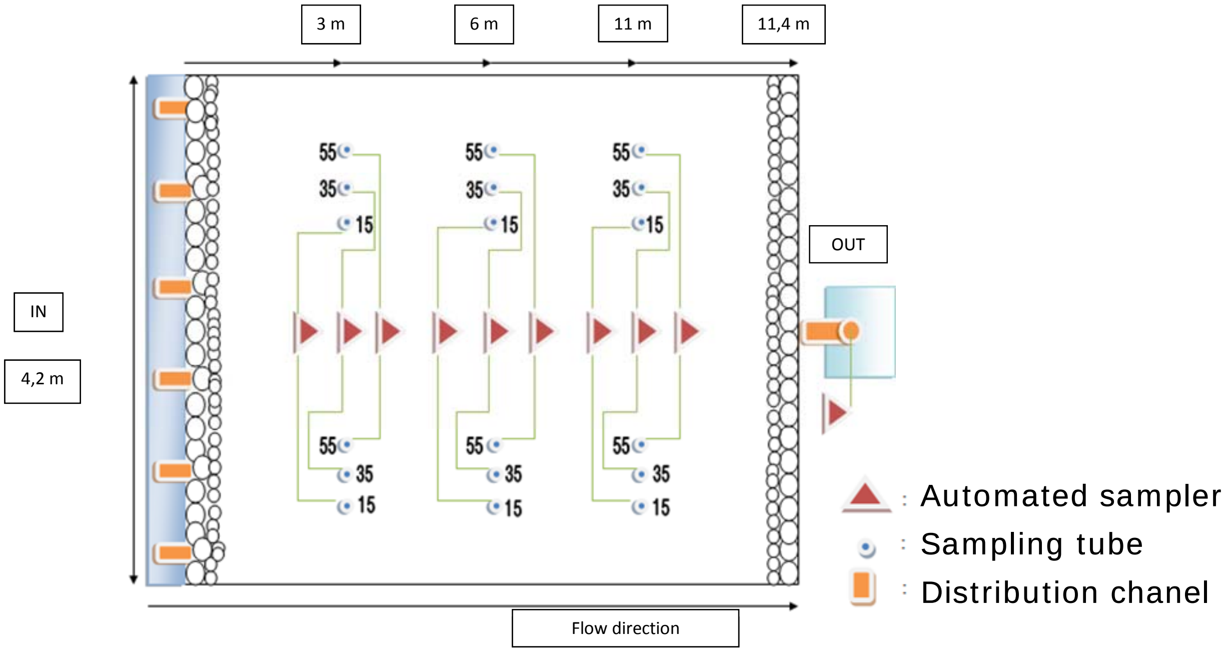


Figure 23 : Sampling layout of the automated samplers. Each sampler mixed two samples from the same depth points, located on the same transect over width

Figure 24 : Photos of the sampling layout with (left) the 3 automated samplers set on lines 1, 2 and 3 to collect the 3 investigated depths over length; (right) detail of one sampler mixing two points from the same depth located on the same transect line over length

II.3.2 Material and methods – greenhouse experiments

The general description of the four pilot wetland cells is detailed in Chapter I.
For the tracer tests, two head tanks of 100 L each, load the pilots with drinking water from the public network through a Watson-Marlow 205S peristaltic pump, equipped with 8 marprene manifold tubes of 2,79 mm in diameter. Each tube delivers a flow rate of 20 L/d at 57 rpm. A pair of tubes is used for a flow rate of 40 L/d per pilot. Tubes deliver the water sub-surface within the 15 first cm of larger gravels of the inlet distribution zone of each pilot wetland cell.

Potassium bromide (KBr) was used as the tracer. One litre was injected through the tubes of the peristaltic pump at the concentration of 1,49 g/L KBr (1 g Br^-/L). Single-shot injections lasted 35 min and may be considered as instantaneous impulses as they lasted less than 2% of the nominal residence time. Background conductivity was assessed during the two days prior to the test start.
Samples were taken manually every hour. Outflow was collected in buckets at the outlet end of the pilots. The water volume was measured and a sample was taken for analyses in the laboratory. Samples required preparation as on-line measurement can not be done for Bromide.
Tests have been performed for duration of five times the nominal hydraulic retention time, and repeated twice.

II.3.3 Bromide content measurement

The measuring instrument was a WTW Br 800 bromide combination electrode connected to a WTW pH/Ion 340i ion meter. The conductivity measuring range of the Br 800 bromide combination electrode ranges from 0,4 mg/L to 79 000 mg/L, its temperature range is from 0 to 80°C (up to 100°C for a short time). Sample preparation consisted of adding 2% of a conditioning solution (ISA, Ionic Strength Adjustment) to the sample. This solution provides a constant ionic strength and similar diffusion potentials as the reference electrode in standard and test samples. In order to prevent deviation, the probe was calibrated every day.

II.4 RESULTS AND DISCUSSION

The first part is presenting data and preliminary graphs and tables; the second part is applying mathematical models; their solutions provide parameters of the system which are discussed.

II.4.1 Data for the site in operation of Nassogne

II.4.1.1 Presentation of TBC results

The graphs on following pages are firstly presenting results of measurements through classical Tracer Breakthrough Curves (TBC) of the measured concentration, with tracer concentration versus time for the two performed tests. The first set of graphs presents TBC per depth over length for the 3 investigated lengths (L1 to L3); and the second set of graphs is reversely showing TBC per line for the 3 investigated depths (-15 to -55 cm deep). Figure 25 and 26 are related to the first test (Test 1), figures 27 and 28 to the second test (Test 2). The duration time for the first test was 10 days, and 20 days for the second tests, which is marked on X axes; the Y axes present various scales in order to present the most relevant TBC at first sight. A common Y axes to all graphs would have lead to very low and flat curves for some of them; and harmonised presentation will be done through the results discussion and interpretation within the next section.
Selected colour code is blue for line 1, green for line 2 and red for line 3, with increasing colours from pale blue, pale green and pink for depths close to surface (-15 cm) on lines 1, 2 and 3 respectively to dark blue, dark green and dark red for depths closer to the bottom of the bed (-55 cm) on the same respective lines.

II.4.1.2 Comments about TBC results

First observation of those TBC is providing the following comments:

- all obtained curves have a general regular bell shape, corresponding to theoretical expectations;
- replication with Test 1 and Test 2 show very different results, even with the closest experimental conditions that field experiments can provide;
- for both Tests 1 and 2, TBC at depth -15 cm (figures 25a and 27a) are presenting a tracer peak decreasing in concentration over length, providing a flattening of the bell shape, likely due to mixing or dispersion conditions;
- Reversely, TBC curves for Test 1 and Test 2 are presenting opposite effects for depth -35 cm, between figure 25b and 27b and depth -55 cm, between figures 25c and 27c. During the first test, the tracer peak concentration presents a high increase on line 3 (11 m from inlet), while the tracer concentration is decreasing over length for test 2;
- Small second peaks are observed around days 7 and 8 for both Tests 1 and 2 at the depth of 55 cm on lines 2 and 3, showing a small back mixing effect;
- Figures 25 and 27 related to the tests 1 and 2 are providing the following major and relevant information :
    1. Tracer peaks are not reached on the same time as a homogeneous system over depth should do,
    2. Tracer peaks do not the same intensity at all over length and with depth, varying in tracer concentration by a factor up to 100,
    3. Tracer peaks are systematically appearing first in the bottom of the bed at depths -55 cm and -35 cm before the surface depth of -15 cm,
    4. Following the previous remark n°3, a high noticeable difference between the 2 tests is observed, as all tracer peak concentrations are much higher for Test 2 than Test 1.

II.4.1.3 First observations from TBC results on hydraulic patterns

These general good bell shapes of all curves demonstrate that the general behaviour of the system is following a plug flow water movement. Without surprises, the plug flow behaviour is neither ideal, nor perfect as the intensities of peaks are not constant over length but rather decreasing.
The delays in tracer peaks with depth at the same distance from inlet injection point are attesting that the velocity of water within the bed is not homogeneous according to depth layers.

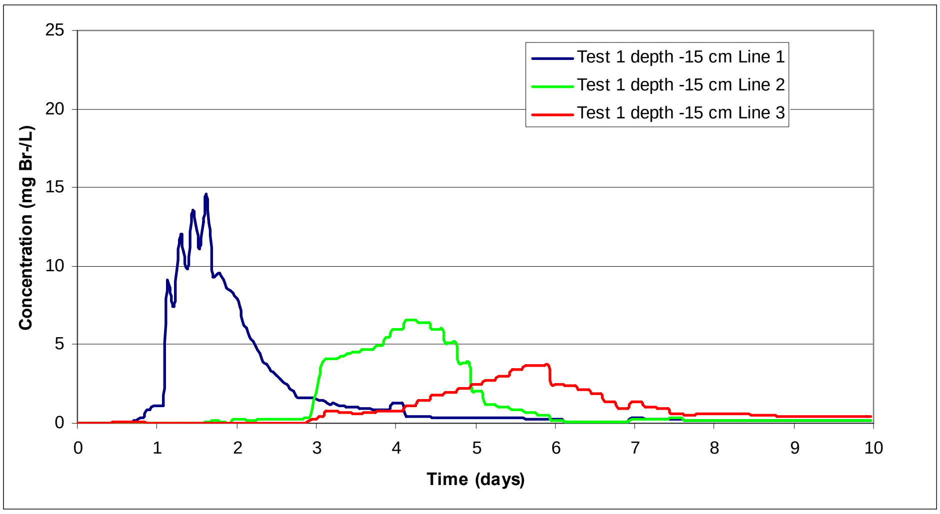
II.4.1.4 Water Budget

As mentioned in the theoretical background section, the water budget has limited parameters in the case of these experiments. As the experiment was covered by tarps to avoid any water added to the system through precipitation, the single inflow (IN) entering the system is the clear water measured by the tipping bucket system with counter. The water exiting the system is the outflow (OUT) measured by the pump equipped with a floating device and counter and the evapotranspiration (ET) of the bed. This summarised by :

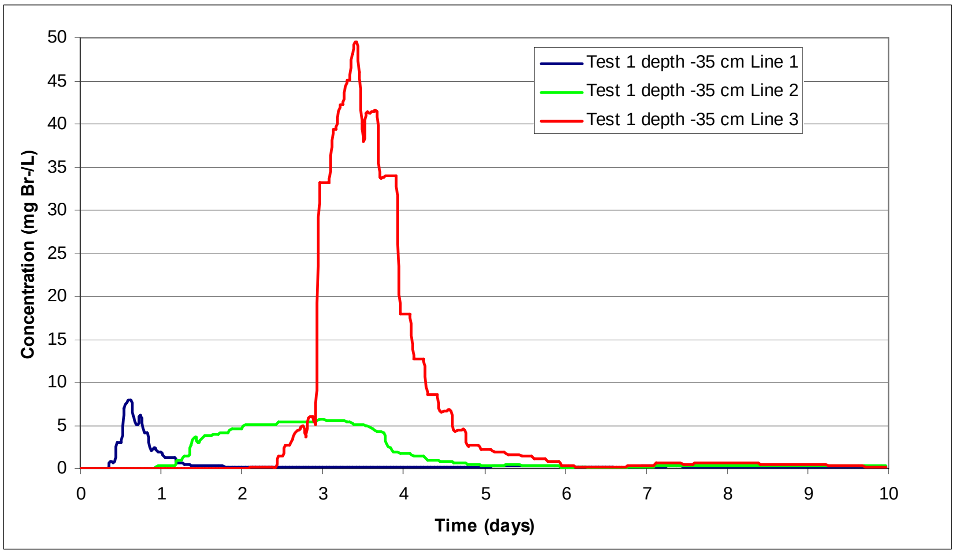
$$IN = ET + OUT$$

Test 1 was performed late February - early March on 2009; the average temperature was 3,6°C. The average inflow delivered over the 10 days of experiment to the system was 2306,68 L/d and the average outflow measured was 2168,28 L/d. The difference is 138,4 L/d with was lost by ET, representing 6 % of the total inflow, or 2,5 mm/d. This value is below the usual range of 4 to 6 mm for gravel beds. It can be explained, by the cold season as the experiment started after one week of snow and, secondly by the harvested reeds for the winter season inducing almost zero evapotranspiration. The experiment was also covered by tarps, which were preventing rainfall water intrusion into the system but allowed gaseous water to evaporate through junction parts of the tarps. The period was cold but sunny, and the isolation induced an increased temperature of the top layers of the bed, and thus loss by water vaporization.

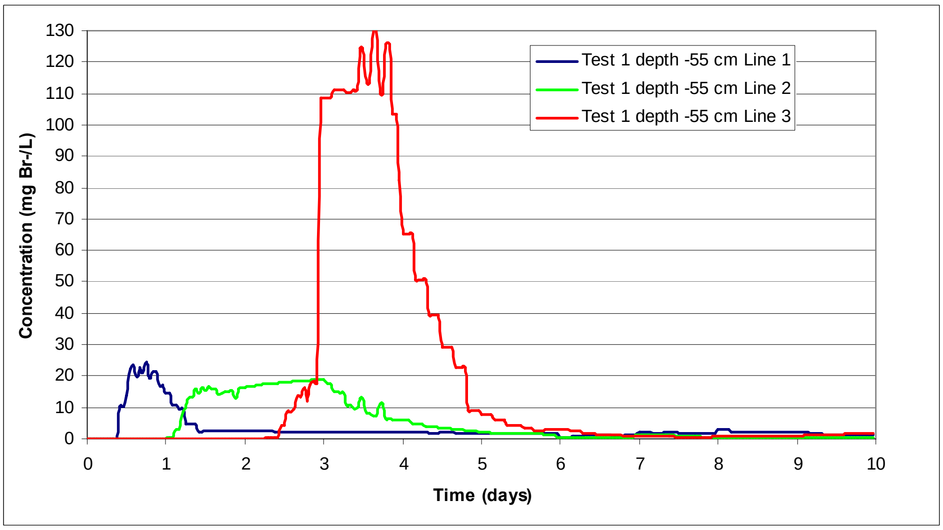
Test 2 was performed on March (9 to 29) over 20 days and the average temperature of the month was 6,7°C. The average inflow delivered to the system was 2690 L/d and the average outflow measured was 2542,5 L/d. The difference is 147,98 L/d which are lost by ET, representing 5,5 % of the total inflow, or 2,6 mm/d. This period was warmer on temperature stand point but also rainier. It has involved less heating of the top layers of the bed below the tarps, inducing a lower ET percentage than during Test 1.



(a)

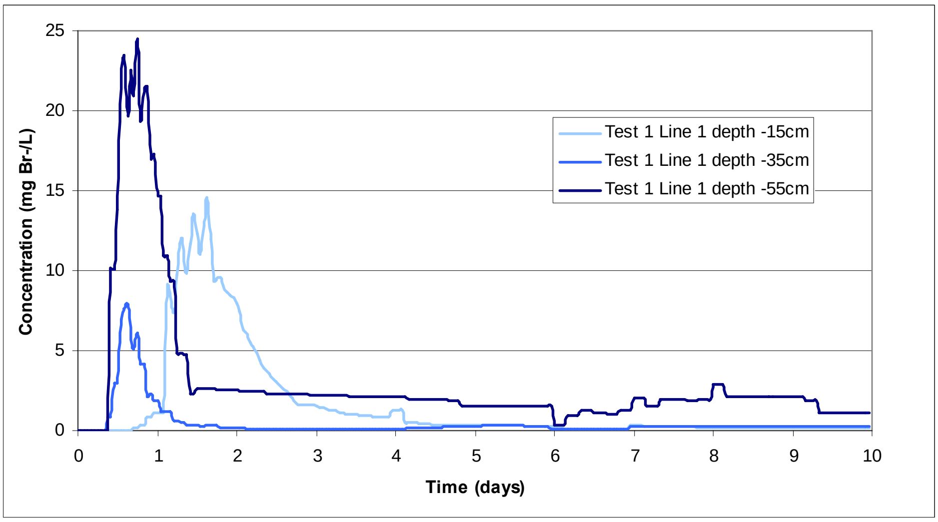


(b)

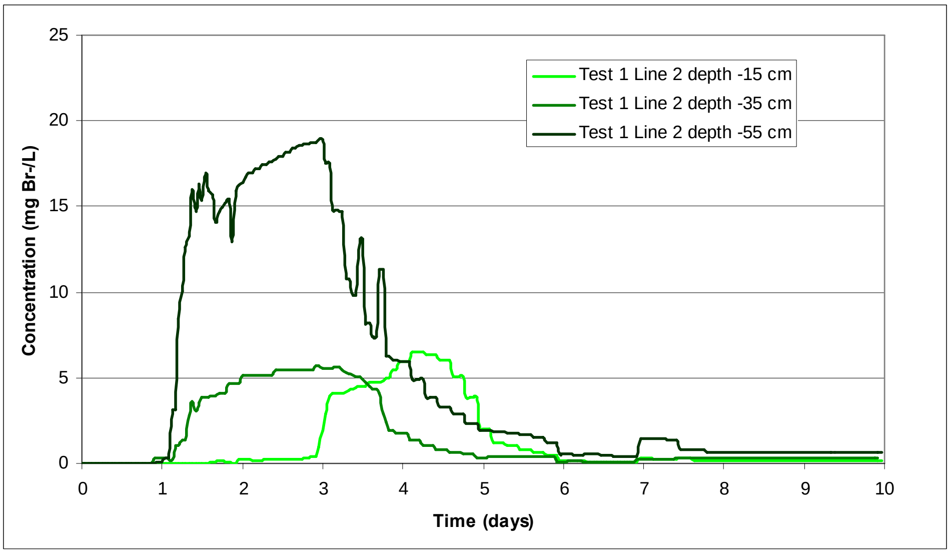


(c)

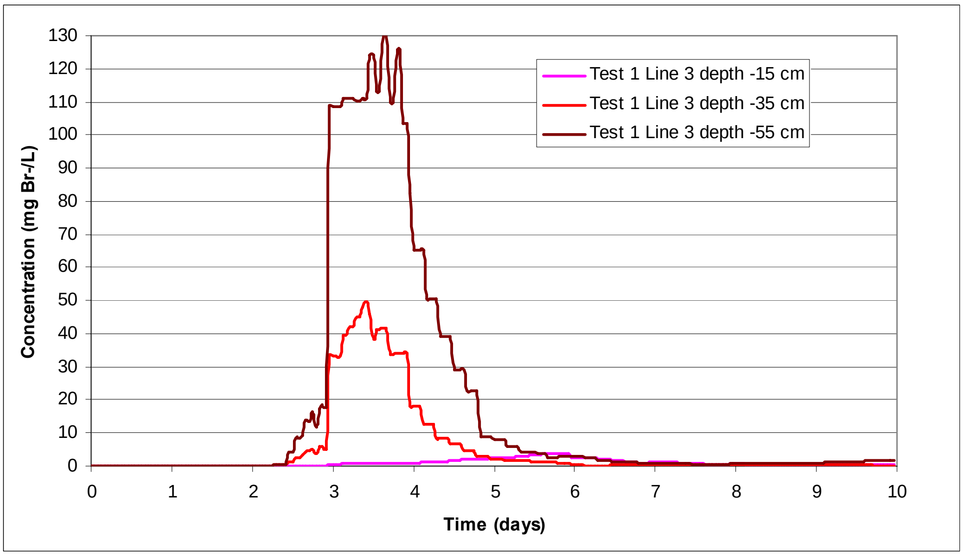
Figure 25 : TBC for test 1 for the 3 investigated depths : -15 cm (a), -35 cm (b) and -55 cm (c) over the 3 lengths, L1 at 3m from inlet, L2 at 6 m and L3 at 11 m from inlet respectively.



(a)

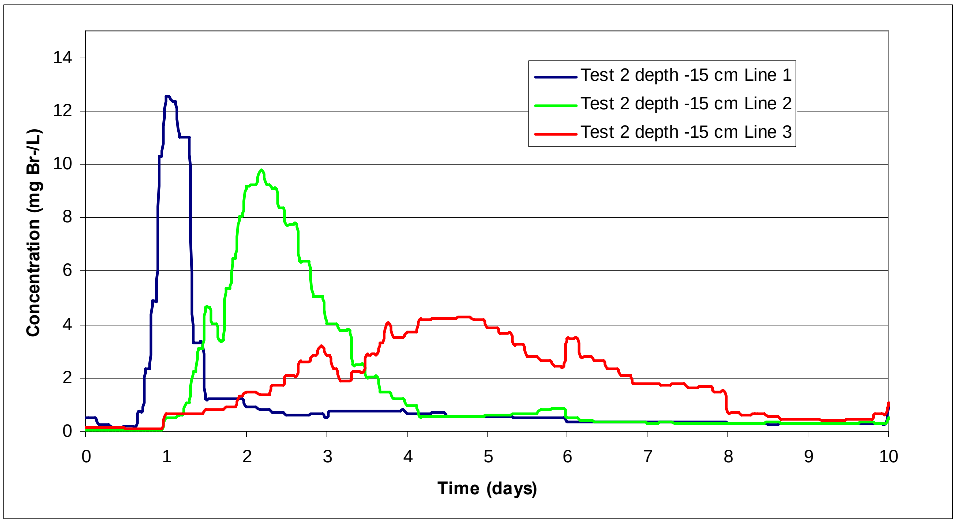


(b)

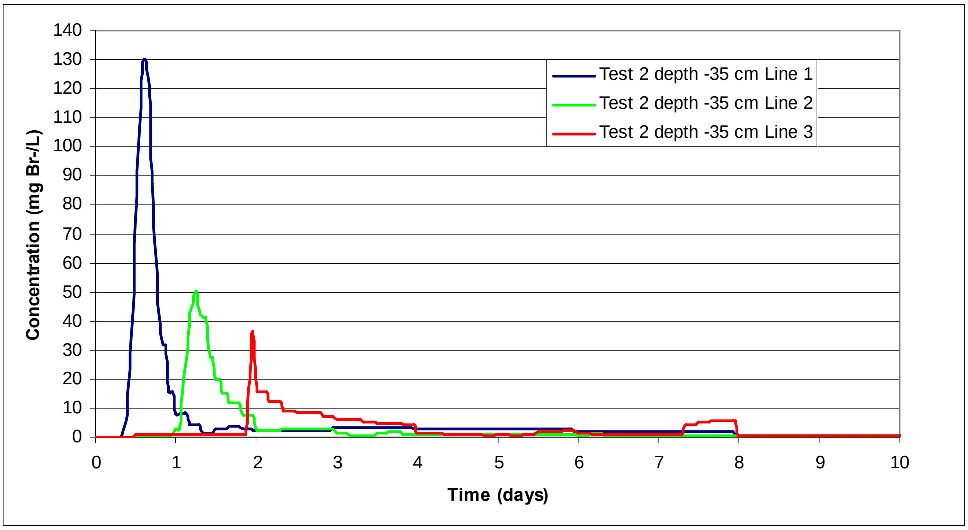


(c)

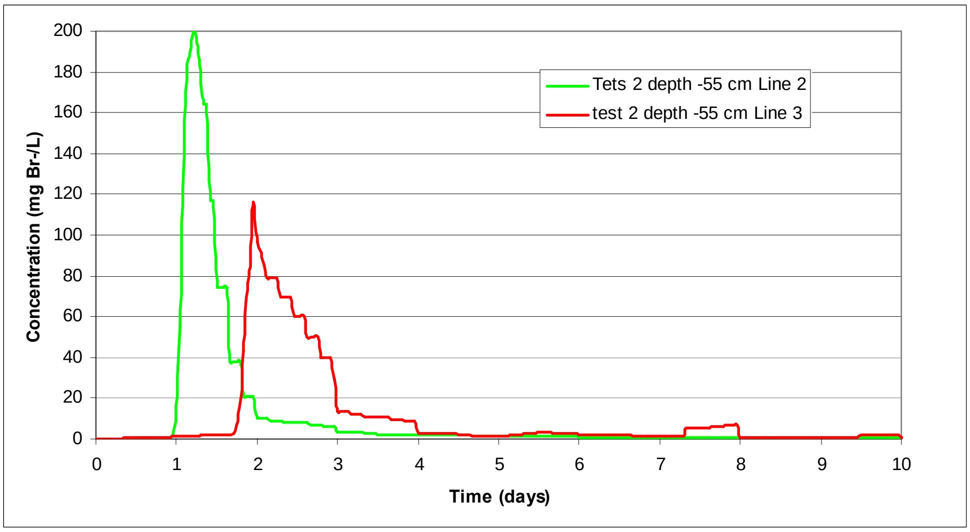
Figure 26 : TBC for test 1 for the 3 investigated lengths: L1 at 3m from inlet (a), L2 at 6 m (b) and L3 at 11 m (c) from inlet respectively through the 3 depths, from -15 cm to -55 cm.



(a)

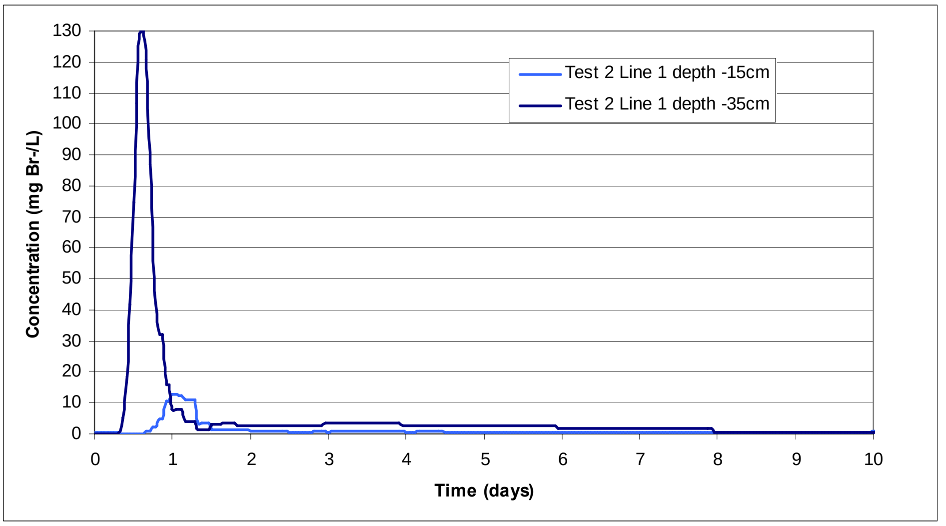


(b)

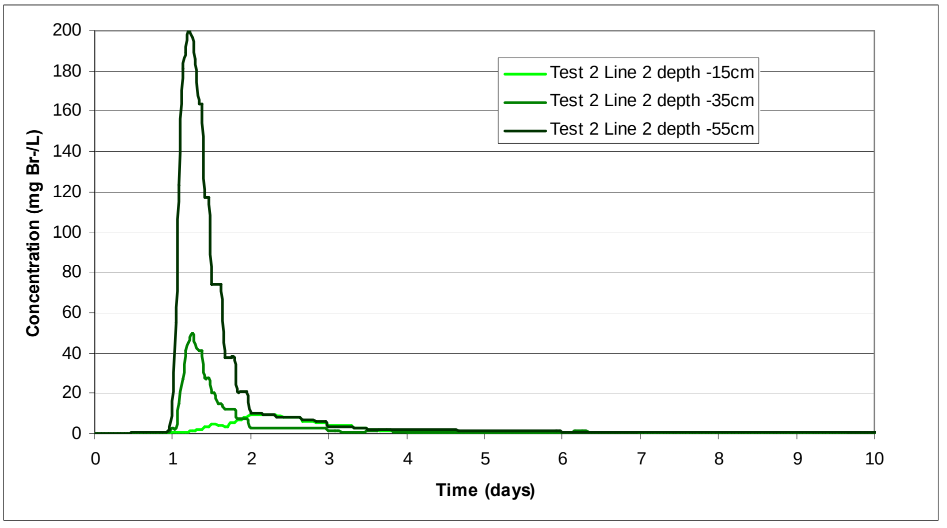


(c)

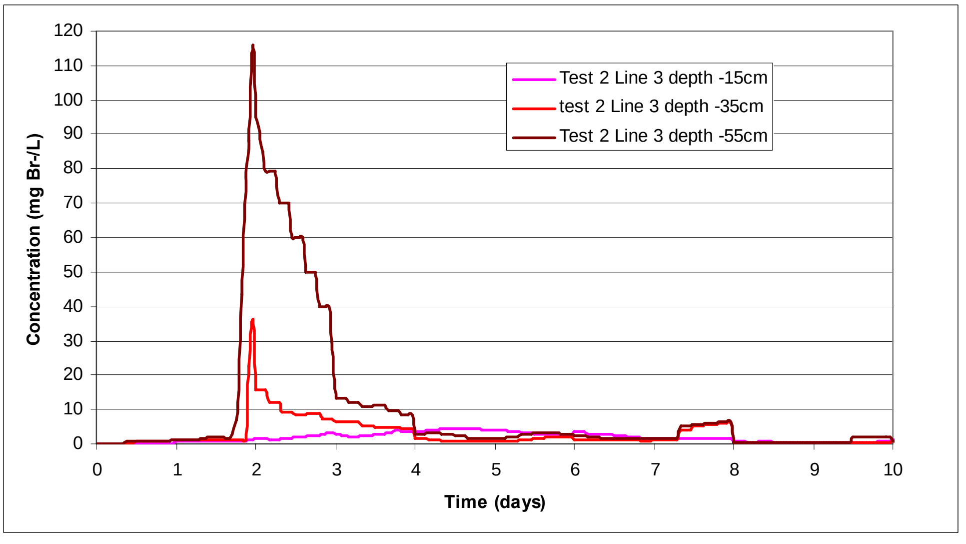
Figure 27 : TBC for test 2 for the 3 investigated depths : -15 cm (a), -35 cm (b) and -55 cm (c) over the 3 lengths, L1 at 3m from inlet, L2 at 6 m and L3 at 11 m from inlet respectively.



(a)



(b)



(c)

Figure 28 : TBC for test 2 for the 3 investigated lengths: L1 at 3m from inlet (a), L2 at 6 m (b) and L3 at 11 m (c) from inlet respectively through the 3 depths, from -15 cm to -55 cm.

II.4.1.5 Regular parameter results

In order to qualify the efficiency of the tests and the system, regular parameters described by equations from the theoretical background paragraph are presented in Table 4.
The data presented are first the design parameters of the horizontal flow bed. Data are organised according length (lines) and depths. The last column per line is a total or mean value of date related to the three depths.

Statistics attest that the set of data (concentration of tracer versus time) are homogenous and acceptable for statistical analysis for both tests. All curves are presenting a log-normal distribution where $\frac{\tau - \tau_p}{\tau_{med} - \tau_p}$ is close to 3. The median time (τ_{med}) corresponds to 50% of the tracer recovery. Statistics are normal and performed with t-distribution tests with confidence intervals of 0.05.

Porosity was measured in laboratory on gravels without roots from plants. The method used was the difference between wet and oven-dried weights of core of samples. The mean value based on three replications of the porosity of the gravels used ranked between 0,37 and 0,4. Samples from the surface and intermediate layers were considered as having a higher porosity due to the numerous roots from reeds and their "pushing-up" effect on the gravels, especially dense in the top level. The selected mean porosity is 0,45.
The **nominal volumes** per layer make the assumption that the three investigated layers are homogenous, each one being 1/3 of the total nominal volume at Lines 1, 2 or 3. The last column per line presents the total nominal volume.

The **inflow** presented in Table 4 for Tests 1 and 2 also make the assumption that the flow is equally distributed over the depth into the three investigated depths. The last column per line is presenting the total inflow per line, which is the inflow adjusted for ET.
Nominal detention time values are based on the above mentioned assumptions of low equal distribution over depth.
Tracer peak times are issued from the measurement campaigns and displayed on graphs above.

Recovery percentages are calculated as the total amount of tracer was supposed to be caught per lines. For Test 1, recovery percentages are low for Lines 1 and 2 and are good for Line 3; a failure of the automated sampler explains the low recovery percentage at the outlet (30%). The recovery distribution is then calculated based on the amount of tracer percentage found per depth, reported to the total of the concerned line. For Test 2, the sampler of Line 1 depth 55 cm collapsed and gave no data. Test 2 performed better as recovery percentages are higher than Test 1. As the whole test in general performed better, the assumption is made for the recovery distribution according to depth on Line 1 that the recovery percentage would have been equal to the percentage found at Lines 2 and 3. Both tests show that the larger amount of tracer is found at the deeper depth of -55 cm. Recovery percentage at the outlet would probably been higher is the sampling procedure had last longer.

The **Morril Index**, MI = t_{90}/t_{10} is a tracer retention time recovery ratio corresponding to the time where 90% of the tracer is recovered divided by time where 10% of the tracer is recovered. It characterises the slope of the curve. A low index value means a curve with a narrow bell shape and a high index value is representative of a flat curve. This index is a pre-assessment whether water movement is closer to plug flow reactor or to the perfect mixing of an ideal complete stirred tank reactor.

Its evaluation is ranking from:
- 1 = very good, (close to plug flow)
- 2 = good,
- 4 = medium,
- 12 = bad. (close to complete stirred tank reactor)

For Test 1 the Morril index improved over length and was almost classified as “bad” for curves related to Line 1, the closest from inlet. The same observation is drawn from Test 2 and the global values of the Morril index are higher for Test 2 than Test 1.

Dimensionless time (Θ) is provided as it will be further used to compare the two tests in situ, as they were not performed over the same time duration (10 days for test 1 and 20 days for test 2), and also with greenhouse experiments on the pilot wetland cells. Tracer tests are recommended to be performed over the duration of 5 Θ, what means 5 times the tracer peak time. It shows that the first test was stopped earlier to be replicated a second time; and the second test was performed for 4 Θ at the outlet, with up to 13 Θ and 7 Θ for lines 1 and 2 respectively, but less than 3 Θ in average for line 3.

Hydraulic efficiencies are showing quite good efficiencies for Test 1 and low for Test 2, with a total average value 74% and 47% for tests 1 and 2 respectively. For both tests 1 and 2, the best hydraulic efficiency is obtained for the line 2 located in the middle over length of the bed.

This parameter shows a strong bias, being dependant on the inflow. The inflow makes the assumption of equal distribution over depth, what is graphically demonstrated as an incorrect assumption by previous presented figures and by the recovery percentages distribution over depths. This aspect will be further developed and analysed in the results discussion section.

Table 4 : Data about the HF system and the 2 performed tracer tests

		Line			**L1**		**Mean /**		**L2**		**Mean /**		**L3**		**Mean /**	**OUTLET**
		Depth		**15**	**35**	**55**	**Tot L1**	**15**	**35**	**55**	**Tot L2**	**15**	**35**	**55**	**Tot L3**	
Parameters	**Nom.**	**Formula**	**test**													
Length	**L**		**m**	3	3	3	3	6	6	6	6	11	11	11	11	13,4
Width	**W**		**m**	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2	4,2
Depth	**D**		**m**	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65	0,65
Volume	**V**	**L*W*D**	**m^3**	2,73	2,73	2,73	8,19	5,46	5,46	5,46	16,38	10,01	10,01	10,01	30,03	36,58
Porosity	**ε**	**Measure**	**-**	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5	0,5
Nominal Volum	**V_n**	**$V*\varepsilon$**	**m^3**	1,229	1,229	1,229	3,686	2,457	2,457	2,457	7,371	4,505	4,505	4,505	13,514	16,462
DATA TEST 1																
Inflow	**Q**	**Measure**	**m^3/day**	0,723	0,723	0,723	2,168	0,723	0,723	0,723	2,168	0,723	0,723	0,723	2,168	2,168
Nom.Det.Time	**τ_n**	**V_n/Q**	**Day**	1,7	1,7	1,7	1,7	3,4	3,4	3,4	3,4	6,2	6,2	6,2	6,2	7,6
Tracer Peak Ti	**τ_p**		**Day**	1,6	0,7	0,8	1,0	4,1	3,0	3,0	3,4	5,8	3,4	2,6	3,9	3,9
Recovery			**%**				21				31				94	30*
Rec. Distribution			**%**	32	13	54	100	21	18	61	100	2	27	71	100	
Hydr.efficiency	**λ**	**$\tau_p/\ \tau_n$**	**%**	96	39	44	60	121	88	88	99	93	55	42	63	52
Morril Index	**MI**	t_{90}/t_{10}		3	10	10	8	2	4	4	4	3	1	2	2	2
dim less time	**Θ**	**t/τ_n**	**-**	5,9	5,9	5,9	5,9	2,9	2,9	2,9	2,9	1,6	1,6	1,6	1,6	1,3
DATA TEST 2																
Inflow	**Q**	**Measure**	**m^3/day**	0,847	0,847	0,847	2,542	0,847	0,847	0,847	2,542	0,847	0,847	0,847	2,542	2,543
Nom.Det.Time	**τ_n**	**V_n/Q**	**Day**	1,4	1,4	1,4	1,4	2,9	2,9	2,9	2,9	5,3	5,3	5,3	5,3	6,5
Tracer Peak Ti	**τ_p**		**Day**	1,0	0,6	-	0,8	2,2	1,3	1,3	1,6	2,0	2,0	1,2	1,7	2,2
Recovery			**%**			-	81**				81				81	66
Rec. Distribution			**%**	4	43	61**	100	12	21	67	100	15	20	65	100	
Morril Index	**MI**	t_{90}/t_{10}		12	10			7	5	4	5	8	9	4	5	10
Hydr.efficiency	**λ**	**$\tau_p/\ \tau_n$**	**%**	69	43	-	56	75	43	43	54	37	37	22	32	34
dim less time	**Θ**	**t/τ_n**	**-**	13,8	13,8	-	13,8	13,8	6,9	6,9	6,9	3,8	3,8	3,8	3,8	3,1

* Failure of the automated sampler
** based on assumption

II.4.2 Data of tracer tests for the pilot-scale wetland cells

Results from tests under greenhouse (the pilot-scale wetland cells) are presented following the same development: preliminary graphs are displayed; data and preliminary indexes are presented.

II.4.2.1 Presentation of TBC results

Figure 29 presents the TBC of the three tests according media sizes and show data with Bromide concentration (mg/l) versus time (hours). Recovery percentage of 100% is displayed by the vertical red lines on graphs. Table 5 is presenting regular parameter results.

II.4.2.2 Comments about TBC curves

The general first comments based on the observation of the curves are the follow ones.

- All obtained curves have a general regular bell shape, corresponding to theoretical expectations;
- As the exact same amount of tracer was injected for the three tests and as all axes have the same scale, it is noticeable that concentration of Bromide measured at the outlet of the systems seems to be relatively homogeneous with replication;
- Nevertheless, a general decrease in tracer peaks is observed from Test 1 to Test 3;
- No difference in tracer concentration between media size can be seen in an initial overlook on curves;
- All curves have a unique tracer peak attesting to neither differential flow velocities with layers within pilots, nor back mixing process;
- Tracer recoveries are good for all performed tests.

II.4.2.3 First assessments from TBC results on hydraulic patterns

First assessments are similar to those provided by the onsite tests. The general good bell shapes of all curves demonstrate that the general behaviour of the system is following a general plug flow water movement. Nevertheless the plug flow behaviour is not ideal, as the curves have a bell shape and are not straight lines as ideal plug flow would display.
No back mixing or delays in tracer peak at the outlet are observed, indicating that all the pilot volume is involved in the flow.

II.4.2.4 Water budget

The water inflow is delivered by a peristaltic pump with constant monitoring in order to respect a constant, regular and measured inflow for the three replicated. The water exiting the system is collected every hour in buckets at the outlet of pilots and was measured for Test 2 and 3, but was not measured for Test 1. Inflow and outflows were under continuous measurement and known for Tests 2 and 3. Outflow from Test 1 were calculated based on evapotranspiration rates observed for Test 3, which was performed during the same season, and tables of evapotranspiration according to the temperature of the greenhouse which was daily recorded.
A significant difference in evaporation rate was observed within the four media sizes during all the tests, with the largest difference observed during the third test (summer): the smallest media size of 3-5 mm had a significantly different rate of water loss on a daily basis than the three other media sizes. For the smallest media size, the mean value was 11% of water loss/ day, whereas it was 4 to 5 % for the larger media sizes.

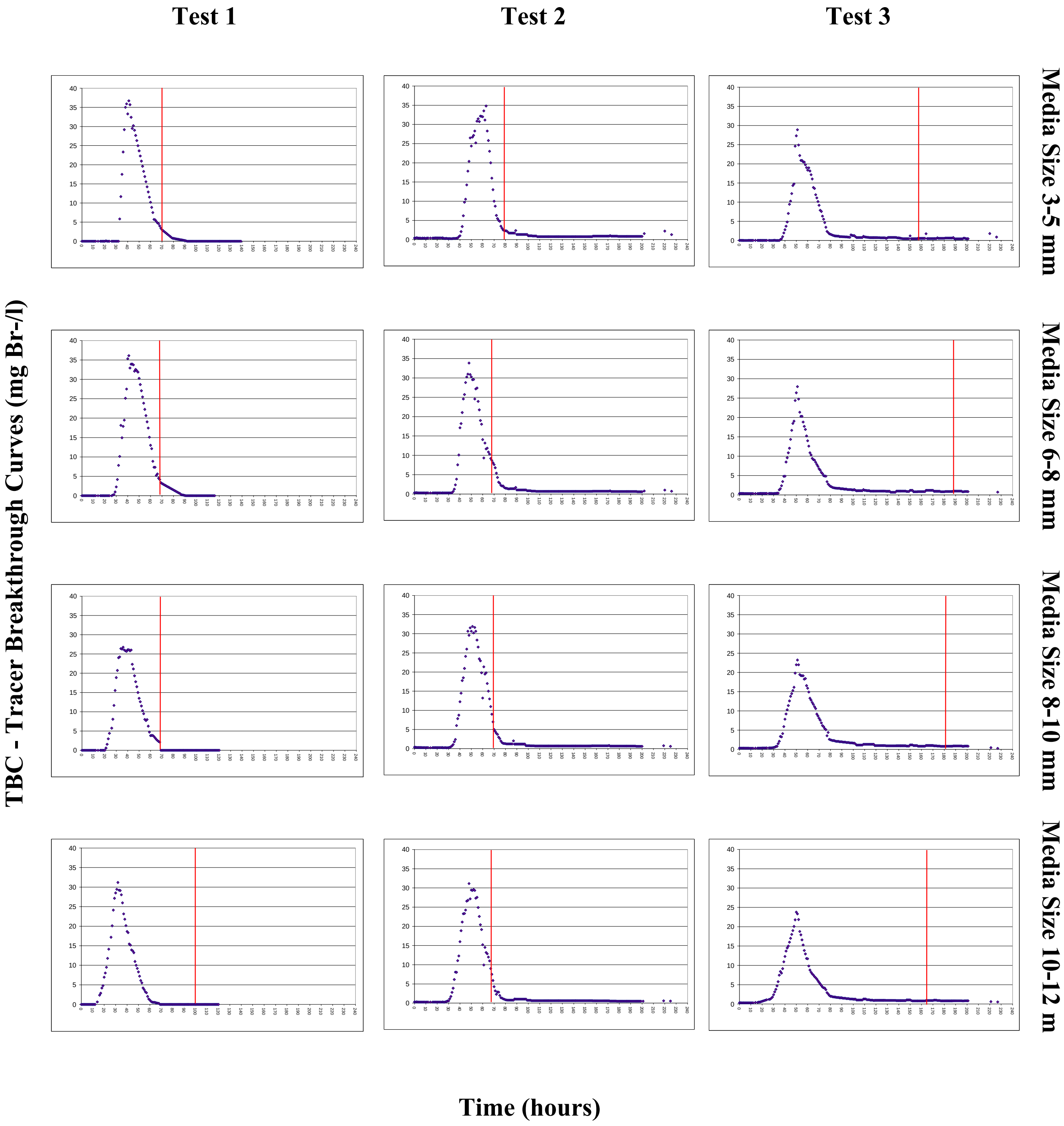


Figure 29 : Graphs of three replicated tracer tests, according to media sizes (mm). Graphs present the TBC – Tracer Breakthrough Curves- of Bromide concentration at the outlet (mg Br-/L) versus Time (hours). Axes have all equal scales. Vertical red lines indicate the point of 100% tracer recovery

II.4.2.5 Regular parameters result

Statistics attest that the set of data (concentration of tracer versus time) are homogenous and acceptable for statistical analysis for all three tests and media sizes. All curves are presenting a log-normal distribution where $\frac{\tau - \tau_p}{\tau_{med} - \tau_p}$ is close to 3. The median time (τ_{med}) corresponds to 50% of the tracer recovery. Statistics are normal and performed with t-distribution tests with confidence intervals of 0.05.

As the first test was performed without measures of the outflow, the tracer mass recovery for Test 1 was estimated taking into account the evaporation rate measured by the two next tests. The first test was done on late summer (August 2007), the second during fall (September 2007) and the third one during the next summer (July 2008).

The mean value **tracer mass recoveries** of tests two and three (including all media sizes) was 96%, which is considered an excellent rate of tracer recovery. Test one was excluded from this calculation (as the mass of the tracer recovered could not be directly measured).

First calculations on data show that the theoretical **nominal detention time** (τ_n), depending on porosity (which is itself increasing with the media size), grows from 2,2 day for media sizes 3-5 mm and 6-8 mm, to 2,3 day and 2,4 day for media sizes 8-10 mm and 10-12 mm, respectively.

The **tracer peak time** (τ_p) varies significantly within replicated tests and between mean values of media sizes. The tracer peak time is smaller for larger media size and ranged from 2.1 for media size 3-5mm down to 1.8 for media size 10-12 mm. The tracer exits the bed faster with coarser media than fine media and peak time is accordingly achieved faster for larger gravels than fine ones.

As no reference of replicated tracer tests under the same experimental conditions has been found in literature to explain the phenomena, the following hypothesis were speculated : (i) tests were not performed during the same season of the year, which has induced variable evaporation of the gravel beds; (ii) repairs of leaks has been done to the beds in-between tests which necessitated removal and replacement of the gravels, this could have induced changes in preferential flow paths for the different tests, and (iii) the pilots were not set under the exact same flushing conditions before starting the test. It seems the first experiment did not adequately flush the gravel beds before starting the test, which is demonstrated by a systematically lower hydraulic efficiency.

The **hydraulic efficiency λ** presents the ratio of tracer peak time divided by nominal detention time. Garcia *et al* (2004b) report that hydraulic efficiency can be categorised as "good hydraulic efficiency" when λ is > 0.75 (or 75%), which is the case for the total average of all tests and media sizes reported as 0,79 or 79%. The pilots can be considered as having a good general hydraulic behaviour. As the hydraulic efficiency is directly depending on tracer peak time, the same observation is made as for tracer peak time, which is that the hydraulic efficiency decreases when media size increases, from 87% as mean value for media size 3-5 mm down to 71% as mean value for media size 10-12 mm.

It is also pointed out that the first test has systematically lower hydraulic efficiencies. It can be explained by the fact that the pilots were not properly flushed and saturated before the start of the first experiment.

The **Morril Index**, MI = t_{90}/t_{10} is defined here above (see § II.4.1.5). MI obtained for the three tests and four media sizes are ranking from 1,1 to 2,2 with a slight increase with media size increase, what is classified as very good to good.

Dimensionless time is calculated for comparison with onsite tests. All tests were performed over almost 4 ϴ, which is close to the ideally recommended 5 ϴ.

Briefly, these first results show that :
- the pilot design combined with the experimental method provides a good hydraulic behaviour of the pilots,
- the replication of tracer tests experiments leads to a high variability within replicated test for measured tracer peak times,
- the tracer exits the beds faster with coarser gravels.

Table 5 : Data about the tracer tests on pilots under greenhouse.

	Media size		3 - 5 mm				6 - 8 mm				8 - 10 mm				10 - 12 mm				3 mm
	Replication		Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	12 mm
Parameters	**Form.**	**Units**																	**Tot Mean**
Length	**L**	**m**	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Width	**W**	**m**	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3
Depth	**D**	**m**	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3	0,3
Volume	**L*W*D**	**L**	270	270	270	270	270	270	270	270	270	270	270	270	270	270	270	270	270
Porosity	**Meas.**	**-**	0,35	0,35	0,35	0,35	0,36	0,36	0,36	0,36	0,38	0,38	0,38	0,38	0,39	0,39	0,39	0,39	0,37
Nom. Vol.	$V*\varepsilon$	**L**	95	95	95	95	97	97	97	97	103	103	103	103	105	105	105	105	100
DATA																			
Inflow	**Meas.**	**L/day**	40,00	40,30	40,62	40,31	40,00	41,34	40,66	40,67	40,00	42,82	40,68	41,17	40,00	41,76	40,32	40,69	40,71
Nom.Det.Time	V_n/Q	**Day**	2,4	2,3	2,3	2,3	2,4	2,4	2,4	2,4	2,6	2,4	2,5	2,5	2,6	2,5	2,6	2,6	2,5
Tr. Peak Time	τ_p	**Day**	1,7	2,4	2,1	2,1	1,7	2,0	2,1	1,9	1,5	2,2	2,1	1,9	1,3	2,1	2,1	1,8	1,9
Recovery		**%**		94	99			94	95			95	99			92	96		
Hydr.efficiency	$\tau_p/\ \tau_n$	**%**	72	100	90	87	70	85	88	81	58	92	83	78	49	83	80	71	79
Morril Index	t_{90}/t_{10}	**MI**	1,5	1,3	1,4	1,4	1,5	1,4	1,7	1,5	1,7	1,4	1,3	1,5	2,2	1,4	2,0	1,9	1,6
dim less time	t/τ_n		3,5	3,6	3,6	3,6	3,4	3,6	3,5	3,5	3,3	3,5	3,3	3,4	3,2	3,3	3,2	3,2	3,4

II.4.3 Results and discussion

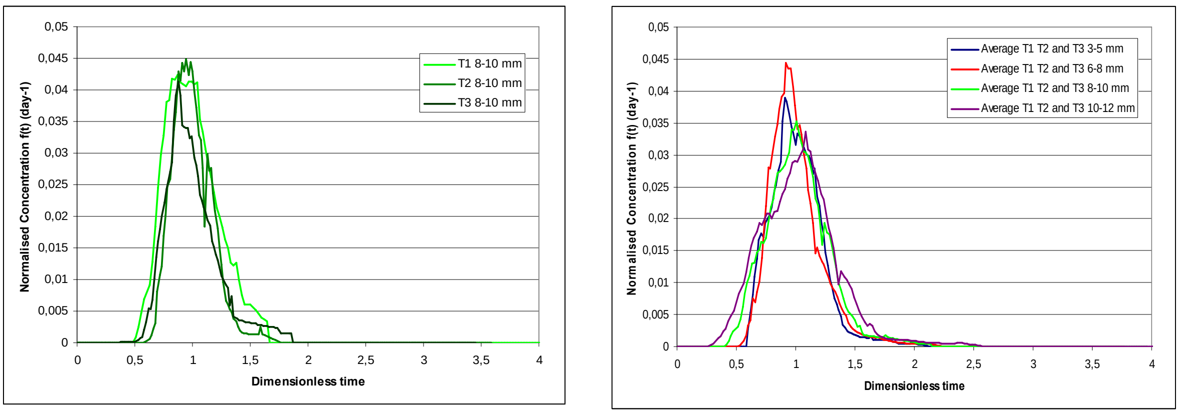
The first part of this chapter has presented raw data and provided preliminary hydraulic indexes.
In this section, results are presented and discussed following three steps.

1. A graphical analysis of data and comparisons between tests onsite and under greenhouse are presented.
2. Then, mathematical models are applied to the set of data from experimental tests and their solutions give the quantitative determination of the system properties with values for feature parameters.
3. The mathematical models are finally calibrated to data obtained from experimental tests, providing adjusted parameters to qualify the systems tested.

II.4.3.1 Graphical analysis and comparison between tests and experimental sites

In order to compare tests on pilots under greenhouse and tests on full scale site of Nassogne, which were performed on variable duration times, graphs are drawn with normalised concentration versus dimensionless time (Θ).
Normalised concentration is given by Equation 17 and is the tracer concentration measured on time *t*, divided by the integration of all concentration on time t for the delta t interval - C(t)dt. The sum of all f(t) is equal to unity and correspond to the area below the curve.

Pilots under greenhouse
The graphs were built for the four media sizes investigated in the pilots under greenhouse, with comparison for the three replicated tests. Normalised concentrations are much closed from one test to another as shown by the Figure 30(a) and no relevant differences were displayed. As graphs for each media sizes are similar, only one granulometry is presented by Figure 30(a).
The average normalised curves of the three tests is then drawn per media size and presented on Figure 30(b). The Figure 30(b) shows a general decrease in the tracer peaks with the increase of the media sizes, except that tracer peaks is higher for media size 6-8 mm; and also that peaks are occurring faster with lower media sizes.

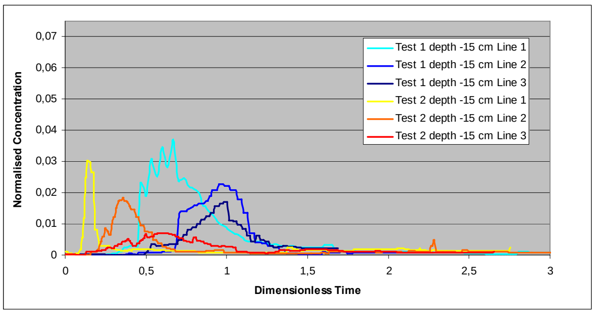


(a) **(b)**

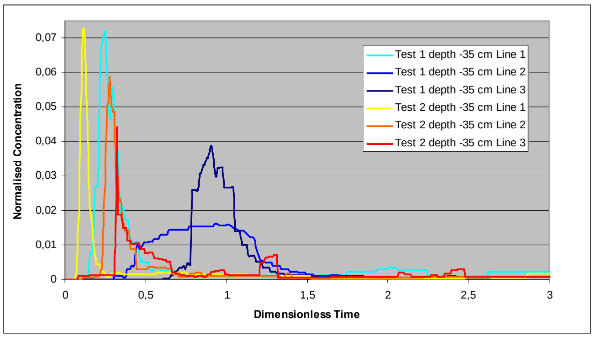
Figure 30: Tracer Breakthrough Curves (TBC) of Retention Distribution Time (RTD).
(a) Comparison of the three replication tests for media sizes 6-8 mm.
(b) Comparison of average TBC from the three replication and the four media sizes.

<u>Full scale site of Nassogne</u>

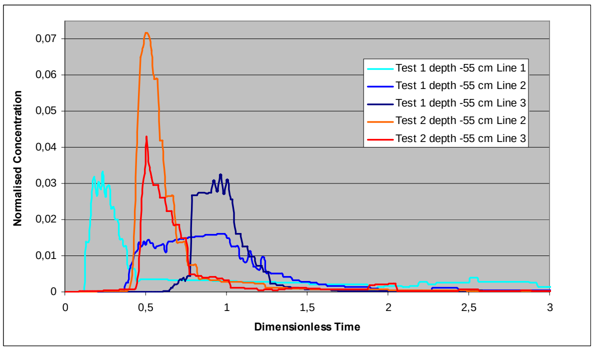
Similarly, graphs of normalised concentration versus dimensionless time are built for the two tests performed on the site of Nassogne. Curves are compared over length and with depth and presented on Figure 31. The figure has to be read per columns. The colour code is the blue family lines for Test 1 and the red family lines for Test 2. X and Y axes have all the same scales for comparison between graphs. As reminder, no data were obtained for test 2 on the line 1 at 55 cm deep due to automated sampler failure.



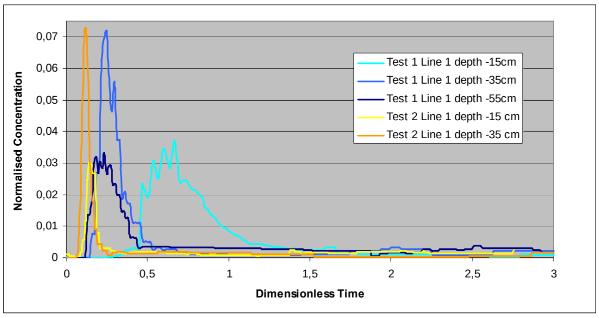
(a) comparison of curves for -15 cm deep over lengths and for the 2 tests



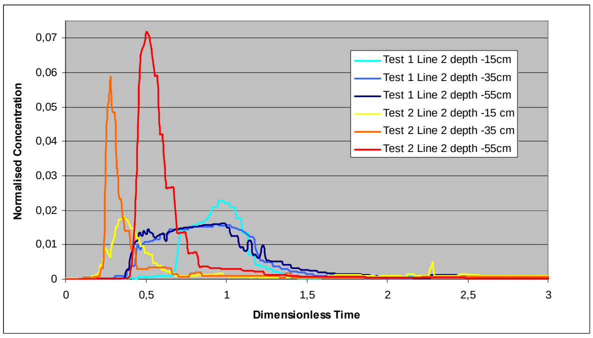
(b) comparison of curves for -35 cm deep over lengths and for the 2 tests



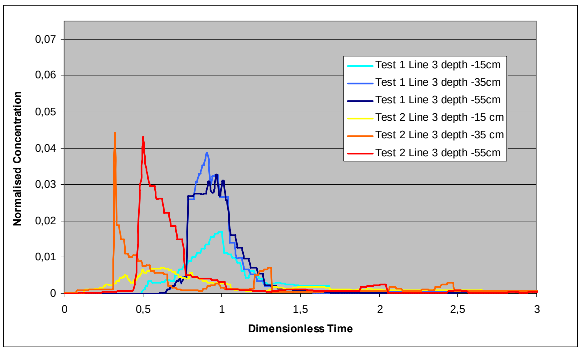
(c) comparison of curves for -55 cm deep over lengths and for the 2 tests



(d) comparison of curves on line 1 for the 2 tests and with depths



(e) comparison of curves on line 1 for the 2 tests and with depths



(f) comparison of curves on line 1 for the 2 tests and with depths

Figure 31 : Normalised concentration versus dimensionless time of the two tests performed on the full scale site of Nassogne. Vertical left column of graphs (a) to (c) is presenting results along a vertical transect with depth and right column of graphs (d) to (f) is presenting result along horizontal transect over length.

Figure 31(a) (focused on 15 cm deep) shows that the two curves on line 1 for Test 1 and 2 (yellow and pale blue lines) present similar tracer peaks but are not equally distributed over time; the bell shape of curves are narrower for Test 2 than Test 1. The same observations are pointed out for Line 2 (orange and mid blue lines). On Line 3, red and dark blue curves are very different for peak intensities, distribution over time and bell shapes of the curves.

Figure 31(b) (focused on 35 cm deep) is again presenting very similar curves for Line 1 between Test 1 and 2, as yellow and pale blue lines have equal peak intensities, similar narrow bell shapes and are delayed with time. Line 2 (orange and mid blue lines) show a very different behaviour with peak intensity being lower for Test 1, the bell shape being flatter for Test 1 and also tracer crossing Line 2 much faster with Test 2 than Test 1. Finally, tracer on Line 3 is presenting similar peak intensities, narrower bell shapes for Test 2 and delay with time for Test 1.

Figure 31(c) (focused on 55 cm deep). No comparison can be done for Line 1 as the second test failed in giving data at this point. The exact same observations are made for Line 2 and 3 at 55 cm deep than at 35 cm on Figure 31(b).

As general comment, the three Figures looking at the horizontal tracer behaviour for each individual depths over length are showing that :

- the tracer crossed the bed faster with Test 2 than Test 1, as the red family curves are systematically appearing earlier with time than blue ones;
- for the red family curves, tracer peaks are distributed gradually over time for 15 cm deep between Lines 1 to 3, then are appearing closer for depth 35 and are almost on the same time on Line 2 and 3 at 55 cm deep;
- for the blue family curves, this phenomena is not observed as the distribution of peak times remains gradually scheduled over Lines 1 to Line 3, whatever the depth;
- for all depths, tracer peak intensities are generally closed for Tests 1 and 2 but decreasing over length from Line 1 to Line 3;
- test 2 is systematically presenting narrower curves than Test 1, which have flatter bell shapes.

Figure 31(d) is focused on Line 1 for all investigated depths and shows that tracer peak intensities are similar for 15 (yellow/pale blue lines) and 35 cm deep (orange/mid blue lines), but the tracer is processing faster through Line 1 with Test 2 than Test 1, as blue curves are delayed over time and yellow and orange curves are much narrower than blue ones.

Figure 31(e) is focused on Line 2 for all depths and shows that tracer peak intensities are similar between tests at 15 cm deep (yellow/pale blue lines) but very differently distributed over time. The two other depths of 35 cm and 55 cm are showing large differences between the two tests for tracer peak intensities, distribution over time and bell shapes of the curves. Test 1 is systemically presenting higher intensities, earlier on time and with narrower curves shapes.

Figure 31(f) focusing on Line 3 for all depths is presenting homogeneous curves for tracer peak intensities between the two tests for depths 35 cm and 55 cm except for 15 cm deep, where Test 2 presents a lower tracer peak intensity (yellow/pale blue lines). Test 1 and Test 2 are not occurring over the same time distribution, and Test 2 is systematically faster than Test 1. Finally, tracer peak are occurring almost on the same time for Test 1 whatever the depths and are spread out over time for Test 2.

General comments about those three graphs investigating vertical tracer behaviour lines are the following.

- Similar tracer peak intensities are observed on Lines 1 and 3, but not on Line 2;
- Test 2 is systematically processing faster over the 3 lines than Test 1;
- All tracer peaks are regularly scheduled over time from Line 1 to Line 3;
- Tracer peaks are scheduled with time for Test 2 on Lines 2 and 3 but are occurring on the same time for Test 1;
- Tracer curves Test 1 and Test 2 are presenting very different results on Line 2, especially for 35 cm and 55 cm deep.

Comparison between small scale pilots and full scale site

Small scale pilot experiments were led to investigate the influence of media size on the hydraulic behaviour. Outlet results are here generally compared between the two tests on the full scale TW and small scale pilots under greenhouse. Normalised tracer concentrations versus time are presented the comparison of the 2 investigated bed scales on Figure 32. The black curve is an average curve of all tests done on pilots, with media sizes and replications as Figure 30(a) and 30(b) displayed that they all had a very similar behaviour when using normalised and dimensionless axes.

Figure 32 is showing that the tracer peak intensities between the two tests on the full scale system (blue and red lines) are similar, even if delayed over time, whereas small scale pilots (black line) is presenting higher intensity. This is not surprising due to the better controlled conditions and best tracer recovery percentages observed for the pilot tests.

The bell shape of the curve from the pilot experiments is also more symmetrical than the ones from the full scale system.

This comparison shows a similar hydraulic flow pattern for small and full scale systems.

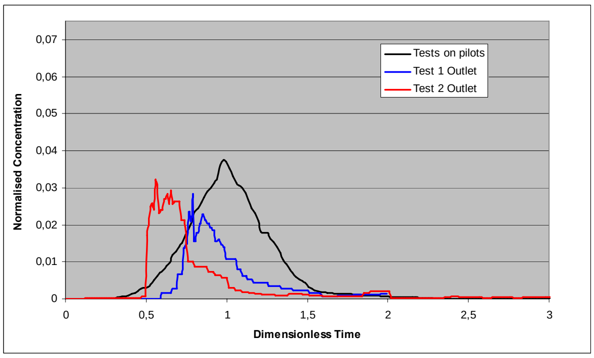


Figure 32 : Comparison of outlet normalised concentration versus dimensionless time for tests on small and full scale systems.

Conclusion of the graphical analysis

The graphical presentation of results provides the following major information.

- Test 1 and Test 2 on full scale system present differences on tracer peak intensities and peak times;
- Test 2 on the full scale system systematically occurred faster than Test 1;
- Test 2 on the full scale system presents systematically higher peak intensities than Test 1;
- Line 1 and Line 3 on the full scale system are presenting similar results for both tests, the major differences are occurring between the two tests at Line 2 for 35 and 55 cm deep;
- Small scale and full scale systems are presenting similar curves and tracer behaviour.

About the hydraulic pattern, this provides the information the bed has general plug flow behaviour closer to ideal for Test 2 than Test 1 with negligible back mixing in both cases. It also demonstrates that the media is not delivering homogeneous condition neither with depth, nor over length.
In order to quantify these results, mathematical models were applied to characterise the parameter values of the system.

II.4.3.2 Application of mathematical models and data fitting process

An estimation of parameter values from tracer experiments is only possible if an adequate mathematical model is used. Different approaches and models are here tested to provide parameter values which best reflect the tracer transport and tracer behaviour in the system being studied.

The calculation of the Reynolds number, based on Eq. 15, allows assessment if the flow considered is to be laminar or turbulent, as

$1 < Re < 10$ is characterising a laminar flow and
$Re > 10$ is representative of the transition to turbulent flow (Kadlec and Wallace, 2009).

In the case of the site of Nassogne in operation as well as for greenhouses:
- the gravel media size of the bed is ranging from 3 to 12 mm, the average of 6 mm is taken for the calculation;
- tests were performed with clear water, which viscosity and density is considered to be 1000 kg/ m.d and kg/m^3 respectively.

The calculated result of Equation 15 is a Reynolds number of 10.
This value is the tipping value between two conceptual modelling theories of laminar or turbulent flows, it is thus acceptable and relevant to investigate both in the frame of this work.

The mathematical models which were solved with data results from experiments to provide system parameters are: (1) the GAMMA Distribution of Tank In Series model (TIS GAMMA), (2) Dispersion Model (DM) also called Dispersed Plug Flow, and (3) the Multi Flow with Dispersion Model (MFDM).

Calculations are done with two different steps:
1. The moment method (MM), which is based on integration processes, was firstly used to estimate parameters and solve the mathematical equations;
2. Secondly, the reversed process named ‘retrofitting’ or ‘model calibration’ uses the least squares method or Sum of the SQuares Errors (SSQE) which is a computational iteration process, to adjust and fit the mathematical model curves found by step one with the set of data obtained from field experiments in order to provide new and more accurate values for the parameters of the investigated system.

II.4.3.3 Graphical presentation of results

Graphical results are presented first for the tests on pilots under greenhouse, secondly, for the full scale site of Nassogne and finally, pilot and full scales results are graphically compared.

The following parameters of the system are provided:

- The moment method provides the tracer residence time (τ, in days);
- The solution of the GAMMADIS model provides the number of tank in series *NTIS*, which is indicative of a hydraulic behaviour close to plug flow or not;
- The SSQE method adjusts the two model parameters (τ and NTIS) to fit with experimental data and provides an adjusted value of the tracer residence time and number of tank in series;
- The ratio of tracer residence time to nominal detention time gives the real volumetric efficiency of the system;
- The solution of the Dispersed Plug Flow model or Dispersion Model (DM) adjusted with SSQE method provides the unitless Peclet and dispersion numbers (Pe and D), corresponding to the TIS and DM models, and the axial dispersion D_L and the flow velocity (v) of the three monitored layers.

The two models (TIS and DM) provide system parameters which quantify and qualify the hydraulic behaviour and efficiency of the tested system. They both make the assumption that the media is homogeneous, what has been graphically demonstrated as not true by Figures 25 to 28.

The last model, Multi Flow with Dispersion Model (MFDM) was applied and the solution of its mathematical equation, adjusted with SSQE method present for each line: the number of identified flow paths (N), the fraction of the flux involved (P_i), the dispersion parameter (P_D) and tracer residence time (τ).
This model could be considered as less accurate due to the number of parameters which have to be adjusted, four for this model next to two for the former ones. However, with the data sets generated by this work such a model can be made to fit with field data without any indication on layers, as in our case, we already have data related to underground layers with measurements from 15 cm, 35 cm and 55 cm depths. The number of flow paths and the tracer residence time are given by previous model parameters and tracer peak times by the data sets. The MFDM can thus be reduced to 2 parameters to adjust, and thus fit as well as the previous developed models.

Figure 33 shows the TBC of the TIS model without adjustment. Figure 18 showed the theoretical curves which are observed according to the number of Tank In Series. Figure 33 presents a couple of curves obtained for Test 2 according to the increasing number of TIS found by the solution of the mathematical equation. The graph is displayed with dimensionless time X axe versus normalised C(t) on Y axe.

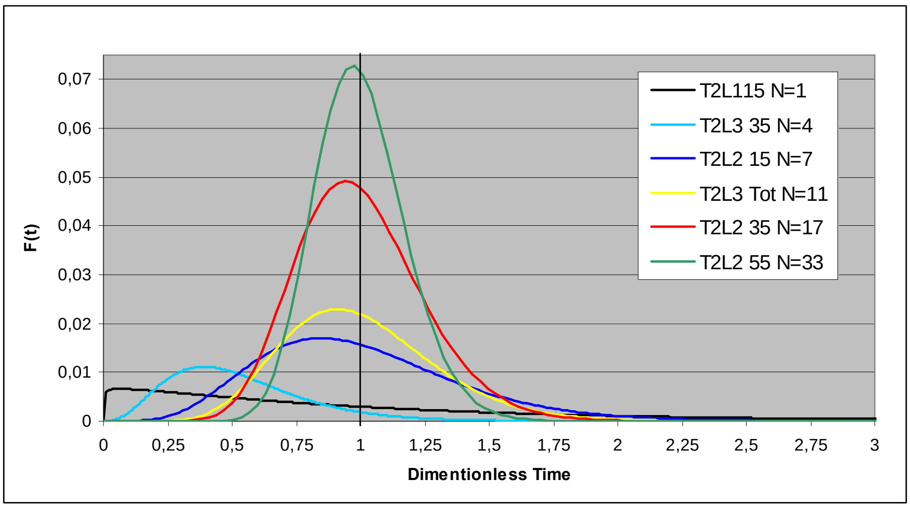


Figure 33: Observed TBC for TIS model

The vertical black line is drawn on dimensionless time equal to 1, what corresponds to one the tracer detention time τ. Standard tracer tests advise to schedule the experiment over 5 τ. In our case, all tests were performed over more or less 3,5 τ.
When N=1, the TBC is a decreasing exponential curve reflective of a CSTR , and when N = ∞, it fits to the black straight line and is reflective of the ideal plug flow.
It is clearly shown with Figure 33 that firstly, the higher the number of TIS N is, the higher and narrower curves are, and secondly, that tracer peaks are getting closer to 1 τ when the number N is increasing, what perfectly fits with the theory.
It also gives us the first indication that when the number N of TIS will be low, the curve will have a flat bell shape, probably fitting badly with the data bell shapes presented by Figures 25 to 28.

<u>Graphical results of the full scale site of Nassogne</u>
Figure 34 is presenting here below the same graphical results of those calculation for the full scale experiment, site of Nassogne. All X and Y axes have the same scale to allow comparison between graphs. Blue dots are measured data of normalised tracer concentration versus dimensionless time; green lines are the mathematical solutions of the TIS equation using the moment method; red lines are the adjustment of TIS model using the Gamma distribution added with the sum of squared error method and blue lines are the solution of dispersion model equation without adjustment.

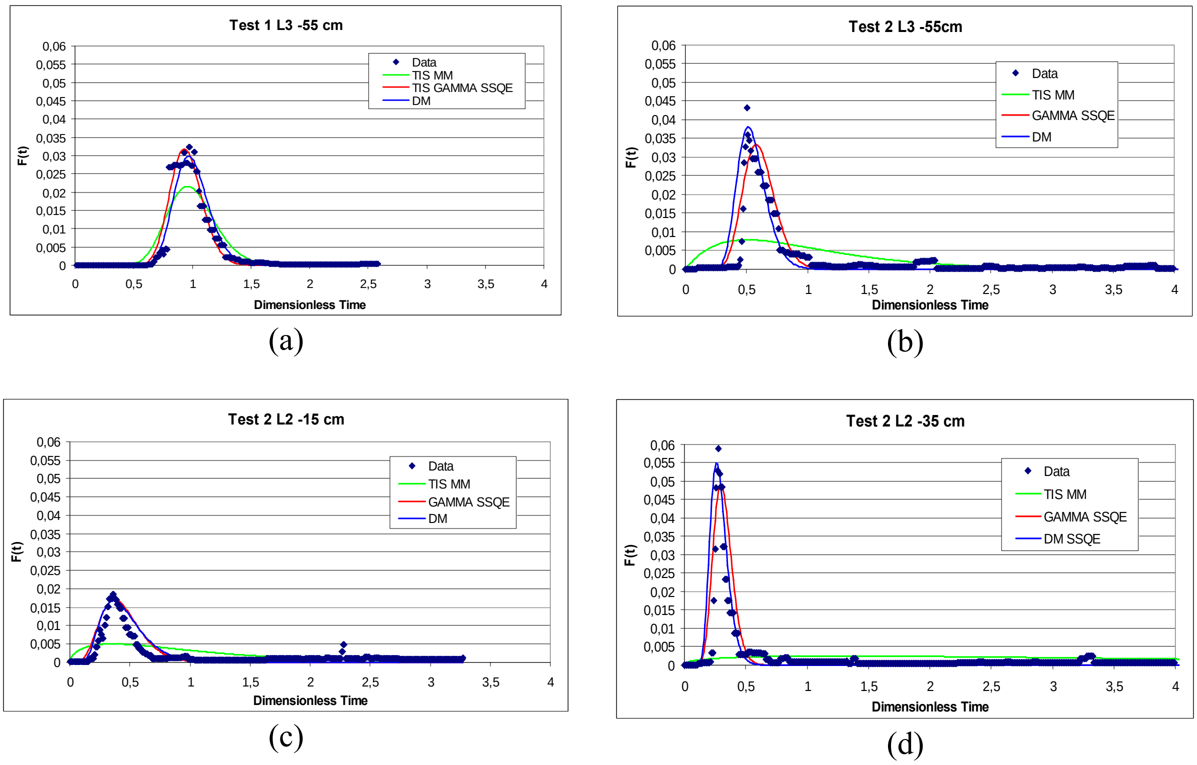


Figure 34 : Field data, model solutions and adjustments from tracer experiments on the full scale site.

For the presented cases, (which were similarly observed for all results), the solution of the TIS equation using the TIS equation poorly fits to the data sets, especially when the number of tank in series N is low. N = 24 for Figure 23 (a), N=2 (b), N=2 (c), N=5 (d).

Again, the least square error method is applied to sum the square of the difference between the observed and calculated curves (green lines and blue dots) and to minimize it with the adjustment the two parameters of the tracer retention time and number of TIS. The new curves (red lines) obtained from this method fit much better to the data set and provide new values of the system parameters.
This demonstrates that the TIS model without adjustment developed by the chemical engineering theories is poorly reflective of the observed data for our horizontal treatment wetland system.
Nevertheless, once we have a good set of data, the calibration of this model can provide good system parameters values, indicative of the general hydraulic behaviour.

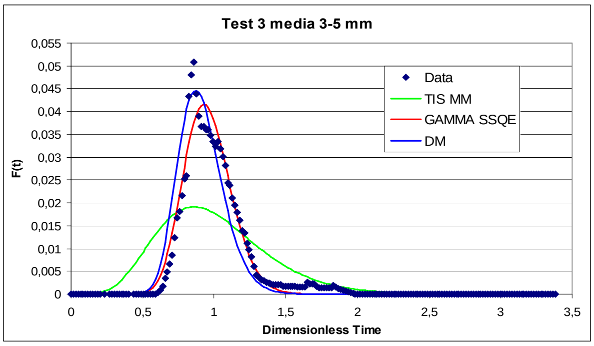
The blue line is presenting the solution of the dispersion model equation and they all fit very well without adjustment. The retrofitting is nevertheless made to obtain the system parameters values and the obtained curves are exactly above the red ones. The adjusted curves are thus not here presented as they are combined with the red ones.
The dispersion model has two unknown parameters, which are the flow velocity and the axial dispersion. In our case, the flow velocity, being a distance over time, was taken as the distance from inlet of measurement point divided by the tracer peak time observed. The axial dispersion was assessed by the equation describing the Dispersed Plug Flow as reminded by equations 27, 28 and 29. These two estimated parameters were very suitable to obtain a good fit of the curves without adjustment. Adjustment was secondly made with the SSQE method.

This shows that the Dispersion Model describes very well the system without the need of adjustments. It will nevertheless provide new and different information on the hydraulic behaviour than the TIS model does.
The system parameters values are presented in Table 6.

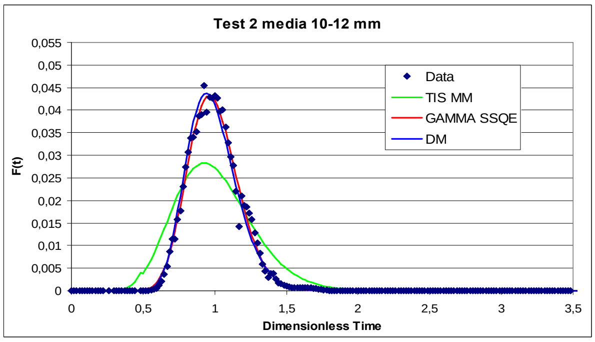
The DM model has to be solved by specific calculation software as it has non linear equations. Calculation software solves and also automatically adjusts the two parameters to the data set. In our case, the equation was set in regular Excel sheets (from Microsoft Office software), but as Excel cannot solve non linear equations, both parameters to adjust were assessed. The assessment resulted from Equations 27, 28 and 29. The Solver routine in Microsoft Excel was then used to adjust results with the data set in order to have the final requested parameters of flow velocity and axial dispersion. Table curve software was used to double check the results obtained, but Excel was kept for its convenience for graph drawings.

Graphical results of pilots under greenhouse

The presentation of all graphs issued from all results would not add to the understanding of the research process. Figure 35 presents two of the graphical results of the mathematical solutions and their adjustments for tests on pilots under greenhouse.



(a) Test 3, smaller media size tested



(b) Test 2 larger media size tested

Figure 35 : Field data, model solutions and adjustments from tracer experiments on small pilots under greenhouse.

Blue dots are the concentration measure reported as normalised concentration on Y axis; green line is the solution of the TIS model equation with the moment method, the red line is the adjustment of the TIS model with the data set obtained and the blue line is the solution of the DM model.

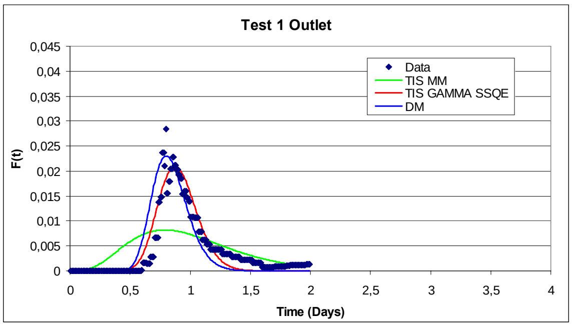
Both Figures are not showing large differences, even as it displays the two extreme tested media sizes of 3-5 mm and 10-12 mm pea gravels, between the two curves neither for the intensities, nor for the belt shapes.
For both presented graphs, as well as for all results, the solution of the TIS model with the moment method (green lines) poorly fits the set of measured data. The use of the Gamma distribution and sum of squared errors to adjust the green line with the data set is efficient to provide curves fitting very well with the data (red lines).
Finally, it is clearly demonstrated that the DM fits immediately very well with the data set (blue line).

Here again, calculations were done with the regular EXEL table sheets provided by Microsoft Office; any mathematical software is able to solve this type of non linear equation (TableCurve, e.g.) to find these two parameters without pre-estimation. TableCurve software was used to double check the results but Excel was chosen to presents all graphs in a fancier way.

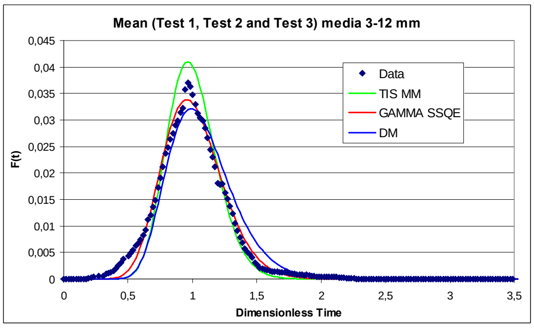
The graphical presentation of results of Figure 35 shows that the solution of the DM equation with assessed parameters from the TIS and related DPF model is providing a very good fit with the data set. This means that the single use of TIS model is poor to predict the real behaviour of a TW but when it is adjusted and jointly used with DM, it provides good dispersion parameters, reflective of the data set obtained.
The calculated parameters values of the system are presented in Table 7.

<u>Graphical comparison between pilot and full scale results</u>
Figure 36 is presenting the graphical comparison of mathematical models solutions and calibrations for results from tests on pilots (a) or on full scale site (b).
The results presented for the full scale site are from the Test 1 which was previously demonstrated as similar to the Test 2, except that Test 2 occurred faster than Test 1.

The TIS model fits again poorly to the data from the full scale test, but the calibration allows a good fit to the data obtained; the DM applied with assessed parameters from the DPF model equation fit very well without adjustment to the set of data.



(a) full scale site of Nassogne, results from test 1



(b) pilot scale, average results of all tests and all media sizes

Figure 36 : Comparison between results from full scale tests (a) and tests on pilots under green house (b)

The results presented for the pilot tests are total average of results obtained for all media sizes and tests. The mean value for each media sizes is firstly calculated for the three tests and the average is then secondly calculated combining all media sizes mean values.
In this case, Figure 36 (b) shows that for small scale pilot systems the TIS model presents a curve which fits quite well to the data, even if it provides a curve above the set of data. As demonstrated till now, the DM provides a curve which fits very well to the set of data, even before any adjustment and with parameters assessed by the DPF equations.
The general shapes of curves from small scale and full size experimental sites are similar; tracer peak intensities are lower for the full scale site; the tracer recovery percentage was also lower on the full scale site.

II.4.3.4 System parameters of the full scale site of Nassogne

Tables 6 and 7 are giving the results of the system parameters values, obtained by the solutions of mathematical models and their calibration for both experimental sites.
Indexes ratios are calculated out of some results.

Table 6 is providing results issued from data of tracer tests on full scale site of Nassogne. For comparison between full and small scale systems, the last column is including the average results from tests under greenhouse, which are detailed by the next table (Table 7).

Table 6 is distinctly organised to present firstly results from the solution of the Tank In Series (TIS) model with the moment method, secondly, results from the TIS model improved with the Gamma Distribution and calibrated with the sum of squared error (SSQE) method and thirdly, results issued from the calibrated Dispersion Model (DM + SSQE).

The moment method of the TIS model provides values of tracer detention time (τ), the dimensionless variance and the number of tank in series (NTIS).
The **tracer detention time** is increasing with depth for line 1 (located at 3 meters from inlet) and reversely is decreasing with depth for Lines 2 and 3, respectively located at 6 and 11 meters from inlet. The replication with Test 2 shows the same effect, even stronger than on

Test 1, as the detention time at 15 cm deep is 2,5 times longer than at 55 cm deep on Line 2 and similarly twice longer at Line 3. The second tests failed on Line 1 at 55 cm deep to collect data and no confirmation can be made for Line 1.

The variance is indicative of the spread of the tracer response curve about the mean of the distribution and is rendered dimensionless by dividing by τ^2 (Equations 48 and 49).
The **dimensionless variance** (σ_θ^2) has an important relationship with the number (NTIS) of equivalent tanks in series in a TIS model and with the Peclet number (Pe) of a Dispersed Plug Flow (DPF) model (Eq 50) which is taken as pre-assessment for the DM model parameters.
For Test 1 and 2, **NTIS** values are low for Line 1, which demonstrates a water flow movement close to CSTR. The dimensionless variances are jointly presenting high values, reflective of flat bell shape TBC. For Lines 2 and 3, Test 1 and 2 present significantly different values of NTIS. Test 1 presents high NTIS values representative of a close to plug flow into the system, whereas Test 2 presents low values closer to a CSTR. Dimensionless variance is varying accordingly. The attention is drawn here as in the previous section has demonstrated that the TIS model poorly fits with the data set and is thus probably provides indicators poorly suited to describe the real tracer movement.

Previous Figures have shown that the TIS moment method had poor fits with the data, so the second calculation method using the Gamma distribution (added with the adjustment by the sum of squared errors). Results are presented in the second part of table 5.
New values of **tracer detention time** and **number of tank in series** are presented.
The new values of tracer detention time (τ) are systematically lower than those calculated by the TIS with Moment Method. They are showing the same trend of lower values with depth on Line 2 and 3 but not for Line 1. Results obtained from replication are significantly different.
NTIS values are no more integer values and are systematically highly increased in comparison to the NTIS values obtained by the TIS MM, except for Line 2 on Test 1. The mean value of all NTIS values is 22, 3, what is representative of a general good plug flow water movement within the bed and highly above the mean value of 11 NTIS summarised by Kadlec and Wallace (2009) for horizontal flow systems.

The last section of Table 6 present results from the hydraulic **dispersion model** with least squares method.
The dimensionless Peclet number (Pe) and dispersion coefficient (or dispersion number, D) are usual parameters from the chemical engineering theory, obtained from the Dispersed plug Flow model. The axial dispersion (D_L) and water velocity (v) are the two main parameters of the hydraulic dispersion model.

In chemical engineering, Levenspiel (1972) considers that the condition to apply the dispersed plug flow model is met when $D < 0{,}025$ what corresponds to about 20 TIS. Kadlec (1994) characterises surface flow systems as having large amount of apparent dispersion with $0{,}07 < D < 0{,}35$. Kadlec and Wallace (2009) consider that generally neither surface flow systems nor horizontal flow systems are within acceptable ranges.
The above NTIS found by the calibration of the TIS Gamma model provided an acceptable average value of 22,3 NTIS to apply the theory of Levenspiel.
Dispersion coefficients are generally low for Test 1, ranking from 0,01 to 0,09 with a few outlier values. Test 2 presents higher values than Test 1 but still representative of low dispersion phenomena.

The **axial dispersion** (D_L) presents highly significant different values between the two tests, with significant higher values for Test 2 (up to 11) on Line 1. Axial dispersion is high on Line 1 in comparison with Lines 2 and 3 and is also higher on the top layer than on the bottom of the bed. The DM explains the dispersion process by the combined action of turbulent and molecular transports and velocity variations. The velocity is the next parameter described.

A very interesting parameter given by the dispersion model is the **velocity of the flow**. It shows significantly that the flow velocity is systematically twice higher on the bottom of the bed at 55 cm deep than on the surface layer at 15 cm deep for Lines 2 and 3. On Line 1, the highest velocity is located in the middle layer of 35 cm deep.

The combination analysis of all these parameters provides the following indications:

- On Line 1 (located at 3 m from inlet) high values of axial dispersion and high flow velocities are representative of the good conditions for high dispersion processes. The low NTIS are concomitantly confirming that within this first section of the bed, the water movement is closer from the CST reactor than a plug flow movement. According to the vertical profile, the degree of mixing is increasing with depth: NTIS is high and axial dispersion value is low on the top layer of 15 cm deep, more representative of a plug flow reactor, where as NTIS is low and axial dispersion is high at 55 cm deep, representative of a mixing reactor. Significantly different flow velocities confirm that two different patterns are occurring at these different depths, with magnitude order varying from 1 to 5 and even almost 10.

- On Line 2 (located at 6 m from inlet) highly significant different results have been found between Test 1 and Test 2. General values of TIS and axial dispersion are nevertheless both attesting a general plug flow movement of the water within the bed. A closer look according to depth are indicative of a slightly more uniform mixing as a function of depth than Test 1, presenting higher NTIS and lower axial dispersion at 15 cm than 55 cm deep; and reversely the mixing effect is higher on the top layer for Test 2, as NTIS is lower and axial dispersion higher at 15 cm than 55 cm deep. Flow velocity is systematically higher for Test 2, which result from the higher inflow rate for Test 2 than Test 1 and presents the same effect of flow moving twice faster on the bottom of the bed at 55 cm deep than on the surface layer at 15 cm deep, with a magnitude of order of 2.

- On Line 3 (located at 11 m from inlet) significant differences are observed between Test1 and Test 2 but they nevertheless present the same trends. Test 1 presents a general plug flow water movement within the bed as NTIS values are high and axial dispersion is low. The general plug flow movement is more pronounced with depth than on the surface. For Test 2, the top and mid layers of 15 cm and 35 cm deep are presenting a poor plug flow movement, (closer to a complete stirred tank reactor) with low NTIS values and high axial dispersion; the bottom layer at 55 cm deep reversely present a good general plug flow behaviour with low values of axial dispersion. Flow velocities are for both tests showing that the mid and bottom of the bed (at 35 and 55 cm deep) conduct flows with similar velocities and the surface layer conduct flows much slower.

Usual studies of tracer tests have monitored tracer at the outlet only. The results at the outlet would lead us to consider the bed as having a general good plug flow water movement pattern within the bed, presenting high NTIS and low axial dispersion values. The observation that the NTIS value are higher than the average 11 NTIS summarised by Kadlec and Wallace (2009) for horizontal systems, would even drive us to consider our bed as a very good plug flow system. This is confirmed by tests on pilots, presenting similar results and values. Based

of small scale and full scale tests, we would consider that we can logically go on for a scale up design. As the difference of 2 days between the tracer detention time and theoretical calculated nominal detention time of the bed would be a design problem, we would only assume that we made a wrong assumption for porosity and would have no other explanation to understand this difference.
BUT, the study of points inside the full scale TW bed demonstrates that a high variability exists within the bed and cares must be taken when trying to qualify the hydraulic pattern of a system.

Briefly, Table 6 summarises the hydraulic behaviour of the system using both parameters from the TIS and DM theories.
It is clearly shown that two onsite tracer tests replicated with the same methodology, having an inflow higher of only 0,5 m^3/day for the second test in comparison with the first one, are producing significant different results.
Significantly data interpretation results are also produced by the TIS MM method or the Gamma Dis with SSQE method.
Tracer detention times are decreasing with depth on Lines 2 and 3 but increasing with depth on Line 1 for both tests. NTIS values are indicative of a general good plug flow water movement within the bed on Line 2 in the middle of the bed and are indicative of a mixed effect between PF and CST reactors on Lines 1 and 3.
The water velocity is significantly different with layers, being at the minimum, twice as fast on the bottom of the bed than on the surface layer.

The results from these parameters demonstrate that considering the bed as a homogenous media, having uniform flow through the whole cross sectional area is a wrong assumption. This means that the nominal detention time should be reviewed accordingly, as it is inversely dependent on the flow, as well as all indexes depending on nominal detention time (hydraulic efficiency, volumetric efficiency and dead zone index).
This is developed by the next section, after presentation of Table 7 with the system parameters for the small scale pilots under greenhouse.

II.4.3.5 *System parameters of the small scale pilots under greenhouse*

Table 7 is providing results issued from data of tracer tests on pilot scale TW under greenhouse. The first part of Table 7 is including Table 4, reminding data of the pilot systems.

The table firstly presents the results obtained for tracer detention time and the number of tank in series (N) calculated by the moment method for the TIS model (see equations 46 to 50). The variance is indicative of the spread of the tracer response curve about the mean of the distribution and is rendered dimensionless by dividing by τ^2 (eq. 49).
The dimensionless variance (σ_θ^2) has an important relationship with the number (N) of equivalent tanks in series in a TIS model and with the Peclet number (Pe) of a Dispersed Plug Flow (DPF) model (Eq 50) which is taken as pre-assessment for the DM model parameters.

The column entitled “mean” gives mean values for the feature of the pilot cells and the data. But for the application of Gamma Dis and DM models, the results presented in the “mean” column are not the average value resulting from Tests 1, 2 and 3 but the application of the models on data from the three tests combined, what provides values of detention time, NTIS and the Peclet number different that the average of Tests 1, 2 and 3.

Table 6 : Parameter values obtained by models and calculation methods for the full scale site of Nassogne

		Line		L1			∑	L2			∑	L3			∑	OUTLET	Pilots
		Depth		15	35	55	L1	15	35	55	L2	15	35	55	L3		3-12 mm
	Nom.	Units	Test														Tot Mean
TIS Model + MM method																	
Det. Time	τ	day	1	2,49	2,66	3,19	2,92	4,39	3,23	3,07	3,11	5,93	3,80	3,87	3,90	5,02	2,16
			2	7,26	5,22			6,11	4,49	2,41	3,31	7,54	6,05	3,89	4,87	3,96	
Dim.less Var.	σ_θ^2		1	0,330	0,750	0,765	0,770	0,050	0,071	0,024	0,081	0,016	0,091	0,410	0,741	0,211	0,036
			2	0,86	0,88			0,65	0,72	0,50	0,64	0,38	0,68	0,50	0,60	0,44	
NTIS	N		1	3	1	1	1	20	14	42	12	62	11	24	1	5	28
			2	1	1			2	1	2	2	3	1	2	2	2	
TIS GAMMADIS Model + SSQE Method																	
Det. Time	τ	day	1	1,81	0,80	1,41	1,83	4,21	2,94	2,77	3,10	5,82	3,55	3,68	3,63	4,53	2,18
			2	5,58	7,72			2,68	1,42	1,32	1,37	6,35	3,33	2,37	2,54	2,67	
NTIS	N		1	10,90	8,27	2,17	2,11	32,12	8,19	6,46	6,04	22,16	62,46	48,98	47,78	31,72	18,81
			2	1,07	0,71			6,57	16,81	32,59	21,05	3,28	3,92	21,67	11,39	21,17	
DM Model + SSQE method																	
Peclet Nr	Pe		1	19,80	14,55	2,34	2,21	64,24	14,38	10,92	10,08	44,32	124,91	97,95	95,55	63,43	35,62
			2	0,14	1,42			11,14	31,61	65,17	42,10	4,56	5,84	43,35	20,77	42,33	
Dispersion	D		1	0,05	0,07	0,43	0,45	0,02	0,07	0,09	0,10	0,02	0,01	0,01	0,01	0,02	0,03
			2	7,06	0,70			0,09	0,03	0,02	0,02	0,22	0,17	0,02	0,05	0,02	
Axial D.	D_L	m²/d	1	0,29	3,76	5,81	6,10	0,15	1,10	1,56	1,63	0,53	0,28	0,36	0,37	0,68	0,13
			2	11,09	3,80			1,59	0,87	0,45	0,71	7,65	9,32	1,32	2,61	1,80	
Velocity	v	m/day	1	1,95	9,33	5,65	4,50	1,51	2,59	2,90	2,69	2,04	3,18	3,10	3,15	3,13	1,51
			2	5,17	8,90			2,95	4,67	4,79	4,75	3,24	5,33	5,03	5,03	5,45	

Table 7 : Parameter values obtained by models and calculation methods for the pilots under greenhouse.

	Media size		3 - 5 mm				6 - 8 mm				8 - 10 mm				10 - 12 mm				3 mm
	Replication		Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	Test 1	Test 2	Test 3	Mean	12 mm
Parameters	**Form.**	**Units**																	**Tot Mean**
Length	**L**	**m**	3			3	3			3	3			3	3			3	3
Width	**W**	**m**	0,3			0,3	0,3			0,3	0,3			0,3	0,3			0,3	0,3
Depth	**D**	**m**	0,3			0,3	0,3			0,3	0,3			0,3	0,3			0,3	0,3
Volume	**L*W*D**	**L**	270			270	270			270	270			270	270			270	270
Porosity	**Meas.**	**-**	0,35			0,35	0,36			0,36	0,38			0,38	0,39			0,39	0,37
Nom. Vol.	**V*ε**	**L**	95			95	97			97	103			103	105			105	100
DATA																			
Inflow	**Meas.**	**L/day**	40,00	40,30	40,62	40,31	40,00	41,34	40,66	40,67	40,00	42,82	40,68	41,17	40,00	41,76	40,32	40,69	40,71
Nom.Det.Time	**V_n/Q**	**Day**	2,4	2,3	2,3	2,3	2,4	2,4	2,4	2,4	2,6	2,4	2,5	2,5	2,6	2,5	2,6	2,6	2,5
Tr. Peak Time	**τ_p**	**Day**	1,7	2,4	2,1	2,1	1,7	2,0	2,1	1,9	1,5	2,2	2,1	1,9	1,3	2,1	2,1	1,8	1,9
Recovery		**%**		94	99			94	95			95	99			92	96		
Hydr.efficiency	**$\tau_p/\ \tau_n$**	**%**	72	100	90	87	70	85	88	81	58	92	83	78	49	83	80	71	79
Morril Index	**t_{90}/t_{10}**	**MI**	1,5	1,3	1,4	1,4	1,5	1,4	1,7	1,5	1,7	1,4	1,3	1,5	2,2	1,4	2,0	1,9	1,6
dim less time	**t/τ_n**		3,5	3,6	3,6	3,6	3,4	3,6	3,5	3,5	3,3	3,5	3,3	3,4	3,2	3,3	3,2	3,2	3,4
TIS Model + MM method																			
Det. Time	**τ**	**day**	1,98	2,50	2,48	2,32	2,02	2,26	2,41	2,23	1,72	2,33	2,43	2,16	1,49	2,25	2,27	1,96	2,16
Dim.less Var.	σ_θ^2		0,139	0,017	0,142	0,097	0,153	0,099	0,120	0,074	0,129	0,050	0,125	0,120	0,103	0,073	0,082	0,090	0,036
Number of TIS	**N**		7	58	7	10	7	10	8	13	8	19	8	9	10	14	12	11	28
TIS GAMMADIS Model + SSQE Method																			
Det. Time	**τ**	**day**	1,90	2,35	2,30	2,30	1,97	2,18	2,26	2,13	1,69	2,30	2,33	2,17	1,49	2,23	2,16	2,04	2,18
Number of TIS	**N**		27,93	47,35	34,23	22,90	25,08	34,45	31,61	27,11	18,12	35,75	24,30	16,83	14,75	32,80	20,11	9,50	18,81
Vol Efficiency	**τ/τ_n**	**%**	80	100	99	98	81	93	95	89	66	96	92	87	57	88	83	79	89
Dead zone index	**$(\tau_n-\tau)/\tau_n$**	**%**	20	0	1	2	19	7	5	11	34	4	8	13	43	12	17	21	11
Short cut index	**$(\tau-\tau_p)/\tau$**	**%**	11	0	9	11	14	8	7	9	11	4	10	11	13	6	3	10	11
DM Model + SSQE method																			
Peclet Number	**Pe**		53,46	94,69	68,46	43,79	50,16	68,91	63,22	52,22	34,25	69,51	46,59	31,66	27,50	63,61	40,21	17,00	35,62
Dispersion coef.	**D**		0,02	0,01	0,02	0,02	0,02	0,02	0,02	0,02	0,03	0,01	0,02	0,03	0,04	0,02	0,03	0,06	0,03
Axial Dispersion	**D_L**	**m²/d**	0,09	0,04	0,06	0,10	0,10	0,07	0,07	0,09	0,18	0,06	0,09	0,15	0,25	0,07	0,12	0,32	0,13
Velocity	**v**	**m/day**	1,69	1,26	1,33	1,41	1,63	1,45	1,40	1,50	2,00	1,37	1,38	1,54	2,25	1,42	1,51	1,79	1,51

Tracer detention time (τ) is systematically presenting the same statistical significant decrease with the increase of media size, ranking from 2,31 for 3-5 mm to 1,96 for 10-12 mm media size.
The number of tank in series (N) identified is around 10 to 13 (except two very different value of 58 and 19) and are representative of a general good plug flow water movement within the beds.Graphs have shown that the moment method was poor to fit with the data, so the second calculation method results was added using the Gamma distribution (with the adjustment by sum of squared errors).
New values of tracer detention time and number of tank in series are secondly presented and are representative of the data sets, as displayed by previous graphs.

Tracer detention time values obtained by the **Gamma Distribution and SSQE** are systematically a little lower than the values issued from TIS and moment method.
Reversely, the **number of tank in series** (N), which is no more an integer value, are much higher than the number of tank in series calculated by the moment method. All the values obtained are very high and demonstrate the general good plug flow water movement obtained by the system pilot design. N is slowly decreasing with media size and remains higher than the average value of 11 TIS identified by Kadlec and Wallace (2009) for horizontal flow systems.
The same high statistical significant differences between test replications are observed as for all parameters previously discussed about the data.

Volumetric efficiency and dead zone index are two parameters which are calculated from ratio between nominal detention time (τ_n), and tracer detention time (τ)
Volumetric efficiency (e_v) = = τ/τ_n is a ratio used by the chemical engineering theory and represent the efficient volume involved in the flow (expressed here in percentage), whereas the **dead zones index** = $100(\tau_n-\tau)/\tau$ is issued from the hydraulics theory and represents the percentage of inactive zones within the bed.
They are complementary and present results in an opposite way.
Two important information provided by those two parameters are :

- the first test (Test 1) had high significant different values than the two next tests (Tests 2 and 3); it had much lower volumetric efficiency and a higher dead zone index, which demonstrate the correct assumption that the four pilots were not properly flushed before the start of the test one;
- when values of Test 1 are removed from the statistics, and Test 2 and Test 3 are compared for media sizes, a statistical significant difference is observed per media size, as dead zones are increasing and volumetric efficiency is decreasing as the media size increases.

Globally, the two indexes indicate that the volume of the bed involved in the water flow is better, creating lesser dead zones, with smaller media size (3-5mm) than with larger gravels (10-12 mm).

The **short cut index** is a ratio of tracer detention time and tracer peak time.
Short Cut Index = $\frac{\tau - \tau_p}{\tau}$ and presents the percentage of the flow which have preferential flow paths, with lower detention time and higher velocity. It ranks between 0 and 14 %, with an average value around 10 %. This is qualified as very good as it is not unusual to have this index having values above 75 % (Edeline, 1998). Test 1 presents higher values for this index, demonstrating that the beds were not properly flushed and preferential paths for the flow were more numerous than for Tests 2 and 3. The size of the gravel has no influence on short cuts.

The last part of the table is presenting parameter values calculated from the **dispersion model adjusted with SSQE method**.
The dimensionless Peclet number (Pe) and dispersion coefficient (or dispersion number, D) are usual parameters from the chemical engineering theory, obtained from the Dispersed Plug Flow Model. Axial dispersion (D_L) and water velocity (v) are the two main parameters of the hydraulic dispersion model.

In chemical engineering, Levenspiel (1972) considers that the condition to apply the dispersed plug flow model is met when D < 0,025 what corresponds to about 20 TIS. Kadlec (1994) characterises surface flow systems as having large amount of apparent dispersion with 0,07 < D < 0,35. Therefore, generally neither surface flow systems nor horizontal flow systems are within acceptable ranges (Kadlec and Wallace, 2009).
The results from Table 4 demonstrate that in our case, we fit within these ranges, as the mean value of all NTIS presented is 29 NTIS and the dispersion coefficient D is very low with 0,01 < D <0,03.

From the dispersion model, the **axial dispersion** D_L (or longitudinal dispersivity α_L) m²/day is low with 0,01 < D_L< 0,3. There is a statistical increase of the axial dispersion with the increase of media size as D_L has higher values with 10-12 mm gravels than with 3-5 mm gravels.
The **flow velocity** (v in m/day) also shows a significant increase with the increase of media size, ranking from 1,41 m/day with 3-5 mm gravels to 1,79 m/day with 10-12 mm gravels.

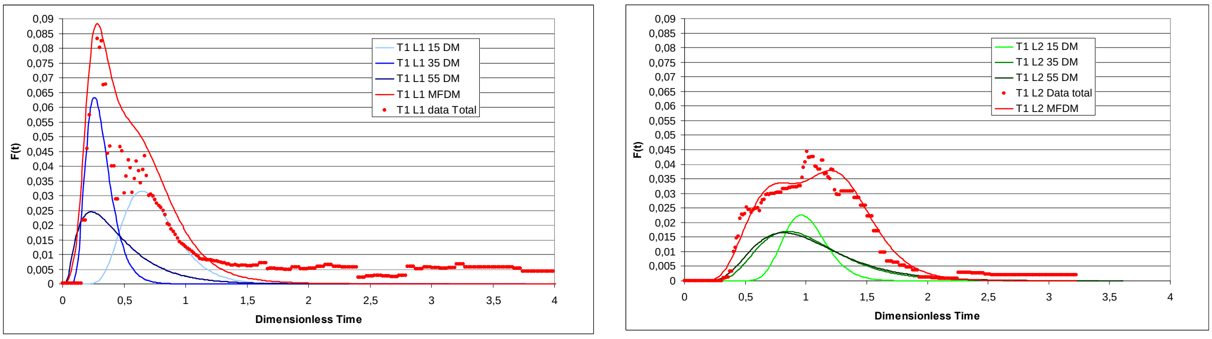
Briefly, Table 7 summarises the hydraulic behaviour of the pilot systems using both parameters from the TIS and DM theories.
The data of the systems confirm that the tests were well performed with good tracer recovery percentages and that they have good hydraulic efficiencies, double checked by the Morril index.
The TIS theory allows finding parameter values for the number of tank in series (NTIS) and tracer detention time (τ), especially when the Gamma Dis and SSQE methods are used to adjust them. The values of NTIS demonstrate that the hydraulic of the pilots are representative of a generally good plug flow movement within the beds. The tracer detention time allows the calculation of ratio showing a small percentage of dead zones, a high volumetric efficiency, with low short cut index, demonstrating that almost the whole bed volume is involved into the water plug flow movement through the beds.
The DM theory allows finding the velocity of the flow and proving the low axial dispersion occurring with the pilots.
Significant differences are pointed out between replication and the first test is significantly different than the two other replications.
The effect of the media size is significant on all discussed parameters. Larger gravels decrease the tracer detention time, the plug flow effect is lower with lower NTIS values as well as lower volumetric efficiency than for fine gravels. Reversely, larger gravels increase dead zones, dispersion coefficient and axial dispersion, and the velocity of the flow is faster with larger gravels than with fine ones.

II.4.3.6 Results of the reviewed full scale site in light with Multi Flow with Dispersion Model

The results from all the parameters presented above demonstrate that considering the bed as a homogenous media, having a uniform flow through the whole cross sectional area is a wrong assumption, especially for the full scale site experiment. The small scale pilots, due to their small dimensions can be considered as homogeneous.

This section develops the MFD model applied by Maloszewski et al. (2006) to the horizontal flow full scale site TW.
This model like any others, has specific boundary conditions which are : it assumes that (i) the tracer transport through the cell can be approximated by a combination of 1-D dispersion equations, here corresponding to our variable depths; (ii) that each flow paths is characterised by a specific volume rate, mean transit time and dispersion parameter; (iii) that the volume of immobile water within the wetland cell is insignificant; (iv) that there are no interactions between flow paths and that (v) the mass of tracer is injected is divided into several flow paths proportionally to the volumetric flow rate.

Figure 37 presents some of the graphical solutions and Table 8 presents the system parameters with reviewed data accordingly of nominal detention time and hydraulic indexes as they are depending on nominal detention time (hydraulic efficiency, volumetric efficiency and dead zone index).
Graphs (a) and (b) from Figure 37 displays with red dots, the total normalised concentration data (sum of the concentration from the three depths) on Line 1 and 2 respectively for Test 1. Blue lines for Line 1 and green lines for Line 2 are the DM solution found previously per depth on the same line. The red line is the combination of them into the MFDM.



(a) Test 1 line 1 (3 m from inlet)

(b) Test 1, line 2 (6 m from inlet)

Figure 37 : Graphical solution of MFDM equations for lines 1 and 2 from Test 1.

The Equation 36 is applied and integrated to obtain the total normalised concentration data per lines over length. Equation 37 is applied to obtain the fraction of water flux for each monitored layers. The solution of Equation 31 can be found having all the requested parameters: (i) the values of the volumetric rates along each flow path, Q_i,
(ii) the mass of tracer transported along each flow path given by tracer recoveries, M_i,
(iii) the dispersion values given by the DM, D_{Li}, and
(iv) the tracer peak times of each layers is measured by the experiments, t_{0i}.
The mathematical solution is graphically displayed by the red lines on Figure 37.

Having data on three depths, the assumption was made to have 3 flow paths (N). We can see with the graphical results that the assumption fits well with the model.

Table 8 provides the summary of parameters from the Dispersion and Multi Flow with Dispersion models. The hypothesis of flow rate equally shared over depth is reviewed according to the fraction of water fluxes found per flow paths. As the flow rates per layers are reviewed, the nominal detention time is thus reviewed accordingly. It allows presenting hydraulic parameters of volumetric and hydraulic efficiencies as well as dead zones index. The application of equation 43 allows calculating the height (thickness) of each layer. Equations 38, 39 and 40 are giving the volume of mobile water involved into the flow and with comparison to nominal volume, propose a filtering medium parameter which can be compared with porosity. Unfortunately, as for the second test (Test 2), the automated sampler from Line 1 at 55 cm deep collapsed, and as all calculation are made out of the total results per lines, the calculation can not be done for the Test 2 on Line 1.

The first parts of Table 8 are summarising data from previous analyses, parameters obtained by the Gamma TIS and the DM calibrated models. The column entitled “Total per Line” gives mean or total values per lines. For the application of models, the results presented in the “Total” column are not the sum of various depths but the result of calculation made for all the data per lines, what provides results different than sum or mean values presented per depths.

The first observation concerns the fraction of **water flux per layer**. Both tests (Tests 1 and 2) are providing non significant different results. It demonstrates that for both tests and along the whole bed, covering the set of data for the 3 lines, more than 50% of the flow rate occurs at the bottom of the bed, ranking from 59% up to 73%. The second major flow path is occurring in the middle of the bed with an average rate of 20% of the total inflow flowing through this mid layer on both Lines 2 and 3. Line 1 is providing rather different results with 30% of the flow on the top layer, only 10% in the mid layer and a short 60% on the bottom.

The calculation of the **thickness of the water flow** paths, based on the data monitored at 15 cm, 35 cm and 55 cm deep, globally shows that the first two paths have probably 15 cm height, but the bottom one is close to 35-40 cm. This could correspond to the first 15 cm of surface non saturated zones, partly colonised by the phragmites roots; the second layer from -15 cm to -30 cm correspond to the major root zones of the phragmites and the last layer from -30 to -65 cm correspond to gravels media not colonised by the roots of the phragmites. Breen and Chick (1995), U.S.EPA (2000), Wallace and Knight (2006) and Whitney et al. (2003) have already mentioned that limited root penetration was able to create preferential flow paths through the lower section of the gravel bed.

The **hydraulic efficiency** is an index giving information about the plug flow water movement and shows that the highest hydraulic efficiency is obtained for the middle of the bed at line 2, with a global value of 96%. It is also very high on Line 1 at surface and bottom depths with 91 and 82% respectively. Hydraulic efficiency rates are decreasing for the top layer from inlet toward outlet being of 91%, 60 % and finally 12% on lines 1, 2 and 3 respectively.

Volumetric efficiency and dead zone index are two parameters which are calculated from ratio between nominal detention time (τ_n), and tracer detention time (τ)

Volumetric efficiency $e_v = \dfrac{\tau}{\tau_n}$ is a ratio used by the chemical engineering theory and represent the efficient volume involved in the flow (%), whereas

the **dead zones index** = $\frac{\tau - \tau_n}{\tau}$ is issued from the hydraulics theory and represents the percentage of inactive zones within the bed.
The volumetric efficiency + dead zones are equal to the totality of the bed, or 100%. Results present on average values per line, a low volumetric efficiency at line 1 (44%), a good one at line 2 (95%) and a medium one at line 3 (61%). The dead zone index of 87% on line 1 at mid layer (35 cm deep) has to be related to the high axial dispersion values which were calculated, as it represents zones with low water movement, inducing higher dispersion process. According to Edeline (1992), it is not unusual to have dead zone index above 75%, we can thus consider that the general efficiency of the system and water volume involved in the flow as quite good. It is also demonstrated that with the second test, having a higher inflow rate (Q), the volumetric efficiency is lower than for first test. It means that the water volume involved in the flow is reduced when the inflow rate increases, with an accentuated preferential flow path along the bottom layer of the bed, as the hydraulic efficiency is there

The **volume of mobile water** (V_m) is also calculated by equation33 and 38; its ration with the nominal volume of the bed gives an indication to the **filtering media** of the system which can be compared to reviewed porosity.
The measured porosity of the gravels without plant was measured as 0,40. The value taken for calculation was 0,45 considering the increase of porosity added by the plant roots. The average filtering media value obtained after calculation is of 0,43 which demonstrates that the initial assumptions were justified. Nevertheless, once more this value varies considerably according to the location over length and with depth. Over length, the average value for Line 1 is of 0,39 and 0,40 for Line 2 but is dropping down to 0,27 for Line 3. With depth, average values are increasing.

II.4.3.7 Results and design explanation

In addition to previous discussions, two specific design features of the investigated system can contribute to explain some results:

1. The first specific design feature is the distribution inlet system, delivering the inflow into the TW bed. Inflow coming from the primary treatment sedimentation tank passes through an access chamber with the inflow rate measurement counter system; and is then poured into a distribution channel, which is small channel of 40 cm wide and 40 cm cm deep, parallel to the TW bed and having PVC pipes every 70 cm to deliver the inflow into the TW system. Those PVC pipes are installed with a strong slope (50°) in order to provide a flush impulse of the wastewater into the TW bed. They are also long in order to distribute the inflow down into the bed, close to the bottom of the wetland cell.
2. The second specific design feature is the collecting pipe which is a drain pipe located in the outlet zone and which is laid on the bottom of the bed. It is connected to a leg pipe which regulates the water level within the bed. Gerard (1992) has demonstrated that in flooded tanks, if the outlet structure is located on the bottom of the tank, it was creating a sucking effect inducing preferential flow paths.

These two specific design features probably highly contribute to deliver the water inflow directly down on the bottom of the bed, proximate to the outlet drain pipe inducing a creeping water movement along the bottom of the bed. The flush impulse created by the steep distribution pipes is also probably responsible of the good mixing occurring on Line 1 having high axial dispersion coefficient, and a low number of tanks in series (NTIS).

Table 8 : Multi Flow with Dispersion Model, inputs, revised data and hydraulic indexes

		Line		L1				L2				L3				OUTLET
	Nom.	**Form.**	**Depth**	**15**	**35**	**55**	**Tot L1**	**15**	**35**	**55**	**Tot L2**	**15**	**35**	**55**	**Tot L3**	
DATA			**Test**													
Total Volume		**L*D*W**		2,73	2,73	2,73	8,19	5,46	5,46	5,46	16,38	10,01	10,01	10,01	30,03	36,582
Nom. Volume	V_n	**V*ε**		1,17	1,17	1,17	1,37	2,35	2,35	2,35	7,04	4,30	4,30	4,30	12,91	14,633
Tracer Peak Time	τ_p		**1**	1,63	0,67	0,75	1,01	4,13	3,00	3,00	3,38	5,79	3,42	2,63	3,95	3,9
			2	1,00	0,63	-	0,82	2,17	1,25	1,25	1,56	1,96	1,96	1,17	1,70	2,2
TIS GAMMADIS Model + SSQE Method																
Det. Time	τ		**1**	1,81	0,80	1,41	1,83	4,21	2,94	2,77	3,10	5,82	3,55	3,68	3,63	4,53
			2	5,58	7,72			2,68	1,42	1,32	1,37	6,35	3,33	2,37	2,54	2,67
NTIS	N		**1**	10,90	8,27	2,17	2,11	32,12	8,19	6,46	6,04	22,16	62,46	48,98	47,78	31,72
			2	1,07	0,71			6,57	16,81	32,59	21,05	3,28	3,92	21,67	11,39	21,17
DM - MFDM Model																
Velocity	v		**1**	1,95	9,33	5,65	4,50	1,51	2,59	2,90	2,69	2,04	3,18	3,10	3,15	3,13
			2	5,17	8,90			2,95	4,67	4,79	4,75	3,24	5,33	5,03	5,03	5,45
Axial Dispersion	D_L		**1**	0,29	3,76	5,81	6,10	0,15	1,10	1,56	1,63	0,53	0,28	0,36	0,37	0,68
			2	11,09	3,80			1,59	0,87	0,45	0,71	7,65	9,32	1,32	2,61	1,80
Fraction of Q	p_i		**1**	0,30	0,09	0,59	0,99	0,16	0,20	0,65	1,00	0,04	0,23	0,73	1,00	1,00
			2					0,13	0,21	0,67	1,01	0,15	0,20	0,65	1,00	1,000
REVISED DATA																
Inflow	Q		**1**	0,72	0,72	0,72	2,17	0,72	0,72	0,72	2,17	0,72	0,72	0,72	2,17	2,17
			2	0,85	0,85	0,85	2,54	0,85	0,85	0,85	2,54	0,85	0,85	0,85	2,54	2,54
Inflow rev.	Q_i	p_i*Q	**1**	0,66	0,19	1,29	2,17	0,34	0,42	1,40	2,17	0,09	0,49	1,59	2,16	2,17
			2					0,33	0,53	1,70	2,55	0,38	0,51	1,66	2,54	2,54
Nom.Det.Time rev.	τ_{ni}	V_n/Q_i	**1**	1,78	6,08	0,91	0,63	6,85	5,52	1,68	3,25	49,63	8,78	2,72	5,97	6,74
			2					7,22	4,46	1,38	2,76	11,44	8,42	2,60	5,07	5,76
Thickness of layer	H_i		**1**	0,20	0,03	0,30	0,64	0,14	0,12	0,38	0,65	0,04	0,14	0,48	0,65	
			2					0,16	0,14	0,42	0,65	0,24	0,17	0,40	0,65	
INDEXES																
Hydr.efficiency	λ	τ_p/τ_n	**1**	91	11	82	62	60	54	56	96	12	39	97	66	58
			2					30	28	91	56	17	23	45	33	38
Vol eff	e_v	τ/τ_n	**1**	98	13	75	44	61	53	61	95	12	40	64	61	67
			2					37	32	96	50	56	40	91	50	46
Dead Zones	DZ	τ_n-τ/τ_n	**1**	2	87	35	66	39	47	39	5	88	60	26	39	33
			2					63	68	4	50	44	60	9	50	54
Volume of mobile water	V_{mi}	τ_{ni}/Q_i	**1**	1,19	0,15	1,81	3,16	1,44	1,25	3,88	6,57	0,50	1,74	5,83	8,08	15,56
			2					0,87	0,75	2,25	3,50	2,39	1,70	3,93	6,46	
Filtering medium	ε rev.	V_{mi}/V_{tot}	**1**	0,44	0,06	0,66	0,39	0,26	0,23	0,71	0,40	0,05	0,17	0,58	0,27	0,43
			2					0,16	0,14	0,41	0,21	0,24	0,17	0,39	0,22	

II.5 CONCLUSION

II.5.1 Hydraulic behaviour of the system

This chapter has devoted a large section to the understanding and study of hydraulic patterns of the investigated system. Very relevant and new insights have been achieved.

It has demonstrated that single tracer tests should not be considered as providing absolute information on the hydraulic behaviour. Replication can provide significantly different results, even for strictly controlled tests on pilots under greenhouse as well as for onsite tests. The information provided by tracer tests should be considered as a snapshot of a system, highly sensitive to spatial and temporal variations.

It has demonstrated that the scale up from pilot systems to large full scale wetland based on regular IN and OUT data can lead to major design errors in full scale systems, even if the global hydraulic patterns are well related.

Pilot tests have demonstrated that the media size (granulometry) of gravels has a significant influence on the tracer detention time and number of tank in series, volumetric and hydraulic efficiencies, dead zone index, axial dispersion and flow velocity. Larger gravels induce significant lower detention times, lower plug flow behaviour (lower NTIS) and lower volumetric efficiencies; they increase the dead zones index, the axial dispersion and the velocity of the flow.

Mathematical modelling has demonstrated that the TIS model solved by the moment method is a poor fit with data sets from onsite tests. The calibration of the model, improved by the Gamma Distribution, which allow considering the NTIS as a non integer value, and the sum of the squared errors (SSQE) fit almost perfectly with the data sets. The Dispersion model fits directly perfectly to the data sets. Simple calculation software unable to solve non linear equations, e.g. Excel from Microsoft Office, can solve the dispersion equation by assessing parameters with the connexion equations of dispersed plug flow model with dispersion model theories and using the Solver routine to apply the SSQE method.

The Gamma Dis TIS model provides accurate detention time and number of tank in series which are important for design considerations and the global evaluation of the hydraulic pattern (being closer to plug flow or complete stirred tank reactor). NTIS has demonstrated that the full-scale TW bed is presenting a good mixing condition close to the inlet, the middle of the bed has a general plug flow water movement behaviour which improves on the last portion of the bed, close to outlet.

The dispersion model provides the additional information of axial dispersion, which is another way to assess the mixing process within the TW bed; and the velocity of the flow, which is a parameter similar to the detention time of the system when it is related to design consideration. The axial dispersion is correlated with NTIS and presents high values where mixing is occurring, and low values where plug flow water movement is dominant. The velocity has proved the differential flow velocity within the bed according to depth, the flow velocity being always higher on the bottom of the bed.

The multi flow with dispersion model has demonstrated that in the case of the full scale site, several flow paths are occurring in parallel at different depths within the bed. The solution of

the model can provide the fractions of water flow rate per the layers identified. It has demonstrated that flow rates are not homogeneous with depth as the total flow is not delivered homogeneously over the cross sectional area of the bed. A strong preferential flow path drives 60% of the total inflow rate along the bottom of the bed and only 20% through the mid layer.
This reviewed inflow rate distribution has allowed the reviewing of nominal detention time and reviewing of hydraulic indexes.

The indexes have provided the information that the horizontal flow bed has general good hydraulic and volumetric efficiencies, with relatively low dead zones, but it cannot be classified as an ideal flow system.
The reviewed porosity or filtering medium is providing a general value of 0,43 which is very close to the 0,4 general value for unplanted gravels and the 0,45 we initially assumed for planted gravels. The filtering medium index is higher on the bottom of the bed than on the surface layers, which demonstrates that the roots complex is reducing the global porosity.

II.5.2 Design consideration

The hydraulic pattern has been described as a rather good plug flow water movement as expected by the design. The general hydraulic flow pattern can hardly be improved as the TW beds have:

- straight walls in concrete blocks,
- width : length ratio (W:L) higher than 1:3, and
- the distribution channel to distribute uniformly the inflow through pipes every 70 cm along width.

Nevertheless, the following advises on future design can be suggested in order to reduce the fractions of flow paths.

1. Reduce the depth of the bed, as partly investigated by Garcia et al. (2004b), in order to avoid porosity gradient induced by the presence of roots or not. This advice takes only into account hydrological concerns but also plant biology aspects have to be jointly investigated to develop this first proposal.

2. Use different media size layers: fine gravels could be set on the bottom of the bed in order to reduce the flow velocity and larger gravels on surface layers in order to increase the porosity and the flow velocity, which would have a general better homogeneous result over depth.

3. Reduce the length of distribution pipes in order to distribute the flow closer from the surface than the bottom of the bed.

4. Set 2 drain pipes to collect the outflow: one could be let on the bottom of the bed and be used only in case of emptying the bed, and a second one could be set within the gravels of the collecting zone, located over width closer to surface than the bottom of the bed in order to reduce the sucking effect and creeping flow along the bottom of the system.

CHAPTER III

Water Quality Parameters and Treatment Processes

III. CHAPTER III : WATER QUALITY PARAMETERS AND TREATMENT PROCESSES

III.1 INTRODUCTION

Treatment wetlands (TW) rely on a variety of physical, chemical and biological processes to reduce, transform and remove pollutants from the influent. Once wastewater enters a TW, the major factor acting in suspended solids removal is the drop in the inflow velocity which settles out suspended solids. The major factor driving COD and BOD removal is the availability of oxygen which impacts the chemical transformation of organic matter (COD, BOD) and nitrogen. Other contaminants like phosphorus, trace metals and pathogens are reduced by diverse treatment mechanisms (such as adsorption and precipitation).
This Chapter III focuses on removal mechanisms for organic compounds (BOD, COD) and suspended solids (TSS), which are the three water quality parameters ruled by Walloon standards for small scale wastewater treatment systems (< 10.000 PE), which have to be respected before discharge of any treated wastewater effluent into the environment.
About other water quality parameters, the next and last Chapter IV develops the nitrogen nutrient cycling occurring within the treatment wetland related with microbial activity, as well as some aspects of phosphorus removal. Trace metals, which are usually out of concerns for domestic wastewaters, and pathogens will not be investigated by this work.

III.2 THEORETICAL BACKGROUND

III.2.1 Walloon legislation

In respect to the Water Frame Directive and the European Directive on urban wastewater treatment, the following norms are applied in the Walloon region for treated wastewater discharge in the environment for small scale wastewater treatment plants, based on punctual sampling. Results are presented in comparison with the Walloon regional standards.

Treatment plant size	BOD_5 (mg O_2/L)	COD (mg O_2/L)	Total Suspended Solids (mg/L)
< 20 PE	70	180	60
20 to 100 PE	50	160	60
> 100 PE	30	125	60

III.2.2 Organic compounds degradation

Wastewater contains a wide range of organic carbon compounds and other oxygen demanding substances. These contaminants vary from being readily biodegradable to highly refractory, and are present in both soluble and particulate forms (Wallace and Knight, 2006). The oxygen demand resulting from organic matter is measured in the laboratory by COD and BOD_5 which are lumped parameters based on specific laboratory procedures; they measure the aggregate total of a variety of organic compounds in the wastewater.

Chemical Oxygen Demand (COD) is the amount of oxygen required to chemically oxidize organic matter. 5-Day Biochemical Oxygen Demand (BOD_5) is the amount of oxygen used by microbial organisms to degrade organic matters. Ultimate Biochemical Oxygen Demand

(BOD_u) is the total oxygen demand that would be exerted by the organic matter if there were enough contact time to biologically degrade all the materials.

These organic compounds have individual degradability, based on their chemical bonds (e.g. glucose is more easily degraded than halogenated compounds) and the size of the particles. Some of the compounds require more than five days to be degraded and they are not measured in BOD_5 test. The COD test oxidizes all the organic matter, including those which could not be degraded by microbial fauna. As a result, COD > BOD_u > BOD_5. As all those parameters measure the same organic matter, BOD_u is a subset of COD, and BOD_5 is a subset of BOD_u (Wallace and Knight, 2006). As the flow goes through the TW the remaining organic compounds become more difficult to degrade and the coefficient rate for BOD removal decreases (Kadlec, 2003). This also implies that the organic matter which enters the TW does not have the same chemical composition as the remaining non-degraded organic matter which is contained in the effluent of the TW system (Wallace and Knight, 2006).
Settleable organics are rapidly removed in wetland systems by deposition and filtration (Vymazal et al., 1998). Organic compounds are degraded aerobically as well as anaerobically. The oxygen required for aerobic degradation is supplied directly from the atmosphere by diffusion or oxygen leakage from the macrophyte roots into the rhizosphere (Brix and Schirup, 1990). Aerobic decomposition tends to be rapid, with minimal by-products of organic matter accumulation (named "sludge" or "biomat" or "biosolids") within the TW. The biosolids which are generated internally within the system consist of plant detritus material (and associated microbial and fungi fauna) and microbial film present on the bed media particles (Kadlec and Wallace, 2009). Anaerobic conditions prevail when the oxygen transfer rate cannot satisfy the oxygen demand. Anaerobic conditions operate slower than aerobic ones and result in greater accumulation of organic matter within the TW.

III.2.3 Aerobic and anaerobic degradations

Microbial cells using carbon as synthesis source are classified into two groups:
- Heterotrophic bacteria are using organic carbon,
- Autotrophic bacteria are using carbon dioxide (CO_2).

The type of decomposition (aerobic or anaerobic) is determined by the balance between the organic matter loads (internal and external) and the oxygen transfer rate of the wetland. If the oxygen transfer is high enough to more than satisfy the oxygen demand exerted by the organic matter loads, aerobic conditions will prevail (redox potential > 300 mV) (Wallace and Knight, 2006).

<u>Aerobic degradation</u> of soluble organic matter is governed by the aerobic heterotrophic bacteria (Mitsch and Gosselink, 2007) according to the following equation of respiration:

$$C_6H_{12}O_6 + 6O_2 \Rightarrow 6CO_2 + 6H_2O \qquad \text{(Eq. 51)}$$

Where *($C_6H_{12}O_6$)* represents the soluble organic matter (carbohydrates)

The autotrophic group of bacteria which aerobically degrade compounds containing nitrogen are called nitrifying bacteria, the process is called ammonification and will be presented in Chapter IV.
Both autotrophic and heterotrophic bacteria groups consume organics within TW systems but the metabolic rate of the heterotrophic groups is faster than the metabolic rate of the autotrophic group, which means that they are the main responsible group for BOD reduction

(Cooper *et al.,* 1996). If the oxygen supply is not limited, aerobic degradation will be governed by the amount of available organic matter (Vymazal *et al*., 1998a).

Anaerobic degradation is a multi-step process that occurs within constructed wetlands in the absence of dissolved oxygen (DO) (Cooper et al., 1996). The process can be carried out by either facultative or obligate anaerobic heterotrophic bacteria. In the first step the primary by-product of fermentation are fatty acids (acetic, Eq.52), butyric and lactic acids (Eq.53), alcohols (Eq.54) and the gases CO_2 and H_2 (Eq.52 and 54) (Vymazal *et al*., 1998b).

$$C_6H_{12}O_6 \Rightarrow 3CH_3COOH + H_2 \quad \text{(acetic acid)} \qquad \text{(Eq. 52)}$$
$$C_6H_{12}O_6 \Rightarrow 2CH_3CHOHCOOH \quad \text{(lactic acid)} \qquad \text{(Eq. 53)}$$
$$C_6H_{12}O_6 \Rightarrow 2CO_2 + 2CH_3CH_2OH \quad \text{(ethanol)} \qquad \text{(Eq. 54)}$$

Acetic acid is the primary acid formed in most of the flooded soils and sediments. Strictly anaerobic sulphate-reducing (Eq.55 and 56) and methanogens (Eq. 57) bacteria utilize the end-product of fermentation to produce sulphur and methane according to the following sulphate reduction and methanogenesis equations:

Sulfate reduction

$$\underset{\text{(lactate)}}{2CH_3CHOHCOO^-} + SO_4^{2-} + H^+ \Rightarrow \underset{\text{(acetate)}}{2CH_3COO^-} + 2CO_2 + 2H_2O + HS^- \qquad \text{(Eq. 55)}$$

$$\underset{\text{(acetate)}}{CH_3COO^-} + SO_4^{2-} + 2H^+ \Rightarrow 2CO_2 + 2H_2O + 2HS^- \qquad \text{(Eq. 56)}$$

Methanogenesis

$$\begin{aligned} &4H_2 + 2CO_2 \Rightarrow CH_4 + 2H_2O \\ &\underset{\text{(acetate)}}{CH_3COO^-} + 4H_2 \Rightarrow 2CH_4 + H_2O + OH^- \end{aligned} \qquad \text{(Eq. 57)}$$

The acid-forming bacteria are fairly adaptable but the methane-formers are more sensitive and will only operate in the pH range of 6,5 to 7,5. Over production of acid by the acid-formers (Eq.52, 53 and 54) can rapidly result in the stepped chain of decreasing pH, stopping the action of methanogens bacteria and producing of odorous compounds. Anaerobic degradation of organic compounds is much slower than aerobic degradation; however, when oxygen is limiting (i.e., at high organic loading) anaerobic degradation will predominate (Cooper *et al*., 1996).

Due to the low oxygen transfer, most of the horizontal flow TW systems are processing with anaerobic degradation of the organic matter, unless the system is very lightly loaded. Most of the oxygen present is in the upper portion of the water column, close to the surface and plant roots. Garcia *et al*., 2004b reported higher BOD removal efficiencies with shallower depth beds of 0,27 m than with bed of 0,5 m deep. Patents were developed in the USA and Canada to provide oxygen to these type of systems. One was developed and patented by Wallace (2001) and is a horizontal flow TW system with forced aeration from bubblers set on the bottom of the bed. Another one is patented by Austin *et al.* (2002) for the fill and drain systems (reported on Chapter I) where the water fluctuation within beds can support aerobic degradation of the organic matter.

It is apparent that TW provide a spectrum of potential pathways for the utilisation of organic compounds, but sufficient information is not available to quantify both of the complex chemistry and the spatial distribution of chemical compounds. Therefore, the interactions must be described via correlations and rate equations, which are supportable by wetland performance data (Kadlec and Wallace, 2009).

III.2.4 Suspended solids

The suspended solids entering a TW may display widely varying characteristics, according to the source involved. Domestic wastewaters at all pre-treatment stages contain suspended materials that are primarily organic. Runoff waters, agricultural or urban stormwater can contain high proportion of mineral matter. Other specific sources can carry colloidal materials (for instance, milking parlors). The two principal ways of describing solids are the soil type and the size distribution. The soil fractions are organic, clay, silt and sand. The Volatile Suspended Solids (VSS) is taken to be a measure of the organic fraction and the Non Volatile Suspended Solids (NVSS) is assumed to be the mineral remaining fraction of the overall Total Suspended Solids (TSS) (Kadlec and Wallace, 2009). In total : TSS = VSS + NVSS.

Horizontal Flow TW systems are mostly used for the secondary or tertiary treatment of wastewaters. The majority of settable solids are thus expected to be removed in a mechanical pre-treatment unit (sedimentation or Imhoff tanks) before the discharge of wastewater into the TW system.

No generalization of soil type and size distribution of TSS can be made across the spectrum of TWs and source waters. It is relevant to mention that organic materials can be subject to decomposition after deposition, and mineral fractions can accrete but without decomposition (Kadlec and Wallace, 2009).

In all types of constructed treatment wetland systems, most of the solids of wastewater origin are filtered out and settled within the first few meters beyond the inlet zone. The accumulation of trapped suspended solids is a major threat for long-term performance of some systems, especially those with subsurface flow, which can be clogged by suspended solids (Vymazal *et al.*, 1998b).

The slow flow velocities and abundance of interception surfaces that exist in TW systems contribute to the following suspended solid removal mechanisms which are: sedimentation, aggregation, filtration and interception.
<u>Sedimentation</u> (Discrete particle settling): the largest and heaviest particles will predominantly settle out in the inlet region of the wetland. Smaller and less dense particles will require additional detention time. Sedimentation s promoted by the bed media (for horizontal flow systems) or by the plant detritus (for surface flow systems) by reducing water column mixing and re-suspension of particles. Discrete particle settling rates can be calculated by the Stoke's Law and appropriate drag coefficients (Kadlec and Knight, 1996). All the particles associated with the influent wastewater are settled out generally within the first 5% of the horizontal flow wetland bed (Puigagut *et al.*, 2006).
<u>Aggregation</u> (Flocculation): Aggregation is the process by which particles naturally tend to flocculate. The aggregation degree depends on the balance between particle attraction, which is controlled by surface chemistry characteristics and the strength of the shear forces on the particles. Shear forces within the water column are a function of mixing and turbulence. Bed media (for HF TW systems) and plant detritus (for SF TW systems) within the water column

greatly reduce shear forces, resulting in enhanced flocculation and settling performance (Wallace and Knight, 2006).
Filtration: the principal mechanisms of granular bed filtration include (Metcalf and Eddy, 1991; Crites and Tchobanoglous, 1998):
- inertial deposition where particles moving fast enough that they impact the bed particles rather than being swept past by the flowing water,
- diffusional deposition where random processes at either micro scale (Brownian motion) or macro-scale (bioturbation, burrowing rodents, e.g.) which move a particle to an immersed surface,
- flow line interception where particles moving with the water and avoiding head-on collisions, but passing close enough to graze the stem and its biofilm, and sticking.

The size range of the bed media in TW systems spans the dominant factor of these mechanisms.
Interception: the smallest particles (e.g. bacteria, colloids) may not aggregate enough to settle out in the detention time available in the TW system. For these particles, the only removal mechanism left is the interception by the biofilm growth located on the wetland media bed (for media based beds) and on emergent wetland plants and detritus (for non-media based SF TW systems).

As a result of these removal processes (and similar to organic compounds measured by BOD), the TSS of the effluent usually is not related to the solids entering the system but represents the solids produced by the decomposition and resuspension of biomat particles within the horizontal flow bed (Kadlec and Wallace, 2009).

An additional removal mechanism is chemical precipitation. Generally, accumulation of chemical precipitates does not occur at a rate significant enough to impact the hydraulic conductivity of a horizontal flow TW. This mechanism is worth being mentioned as it is the basis of specific wastewater treatments of mainly mine wastes, using the sulphate-reducing process to precipitate Copper, Nickel, Zinc, Chromium and other metals (i.e. Pollman *et al*., 2008; Veena Devi *et al*., 2008; Mayes *et al*., 2008; Vymazal *et al*., 2006; Garcia *et al*., 2006; Paredes *et al*., 2006).

III.2.5 Clogging process and effect of biomat on hydraulic conductivity

Microbial biofilms form in response to both particulate and soluble organic loading rates. The biofilm entraps organic and inorganic solids (Winter and Goetz, 2003) and forms the composite materials which is named “biomat”, or “biosolids” or “sludge”. The biomat can either be attached to any support (media, plants or roots), or be present as colloidal material within the media pores. If the biomat formation is over abundant, it fills the pore spaces between bed media particles. When the media particle sizes are small, void volumes are small and flow paths through the bed media can became blocked and no more connected, inducing a surface flow in systems originally designed for subsurface flow. The bed is then considered as clogged (Wallace and Knight, 2006). The reason for the reduction of pore volumes leading to clogging can be complex and there is no simple cause-and-effect chain. Winter and Goetz (2003) made the following literature review which discusses several possibilities:
- Blocking of the pore space by deposition of organic and inorganic particles (Hill, 1983; Borner, 1992; Ellis and Aydin, 1995; Müller and Lützner, 1999; Nguyen, 2000);
- Precipitation and deposition of CaCo3 (Baveye *et al.*, 1998; Blazejewski and Murat-Blazejeweska, 1997);
- Clogging of the pores from microbial biomass (Kristiansen, 1981; Ronnern and Wong, 1994; Bihan and Lessard, 2000);

- Root influence (Böener, 1992);
- Mechanical compaction of the soil (in soil-based TWs)(Otis, 1985; Kristiansen, 1982);
- Entrapped gas blocking the pores (Rice, 1974; Soares *et al.*, 1989) (Winter and Goetz, 2003);

The clogging phenomenon occurs at the filter surface or at soil layer changes (*in* Winter and Goetz, 2003: Otis, 1985; Rice, 1974; Thomas *et al.*, 1966; Siegrist and Boyle, 1987; Platzer and Mauch, 1997).
Biomat formation is the more active at the inlet and distribution zone of the TW where the organic load is the highest (Ragusa *et al.*, 2004); it reduces the pore volumes which impacts on the hydraulic conductivity (Zhao *et al.*, 2004). Organic matter is removed as wastewater flows through the TW, resulting in declining biomat growth. At the outlet or collecting zone of the TW, as only small quantities of organic matter are available to the microbial biofilm, and biomat formation is minimal (Wallace and Knight, 2006). The biomat is thus non equally distributed over length within a horizontal flow TW system and it results in a non uniform hydraulic conductivity throughout the bed as shown in Figure 38.

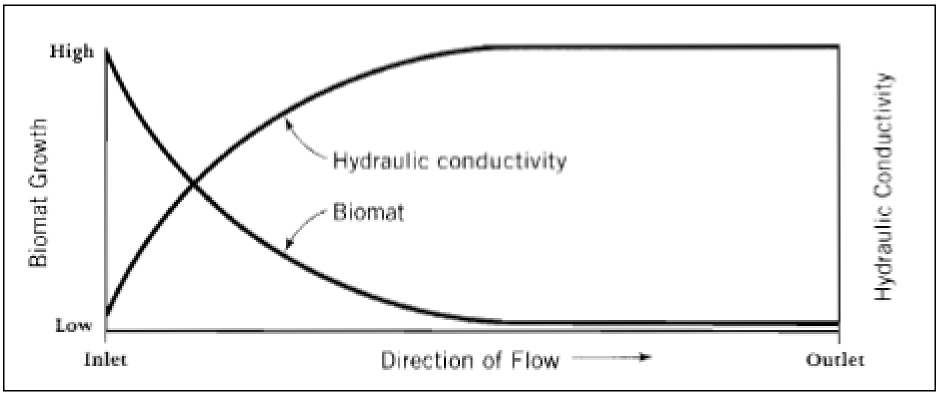
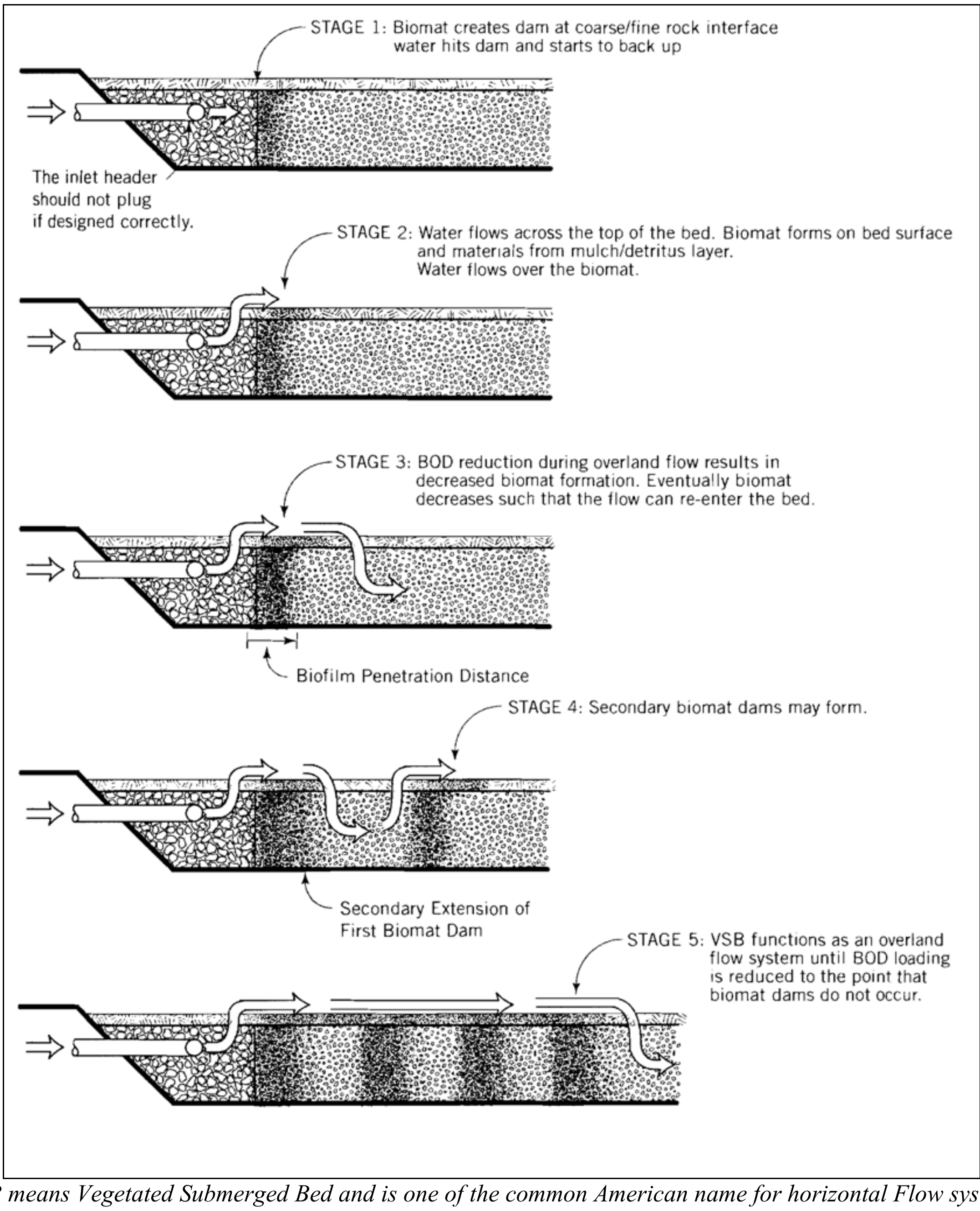


Figure 38: Relationship between hydraulic conductivity and biomat formation (Wallace and Knight, 2006)

Most organic matter is removed within the first few meters of a HF TW system (Wallace and Knight, 2006) and this zone of heavy biosolids accumulation is termed the “biosolids clogging distance” or “biomat penetration distance”. The biomat penetration distance is analogous to the clogging mat that develops in soil infiltration systems that treat septic tank effluent (U.S.EPA, 2002).
When the media size is small, the surface area available per unit of flow path is higher; and according to organic loading, more microbial biofilm can form. The fine media size creates small pore volumes and the biomat is thus more efficient at trapping organic and inorganic solids. But the resulting biomat can fill the pore spaces, the hydraulic conductivity will drop and the wastewater will flow up on surface.
Reversely, when the media size is coarse, the surface area available for biomat formation per unit of flow path is low, less biomat can form and thus void pore volumes cannot be filled. The flow path through the media can thus still perform without surfacing flow. The biomat penetration is longer for coarser media than for small media but coarse media decrease the overland flow potential of the system.
If biomat plugging occurs, the hydraulic conductivity is no longer a function of the size of the bed media; instead, the hydraulic conductivity is controlled by the biomat which progressively leads to a clogging failure of the TW, ending up functioning as a surface flow

TW system (Wallace and Knight, 2006). This phenomenon of biomat formation linked with changes in media size and surfacing flow is explained by Figure 39.
The traditional restoration procedure to reverse bed clogging is to remove the clogged bed media and replace it by clean media. Another restoration option for gravel-based systems is to remove the clogged media, wash it and return it to the wetland bed (Cooper *et al.*, 2006), although this technology is still being developed (a "washing gravel machine" is in use in the UK) and requires care not to damage the liner during the excavation/replacement process. Recently, alternate approaches are testing the chemical oxidation of organic matters by means of hydrogen peroxide (H_2O_2). Nivala and Rousseau (2008) report substantial clogging reduction and also that H_2O_2 did not seem to have a negative effect on plants and microbial biofilm.



(VSB means Vegetated Submerged Bed and is one of the common American name for horizontal Flow systems)

Figure 39: Stages of clogging in Horizontal subsurface flow TW systems (Wallace and Knight, 2006)

III.2.6 Representing treatment performances and modelling

Wetlands are "open" systems influenced by a variety of environmental factors, which makes them more complex than other biological treatment reactors (activated sludge, trickling filters) described in the environmental engineering literature. Some authors, like Burgoon *et al.*, 1999; McBride and Tanner, 2000; Langergraber, 2001; Rousseau *et al.*, 2005; Wu and Huang, 2006, have made the attempt to adapt models from these other technologies to treatment wetlands to predict degradation of pollutant through the TW and to allow engineering design.

The most common and useful graphical representation of results of BOD, COD and TSS are presenting output concentration versus input concentration or input areal loading. In/out concentration graphs essentially extend the idea of percent removal and it is useful to obtain a first estimation of the potential of a TW to reduce a particular contaminant. Nevertheless, as it does not include any information on the detention time or hydraulic loading, it provides no value in sizing the wetland.

III.2.6.1 The k-C model*

Previously, old kinetic removal models (e.g. the *k-C** model) based on the plug flow models were developed and synthesised by Kadlec and Knight, 1996. They knew that plug flow did not apply to the reality of TW, especially for low loaded systems but they reasoned that the plug flow assumption would be conservative. It is now known that this assumption is wrong, even if the plug flow model is often an acceptable interpolator on existing data sets (Kadlec, 1999).
The *k*-value is easily calculated for each pair of input-output concentrations according to the following equation:

$$k = q \ln\left(\frac{C_i}{C_0}\right) \qquad \text{(Eq. 58)}$$

Where, k = first order areal constant (m/d)
q = hydraulic loading rate (m/d)
C_i = inlet concentration (mg/L)
C_0 = outlet concentration (mg/L)

The average *k*-values for BOD in HF TW systems so calculated average 32 m/yr, but this model fails at predicting concentrations which scatter randomly within observed concentration ranges. On 2000, Kadlec explained the inadequacy of those models and developed the *P-k-C** model which is now considered as the preferred design approach. Only the latter is developed, briefly explained and applied by this work.

III.2.6.2 The P-k-C model*

The concepts of local pollutant reduction are blended with hydraulic considerations, as developed in Chapter II, but also environmental and ecosystem features. The pollutant removal model is developed based on the Tank-In-Series (TIS) model, developed in Chapter II. HF TW systems with continuous flow are not spatially uniform and the TIS model decomposes the system into a number (*N*) tanks in series, where contaminants are removed in each of the internal compartments (see Figure 4 on Chapter II). For the entire sequence of tanks, the mass balance is given by Eq. 59, where it has been presumed that the k-constant rate does not vary with the time of exposure to the wetland. This is known not to be the case typically for mixtures of contaminants such as BOD where removal rates slow down as a result of the treatment process (Kadlec and Wallace, 2009).

$$\frac{(C - C^*)}{(C_i - C^*)} = \left(1 + \frac{k\tau}{Nh}\right)^{-N} \qquad \text{(Eq. 59)}$$

Where,
- C = predicted concentration at the outlet (mg/L)
- C_i = inlet concentration (mg/L)
- C* = background concentration (mg/L)
- *k* = degradation rate constant (m/d)
- τ = detention time (days)
- *N* = number of TIS
- *h* =water depth in the TW (m)

A longitudinal concentration profile can be derived from this model and provides the following result:

$$\frac{(C - C^*)}{(C_i - C^*)} = \left(1 + \frac{k\tau y}{Nh}\right)^{-N} \qquad \text{(Eq. 60)}$$

Where, *y* = fractional distance through the wetland, dimensionless

This profile is theoretically a smoothly decreasing concentration curve that starts at the inlet concentration and levels off at a plateau value of C*. The parameters of this model were determined in this study by the longitudinal transect data monitored over 2 years, which provided C and *y* values.

III.2.6.3 The relaxed TIS concentration model

Equations 59 and 60 from the *P-k-C** model represent the reduction of a single compound based on transit through a TW. However, many pollutants are in fact mixtures and water quality parameters are measured by procedures which lump individual chemical compounds into an overall or total concentration, such as BOD and TSS (Kadlec and Wallace, 2009). The individual components of such mixtures are degraded or removed at different rates, and present a distribution of rate constants across the various mass fraction of the mixture (Crites and Tchobanoglous, 1998; Shepherd *et al.*, 2001; Kadlec, 2003). As wastewater containing such mixtures passes through the TW, its composition changes because different fractions of the mixture are reduced at different rates, and the mixture become "*weathered*"(Kadlec and Wallace, 2009). Each fraction of the lumped material will possess its own *k*-value and there is a distribution of k-values designated by *f(k).* This *k*-value frequency distribution across the mass fractions of the lumped material is termed the *k*VD.

f(k) dk = mass fraction of material with rate constant in the range *k* to *k* + *dk*.

It has been noted that observed weathering behaviour in real TW situations may be represented by the TIS model and Equation 59, wherein the parameter values are relaxed to be fitting parameters (Kadlec, 2003).
This relaxed TIS concentration model is defined by the following equation:

$$\frac{(C_0 - C*)}{(C_i - C*)} = \left(\frac{1}{(1 + k/Pq)^P}\right) \quad \text{(Eq. 61)}$$

Where,
C* = background concentration (mg/L)
C_i = inlet concentration (mg/L)
C_0 = outlet concentration (mg/L)
k = apparent TIS rate constant (m/yr)
P = apparent number of TIS
q = hydraulic loading rate (m/yr)

The apparent number of *P*TIS can vary but is always ≤ *N*TIS.
The number of *N*TIS is related to hydraulic water fraction and detention time, where the apparent number of *P*TIS is related to the organic fractions and detention times. In this regard, N is always higher than P as organic materials are moving slower inside TW than wastewater: *N*TIS ≥ *P*TIS.

Kadlec and Wallace (2009) report that there is not much difference in concentration reduction among the various models for low removal rates, e.g. when removal is lower than 50%. However, there is a large difference when predicted removals are in the high range, and there can be a factor of 10 in concentrations when expected reductions are above 99%.

In the results section, data from the monitored TW of Nassogne has been used to calibrate the *P*TIS model, which allows determining the frequency distribution of *k*-values. For tracer test experiments, one cell was isolated from the system and received only rainwater during more than one year. The BOD measure in this cell after one year was taken as C*, even if inter-annual and inter-system variability have been demonstrated to occur (Kadlec and Wallace, 2009).

Nevertheless, when we talk about design, it is necessary to understand the components of the wetland environment and layouts that contribute to either high or low *k*-values, as the distribution observed is too broad to give confidence in design for a mean *k*-value.

This sector is clearly one of the fields which is necessary to produce accurate TW designs, and where comprehensive data are still missing. Future investigations still have to be made to provide real data of *k*-values according to pollutants, TW design, loading rates, etc. and enhance the understanding of degradation processes.

In this regard, this study is unique to apply, use and calibrate the existing modelling with onsite data. Especially as data are distributed over length and with depth within one TW and monitored over a long enough period to account for seasonal effects, start up effects and others. This study is thus a tool key to the building of a *k*-values data base which still has to be developed in order to have appropriate designs for TW in the Walloon Region of Belgium.

III.3 EXPERIMENTAL METHOD

III.3.1 Sampling layout

The monitored constructed wetland has been in operation at a rural guesthouse with a café-restaurant since 2003. The detailed description of the beds has been provided in chapter I. The main aspects important are summarized here:

- It is a horizontal flow system;
- Stones of 4-12 cm are used in the distribution and collection zones, followed for the distribution zone and preceded for the collection zone with large gravels of 10-20 mm and pea gravels of 3-8 mm in the treatment zone;
- It is composed of two cells in parallel, each of which is 13,4 m long, 4,2 m wide and 0,8 m deep;
- Water depth is 0,60 m;
- The two beds are planted with *Phragmites australis sp*.

There have been some operational challenges encountered in the frame of this study resulting from the low loading rate applied on the CW, due to management troubles for starting of the guest house and the high variability of the attendance depending on the tourist season and meteorological weather. Water samples were collected once or twice a month inside the TW, influent and effluent samples were also collected. Water samples were monthly collected during winter and spring season inside the wetland due to the low load applied, as the rural guest house was not much visited during the low tourist season. Sample distribution was spread between 4 points over width, with 2 points in each cell, and 3 points along the flow path of 3 m , 6 m and 11 m from the inlet and at three depths, 0,15 cm , 0,35 cm and 0,55 cm from the surface.

One sample collected water was of one depth over one distance, and was a mix of four points over width, having two points in the two cells. Influent and effluent samples from the wetland were also collected, 1 at the entry and 1 at the exit (a mixed sample of each cell). Each collecting campaign totalled 11 samples.

The sampling campaign started on June 2006 and ended on October 2008. On September 2007, one cell was isolated and received no more influent from the rural guest house. The purpose was double: (i) to increase the organic loading rate of the TW which was very low due to the low starting of the activity of the guest house and (ii) to isolate one cell to conduct tracer test experiments with clear water, with controlled and constant inflow rates. Sampling and analysis continued for the remaining cell, and every sample was a mix of 2 points over width instead of the previous 4.

The sampling systems were small piezometers (permanently installed): these consisted of small PVC pipes (such as those used for sanitary conduits), of 2 to 3 cm of diameter. One tip section was perforated with holes on its periphery and closed with an end plug, driven into the gravel of the TW. Water was collected with a manual vacuum system; a first release empties the stagnant water, and then water was collected directly into the sample bottles (Figure 40).
Samples were stored inside dark glass bottles, and placed in a fridge-box (at 4°C) till their arrival the same day at the laboratory.
Usually, samples were collected in the morning and the laboratory analyses started in the afternoon.

Parameters analyzed were : BOD, COD, pH, EC, Total Nitrogen (TN), Nitrate (N-NO3), Total Phosphorus (TP) and TSS. Samples were not filtered before analyses.

(a)

(b)

Figure 40 : Sampling ports (a) and sampling with manual vacuuming pump (b)

III.3.2 TSS sampling errors

Most of the solids present inside a horizontal flow TW is an accumulation of microbial biofilms, biomat, intercepted particulate matter and plant roots.
The first sampling error is induced by a flow velocity change. As the velocity of the flow is strongly reduced inside the TW bed, sampling events can induce localised flow velocities at the sampling point that are much higher than the ambient flow velocities. This disturbs the *in situ* biomat and leads to sampling errors (Kadlec and Wallace, 2009). In the current case, permanent sampling ports were installed within the TW bed and could have reduced this impact.
The second effect of sampling error is depending on the spatial representativeness. The sampling ports are set permanently and can induce a specific biomat coating around the subsurface part of the ports. Additionally, studies report a variability for TSS samples with coefficient of variation of 145 % (with *n* samples = 215) (Kadlec and Wallace, 2009).
As a consequence of this, in the frame of this work, it was decided not to collect samples inside the TW bed but only focus on IN- and OUT- samples for TSS measures.
Future recommendations about TSS measures would be to analyse their composition (mineral and/or organic, microbial and chemical composition, etc.) if samples were collected inside the TW bed.
The same care had to be taken for the sampling at the outlet pipe. The biomat is present as a coating layer on the outlet pipe and any agitation of the water with the sampling process contaminates immediately the sample. The sampling procedure at the outlet was using a small tube connected to a manual pump. The tube was carefully set in the middle of the outlet pipe and delicately vacuumed the collected sample.

III.3.3 Laboratory analysis

III.3.3.1 Theoretical background

The oxygen demand resulting from organic matter in the wastewater influent can be measured by laboratory tests (Crites and Tchobanoglous, 1998):

- Chemical Oxygen Demand (COD): the oxygen equivalent is determined by chemically oxidizing organic matter using dichromate in and acid solution.

- 5-Day Biochemical Oxygen Demand (BOD_5): a sample of wastewater is placed in a 300 ml bottle, filled with oxygen saturated dilution water (containing nutrients), and incubated for 5 days at 20°C. The amount of oxygen consumed through microbial degradation is measured. Since this laboratory procedure measures oxygen demand resulting from breakdown and carbonaceous and nitrogen compounds, an nitrification inhibitor is add in the bottle so only the carbonaceous biochemichal oxygen demand ($CBOD_5$) is measured (Wallace and Knight, 2006).
- Ultimate Biochemical Oxygen Demand (BOD_u) is the total oxygen demand that could be exerted by organic matter if there were enough contact time to degrade all the material.

III.3.3.2 *<u>BOD</u>*

The laboratory analyses were done with the use of chemical kits, followed by colorimetric measurements. Laboratory analysis measured BOD_5 and COD but did not measured BOD_u. Samples were not filtered for BOD and COD measures.
Many of the limitations about the number of samples that can be analyzed were coming from the BOD analysis equipment. The laboratory has two racks of 6 bottles and can thus measure 12 BOD maximum per week. Analyses must be shared between the PhD site, tests performed by students on the pilot under greenhouse and the studies Epuvaleau leads on private orders.

BOD measures were done by the BODTrak system of HACH company (Model 2173B) based on manometer technology: The sample volume is defined by the expected BOD range from 95 ml to 420 ml of sample volume for BOD range of 0-35 mg/L to 0-700 mg/L. (Higher BOD range expectation involves the dilution of the samples). The sample volume is poured into the 473 ml BODTrak amber bottle. A magnetic stir bar is added inside the bottle, as well as a nutrient buffer pillow for optimum bacteria growth (HACH reference number 14861-98) and a nitrification inhibitor (HACH reference number 2533-35). A capsule of Lithium hydroxide crystal is delicately set in the seal cup having no direct contact with the sample.

Figure 41 : BODTrak System (HACH)

The bottle is set on the chassis of the BODTrak which can host 6 sample bottles. The appropriate tube is connected to the sample bottle and the cap is firmly tightened. Each tube is tagged with the channel number and the channel number setup is reflected on the control panel (Figure 41). The BODTrak is the incubator (20°C). The magnetic stir bars are active through the incubator chassis. The incubator is closed and the samples are left for 5 days without any additional manipulation. The BODTrack has an initial head space containing 21% oxygen above the water sample. Continuous stirring replenishes dissolved oxygen to the sample. This makes the BODTrak results similar to occurrences found in a natural environment. Bacteria use the dissolved oxygen from the sample; the CO2 gas produced by this oxidation is absorbed by the Lithium hydroxide and it creates a drop of pressure. The method continuously removes the carbon dioxide from the system so that the pressure difference observed is proportional to the amount of oxygen used. The drop of pressure is directly converted into mg BOD/L by the manometer system and can be read on LCD screen. The BOD rates can be observed daily without disturbing the sample. Pressure changes within the closed BODTrak system are displayed graphically in mg/L on a LCD. The final reading is done after five days and corresponds to the BOD_5 of the sample.

III.3.3.3 *COD*

COD was analyzed by the HACH chemical kit, with the reactor and the colorimetric spectrophotometer DR2500 Odyssey, Method 8000 for COD range 0-1500 mg/L COD. To determine the COD, samples were heated for 2 hours (150°C) a powerful Potassium dichromate oxidant in the COD reactor and then cooled down. The organic compounds which can be oxidized reduce the dichromate ion (Cr^{6+}) into chromium ion (Cr^{3+}) having a specific green color which is then measured by the spectrophotometer. The measure is comparative relative to a standard reference of de-ionized solution. The COD reactant contains salts of silver and mercury. Silver is a catalyst and the mercury salt complexes the interaction with Chloride ion. The spectrophotometer is set on 620 nm and calibrated with the reference standard. The sample reading is directly converted into mg/L COD.

III.3.3.4 *TSS*

TSS is measured by filtration. WHATMAN filters of microfibers, 47 mm in diameter, are used for the filtration. Filters are firstly set in the autoclave at 105°C during 15 min and weighted. The filter is then set on the filtration support and the sample is filtered. The volume which is filtered is noted. The filter is removed and set back in the autoclave at 105°C for 1 hour and is finally weighted back. The difference between the 2 weights reported to the filtered volume provides the mg/L TSS.

III.4 RESULTS AND DISCUSSION

The section is divided into three parts (devoted to BOD, COD and TSS results and discussion). Data are first presented and each of the water quality parameters are then presented and discussed. When adequate, models developed to predict the degradation rates coefficient (*k*-values) have been calibrated.

III.4.1 General presentation of data

Table 9 displays results of pH, Electric Conductivity (EC), Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), the COD/BOD ratio and Total Suspended Solids (TSS) with the regular statistical indexes of mean, standard deviation, minimum and maximum, median values and the number of samples on which results are based.
The sampling campaign started on June 2006 and ended on October 2008 and had more or less a monthly base. The number of samples provided varied from 35 to 41, according to laboratory analysis problems and also some lack of samples for surface samples (at 15 cm deep) when the water level of the TW was too low.

pH
The mean pH of influent was 7,4 and 6,9 for the effluent. The highest pH was 7,9 for wastewater and the minimum was 6,0 inside the TW bed. Mean values of pH show that there is a pH drop from the inlet to the first sampling line, and almost no more pH changes along length and with depth afterwards. pH was nevertheless systematically a little higher on bottom layers than on top layers.

Electrical Conductivity
The mean value of EC was 1000 µS/cm for wastewater influent and of 790 µS/cm for the effluent. There is a magnitude order of 10 between minimum and maximum values as the maximum value was recorded inside the bed at 1638 µS/cm and the minimum at 115 µS/cm. Even with a large standard deviation (SD), differences are systematically observed between surface samples from 15 cm, 35 cm and 55 cm deep with a decrease from surface to bottom layers. The two major reasons are believed to be the rain inputs on top surface layers and the low flow fractions (15 %) demonstrated by Chapter II for surface layers. The difference between influent and results inside the bed and effluent are not very high. The effluent sampling was also subject to the same sampling effect as TSS, which is discussed further.

Biochemical Oxygen Demand
The mean value of wastewater influent was 328 mg BOD/L what is considered as high for a secondary wastewater system influent. Kadlec and Wallace (2009) classify BOD concentration related to tertiary treatment with values of 3 to 30 mg BOD/L, to secondary when ranking from 30 to 100 mg BOD/L, to primary from 100 to 200 mg BOD/L and as super-loaded when > 200 mg BOD/L. The current influent set to this system is highly loaded when looking at the concentration level. The generally high BOD values can be attributed to the main dedicated role of the guest house as a restaurant, which produces high biodegradable wastes from cooking and over left food.
The mean outflow BOD concentration was 32 mg BOD/L, which demonstrates that based on long term data, the treatment efficiency is respectful of Walloon guidelines (the standard is 70 mg BOD /L for a treatment plant < 20 PE, see §III.2.1). It is interesting to point out that the median values are lower than the mean values. Nevertheless, the maximum value measured at the outlet is 135 mg BOD/L, which would not be respectful of the requested standard.

One cell of the system was closed for more than one year to any wastewater inflow to conduct the tracer tests. The cell was only loaded with rain water during that period. One sampling was done before starting tracer tests and it provided the minimum BOD values (also named C*) and considered as the BOD background of the system. This measure was done on September and it can be demonstrated that the TW has a seasonal variability. This value can only be considered as indicative, but is useful for future modelling, allowing calculations to have less unknown parameter values. It is nevertheless interesting to observe that within a TW in operation, there is BOD stratification with depth as the C* or BOD background is varying with length and depth and is ranking from 2 to 9 mg BOD/L. The rain input is probably responsible of this "flush down" effect through the pea gravel media.

Chemical Oxygen Demand

The general COD content is low for the system was indicated by the BOD/COD ratio. Metcalf and Eddy (1991) proposed the range of 0,4 to 0,8 for untreated wastewater and Crites and Tchobanoglous (1998) are classifying this ratio ranking from 0,3 to 0,8 for untreated wastewater and 0,4 to 0,6 after primary settling. The values observed should be within the range for wastewater after primary settling in this case. The mean value of this ratio is of 0,6 what is the tipping value between untreated wastewater and primary settling. General comments on BOD values indicated that the BOD values were high due to high content of biodegradable wastes from the restaurant of the wastewater. This also implies low COD values and the general ratio BOD/COD to be classified at the edge of untreated wastewater and primary settling.

Total Suspended Solids

As explained in the methodology section, TSS is measured on influent and effluent only. In this case, the number (*n*) of samples severely drops in comparison to the other parameters previously discussed, and is representative of the sampling problems with samples polluted by the agitation inside the outlet pipe as as described in the methodology section.
This phenomena points out a serious problem when treatment plants are sampled at the outlet by administrative workers to check the respect of the requested Walloon standards; and when they are not aware of the sampling methodology to respect for this specific type of treatment station.
The mean value of TSS of the influent is 99 mg/L with a median value of 70 mg/L and a maximum value of 330 mg/L. The mean value of TSS of the effluent is 34 mg/L and is respectful of the Walloon standards (60 mg TSS/L for treatment plant < 20 PE, see §III.2.1). The median value is lower (24,8 mg/L) but the maximum value observed would be too high to respect the Belgian norms (max value of 120 TSS mg/L). The removal percentage of 66 % could be considered as low but it has to be reported as a function of the low load of the influent.

The general results of Table 9 demonstrate that on the long term, the system in operation of Nassogne was respectful of Walloon guidelines related to BOD, COD and TSS.
pH and EC are showing a stratification with depth, which is assumed to be related to rainfall input and the low fractional flow on surface layers as demonstrated in chapter II.
It also shows that the BOD_{in} is high for a secondary effluent but the removal rate through the system is good at satisfying Walloon standards and is achieved within the first meters of the bed. Reversely, COD values are relatively low and logically, the BOD/COD ratio linking both parameters is high for a secondary effluent.

Table 9: General results of the monitoring campaign

Distance and depth				3 m from inlet			6 m from inlet			11 m from inlet			
Parameter	Stat	Unit	Inlet	0,15	0,35	0,55	0,15	0,35	0,55	0,15	0,35	0,55	Outlet
pH	n		38	39	40	40	39	40	40	40	40	39	40
	Mean		7,4	7,0	7,1	7,1	7,0	7,0	7,1	6,9	6,9	7,0	6,9
	Std Dev		0,2	0,3	0,2	0,2	0,3	0,2	0,2	0,3	0,2	0,2	0,2
	Median		7,4	7,0	7,1	7,1	7,0	7,0	7,1	6,9	6,9	7,0	7,0
	min		6,9	6,6	6,5	6,8	6,0	6,5	6,6	6,3	6,3	6,7	6,0
	Max		7,9	7,5	7,6	7,6	7,7	7,6	7,6	7,6	7,4	7,4	7,5
EC	n		40	41	42	42	41	42	42	42	42	41	42
	Mean	µS/cm	1000	682	878	1010	693	806	948	493	756	930	790
	Std Dev		351	332	366	352	613	309	336	194	313	288	299
	Median	µS/cm	1019	663	856	1004	604	814	1022	463	687	915	835
	min	µS/cm	170	223	256	324	195	355	251	224	115	449	312
	Max	µS/cm	1598	1330	1514	1638	4121	1342	1498	991	1379	1407	1328
BOD	n		39	38	38	38	36	39	38	37	37	38	36
	Mean	mg/l	328	46	51	44	34	40	36	23	39	37	32
	Std Dev		348	44	48	35	35	42	27	27	54	50	34
	Median	mg/l	235	38	38	35	27	25	29	12	20	22	19
	min	mg/l	11	6	7	8	3	5	7	2	9	9	6
	Max	mg/l	1500	194	226	170	166	215	135	130	304	240	135
	Remov.	%		86	84	86	90	88	89	93	88	89	90
COD	n		40	37	40	41	35	37	39	37	40	39	40
	Mean	mg/l	341	71	71	77	60	63	67	36	59	61	49
	Std Dev		319	61	46	64	44	32	43	34	71	52	39
	Median	mg/l	236	53	67	68	42	53	57	32	43	51	47
	min	mg/l	9	4	3	7	2	12	7	1	9	20	3
	Max	mg/l	1500	330	200	380	155	157	164	155	452	320	183
	Remov.	%		79	79	77	83	82	81	90	83	82	86
BOD/COD	Mean		1,0	0,6	0,7	0,6	0,6	0,6	0,5	0,7	0,7	0,6	0,7
TSS	n		23										22
	Mean	mg/l	99,1										33,9
	Std Dev		85,1										24,8
	Median	mg/l	70,0										30,0
	min	mg/l	16,0										4,0
	Max	mg/l	330,0										120,0
	Removal	%											66

n = Number of samples

III.4.2 Water Budget

In order to calculate the Organic Loading Rate (OLR, g BOD/m².day) applied to the system, the inflow rate per line over the TW must be known, and the water budget has to be defined. The system was monitored during 851 days, what represents almost two years and a half. Inflow was measured by a tipping bucket with a counter device (counting seesaw motions) and the outflow with a floating pump equipped with a flow meter. Rainfall (R) was measured with a pluviometer. Data were collected once or twice a month.

The water budget is given by the Equation 62:

$$IN + (P - ET) = OUT \qquad \text{(Eq. 62)}$$

Where, IN = Influent (m^3),
P = Precipitation (m^3)
ET = Evapotranspiration (m^3)
OUT = Effluent (m^3)

The first assumption is to use the common accepted one for large wetlands, and represent ET by the reference crop ET_0 computation given by Equation 63:

$$ET = K_c * ET_0 \qquad \text{(Eq. 63)}$$

Where, ET = Evapotranspiration
K_c = wetland crop coefficient, dimensionless
ET_0 = Evapotranspiration of the reference crop

The reference crop is grass and ET_0 values were provided by the Institut Royal of Meteorology (IRM) for a station located close to Gembloux. Even if this station is not close from the experimental site, the second assumption considers that ET_0 does not vary significantly between the two sites.

The coefficient K_c is taken out of a study from Fermor *et al.* (1999) giving the K_c values for reed beds in the United Kingdom based on the Penman - Montieth equations method of determination of ET_0. The reeds crop coefficient (K_c) is given by Table 10. In agriculture, every crop has its crop coefficient (K_c) that influences ET at specific sites. Free databases of crop coefficients are provided by FAO (Food and Agriculture Organisation of the United Nations).

Table 10 : Crop coefficient for horizontal reed beds in the UK (Fermor *et al.*, 1999)

Month	April	May	June	July	August	Septmber	October
K_c value	**0,76**	**0,90**	**1,24**	**1,61**	**2,16**	**3,38**	**1,98**

Data of the total water budget are the following :

- The total inflow (IN) is 155,4941 m^3,
- The total outflow (OUT) is 163,5492 m^3,
- The total precipitation is 887 mm, or 49,9035 m^3 over the TW area,
- The total ET is 40,8245 m^3

- The size of the TW is 56,28 m² (13,4 m * 4,2 m),
- The cumulative days are 344.

Out of Equation 62, the total outflow should have been of 164,5731 m^3 instead of the 163,5492 m^3 measured. The missing 1 m^3 is assumed to be measurement system errors combined with possible leaks of the TW.

Figure 42 displays the water budget drawn with data over 344 days having accurate data in a row with in- and outflow (m^3) with P-ET (m^3) on the left Y axis and precipitation (mm) on the right Y axis, versus the X axis with days (in decade).

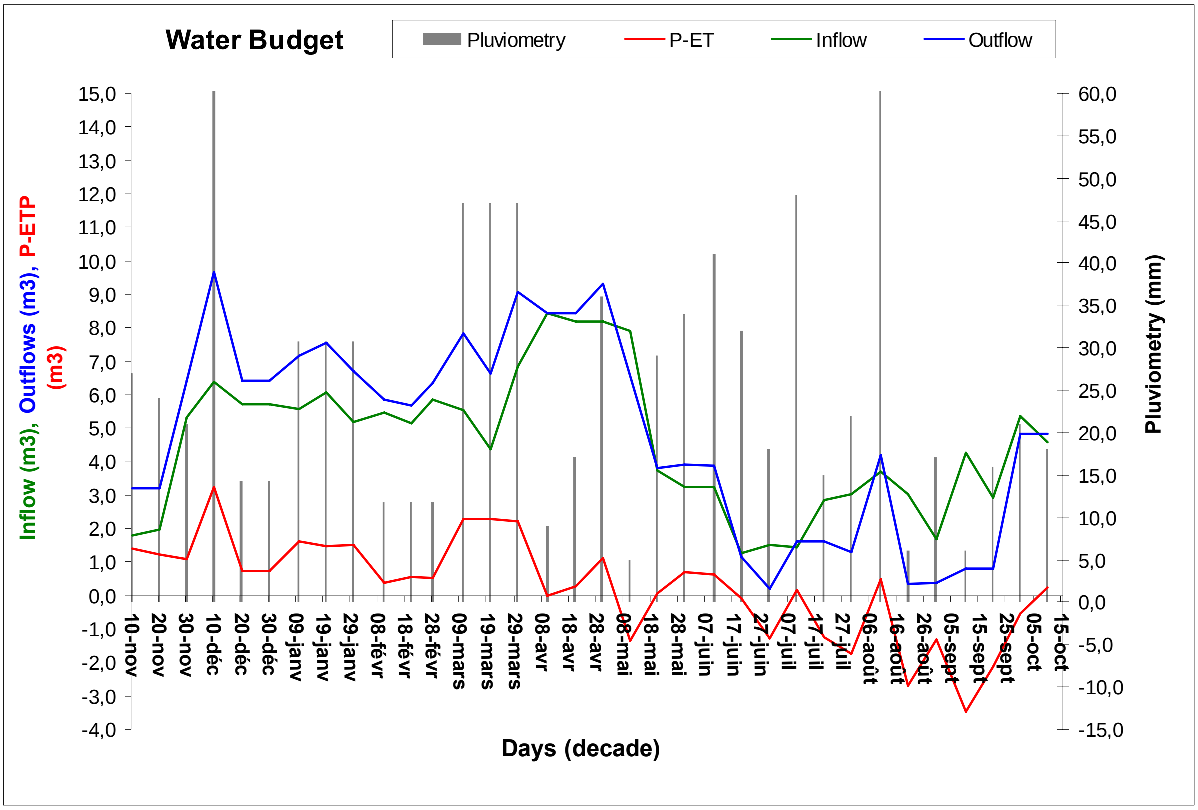


Figure 42 : In and Out flow (m^3) versus days (decades) over 344 days on 2007-2008

Most of the time, the inflow (green curve) is below the outflow (blue curve), meaning that more water exits the wetland, due to rainfall input (grey bars) and low evapotranspiration rate. The inflow curve is above the outflow one, meaning that less water exits the TW than it enters, when the evapotranspiration rate is higher than rainfall and the cumulative effect of 'P-ET' is negative, as shown by the red curve with negative values. If the reed bed was a crop, when the red curve has negative values, it would mean that the crop has a water deficit and the water need of the crop would not be provided. In this case of a TW bed, when the evapotranspiration (ET) is higher than rainfall input, the deficit is taken out of the water stock stored in the TW volume, as it is a permanent saturated bed, and the result is an outflow lower than the inflow.

As data collected about in- and out-flows were not always reliable due to pump failures, electricity cuts, clogging of flow meters, the inflow rate which is taken for further calculation as an average flow based on the elapsed time here above presented, having continuous reliable data.

In order to calculate the organic loading rate per line over the length of the wetland, the last assumption considers that 1/4 of the total ET is removed from inflow rate on Line 1, half the total ET on Line 2 and ¾ on Line 3 and the total at the outlet. Precipitation inputs are calculated as falling on the area including the 3 first meters of the length for Line 1, the 6 first meters of the bed for Line 2 and 11 m for Line 3. The thickness of layers and the fraction of flow per layers with depth was provided by Chapter II. Surface area is calculated by the multiplication of the distance from inlet (3, 6 and 11 m) by the width (4,2 m); the cross sectional area is the multiplication of the thickness of the layers per width. The loading rate values over length and with depth are summarised by Table 11.

Table 11 : Hydraulic loading rates over length and with depth

Line - Distance from Inlet			**L1**	**3**	**m**	**L2**	**6**	**m**	**L3**	**11**	**m**	**OUT**
	Nom.	**Unit**	**15**	**35**	**55**	**15**	**35**	**55**	**15**	**35**	**55**	
Surface area	**A**	**m²**	12,60	12,60	12,60	25,20	25,20	25,20	46,20	46,20	46,20	56,28
Thickness of layer	**H_i**	**m**	0,30	0,10	0,30	0,16	0,14	0,40	0,14	0,12	0,44	0,65
Cross sectional area	**A_c**	**m²**	1,26	0,42	1,26	0,67	0,59	1,68	0,59	0,50	1,85	2,73
Fraction of Q	**p_i**	**%**	30	10	60	14	20	66	9	21	69	100
Precipitation	**P**	**m^3/d**	0,032	0,032	0,032	0,065	0,065	0,065	0,119	0,119	0,119	0,145
Evapotranspiration	**ET**	**m^3/d**	0,030	0,030	0,030	0,059	0,059	0,059	0,089	0,089	0,089	0,119
Inflow : $(Q*p_i)$+P-ET	**Q_i**	**m^3/d**	0,138	0,048	0,274	0,069	0,096	0,304	0,071	0,125	0,342	0,478
HLR surf. Area (q)	**Q_i/A**	**m/d**	0,011	0,004	0,022	0,003	0,004	0,012	0,002	0,003	0,007	0,008
HLR Cross sect. Area	**Q_i/A_c**	**m/d**	0,110	0,114	0,217	0,103	0,164	0,181	0,120	0,248	0,185	0,175

III.4.3 Biochemichal Oxygen Demand (BOD) results

III.4.3.1 Frequency percentile

One typical method to present BOD results is the probability distribution for concentration entering and leaving the TW. Figure 43 displays the graph for inlet and outlet. The median inlet (50^{th} percentile) BOD is 235 mg/L, while the median outlet is 19 mg/L (as reported on Table 9). The Walloon standard of 70 mg/L BOD is achieved at the 87^{th} percentile.

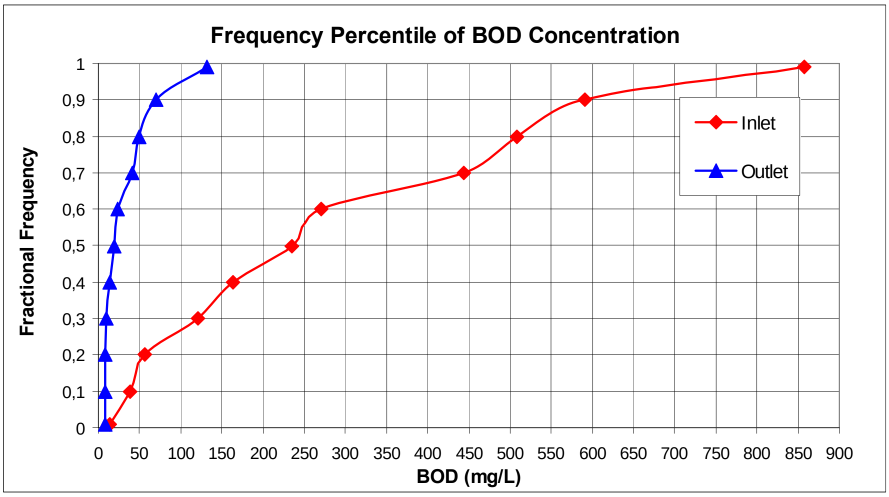


Figure 43 : Distribution of BOD concentration for inlet and outlet.

Figure 44 is showing the same probability distribution for all depths (15, 35 and 55 cm deep) and Lines 1, 2 and 3 located at 3, 6 and 11 m from the inlet. (The inlet curve is removed to allow a zoom in the BOD concentration scale). It is obvious to see that all curves are very similar and closed, what demonstrates that the BOD removal is achieved very quickly within the first meters of the TW. The same colour code is used as in Chapter II, which is blue family for Line 1, green for Line 2 and red for Line 3 with light colours for surface layer (15 cm deep) to dark colours for bottom layer (55 cm deep). It shows stratification in performances from the blue family curves located lower on percentile performances, followed by the green family curves with middle performances and the red family curves at the top percentile performances. The outlet curve being logically in the middle of red family curves. This reports the progressive BOD removal over length in the TW. The Walloon BOD standard is achieved for 70 to 90% of the samples.

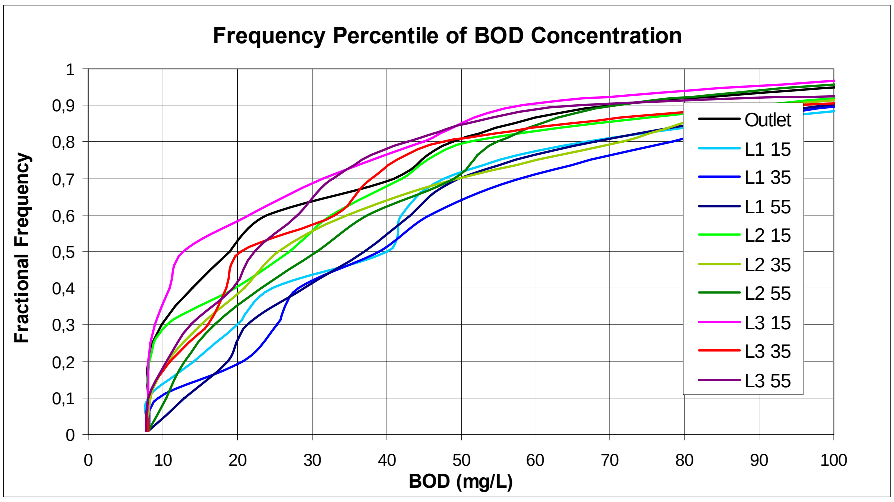
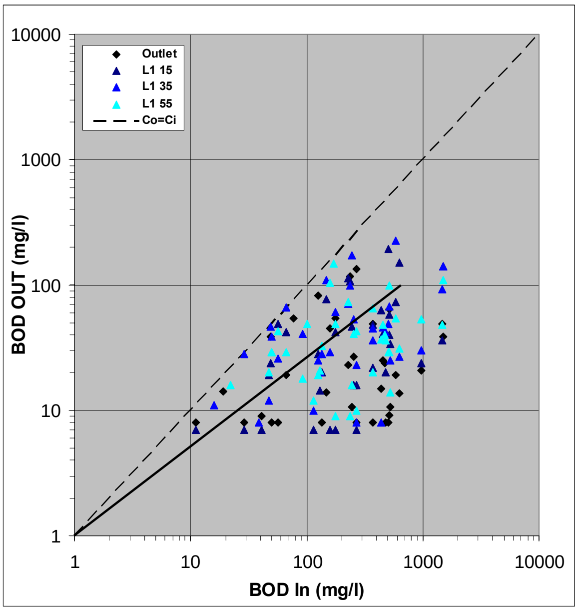


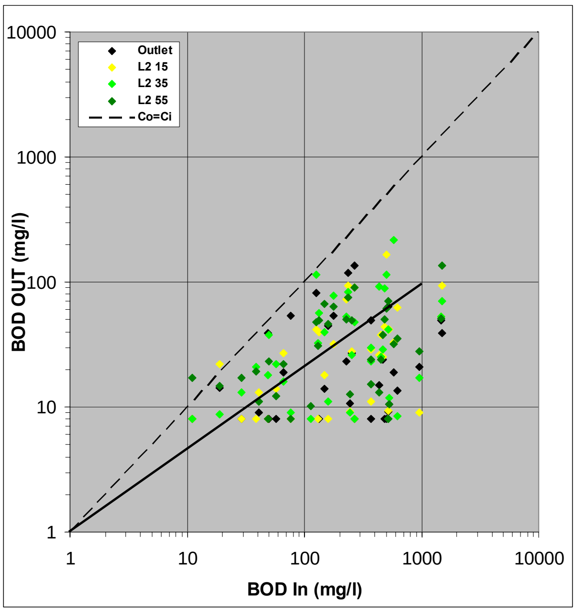
Figure 44 : Distribution of BOD performance over length and with depth.

III.4.3.2 <u>In- and Out- concentrations</u>

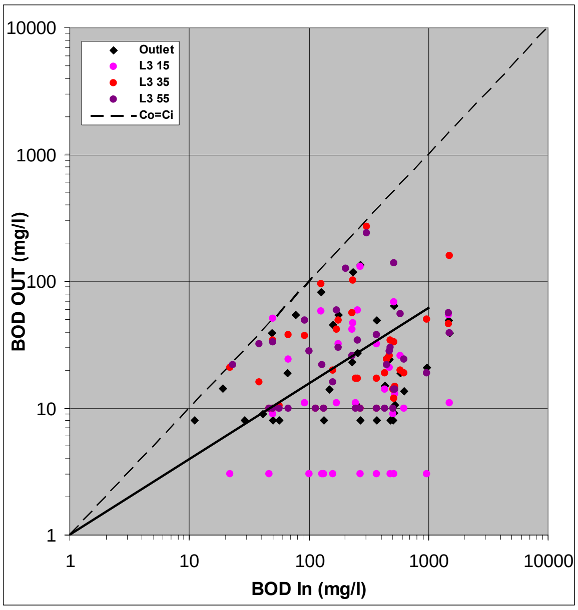
The other graphical displays which are often adopted in literature the TW plot outlet versus inlet concentration in mg/L (Figure 45) or BOD concentration (mg/L) versus BOD Loading In, also named BLI. BLI is expressed in g/m².d related to cross sectional area or on surface area. Figure 45 is presenting BOD Out versus BOD In (mg/L) per lines.



(a) BOD Input-Output for Line 1



(b) BOD Input-Output for Line 2



(c) BOD Input-Output for Line 3

Figure 45 is split into the 3 graphs above and on the left corresponding to Line 1, Line 2 and Line 3 at 3 m, 6 m and 11 m from inlet respectively. The same colour code per family (line1 = blue, line 2= green; line 3= red) is reproduced with darker colours corresponding to lower layer.

X and Y axes are all similar and on logarithmic scale to avoid scatter cloud of points and to show a more consistent central trend. The diagonal dashed line represents BOD concentration at the outlet equal to BOD concentration ate the inlet, what means no BOD removal. A Log-Linear central tendency regression is shown per line. The slope of those curves is increasing from line 1 to line 3 showing BOD removal efficiency higher as the wastewater is going through the bed with length. The slope coefficients are given here below.

Figure 45 : BOD Input versus output per lines over length.

The log linear central tendency regression per line and depth are the following: (C_o= BOD concentration at the outlet, C_i = BOD concentration at the inlet, corresponding here to a specific line and depth):

- Line 1 15 cm deep : $\quad$ $\ln C_o = 0{,}561 \ln C_i$
- Line 1 35 cm deep $\quad$ $\ln C_o = 0{,}545 \ln C_i$
- Line 1 55 cm deep $\quad$ $\ln C_o = 0{,}562 \ln C_i$

- Line 2 15 cm deep $\quad$ $\ln C_o = 0{,}616 \ln C_i$
- Line 2 35 cm deep $\quad$ $\ln C_o = 0{,}597 \ln C_i$
- Line 2 55 cm deep $\quad$ $\ln C_o = 0{,}616 \ln C_i$

- Line 3 15 cm deep $\quad$ $\ln C_o = 0{,}717 \ln C_i$
- Line 3 35 cm deep $\quad$ $\ln C_o = 0{,}629 \ln C_i$
- Line 3 55 cm deep $\quad$ $\ln C_o = 0{,}633 \ln C_i$

III.4.3.3 Seasonal effect on BOD

The residence time of organic matter within the TW bed may be considerably longer than the hydraulic residence time (defined in Chapter II), because particulate organic matter (and associated nutrients) are initially trapped by sedimentation, filtration and interception processes prior to microbial degradation (Wallace and Knight, 2006). During winter and cold months the degradation of organic matter proceeds slower than on spring and summer warm months, which leads to the trapping and accumulation of organic matter within the TW bed. When warmer months return, the organic matter restarts to be degraded, leading to the reduction of accumulated solids and a release of BOD. This theory explains why warm-season removal BOD rates do not appear to be significantly different than cold-season removal rates

and why HF TW systems are still efficient on water quality parameters removal during cold months (U.S.EPA, 2000).
Figure 46 shows the BOD removal efficiencies observed over months. A general sinusoidal trend points out the variations in removal efficiencies along the year. It also has to be related to the start up of the system which occurred on May 2005. It means that the first month displayed on the Figure 46 (June 2006) was during the first summer of the system, when the wetland was still not fully commissioned for purification processes. A drop is observed on spring 2007 and a second drop at the autumn 2007. Highest removal efficiency values are obtained around June on 2007. Seasonal effects were less impacted on 2008.

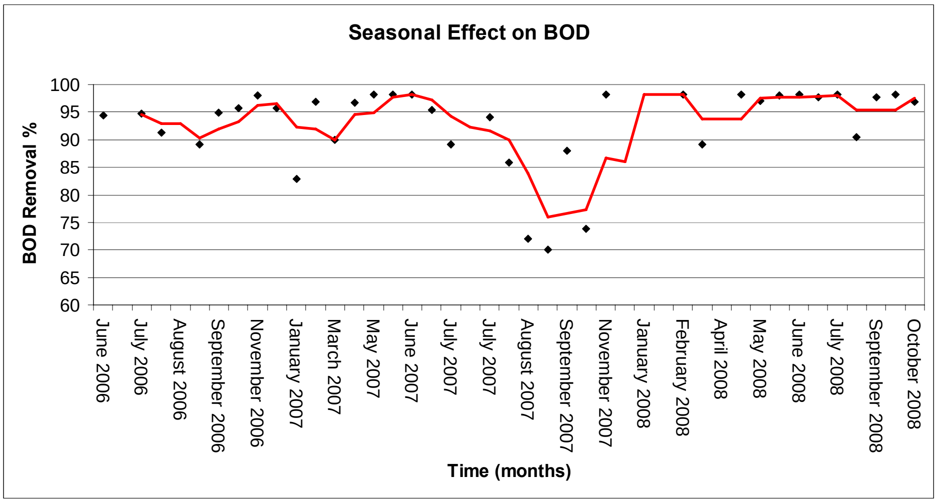


Figure 46 : seasonal effect on BOD

III.4.3.4 Model application and k-values

The major purpose of this work was to investigate what occurs and where inside the TW. Chapter II has identified the water flow pattern and this chapter is devoted to pollutant removal. The degradation rate for BOD (*k*-values) is calculated below in order to point out differences in BOD degradation rates over length and with depth.
The previously defined relaxed *P-k-C** TIS concentration model has been applied to all BOD data along the 385 days of measurements. The mathematical relationship is given by Equation 61, repeated here as:

$$\frac{(C_0 - C^*)}{(C_i - C^*)} = \left(\frac{1}{(1 + k/Pq)^P}\right) \qquad \text{(Eq. 64)}$$

Where,
- C^* = background concentration (mg/L)
- C_i = inlet concentration (mg/L)
- C_o = outlet concentration (mg/L)
- k = apparent TIS rate constant (m/yr)
- P = apparent number of TIS
- q = hydraulic loading rate (m/yr)

This equation has been applied for all BOD data and provides the overall *k*-rate constant, which is the single unknown of the equation.

The C* (mg/L) value is taken as constant per line and per depth and is the value presented in Table 1 as the minimum BOD concentration, corresponding to the isolated cell receiving rainwater only, and sampled prior to the tracer test experiments.
C_i (mg/L) values are the data collected of the inflow BOD concentration varying with all dates of measurements.
C_O (mg/L) values are the values of BOD concentration observed for all lines and depth, also varying with all dates of measurements.
The apparent number of tank in series *P* (dimensionless), is based on the *NTIS* calculated in Chapter II and divided by 2 as suggested by Kadlec and Wallace (2009). The *NTIS* represents the hydraulic number of TIS and *P* represents the organic TIS. The movement of organic particles is known as being slower than the water movement within the bed, as described previously.
The HLR *q* (m/yr), is the inflow rate (m^3/yr) (as defined by the water budget) and given by Table 11, divided by the surface area (m²). As reported for the water budget in Chapter II, *q* is varying for all dates of measurements and is given by the inflow (m^3/yr) weighted according to fraction of the flow identified per layer in Chapter II, combined with precipitation (m^3/yr) which have been weighted according the area related to the position of the concerned line of sampling over length and minus the evapotranspiration (ET) also varying with the position of the concerned line over length (ET is ¼ of total ET for line 1, ½ of total ET on line 2 and ¾ of total ET on line 3).
The *k*- value (m/yr) is given by the solution of Equation 61 (also Eq. 64).

The *k* degradation rate constants obtained are presented with the following frequency percentile table with statistics and figures here below (Table 12, Figure 47).

Table 12 : Annual area rate coefficient k (m/year) for BOD per lines and depths, percentile and statistics results

Line - Distance from Inlet / depth			L1	3	m	L2	6	m	L3	11	m	OUTLET
	Nom.	Units	15	35	55	15	35	55	15	35	55	
Data												
Inlet Conc.	C_i	mg/L	11-1500									
Background Conc.	C*	mg/L	6	7	8	3	5	7	2	9	9	6
Apparent TIS	P		3	2	1	10	6	10	6	16	17	13
Number of TIS	NTIS		6	4	2	19	12	19	12	32	34	26
k-Values Percentile												
0,05		m/yr	0,5	0,0	1,0	0,2	0,1	0,9	0,0	0,1	0,3	0,5
0,1		m/yr	1,0	0,2	4,0	0,4	0,3	1,5	0,2	0,1	0,8	1,3
0,2		m/yr	2,9	0,4	6,4	0,8	0,6	2,7	0,4	0,3	1,1	2,4
0,3		m/yr	5,4	1,1	20,7	1,3	1,1	3,8	0,6	1,0	3,2	3,7
0,4		m/yr	8,6	1,7	37,2	1,6	1,8	5,3	0,6	1,1	4,4	4,9
0,5		m/yr	10,4	2,7	46,7	2,6	2,7	7,9	0,8	1,4	5,6	5,8
0,6		m/yr	12,8	4,3	90,5	3,0	4,0	9,0	1,2	1,9	7,4	6,5
0,7		m/yr	20,0	8,1	193,8	3,5	4,9	14,2	1,7	3,5	8,6	7,7
0,8		m/yr	31,8	14,3	246,1	5,4	7,5	19,4	3,4	4,6	16,3	19,9
0,9		m/yr	39,8	17,9	291,4	8,3	13,0	33,9	5,2	5,7	20,7	26,1
0,99		m/yr	66,3	23,8	359,5	11,6	18,9	50,8	10,1	6,8	29,1	56,8
Statistic data												
Number of samples	N		36	35	37	35	36	36	35	35	36	34
Mean	Mean	m/yr	17	6	113	3	5	13	2	2	8	11
Standard Deviation	SD	m/yr	17	7	119	3	5	14	3	2	8	14
Median	Md	m/yr	10	3	47	3	3	8	1	1	6	6
minimum	min	m/yr	6	7	0,3	3	5	7	2	0,1	0,2	0,4
Maximum	Max	m/yr	75	24	362	12	19	53	11	7	32	66

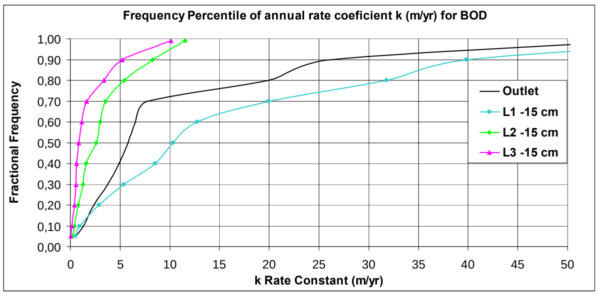
Table 12 shows that the influent concentration ranges from influent classified as "tertiary" influent (ranking from 3 to 30 mg BOD/L) to higher than "primary" influent (ranging from 100 to 200 mg BOD/L and above).
The hydraulic number of tank in series (*NTIS*) calculated from chapter 2 is divided by 2 to obtain the apparent number of organic tank in serie *PTIS*. The usual *PTIS* chosen in literature is typically around 3 (Kadlec and Wallace, 2009; Tanner and Sukias, 1997). Chapter II has demonstrated high *NTIS* in comparison to average values of *NTIS* usually accepted and thus high values for *PTIS* have been kept. The percentile frequency distribution is depicted by Figures 47.
Statistics indexes are based on lower number of samples than presented on Table 9, due to the fact that for the current calculation, both data on BOD from the influent and the concerned line and depth had to be reliable or existing.
The median value is corresponding to percentile 0,5 of the Table 12 and is ranking from 1 to 47 m/yr. The mean value is ranking from 2 to 113 m/yr, and a maximum value at 362 m/yr on Line 1 at the bottom layer, what is within the interval calculated by Kadlec and Knight, 1996 and Kadlec and Wallace, 2009.

The analysis of the statistical variance (ANOVA) with 3 factors (fixed factors for depth and location over length, random for sampling dates) with a confidence interval range of 0,01 and after the exclusion of a few erratic data provides the following observations.

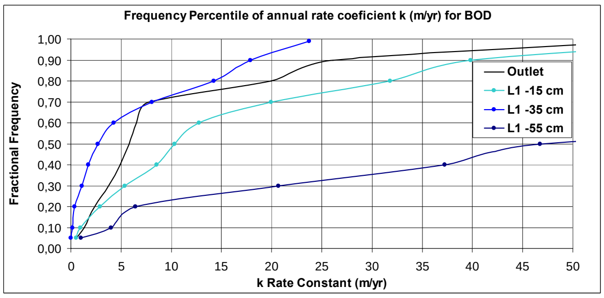
- The three locations over length have all very high significant influence of the degradation rate values.
- The three depths also have a very high significant influence of the degradation rate values.

It respects the logic that degradation rates are decreasing over length as the influent reaching the end of the bed has already been degraded by its path along length, the influent reaching the end of the bed is less strong and degradation rates cannot be as high as on the first few meters of the bed. Nevertheless, the higher degradation rates are obtained on bottom layers, and especially on Line 1. On Line 1, the best good coefficient is obtained for the surface layer. These results have to be considered with the high detention time and fraction of the flow involved according to depth as calculated on Chapter II.

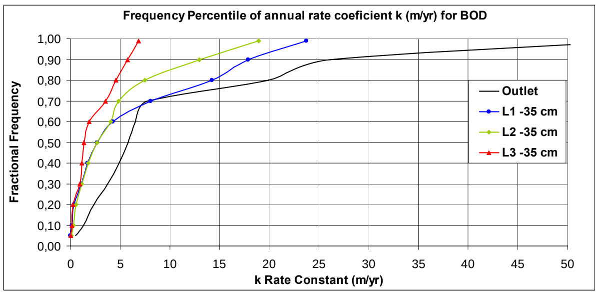
Figure 47 is providing the frequency distribution profiles of *k*-rate percentiles. The figure has to be read by columns. The left column presents results according to depth (Figure 47a to 47c) and the right column presents results according to lines (Figure 47d to 47f). The same colour code is kept as previously with blue family curves for line 1, green family for line 2 and red family for line 3. Darker colours correspond to bottom layers and light colours to surface layers. The outlet is represented on all graphs by the black line.
It shows that from Figure 47a to Figure 47c, curves are straighter in the surface layer and more inclined with depth demonstrating higher *k*-rate constants.
Figure 47d to 47f presents profiles per line and show a general tendency to be more inclined on Line 1 (Figure 47d) and getting straighter with Lines 2 (Figure 47e) and 3 (Figure 47f). For Line 2 (Figure 47e) and 3 (Figure 47f), curves with lighter colours are straighter than the darker ones, demonstrating the *k*-rate constants increase with depth. On Line 1 (Figure 47d) the degradation constants are higher on 15 cm deep than 35 cm deep.



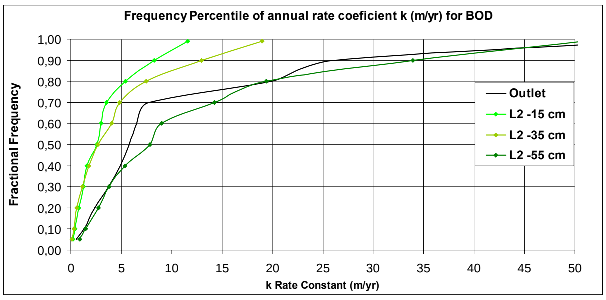
(a) Distribution frequency at 15 cm deep



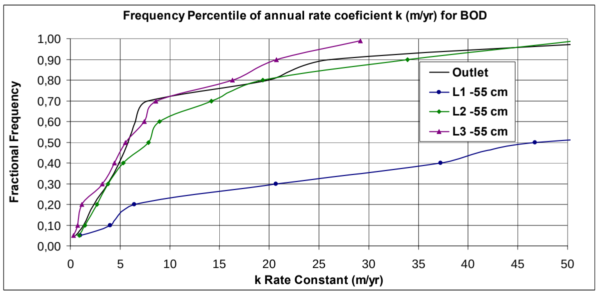
(d) Distribution frequency on line 1



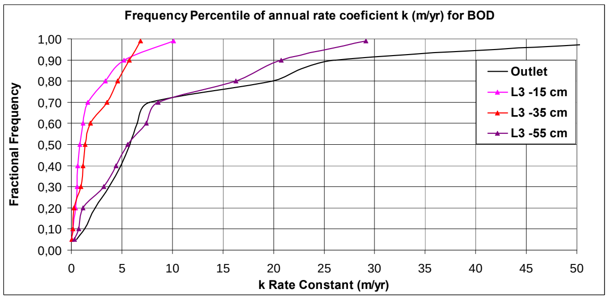
(b) Distribution frequency at 35 cm deep



(e) Distribution frequency on line 2



(c) Distribution frequency at 55 cm deep



(f) Distribution frequency on line 3

Figure 47 : Distribution frequency percentile of annual rate k coefficient for BOD

In conclusion, the results of BOD concentration over length and with depth have shown that 80 % of the samples collected satisfy the Walloon standard at the outlet. A very large decrease in BOD concentration occurs within the first few meters of the TW bed, and more than 60 % of the samples already met the Walloon standard after 3 m through the TW bed, as sampled on Line 1. Logically, the removal efficiencies are increasing from Line 1 to Line 2 and from Line 2 to Line 3, located at 3 m, 6 m and 11 m from the inlet respectively. A slight seasonal effect was observed along the two years and a half of measurements, with higher removal rates on June and lower ones during the autumns and during the winters. Finally, the application of the relaxed *P-k-C** TIS model has provided degradation rate coefficients for BOD ranking from less than 10 m/yr to more than 300 m/yr. Those *k*-rates are very significantly different and higher on Line 1 than for the 2 other lines and also significantly higher on the bottom layer of 55 cm deep than for other intermediate or surface layers, demonstrating a higher BOD degradation in those locations.

III.4.4 Chemical Oxygen Demand (COD) results

The same analysis can be done on COD data and are presented in a similar but simplified way than the BOD results.

III.4.4.1 Frequency percentile

Figure 48 presents the typical probability distribution for COD concentration entering and leaving the TW. The median inlet is 236 mg COD/L and the median outlet is 47 mg COD/L. The Walloon standard of 180 mg COD/L is achieved for 100 % of the samples.

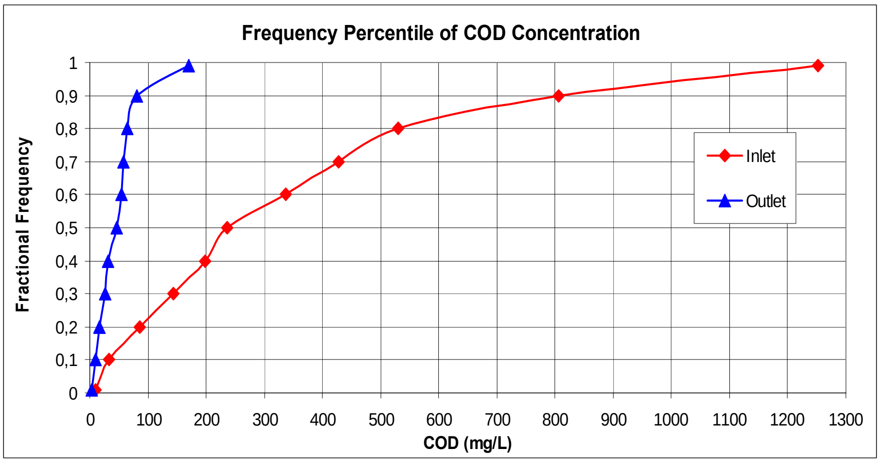
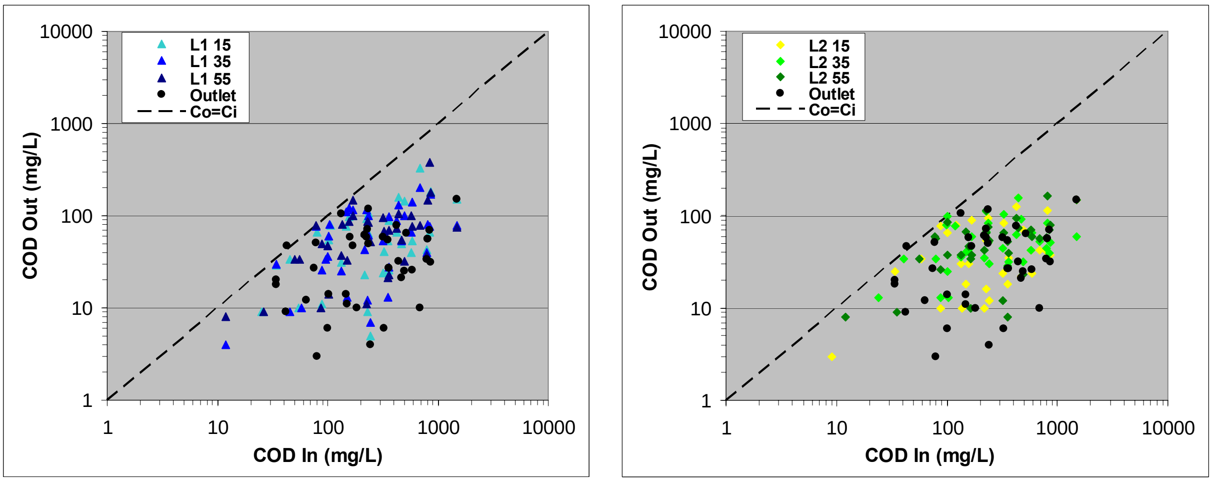


Figure 48 : Distribution of COD concentration for inlet and outlet

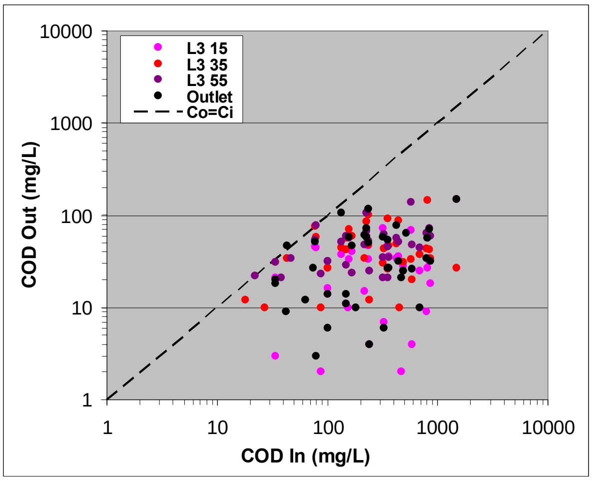
The percentile distributions with length and depth are not presented here as it displays no relevant visual trend.

III.4.4.2 In- and Out- concentrations

Results over lengths and depths are presented by Figure 49, which plots COD concentration at the outlet versus COD at the Inlet (mg/L).



(a) COD In-Out on line 1 (b) COD In-Out on line 2



(c) COD In-Out on line 3

The three sub-figures are presenting In and Out COD concentration. All X and Y axis are similar and with logarithmic scale to present a more consistent central cloud of points. The diagonal dashed black line represents no COD removal, with COD out equal to COD In.

At first sight, no specific tendency can be drawn from these graphs neither with distance over the TW from inlet, nor with depth.

Figure 49 : COD input versus output per lines over depth.

III.4.4.3 Model application and k-values

In order to investigate whether differences are existing in COD removal depending on distance from inlet and with depth, the degradation rate coefficient *k* was calculated for COD similarly as it was done for BOD. The reference *P-k-C* TIS* concentration model is applied to all collected data over the two and a half year of monitoring.

The C* (mg/L) value is taken as constant per line and per depth and is the value presented in Table 9 as the minimum COD concentration, corresponding to the value of COD concentration measured on the isolated cell, which was sampled before the tracer test experiments.

All other values of C_i (mg/L), C_0 (mg/L), the apparent number of tank in series (PTIS), the hydraulic loading rate *q* (m/yr) are similarly provided as they were described for the BOD calculations. The *k*- value (m/yr) is given by the solution of Equation 61.

The *k* degradation rate constant obtained are presented in Table 13 and Figures 50.

Table 13 presents the *k*-values percentile and the usual statistic indexes. The frequency percentiles are presented graphically with figure 50.

As discussed previously with table 9, the COD concentration of the influent is quite low for a domestic wastewater. The low values are explained by the major activity of the rural guesthouse as a restaurant, producing wastewater mainly loaded with biodegradable residues. As the COD concentration is low, the difference between influent and background concentration is low too, the ratio between influent and effluent is low, what induces logically low degradation rates. Degradation rate *k* is varying from 0 up to 337 m/yr, with mean values around 10 and median ones around 5 m/yr.

The statistical variance analysis (ANOVA) with 3 factors (fixed for lines and depths, random for sampling dates) with a confidence interval range of 0,01 and with the exclusion of a few erratic data reveals the following significant differences.

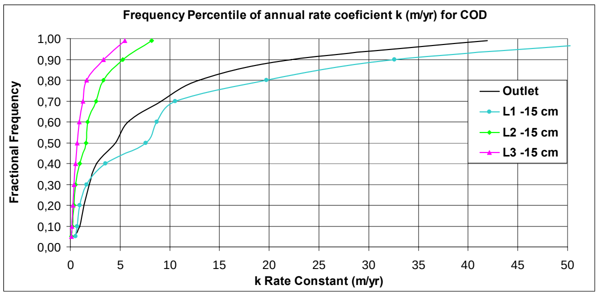
- The analysis per line present very high significant differences between all lines.
- The analysis per depth presents very high significant difference for all depths.

As observed for BOD *k*-value results, the degradation coefficient is decreasing over length. The COD load of the influent arriving at the end of the bed (location of Line 3) being lower, the degradation rate cannot be as high as for Line 1 located at the entry of the bed and receiving highly loaded influent. As for BOD also, there is a severe drop in COD degradation from inlet to Line 1 and a softened reduction along Lines 1, 2 and 3.
As for BOD finally, COD *k*-values are systematically higher in the bottom layers, demonstrating that the majority of the wastewater treatment occurs in the bottom of the bed, where the majority of the water movement has been demonstrated to flow on Chapter II.

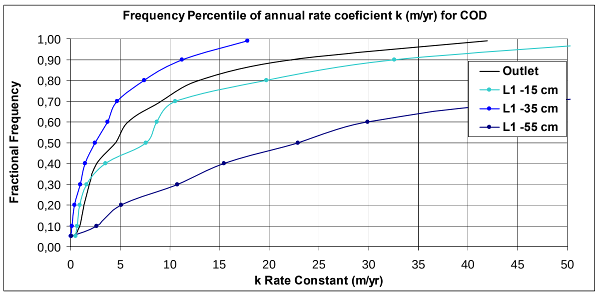
Table 13 : Annual area rate coefficient *k* (m/year) for COD per lines and depths, percentile and statistics results

Line - Distance from Inlet / depth			**L1**	**3**	**m**	**L2**	**6**	**m**	**L3**	**11**	**m**	**OUTLET**
	Nom.	**Units**	**15**	**35**	**55**	**15**	**35**	**55**	**15**	**35**	**55**	
Data												
Inlet Conc.	**Ci**	**mg/L**	9-1500									
Background Conc.	**C***	**mg/L**	4	3	7	2	12	7	1	9	20	3
Apparent TIS	**P**		3	2	1	10	6	10	6	16	17	13
Number of TIS	**NTIS**		6	4	2	19	12	19	12	32	34	26
k-Values Percentile												
0,05		**m/yr**	0,5	0,0	0,1	0,1	0,1	0,5	0,1	0,0	0,1	0,4
0,1		**m/yr**	0,7	0,2	2,7	0,2	0,4	1,0	0,1	0,2	0,7	1,0
0,2		**m/yr**	1,0	0,4	5,1	0,3	0,5	2,2	0,3	0,3	1,4	1,4
0,3		**m/yr**	1,7	1,0	10,7	0,5	0,9	2,6	0,4	0,5	1,9	1,9
0,4		**m/yr**	3,5	1,5	15,5	1,0	1,0	3,5	0,5	0,7	2,1	2,6
0,5		**m/yr**	7,6	2,5	22,9	1,6	1,8	4,8	0,7	0,9	3,5	4,5
0,6		**m/yr**	8,7	3,7	29,9	1,7	2,7	6,7	0,9	1,6	5,9	5,8
0,7		**m/yr**	10,5	4,7	46,9	2,6	3,9	10,8	1,3	2,3	7,1	9,1
0,8		**m/yr**	19,7	7,5	88,8	3,3	5,6	22,0	1,6	2,5	10,8	13,0
0,9		**m/yr**	32,6	11,2	130,2	5,3	9,8	26,7	3,3	4,5	19,1	22,2
0,99		**m/yr**	57,4	17,8	309,4	8,2	14,8	46,8	5,5	8,6	27,7	41,9
Statistic data												
Number of samples	**N**		35	38	38	34	35	36	34	37	36	37
Mean	**Mean**	**m/yr**	12	4	55	2	4	10	1	2	7	7
Standard Deviation	**SD**	**m/yr**	15	5	80	2	4	12	1	2	8	10
Median	**Md**	**m/yr**	8	2	23	2	2	5	1	1	4	5
minimum	**min**	**m/yr**	0	0	0	0	0	0	0	0	0	0
Maximum	**Max**	**m/yr**	67	18	337	9	15	53	6	9	28	47

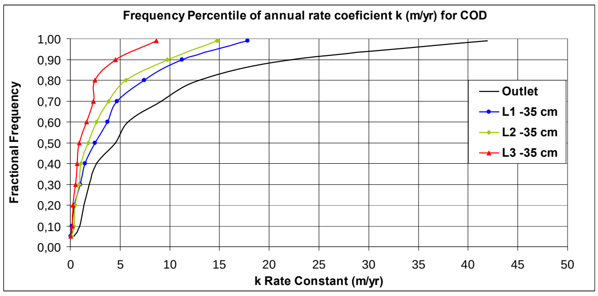
Figure 50 is represents graphically the percentile results of *k*-values for COD. The figure is split into 2 columns: the left one presents results with depth and the right column is presenting results over length. The colour code of blue for Line 1, green for Line 2 and red for Line 3 is replicated as previously. Darker colours are representing lower layers and lighter colours the upper ones. All axes have the same scales. The purpose of these graphs is to visually show the general trend expressed in Table 13.
Graphs (a) to (c) varying with depth show that curves for the 55 cm deep are flatter than the curves related to 15 and 35 cm deep, demonstrating higher degradation rates.
Graphs (d) to (f) varying over length are showing that the farer from inlet, the straighter the curves are, reporting the decrease in degradation rates over length.



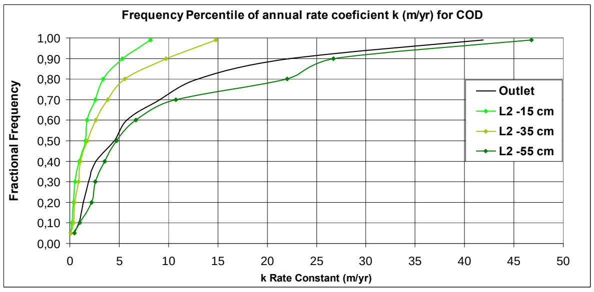
(a) Distribution frequency at 15 cm deep



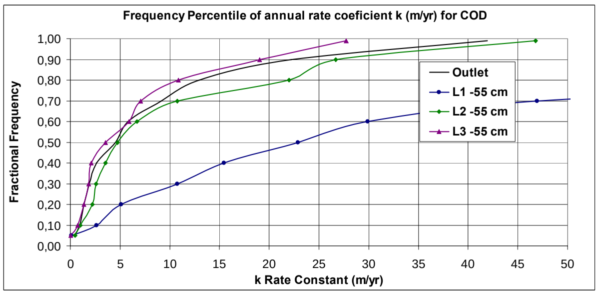
(d) Distribution frequency on line 1



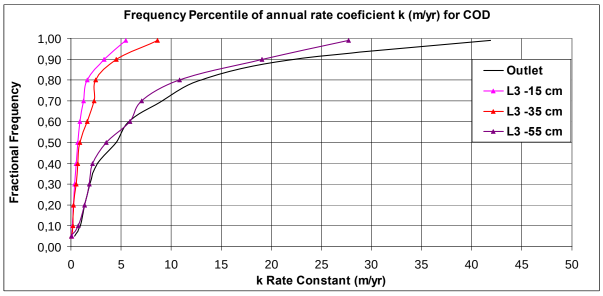
(b) Distribution frequency at 35 cm deep



(e) Distribution frequency on line 2



(c) Distribution frequency at 55 cm deep



(f) Distribution frequency on line 3

Figure 50 : Distribution frequency percentile of annual rate *k* coefficient for COD

In conclusion, the results of COD concentration over length and with depth have shown that more than 100 % of the samples collected were satisfying the Walloon standard of 180 mg COD/L at the outlet. The percentage has to be taken as the first years of the start up of the system, which is probably still not fully colonised by micro-fauna. A substantial decrease in COD concentration occurs within the first few meters of the TW bed, and more than 60% of the samples on Line 1 located at 3 m from inlet are already satisfying this norm. Logically, the removal efficiencies are increasing from Line 1 to Line 2 and from Line 2 to Line 3, located at 3 m, 6 m and 11 m from inlet respectively, but with no clear distinction. Finally, the application of the relaxed *P-k-C* TIS* model has provided degradation rate coefficients for COD ranking from 0 m/yr to more than 300 m/yr. Those *k*-rates are all significantly different with the distance over the bed. The degradation rates are systematically higher and highly significantly different for the bottom layers, demonstrating higher COD degradation with depth.

III.4.5 Total Suspended Solids (TSS) results

III.4.5.1 Frequency percentile

The probability distribution for TSS concentration entering and leaving the TW is displayed by Figure 51. The data presented indicate that the HF system returns an average effluent concentration of 33,9 mg/L over the monitored period with a 90^{th} percentile limit of 53 mg TSS/L. The median inlet TSS was 70 mg/L and the median outlet was 30 mg TSS/L. This is consistent with other statistical analyses of TSS removal in horizontal systems (Wallace and Knight, 2006).

The system is designed for 20 PE and the Walloon standard for this station size is of 60 mg/L of TSS was met for 92% of the collected samples. The conclusion is that the system fully satisfies and is respectful of the Walloon standards over time.

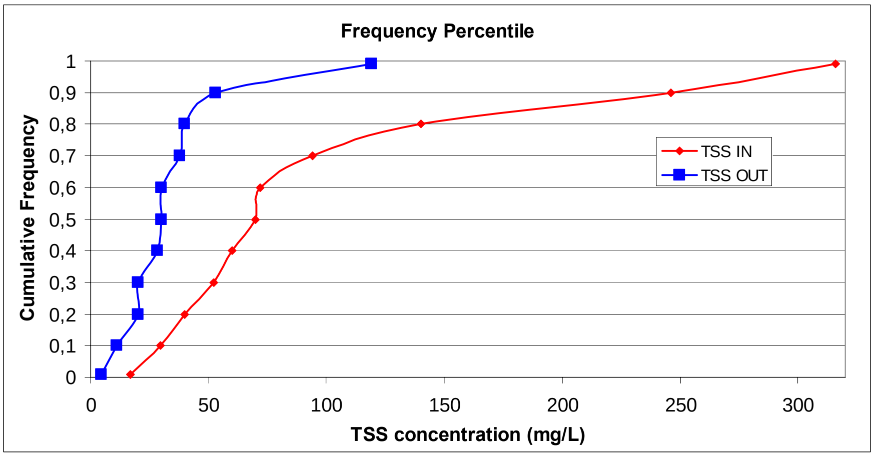


Figure 51 : Distribution of TSS concentration for inlet and outlet.

III.4.5.2 In- and Out- concentrations

TSS leaving the TW is a function of internal biological processes, as described within the theoretical background section, and there is a non zero background concentration (C*) for effluent TSS. The most commonly used graphical relationship is expressed by outlet concentration versus the inlet loading (Figure 52). The TSS inlet load is the TSS concentration of the influent (mg/L) multiplied by the inflow (m^3/yr) and divided by the surface area (m²). The result is expressed in g/m².yr. TSS in the effluent of horizontal TW system is not a function of inlet TSS loading (Kadlec and Wallace, 2009), as demonstrated by Figure 52 where no trend curve can be drawn out of the results. Performance criteria postulated by the Walloon standard and the mean value is displayed with data results on Figure 52.

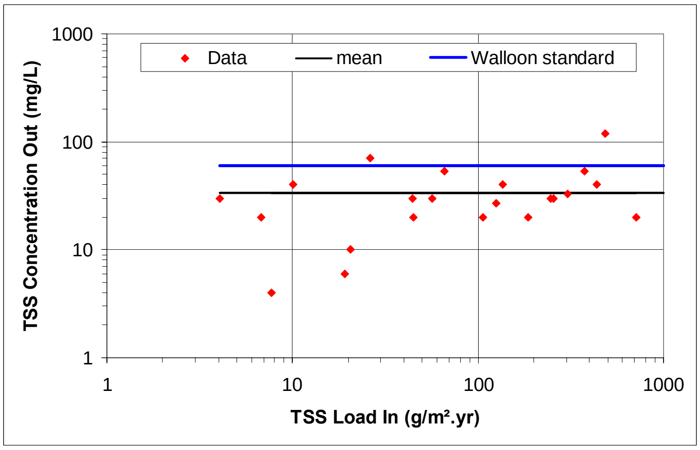
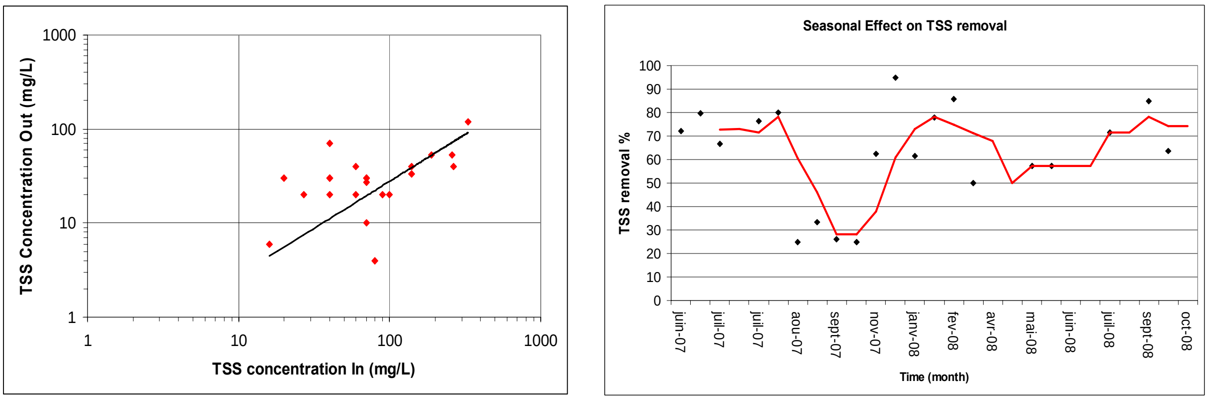


Figure 52 : Effluent TSS concentration (mg/L) versus Influent TSS load (g/m².yr).

III.4.5.3 *Seasonal effect on TSS*

TSS data series often display some degree of sinusoidal behaviour through the course of a calendar year (Kadlec and Wallace, 2009). The linear trend drawn on the graph presenting TSS concentration In (mg/L) versus TSS concentration Out (mg/L) (Figure 53a) reports a variability of 32 %, which can be attributed to stochastic and seasonal effects. Usual ranges have upper limits of 20 % (Kadlec and Wallace, 2009). The high values observed for our data can be attributed to the sampling problems described in the experimental method section.
As for BOD, a minor seasonal variation is observed on TSS removal efficiency, which is displayed by Figure 53b. The seasonal effect has a lower impact on 2008.



(a) (b)

Figure 53 : Seasonal effects on TSS removal

III.4.5.4 *Model application*

Suspended Solids removal is a very specific concern for surface systems and specific modelling has been developed. Nevertheless, in the case of horizontal subsurface system the application of these equations is of limited utility, as in many instances the TSS entering the TW is removed very rapidly and the TSS effluent leaving the TW is determined by internal biological processes, but not by the external loading of incoming TSS (Kadlec and Wallace, 2009). No model was thus applied for TSS removal.

III.5 CONCLUSIONS

This third chapter presents the observed results for three wastewater quality parameters of BOD, COD and TSS which are ruled by Walloon standards.

The theoretical background summarised the current state of the art related to organic compound degradation and processes occurring within horizontal flow treatment wetlands for BOD, COD and TSS removal, as well as clogging processes and impacts. Models which are developed to express and predict the treatment performances for BOD and COD were presented.

The experimental layout explains the sampling methodology, through the set of sampling port tubes inserted permanently in the TW bed and distributed over three location along the length of the bed, always replicated three times in order to sample three depths. The sampling error associated with the analysis of TSS led to the decision not to investigate TSS within the TW bed in the frame of this work but only focus on in- and outlets analysis. If TSS had to be sampled within the TW bed, recommendations are to analyse the TSS in mg/l, as requested by the Walloon standard, but to jointly analyse their inner biochemical composition (mineral/organic, microbial fauna, chemical composition, etc.). This would be the relevant way to investigate TSS over length and with depth inside a horizontal flow TW bed.

Results are explained based on the background of the hydraulic pattern of water issued from Chapter II. This provides explanations to the low general results of pH and EC on surface layers, as the combined effect of surface dilution and flush-down of pollutants due to rainfall associated with the low fraction of the flow involved in the water movement for the upper layers inside the TW bed. Mean results demonstrate high values for the BOD in the inflow, especially for a wastewater issued after primary treatment. Reversely, COD content of influent is low. This is attributed to the main activity of the rural guesthouse as a restaurant, producing wastewater with a high biodegradable fraction. The three water quality parameters of BOD, COD and TSS are respectful of Walloon guidelines on mean and median values at the outlet but are occasionally above the maximum values.

The global water budget shows that the outflow is usually higher than inflow due to the rain input, except for a few months from May to September where the evapotranspiration (ET) of the bed can be higher than the inflow and combined rain inputs. The deficit is then taken out of the water stock enclosed in the wetland bed. The water budget data of inflow, rain, ET and outflow combined with the fraction of the flow identified in Chapter II allowed the calculation of the inflow per layers for the three locations over the length of the bed. This has allowed the use of the mathematical relaxed *P-k-C** model to calculate the BOD and COD degradation coefficient values (*k*) for all the collected samples. Their percentile frequency distribution and statistical analysis quantified the degradation rates over length and with depth, which is a new work in the TW field. BOD and COD *k*-values, associated with usual graph and table presentations, demonstrate the same effect which is a substantial decrease in BOD and COD from the inlet to the first line of sampling located at 3 m from the inlet. 67% and 73 % of BOD and COD respectively were removed prior to this first internal sampling line. The degradation *k*-coefficients are statistically different from lines to lines over the length of the bed and the degradation rates are significantly higher for the bottom layers, whatever the lines. Seasonal effects were displayed for BOD and TSS especially with lower removal percentages observed during winter months.

CHAPTER IV

Nitrogen and Phosphorus Removal Processes and Performances

IV. 4. CHAPTER IV: NITROGEN AND PHOSPHORUS REMOVAL PROCESSES AND PERFORMANCES

IV.1 INTRODUCTION

This chapter reviews with the theoretical background the various forms of nitrogen and their transformation within horizontal subsurface flow beds, based on current knowledge adapted to Belgium. Sampling layout and laboratory analysis are detailed. Results about total nitrogen, nitrate and total phosphorus are presented. Additional microbial analyses are performed to assess the activity of the aerobic bacteria and the presence of nitrifying and denitrifying bacteria.

IV.2 THEORETICAL BACKGROUND

IV.2.1 Walloon legislation

The European Water directive 91/271/CEE on urban wastewater treatment require no treatment obligation on Nitrogen and Phosphorus purification for treatment plants having a size lower than 10.000 PE. The present case study has thus no legal obligation for TN and TP removal efficiencies. The European standard for larger plants is presented by table 14 in a simplified version. Results will be discussed on its basis.

Parameter	Number of PE	Concentration (mg/L)	Minimum removal percentage (%)
Total Phosphorus	10.000-100.000	2	80
	> 100.000	1	80
Total Nitrogen	10.000-100.000	15	70-80
	> 100.000	10	70-80

Table 14 : Simplified table of 91/271/EEC, annex 1.

IV.2.2 Nitrogen degradation and cycle within horizontal flow TW

Municipal or domestic wastewaters contain varying amounts of organic nitrogen depending upon the source. Regular sewage wastewater usually contains 40% of nitrogen under organic form and 60% as ammonia. (U.S. EPA, 1993). The most important inorganic forms of nitrogen in wetlands treating domestic wastewater are ammonia (NH_4^+), nitrite (NO_2^-), nitrate (NO_3^-), nitrous oxide (N_2O) and dissolved elemental nitrogen (N_2).
Nitrogen compounds can be moved inside the wetland without molecular transformation; the main physical processes are: particulate settling and re-suspension, diffusion of dissolved forms, plant translocation, litterfall, ammonia volatilization, and sorption of soluble nitrogen on substrates. Besides the physical translocation of nitrogen compounds, five major processes transform nitrogen form one form to another, which are: ammonification, nitrification, denitrification, assimilation and decomposition (Kadlec and Wallace, 2009).
Figure 54 depicts the global nitrogen cycle for a surface flow TW. The degradation processes are the same for media based subsurface systems, with different proportionalities ratio and location of occurrences.

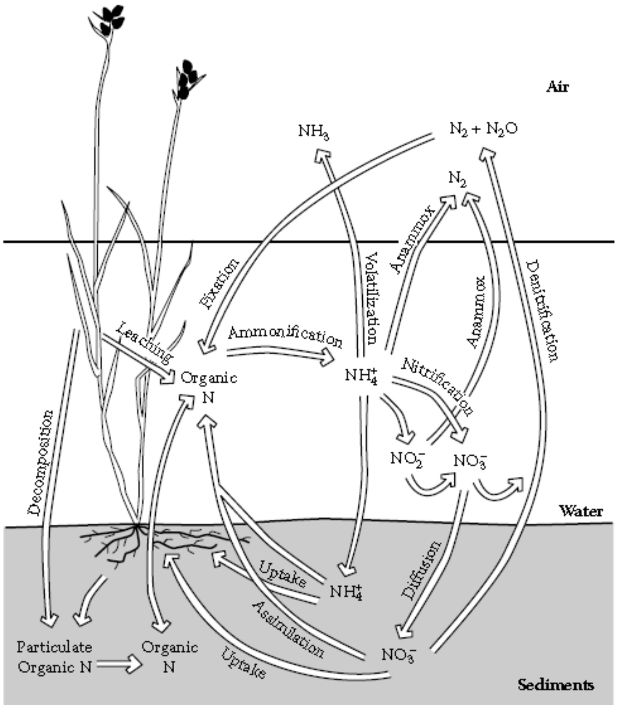


Figure 54 : Sketch of nitrogen cycle for a surface TW (Kadlec and Wallace, 2009)

IV.2.2.1 Nitrogen forms in TW for domestic wastewater

Organic nitrogen in domestic wastewater includes a variety of compounds such as amino acids (main component of proteins); urea and uric acid issued from mammals (the simplest form of nitrogen in aquatic systems); and purines and pyrimidines (heterocyclic organic compounds in which nitrogen replaces two or more of the carbon atom in the aromatic ring).

Ammonia exists in water solution under un-ionized ammonia (NH_3) or ionized ammonia (NH_4^+, ammonium ion) depending in pH and temperature. This physico-chemical process where ammonium N is known to be in equilibrium between gaseous and hydroxyl form as indicated by Equation 1, is called the ammonia volatilization.

$$NH_3 + H_2O \Leftrightarrow NH_4^+ + OH^- \qquad \text{(Eq. 65)}$$

Based on a broad literature review, Vymazal (1995) summarized that volatilisation rate is controlled by the NH_4^+ concentration in water, temperature, wind velocity, solar radiation, the nature and number of aquatic plants, and the capacity of the system to change the pH value in diurnal cycles (the absence of CO_2 increases volatilisation).

The ionized form (NH_4^+) is predominant in most wetlands systems because of moderate pH and temperature (Kadlec and Wallace, 2009). Un-ionized ammonia (NH_3) is toxic to fish and aquatic life, typically at concentration < 0,2 mg/L and is subject to standard regulation for discharge in many countries, especially those with coasts and rivers having fisheries activities well developed (UK and Sweden, e.g.). The volatilization is a process which is much more used for Nitrogen removal in countries having warm climates (Bilore *et al.*, 2006).

Nitrite (NO_2^-) is an intermediate oxidation stage (+3) of nitrogen between ammonia (oxidation stage -3) and nitrate (NO_3^-, oxidation stage +5). Because of this intermediate energetic condition, nitrite is chemically not stable in most wetlands and usually found at very low concentration.

Nitrate (NO_3^-) can serve as essential nutrient for plants, but in excess and in combination with phosphorus to the eutrophication of surface water, like lakes and rivers. The current regulatory consent for nitrate in drinking water supply in Wallonia is < 50 mg/L.

Nitrite is typically near to zero in sewage and in secondarily treated effluents; however nitrate may be the dominant form in nitrified secondary effluents. Nitrate is also present in agricultural runoff due to the oxidation of ammonia fertilisers in the non saturated zone of agricultural fields (Kadlec and Wallace, 2009). Measures of nitrate leaching with the percolation water and migrating towards the underground aquifer under agricultural fields cultivated with industrial vegetable crops have reached up to 230 mg /L in the Walloon region (Fonder *et al*., 2010).

IV.2.2.2 Nitrogen physical translocation, without molecular transformation

Biomass. The potential rate of nutrient uptake by plant is limited by its net growth rate and the concentration of nutrients in plant tissues. Therefore, desirable traits of a plant used for nutrient assimilation and storage would include rapid growth, high tissue nutrient content and the capability to attain a high standing crop (biomass per unit area) (Reddy and Debusk, 1987). Borin and Bonati, 2000 have quantified the biomass produced by Typha latifolia and Phragmites australis of 33 and 39 $t.ha^{-1}$ of dry weight respectively and the immobilisation of nitrogen showed the high levels of 270 and 250 kg ha^{-1} respectively. The Total Nitrogen (TN) content of living biomass in wetlands varies considerably among species, plant parts and wetland sites (Kadlec and Wallace, 2009). The biomass is often analysed for aboveground tissues, standing dead and litter, and belowground roots and rhizomes. In The Netherlands and for Phragmites australis, Mueleman *et al.* (2002) shown that a considerable fraction of the biomass is belowground, which is particularly troublesome form the standpoint of sampling. This study did not develop the biomass stand point.

Matrix sorption. Oxidised nitrogen forms (nitrite and nitrate) do not bind to solid surface but in a reduced state ammoniacal N is stable and can be adsorbed onto active sites of the bed matrix. Because of the positive charge of the ammonium ion, it is subject to cation exchange and is not considered to be a long term sink for NH4+-N removal as sorption is presumed to be rapidly reversible (Vymazal *et al*., 1998b). As the NH4+-N is lost from the system by nitrification, the exchange equilibrium are expected to redistribute itself. This concept is applied by vertical fill and drain TW systems being alternately fed and drained to switch aerobic and anaerobic chemical reactions. The Freundlich equation can be used to model NH4+-N sorption (Cooper *et al*., 1996). The character of the substrate is important to determine the amount of sorption and exchange and the median ammonia loading for horizontal systems is about 1g/m².day, and the median concentration is 20 mg/L (Kadlec and Wallace, 2009).

In surface flow TW, organic material can form ***accretion*** in soils and sediments (Craft *et al*., 2008).

IV.2.2.3 Nitrogen chemical transformation

Microbial classification

Bacteria are numerous and strongly involved in chemical reactions with nitrogen in TW. Nitrification is usually carried out by autotrophic bacteria (capable to acquire energy and carbon from inorganic sources) and denitrification often, but not always, by heterotrophic bacteria (requesting a source of carbon). Bacteria also produce enzymes that break down complex molecules and the principal nitrogen processes are carried out in biofilms located on soil, sediments, media and submerged plant parts (Kadlec and Wallace, 2009).

Primary nutritional groups are groups of organisms, divided according to the sources of energy and carbon, needed for living, growth and reproduction. The sources of energy can be light and organic or inorganic compounds; the sources of carbon can be of organic or inorganic origin. Table 15 summarizes the name of those organisms which will be used within the following sections.

Energy source	Reducing equivalent source	Carbon source	Name
Light *Photo*	Organic *-organo-*	Organic *-heterotroph*	*Photoorganoheterotroph*
		Carbon dioxide *-autrotroph*	*Photoorganoautotroph*
	Inorganic *-litho-*	Organic *-heterotroph*	*Photolithoheterotroph*
		Carbon dioxide *-autrotroph*	*Photolithoautotroph*
Chemical compound *Chemo*	Organic *-organo*	Organic *-heterotroph*	*Chemoorganoheterotroph*
		Carbon dioxide *-autrotroph*	*Chemoorganoautotroph*
	Inorganic *-litho-*	Organic *-heterotroph*	*Chemolithoheterotroph*
		Carbon dioxide *-autrotroph*	*Chemolithoautotroph*

Table 15 : name of organisms according to the source of energy and carbon (Culot, 2005)

In horizontal TW systems the oxidation of nitrogen is governed primarily by oxygen availability in the rhizosphere , where the dynamic of oxygen is complex as oxygen transfer to the roots are driven by a variety of physiologic and physical processes (Brix, 1994b). Within the rhizosphere, bacteria are competing for oxidation of organic matter, nitrogen compounds and sulfides; the influent composition and loading rates affect the overall redox condition of the root zone and influence the degree to which oxygen is available for oxidation of nitrogen compounds (Wallace and Knight, 2006). Horizontal flow TW in Wallonia are used to treat septic effluents, having very little dissolved oxygen available to support nitrogen oxidation and as a result, most horizontal TW systems typically process nitrogen no further than the mineralisation stage, producing ammonia as the primary form of nitrogen in the effluent (Wallace *et al*., 2006).

Ammonification
Ammonification is the biological transformation of organic nitrogen to inorganic nitrogen, especially N-NH_4^+, and is the first step to mineralization of organic nitrogen (Reddy and Patrick, 1984). The process occurs both aerobically and anaerobically, heterotrophic microorganisms are considered to be the group involved (U.S. EPA, 1993). Mineralization rates are fastest in the oxygenated zone and decrease as mineralization switches from aerobic to facultative anaerobic and obligate anaerobic fauna (Vymazal *et al*., 1998b). The rate of ammonification in wetlands depends on temperature, pH, C/N ratio of te residue, available nutrients in the system, and soil conditions such as texture and structure (Reddy and Patrick, 1984).

Nitrification is the principal mechanism that converts ammonia nitrogen to oxidised nitrogen. Van de Graaf *et al.* (1996) defined nitrification as the biological formation of nitrate or nitrite from compounds containing reduced nitrogen with oxygen as the terminal electron acceptor. Nitrification typically relies on chemoautotrophic bacteria, but it is now also recognized that nitrification can occur through heterotrophic bacteria (Paul and Clark, 1996).

- *Classical nitrification with autotrophic bacteria in conventional systems*:

In conventional wastewater treatments, nitrification is a two step mediated process (U.S. EPA, 1993):

Nitritation $2NH_4^+ + 3O_2 \xrightarrow{Nitrosomonas} 2NO_2^- + 2H_2O + 4H^+$ (Eq. 66)

Nitrification $2NO_2^- + O_2 \xrightarrow{Nitrobacter} 2NO_3^-$ (Eq. 67)

Nitritation is mediated in a first step by autotrophic bacteria (*Nitrosomonas*) and nitrification as a second step by *Nitrobacter* bacteria. Both steps relies on oxygen and the nitrification rate may be controlled by the flux of dissolved oxygen into the system. The oxidation reactions release energy used by both Nitrosomas and Nitrobacter for cell synthesis. Nitrification lowers the alkalinity and pH of the water (U.S. EPA, 1993). The optimal pH range for nitrification in treatment systems is between 7,2 to 9,0 (Metcalf and Eddy, 1991)

- *Nitrification with heterotrophic bacteria in natural wetlands*

Treatment wetlands almost always operate at neutral pH and are considerably more complex than the biological nutrient removal of conventional systems (e.g., activated sludge or trickling filters) (Kadlec and Wallace, 2009). Bothe *et al*. (2000) in Kadlec and Wallace, (2009) identified *Nitrosospira* and *Nitrosococcus* as Ammonia Oxidizing Bacteria (AOB) in addition to *Nitrosomonas*. Austin *et al*. (2006a) found *Nitrospira* as abundant as *Nitrosomonas* and much more prevalent than *Nitrobacter* to oxidise nitrite. They also found heterotrophic bacteria as *Paracoccus denitrificans* and *Pseudomonas butida* capable of nitrification. They suggest the oxidation of ammonia in nitrite as a two step process, catalysed by enzymes :

$NH_3 + O_2 + 2H^+ + 2e^- \xrightarrow{Ammonia\,Monooxygenase} NH_2OH + H_2O$ (Eq. 68)

$NH_2OH + H_2O \xrightarrow{Hydroxylamne\,Oxidoreductase} NO_2^- + 5H^+ + 4e^-$ (Eq. 69)

These considerations cast doubt about the applicability to TW of the stoichiometry applied to conventional wastewater treatment systems. From equations 68 and 69, the dissolved oxygen requirement is 1,14 g O_2 per gram of ammonia nitrogen, and equation 2 requires 3,43 g O_2 per gram of ammonia nitrogen. The stoichiometric factor of 4,3 g O_2 per gram of NH_4-N oxidized is used in many papers as the maximum amount of oxygen transferred into the water , but in many wetlands situations, the factor 4,3 does not seem to be applicable (Tanner and Kadlec, 2002). These alternative pathways having the potential to substantially reduce the oxygen fluxes need to be investigated further in both natural and constructed wetlands to develop an understanding of their role in wetland nitrogen removal (Kadlec and Wallace, 2009).

In horizontal systems treating domestic wastewaters, nitrification typically does not occur unless the system is very lightly loaded with an areal loading rate of 10m²/person.day (Geller, 1997). Because of their low oxygen transfer rates, horizontal systems were mostly developed in Europe as denitrification processes to remove nitrogen from nitrified trickling filters effluent (Cooper *et al*., 1999). A comparative study from Sundberg *et al*. (2006) of filter beds and ponds indicated that nitrification activity was not correlated with depths and independently from ponds or filter beds; no relationship was detected between the activity and the corresponding bacterial community.

Denitrification is commonly defined as the process in which nitrate is converted into dinitrogen via intermediates nitrites, nitric oxide and nitrous oxide (Paul and Clark, 1996).

- *Classical denitrification with carbon source*

Bacteria responsible of denitrification are facultative heterotrophs, what means that they can use either oxygen or nitrate as terminal electron acceptors. The sequential starting from nitrate is the following:

$$\underset{Nitrate \rightarrow Nitrite \rightarrow nitric oxide \rightarrow Nitrous oxide \rightarrow Nitrogen gas}{2NO_3^- \rightarrow 2NO_2^- \rightarrow 2NO \rightarrow N_2O \rightarrow N_2} \quad \text{(Eq. 70)}$$

Diverse organisms are capable of denitrification and can be :

- Organotrophs (e.g., *Pseudomonas*, *Alcaligene*, *Bacillus*, *Agrobacterium*, *Flavobacterium*, *Propioni-bacterium*, *Vibrio*);
- chemolithothrophs (e.g., *Thiobacillus*, *Thiomicrospira*, *Nitrosomonas*);
- photolithotrophs (e.g., *Rhodopseudomonas*);
- diazotrophs, bacteria that fix atmospheric nitrogen gas (e.g., *Rhizobium*, *Azospirillum*);
- Archaea, group of single-celled microorganisms (e.g., *Halobacterium*);
- And others such as *Paracoccus* or *Neisseria* (Foch and Verstraete, 1977; Knowles, 1982; Kilham, 1994; Paul and Clark, 1996 *in* Kadlec and Wallace, 2009).

The stoichoimetry of nitrate reaction with methanol or glucose as a source of carbon is translated to the optimum carbon level of 2,3 g BOD per g NO3-N (Gersberg *et al.*, 1984), as the most labile form of organic carbon in wetland environments is the influent BOD, which is preferentially used to reduce oxidized forms of nitrogen (Kadlec and Wallace, 2009).
Theoretically, denitrification does not occur in the presence of dissolved oxygen. However, denitrification has been observed in conventional treatment systems that have relatively low measured dissolved oxygen concentrations, but not above 0.3–1.5 mg/L (U.S. EPA, 1993).
The reaction is irreversible and occurs in the presence of available organic substrate only under anaerobic or anoxic conditions, with redox potential ranking from +350 to +100 mV and where nitrogen is used as an electron acceptor in place of oxygen (Kadlec and Wallace, 2009). As most of the denitrification is done by heterotrophic bacteria, the process is strongly dependant on the carbon source and there is a general correlation between total soluble organic matter content and denitrification potential (Wallace and Knight, 2006). Horizontal flow TW systems producing a reducing environment are favourable and efficient to denitrification when they are fed with nitrified influent; the lack of organic carbon can be a limiting factor (Liehr *et al.*, 2000).

- *Denitrification with sulphur*

Koenig and Liu (2001) and Soares (2002) have demonstrated that sulphur driven autotrophic denitrification can alternate to heterotrophic denitrification. *Thibacillus denitrificans* bacterium can reduce nitrate to nitrogen gas while oxydising elemental sulphur, or reduced sulphur compounds including sulfides (S^{2-}), thisulfates ($S_2O_3^{2-}$), and sulfite (SO_3^{2-}). Treatment wetlands can have many forms of sulphur arising from the introduction of sulphate in the influent.

- *Aerobic denitrification*

The spatial zonation of oxygen and dissolved oxygen in a wetland is complex: there is an oxygen gradient from surface water to bottom sediments; significant quantities of oxygen pass down through the airways to the roots which can be used for respiration (Brix and Schirup, 1990; Brix, 1993); other gases pass upward from the roots zone; bed media and changes in loading rates induce capillary edge level being variably saturated or non saturated; all those effects allow aerobic and anoxic conditions to proceed in a very close proximity, conducting to reactions easily connected by diffusion of nitrification and denitrification occurring only microns away from each other. Most of the reactions occur thanks to bacteria attached to the bed media, and the biofilms therefore comprise a third spatial no uniformity in the wetland environment (Kadlec and Wallace, 2009). Additionally, *Thiosphaera pantotropha* and other organisms have been found being not only heterotrophic nitrifiers but also aerobic denitrifiers, what forced the reevaluation that nitrite reduction to gaseous products can not to be a strict anaerobic process only (Jetten, 2001).

- *Anaerobic ammonia oxidation (anammox)*

The bacteria mediating this process were identified only 20 years. It takes place in many natural environments and anammox is also the trademarked name for an ammonium removal technology that has been developed by the Delft University of Technology. The reaction commercially demonstrated is given by Equation 71:

$$NH_4^+ + NO_2^- \xrightarrow{Plantomycetes Nitrosomonas\, eutrophora} N_2 + 2H_2O \qquad \text{(Eq. 71)}$$

The process proceeds through nitrite, formed according Equations 68 and 69 and carries an oxygen requirement of 1,94 g O per gram of NH4-N. It is autotrophic and has no carbon requirement (Kadlec and Wallace, 2009). The importance of this process lies in the reduce carbon and oxygen requirements which are less than half and no carbon, in comparison to conventional routes. Various commercial processes are now available which capitalize on the advantage of this route for nitrogen removal (Anammox ®).
A striking feature of the organism is the extremely slow growth rate. The doubling time is nearly two weeks. They have been identified in both surface and horizontal subsurface flow TW systems (Austin *et al.*, 2006b). In many TW there is adequate oxygen to allow classical nitrification and in others enough carbon to fuel classical denitrification, but there are some for which ammonia and oxidized nitrogen are removed in amounts that considerably exceed the estimated supplies of carbon and oxygen, opening the route for anammox (Tanner and Kadlec, 2002).

Fixation. Biological nitrogen fixation is the process by which nitrogen gas in the atmosphere diffuses into solution and is reduced to ammonia by autotrophic and heterotrophic bacteria, cyanobacteria (blue-green algae) and higher plants. The reduction of gaseous nitrogen (N_2) to ammonia (NH_3) takes place very rapidly and individual steps have not been investigated separately. It is supposed that the reaction is a three-step, two electrons per steps mechanism, given by Equation 72 (Winter and Burris, 1976 *in* Kadlec and Wallace, 2009):

$$N \equiv N \rightarrow HN = NH \rightarrow H_2N - N_2H \rightarrow NH_3 \qquad \text{(Eq. 72)}$$

Results from different studies do not permit quantification of the fixation occurring in TW but indicate the ability of wetland plants and soils to fix nitrogen. It is unlikely that the rates of fixation in TW contribute materially to nitrogen cycling in nitrogen-rich systems (Kadlec and Wallace, 2009).

IV.2.3 Phosphorus removal within Horizontal Flow TW

Phosphorus is often the limiting nutrient in fresh water systems and can have significant impacts on downstream receiving water bodies. Due to the high content of phosphorus and nitrogen in domestic wastewater, constructed wetlands are very nutrient-rich.
The mechanisms which can store phosphorus in horizontal TW systems are: sorption by the bed media, plant biomass, chemical precipitation, accretion and particulate settling in

sediments. These storages have all finite capacities. The forms of phosphorus in the wetland environment can be dissolved forms (ortho-phosphate, PO_4-P; soluble reactive P, total dissolved P, dissolved organic P), dissolved forms associated with suspended solids (total reactive P, Total P, Total organic P and particulate P), sorbed to the surface of soil or media particles, contained in the structure of biomass and contained in the structure of soil particles (Kadlec and Wallace, 2009). Biological oxidation results in the conversion of most phosphorus to the orthophosphate form (Cooper *et al*., 1996). A large body of knowledge was developed last 15 years for large scale surface flow systems in Northern America for lightly loaded systems (typified by the Everglades project in Florida) and has no equivalent to horizontal compact small systems developed for domestic wastewater treatment in Northern Europe. Research is focusing for HF systems on bed media and retention time to maximize the short-term phosphorus removal and adsorption capacities fro wastewater combination (Wallace and Knight, 2006).

IV.2.3.1 Sorption

The amount of phosphorus which can be sorbed is a function of the influent phosphorus concentration and the properties of the bed media. Isotherms of Langmuir and Freundlich describe the sorption process (Del Bubba *et al*., 2003). Media which were investigated are mineral aggregates (sand, gravels, crushed rocks, dolomite, calcite, limestone, i.e.), soils, marine sediments (maerl, shells), man-made aggregates (LECA- Light Expanded Clay Aggregate) and industrial by-products (furnace slag, crushed concrete, wood chips, i.e.) (Arias *et al*., 2001, 2003; Arias and Brix, 2004, 2006; Chazarenc *et al*. , 2007; Drizo *et al*., 1999, 2006; Harouya *et al*., 2008; Kadlec and Wallace, 2009; Korkuzus *et al*., 2006; Kurup, 2008; Molle *et al*., 2005; Tsihrintzis and Gikas, 2009).

The problem about phosphorus sorption is that it is limited by the finite extent of the reservoir of adsorption sites; once sites are saturated, the phosphorus sorption ceases. As the available P retention capacities of the media may be exhausted in the first years of operation although it depends on the influent P loading and required effluent P concentration (Kurup, 2008), it implies the regular replacement of the sorbent. Pre and post-treatments have so been added to horizontal beds in order to avoid their complete refurbishment (Arias *et al*., 2003).

IV.2.3.2 Plant biomass cycling

Phosphorus being an essential nutrient for plant growth, there is a phosphorus plant uptake during the growing season and a release of phosphorus during the decomposition stage. The standing crop biomass of Phragmites is of 1-2 kg/m², with a phosphorus content of about 2% (Vymazal *et al*., 1999). Even if 50 % of the phosphorus is taken during the maximum growth month, on a four month growing season, only 4g/m².month will be up taken, what is considerably much lower than the average influent phosphorus load of domestic wastewater, which is of 12,6 g/m².month (Kadlec and Wallace, 2009). When plants are not harvested it maybe speculated that phosphorus will leach out of the detritus layer and will find it sway back to the water, at a rate which is much slower than surface flow systems. Because decomposition is not complete, there maybe a build up of the refractory, indecomposable fraction of the litter on the top of the bed (Kadlec and Wallace, 2009). When plants are harvested, the amount of phosphorus which can be recovered through harvesting part is about 2-4,9 g/m².year (Vymazal *et al*., 1999). This is again a very small amount next to the primary influent phosphorus loading which are typically about 150 g P/m².yr for horizontal flow systems (Kadlec and Wallace, 2009). Nevertheless, lightly loaded systems can reach nearly 40 % removals by harvest (Vymazal, 2004).

IV.2.3.3 *Chemical precipitation*

A variety of cations can precipitate phosphate under certain conditions. In important potential mineral precipitates in the wetlands environment include apatite ($Ca_5(Cl,F)(PO_4)^3$) and hydroxyl apatite ($Ca_5(OH)(PO_4)^3$). In addition to direct chemical reaction, phosphorus can precipitate with other minerals, such as ferric oxydoxide and the carbonate minerals (e.g. calcite, $CaCO_3$) (Reddy and D'Angelo, 1994). According to pH, the phosphorus may be fixed: in acid soil, by aluminium and iron if available; in alkaline soils by calcium and magnesium if available and forming amorphous or poorly crystalline solids. In reducing conditions, iron minerals are solubilised and release phosphorus co-precipitates (Reddy and D'Angelo, 1994). The nature and extent of precipitation reactions are dependent on the geochemistry of the water to be treated, as well as the pH, temperature and redox conditions (Kadlec and Wallace, 2009). The regeneration mechanism of slag furnace to phosphorus retention capacity is explained by Drizo *et al*. (2002) by nucleation sites which promote the adsorption/precipitation/crystallisation in calcium, iron and hydroxyl supersaturated pools, which can be drained and oxidized. In Denmark, Arias and Brix (2006) report the chemical precipitation of phosphorus as pre-treatment for onsite small scale systems, using ferric chloride or alum, which prevents the wetland media from short-term finite sorption. Precipitates depose as sediments in the sludge which is regularly removed by the cleaning and maintenance of the system.

IV.2.3.4 *Accretion*

Accretion is an important removal mechanism for surface flow systems which is less sustainable for horizontal flow systems due to the loss of porosity and hydraulic conductivity that accompanies solids deposition (Kadlec and Wallace, 2009). The phosphorus up took during the growing season returns to the system by biomass decay, generally within one annual cycle. The refractory fraction can range 10 to 15% of the overall biomass production, what would result in the accumulation of 0,8 – 1,2 g/m².yr for *Phragmites* (see above) in refractory organic material, which can be considered as not significant next to the influent phosphorus load (Kadlec and Wallace, 2009).

IV.2.3.5 *Particulate settling*

Horizontal flow systems, which are media based present a favourable environment to particulate settling and removal mechanisms issued from deposit and trap of solid particles (see Chapter II on hydraulic). The deposits contribute to a loss of porosity and hydraulic failures leading to flooding conditions (see Chapter III on clogging). The finite pore volume within the inlet region was used in very punctual study (Tanner *et al*., 1999) to estimate the limit of phosphorus retention via particulate settling, which was of 170 g/m² after 5 years of operation in New Zealand.

IV.3 EXPERIMENTAL METHOD

IV.3.1 Sampling layout

Chapter I described the horizontal Flow TW system which is investigated in this study. The sampling layout for total nitrogen, nitrates and total phosphorus is the same as described in Chapter III which was applied for BOD and COD sampling.
The important aspects to keep in mind for this chapter are:

- Influent is delivered on subsurface layer and is not a surface discharge,
- Stones of 4-12 cm are used in the distribution and collection zones, followed for the distribution zone and preceded for the collection zone with large gravels of 10-20 mm and the treatment zone is filled with pea gravels of 3-8 mm;
- Pea gravels are not calcareous but siliceous,
- The system is composed of two cells in parallel, each of which is 13,4 m long, 4,2 m wide and 0,8 m deep;
- Water depth is 0,60 m;
- The two beds are planted with *Phragmites australis sp*.

The operational challenges resulting from the low loading rate applied on the CW, due to management troubles for starting of the guest house and the high variability of the attendance depending on tourist season and meteorological weather, were the same as previously described for BOD and COD measurements. Water samples were collected once or twice a month inside the TW, influent and effluent samples were also collected. Water samples were monthly collected during winter and spring season inside the wetland due to the low load applied, as the rural guest house was not much visited during the low tourist season. Samples distribution was spread between 4 points over width, with 2 points in each cell, and 3 points along the flow path of 3 m , 6 m and 11 m from the inlet and at three depths, 0,15 cm , 0,35 cm and 0,55 cm from the surface.

As a reminder, the sampling campaign started on June 2006 and ended on October 2008. On September 2007, one cell was isolated and received no more influent from the rural guest house. The purpose was double: (i) to increase the organic loading rate of the TW which was very low due to the low starting of the activity of the guest house and (ii) to isolate one cell to conduct tracer test experiments developed in chapter II. Sampling and analysis continued for the remaining cell, and every sample was a mix of 2 points over width instead of the previous 4.

IV.3.1.1 Biofilm sampling for microbial analysis

The bed media (gravels and roots) was sampled at the three locations over length and with the depths of 15 cm, 20 cm and 40 cm. Gravels were collected with a graduated sanitary tube which is driven down in the gravels. Gravels are manually extracted and collected per location over length and with depth in dark glass bottles. Samples are refrigerated at 4°C and analysed at laboratory within five days. Figure 55 shows sampling material (b) and field operation (c).

IV.3.1.2 Total Nitrogen, Nitrate and Total Phosphorus sampling

One sample collecting water was of one depth over one distance, and was a mix of four points over width, having two points in the two cells. Influent and effluent samples from the wetland were also collected, one at the entry and one at the exit (a mixed sample of each cell). Each collecting campaign totalled 11 samples.

The sampling systems were small piezometers (permanently installed): these consisted of small PVC pipes (such as those used for sanitary conduits) of 2 to 3 cm of diameter. One tip section was perforated with holes on its periphery and closed with an end plug, driven into the gravel of the TW. Water was collected with a manual vacuum system; a first release empties the stagnant water, and then water was collected directly into the sample bottles (Figure 55 (a)). Samples were stored inside dark glass bottles, and placed in a fridge-box (at 4°C) till their arrival the same day at the laboratory. Usually, samples were collected in the morning and the laboratory analyses started in the afternoon.


(a)


(b)


(c)

Figure 1

Figure 55: (a) sampling of TN, nitrate, TP with manual vacuum system into dark glass bottles, (b) graduated sanitary tube, gloves and dark glass bottles to sample bed media for biofilm analysis; (c) manual extraction of the media for biofilm analysis

IV.3.2 Laboratory analysis for microbial biofilm

In order to set the biofilm attached to gravels and roots in an aqueous solution, 10 g of gravels are added to 10 ml of Ringer solution (isotonic solution adapted to avoid osmotic shocks when bacteria are removed from their natural environment) and shake for one minute with a vortex (Genie II, STUART brand) set on power 6 (1800 T/min).

IV.3.2.1 Respirometry

Respirometry measures the oxygen rate consumed by aerobic microorganisms to assess their activity. 40 g of gravels are set in a 100 ml solution saturated with oxygen, an oxygen probe is connected to an oxymeter (Oximeter OXI 323, WTW brand) through a sealed cap. The oxymeter is connected to a recorder with a paper band where the concentration of oxygen is draft over time with a regular decreasing slope. The slope drawn is proportional to the microbial activity. Three blank samples are also prepared and measured to check that no intrinsic oxygen is provided by the device. Depending on the exact volume of demineralised water saturated with oxygen, the amount of oxygen breathed in is expressed by gram of calcined gravels per gram of volatile matter.

IV.3.2.2 Nitrifying and denitrifying bacteria in biofilm

The method used to quantify bacteria is the most probable number method. 40 g of gravels are diluted in 20 ml of Ringer solution and shake with the vortex to prepare the initial solution; 7 dilutions are made in vials (from 10^{-1} to 10^{-7}) out of this initial one and replicated three times. The three replications are prepared with three different media: two media are favourable to the growth of bacteria responsible of nitrification with the production of nitrite and nitrate, the third is to observe denitrifying bacteria; they are seeded with the 7 different prepared dilutions. All vials are incubated for 3 weeks. The presence of nitrites is obtained by the reaction with specific reagents (Griess 1 and Griess 2). If no nitrite is observed, a zinc powder is added, which reduces nitrates in nitrite (reacting then with the reagents). Denitrifying bacteria are observed by the production of gas trapped in the Durham small bell. This method has the disadvantage that it can be a mix of gases and not N_2 only issued from the denitrification process. According to the bacteria observed per dilutions, a characteristic profile corresponding to the most probable number is given by the statistic tables from Mac Grady, and reported to the initial solution.

IV.3.3 Laboratory analysis for chemical compounds

The laboratory analysis for TN and TP are the chemical kits 'Test'N Tube^{TM} vials' method, provided by HACH, with the reactor and the colorimetric spectrophotometer DR2500 Odyssey, using the COD reactor for heating. These laboratory analyses are accepted for reporting by USEPA for wastewater. The nitrate content were measured by the onsite colorimeter box 'Nitracheck'.

IV.3.3.1 Total Nitrogen

For low Total Nitrogen range (0,5-25,0 mg/L TN), the method 10071 is applied while for high TN range (10-150 mg/L TN) the method 10072 is applied. They are both based on Persulfate Digestion Method. This test is technique sensitive due to upside-downs inversion of the vials and was always completed by the same operator during the whole study.
Vials are provided with an initial Total Nitrogen Hydroxide Reagent; one powder pillow of Nitrogen Persulfate Reagent is added to vials and 2 ml of sample to analyze are added to the so prepared vials (0,5 ml in case of method 10072); one vials is added with deionized water, instead of wastewater, which is included in the kit as reagent blank; they are all set for 30 min into the COD reactor warmed up at 105°C. The alkaline persulfate digestion converts all forms of nitrogen into nitrate. After 30 min, all vials are removed from the reactor and cooled down to room temperature. Sodium metabisulfite, provided by reagent powder pillows, is added after the digestion to eliminate halogen oxide interferences into two steps. Tubes are shaken for 15 seconds and let for a 3 min rest before restarting a second time, with a 15 sec shake and 2 min rest. Out of those vials, 2 ml are taken and added to the second set of vials provided for the test within the kit. They content a chromotropic acid and nitrate will react under the strong acidic conditions to form a yellow complex. Vials are inverted 10 times to mix, they are getting warm due to the chemical reaction. As the reaction begins, the yellow color intensifies. Results are read after 5 min of reaction time with an absorbance maximum at 410 nm. The calibration is made by the prepared blank vial.

IV.3.3.2 Nitrate

Nitrate are measured by the colorimeter for onsite tests "Nitracheck 404" provided by the French factory *Challenge Agriculture*. A reagent strip "Nitra-test" from MERK factory is inserted in the colorimeter box before any measure for calibration related to local temperature. The method is technique sensitive and has to respect a strict timing. The measures were always done by the same laboratory operator for the whole study. The reagent strip is dipped in the wastewater sample for 3 seconds; it is drained during 5 seconds and let rested for 1 minute. Within the 10 last second of the resting minute, the strip is inserted into the measure slot of the colorimeter box. The concentration is given in mg NO_3^-/l. Three replications must be made per sample with less than 10% of deviation between them. If a higher deviation is observed, the test must be replicated.

The reactive strips have 2 reactive zones: one is for analysing of NO_3^- content and the second one is to detect any interfering substances, due to nitrites. If the colour of the second reactive zone changes, the current test is not valid and the analyses has to be done following laboratory procedure. The nitracheck test ranges from 5 to 500 mg NO_3^-/l ; no interfering substances have ever been observed in this case study.

IV.3.3.3 Total Phosphorus

The analyse of Total Phosphorus is made by the method 8190, which range from 0,02 to 1,10 mg TP/L or 0,06 to 3,5 mg/L PO_4^{3-}. Higher concentration requires the dilution of the samples. The method is based on PhosVer®3 with acid persulfate digestion method.
5 ml of the samples of wastewater to analyse is added to the vials provided by the kit containing an acid hydrolysable medium, as well as a powder pillow of potassium persulfate for phosphonate. Vials are shaken to dissolve the reagents and placed into the COD reactor warmed up at 105°C for 30 minutes. The vials are then removed form the reactor and cooled down to room temperature. Orthophospate reacts with molybdate in an acid medium to produce a mixed phosphate/molybdate complex. 2 ml are of 1,54N Sodium hydroxide standard solution is added to the vials, which are capped and mixed. Ascorbic acid then reduces the complex, giving an intense molybdenum blue colour. Test results are measured at 880 nm, and calibrated by a blank prepared vial (without wastewater but deionised water).

IV.4 RESULTS AND DISCUSSION

As the Walloon region is not subject yet to any regulation either on any nitrogen form or phosphorus content for treated wastewater discharged to the environment for small treatment plants, nitrogen was studied and analysed in the frame of this work based on Total Nitrogen (TN) and nitrate (NO_3^-) and phosphorus for Total Phosphorus (TP) only. Investigation of the total nitrogen cycle or the phosphorus forms over length and with depth would have required complete studies by themselves which would also require higher laboratory equipments. The current work focuses on the global removal processes for TN and TP occurring along length and with depth. The additional analyses on microbial fauna corroborate the removal processes locations.

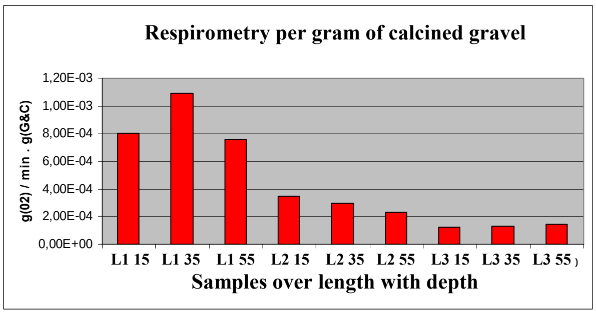
Data and results of TN, TP, and nitrates are presented and discussed. Total Nitrogen results are inserted into the *P-C*-k* model developed to provide the degradation rates coefficient (*k*-values). Results of microbial study are presented and related to observed results of TN, nitrate, TP but also BOD and COD removals.

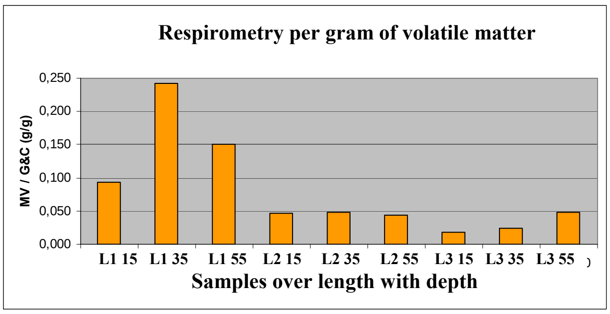
IV.4.1 Microbial biofilm analysis

IV.4.1.1 Respirometry

Figure 56 presents the results of respirometry activity reported to 1g of calcined gravel (Figure 56a) and reported to 1g of volatile matters (Figure 56b). When respirometry is reported to calcined gravel, it is reflective of the activity of the bacteria composing the biofilm but also of the amount of biofilm attached to the sample and is named the 'total respirometry'. It could reflect e.g. a biofilm having a low activity but being important in term of quantity. In order to obtain the 'real respirometry' of the biofilm, results are expressed based on volatile matter.

Figure 56a demonstrates a higher activity on the first meter of the TW, decreasing with length. Variations in respirometry rates are also observed with depth, with a higher activity on top layers than on the bottom layers. This is related to the aerobic bacteria involved in this measures which are more numerous closer to surface, having the benefit of gases exchanges with the atmosphere than on the bottom of the bed. The lower peak observed at the surface sample from the inlet is explained by the low level of the wastewater in the bed when the sampling was done, resulting in a dried sample, having suffered from drought.





(a) (b)

Figure 56 : Respirometry reported to one gram of calcined gravel (a) and reported to one gram of volatile matter (b)

When respirometry is based on the volatile matter (Figure 56b), the general shape of the diagrams remain the same as for figure 56a but peaks for surface samples are strongly decreased. This demonstrates that the high peaks of figure 56a are the result of a large

quantity of biofilm rather than due to a high activity. Due to the dried sample of surface layer close to inlet, its low measurement phenomenon is accentuated from figure 56a to figure 56b. Graph 56b also shows the highest activity and amount of biofilm is located close to inlet in middle layer; the activity is less influenced with depth in the middle of the bed and still has a good activity at the end of the bed at the bottom layer.
The respirometry activity measured does not provide any indication about the type of organisms which are involved in the respirometry. Microscope observations confirmed the higher presence of protozoa and metazoans on surface samples and also at the inlet of the bed. Theses observations could suggest that beyond aerobic bacteria involved in this measure, microscopic fauna is also probably responsible of higher values of respirometry on surface samples.

IV.4.1.2 *Nitrifying and denitrifying bacteria*

Figure 57 presents results of the count based on Most Probable Number (MPN) of bacteria for nitrification (nitrite and nitrate) and denitrification. Globally, the high amount of bacteria found demonstrates the good potential of the bed to nitrify and denitrify. Nevertheless, the different magnitude order of figure 57c next to figure 57a and 57b shows the domination of dentrifying bacteria next to nitrifying ones.

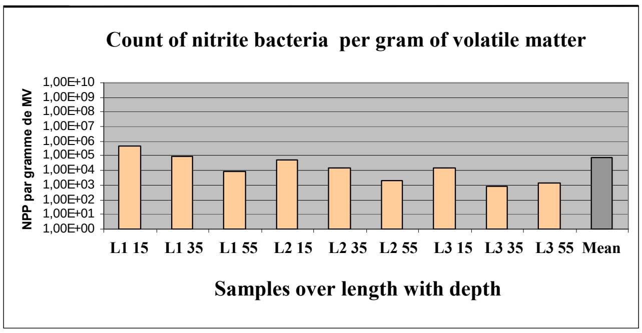


Figure 57 (a) : Count of nitrite bacteria
Bacteria responsible of nitrites (figure 57a) and nitrates (figure 57b) both display a peak for the surface samples, whatever the location over length. This is coherent with the fact that they are aerobic bacteria.
Average MPN = 10^4

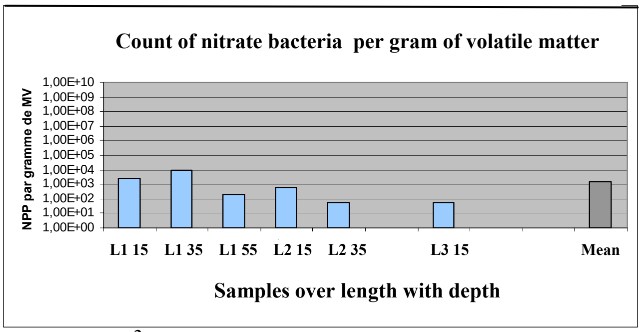


Figure 57(b): Count of nitrate bacteria
Bacteria are more numerous for nitrite then nitrate, some results for nitrate are non detected. No specific explanation can be given except that it confirms the low ability of horizontal flow system to nitrification and also the short cut existing for bacteria responsible of denitrification, starting it at the nitrite stage. Average MPN = 10^3

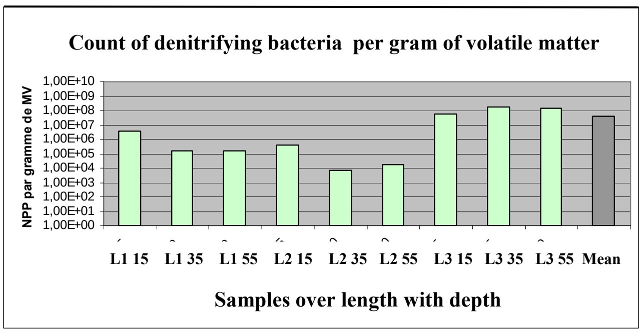


Figure 57(c): Count of dentrifying bacteria
Denitrifying bacteria are much more numerous with an average MPN =10^7.
Depth was not found as having any influence on the number of denitrifying bacteria found in the bed. This is reflective of the fact that denitrifying bacteria can work aerobically or anaerobically. An increase of bacteria is observed with length, what could be related with the low organic loading rate, allowing nitrification to occur due higher aerobic conditions within

the first part of the bed and having a higher denitrification processes occurring on the second part of the bed.

This study was lead before the closing of one cell of the system, and the organic loading rate of the bed was still very low. The deviation of all wastewater flow over one single cell has increased the organic loading rate for the sampling campaign done in the frame of this work.

IV.4.2 General presentation of data of TN, nitrate and TP

Table 16 presents then results of chemical analysis with the regular statistical indexes of mean, standard deviation, minimum, maximum values and the number of samples on which results are based. The sampling campaign started (as for BOD COD and TSS) on June 2006 and ended on October 2008 and had more or less a monthly base. The number of samples varies from 31 to 35 for TP and TN, according to laboratory problems or lack of surface samples. The number of samples for nitrate is lower as it is a measure which started later.

IV.4.2.1 Total Nitrogen

Total Nitrogen is defined as the combination of organic, ammonia and oxidised nitrogen and is subject to consideration as a group of compounds that are reduced in TW. This group is known to possess sequential conversions including primarily ammonification, followed by nitrification, followed by denitrification, all proceeding at varying rates. In this case study, as the TW is a secondary treatment, it is expected to observe high TN removal rates for nitrified effluent, as the precursor conversion of organic and ammonia should already have occurred in pretreatment, especially when the system is low loaded and having the additional distribution channel providing high gas exchanges (aerobic conditions favourable to nitrification) before entering the TW.
The mean value of influent and effluent is 89,9 mg TN/L and 21 mg TN/L for the effluent. The standard deviation of the influent is of 100 and 21 respectively, what shows a higher variability in the influent TN than in the effluent concentration. Median values are much lower, 63 mg TN/L for the influent and 13 mg TN/L for the effluent. Minimum values are providing the background concentration, decreasing along the bed. Maximum values are impressively high, being more than 6 times the mean value with 540 mg TN/L at the inlet, and 5 times the mean value at the outlet with 96 mg TN/L. Total removal percentage, based on concentration, is of 77 % at the outlet. Based on standards for larger treatment plants, the removal efficiency percentage is satisfying but total nitrogen is higher than the acceptable level of the norm.

IV.4.2.2 Nitrate

The mean value of the influent is low (7 mg NO_3^-/L), with a low standard deviation (8) which reveals very stable conditions. Having a low value of TN at the outlet (21 mg TN/L) and low nitrate value (6 mg NO_3^-/L) demonstrate that nitrification occurred either prior to the entrance of the wastewater in the TW bed or along the flow path inside the bed and denitrification must occur too, providing low nitrate residues as well as TN at the outlet of the system. Median, maximum and minimum values are all very close, varying from background concentration, or detection limit of laboratory analysis, of 1 to values of 7 mg NO_3^-/L which cannot be considered as high.
Results of bacteria analysis here above presented combined with the results of TN and nitrate can lead to the assumption that nitrification and denitrification probably occur simultaneously in the system as TN decreases from the inlet to the outlet. The nitrogen still present at the

outlet is probably not nitrified with nitrogen issued from the plant biomass cycle of the system. Ammonia levels could still be too high for specific regulation, but cannot be assessed by the data gained from this sampling and measurement campaign. The Walloon regulatory limit of 50 mg NO_3^-/L is respected at the outlet of the system.

IV.4.2.3 Total Phosphorus

The mean value of influent concentration in TP is of 21 mg TP/L with a maximum value of 93 mg TP/L. This value quite high and is likely induced by the main activity of the guesthouse as a restaurant, using a lot of chemical products for dishwashing, still containing phosphorus in Belgium. Values are very quickly decreasing and follow the hydraulic pattern identified at chapter II, as higher values are observed in bottom layers. The bed was still young (2 years of operation at the beginning of the sampling campaign) and the sorption capacity of the media is still high as the value of TP at the outlet is of 0,8 mg TP/L. Results presented further will show the exhaustment rate of the media sorption capacity of the bed. The standards are still respected for both removal percentage and TP concentration of the effluent, based on standards for larger wastewater treatment plants.

Table 16 : General results of the monitoring campaign

		Distance	Inlet	3 m from inlet			6 m from inlet			11 m from inlet			
Parameter	**Stat**	**Unit**	**Depth**	**0,15**	**0,35**	**0,55**	**0,15**	**0,35**	**0,55**	**0,15**	**0,35**	**0,55**	**Outlet**
N Tot	n		34	34	34	34	32	35	34	34	31	34	31
	Mean	mg/l	89,9	47,9	43,2	57,3	30,4	39,8	52,8	23,5	20,2	48,4	20,9
	Std Dev		100,4	54,9	32,5	43,8	38,0	37,7	51,6	25,7	15,2	52,3	20,8
	Median	mg/l	63,0	25,5	31,5	46,5	11,5	24,6	36,0	14,7	17,0	32,0	13,3
	min	mg/l	3	2	5	10	2	2	3	1	1	1	1
	Max	mg/l	540	240	118	210	139	141	290	100	71	280	96
	Remov.	%		47	52	36	66	56	41	74	78	46	77
Nitrates	n		38	37	37	36	37	37	36	38	37	36	36
	Mean	mg/l	7	5	5	5	5	6	6	7	6	6	6
	Std Dev		8	4	5	6	3	6	6	5	5	7	4
	Median	mg/l	6	5	5	5	6	6	5	7	6	6	7
	min	mg/l	5	1	1	2	2	1	1	1	1	1	1
	Max	mg/l	11	1	3	5	6	1	2	5	6	1	1
P tot	n		35	34	35	35	32	35	35	33	33	32	35
	Mean	mg/l	21,3	4,0	4,7	4,9	2,7	3,9	4,3	1,2	1,2	2,6	0,8
	Std Dev		22,1	3,1	2,6	2,5	3,2	3,5	3,1	3,3	1,5	2,7	1,8
	Median	mg/l	14,0	3,1	4,4	4,6	1,2	2,6	3,2	0,3	0,5	1,4	0,3
	min	mg/l	0,4	0,2	0,1	0,4	0,1	0,3	0,5	0,1	0,1	0,0	0,0
	Max	mg/l	93,0	12,0	12,9	9,5	10,3	12,0	12,0	19,0	5,1	12,1	10,1
	Remov.	%		81	78	77	87	81	80	95	94	88	96

IV.4.3 Total Nitrogen (TN) results

The reduction in TN in TW can be represented by loading analysis and the *P-k-C** model. Loading analysis is presented by frequency percentile distribution and In/Out loading charts. *P-k-C** model presents the degradation coefficient rates obtained.

IV.4.3.1 Frequency percentile

The typical method to present treatment performance is the probability distribution for concentration entering and leaving the TW system. Figure 58 displays the graph for inlet and outlet. The median TN inlet (50th percentile) is 63 mg TN/L, while the median outlet is 13 mg TN/L, as reported on Table 16.

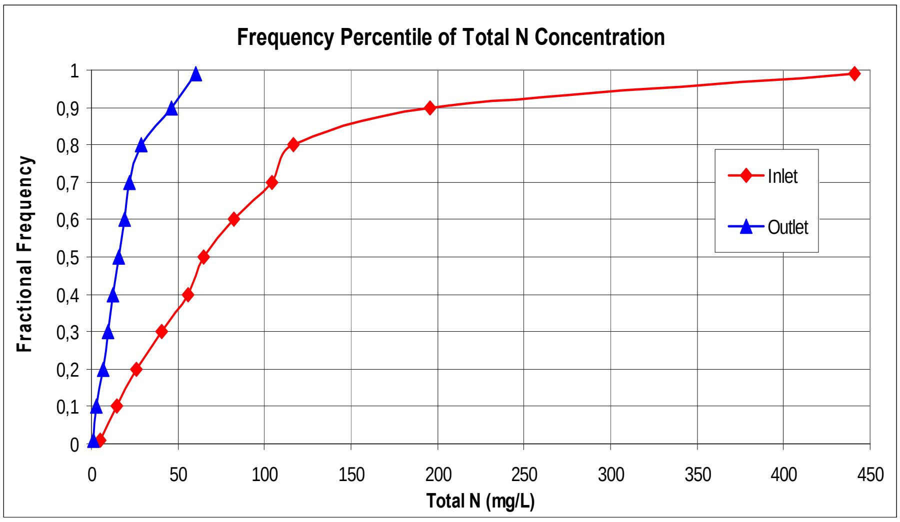


Figure 58 : Distribution of Total Nitrogen concentration for inlet and outlet

Figure 59 is showing the same probability distribution for all depths (15, 35 and 55 cm deep) and Lines 1, 2 and 3 located at 3, 6 at 11 m from inlet respectively. It is clear that all curves have a similar shape, what demonstrates again that the main removal is achieved within the few meters of the bed. The same colour code is used as in Chapter II, which is the blue family for Line 1, green for Line 2 and red for Line 3 with light colours for surface layer (15 cm deep) to dark colours for bottom layer (55 cm deep). It shows stratification in performance from the blue family curves (located lower on percentile performances), followed by the green family curves with middle performances and the red family curves at the top percentile performances. The three lower curves are also the bottom layers where concentrations appear to be higher. Chapter II has shown that flow rates are more important on the bottom which indicates in terms of removal efficiency that the bacteria located on the bottom layer have to work more to achieve the same results as on surface layers, based on loading rates rather than concentration content. Degradation *k*-rate values will allow us to quantify the efficiencies over length and with depth.

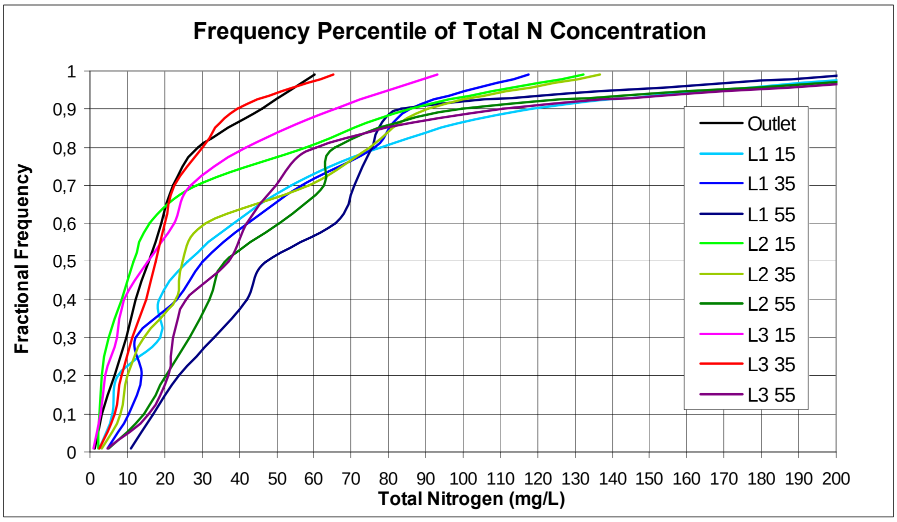
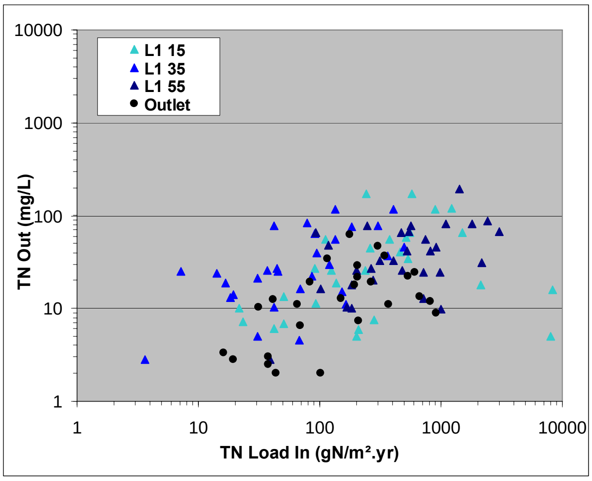


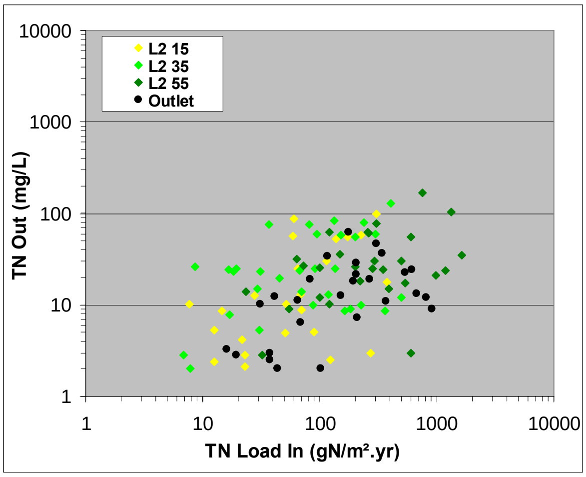
Figure 59 : Distribution of Total Nitrogen performance over length and with depth

IV.4.3.2 In- and Out- concentrations

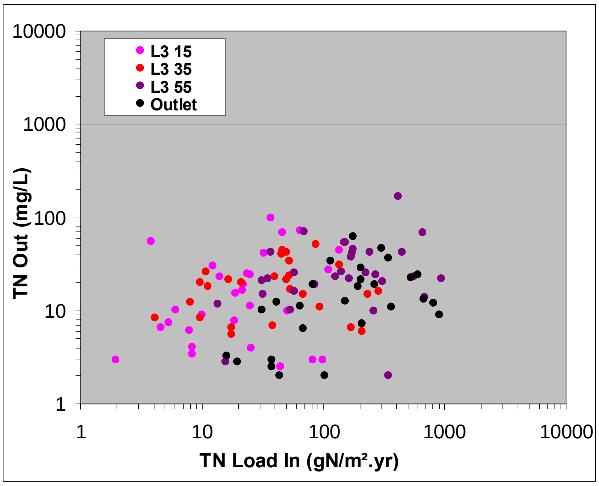
The other usual graphical displays which are adopted in the TW literature plot outlet concentration in mg/L versus inlet concentration loading (Inlet Loading Rate – LRI). LRI is the concentration multiplied by the Hydraulic Loading Rate and is expressed in g/m².year. Figure 60 is split into the 3 graphs corresponding to Line 1, Line 2 and Line 3 at 3 m, 6 m and 11 m from the inlet respectively. The same colour code per family (Line1 = blue, Line 2= green; Line 3= red) is reproduced with darker colours corresponding to lower layer. X and Y axes are all similar and on logarithmic scale to avoid scatter cloud of points and to show a more consistent central trend. The data are all showing the trend of increasing Concentration Out with increasing LRI, having overall slope slightly less than 1,0 and similar for all graphs (a, b and c) variable in distance from inlet.



(a) TN input-output for Line 1



(b) TN input-output for Line 2



(c) TN input-output for Line 3

Figure 60 : Total Nitrogen concentration input versus output per lines over depth

IV.4.3.3 Model application and k-values

As developed in Chapter III, the major purpose of this work was to investigate what occurs and where inside the TW. Chapter II has identified the water flow pattern and this chapter is devoted to pollutant removal. The degradation rate for TN (*k*-values) is calculated in order to point out differences in TN degradation rates over length and with depth.

The previously defined relaxed *P-k-C** TIS concentration model has been applied to all TN data. The mathematical relationship is given by Equation 11 in Chapter III, repeated here as Equation 73:

$$\frac{(C_0 - C^*)}{(C_i - C^*)} = \left(\frac{1}{(1 + k/Pq)^P}\right) \qquad \text{(Eq. 73)}$$

Where,
- C^* = Background concentration (mg/L)
- C_i = inlet concentration (mg/L)
- C_o = outlet concentration (mg/L)
- k = apparent TIS rate constant (m/yr)
- P = apparent number of TIS
- q = hydraulic loading rate (m/yr)

This equation has been applied for all TN data and provides the overall *k*-rate constant, which is the single unknown of the equation.
The C* (mg/L) value is taken as constant per line and per depth and is the value presented in Table 16 as the minimum TN concentration, corresponding to the isolated cell receiving rainwater only, and sampled prior to the tracer test experiments.
C_i (mg/L) values are the data collected of the inflow TN concentration varying with all dates of measurements.
C_O (mg/L) values are the values of TN concentration observed for all lines and depth, also varying with all dates of measurements.
The apparent number of tank in series *P* (dimensionless), is based on the *NTIS* calculated in Chapter II and divided by 2 as suggested by Kadlec and Wallace (2009). The *NTIS* represents the hydraulic number of TIS and *P* represents the organic TIS. The movement of organic or mineral particles is known as being slower than the water movement within the bed, as described previously. It worth being mentioned that according to the *NTIS* found in Chapter II and according the division by a factor 2 as suggested by literature, the number of *PTIS* is much above other any literature sources, presenting usual *PTIS* varying from 3 to 6 at the maximum.
The HLR *q* (m/yr), is the inflow rate (m^3/yr) (as defined by the water budget) and given by Table 17, divided by the surface area (m²). As reported for the water budget in Chapter II, *q* is varying for all dates of measurements and is given by the inflow (m^3/yr) weighted according to fraction of the flow identified per layer in Chapter II, combined with precipitation (m^3/yr) which have been weighted according the area related to the position of the concerned line of sampling over length and minus the evapotranspiration (ET) also varying with the position of the concerned line over length (ET is ¼ of total ET for line 1, ½ of total ET on line 2 and ¾ of total ET on line 3).
The *k*- value (m/yr) is given by the solution in Chapter III (also Eq. 73 here above).
The *k* degradation rate constants obtained are presented with the following frequency percentile table with statistics and figures here below (Table 17, Figure 61).

Table 17 : Annual rate coefficient k (m/yr) for TN per lines and depths, percentile and statistics results.

Line - Distance from Inlet / depth			**L1**	**3**	**m**	**L2**	**6**	**m**	**L3**	**11**	**m**	**OUTLET**
	Nom.	**Units**	**15**	**35**	**55**	**15**	**35**	**55**	**15**	**35**	**55**	
Data												
Inlet Conc.	**Ci**	**mg/L**	2,8 - 540									
Background Conc.	**C***	**mg/L**	1,5	1,0	2,0	1,5	1,0	1,5	2,0	1,5	1,0	1,0
Apparent TIS	**P**		3	2	1	10	6	10	6	16	17	13
Number of TIS	**NTIS**		6	4	2	19	12	19	12	32	34	26
HLR												
Mean		**m/yr**	3,7	1,2	7,4	0,9	1,2	4,0	0,3	0,7	2,3	2,4
SD		**m/yr**	3,0	1,0	6,1	0,8	1,0	3,3	0,3	0,6	1,9	2,3
Median		**m/yr**	2,5	0,8	5,1	0,6	0,8	2,7	0,2	0,5	1,6	1,9
min		**m/yr**	0,6	0,2	1,3	0,2	0,2	0,7	0,1	0,1	0,4	0,5
Max		**m/yr**	11,6	3,9	23,2	2,9	3,9	12,6	1,1	2,2	7,3	8,6
k-Values Percentile												
0,05		**m/yr**	0,4	0,0	0,2	0,1	0,0	0,1	0,0	0,0	0,0	0,0
0,1		**m/yr**	1,0	0,0	1,7	0,3	0,1	0,6	0,1	0,1	0,2	0,6
0,2		**m/yr**	1,6	0,3	2,3	0,7	0,4	1,1	0,2	0,2	0,7	1,5
0,3		**m/yr**	2,1	0,4	3,6	0,9	0,6	1,6	0,3	0,3	0,9	2,2
0,4		**m/yr**	2,7	0,7	4,8	1,1	0,9	2,9	0,3	0,4	1,5	2,8
0,5		**m/yr**	3,1	0,8	6,9	1,5	1,0	3,9	0,4	0,5	1,8	3,4
0,6		**m/yr**	3,8	1,3	12,8	1,8	1,4	4,3	0,5	0,7	2,1	4,1
0,7		**m/yr**	4,3	1,7	14,5	2,1	1,8	4,7	0,7	0,9	2,5	4,4
0,8		**m/yr**	9,9	2,3	20,5	2,9	2,3	10,3	1,2	1,7	4,1	7,1
0,9		**m/yr**	19,1	5,4	41,9	5,4	6,8	17,9	1,7	3,3	8,7	10,6
0,99		**m/yr**	129,1	7,6	91,9	12,5	11,4	26,6	6,2	6,1	16,1	17,5
Statistic data												
Number of samples	**N**		30	34	31	28	31	31	32	29	31	29
Mean	**Mean**	**m/yr**	17,0	1,7	15,4	2,5	2,2	6,0	0,9	1,2	3,3	4,7
Standard Deviation	**SD**	**m/yr**	45,3	2,2	22,2	3,1	3,0	7,0	1,5	1,6	4,3	4,6
Median	**Md**	**m/yr**	3,8	0,8	6,9	1,5	1,0	3,9	0,4	0,5	1,8	3,4
minimum	**min**	**m/yr**	0,4	0,0	0,1	0,1	0,0	0,0	0,0	0,0	0,0	0,0
Maximum	**Max**	**m/yr**	238,1	7,9	108,0	13,0	11,5	29,2	6,3	6,5	17,1	18,3

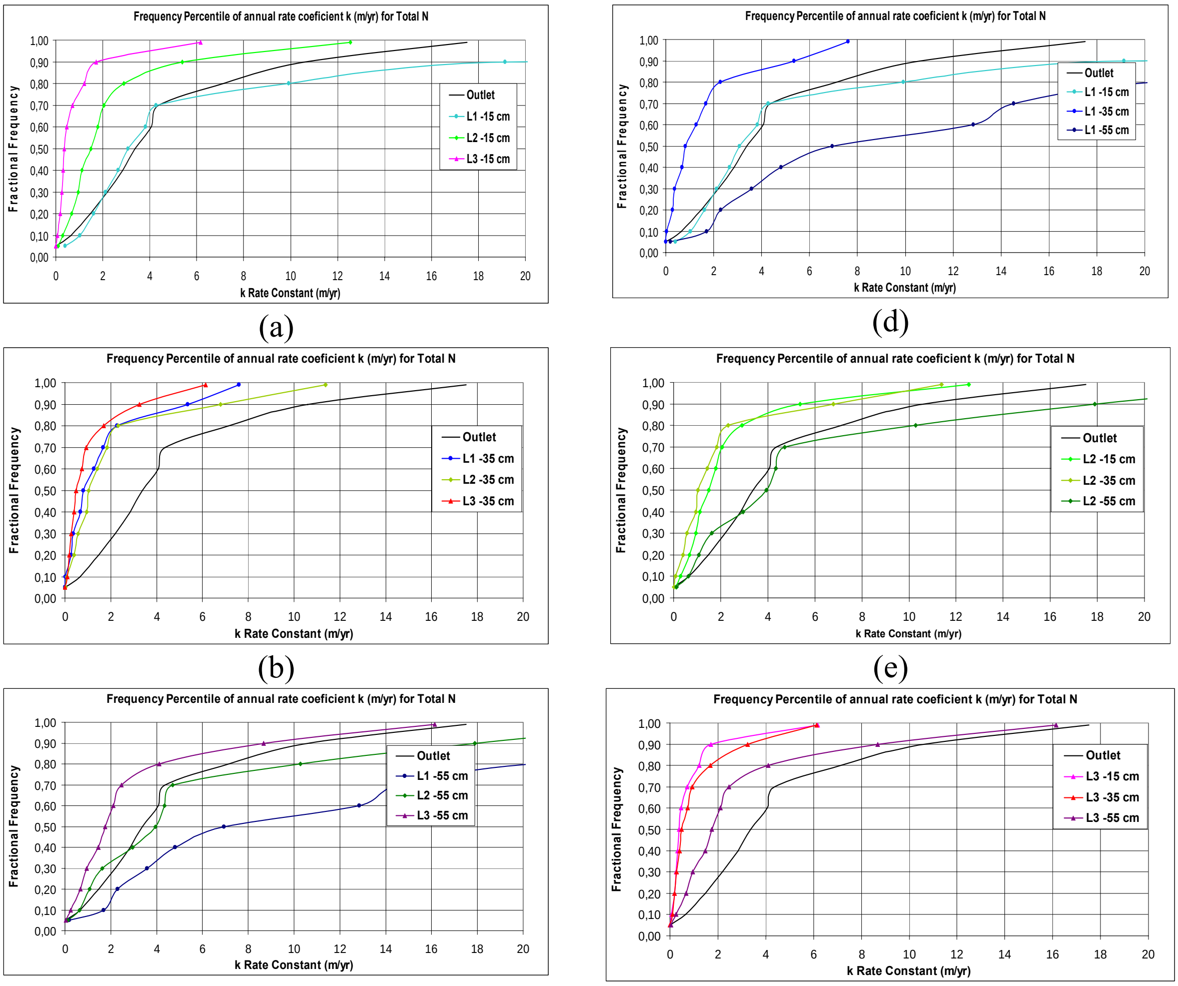
Table 17 shows that the influent concentration ranges from 3 to 540 mg TN/L what is above the usual range of 20-85 mg TN/L of raw municipal wastewater (Kadlec and Wallace, 2009).
The hydraulic loading rate (HLR) shows variability of a factor 3 for the influent concentration between mean value and maximum ones as well as for the outlet. It is reflective of the typical small scale decentralised system implemented in rural areas having seasonal, weekly, monthly or other high and low loading rate peaks.
The mean value for *k*-rate coefficient calculated by Kadlec and Wallace (2009) to evaluate the wetlands performance with data from 112 HF TW systems, is of 8,4 m/yr. The *k*-rate values presented in Table 17 are lower, except on Line 1. This can be explained by the design of the TW system which is investigated. The system is long and narrow what leads to a high number of tank in series (*NTIS*) and thus also high values for the apparent number of tank in series (*PTIS*). Kadlec and Wallace have fixed *PTIS* to the value of 6. In this case, *PTIS* values are much higher (up to 34) what induces lower *k*-rate values when mathematical equation given by the model is solved.
The percentile distribution is graphically depicted by Figure 61.
The statistical analysis is a variance analysis with 2 fixed factors (depth and distance from inlet), and 1 random factor (sampling) with confidence interval range of 0,01 and checks the equality of mean values. Comments are the following after the exclusion of a few erratic data.

- Median values corresponds to 50th percentile and have systematically a magnitude of order of 2 smaller than mean values.
- All Lines present significant difference between them.
- Depths do not present significant different *k*-rate values on Line 1, Line 2 or Line 3.

The global trend which emerges from those results is a slow and regular decrease of the degradation rate for TN with length with systematically higher degradation coefficients for the bottom layers, decreasing also over length.

Higher coefficient rates on the bottom layers related to dentritrifying bacteria and the majority of the flow creeping along the bottom of the bed could indicate that nitrogen degradation is achieved for a large part with anaerobic processes occurring on the bottom layers, what is the usual trend for horizontal flow systems.

Figure 61 is providing the frequency distribution profiles of *k*-rate percentiles. The Figure has to be read by columns. The left column presents results according to depth (Figure 61a to 61c) and the right column presents results according to Lines (Figure 61d to 61f). The same colour code is kept as previously with blue family curves for Line 1, green family for Line 2 and red family for Line 3. Darker colours correspond to bottom layers and light colours to surface layers. The outlet is represented on all graphs by the black line.



(a) (d) (b) (e) (c) (f)

Figure 61 : Distribution frequency percentile of annual *k* rate coefficient for Total Nitrogen

It shows that from Figure a to Figure c, curves are straighter and closer to each other in the middle layer, having lower *k*-rates and they are more inclined and screening broader *k*-rate values for surface and deep layers.

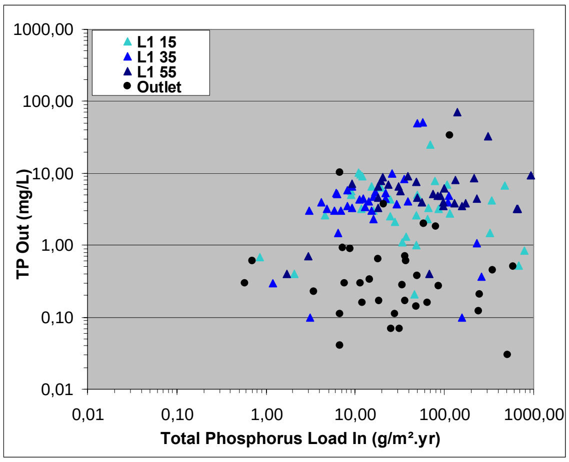
Figure 61d to 61f presents profiles per line and show a general tendency to be more inclined on Line 1 (Figure 63d) and getting straighter with Lines 2 (Figure 61e) and 3 (Figure 61f). For all Lines, curves with the darker colours are more inclined what demonstrates higher *k*-rate constants with depth. The general decrease of *k*-rates coefficients with length attests the regular degradation of Total Nitrogen as the flow passes through the bed. Total Nitrogen cannot provide us more detailed information on the nitrogen processes which are occurring on each depth and length. The combined analysis with bacteria results can lead us to assume that nitrogen is mainly degraded with anaerobic processes occurring on bottom layers.

IV.4.4 Total Phosphorus (TP) results

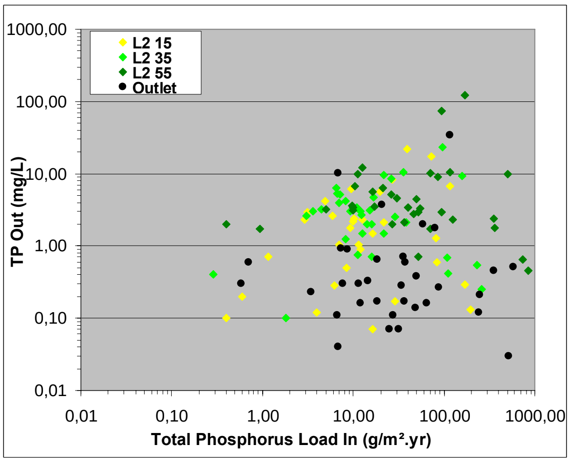
The reduction in TP in TW can be represented by loading analysis through In/Out loading charts. The *P-k-C** model to presents the degradation coefficient rates is not relevant to apply as the main removal processes involved in this case is the media sorption capacity. Phosphorus is usually not a primary design consideration for HF system treating domestic wastewater and the examination of data set reflects this.

IV.4.4.1 In- and Out- concentration

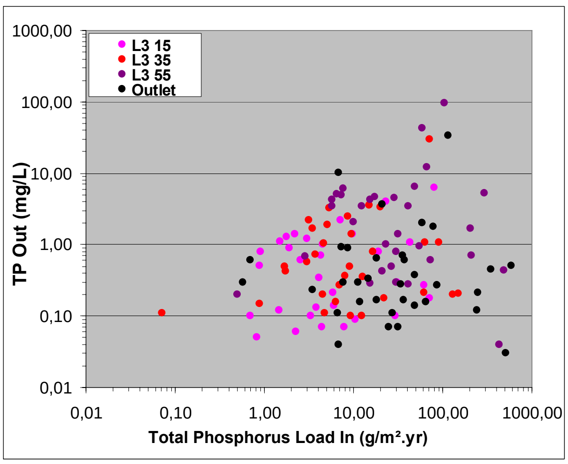
The most useful presentation involves effluent concentration (C_{out} in mg TP/L) versus influent loading – Phosphorus Loading Rate , PLR- (g/m².yr) as shown by Figure 62.



(a) TP input-output for Line 1



(b) TP input-output for Line 2



(c) TP input-output for Line 3

Figure 62 : Total Nitrogen concentration input versus output per lines over depth

Figure 62 shows the potential drawback to combine data over many inlet concentration ranges to produce phosphorus loading rates In (PLRs). Aggregation of the entire data set produces a data cloud that has a general trend line with a slope about 1,0 or lower. But if effluent are classified according influent concentration, the slope of the cloud can be down to 0,2. It indicates that at high PLR, $C_{Out} = C_{In}$ and there is no apparent removal but at low PLR, C_{Out} does not decrease accordingly. It means that if the wetland receives high influent concentration, it can still return high effluent concentration even at low PLRs. The PLR is thus not an independent variable to predict effluent phosphorus concentrations in this type of bed. The conservative assumption at this stage of the technology development is that long-term, sustainable phosphorus removal in HF TW systems is essentially zero ($C_{Out} = C_{In}$) (Kadlec and Wallace, 2009).

IV.4.4.2 Seasonal effect

The growth and die-back of emergent wetland plants impose a cycle of phosphorus uptake and release on HF TW system (Kadlec and Wallace, 2009). Although the phosphorus biomass cycle tends to be small relative to total phosphorus influent loadings, a seasonal trend can be observed, as illustrated in Figure 63.

Two comments arise from the analysis of the red curve displayed on Figure 65:

- The wave effect present low peak during spring and summer months on 2007 when the phosphorus uptake by plants is at the maximum for the macrophytes growth, and higher peak on autumn months on 2007 when the phosphorus is released at the annual die-off of the plants;

- The curve is slowly shifted upward from the starting to the end of the sampling campaign, as the media sorption decreases and the TP concentration at the outlet is increasing.

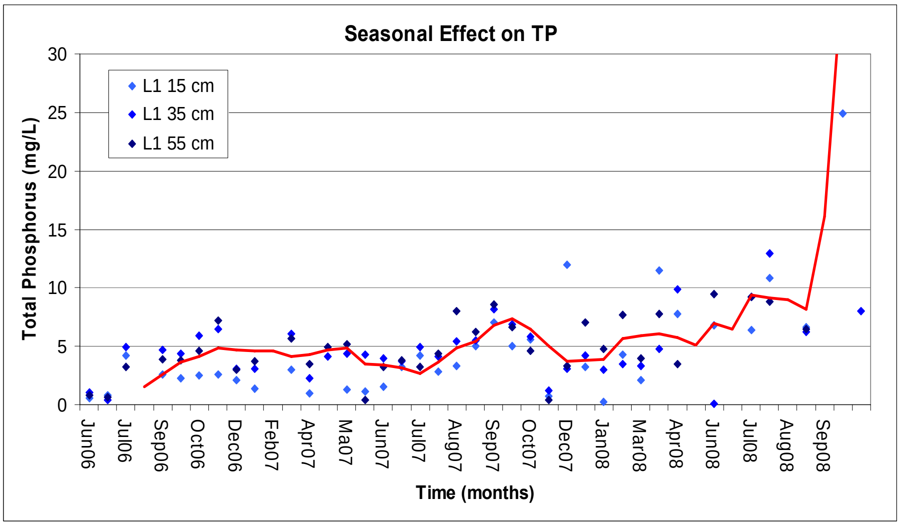


Figure 63 : Seasonal effluent phosphorus trend over the two years of sampling campaign

IV.4.4.3 Media sorption capacity over time

The long-term effluent phosphorus concentrations produced in HF TW are a function of the three primary variables of area, hydraulic loading and influent concentration, as discussed previously. The sorption characteristics of the media are important factors that can be dominant in treatment performances over the initial stage of operation until the sorption capacity is saturated. Figure 64 depicts the capacity sorption exhaustion.

Samples are located on the X axis over length and with depth; Y axis is the measured TP concentration (mg TP/L) and the Z axis is various dates over time. The system started on March 2005 and was in its first stage of operation as the experiment with sampling campaign started (July 2006). First measures of TP were all low for any location and depth of the TW system. The sorption capacity is considered as exhausted when the TP concentration is above 2 mg TP/L, as it corresponds to some specific regulations issued from the Water Frame Work European Directive. The diagram clearly points out that the media was first saturated for the Line 1 located at 3 m from the inlet; on Line 2, the exhaustion was achieved for the middle and bottom layers (35 cm and 55 cm deep) before the surface layer (15 cm deep); and finally, for Line 3, the capacity sorption maximum was achieved for the bottom layer first (55 cm deep) and was still not reached at the end of the sampling campaign for the surface layer (15 cm deep).

The capacity of the media to sorbe phosphorus is clearly connected with the hydraulic study made in Chapter II, showing higher flows on the bottom of the bed. The sorption sites of the gravels were thus logically saturated for bottom layers, starting from the inlet to the outlet, according the wastewater flow and its phosphorus load.

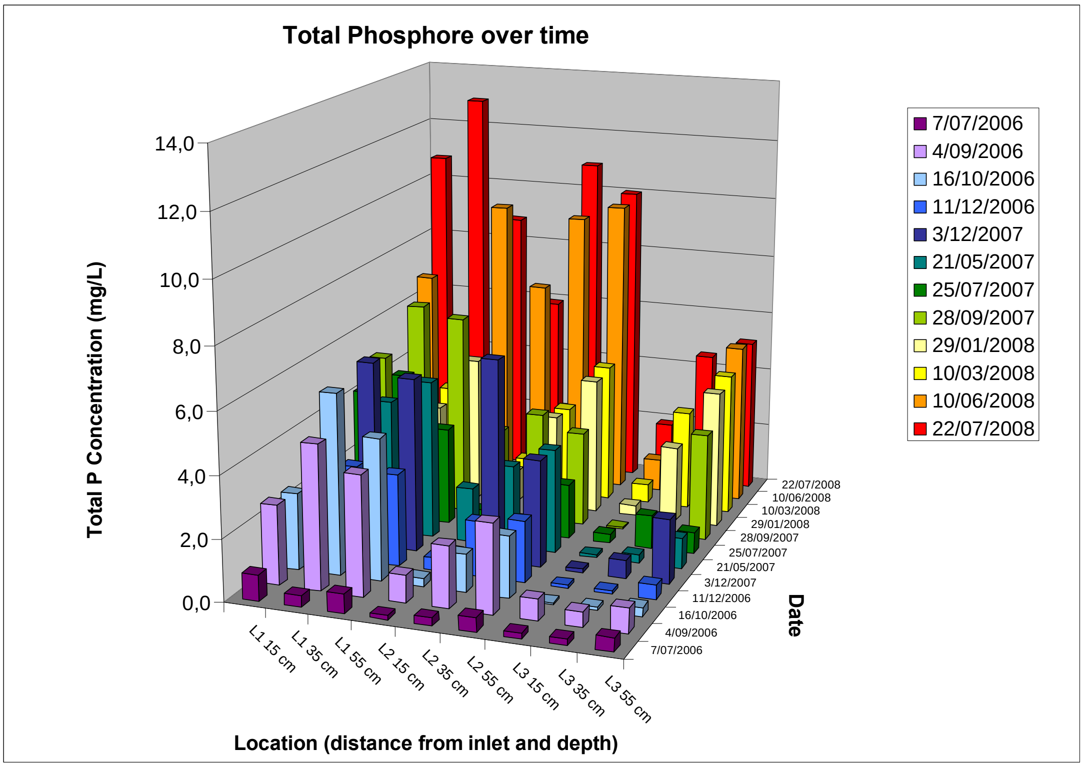


Figure 64 : media sorption saturation according to location (length and depth) over time

The total percentage of TP removal efficiencies in percentage, calculated on loading rates (g TP/m².yr), over the two years of monitoring campaign are given by Table 18. Due to high removal efficiencies at the starting of the monitoring campaign, the removal percentage are still good, but the location and layers where the capacity sorption of the media is decreasing can be pointed out. Table 18 shows that the capacity sorption is probably exhausted on all bottom layers, inducing a drop in removal percentages. The middle layer of Line 1 is also exhausted, being exhausted on Line 2 and starting to be exhausted on Line 3. And for surface layers only Line 3 is still not yet concerned by a saturation of adsorption sites of the gravels.

Table 18 : Total Phosphorus removal efficiencies, calculated on mass loading (g/m².yr)

Lines over Length		L1			L2			L3		Outlet
Depth (cm)	**15**	**35**	**55**	**15**	**35**	**55**	**15**	**35**	**55**	
TP removal (%)	**84**	**74**	**70**	**89**	**86**	**61**	**97**	**94**	**74**	**94**

Table 18 gives the indication that the sorption capacity of the TW system has started to be saturated and according to last measures sampled at the end of the measurement campaign, the system is predicted to be saturated by now.

IV.5 CONCLUSIONS

This fourth and last chapter presents the observed results for total nitrogen and total phosphorus connected with some microbial analysis.
The theoretical background summarised the current state of the art related to nitrogen degradation and phosphorus removal within horizontal flow treatment wetland systems.
The experimental methods for sampling layout, through the set of sampling port tubes inserted permanently in the TW bed and distributed over three location along the length of the bed, always replicated three times in order to sample three depths, were presented as well as laboratory procedures.
Microbial analysis through the total respirometry measures showed that the biofilm had a higher activity within the first few meters of the bed, decreasing with length. Higher repirometry measures are also demonstrating a higher activity of aerobic fauna on surface layer in comparison with bottom layers. The measure of real respirometry demonstrated that the higher activity measured by the total respirometry for the few first meters of the bed, was the result of a higher amount of biofilm rather than due to a higher active biofilm. Those measured provided neither any information about the composition of the biofilm nor any type of organisms constituting the biofilm. Binocular observations have noticed the presence of organisms such as protozoa and metazoans, which are included in the respirometry measures, in addition to aerobic bacteria.
The count of bacteria active in nitrification and denitrification, based on the most probable number method, showed the presence of bacteria involved in nitritation, nitratation and denitrification over all location and depths investigated. Denitrifying bacteria were more numerous in general than nitrifying bacteria. Nitrifying bacteria were more numerous on surface layers; whereas denitrifying bacteria were not dependent with depth, demonstrating that they can be aerobic or anaerobic.
Results of total nitrogen analysis showed that the influent was highly loaded in TN. It severely decreases in the few first meters of the bed and gently goes on regularly decreasing along the length of the bed. The median TN inlet (50^{th} percentile) is 63 mg TN/L, while the median outlet is 13 mg TN/L. The mathematical relaxed *P-k-C** model was applied to calculate the TN degradation coefficient values (*k*) for all the collected samples. The global trend which emerges from those results is a slow and regular decrease of the degradation *k*-rate for TN with length, having systematically higher degradation coefficients for the bottom layers, decreasing also over length. Higher coefficient rates on the bottom layers related to dentritrifying bacteria and the majority of the flow creeping along the bottom of the bed could indicate that nitrogen degradation is achieved for a large part with anaerobic processes occurring on the bottom layers, what is the usual trend for horizontal flow systems.
The analysis of total phosphorus showed graphically the danger to predict the concentration of TP which could exit the TW system based on phosphorus loading rates. The mean value of influent concentration in TP was of 21 mg TP/L with a maximum value of 93 mg TP/L. This high value is induced by the main activity of the guesthouse as a restaurant. The mean value of effluent was of 0,8 mg TP/L and of 0,3 mg TP/L for the median value. A seasonal trend in the TP of the effluent was graphically displayed and the saturation of the media sites for sorption capacity was demonstrated being in progress. Bottom layers receiving a high loading rate, as demonstrated in Chapter II, are already saturated and the surface layer at the end of the bed starts to be exhausted too. The percentage based on phosphorus mass removed is starting to decrease and a fast low percentage removal of TP is predicted.

GENERAL CONCLUSIONS

V. GENERAL CONCLUSIONS

With this investigation work on treatment wetlands, a clarification of the current terminology used to define and describe constructed treatment wetlands was proposed. The standardised classification system proposed is based on the water position, the saturation of the media, the flow direction and the vegetation sessility; all standard types so identified are described. Variants and intensified types are identified and described as well. A classification tree presented as a flow chart summarises the classification. The purpose of this work is already partly achieved as this classification starts to be in use and is already reported by several authors.

The system investigated in the frame of this study is reported into this new classification system and its design is described. The sampling layouts and methods used for experimentations are explained. Experiments under greenhouse on small scale pilots, additional to the onsite tests and measures, are also described.

The hydraulic efficiency and water flow pattern inside the TW was investigated through onsite three dimensional tracer tests experiments, replicated twice, what is unusual for tracer tests. Additional tests on the pilots under greenhouse investigated the media size factor impact on the hydraulic behaviour, which were replicated three times on non planted gravel horizontal flow beds. Both tests demonstrated results significantly different with replication. The experiments under greenhouse showed that the effect of the media size is significant on all investigated parameters: the tracer detention time decreases with larger gravels; the plug flow effect is lower with lower number of tank in series (*NTIS*) values as well as lower volumetric efficiency than for fine gravels; larger gravels increase dead zones, the dispersion coefficient and the axial dispersion; and finally, the velocity of the flow is faster with larger gravels than with fine ones.

The onsite tracer tests demonstrated that the horizontal flow bed has general good hydraulic and volumetric efficiencies, with relatively low dead zones. It demonstrated also that the internal hydraulics of the system was non homogeneous over length and with depth, as it presented variable *NTIS* indicative either of a plug flow water movement for the central portion of the bed, or a internal good mixing of the water for portions close to the inlet and the outlet parts of the TW, and presented also variable tracer detention time. The application of mathematical models developed by chemical engineering or hydraulics provided complementary information. The models developed by chemical engineering provided the *NTIS* and the calculated detention time were injected as one of the parameters in the multi flow with dispersion hydraulic model. This second model identified that water fluxes were not homogeneous with depth inside the TW and 60% of the flow was creeping on the bottom layer of the bed. It also provided the water flow velocities, which were logically faster on the bottom of the bed, and the axial dispersion, which was higher where flow velocities were lower. The reviewed inflow rate distribution allowed the reviewing for all layers of the nominal detention time and of the hydraulic indexes, which are developed by the chemical engineering theory, and based on the assumption of homogeneous systems.

Once the water movement inside the bed was identified, the regular pollutants (BOD, COD, TSS, Total Nitrogen and Total Phosphorus) which are carried out by the wastewater flow inside the TW and their removal processes and efficiencies were investigated. Treatment performances were expressed by graphical analysis of concentration IN or Loading Rates IN versus concentration OUT over length and with depths for all the samples. The *P-k-C** degradation model was applied in order to define degradation *k*-rate values and the frequency distribution profiles of *k*-rate percentiles were drawn to depict what is degraded and where on the TW. Although the mean value of wastewater influent was 328 mg BOD/L what is considered as high for a secondary wastewater system influent, the results of BOD concentration over length and with depth have shown that 80 % of the samples collected satisfy the Belgian standard of 60 mg BOD/L at the outlet. A very large decrease in BOD concentration occurs within the first few meters of the TW bed, and more than 60 % of the samples already met the Belgian standard after 3 m through the TW. A slight seasonal effect was observed along the two years and a half of measurements, and the application of the relaxed P-k-C* TIS model has provided degradation rate coefficients for BOD ranking from less than 10 m/yr to more than 300 m/yr. Significant higher degradation rates were observed for all the bottom layers and for the closest sampling line from the inlet.

The results of COD concentration over length and with depth showed that more than 70 % of the samples collected were satisfying the Belgian standard of 60 mg COD/L at the outlet and were similar to those observed for BOD.

Due to complex sampling procedure, TSS were measured at the inlet and outlet only. HF system returns an average effluent concentration of 33,9 mg TSS/L over the monitored period with a 90^{th} percentile limit of 53 mg TSS/L. The median inlet TSS was 70 mg/L and the median outlet was 30 mg TSS/L.

Finally, the specific pollutants of nitrogen and phosphorus, not submitted to any regulation in the Walloon region for small scale wastewater treatment systems, were analysed for total nitrogen (TN) and total phosphorus (TP). Additional microbial tests were performed on the presence and location of nitrifying and denitrifying bacteria. The high removal rates for all parameters in the first few meters of the bed are demonstrated by the measure of real respirometry which showed that the higher activity measured by the total respirometry for the few first meters of the bed, was the result of a higher amount of biofilm rather than due to a higher active biofilm. Bacteria involved in nitritation, nitratation and denitrification were found over all location and depths investigated. Denitrifying bacteria were more numerous in general than nitrifying bacteria. Nitrifying bacteria were more numerous on surface layers; whereas denitrifying bacteria were not dependent with depth, demonstrating that they can be aerobic or anaerobic.

About TN, the median TN inlet (50^{th} percentile) is 63 mg TN/L, while the median outlet is 13 mg TN/L. The mathematical relaxed *P-k-C** model was applied to calculate the TN degradation coefficient values (*k*) for all the collected samples. The global trend was a slow and regular decrease of the degradation *k*-rate for TN with length, having systematically higher degradation coefficients for the bottom layers, decreasing also over length.

About TP, the mean value of influent concentration in TP was of 21 mg TP/L with a maximum value of 93 mg TP/L and the mean value of effluent was of 0,8 mg TP/L and of 0,3 mg TP/L for the median value. A seasonal trend in the TP of the effluent was graphically displayed and the saturation of the media sites for sorption capacity was demonstrated being in progress.

At the end of this study, the experiments, tests, analysis and results have, as expected, qualified and quantified the hydraulic efficiency of the horizontal flow treatment wetland which was investigated, as well as characterised the water quality parameters removal processes and efficiencies.

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VI. REFERENCES

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