# Life Cycle Assessment of urban wastewater systems: capturing temporal variability of water discharges in an urban catchment

Eva Risch

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# Life Cycle Assessment of Urban Wastewater Systems – capturing temporal variability of water discharges in an urban catchment

Présentée par Eva RISCH
Le 6 Décembre 2018

Sous la direction de Carole SINFORT

Devant le jury composé de

| | |
|---|---|
| Mme Marie-Noëlle PONS, Directrice de Recherche, Université de Lorraine | Rapportrice |
| Mme Ligia BARNA, Professeure, INSA de Toulouse | Rapportrice |
| Mme Carole SINFORT, Professeure, Montpellier SupAgro | Directrice de thèse |
| Mr Christian SALLES, Professeur associé, Université de Montpellier | Examinateur |
| Mme Dominique PATUREAU, Directrice de Recherche, INRA-LBE | Examinatrice, Présidente du jury |
| Mr Thibaud MARUEJOULS, Ingénieur de Recherche, SUEZ-LyRE | Invité |

*A mes parents et grands-parents*

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*Finally I am forever grateful for my parents for giving me the most valuable tools to overcome the barriers set on my path as a deaf kid between a world of golden, deep-space silence and another world full of sound to be later discovered with the help of bionic ears: fantastic support, personal beliefs and French Cued Speech (Langue française Parlée Complétée), so I would not be lost in translation at home or at school. They also taught me to seek the help I needed, and provided me with opportunities and experiences that have made me who I am. This journey would not have been possible if not for them, and I dedicate this milestone to them.*

*“Non credo nel contrasto tra civiltà urbana e civiltà rurale. Il contrario di città non è campagna; è deserto. Deserto come luogo fisico e come solitudine esistenziale. La campagna europea non è l'Amazzonia; è antropizzata, è un luogo dell'uomo. L'Europa è tutta una grande città, e il treno è la sua metropolitana. Da Parigi si va in treno a Londra, Bruxelles, Amsterdam. L'Europa è il mio Paese, è la mia città.”*

Renzo Piano

“*I don’t believe in the contrast between urban civilization and rural civilization. The contrary of a city is not the countryside; it is desert. Desert in the sense of a physical place and an existential loneliness. The European countryside is not the Amazone; it is anthropized, it is a man-made place. Europe is like a whole great city, and the train is its subway. From Paris, one travels by train to London, Bruxelles, Amsterdam. Europe is my Country, it is my city.”*

Renzo Piano

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# Glossary

## Terminology used in the thesis

| | |
|---|---|
| *Build-up and wash-off Models* | These are the most commonly used approaches for modelling the water quality in an urban catchment. These are conceptual models that represent the simplified mechanism of pollutant build-up and wash-off processes. The model parameters need to be calibrated using data from the catchment. |
| *Catchment (or watershed)* | The impervious area that contributes to stormwater runoff and related pollutant load; irrespective that the area is served by a separate sewer network or a combined sewer network. |
| *Catchment area* | The impervious surfaces that contributes (drains) to stormwater runoff and related pollutant load. The words catchment and watershed are used identically in the text. |
| *Centralized UWS* | Urban wastewater systems in which the wastewater is piped from all communities in the urban area served by a single large treatment plant, thus requiring an extensive pipe network so as to reach even the most remote communities. |
| *Characterization factor (CF)* | factor derived from a characterization model which is applied to convert an assigned life cycle inventory analysis result to the common unit of the category indicator  (ISO 14044, 2006) |
| *Combined sewers* | Combined sewers are large networks of underground pipes that collect wastewater from individual households and stormwater (surface runoff) from impervious areas to a (semi) centralized treatment facility (WWTP) using gravity or pumps if necessary. |
| *Combined Sewer Overflow* | A relief structure (called storm-water regulator in American English or combined sewer overflow in British English) which is constructed in combined sewer systems where excess sewage can be diverted from the combined sewer system directly into a receiving watercourse. The mixed untreated flow of wastewater and stormwater from impervious urban surfaces and roads that are discharged from an overflow structure in a combined sewer network. The discharge takes place when the capacity of the downstream network (including the treatment plant) is exceeded, and flows are discharged directly into a receiving watercourse. |
| *Continuous discharges (CDs)* | These discharges are defined as final effluent discharge to receiving waters from municipal wastewater treatment plants (WWTP) and discharges from industrial premises. These discharges are routinely monitored to meet environmental quality standards to safeguard receiving waters quality (e.g. Water Framework Directive in the EU) |
| *Control structure* | Structure that regulates water discharge from a best management practice. |
| *Discharge (or flow)* | Volume of water that passes a cross-section of a river or network in a specified amount of time (e.g., m3) |
| *Discharge rate (or flow rate)* | Instantaneous rate of water discharge from a source expressed as a volume per unit time (e.g. L/sec). |
| *Domestic wastewater* | Wastewater from residential settlements and services which originates predominantly from the human metabolism and from household activities (91/271/EEC Directive, Article 2(2)) |
| *Emission factor* ($EF_{p,s}$) | Defined for a pollutant p and a source s, describes the rate of pollutant p emission for this source s (e.g., atmospheric deposition rate, g/(m².d)) |
| *End-of-pipe technologies* | Methods used to remove already formed contaminants from a stream of air, water, waste, product or similar. These techniques are called 'end-of-pipe' as they are |

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| | normally implemented as a last stage of a process before the stream is disposed of or delivered. |
| *Eutrophication* | The enrichment of water by nutrients, especially compounds of nitrogen and/or phosphorus, causing an accelerated growth of algae and higher forms of plant life to produce an undesirable disturbance to the balance of organisms present in the water and to the quality of the water concerned |
| *Event Mean Concentration (EMC)* | This method assumes that the concentration of pollutant remains constant during the rain event. This method is suitable to use along with a detailed flow model. The flow rate at each time step can be integrated with the EMC to produce the pollutant load. While this method can be useful to predict the total pollution load during a rain event, it cannot predict the concentration changes during the duration of a rain event. A log normal distribution can be used to predict the variation of pollutant concentration during the rain event. |
| *Geographic information system (GIS)* | A computer-based software package for storing, displaying, and querying location and attribute data. |
| *Heavy metals* | Elements such as zinc and copper that accumulate in urban areas, mainly due to automobile use. These metals are readily available to bind to soil and clay particles, but in certain conditions can be transported with runoff and contaminate groundwater. |
| *Impact category* | Class representing environmental issues of concern to which life cycle inventory analysis results may be assigned (ISO 14044, 2006) |
| *Impervious (or impermeable)* | Hard surfaces that do not allow infiltration of rainfall into them; not pervious. |
| *Infiltration* | the movement of water from the surface of the land into the subsurface (vadose zone). |
| *Intensity (of rainfall)* | The time rate of precipitation [L/t - in/hr or cm/hr] |
| *Intermittent discharges (IDs)* | Discharges generated during wet-weather events, occurring intermittently and function of the rainfall temporal distribution. They include stormwater and combined sewer overflows. |
| *Life cycle impact assessment (LCIA)* | phase of life cycle assessment aimed at understanding and evaluating the magnitude and significance of the potential environmental impacts for a product system throughout the life cycle of the product (ISO 14044, 2006) |
| *Life cycle inventory (LCI)* | phase of life cycle assessment involving the compilation and quantification of inputs and outputs for a product throughout its life cycle (ISO 14044, 2006) |
| *Midpoint indicator* | Impact category indicator located somewhere along the impact pathway between emission and category endpoint (Hauschild and Huijbregts, |
| *Nonpoint source* | A source of pollution that cannot be traced to or is released at a definable single place, but rather is sourced from a number of points that are widespread. |
| *Nutrient* | Any element or compound, including pollutants, that fuels high organic aquatic systems. |
| *Particulate pollutants* | A mixture of small (2.5 to 10 micrometers) particles of acids, organic chemicals, metals, and soil or dust particles. |
| *Performance of a system* | A general measure of how well a system, according to its design, meets the goals in terms of both hydraulics and water quality characteristics. An extended understanding of performance includes regulatory requitements on water quality of discharges, use of resources, and aspects of risk and safety. |
| *Point source* | A source of pollution that can be traced to or is released at a single place (e.g. factory, oil or chemical spill, municipal wastewater treatment plant or a stormwater discharge pipe). |
| *Pollutant* | A contaminant existing at a concentration high enough to endanger the environment or the public health or to be otherwise objectionable. |

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| *Pollutant load (or export)* | Mass of pollutant passing a specified point during a specified time period (e.g. 1 day or 1 year) or during a specific event (e.g. kg or ton). Mathematically, load is essentially the product of water discharge and the concentration of a substance in the water. It is generally used to describe the daily pollutant mass arriving to a treatment facility. |
| *Pollutant generation* | The diurnal concentration profiles for wastewater pollutants can be used to model the pollutant generation with an influent generator model. Surface pollutants are accumulating on urban surfaces during dry weather. Buildup models are commonly used to model the rate of accumulation for pollutants on urban surfaces. |
| *Pollutant transport* | Pollutant transport from the urban catchment to sewers takes place during both wet and dry periods. During dry weather conditions, pollutants from domestic and industrial sources are transported directly to the sewer system. Surface pollutants can also be transported to the sewer system through street cleaning and winds during dry days. During rain events, the pollutants in the streets and buildings are washed off along with the rain water and are transported to the sewer system. Other simpler models based on event mean concentrations and regression equations also estimate pollutant wash-off. |
| *Pollution* | Any aspect of water quality (physical, thermal, chemical, or biological) that interferes with an intended use. |
| *Population Equivalent* | A measure defined in the 91/271/EEC directive to define the daily organic biodegradable load generated by an average citizen. This load corresponds to a five-day biochemical oxygen demand (BOD5) of 60 g/pe/d. In the following, we propose to extend the definition of this load with conventional wastewater pollutants based on the French person-equivalent (Arrêté du 9 Décembre 2004): total Kjeldahl nitrogen of 15g/pe/d; total phosphorus of 2 to 4 g/pe/d. |
| *Rainfall-runoff model* | Model describing of a part of the water cycle, and therefore the movement of a fluid - water. It is explicitly or implicitly based on the laws of physics, and in particular on the principles of conservation of mass, conservation of energy and conservation of momentum. Depending on its complexity, such a model can also simulate the dynamics of water quality, ecosystems, and other dynamical systems related to water. |
| *Retention Tank* | Underground structure to allow the retention of a volume of stormwater |
| *Return flow* | Water which is pumped from a stream or basin that is not consumptively used and which returns to the stream or basin (e.g. return flows to WWTP are flows that were stored in the sewer system and pumped back for treatment in the WWTP). |
| *Receiving waters* | Bodies of water or surface water systems that receive water from upstream sources |
| *Sanitary landfill* | A facility that handles and stores nonhazardous waste, such as household garbage. |
| *Secondary treatment* | Technology-based requirements for direct discharging municipal sewage treatment facilities. Standard is based on a combination of physical and biological processes typical for the treatment of pollutants in municipal sewage. Standards are expressed as a minimum level of effluent quality in terms of: BOD5, suspended solids (SS), and pH. Usually this treatment step is preceded by primary treatment which is a physical and/or chemical process involving settlement of suspended solids. |
| *Separate sewers* | Separate sewerage consists in the separate collection of municipal wastewaters (blackwater from toilets, greywater and industrial wastewater) and stormwater (surface runoff). The separate collection prevent the overflow of sewer systems and WWTPs during rainy periods and the mixing of the relatively little polluted stormwater with chemical and microbial pollutants from the municipal wastewater. |
| *Site Mean Concentration* | Geometric average of EMC measured at a site during several storms |
| *Source activity factor* ($a_s$) | Defined for an urban zone, variable parameter describing the level of an activity generating nonpoint emissions of pollutants (e.g. metallic roof area, m²; daily road |

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| | traffic volume, vkm.$d^{-1}$) |
| *Source control* | Removing, enclosing, or otherwise controlling a source of contamination to prevent further water pollution. |
| *Stormwater (or runoff)* | Water that originates from rainfall or snowmelt on impervious surfaces during rain events; often associated with urban areas. Also called runoff. |
| *Stormwater management* | The management of runoff from pre- to post-development, often using stormwater treatment practices and best management practices to manage quality and control release into receiving bodies of water |
| *Surface depression storage* | Runoff occurs from impervious areas if rainfall exceeds the depression storage depth |
| *Surface water* | Water in streams, rivers, lakes, wetlands, and reservoirs; |
| *Total Suspended Solids (or Suspended Solids)* | A measure of the dry weight of filterable (non dissolved) solids present in a sample. |
| *Urbanization* | The changing of a landscape to a an urban or suburban setting, usuall y with the addition of pavements, buildings, and utility systems |
| *Urban wastewater* | Domestic wastewater or the mixture of domestic wastewater with industrial wastewater and/or stormwater runoff (91/271/EEC Directive, Article 2(1)) |
| *Wastewater* | Wastewater is defined as "a combination of one or more of: domestic effluent consisting of blackwater (excreta, urine and faecal sludge) and greywater (kitchen and bathing wastewater); water from commercial establishments and institutions, including hospitals; industrial effluent, stormwater and other urban run-off; agricultural, horticultural and aquaculture effluent, either dissolved or as suspended matter”. |

# List of Abbreviations

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## Abbreviations

| | |
|---|---|
| ADT | Average Daily Traffic |
| AoP | Area Of Protection |
| BOD | Biological Oxygen Demand |
| CD | Continuous Discharge |
| CF | Characterization Factor |
| COD | Chemical Oxygen Demand |
| COV | Coefficient Of Variation |
| CSO | Combined Sewer Overflow |
| DWF | Dry Weather Flow |
| EMC | Event Mean Concentration |
| EU | European Union |
| ID | Intermittent Discharge |
| LCA | Life Cycle Assessment |
| LCI | Life Cycle Inventory |
| LCIA | Life Cycle Impact Assessment |
| LCZ | Local Climate Zone |
| OPUR | Observatory of Urban Pollutants |
| PAH | Polycyclic Aromatic Hydrocarbon |
| P.E. | population equivalent |
| RT | Retention Tank |
| RE | Removal Efficiency |
| SIAAP | Paris public sanitation service |
| SMC | Site Mean Concentration |
| SWR | StormWater Runoff |
| SWMM | Storm Water Management Model |
| TKN | Total Kjeldahl Nitrogen |
| TN | Total Nitrogen |
| TP | Total Phosphorus |
| SS | Suspended Solids |
| U.S. EPA | U.S. Environmental Protection Agency |
| UWS | Urban Wastewater System |
| WFD | Water Framework Directive |
| WWTP | Waste Water Treatment Plant |

# Main Variables

| Used in Chapter 3 | |
|---|---|
| $EF_{p,s}$ | Specific emission factor for each of the key stormwater pollutants *p* and for each primary nonpoint source *s*. Fixed parameter, whose value is already defined and constant across catchments |
| $a_s$ | Source activity factor. Variable parameter, whose value is to be determined for the catchment |
| $E_{p,s}$ | Potential available amount of pollutant *p* from source *s* (stocks & funds) |
| $SPP_p$ | Stormwater pollution potential (annual load) in pollutant *p* from all sources *s* |
| $rWPP_p$ | Raw wastewater pollution potential (annual load) in pollutant *p* |
| $Inf_p$ | Influent pollution potential (annual load) in pollutant *p* |
| $dSS$ | Distribution of suspended solids (SS) load across UWS components |
| $dQ$ | Distribution of water flows (Q) across UWS components |
| $X_{part,p}$ | Particulate fraction of pollutant *p* in influent |
| $X_{diss,p}$ | Dissolved fraction of pollutant *p* in influent |
| $DEP_p$ | Mass vector of pollutant *p* in deposits (annual load) |
| $CSO_p$ | Mass vector of pollutant *p* in CSO discharges (annual load) |
| $WTP_p$ | Mass vector of pollutant *p* in treated WTP effluents (annual load) |
| $SWR_p$ | Mass vector of pollutant *p* in stormwater discharges (annual load) |
| $RE$ | Removal Efficiency (RE) to specify for pollutants if different. Abatement during wastewater treatment applied is coefficient (1-RE) |

# Abstract / Résumé

With unprecedented urban growth at the planetary scale, urban wastewater systems (UWS) are faced with the operational challenge of managing ever increasing and polluted stormwater to mitigate impairments of the quality of aquatic ecosystems. However, to which extent do wet-weather control strategies actually help reduce overall environmental impacts? With the general aim of assessing in a whole-systems perspective to improve the environmental assessment of UWS with Life Cycle Assessment (LCA), by including impacts from operation and infrastructure, and impacts from discharged pollutants, the objective of the thesis is: “Is it possible to perform the LCA of Urban Wastewater Systems that captures the temporal variability of water discharges in an urban catchment?”. The first step in this work demonstrates the relative importance of the impacts caused by intermittent discharges (ID), generated during storm events, compared to continuous discharges from municipal wastewater treatment plants. Then, the core of this thesis is the development of a site-dependent framework to estimate, for life cycle inventory (LCI) purposes, IDs with consideration for their important spatiotemporal variability at the urban catchment scale. The model (named STOIC) satisfies requirements on inventory data quality to support sufficiently accurate modelling of IDs and conceptualizes stormwater processes occurring within an urban catchment and its UWS. These stormwater processes include the generation of water pollutants from nonpoint source emissions to the routing and distribution of pollution in the UWS components, resulting in spatiotemporally differentiated LCIs for water pollutants within an urban catchment. The STOIC framework is applied to an UWS at sub-catchment-scale to assess global performances of different control strategies for IDs in terms of environmental impacts in a real-world case study in Bordeaux, southwest France. This demonstrates the interest and applicability of an extended environmental assessment which considers the temporal variability of UWS discharges from an urban sub-catchment.

**Key words:** life cycle assessment (LCA), environmental impacts, stormwater, urban catchment

Avec une croissance urbaine sans précédent à l'échelle planétaire, les systèmes d’assainissement urbains (SAU) sont confrontés au défi opérationnel de la gestion d’eaux pluviales toujours plus importantes et polluées afin d'atténuer les dégradations de la qualité des écosystèmes aquatiques. Toutefois, dans quelle mesure les stratégies de contrôle par temps de pluie contribuent-elles réellement à réduire les impacts environnementaux globaux? L'objectif général de cette thèse est d'évaluer avec une vision systémique globale, pour améliorer l'évaluation environnementale des SAU par l’analyse du cycle de vie (ACV), en incluant les impacts opérationnels et liés aux infrastructures, et les impacts des polluants rejetés: "Est-il possible d'effectuer l'ACV des SAU en considérant la variabilité temporelle des rejets d’un bassin versant urbain? La première étape de ce travail démontre l'importance relative des impacts causés par les rejets urbains de temps de pluie (RUTP) intermittents, par rapport aux rejets continus des stations d'épuration municipales. Le cœur de cette thèse est l'élaboration d'un cadre dépendant du site pour estimer, en vue d’établir un inventaire du cycle de vie (ICV), les RUTP en considérant leur importante variabilité spatiotemporelle à l'échelle du bassin versant urbain. Le modèle (appelé STOIC) satisfait aux exigences en matière de qualité des données d'inventaire pour soutenir une modélisation suffisamment précise des RUTP et conceptualiser les processus de pollution des eaux pluviales se produisant dans un bassin versant urbain et dans son SAU. Ces processus de pollution des eaux pluviales comprennent la production des polluants des RUTP à partir d'émissions de sources non ponctuelles jusqu'au transfert et à la distribution de la pollution dans les composantes du SAU. Des ICV différenciés spatio-temporellement pour les polluants des RUTP sont calculés pour un bassin versant urbain. Le modèle STOIC est appliqué à un SAU à l'échelle du sous-bassin versant pour évaluer les performances globales de différentes stratégies de gestion des rejets par temps de pluie en termes d'impacts environnementaux dans une étude de cas réel à Bordeaux, dans le sud-ouest de la France. Cela démontre l'intérêt et l'applicabilité d'une évaluation environnementale approfondie qui tient compte de la variabilité temporelle des rejets de SAU d'un sous-bassin versant urbain.

**Mots-clés:** Analyse du cycle de vie (ACV), impacts environnementaux, eaux pluviales, bassin versant urbain

# Scientific production

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### *Articles*

This thesis has three core chapters, one was published in an international peer-reviewed journal, and two are ready for submission:

- Chapter 2 has been published in Water Research, Volume 128, January 2018, p. 412-423: 10.1016/j.watres.2017.10.039: Risch, E.; Gasperi, J.; Gromaire, M.C.; Chebbo, G.; Azimi, S.; Rocher, V.; Roux, P.; Rosenbaum, R.K.; Sinfort, C.: "Impacts from urban water systems on receiving waters – How to account for severe wet-weather events in LCA?".

- Chapter 3 will soon be submitted as: "A parsimonious site-dependent framework linking human activities, land use and climate to determine loads in intermittent discharges to receiving waters from urban catchments for LCI purposes".

- Chapter 4 will soon be submitted as: "Life-cycle perspectives for the management of intermittent discharges in urban wastewater systems: lessons learnt from a French suburban catchment".

### *Oral presentations*

SETAC Europe 22nd LCA Case Study Symposium ELSA-PACT/Irstea (France), CIRAIG (Canada), Montpellier, September 2016: "Impacts of storm events on the emissions of an urban wastewater system: the case of the Greater Paris watershed"

SETAC Nantes SETAC Europe, Nantes, 22-26 May 2016: "Impacts of severe wet-weather events: How to account for temporal variability of unmanaged peak flows in an urban sewage treatment system?"

The 18th International Conference on Diffuse Pollution and Eutrophication Los Angeles, USA, August 13-17, 2017 International Water Association (IWA), Los Angeles, USA: "LCA of wet-weather emissions of an urban wastewater system: the case of the Greater Paris watershed"

### ***Posters***

International conference SETAC, Barcelona, May 2015 “Assessment of wastewater treatment technologies including their water consumption impacts at endpoint level”

International conference SETAC, Rome, May 2018: “Building a Life Cycle Inventory of stormwater pollutant fluxes: model evaluation for a separate residential urban catchment”

# Chapter 1.: General introduction

# *Contents of Chapter 1*

## 1.1 Metabolism of urban wastewater systems

### 1.1.1 Can we truly liken a city to a living organism?

The essence of life is process, according to General Systems Theory[1]. In turn, processes require energy to function. Energy is harnessed and manipulated in systems at a variety of scales ranging from cells to ecosystems, or from batteries to entire industries. In the 19th century, studying the social organization of human labor, Marx and Engels hypothesized that the harvesting of resources from the Earth by humans for sustenance and shelter is itself a metabolic process that generates and sustains civilization. In 1965, Abel Wolman first coined the term metabolism of cities, making a historical contribution to the field of industrial ecology. In his pioneering work, a modern city is like a living organism with metabolic requirements to sustain its inhabitants at home, at work and at play. In his metabolic model (input-output chart) for a hypothetical American city of one million population (Figure 1), the wastewater is the largest component: "water, which enters the city silently and unseen, overshadows all other inputs in volume. (…) After about 10 per cent of the water has been diverted to lawns and other unrecoverable uses, it returns, contaminated, to the city's sewers."

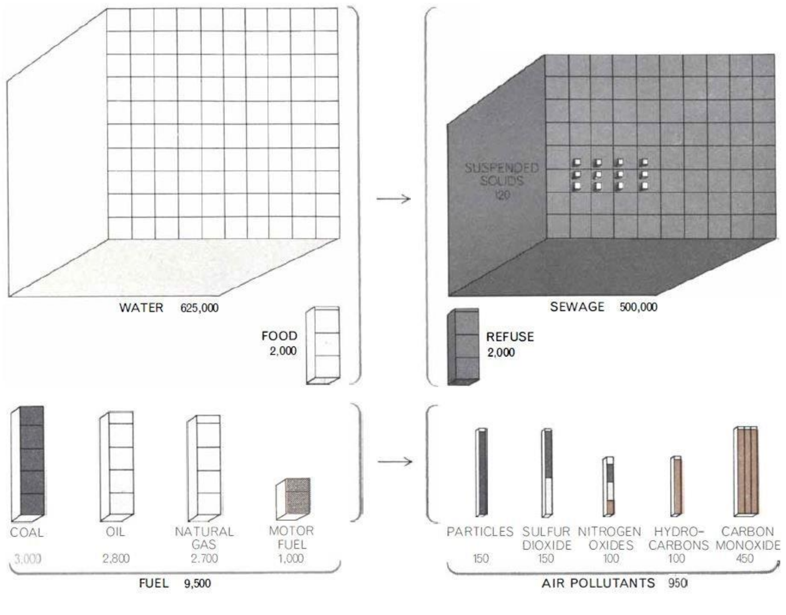

*Figure 1. Input-output transactions of water, food, fuel, sewage, solid refuse and air pollutants of an hypothetical American city of 1 million + people (Wolman, 1965)*

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[1] *http://www.panarchy.org/miller/livingsystems.html*

However, equally important is stormwater runoff as a carrier of urban pollutants which is described in the section below.

### *1.1.2 Another metabolic product of cities: urban stormwater runoff*

In 2018, about 55% of the world population lived in urban areas and this share is expected to increase to about two-thirds by 2050 (United Nations, 2018). Furthermore, there is strong evidence that precipitation regimes will change in the future because of anthropogenic climate change (IPCC 2014). Due to both the individual and combined effects of these two factors, regions with increased precipitation will experience increasingly intense rainfall and higher urban stormwater runoff flow rates. The growth of urban areas and the intensification of urban land use increase the conversion of rainfall into runoff.

Urbanization also leads to a greater abundance of pollutant sources, which, when considered with the increase in runoff, will increase the pollutant loads conveyed by stormwater runoff and negatively affect receiving waters (Goonetilleke et al., 2014; Marsalek et al., 2006). Indeed, cities are hotspots of nonpoint sources due to urban activities (traffic and other combustion processes) and a large and mostly unknown mass of substances stored in the technosphere with long residence times (e.g. buildings, infrastructures)(Brunner et al., 2001). Additionally, many urban areas are facing problems caused by aging drainage infrastructure, which may require major maintenance or reconstruction in the near future.

Quantifiying all environmental impacts associated with urban catchments and their urban wastewater systems (UWS) is a necessary condition to reduce their footprint. Amongst the available tools for assessing environmental impacts of such systems, life cycle assessment (LCA) has already proven its worth. The LCA method is explained in the following section by underlining briefly its main forces and its limitations for the assessment of urban water systems.

## 1.2 Introduction to Life Cycle Assessment (LCA)

The LCA method is standardized by the ISO standards 14040 (2006) and 14044 (2006), and is an internationally recognized approach (ISO, 2006a; ISO, 2006b). This decision-making tool quantifies the potential environmental impacts of a product or a service by analysing all extracted resources and released emissions over its entire life cycle (from cradle to grave). The LCA method has four phases: (1) goal and scope definition, (2) life cycle inventory (LCI) analysis, (3) life cycle impact assessment (LCIA), and (4) interpretation. Among the main principles that make the LCA strengths are described in the ILCD Handbook (JRC European commission, 2011):

- A wide range of environmental impact categories are addressed in LCA, which makes it as comprehensive as possible. Burden shifting from one category to another can be identified, unlike monocriteria methods such as the carbon footprint (Figure 2).
- Environmental impacts are quantified on the basis of a transparent, scientific and quantitative framework.
- LCA makes it possible to assess the potential environmental impacts of any given system, such as a particular type of goods, a service, a company, a technology strategy, a country, etc.
- LCA integrates resource extraction and emissions throughout the whole lifecycle of the system under study, which prevents burden shifting from a system component to another.
- The structure of LCA allows comparisons of environmental performances of different alternative systems and identifies opportunities for improvement on an equal basis: the functional unit. The functional unit is the precise quantitative description of the function(s) provided by the system under study, answering to the questions: “what”, “how much”, “how well”, and “for how long”.

Collectively, these aspects of the LCA methodology are intended to help reduce the risk that something important gets overlooked in any particular options comparison, or any particular decision making context.

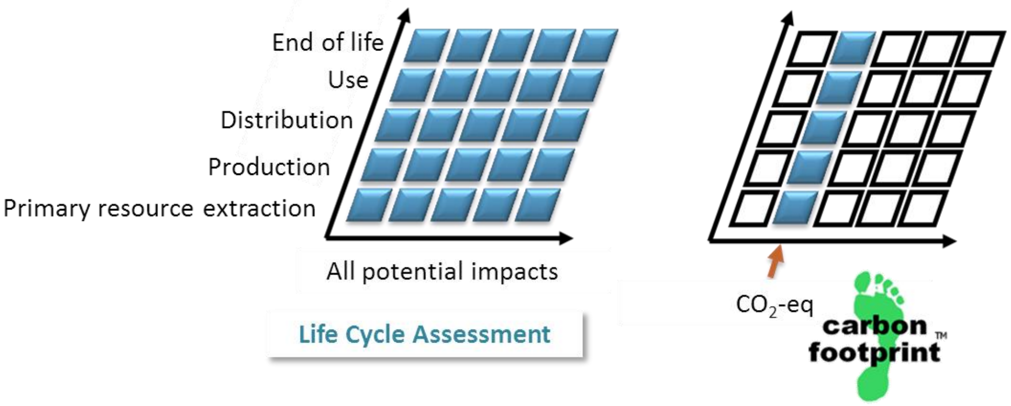


*Figure 2. Capturing all impacts over a life cycle perspective: differences in scope and completeness of LCA and footprinting methods (e.g. carbon footprint™)*

In LCA, an environmental intervention (i.e. an emission into or resource extraction from the environment, quantified in the inventory as elementary flows, phase 2) is linked to potential environmental impacts (phase 3), within causality chains (also called impact pathways). In a causality chain, a specific characterization factor (CF) is assigned to each elementary flow, defined in the inventory, allowing to express the result of its potential impact in terms of a common unit for a given impact category (e.g. all indicator results for the climate change impact category are expressed in $CO_2$-equivalents). There are two kinds of impact indicators along the same causality chain. An impact indicator "early in the environmental mechanism gives a more measurable result but with less environmental relevance and more remote from the concerns directly observable in the environment" whereas an impact indicator "downstream in the environmental mechanism gives more relevant but hardly verifiable information" (Hauschild et al., 2018). Thus, two levels of indicators are defined and used in LCA: the midpoint impact indicators that can be further modelled into endpoint impact indicators, which are at the end of the causality chain. The endpoint indicators reflect specific damages that relate to broader topics of interest, called Areas of Protection (AoP). The overall LCA framework endorsed by the United Nations Environment Life Cycle Initiative Environment is described in Figure 3 (Verones et al., 2017).

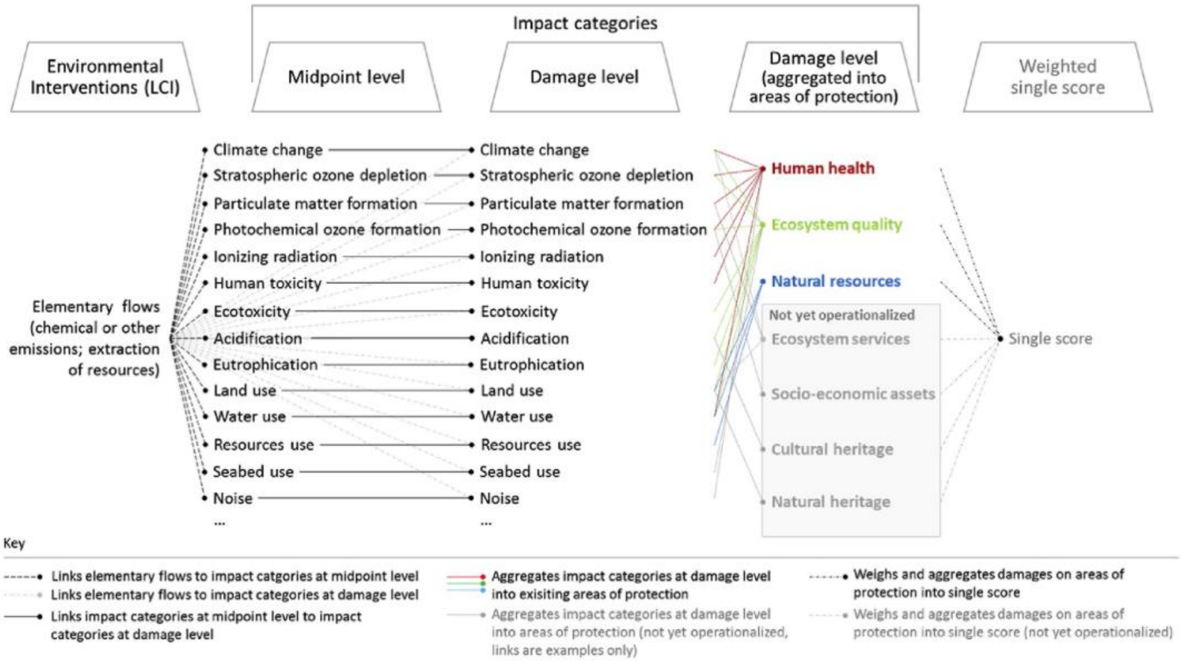


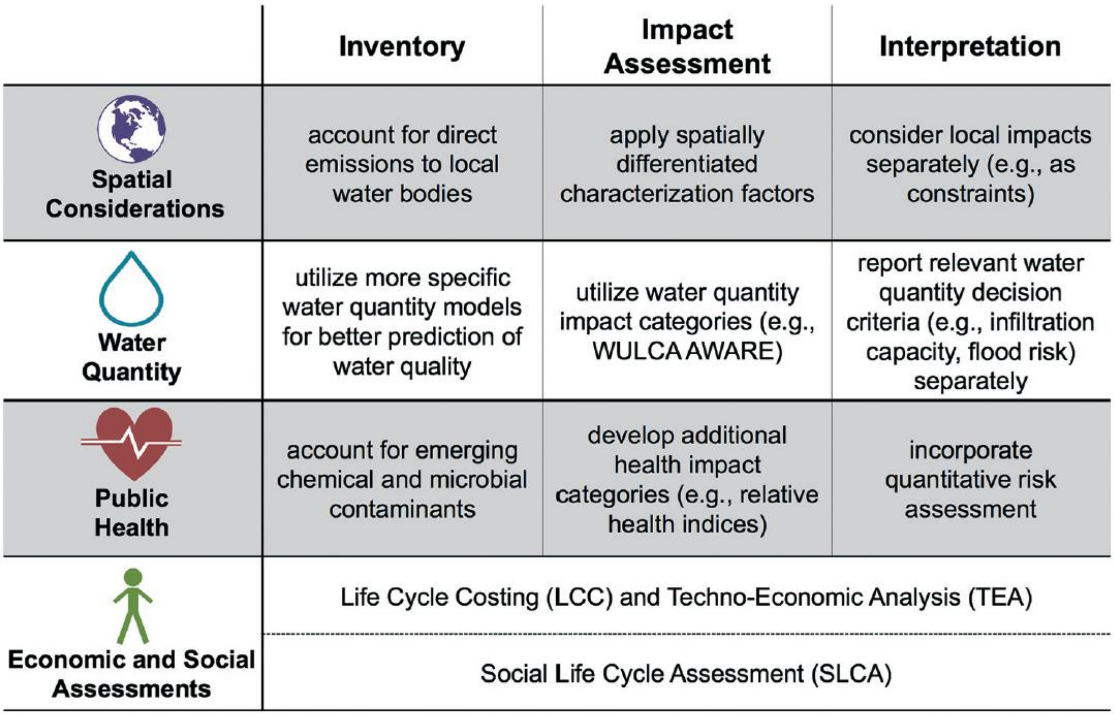
*Figure 3. Updated LCIA framework of the United Nations Environment Life Cycle Initiative linking elementary flows from the inventory results to indicator results at midpoint level and damage (endpoint) level for aggregation into 3 areas of protection. Areas of protection that are operational are indicated with colors, those that are not yet fully operational are shown in the grey box. Retrieved from Verones et al. (2017).*

## 1.3 Towards a comprehensive assessment of UWS including intermittent discharges

### *1.3.1 State of the art of LCA applied to urban water infrastructure*

A recent review revealed that there is now more than 250 research articles on LCA applications to urban water infrastructure (including wastewater, drinking water, stormwater, and integrated urban water systems) since the first applications in the 1990s (Byrne et al., 2017b). It is now clear that LCA is a valuable tool to explore life-cycle environmental burdens across full urban water systems at the city scale, and elucidate the broader implications of design and operational decisions on environmental impacts (Corominas et al., 2013; Guest et al., 2009; Lane et al., 2015; Loubet et al., 2014).

Urban water infrastructure studies should correspond to the primary objectives of urban water systems, which are to manage water quantity and water quality in a way that protects local and regional human and environmental health. To develop this effort, emerging approaches to include spatial considerations, water quantity, public health, and economic and social factors should be considered to better inform decision-making (Figure 4). Some of these opportunities are for advancements within LCA (e.g., apply spatially differentiated characterization factors) while others are for integration of LCA within broader sustainability assessment frameworks (e.g., those which incorporate quantitative risk assessment) (Byrne et al., 2017b).



| | Inventory | Impact Assessment | Interpretation |
|---|---|---|---|
| **Spatial Considerations** | account for direct emissions to local water bodies | apply spatially differentiated characterization factors | consider local impacts separately (e.g., as constraints) |
| **Water Quantity** | utilize more specific water quantity models for better prediction of water quality | utilize water quantity impact categories (e.g., WULCA AWARE) | report relevant water quantity decision criteria (e.g., infiltration capacity, flood risk) separately |
| **Public Health** | account for emerging chemical and microbial contaminants | develop additional health impact categories (e.g., relative health indices) | incorporate quantitative risk assessment |
| **Economic and Social Assessments** | Life Cycle Costing (LCC) and Techno-Economic Analysis (TEA) | | |
| | Social Life Cycle Assessment (SLCA) | | |

*Figure 4. Opportunities for integration of spatial considerations, water quantity, public health, and economic and social assessments within each LCA phase (inventory, impact assessment, interpretation). Retrieved from Byrne et al. (2017b).*

### *1.3.2 Stormwater consideration in LCA*

While it is well known that growing urbanization with its increasing intermittent discharge (ID) flows due to stormwater runoff is a leading contributor to impairments of the quality of aquatic ecosystems (Gosset et al., 2016) yet there is a dearth of LCA research on this topic. The majority of stormwater LCA studies have focused on the quantitative aspect (runoff volume reduction) in specific stormwater control measures (e.g. gray and green infrastructures)(Petit-Boix et al., 2017, 2015; Spatari et al., 2011), or in CSO control strategies (De Sousa et al., 2012). Only two studies investigated the links between land use and stormwater in the life cycle of UWS in terms of changes in the availability of water for aquatic ecosystems (Berger and Finkbeiner, 2010; Saad et al., 2013). Only one study addressed the links between urban land use and pollution from untreated stormwater by defining a stormwater pollution burden for a given system which results from urban land use called in background processes (Phillips, 2015; Phillips et al., 2018).

Thus, several challenges remain in the evaluation of impacts arising from IDs, which are not taken into account in any of the reviewed papers and for which there are no inventory flows. In line with the findings of Byrne et al. (2017b), the main identified challenges/opportunities in the inventory step are the consideration of the spatial and temporal variability of IDs due to the climatic conditions, urban catchment characteristics (e.g. land use and human activities), and the type of UWS (infrastructure and management).

## 1.4 Objectives of the thesis

With the general aim of assessing in a whole-systems perspective to improve the environmental assessment of UWS with LCA, by including impacts from operation and infrastructure of UWS and impacts from treated pollutants in discharges, the general research question of this thesis is:

> *"Is it possible to perform the LCA of Urban Wastewater Systems that captures the temporal variability of water discharges in an urban catchment?"*

Three research questions were identified for this thesis:

- *RQ1. Are direct intermittent water discharges impacts significant compared to continuous discharges impacts at urban catchment scale?*
- *RQ2. How to estimate intermittent discharges from an urban catchment in relation to urban activities, climate and land use for LCI purposes?*
- *RQ3. How do induced impacts from wet-weather control infrastructure compare to avoided impacts from intermittent discharges from a UWS?*

Following this general introduction (**Chapter 1**), each objective will be addressed by a chapter of the thesis referring to a scientific publication (either published or in preparation to be submitted) as described hereafter and summarized in Figure 5.

**Chapter 2** raises the question of the pertinence of considering IDs and their associated impacts at the urban catchment scale. The relative significance of wet-weather IDs versus dry-weather continuous discharges (CDs) is assessed on impact categories concerned by water pollution (e.g. freshwater and marine eutrophication, and ecotoxicity). Measurement data on discharges from the UWS in the Paris area is analysed and organized to develop inventory flows which are used as LCA input data to assess the associated impacts of UWS discharges.

**Chapter 3** tackles the challenge of the spatiotemporal consideration outlined in **Chapter 1**, to estimate IDs from an urban catchment in relation to urban activities, climate and land use for LCI purposes. A description of the resulting site-dependent framework is provided, leading to spatiotemporally differentiated LCI of intermittent discharges from the UWS, which can be used as inputs to a classical LCA to assess wet-weather management strategies.

**Chapter 4** is the application of the framework on a real case study. It aims at evaluating the capacity of the framework to assess induced impacts from wet-weather control infrastructure against avoided impacts from IDs of the UWS. The chosen case study is a sub-catchment in the Bordeaux Metropolis. The global performances of several wet-weather control strategies are compared in terms of net environmental impacts. In light of the case study application, **Chapter 4** discusses the framework applicability, its limitations and opportunities as a decision-support tool.

Finally, the general conclusions of the thesis as well as future research perspectives are provided in **Chapter 5**.

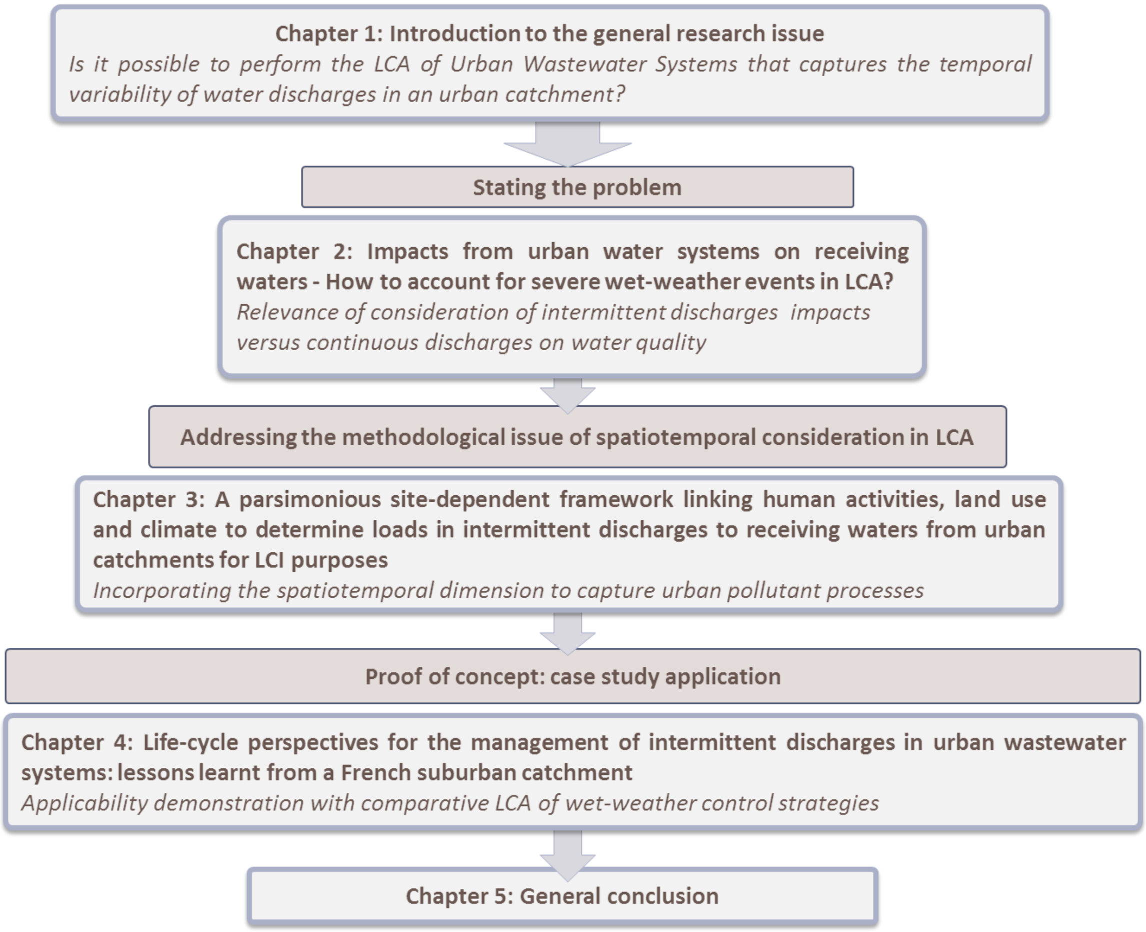


*Figure 5. Structure of the thesis*

# Chapter 2.: Impacts from urban water systems on receiving waters - How to account for severe wet-weather events in LCA?

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*Eva Risch, Johnny Gasperi, Marie-Christine Gromaire, Ghassan Chebbo, Sam Azimi, Vincent Rocher, Philippe Roux, Ralph K. Rosenbaum, Carole Sinfort*

## *Contents of Chapter 2*

*This Chapter investigates the contributions of these wet-weather-induced discharges relative to average dry-weather conditions in the life cycle inventory for UWS. In collaboration with the Paris public sanitation service (SIAAP) and Observatory of Urban Pollutants (OPUR) program researchers, monitoring data on discharges from the UWS in the Paris area is analysed for a selection of routine wastewater parameters and priority pollutants. This data is organized according to archetypal weather days during a reference year. Then, for each archetypal weather day and its associated flows to the receiving river waters (Seine), the parameters of pollutant loads (statistical distribution of concentrations and volumes) are determined. The resulting inventory flows (i.e. the potential loads from the UWS) are used as LCA input data to assess the associated impacts.*

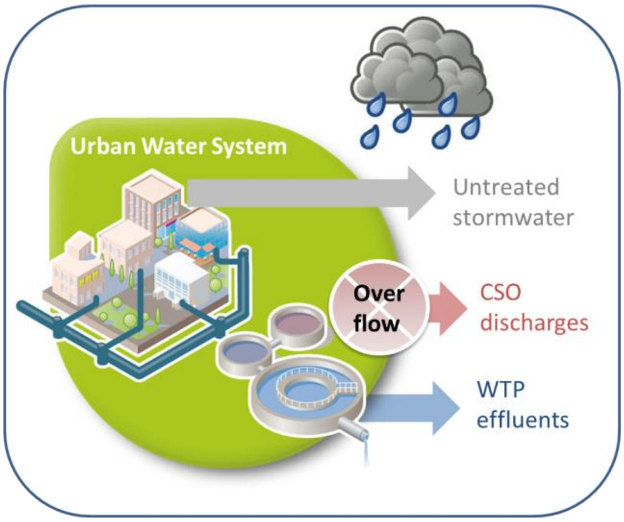


*Figure 6. Graphical abstract of Chapter 2*

## 2.1 Introduction

Handling of water resources (scarcity and/or flooding) is needed for all types of catchments. With an ever increasing rate of urbanisation on a planetary scale, stormwater management has become a tremendous challenge and deals with pollution, additional stormwater and increased flooding. Stormwater sewers are exclusively dedicated to the collection of stormwater whereas combined sewers also collect wastewater from domestic and/or industrial sources in an urban area. Sewers provide drainage, which protects public health, prevents the flooding of property and protects the water environment around urban areas. Usually combined systems are found in temperate – and to some extent subtropical – climates and are concentrated in North America, Canada, Europe, Australia and, to some extent, Japan. In these countries, combined sewer networks dominate the central and old parts of the large cities whereas separate systems exist in the new developments and suburbs (Hvitved-Jacobsen et al., 2010).

During dry weather, the combined sewage collection system, which transports wastewater to the wastewater treatment plant, operates effectively. However, during a heavy rainstorm, extra stormwater gets into the sewage collection system through direct connections. This extra volume of water overloads the sewage collection system and raw untreated sewage overflows at several locations potentially causing impacts on the environment. Historically, sanitary and civil engineering have focused on handling the physical aspects of wet-weather (protection against flooding), omitting the eutrophication and ecotoxicity impacts of combined sewer overflows (CSO) and stormwater on receiving water ecosystems. It is well-known today that stormwater from urban and industrial areas and from roads constitutes one of the main transport mechanisms introducing non-point source pollutants into receiving waters (Pitt et al., 1995; Zgheib et al., 2011). This is why urban stormwater is currently the most significant pollution hazard for coastal waters downstream of densely urbanized areas with separate sewer systems such as the Southern California Bight (DiGiacomo et al., 2004; Schiff et al., 2000). There is a growing number of studies evaluating ecotoxicological impacts of urban wet-weather discharges (stormwater and CSO) on stream ecosystems using an array of tools (Angerville, 2009; Becouze-lareure et al., 2012; Gosset et al., 2016).

In Europe, the implementation of the control policy of pollutants entering the receiving waters in the Water Framework Directive 2000/60/EC (WFD) in 2001, has led to increased research efforts to identify possible pollutant reduction measures. Given the lack of accurate and extensive data on the contamination by priority pollutants of urban flows which enter receiving waters, the Observatory of Urban Pollutants (OPUR) research programme has sought to investigate the production and transfer of pollutants in these flows, which include treated effluents from wastewater treatment plants; CSO discharges and untreated stormwater. This research has delivered data on the occurrence and concentration of priority pollutants listed in the WFD and additional pollutants -typically measured in urban stormwater- from both separate and combined sewers of the Greater Paris area (Gasperi et al., 2014, 2012; Zgheib, 2009; Zgheib et al., 2012). In parallel to the identification of pollutant flows, the need for holistic methods for the environmental assessment of water and wastewater systems was identified and Life Cycle Assessment (LCA) is now increasingly used as a promising approach (Loubet et al., 2014). Indeed, LCA enables the comparison of environmental performances of products or services, supporting decision-making by consumers, industries and governments. LCA in the context of urban wastewater systems (UWS) aims to provide quantitative information to policy makers on UWS environmental profiles, hot spots, and forecasting scenarios that can involve (depending on the chosen goal): (i) change or improvement of a technology (e.g., construction of a new treatment plant or an increase in the connection rate of a waste water collection system), (ii) change of water resources (e.g., water abstraction from another river, wastewater release into the sea) and (iii) change of users (e.g., increase of the population, change of users' behavior). Loubet et al. (2014) concluded that for decisions about future investments done by regional and local authorities at the scale of a river basin or a city, forecasting scenarios should be evaluated in order to inform on their potential environmental impacts. For day-to-day management of water services by operators, LCA could be used to select the most interesting solution on an environmental point of view (e.g. management of water production from different drinking water plants which withdraw water at different locations). However, there are methodological limits in LCA application at smaller temporal and spatial scales as well as uncertainties (e.g. traditional LCA models with annual averages are not suited for this goal, which require dynamic models running at hourly or daily time steps).

The level of sophistication of a LCA model should reflect the robustness required to support a particular decision-making context (Bare et al., 1999). In typical LCA studies of UWS, only average dry-weather conditions are modelled while wet-weather flows originating from UWS are typically not considered, with the exception of Hadjimichael et al. (2016) who proposes to account for reduced wet-weather discharges in the LCA of different UWS upgrades in terms of nutrient inputs to the receiving waters. By nature, wet-weather flows are highly variable. For example, it is readily seen that the concentrations in wet-weather flows are often relatively high in the beginning of an event in contrast to what follows, also called the first flush phenomenon which was studied for a range of catchments (Bertrand-Krajewski et al., 1998; Kim et al., 2005; Lee and Bang, 2000). Yet traditionally in LCA, pollutant discharges are aggregated over a certain time horizon and their impact characterised using steady-state mass-balance fate models. Currently it is a methodological challenge in LCA of UWS to account for discharge dynamics occurring during wet-weather flows (e.g. concentration variations at hourly or daily time steps).

Ecotoxicity impacts are particularly sensitive to time since dynamic processes drive the fate of contaminants. Traditionally in LCA, the mass of a pollutant released in the environment (i.e., the inventory result) is proportionally linked to its corresponding impact by a constant characterization factor, but time-dependent processes such as mass transfer phenomena and chemical reactions are responsible for transitory states and induce the nonlinear distribution of the impact over time (Lebailly et al., 2014; Reap et al., 2008). However, it was shown that the generated impacts can be sensitive to the mode (e.g. continuous or pulsed) and timing of discharges (Owens, 1997), which have a large influence on how they are assimilated within the river system. For example, toxic pollutants (e.g. heavy metals and organic micropollutants) may have acute impacts if released in pulse discharges (resulting in shock effects subsiding relatively fast) or cumulative impacts with continuous discharges (resulting in long term effects)(Hvitved-Jacobsen et al., 2010, 1994). Concerning freshwater eutrophication there is a clear distinction between impacts of concentrated phosphorus discharges during ecologically sensitive periods (e.g. warmer temperatures and increased light for algal growth) and impacts associated with particulate phosphorus transported in stormwater during winter storm events (Withers and Jarvie, 2008).

The objective of this study was to assess potential impacts from priority compounds and nutrient pollution in UWS discharges during wet-weather events. These discharges concern the impact categories of freshwater ecotoxicity as well as freshwater eutrophication. Marine eutrophication is also considered as rivers ultimately discharge into the oceans.

The goal and scope of the present paper was to assess inventory flows (i.e. pollutant loads from the UWS) at two temporal resolutions (year (annual average) and wet-weather event) and to use these inventory flows as data inputs in a LCA with two sub-objectives: (i) to assess the significance of pollutant discharges and impacts from “intermittent” wet-weather discharges relative to average “continuous” dry-weather conditions at the year scale; and (ii) quantify pollutant discharges and impacts from additional wet-weather flows at the scale of a “wet-weather event” by including temporal variability in the life cycle inventory (LCI) for UWS.

## 2.2 Materials and methods

The general LCA methodology described in the ISO 14040 and 14044 standards (ISO, 2006a, 2006b) was applied in this study through the four following phases: LCA goal & scope definition, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and interpretation of results which are detailed hereafter. In this study, the urban catchment of the Greater Paris area (Figure 7) was chosen to develop the assessment of pollutant discharges from a European megalopolis with a combined system servicing its inner city area.

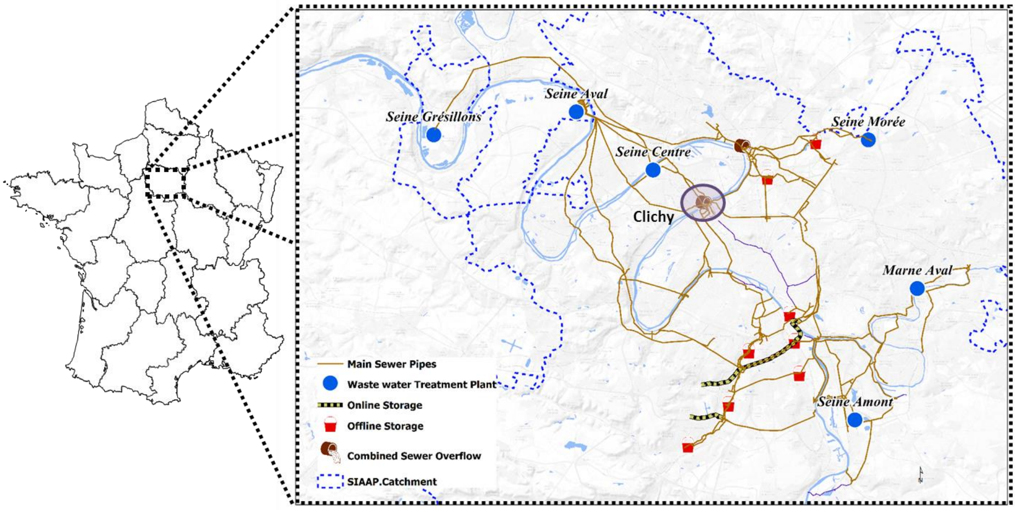


*Figure 7. Study area: SIAAP urban catchment and the main sewer network servicing the Greater Paris. Wastewater treatment plants discharging into the Seine and the Marne rivers are included (blue dots). Source: SIAAP.*

The modelling of pollutant loads from this UWS was made possible thanks to recent, qualitative and quantitative data from the Paris public sanitation service (SIAAP) and OPUR. Moreover, there is a great anthropic pressure from this densely urbanized catchment (8.9 million inhabitants as of 1st January 2014 were connected to sewer systems), which may result in significant impacts on the receiving waters (the Seine river with a median flows at Paris of about 300 $m^3.s^{-1}$). This catchment comprises Paris and 284 neighbouring towns spanning 1830 km². The inner city area of Paris (105 km²) is characterized by a continuous dense urban fabric (i.e. approximately 20,500 inhabitants/km²) with a high degree of imperviousness (>

80%) and is dominated by residential and commercial/institutional land uses and very little industrial activity. Suburbs are areas of discontinuous dense urban fabric with a medium-high imperviousness degree (50 – 80%), which are mostly residential zones with industrial activities (European Environment Agency, 2012). The Greater Paris network is mostly separate (56%), and impervious areas serviced by separate sewers were considered as the only active areas contributing to stormwater generation. At the scale of the SIAAP network, impervious surface was estimated to be 190 km² (10% of the total catchment area). A runoff coefficient value of 0.6 was chosen to calculate stormwater volumes generated from these active areas (Table 1).

*Table 1. Characteristics of the Greater Paris catchment*

| Sewer system | Drainage area (km²) | Share of total area (%) | Corresponding impervious area (km²) | Runoff coefficient value chosen for active area (1) |
|---|---|---|---|---|
| Separate | 1032 | 56% | 190 | 0.6 |
| Combined | 550 | 30% | 235 | n.a.(2) |
| Hybrid | 249 | 14% | 85 | n.a.(2) |
| *Total area (km²)* | *1830* | | *509* | |

*(1) Effective impervious area which generates stormwater and is directly connected to stream channels*

*(2) Non relevant since storm water is (partially) collected with domestic wastewater. It may contribute to CSO discharges which are assessed separately in this study.*

The mean annual rainfall is of 637 mm, for a warm temperate climate, with significant rainfall even during the driest month. There are five wastewater treatment plants (WWTP) which released 922 million $m^3$ of treated wastewater in 2013, of which 533 million $m^3$ were produced during dry weather, and 389 million $m^3$ during wet-weather. This treatment capacity amounts to an average of 2.53 million $m^3$/day (Tabuchi and Penouel, 2014).

### 2.2.1 LCA goal and scope definition

The main goal of this study is to compare relative contributions of pollutants discharged in dry-weather flows and wet-weather flows. Using the LCA framework, this study compares inventories of discharged pollutants as well as their associated impacts to receiving waters. The scope of the study covers discharges generated by an urban catchment with combined and separate sewer systems at two temporal scales, (i) during one year, and (ii) during wet-weather events. The potential impacts will be assessed for three categories relevant to waterborne pollutants, e.g. eutrophication (marine and freshwater) and freshwater ecotoxicity. Recent research carried out in the framework of the OPUR programme has provided the first comprehensive, broad overview of priority pollutant contamination of stormwater and CSOs. Since CSOs are an untreated mixture of raw wastewater and stormwater, the wastewater quality was also investigated.

The conceptual diagram in Figure 8 presents the system boundaries and inventoried flows towards the ecosphere (receiving waters) generated in the urban technosphere (man-made processes and systems) with both types of sewer networks. All flows were assessed in terms of volume and pollutant concentrations, but only flows from the technosphere are converted into potential impacts (LCIA). In Paris, the high-density inner area is drained by a mostly combined sewer system, collecting wastewater and stormwater for treatment in a wastewater treatment plant, sometimes leading to CSO discharges during storm events with a total annual volume of 13.7 million $m^3$ in 2013 (Tabuchi and Penouel, 2014). The surrounding urban developments are serviced by a separate sewer system at the end of which polluted stormwater is discharged into local watercourses (See Figure S1 in Annex A-1). The UWS includes several large storage capacities for combined flows (of wastewater and stormwater) during storm events, as well as an automated management system for these flows which enabled a significant reduction in CSO discharges in this urban catchment. Thus, other cities may have a higher proportion of CSO discharges relative to the total volume in their sewage system.

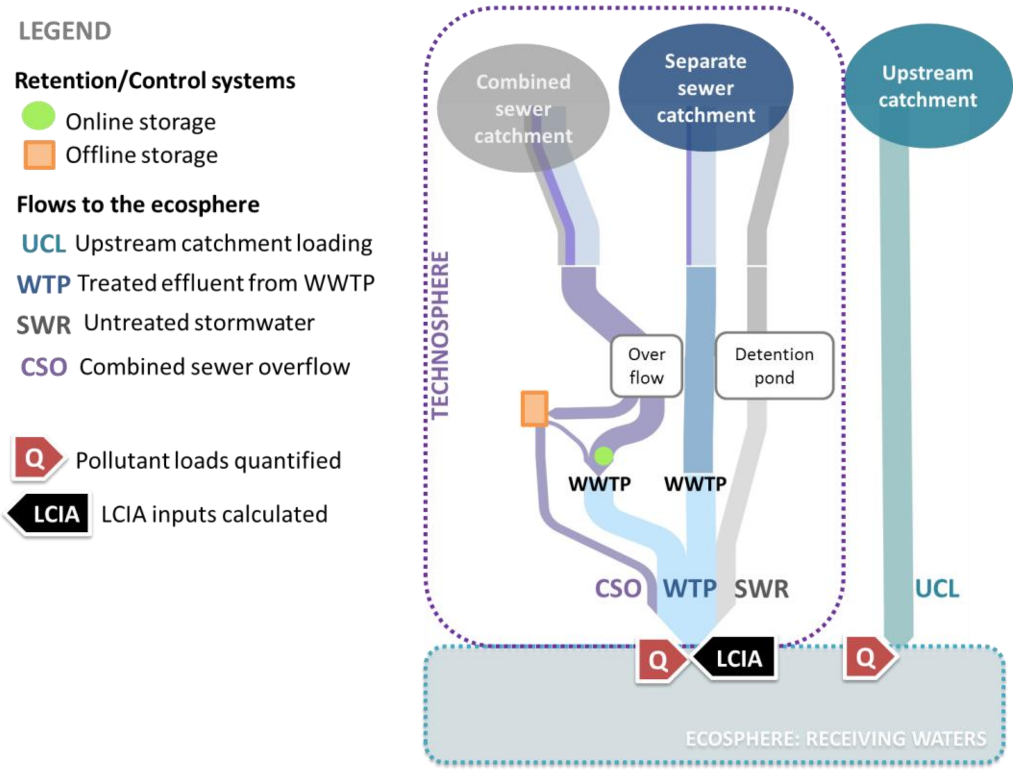


*Figure 8. Conceptual diagram of the modelled urban catchment with flows reaching the receiving waters.*

### 2.2.2 Life Cycle Inventory analysis: available data regarding flows reaching receiving waters

This section describes the available data to conduct the Life Cycle Inventory (LCI) analysis on the Paris case study.

#### 2.2.2.1 Classification of wet-weather events

Since the goal and scope of the study is to determine, at the year and event scales, the significance of wet-weather pollutant discharges to receiving waters, three categories of wet-weather events are proposed. The first event category is representative of a dry weather day (T1) where the only flows reaching the river are treated effluents. Wet-weather events (where a minimum of 1mm of rainfall has been recorded) were grouped into the two other categories: (i) T2, wet-weather with untreated stormwater reaching the river from the separate sewer systems (mild storms); and (ii) T3, wet-weather with untreated stormwater and CSO discharges reaching the river (severe storms). Table 2 summarises the characteristics of the three types of events.

*Table 2. Proposed classification for wet-weather events for the Paris case study.*

| Event type | Category | Rainfall event specificity | Flows reaching surface water | | |
|---|---|---|---|---|---|
| | | | Treated effluents (WTP) | Stormwater (SWR) | CSO discharges (CSO) |
| T1 | Dry weather | <1mm/event | X | | |
| T2 | Wet-weather mild storm | ≥ 1mm/event, no overflows | X | X | |
| T3 | Wet-weather severe storm | ≥ 1mm/event, overflows | X | X | X |

The year 2013 was chosen as reference year for flows since it was a year with an average number of wet-weather events compared to the historic time series of the Paris area. It had 231 days of dry-weather, 75 days with wet-weather events of type T2, and 59 days of type T3 were recorded, which amounts to a total of 707 mm of significant rainfall (i.e. rainfall depth greater than 1 mm) in 2013. Volumes of treated effluents and CSO discharges were measured over 24-hour periods for 2013. Surface runoff volumes were derived using the total area of active impervious sub-catchments serviced by a separate-only sewer system (190 km$^2$) and the adjusted recorded daily rainfall depth with a runoff coefficient of 0.6, which corresponds to a high density urban area. More precise stormwater volumes would be obtained with a calibrated rainfall-runoff model, using spatial differentiation and varying runoff coefficients depending on the land uses.

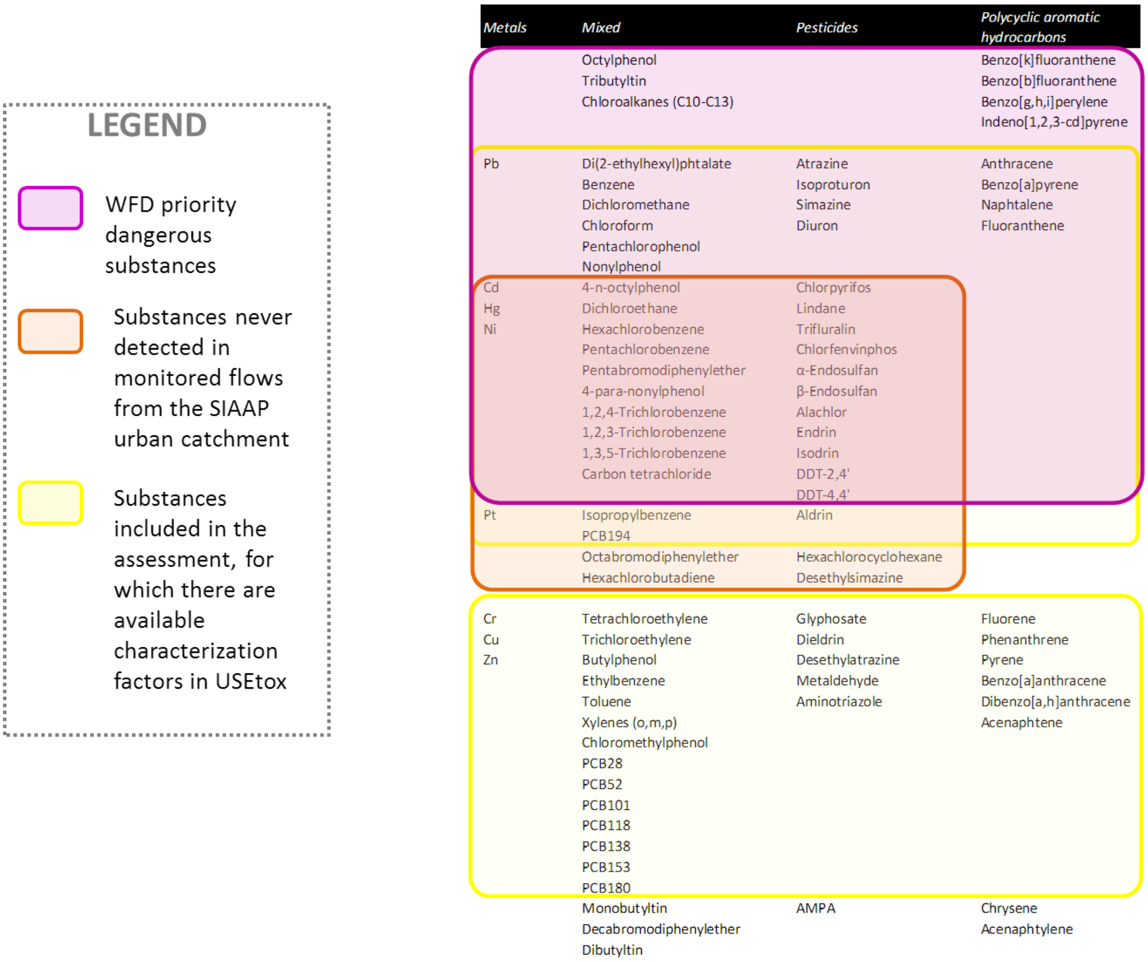
At an annual scale, untreated stormwater volume was hence estimated at 80.7 million m$^3$ (Tabuchi and Penouel, 2014).While this approach, also known as the rational method, does not acknowledge variations in stormwater volumes by using a single value for runoff coefficient for each contributing sub-catchment with impervious surfaces and separate sewers, this choice was motivated by the goal of assessing in a first order analysis, the scale of stormwater pollutant discharges in terms of inventory and impacts. Indeed, with no existing LCA studies of urban wastewater systems (UWS) during wet-weather, this study is a first step towards the inclusion of wet-weather discharges in the LCA models of UWS. For each of the flows described in Figure 8, quality parameters include common pollutants routinely measured in wastewater, and a broad range of priority pollutants.

#### 2.2.2.2 Routine water quality parameters (common pollutants)

Quality parameters measured in the inventoried flows from all WWTPs managed by SIAAP include routine wastewater quality parameters, such as ammonium, phosphates and total oxidized nitrogen (nitrates and nitrites). Data for the characterization of flows from Greater Paris include daily loads for treated effluents from the wastewater treatment plants (summarised in 2014 SIAAP's annual activity report), concentrations in stormwater from dense urban and sub-urban (separate sewer) catchments for 20 storms between 2008 and 2009 (Zgheib et al., 2012) and 52 measurements of CSO discharges from Greater Paris by Gasperi et al. (2012) in the CSO database. For stormwater, concentrations on classical parameters (ammonium, phosphates and total phosphorus) were measured, however for oxidized nitrogen forms (nitrates and nitrites), concentration mean and standard deviation in stormwater were taken from another study (Taylor et al., 2005). Data for CSO discharges was retrieved from the CSO database with 52 measurements, however there were no measured concentrations on nitrates and nitrites, which are negligible in these flows.

#### 2.2.2.3 Monitored priority pollutants

The OPUR research programme provided new insights into the stormwater quality from various urban watersheds, including the Greater Paris area (Zgheib et al., 2012, 2011). In order to include specific urban pollutants, the WFD list of priority pollutants has been extended from 33 substances to a total of 88 (80 organic substances and eight metals) chosen for their representativeness in stormwater (Zgheib, 2009; Zgheib et al., 2008). Pharmaceutical compounds were not included in the WFD list, and as the OPUR programme focussed on pollutants relevant to stormwater, these compounds were not monitored. The same methodology was applied to monitor CSO discharges from the largest CSO outfall in the Paris metropolitan area as well as raw wastewater (Gasperi et al. 2012). Of the 88 priority pollutants monitored in wet-weather flows (stormwater and CSOs), 58 were consistently found and 30 were never detected (Figure 9). Total event mean concentrations and the detection frequency are given as well as details in the measurement procedure of Gasperi et al. (2012, 2014) and Zgheib et al. (2009, 2012).



LEGEND

- 🟪 WFD priority dangerous substances
- 🟧 Substances never detected in monitored flows from the SIAAP urban catchment
- 🟨 Substances included in the assessment, for which there are available characterization factors in USEtox

| Metals | Mixed | Pesticides | Polycyclic aromatic hydrocarbons |
|---|---|---|---|
| | Octylphenol | | Benzo[k]fluoranthene |
| | Tributyltin | | Benzo[b]fluoranthene |
| | Chloroalkanes (C10-C13) | | Benzo[g,h,i]perylene |
| | | | Indeno[1,2,3-cd]pyrene |
| Pb | Di(2-ethylhexyl)phtalate | Atrazine | Anthracene |
| | Benzene | Isoproturon | Benzo[a]pyrene |
| | Dichloromethane | Simazine | Naphtalene |
| | Chloroform | Diuron | Fluoranthene |
| | Pentachlorophenol | | |
| | Nonylphenol | | |
| Cd | 4-n-octylphenol | Chlorpyrifos | |
| Hg | Dichloroethane | Lindane | |
| Ni | Hexachlorobenzene | Trifluralin | |
| | Pentachlorobenzene | Chlorfenvinphos | |
| | Pentabromodiphenylether | α-Endosulfan | |
| | 4-para-nonylphenol | β-Endosulfan | |
| | 1,2,4-Trichlorobenzene | Alachlor | |
| | 1,2,3-Trichlorobenzene | Endrin | |
| | 1,3,5-Trichlorobenzene | Isodrin | |
| | Carbon tetrachloride | DDT-2,4' | |
| | | DDT-4,4' | |
| Pt | Isopropylbenzene | Aldrin | |
| | PCB194 | | |
| | Octabromodiphenylether | Hexachlorocyclohexane | |
| | Hexachlorobutadiene | Desethylsimazine | |
| Cr | Tetrachloroethylene | Glyphosate | Fluorene |
| Cu | Trichloroethylene | Dieldrin | Phenanthrene |
| Zn | Butylphenol | Desethylatrazine | Pyrene |
| | Ethylbenzene | Metaldehyde | Benzo[a]anthracene |
| | Toluene | Aminotriazole | Dibenzo[a,h]anthracene |
| | Xylenes (o,m,p) | | Acenaphtene |
| | Chloromethylphenol | | |
| | PCB28 | | |
| | PCB52 | | |
| | PCB101 | | |
| | PCB118 | | |
| | PCB138 | | |
| | PCB153 | | |
| | PCB180 | | |
| | Monobutyltin | AMPA | Chrysene |
| | Decabromodiphenylether | | Acenaphtylene |
| | Dibutyltin | | |

*Figure 9. List of 88 substances frequently found in stormwater, complementing the WFD list of priority pollutants (Zgheib et al., 2008). Among these substances, there are EU WFD priority dangerous substances (purple), substances never detected in monitored flows.*

Concentrations (average and standard deviation values) of priority pollutants in treated (secondary) effluents discharged from wastewater treatment plants in the Paris UWS have been monitored in a recent study of the removal efficacy of a tertiary treatment step (Mailler et al., 2015) at the Seine Centre WWTP (Paris Agglomeration). Where data concerning relevant pollutants was missing from this study, concentration and occurrence data in raw wastewater given by Gasperi et al. (2012) was used for some priority pollutants, along with removal efficacies in a conventional activated sludge treatment step (Ruel et al., 2012, 2010). Priority pollutant concentration data in stormwater was retrieved from the campaign on 20 storms by Zgheib et al. (2012), while for CSO discharges only four rain events in 2010 were monitored (Gasperi et al., 2012).

Pollutant discharges which occurred upstream of the UWS in this study (upstream catchment loading) and measured at Choisy-le-Roi, were included in the inventory for information purpose despite the scarcity of data on the range of priority pollutants. However, these upstream discharges were not converted into impacts as they are not UWS discharges per se. A summary of the data used is available in the Supporting Information (See Table S1 in Annex A-1).

In this study it was assumed that the treatment efficacy in the WWTP remains unchanged even during wet-weather periods with CSO discharges where it is evident that the WWTP is overloaded. This assumption can hold for the SIAAP wastewater treatment plants since it has been shown that the treatment efficacy of the Seine-Centre WWTP was still good during wet-weather periods (Gilbert et al., 2012). This is a simplification made for this study as the focus is not on the WWTP treatment performance but rather on the unmanaged flows reaching the receiving waters.

#### 2.2.2.4 Statistical treatment of LCI data

Pollution in runoff from urban areas and roads is complex compared to the pollution originating from e.g. a specific treatment plant. Its chemical characteristics are dependent on the nature of urban surfaces which are washed off, as well as natural processes (e.g. wet and dry deposition) and anthropogenic activities in the catchment. Rainfall is a stochastic phenomenon hence it becomes relevant to describe this pollution in terms of a probability, a frequency of occurrence or a return period (Hvitved-Jacobsen et al., 2010).

As a preliminary analysis, the concentrations of common pollutants in wastewater (ammonium, phosphates and total oxidized nitrogen) were tested for lognormality using a Chi-square goodness-of-fit test. These parameters indeed followed a lognormal distribution in wastewater which was also demonstrated for organic matter and total suspended solids in stormwater across several sites in France (Gasperi et al., 2014). The number of priority pollutants measurements was not sufficient to conduct proper lognormality tests. Their concentrations were therefore assumed to follow lognormal distributions similar to the common pollutants.

In order to assess the variability in inventory flows at the scale of a year and during wet-weather events, it is required to determine the mass load of pollutant $i$ (mean and standard deviation, in kg) in a given flow emitted into the environment from a single storm or a series of storm events (year scale). Using the pollutant concentration value and volume of flow emitted, the mass load of pollutant $i$ is computed as follows:

$$\left(\overline{m_\iota}, \sigma_{m_i}\right) = \left(\overline{c_\iota}, \sigma_{c_i}\right) * V$$

With:

$(\bar{c_i}, \sigma_{C_i})$, mean and standard deviation in the concentration in pollutant *i* (mg/L or µg/L); $(\overline{m_i}, \sigma_{m_i})$, mean and standard deviation mass of pollutant *i* emitted (kg), and V the volume of flow reaching the river ($m^3$) which can be a yearly volume or an event volume.

However, given the various sources and sampling strategies of urban flows, the obtained data on pollutant concentrations is heterogeneous in quality and the statistical treatment resulting in inventory flows (mean and standard deviation) is detailed in Figure 10.

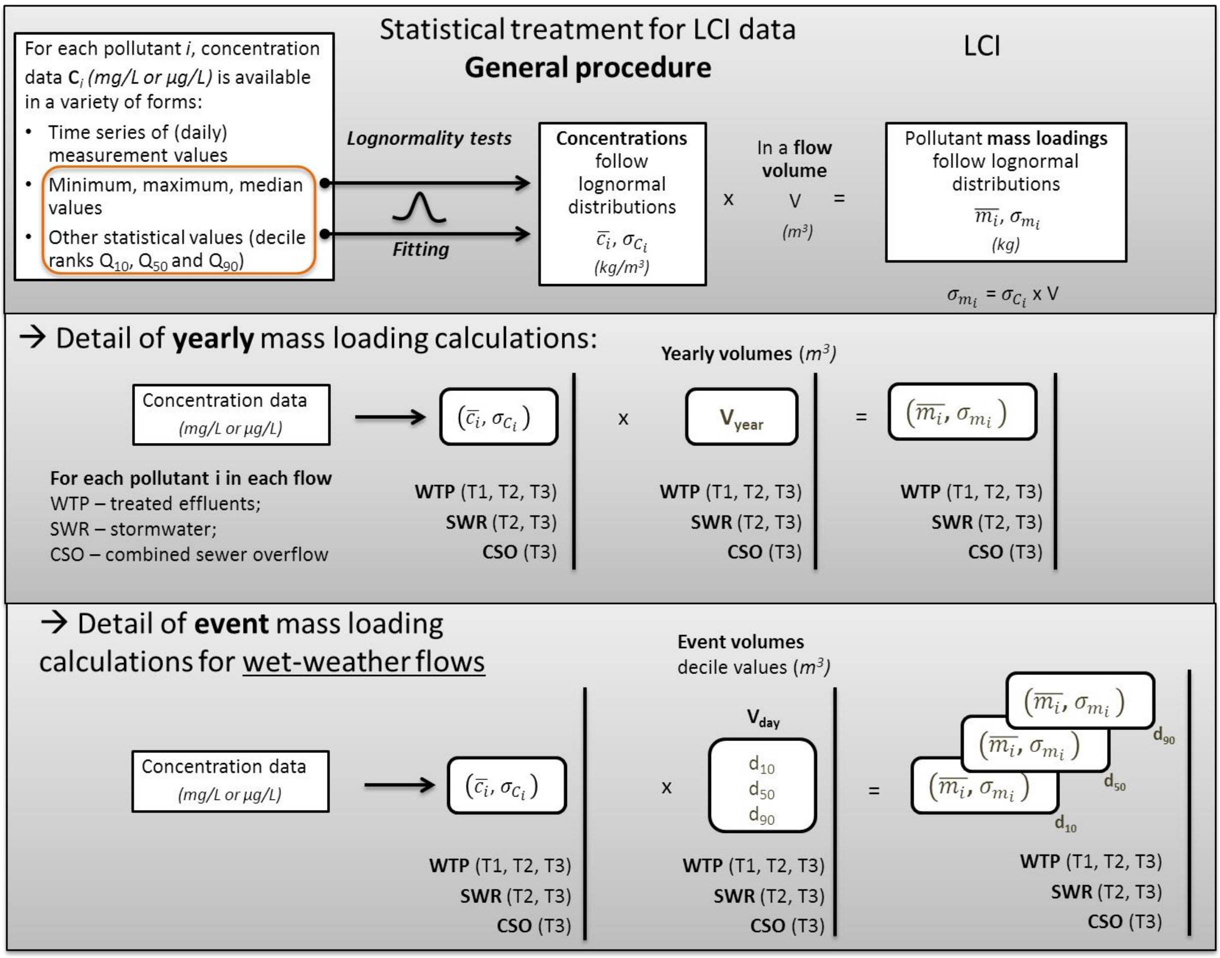


*Figure 10. Statistical treatment of data to compute inventory flows for the pollutants found in treated wastewater effluents (WTP), stormwater (SWR) and combined sewer overflow (CSO) discharges.*

On one hand, for some flows, time series or daily measurements (e.g. for common pollutants in treated WWTP effluents) are available for which the concentration mean and standard deviation are straightforward to compute. While on the other hand, statistically-processed concentration data are available in a variety of forms such as (minimum, maximum, median values) or decile ranks (e.g. d10, d50, d90). In this case, under the hypothesis of lognormality, the heterogeneous concentration data is fitted to a lognormal probability distribution using the Matlab® original "fitdist" function, which then yields a concentration mean and standard deviation. Concerning priority pollutants which are measured with variable frequencies in samples (because they are often found at low concentrations, sometimes below the current limits of quantification in the order of the ng/L), we chose to account within each flow for frequencies of occurrence (in %) for each priority pollutant. Mean pollutant concentrations were adjusted with the associated frequencies of occurrence (resulting in weighted average concentrations) within each flow.  Indeed, a pollutant which was measured with a frequency of 25% (i.e. in one out of four samples) should be less represented than a pollutant systematically measured in a given flow.

In order to characterize the specific volume distribution in wet-weather flows at the scale of a wet-weather event (T2 and T3), these flow volumes were estimated with three statistical values: 1st, 5th (median) and 9th deciles. The volumes used for the inventoried flows at year and event scales are summarized in Supporting information (See Table S2 in Annex A-1). Finally, for each flow pollutant mass loads at the year scale were estimated using concentration values and total volume. At the event scale, daily event volumes were used instead of the total volume to compute event pollutant mass loads.

### *2.2.3 Life cycle impact assessment*

#### *2.2.3.1 LCIA method selection*

This specific application of LCA concerns only discharges to water and does not comprise the entire life cycle of the studied urban water system including infrastructure and operation. In this context, impact categories such as resource depletion or climate change are not relevant and only a small set of indicators related to water pollution must be selected from the available methods.

A requirement of the ISO14044 standard (ISO, 2006b) is that the characterisation factors be based on environmental mechanisms that link human interventions to a set of areas of protection. For freshwater eutrophication, the generally accepted assumption in LCIA is that only phosphorus compounds are contributing to this impact, as the majority of freshwater ecosystems are limited by phosphorus availability. However, coastal marine waters are assumed to be nitrogen-limited; hence nitrogen compounds reaching the ocean potentially cause marine eutrophication.

The ReCiPe Midpoint v1.09 (H) method (M. Goedkoop et al., 2009) was chosen for the impact assessment of wastewater pollutants causing freshwater eutrophication and marine eutrophication as it is based on the latest recommendations by the European Commission (JRC European commission, 2011). Freshwater eutrophication and marine eutrophication potentials for phosphates, total oxidized nitrogen and ammonium are expressed respectively as kgP and kgN.

Regarding the impact assessment of priority pollutants on freshwater ecotoxicity, USEtox was used (Henderson et al., 2011; Rosenbaum et al., 2008). In the latest version USEtox v2.01, an updated ecotoxicological characterization model for metals was included (Diamond et al., 2010; Dong et al., 2014; Gandhi et al., 2010). The freshwater ecotoxicity potential is expressed in potentially affected fraction of species (PAF) at midpoint level due to change in substance concentration in freshwater, integrated over the freshwater volume ($m^3$) per substance mass emitted to freshwater (kg), PAF.$m^3$/kg. Of the 58 priority pollutants monitored in all urban flows, 43 had ecotoxicity characterization factors and were included in the study.

## 2.3 Results

Details on inventory results (i.e. mass loads of pollutants entering the receiving waters) and impact assessment scores on the aquatic categories for the monitored flows are available Annex A-3. The section below presents the general results of the calculated mass loads and impact contributions for the three classes of archetypal weather days for the Greater Paris area: dry weather (T1), wet-weather without discharges of untreated sewage from CSOs (T2), and wet-weather with discharges of raw untreated sewage from CSOs (T3).

### *2.3.1 Pollutant loads (LCI)*

#### *2.3.1.1 Year scale*

In 2013, WWTPs discharged a much greater volume aggregated over dry- and wet-weather days (922 million $m^3$) than the runoff due to wet-weather events (respectively 27 million $m^3$ and 53.7 million $m^3$ across resp. all T2 events and T3 events) and CSO discharges (13.7 million $m^3$) (See Table S2 in Annex A-1).

Some common pollutants showed a great range of variation across the monitored flows. The load of total oxidized nitrogen was found to be three orders of magnitude larger in treated effluents compared to wet-weather loads (See Figure S2 in Annex A-3). Generally, the loads from treated effluents were higher than the wet-weather loads (aggregated wet-weather flows in 2013). Data on yearly loads of common pollutants from the upstream catchment was available only for ammonium and total phosphorus.

Most wet-weather loads of priority pollutants were in the same order of magnitude as those in treated effluents (See Figure S3 in Annex A-3). Regarding metals, the inventory results are expressed as total metal loads, using the free ion fraction calculated in the LCIA method, USEtox.

All metal species were found in comparable loads across all flows, with copper and zinc also present in the upstream catchment loads. The loads of four polycyclic aromatic hydrocarbons (PAH) (anthracene, fluoranthene, pyrene, and dibenzo[a,h]anthracene) were an order of magnitude higher in stormwater than in treated effluents and CSO discharges. Of these PAHs, the first three and benzo[a]pyrene showed important loads from the upstream catchment. Pesticides did not show a clear tendency with some showing higher loads in treated effluents (isoproturon) while some were dominant in stormwater (glyphosate). Two alkylphenols (4-nonylphenol and butylphenol) were found in similar amounts and almost always detected (measurement occurrences greater than 90%) in treated effluents and stormwater.

From a quantitative point of view, the total metal forms of copper and zinc were the two compounds with the greatest annual discharged loads, in the range of $10^3$ to $10^4$ kg per year. They are followed by lead, DEHP, chromium, and glyphosate in the range of $10^2$ to $10^3$ kg per year.

#### *2.3.1.2 Wet-weather event scale*

Event scale loads are shown in the following section for median volume values (5th decile, d50) of wet-weather flows. At the wet-weather event scale common pollutants followed the same trends as the year scale, without any marked differences in loads of common pollutants between dry-weather treated effluents and wet-weather flows. Hence the pollution generated during wet-weather events was quantitatively similar to the pollution of the treated effluents (See Figure S4 in Annex A-3).

At the event scale priority compounds also showed similar trends as the year scale, only a bit more marked. Copper and zinc were found in comparable amounts in dry weather treated effluents and wet-weather flows. However lead showed a load one order of magnitude greater in stormwater and CSO discharges than in treated effluents. PAHs that were predominant in stormwater at the year scale are even more so at the event scale with a difference in substance loads of up to two orders of magnitude.

From a quantitative point of view, discharges of all metal elements, nonylphenol, DEHP and glyphosate were in the range of 1 to 10 kg/d when most other priority compounds loads ranged between $10^{-4}$ to $10^{-1}$ kg/d (See Figure S5 in Annex A-3).

### 2.3.2 Impact assessment

Routine water quality parameters are the only substances counting towards the aquatic categories of eutrophication (marine and freshwater) while all priority compounds inventoried in this study are counting towards the freshwater ecotoxicity category.

At the year scale, contribution analysis for freshwater ecotoxicity showed that wet-weather flows (stormwater from T2 and T3 events, and CSO discharges from T3 events) moderately contributed to the total impact on freshwater ecotoxicity (34%), with treated effluents accounting for the remaining 66% (Figure 11). Of the wet-weather flows, CSO discharges were the least contributing, followed by untreated stormwater. Results for the eutrophication categories (marine and freshwater) showed a similar trend albeit less marked, and are presented in Annex (See Figures S6 to S11 in Annex A-3).

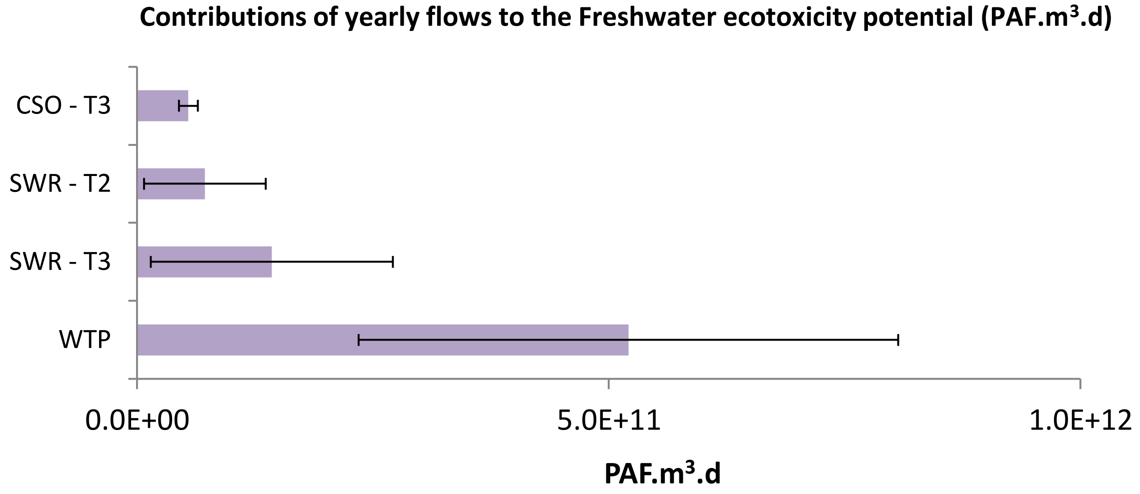


*Figure 11. Contributions to the Freshwater ecotoxicity impacts (PAF.m3.d) of urban dry- and wet-weather flows at year scale.*

At the event scale, wet-weather flows had even greater contributions on freshwater ecotoxicity with up to 62% of the total impact in a T3 wet-weather event (Figure 12).

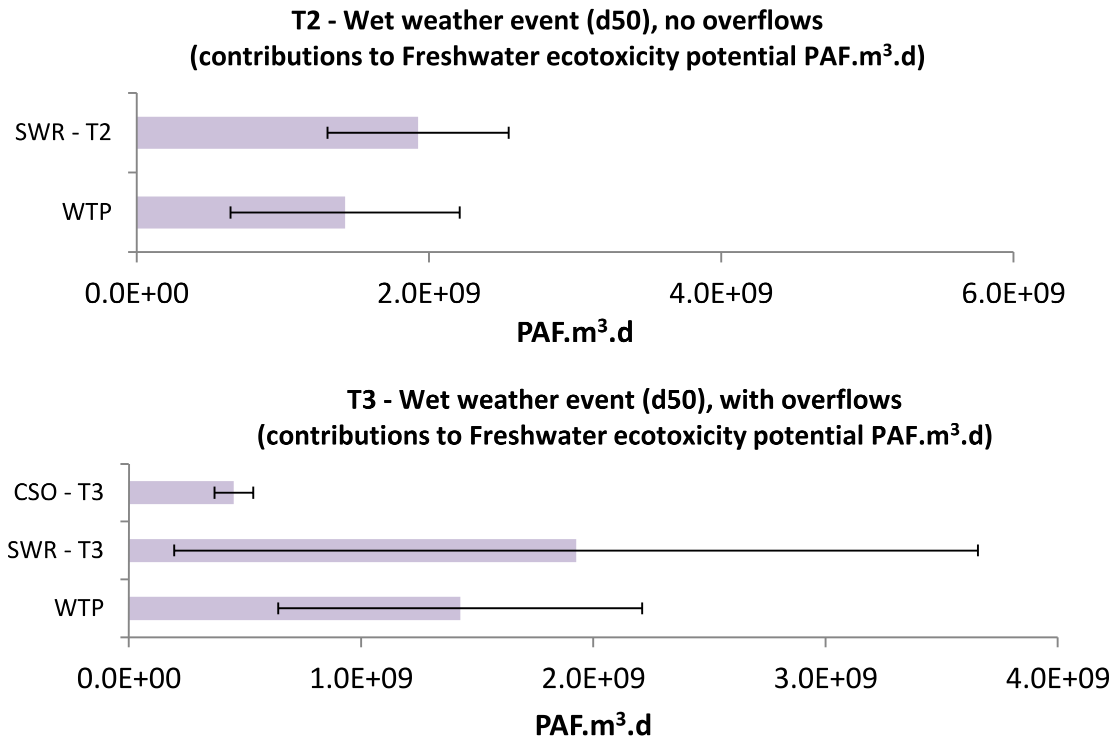


*Figure 12. Contributions to the Freshwater ecotoxicity impacts (PAF.m3.d) of urban dry- and wet-weather flows at event scale (d50, median wet-weather volumes.)*

A detailed contribution analysis showed the notable contributors on the freshwater ecotoxicity impact at the year scale (Table 1). The ranking was not modified when considering the event scale as the contributions of the three most important pollutants did not change, therefore masking variations from the minor contributors (under 5%).

*Table 1. Top 6 contributors to freshwater ecotoxicity. B[a]a, Benzo[a]anthracene; Flu, Fluoranthene; 4-NP, 4-nonylphenol.*

| **At year scale** Freshwater ecotoxicity impact (%) | **T1** *treated effluents* | | **T2** *untreated stormwater* | | **T3** *untreated stormwater* | | **T3** *combined sewer overflows (untreated)* | |
|---|---|---|---|---|---|---|---|---|
| Rank # | **WTP** | | **SWR** | | **SWR** | | **CSO** | |
| 1 | Cu (II) | 9.2E+01 | Cu (II) | 9.4E+01 | Cu (II) | 9.4E+01 | Cu (II) | 8.7E+01 |
| 2 | Zn (II) | 7.9E+00 | Zn (II) | 6.1E+00 | Zn (II) | 6.1E+00 | Zn (II) | 1.3E+01 |
| 3 | 4-NP | 2.4E-02 | Pyrene | 1.9E-01 | Pyrene | 1.9E-01 | Pyrene | 4.6E-02 |
| 4 | Cr (III) | 5.5E-03 | B[a]a | 2.7E-02 | B[a]a | 2.7E-02 | Dieldrin | 3.1E-02 |
| 5 | Pyrene | 2.7E-03 | 4-NP | 2.4E-02 | 4-NP | 2.4E-02 | B[a]a | 1.8E-02 |
| 6 | Isoproturon | 9.1E-04 | Flu | 9.4E-03 | Flu | 9.4E-03 | Pb (II) | 6.2E-03 |

Table 1 underlines that two metal elements (Copper (II) and Zinc (II)) clearly dominated this impact, as they alone made up for over 99% of the ecotoxicity score, in all considered flows.

Then in 3rd position there was 4-nonylphenol in treated effluents and pyrene in wet-weather flows.

Detailed contribution analysis of the marine eutrophication results demonstrated that this impact was mostly driven by total oxidized nitrogen ($NO_2^-$ and $NO_3^{2-}$) in treated effluents and by ammonium ($NH_4^+$) in both wet-weather flows at the year scale (See Table S2 in Annex A-3). Freshwater eutrophication was mostly driven by total phosphorus in all flows.

## 2.4 Discussion

### *2.4.1 Discharges from the UWS*

Despite a reduced total annual volume for both wet-weather flows (i.e. stormwater and CSO discharges, arising to 94.4 million $m^3$) in comparison with WWTP effluents (922 million $m^3$), the computed loads at a year scale for most priority compounds in these flows were within the same order of magnitude as in treated WWTP effluents. This is due to relatively high measurements in wet-weather flows. The predominance of some PAHs in stormwater is expected as these compounds originate from road traffic.

- ***Representativeness of available data used to calculate mass loads***

Data representativeness in stormwater pollutants could be increased by accounting for the significant site variability by using pollutant concentrations from other catchments. Concerning available data on priority compounds which is summarized in Table S2 (Annex A-2), the heterogeneity in representativeness of measurements to determine concentration values across all flows is notable, with CSO discharges (n=4) (Gasperi et al., 2012), treated wastewater effluents (n=11 or 14) (Gasperi et al., 2012; Mailler et al., 2015), and stormwater (n=17) (Gasperi et al., 2012; Zgheib et al., 2012). Also, priority compounds were generally detected across all flows during the measurement campaigns but some were detected at a frequency of 30% or lower, especially in stormwater and CSO discharges (e.g. volatile organic compounds such as benzene, ethylbenzene, toluene, xylenes, tetrachloroethylene). Accounting for the frequency of detection in the load calculation has given a better representation of the contamination by these compounds. However, uncertainties around the concentration values used may be of the same order of magnitude as the mean concentration values; this is why care should be taken in the interpretation of associated impacts on ecotoxicity. Concerning volumes derived from daily recorded rainfall depths, the associated uncertainties were low compared to uncertainties associated with concentration measurements primarily due to the sampling procedure (few samples for wet-weather flows). Results using most conservative volume values ($9^{th}$ decile, d90) of wet-weather flows are given in the Supporting information file (See Figures S8, S11, S12 in Annex A-3), and the general conclusions remain the same.

- ***Consideration of point-source and non-point source pollutants***

This study included point source pollutants (e.g. alkylphenols and PCBs in treated wastewater) and non-point source pollutants (e.g. PAH and pesticides from untreated stormwater). LCA studies of UWS traditionally focused on continuous point-source pollutants (discharges from WTP), omitting non-point source pollutants linked with untreated stormwater from episodic storm events. Indeed, current LCIA methods are steady-state models and do not account for discharge dynamics. Concerning freshwater ecotoxicity, the USEtox LCIA model assesses toxicological effects of over 3000 chemicals emitted into the environment, and the outputs of this model are characterization factors for these chemicals. They represent the cause-effect chain of freshwater ecotoxicity and are modelled by a fate component, as well as an exposure and toxicity components (Henderson et al., 2011). Then, characterization factors are used to make quantitative rankings for chemicals. However, this model does not differentiate between point-source, chronic discharges and non-point source, episodic discharges. In order to cope with this issue, we propose an LCI of non-point source pollutant discharges for several weather categories calculated at the scale of a day. Then, the impacts of these pollutant discharges are calculated by using USEtox characterization factors.

### *2.4.2 Upstream catchment loadings*

It is interesting to discuss the significance of the mass loadings from the UWS relative to upstream catchment loadings in the river Seine, which are flows originating upstream of the SIAAP urban catchment and measured at Choisy-le-Roi. As shown in Figure S3 in Annex A-3, mass loadings for six priority pollutants which were monitored in the upstream river section indicate that there may be a significant “legacy” of pollutants from sources upstream of the UWS which include highways and roads. Indeed, the link between vehicular combustion products and runoff quality has been confirmed in various studies that show that highway discharges have a specific chemical fingerprint including PAHs and metal species (D’Arcy et al., 2000; Ellis et al., 1997; Ellis and Mitchell, 2006). However, the scarcity of the monitoring data on priority pollutants in upstream river sections (only seven compounds) is notable, given that the scope of the regulatory analysis of upstream flows is rather limited.

The research on urban flow contamination by priority pollutants could certainly benefit from a broader range of priority pollutant concentrations to help assess the relative importance of the mass loadings from the SIAAP urban catchment.

### *2.4.3 Life cycle impact assessment results*

- ***Comprehensiveness of ecotoxicity assessment***

Application of the latest LCIA method USEtox to the mass loadings defined in the inventory step yields results for the priority compounds included in this study. These first results are in agreement with other studies on different catchments where heavy metals (especially copper and zinc), PAHs, DEHP and pesticides were found to be the main pollutants of general concern in stormwater runoff (Birch et al., 2011).

However, it is important to bear in mind that they show only a fraction of the total impact on ecotoxicity, given that there are many substances that are missing from the inventory because of a lack of characterization factors to date. Also, in literature, the presence of other pollutants such as perfluoroalkyl compounds, polybrominated diphenyl ethers, and bisphenol A in urban flows is confirmed (Gasperi et al., 2014). Hence including more priority compounds requires new characterization factors, and some compounds that should be added are listed in Figure 9.

- ***Impacts from metal species on freshwater ecotoxicity***

Two metal species (Cu(II) and Zn(II)) are consistently ranked as top contributors in freshwater ecotoxicity across all flows. These metals are therefore of high priority. Besides significant amounts of metals discharged (described in the inventory section 3.1), the characterization factors of these metals are also high with Pb(II) having the greatest ecotoxicity potential in freshwater followed by Cu(II) and Zn(II) (Gandhi et al., 2011). Characterization factors and the parameter values for the most contributing priority compounds identified in Section 3.2 are given in Table S4 in Annex A-3. Cu(II) has the highest factor followed by dieldrin and pyrene, and Zn(II). It should be noted that as a step ahead of its predecessor USEtox 1.0, recent developments implemented in USEtox 2.01 have addressed metal-specific issues such as speciation, bioavailability and freshwater chemistry (Diamond et al., 2010; Dong et al., 2014; Gandhi et al., 2010), as well as updated ecotoxicological effect data as described in Fantke et al. (2015).

## 2.5 Conclusions

This paper proposes an approach to account for intermittent wet-weather discharges from an urban wastewater system in LCA and assess their relative importance regarding water quality impacts (eutrophication and ecotoxicity). This method was applied to the Greater Paris catchment, whose urban wastewater system is representative of a European megalopolis. Major results for this case study are highlighted in the following, as well as recommendations and challenges on the way to conduct such a LCA:

- Data quality was very heterogeneous, which required a probabilistic approach to get a consistent life cycle inventory for all flows. For example, data on classical parameters in wet-weather flows was scarce. While priority pollutants were the recent focus of research in dry- and wet-weather discharges, there are still research efforts to be made to better characterize CSO discharges (only four events monitored for the case study).
- The proposed choice of wet-weather event classification into categories allowed to better define wet-weather discharges and underlined the importance of their associated pollution relative to dry-weather discharges from an urban wastewater system at the scale of a day. This study demonstrated that for the (climatic and infrastructure-related) characteristics of the UWS servicing the Greater Paris, the potential impacts from episodic wet-weather discharges were significant also at a year scale, despite the stormwater management.
- Compared to untreated stormwater, CSO discharges were potentially less contributing to (ecotoxicity and eutrophication) impacts. It may be explained by the low vulnerability of the Greater Paris UWS which features an important storage capacity during wet-weather events, coupled to the dynamic management of flows (routing inside the sewer system), which allow for few combined sewer overflows.
- In the case of the Greater Paris catchment, it was demonstrated that at the event scale, stormwater flows are the greatest contributors to the freshwater ecotoxicity impact because of two metal elements (copper and zinc) found in those flows, even with a conservative runoff coefficient value. Impacts on freshwater ecotoxicity may therefore be greater with higher runoff coefficients (e.g. 0.8 or 0.9).
- Stormwater management upstream of the catchment would be interesting to reduce stormwater volume using a blue-green infrastructure to promote local infiltration (e.g. with raingardens or bio-retention systems) and where necessary, to treat locally more

polluted stormwater from urban surfaces with greater release rates (Bressy et al., 2012).

- Given the significance of the wet-weather discharges, further research into how to include temporally-differentiated discharges in the LCIA framework for eutrophication and ecotoxicity is needed to better understand how the performance of an UWS system affects the receiving environment at different local weather conditions.
- The LCA approach is well-suited to a comparative assessment of wet-weather infrastructures in urban water systems, using physically-based drainage systems models coupled with an inventory (LCI) of wet-weather discharges to assess the changes in impacts at the event scale for different event categories. However, the inclusion of acute toxicity impacts following a peak discharge at the beginning of a storm (e.g. first flush phenomenon) remain a methodological challenge.

## Acknowledgements

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# Chapter 3.: A parsimonious site-dependent framework linking human activities, land use and climate to determine loads in intermittent discharges to receiving waters from urban catchments for LCI purposes

*Chapter 3. A parsimonious site-dependent framework linking human activities, land use and climate to determine loads in intermittent discharges to receiving waters from urban catchments for LCI purposes.*

## *Contents of Chapter 3*

*The previous Chapter identified several methodological challenges for including intermittent discharges in terms of quality and quantity in LCA studies of UWS. It emerged that the missing piece of the puzzle to enable comprehensive assessments of global environmental impacts of UWS was the characterization of stormwater loads in relation to the importance of nonpoint sources of urban pollutants resulting from activities within the urban catchment. Thus, Chapter 3 establishes a site-dependent framework for estimating annual wet-weather pollutant exports to receiving waters from urban catchments under given climatic conditions. The question of the influence of climatic conditions (especially the temporal succession of dry and wet-weather periods) on stormwater pollution generation is considered and further developed in Annex B-1.*

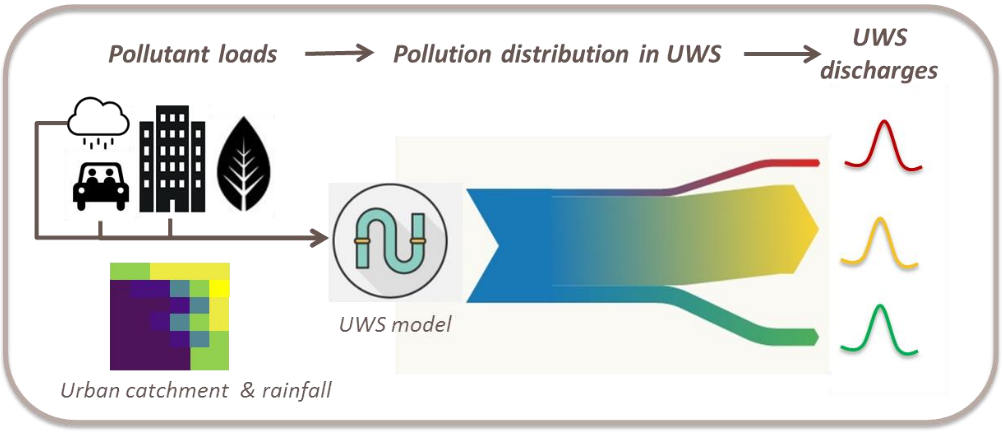


*Figure 13. Graphical abstract of Chapter 3*

## 3.1 Introduction

Globally, more people live in urban areas than in rural areas, with 55% of the world's population residing in urban areas in 2018, and by 2050 68 % of the world's population is projected to be urban (United Nations, 2018). Urban growth is synonymous with growing impervious areas with limited flow infiltration. Future projections (Seto et al., 2012) suggest that, if current trends continue, urban land cover will increase by 1.2 million km2 by 2030, nearly tripling global urban land area between 2000 and 2030. Over the last few decades, this has given rise to the challenge of managing ever increasing and polluted stormwater with up to 600 compounds identified (Chebbo and Gromaire, 2004; Eriksson, 2002; Gasperi et al., 2014; Gosset et al., 2016; Makepeace et al., 1995). In addition to the complex chemical composition of stormwater discharges, their pollution is also subject to considerable variability from runoff event to event and from site to site (Gromaire-Mertz et al., 1999; Kayhanian et al., 2003). This is due in particular to the diversity of urban surfaces leached by rain, temporal variations of runoff flows through time and climatic conditions prior to rain events (Kafi et al., 2008).

During moderate to heavy rains, stormwater generated in urban areas can overwhelm combined sewer systems (collecting together stormwater and wastewater) when high volume flows exceed the capacity of the downstream network (including the treatment plant). This leads to the discharge of the excess water into adjacent receiving water through combined sewer overflow (CSO) structures. In Chapter 2, we investigated the importance of intermittent discharges (IDs) (e.g. untreated CSO and stormwater discharges) in relation to continuous discharges (CDs) from point sources (e.g. effluents from wastewater treatment plants) using a life cycle analysis. At the scale of a year, it was found that IDs contributed significantly to freshwater eutrophication and ecotoxicity. These results are in agreement with findings that wastewater treatment plant (WWTP) effluents and IDs can have equivalent contributions to annual pollutant loadings to aquatic ecosystems (Bertrand-Krajewski, 2006; Brombach et al., 2005; Lijklema et al., 1993; Niemann and Orth, 2001).

However, the continuous increase in stormwater discharges raises the question of how conventional centralized "end-of-pipe" UWS infrastructures (e.g. combined sewers) with limited storage capacity and flexibility will adapt to future climatic variability and urbanization. It is then critical to assess the environmental performances of urban water systems. Among tools and methods, life cycle assessment (LCA), a standardized life cycle and multi-criteria approach, is more and more often used to assess impacts of UWS (Loubet et al., 2014). One of the major advantages of this tool is that it avoids shifting burdens between life cycle stages or between different environmental issues (Hauschild, 2005; ISO, 2006a).

Yet, LCA was originally designed as a steady-state tool (Collet et al., 2014; Udo Haes, 2006) that excludes temporal information necessary to obtain results with sufficient environmental relevance in time-dependent environmental processes. The lack of a time dimension in both life cycle inventory (LCI) and life cycle impact assessment (LCIA) steps is a recognized limitation of the LCA method (Finnveden et al., 2009). However, recent methodological developments now support the integration of the time dimension in dynamic LCA (DLCA) as reviewed in (Shimako, 2017). In full DLCA, temporally differentiated LCI that describe system dynamics are calculated and used as inputs to time-dependent LCIA models to compute environmental impacts through time (Beloin-Saint-Pierre et al., 2016; Shimako et al., 2018, 2017). The remaining implementation challenge of full DLCA studies seems to concern the management of temporal information for system description and modeling (Beloin-Saint-Pierre et al., 2016).

Chapter 2 discussed a number of problems caused by ignoring temporal dynamics in life cycle assessment for ecotoxicity and eutrophication. It is therefore necessary to tackle the methodological challenge of capturing temporal variability in UWS dynamics with IDs during wet-weather events. Hence, a complete assessment of UWS requires considering IDs in addition of dry-weather CDs in the LCI step to fully compare the environmental implications of present-day and future UWS management strategies over the lifetimes of their infrastructures.

The general aim of this work is set on developing a methodology to characterize urban IDs for LCI purposes in terms of annual volumes and annual pollutant loads for a given urban catchment under a specific climate. The methodology is named “STOIC” as an abbreviation for “STOrmwater Inventory Conceptualization”. It satisfies requirements on inventory data quality to support sufficiently accurate modelling of IDs and conceptualizes stormwater processes occurring in the UWS of an urban catchment. More particularly the different objectives of this Chapter are:

- To align the STOIC framework on defined objectives and specifications for consistent LCI modelling of UWS (section 3.2);
- To explain how the STOIC framework was developed (section 3.3) resulting in the proposed modelling approach and its practical implementation (section 3.4).

## 3.2 Objectives and specifications for LCI modelling of UWS

### *3.2.1 Goal and scope of targeted LCAs*

The challenge related to the goal and scope of LCA applied to UWS is to enable different UWS management strategies or alternatives involving additional infrastructures (e.g. for increased water retention in combined UWS). Ultimately, the comparison between strategies or alternatives would be based on net environmental impacts resulting from (i) induced impacts with the operation and infrastructure of UWS, and (ii) avoided impacts with the handling of pollution in urban IDs. As LCA is a global approach, the LCI modelling should be sufficiently generic to adapt to any location around the world by capturing to some extent site specificities. The framework should be based on data and models available across countries and catchments. In addition, adapting the necessary model resolution to the LCI requirements is a key point to be solved.

### *3.2.2 Resulting LCI requirements*

In order to develop LCI of urban IDs from UWS with the objective of comprehensive LCA results of the whole UWS or comparisons of UWS scenarios, the data collection and modelling in this framework will address issues and specifications related to the following points based on best practice in LCA data quality requirements (hereafter referred to as R) (Weidema and Wesnaes, 1996):

R1. ***Water quantity (water flow balance):*** the framework should ensure that within the UWS, input and output water flows are balanced.

R2. ***Water quality (pollutant mass balance):*** With the principle of mass conservation, inputs and outputs of pollutant loads should be balanced within the UWS ensuring there is no burden shifting between different UWS discharges (e.g. stormwater, untreated CSO discharges or WWTP effluents) (Loubet et al., 2016; Risch et al., 2011).

R3. ***Data reliability:*** For such a simplified framework (i.e. sufficiently generic to adapt to any location worldwide), data quality will be compromised as getting verified data based on measurements would not be feasible. Hence, the data will be mostly non-verified and based on measurements (on site, or on a similar site), expert knowledge or assumptions.

R4. ***Appropriate spatial scale:*** In a conventional LCA of UWS, the studied urban catchment is rarely described and at best, the description is a generic, lumped land use approach based on basic input data such as runoff coefficients, areas per land use and standard pollutant concentrations (See Chapter 2). While this approach can estimate pollutant loads correctly, because no physical parameter describing the catchment is used, forecasting scenarios (e.g. to evaluate effects of changes in land use, urban activities or UWS management) cannot be built or evaluated. In order to capture the diversity of urban catchments and UWS it is necessary to model urban catchments with a finer resolution. Hence we propose to characterize the land use and land cover at the scale of sub-catchments with homogeneous land use (i.e. neighborhood blocks). The resulting LCI modeling will hence be site-dependent.

R5. ***Appropriate temporal scale:*** Generally, in LCA of UWS, impacts are calculated around the yearly horizon (over the life cycle of systems), with UWS infrastructures typically lasting several decades (25 to 100 years). The time scale to capture a single wet-weather event in a UWS is of the order of the minute to describe variations of rainfall intensity but can be extended to one hour or one day. It is why models typically used to simulate urban hydrology/hydraulics are computing flows at short time intervals to ensure numerical stability (generally sub-hourly and often using 10-minute time intervals with dynamic flow equations). Therefore, in order to estimate ID loads from UWS over a sufficiently long temporal scale to include wet-weather events that are representative of climatic conditions, the resolution of flow transport will be on short time intervals (e.g. 10-minute time intervals) and the modelling period will be over a yearly scale.

## 3.3 Overview of the proposed framework

The general STOIC framework for IDs combines two sub-models as shown in Figure 14. First, the inventory of pollutant loads determines the specific stormwater pollution potential

($\boldsymbol{SPP_p}$, kg.y$^{-1}$) resulting from urban nonpoint sources emissions ($\boldsymbol{E_{p,s}}$, kg.y$^{-1}$) in a year of rainfall within the catchment. The catchment is described in generic urban zones (requirement R4) with identified land use type and urban form. These zones are associated with levels of nonpoint sources. Second, in the case of combined UWS the raw wastewater load from households serviced by the UWS ($\boldsymbol{rWPP_p}$, kg.y$^{-1}$) is determined using an average wastewater composition and the extent of population serviced. Third, complying with requirements R1, R2 and R5, the distribution of water flows ($\boldsymbol{dQ}$, m$^3$.y$^{-1}$) carrying the suspended solid load ($\boldsymbol{dSS}$, kgSS.y$^{-1}$) are modelled through the UWS during one year rainfall time series at a high temporal resolution (10-min intervals) using an integrated hydrology/hydraulics model. The resulting spatiotemporally differentiated LCI of IDs during a year of rainfall from the UWS ($DEP_p$, $CSO_p$, $DEP_p$, $WTP_p$, $SWR_p$, kg.y$^{-1}$) can then be used as inputs to a LCA of UWS to assess wet-weather management (requirement R3).

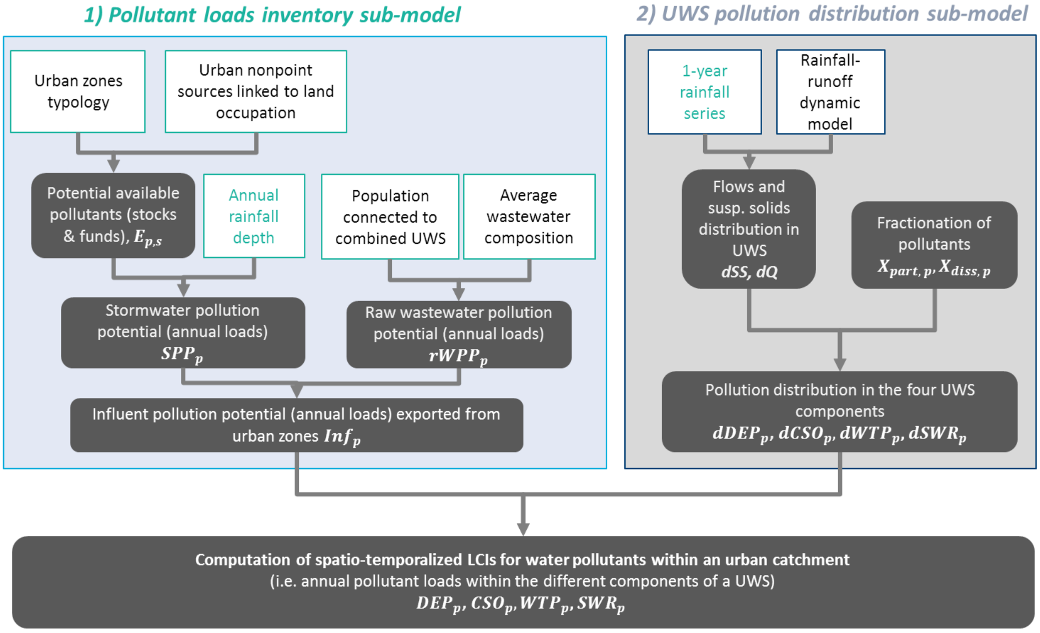


*Figure 14. STOIC framework based on two sub-models to build spatiotemporally differentiated LCIs for water pollutants within an urban catchment, from nonpoint source emissions to the routing and distribution of pollution in the four UWS components (e.g. accumulated deposits in the network (DEP), combined sewer overflow discharges (CSO), discharges of treated effluents from WWTPs (WTP), untreated stormwater discharges (SWR)). Legend: dSS, total suspended solids distribution; dQ, flows distribution; $X_{part,p}$, particulate fraction of pollutant p; $X_{diss,p}$, dissolved fraction of pollutant p; $dDEP_p$, distribution of pollution in accumulated deposits in network; $dCSO_p$, distribution of pollution in combined sewer overflow discharges; $dWTP_p$, distribution of pollution in discharges of treated effluents from WWTPs; $dSWR_p$, distribution of pollution in untreated stormwater discharges.*

### *3.3.1 Key stormwater pollutants*

Among stormwater pollutants, suspended solids, heavy metals and PAHs are widely considered as the major causes of contamination in receiving environments (Fletcher et al., 2013; Zoppou, 2001). The framework developed in this research focuses on pollutants found in stormwater contributing significantly to freshwater ecotoxicity: heavy metal species (e.g. copper and zinc) and polycyclic aromatic hydrocarbons (PAH) as highlighted in Chapter 2. These pollutants are the result of human activities (e.g. diffuse spills, airborne pollution, sediments from traffic or construction/demolition sites) in the urban catchment where they accumulate on impervious surfaces before wash-off and transport in stormwater. Given the numerous widespread and different sources this diffuse (or nonpoint) pollution is typically considered as a surface area related load, in $kg.m^{-2}$ (Hvitved-Jacobsen et al., 2010). Therefore, to determine pollution loads of generic urban areas the framework will require a proper description of urban areas. In the following we will not consider pollutant emissions from industrial areas or specific activities (e.g. mining, construction sites) however these particular nonpoint sources could be included in the framework when necessary.

### *3.3.2 Pollutant loads inventory sub-model*

In order to obtain pollutant loads inventories for the key stormwater pollutants, the main pathways linking urban nonpoint sources to these pollutant emissions need to be identified (section 3.3.2.1), and these nonpoint sources should be quantified in a generic manner for any urban catchment (section 3.3.2.2).

#### *3.3.2.1 Main pathways describing source-pollutant emissions*

Within urban catchments, an array of nonpoint sources are contributing to pollutant emissions and some researchers investigated in source-flux analyses (SFA) the relative weight of each source on the global urban flow of pollutants emitted by urban areas to storm water and/or to the receiving water bodies (Björklund, 2010; Chèvre et al., 2011; Davis et al., 2001; Petrucci et al., 2014). Principal sources identified are road traffic, building and infrastructure materials exposed to runoff (e.g. roof covers and gutters which have potentially leaching metallic elements due to corrosion), and atmospheric deposition. Therefore these principal nonpoint sources are included in the framework with up to three possible pathways describing source-pollutant emissions (Figure 15):

1. Local deposits: direct deposition on roads;
2. Local funds: leaching of corrosion products from metallic surfaces;
3. Atmospheric deposition;

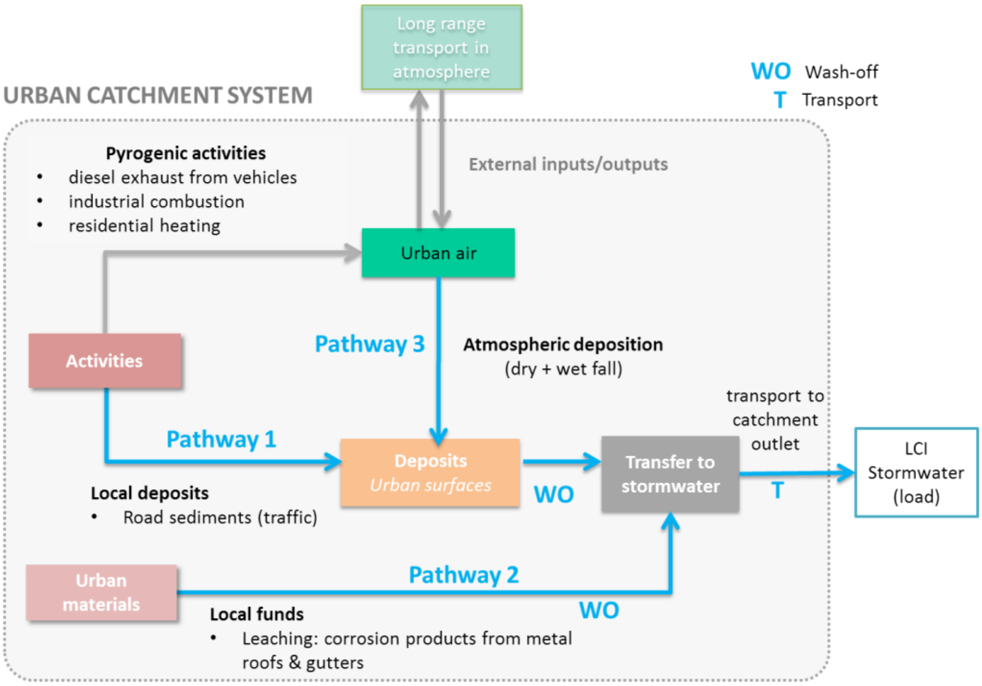


*Figure 15. Pathway modelling for stormwater pollutants generated by urban nonpoint sources (pathways considered within the STOIC framework in blue, pathways not included in grey)*

The characterization and quantification of these principal nonpoint sources are further detailed in Section 3.4.1.2.

#### *3.3.2.2 Urban catchment decomposition*

Ideally, the proposed framework should include a sufficient description of the urban catchment to estimate levels of nonpoint pollutant sources for the targeted pollutants (requirements R3 and R4). Hence, we propose a decomposition of the urban catchment into elementary surfaces including i) impervious road surfaces to describe road traffic, ii) building surfaces to specify roof area and roof cover materials, and iii) pervious surfaces which are not contributing to runoff generation.

To perform this urban catchment decomposition at a global scale, the Local Climate Zone (LCZ) framework was used (Stewart and Oke, 2012). The aim of this classification system is to associate objects/areas in the real world with an organized system of generic classes with quantitative attributes (site metadata), since it is impossible to give a universal definition of the term "urban" for its physical structure, its surface properties, or its thermal climate (Stewart and Oke, 2012). The LCZ framework classifies urban and semi-rural areas into ten "regions of uniform surface cover, structure, material, and human activity that span hundreds of meters to several kilometers in horizontal scale" (Stewart and Oke, 2012). The six common LCZs represented in European cities are described in Figure 16. Originally developed for the research of urban heat island effects, the LCZ framework is interesting in its universal applicability to characterize urban and rural areas which was demonstrated with 99 major cities across continents (8 in Africa, 18 in the Americas, 34 in Asia, 2 in Australasia and 37 in Europe). Indeed, the World Urban Database and Access Portal Tools (WUDAPT) initiative (Bechtel et al., 2015) aims to collect data on the form and function of cities around the world and disseminate it in response to the dearth of information on urban areas which was highlighted in the IPCC's 5th Assessment Report on impacts, adaptation and vulnerability of urban areas to climate change (Revi et al., 2014).

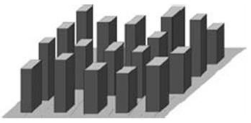
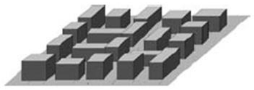
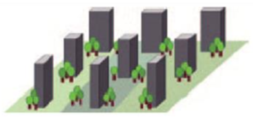
| LCZ 1, Compact high rise | LCZ 2, Compact mid rise | LCZ 3, Compact low rise |
|---|---|---|
|  |  |  |
| Dense mix of tall buildings<br>Land cover mostly paved<br>Core, periphery<br>Commercial, Residential | Dense mix of midrise buildings<br>Land cover mostly paved<br>Core, periphery<br>Residential, Commercial, Industrial | Dense mix of low-rise buildings<br>Land cover mostly paved<br>Old/densely populated cities, towns or villages, core, periphery<br>Residential, Commercial |
| LCZ 4, Open high rise | LCZ 5, Open mid rise | LCZ 6, Open low rise |
|  |  |  |
| Open arrangement of tall buildings<br>Lot of pervious land cover<br>Periphery. Densely populated cities,<br>Residential | Open arrangement of mid rise buildings<br>Lot of pervious land cover<br>Periphery<br>Residential, Institutional, Commercial | Open arrangement of low rise buildings<br>Lot of pervious land cover<br>Medium density cities, periphery (suburbs), commuter and rural towns |

*Figure 16. Abridged definitions for six common local climate zones (LCZs) represented in European cities including description of built types, location and function(s) (see Annex B-2, Table S11 for the full definitions including functions and surface property values of the ten LCZ defined in the LCZ framework (Stewart and Oke, 2012))*

#### *3.3.2.3 Flows inventory sub-model*

Urban water quality models such as rainfall-runoff models simulate surface/subsurface hydrology in urban catchments through a set of physical processes: precipitation, evaporation, infiltration losses on impervious surfaces then the hydraulic processes which describe the routing and storage of water flows and their associated pollutant loads in UWS to its outfalls in receiving waters. Hydraulic equations of unsteady flows in pipes and channels solve the conservation of mass and momentum with a fine temporal resolution (generally on the order of the minute) to maintain numerical stability. Therefore, we can use such urban water quality models to determine, for a rainfall time series, the distribution of pollutants in water flows routed to UWS sub-systems (e.g. WWTP, sewer network and CSO infrastructure if the UWS is combined). Usually, these water flows are described in terms of quantity (volume) and quality (pollutant concentrations and loads of the indicator pollutant, often on total suspended solids).

## 3.4 Modelling approach and practical implementation

In the following sections we describe the two model components and how they are integrated together to produce LCI of urban IDs.

### *3.4.1 Emissions inventory sub-model*

#### *3.4.1.1 Urban catchment characterization*

Urban land cover can be classified in impervious built-up (e.g. road surfaces and building surfaces) and pervious surfaces (e.g. parks, lawns, bare soil). The STOIC framework does not require detailed land-use land-cover data, but rather relies simply on an existing typology of landscape classes, for which average urban area ratios (e.g. building, road and impervious surfaces) are defined (Stewart and Oke, 2012)(See Annex B-2 Table S11).

#### *3.4.1.2 Pollutant emissions in urban catchment*

In this section we describe how pollutant processes are modelled from the generation of pollutants (i.e. source-emission pathways) and their subsequent wash-off during rainfall events. These processes refer to the three primary nonpoint sources identified in Section 3.3.2.1.

### *a. Pollutant generation from nonpoint sources*

Nonpoint sources are estimated in terms of potential annual stormwater pollutant loads discharged into the environment following a procedure adapted from SFA (Petrucci et al. 2014). For each pollutant $p$ and nonpoint source $s$, emissions $\boldsymbol{E_{p,s}}$ (kg.y$^{-1}$) are expressed as the product of a source activity factor, $a_s$ (variable parameter, whose value is to be determined for the catchment), and an emission factor for pollutant $p$ and primary activity $s$, $EF_{p,s}$ (fixed parameter, whose value is already defined and constant across catchments):

$$\boldsymbol{E_{p,s}} = a_s \cdot EF_{p,s} \qquad \text{(Eq.1)}$$

Then the annual stormwater pollution potential, $\boldsymbol{SPP_p}$ can be estimated as the sum of the emissions $\boldsymbol{E_{p,s}}$ from each source $s$:

$$\boldsymbol{SPP_p} = \sum_s E_{p,s} \qquad \text{(Eq.2)}$$

Each identified source is characterized for its variable activity level and chosen emission factor (EF) for the key pollutants identified in the following sections. For sources contributing to pollutant deposits (accumulating over time) such as atmospheric deposition and road traffic, the EF will be expressed in kg.m$^{-2}$. Nonpoint sources representing funds of pollutants stored in urban materials (e.g. metal roof covers) will have EF expressed per millimeter of rainfall and per unit area, g.(m².mm)$^{-1}$ or per unit length, g.(m.mm)$^{-1}$.

We propose to define, for each primary nonpoint source $s$, a specific emission factor ($\boldsymbol{EF_{p,s}}$) for each of the key stormwater pollutants (Cu, Zn, and the PAH class), using the estimation procedure with uncertainty quantification in Petrucci et al. (2014). The values of $EF_{p,s}$ (mean and standard deviation) for the three primary activities are summarized in Table 2. Further details including modelling hypotheses, data sources and references are given in Annex B-3 Table S12.

#### i. ***Emissions from road traffic***

Traffic is one of the primary sources of contamination in urban areas (Björklund, 2010; Markiewicz et al., 2017).

- Variable $a_s$

For traffic activity in the urban area, the selected activity parameter is determined by the daily traffic flow. Traffic flow in a traffic network is calculated by multiplying the number of vehicles on a given road or traffic network by the average length of their trips measured in kilometers during a unit time. It is possible to estimate traffic flow using measurements (e.g. traffic counts) over all the road sections during a day in the catchment and summed up to obtain a total (average) daily traffic in vehicle.km per day and per hectare (vkm.(d.ha)$^{-1}$). Spatial traffic activity and associated emissions inventories may also be a possible alternative, for example with neural network analysis (Fu et al., 2017).

*Other alternative if no available measurement of traffic activity*

Within a same land use (e.g. residential) the daily traffic activity of two suburban sub-catchments can vary importantly in relation with the urban form (individual or collective housing). For example, in France, Petrucci et al. (2014) using daily traffic measurements estimated a daily traffic volume of 1228 vkm.(d.ha)$^{-1}$ in Sucy-en-Brie, in the greater Paris suburb with individual housing while the daily traffic volume in the Pin Sec sub-catchment with collective housing, in Nantes was of 679 vkm.(d.ha)$^{-1}$. Consequently, we can hypothetize that urban form is a good predictor for traffic activity at a sub-catchment level. Moreover, population density has been shown to be a key aspect of urban form and this parameter was used in studies on the effects of traffic on human exposure via inhalation of motor vehicle emissions (Marshall et al., 2005), noise exposure (Salomons and Berghauser Pont, 2012) and atmospheric deposition of road-derived particulates (Davis and Birch, 2011). We propose to estimate local average traffic activity (in number of vehicle kilometers) through the power-relationship with population density proposed by Salomons and Berghauser Pont (2012), valid for European cities where there is higher traffic volume in the suburbs than in the downtown areas. Population density data can be found easily with land occupation databases. Then, the daily traffic per inhabitant, $W_{inh}$ is estimated with the population density, ρ and two variables to be adjusted to urban areas, $A$ and $\varepsilon$:

$$W_{inh} = A \cdot \rho^{\varepsilon}$$

- $\boldsymbol{EF_{p,Traffic}}$

There are two main pathways of pollutant generation from traffic activity, exhaust and non-exhaust emissions. Vehicle exhaust emissions result from fuel combustion and most of these particles tend to be retained in the air compartment, while a small fraction deposits on roads (Gunawardena et al., 2013; Hewitt and Rashed, 1990). Non-exhaust emissions are the result of the wear and tear of brakes, tires, and road materials, which deposit mostly on roads and will be mobilized by runoff. Exhaust emissions are already included in the average atmospheric deposition rate, hence we propose to characterize traffic activity with a global EF value accounting for non-exhaust emissions resulting from the processes above, based on specific EF values for each process (e.g. brake, tire and road wear) calculated in Petrucci et al. (2014).

#### ii. Metal corrosion

- Variable $a_s$

The activity parameter can be defined as the total area of metal cover and length of metal gutters. Hence the areas of zinc-emitting metal covers and lengths of metal gutters (copper and zinc) are estimated for buildings in the urban area. Set values for these elements are defined for different land uses (see Annex B-3, Table S12).

- $EF_{p,corrosion}$

When exposed to runoff, some land cover materials are known to emit significant metallic pollution due to corrosion (Charters et al., 2016; Petrucci et al., 2014; Robert-Sainte et al., 2009; Sellami-Kaaniche et al., 2014). Robert-Sainte (2010) demonstrated a linear relationship between annual metallic emissions and rainfall height. We propose to classify metal roofing materials contributing to zinc emissions using the typology defined by Petrucci et al. (2014) based on the potency of zinc emissions (low-emission materials such as aluminium, and high emission materials such as galvanized steel) and to use the EF values provided in this work.

#### iii. Atmospheric deposition

- Variable $a_s$

  For atmospheric deposition the selected parameter for the activity level is simply the total area of impervious surfaces.

- $EF_{p,atm.dep.}$

For atmospheric deposition, the EF value is an average daily deposition rate on all impervious surfaces, determined from studies of relatively unpolluted residential sites (e.g. without strong industrial influences). We assume that there is a uniform atmospheric deposition on the urban area (e.g. constant deposition rate on all impervious surfaces), hence no necessary differentiation between inclined roofs and asphalt or concrete roads.

Hence, to determine stormwater pollutant potential generated within the catchment in a year, it is necessary to determine the value of 6 parameters as shown in Table 2: total impervious area, area of high zinc-emitting roof covers, area of low zinc-emitting roof covers, copper and zinc gutter lengths and traffic volume.

*Table 2. Summary of the general estimation procedure of emissions per pollutant and per source.* $a_s$, *activity level per source,* $EF_{ps}$, *emission factor for source s and pollutant p; Cv, estimated coefficient of variation for* $EF_{ps}$.

| Source | Source activity factor | | Pollutant | Emission factor, pollutant p, source s | | |
|---|---|---|---|---|---|---|
| *s* | $a_s$ | *Unit* | *p* | $\boldsymbol{EF_{p,s}}$ | *Cv* | *Unit* |
| Atmospheric deposition | Total impervious area in catchment | [m²] | Cu | 8.68E-06 | 30% | $[g.(m^2.d)^{-1}]$ |
| | | | Zn | 2.41E-05 | 39% | $[g.(m^2.d)^{-1}]$ |
| | | | PAH | 6.28E-07 | 89% | $[g.(m^2.d)^{-1}]$ |
| Metal corrosion | High Zn-emitting roof covers | [m²] | Zn | 5.41E-03 | 41% | $[g.(m^2.mm)^{-1}]$ |
| | Low Zn-emitting roof covers | [m²] | Zn | 3.40E-05 | 90% | $[g.(m^2.mm)^{-1}]$ |
| | Copper gutters | [m] | Cu | 7.18E-04 | 42% | $[g.(m^2.mm)^{-1}]$ |
| | | | Zn | 4.70E-05 | 81% | $[g.(m.mm)^{-1}]$ |
| | Zinc gutters | [m] | Cu | 4.00E-06 | 128% | $[g.(m.mm)^{-1}]$ |
| | | | Zn | 1.05E-03 | 45% | $[g.(m.mm)^{-1}]$ |
| Traffic activity | Traffic volume (daily) | [vkm.d-1] | Cu | 6.03E-07 | 65% | $[kg.vkm^{-1}]$ |
| | | | Zn | 1.46E-06 | 31% | $[kg.vkm^{-1}]$ |
| | | | PAH | 8.12E-09 | 55% | $[kg.vkm^{-1}]$ |

### *iv. Characterization of the PAH load for toxicity modelling*

There are 16 priority PAHs identified by the US EPA, which are known to have different characterization factors in toxicity and ecotoxicity LCIA models (Henderson et al., 2011; Rosenbaum et al., 2008). Hence we proposed to further differentiate the PAH load (estimated as a sum of PAHs) using a decomposition in individual PAH compounds. It was demonstrated that the PAH profile varied in deposits depending on their origin. Higher molecular weight PAHs are mostly deposited locally in contrast to lighter PAHs which are more likely to be dispersed away in the atmosphere (Hewitt and Rashed, 1990; Ma et al., 2017). Therefore, the PAH profile for atmospheric deposits is based on statistical data obtained in Motelay-Massei et al. (2006); while the PAH profile for traffic-generated road dust uses collected data in Ma et al. (2017). These PAH profiles are described in Table 3.

*Table 3. Proposed PAH characterization for atmospheric deposits and traffic-generated road dust*

| | *PAH composition relative to total weight in deposits (%)* | |
|---|---|---|
| *Compound name* | *Atmospheric deposits* | *Traffic deposits (road dust)* |
| Naphthalene | NQ | 4.4% |
| Acenaphthene | 2.0% | 0.3% |
| Fluorene | 7.0% | 1.2% |
| Phenanthrene | 23% | 11% |
| Anthracene | 1.6% | 1.5% |
| Fluoranthene | 18% | 7.9% |
| Pyrene | 13% | 22% |
| Benzo[a]anthracene | 3.5% | 2.5% |
| Benzo[a]pyrene | 3.9% | 6.3% |
| Dibenz[a,h]anthracene | 0.5% | 2.7% |

### *b. General approach for the estimation of pollutant wash-off*

On impervious surfaces, accumulated deposits of pollutants from nonpoint sources are available for wash-off through mobilization by rainfall and subsequent entrainment in runoff (Figure 17). There are two main modelling approaches to predict pollutant loads effectively washed-off at the catchment scale: deterministic or statistical models. Hybrid models combine characteristics of both approaches to different degrees (Obropta and Kardos, 2007). In deterministic models, physical processes and catchment characteristics are described, which allow for simulation of scenarios. However these complex models require extensive datasets for their calibration and are difficult to verify and calibrate, so that even the most physically based models contain high levels of uncertainty. On the contrary, statistical models are based on empirical probabilistic relationships based on limited, measured data and do not describe physical processes or the catchment, hence scenarios cannot be built or evaluated (Bonhomme and Petrucci, 2017). A description of these model characteristics is given in Annex B-1.

In line with requirement R5, the objective of the pollutant loads inventory sub-model is to predict stormwater pollutant loads for a given catchment at the scale of a year. Hence, we investigated the suitability of using a deterministic model using dynamic accumulation and wash-off processes (SWMM, Rossman, 2015) to estimate the stormwater pollutant loads from nonpoint urban sources. The ability of deterministic accumulation-washoff models to replicate temporal variations of concentrations in stormwater loads was questioned recently (Al Ali et al., 2017; Sage et al., 2015; Shaw et al., 2010). Sage et al. (2015) stated that accounting for temporal variability in stormwater loads during rain events may not always be necessary to predict pollutant loads, as these loads are in fact mostly explained by stormwater volumes. Indeed, volume alone explained 85% of the variability in stormwater suspended solids loads in combined sewers (Joannis et al., 2015) and 69% in separate sewers (Shaw et al., 2010).

Therefore, we developed a simpler modelling approach also deterministic (which we call "average annual approach" in the following) in this Chapter to evaluate if dynamic accumulation-washoff models (using time-dependent build-up and washoff processes) are required to predict annual loads for a sub-catchment. Both approaches estimate the quantity of available pollutants on urban surfaces (after accumulation during dry-weather periods). The "average annual approach" assumes a complete transfer of the accumulated pollutants to stormwater (i.e. 100% wash-off). In the dynamic approach with deterministic models (such as SWMM), accumulation and wash-off coefficients must be determined. Table 4 compares the two approaches to estimate the total pollutant load transported in stormwater, and the modelled processes called in the two approaches are explained in Figure 17.

*Table 4. Comparison of model elements used in the two approaches for estimating pollutant loads at the catchment scale.*

| *Model elements* | ***Dynamic approach:*** *time-dependent load calculated from a 1-yr dynamic hydrograph* | ***Average annual approach:*** *load calculated from steady-state emissions* |
|---|---|---|
| *Hydrology* | Dynamic hydrograph using measured or synthetic rainfall time series for a one year period. Time steps: order of the minute | No hydrograph |
| *Pollutant generation on impervious surfaces* | Time-dependent build-up of pollutants, function of antecedent dry period. | Constant generation of pollutant each day during a one year period. |
| *Transfer to stormwater* | Time-dependent pollutant load washed-off: function of available stock and rainfall discharge rate. | 100% of available quantity of pollutant is washed-off (all pollutant generated is transferred to stormwater). |
| *Estimation of annual pollutant fluxes (kg.y-1)* | Dynamic equations solved with a hydraulic/hydrologic model (e.g. SWMM). | Sum of all estimated sources (steady-state). |

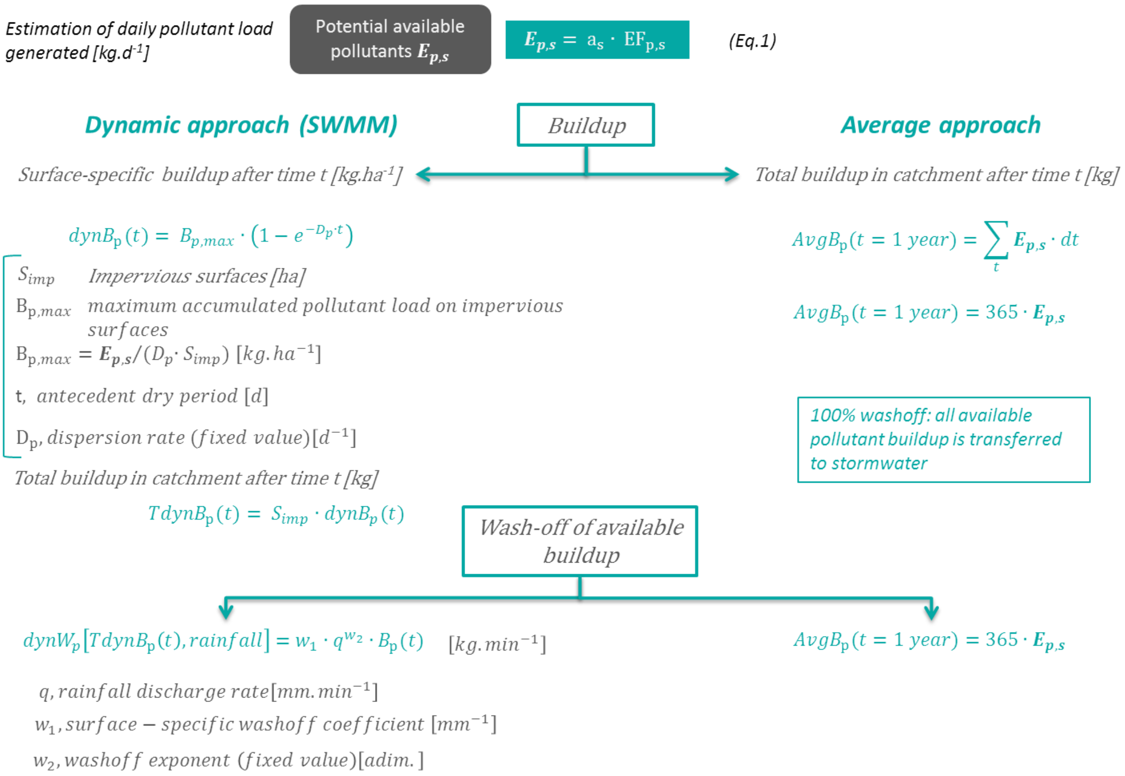


*Figure 17. From buildup to estimated pollutant load exported from the catchment after one year*

We compared these two approaches to estimate stormwater pollutant loads from nonpoint sources contributing to deposits in a French suburban catchment. Results showed that, using the same source characterization, the average approach predicted slightly larger stormwater loads (up to a factor 4) compared to the estimations from the dynamic model (for complete results refer to Annex 3-1). Since the estimated stormwater loads of accumulating pollutants did not show a significant sensitivity to the succession of wet and dry periods over the modelled year, we posit that the average annual approach is sufficient for a first order estimation of the annual pollutant loads from an urban catchment.

### 3.4.2 Water flows inventory sub-model

In the previous section, annual stormwater pollutant loads were determined to characterize the stormwater pollution potential of urban catchments SPP (Figure 14). The second sub-model in the STOIC framework determines the time-dependent routing of water flows (***dQ***, m$^3$.y$^{-1}$) and the distribution of the associated pollution described by suspended solids (***dSS***, kgSS.y$^{-1}$)

through the UWS during a one year rainfall time series at a high temporal resolution (10-min intervals) using an integrated hydrology/hydraulics model.

#### *3.4.2.1 Urban stormwater and combined sewer model*

We chose the SWMM (v.5.1) software (Rossman, 2015) to perform the urban hydrology simulations of water flows in the UWS. Continuously developed by the US EPA since 1969, the original SWMM was designed for the evaluation of combined sewer overflows (CSOs). Now, SWMM is a freeware program with a dedicated support community of expert users which is easily available to small municipalities, companies and universities. We considered this software also because its adoption has become widespread outside the U.S. with between 100 and 1000 model applications in cities world-wide, which means that there are calibrated UWS models running and updated. However, another hydrology-hydraulics model could be used in place of SWMM to describe temporal dynamics in UWS. The STOIC framework is generic enough to adapt to any available urban water quality models calibrated at catchment scale.

The required inputs for the SWMM model include (i) the UWS network (conduits, nodes and storage elements), (ii) a basic description of the sub-catchments drained by the UWS network (impervious surfaces areas, infiltration method), (iii) a calibrated water quality model to describe pollutant dynamics such as sedimentation and resuspension in the UWS and (iv) a rainfall time series with at least 10-minutes time intervals to ensure numerical stability of the computed flows. The SWMM model computes pollutant flows (e.g. SS) with concentrations and volumes for every time interval and yields four outputs or destinations as described in Figure 14:

- $DEP_p$: pollutants trapped in deposits in the UWS network (conduits and storage tanks);
- $CSO_p$: exported pollutants in flows to the receiving waters (CSO discharges occurring in combined UWS);
- $WTP_p$: exported pollutants in flows returned to the WWTP for treatment;
- $SWR_p$: exported pollutants in stormwater flows (for separate UWS).

#### 3.4.2.2 Using SWMM outputs to include other pollutants in the wet-weather LCI

In urban areas, suspended solids (SS) are important vectors of anthropic pollutants such as metals and PAHs since these substances are adsorbed on the surfaces of particles, and predominantly found in this phase (Angerville, 2009; Bressy et al., 2012; Gasperi et al., 2014; Zgheib et al., 2011). Moreover, SS concentrations can be estimated using real-time measures of turbidity as a proxy. Hence, the majority of urban water quality models such as SWMM use SS as primary indicators for sediment transport during stormwater events (Dotto et al., 2012; Hong et al., 2016). Therefore, we propose to include other relevant pollutants using information on their binding affinity to particles.

*Pollutant fractionation in influent wastewater*

Primary pollutants (e.g. organic matter described by chemical oxygen demand (COD), ammonium ($NH_4$-N), organic nitrogen (Org-N), phosphate ($PO_4$-P) and organic phosphorus (Org-P)) and PAHs were fractionated between dissolved ($\boldsymbol{X_{diss,p}}$) and particulate states ($\boldsymbol{X_{part,p}}$), while metals were fractionated between truly dissolved, colloidal and particulate states (see Table 5). Hence copper and zinc were estimated to be predominantly in particulate fractions with truly dissolved fractions in influent estimated at 10% (Hargreaves et al., 2017). Regarding PAHs with a negligible dissolved fraction in influent, these compounds were estimated to be completely in the particulate form (Liu et al., 2016).

*Table 5. Proposed fractionation (% of total pollutant mass) for pollutants according to their binding affinity to particles (SS). Pollutants marked with an asterisk (*) are not characterized in some LCIA models (e.g. ReCiPe 2016) but are still considered in the STOIC framework to enable their inclusion.*

| Pollutant | Dissolved fraction ($\boldsymbol{X_{diss,p}}$) in influent | Particulate fraction ($\boldsymbol{X_{part,p}}$) in influent | References |
|---|---|---|---|
| COD (*) | 0.4 | 0.6 | (Hvitved-Jacobsen et al., 2010) |
| $NH_4$-N (*) | 0.1 | 0.9 | (Hvitved-Jacobsen et al., 2010) |
| Org-N (*) | 0.3 | 0.7 | (Hvitved-Jacobsen et al., 2010) |
| $PO_4$-P | 1 | 0 | (Hvitved-Jacobsen et al., 2010) |
| Org-P | 0.3 | 0.7 | (Hvitved-Jacobsen et al., 2010) |
| Cu | 0.1 | 0.9 | (Hargreaves et al., 2017) |
| Zn | 0.1 | 0.9 | (Hargreaves et al., 2017) |
| PAH | 0 | 1 | (Liu et al., 2016) |

Following the principle of mass conservation, SS inputs and outputs are balanced in the UWS. The urban water quality model (e.g. SWMM) specifies the SS distribution (***dSS***) among the four possible destination flows in the UWS (Figure 14): return flows to WTP (SS_WTP), deposits (SS_DEP), CSO discharges (SS_CSO) and stormwater discharges (SS_SWR).

Regarding water volumes, flows are distributed (***dQ***) between three destination flows: return flows to WWTP (Q_WTP), CSO discharges (Q_CSO), and stormwater discharges (Q_SWR) assuming the water contents in deposits is negligible.

#### 3.4.2.3 LCI wastewater loads from domestic sources

In combined sewers, the composition of the combined influent is determined by a mix of untreated stormwater and wastewater from domestic sources. The stormwater pollution potential was estimated in Section 3.4.1.

The wastewater load generated by an average citizen ($\boldsymbol{rWPP_p}$) can be characterized using the person-equivalent (PE) measure defined for the European context (91/271/EEC directive). This load corresponds to a five-day biochemical oxygen demand ($BOD_5$) of 60 g.$(PE.d)^{-1}$. In the following, Table 6 extends the definition of this average untreated domestic wastewater load (in g.$(PE.d)^{-1}$) with conventional wastewater pollutants based on the French person-equivalent (Arrêté du 9 Décembre 2004: total Kjeldahl nitrogen of 15g.$(PE.d)^{-1}$; total phosphorus of 2 to 4 g.$(PE.d)^{-1}$) and literature data concerning French households (Deronzier et al., 2001; Gasperi et al., 2014; Mercoiret, 2010; Stricker and Héduit, 2010). The total wastewater load is estimated from the number of connected PE given by the wastewater operator. Treated effluent quality is determined using a mean removal efficiency (RE) in WWTP of 70% as PAHs and metals (Cu and Zn) are efficiently removed in most secondary treatment processes (Choubert et al., 2011).

*Table 6. Untreated domestic wastewater influent composition for an average French citizen ($\boldsymbol{rWPP_p}$) in g.$d^{-1}$ per capita*

| Wastewater pollutants | g.$(PE.d)^{-1}$ |
|---|---|
| COD, total (*) | 90 |
| BOD, total (*) | 60 |
| $NH_4$-N (*) | 11 |
| Org-N | 3.8 |
| Kjeldahl-N | 15 |
| $NO_3$-N | 0 |
| $NO_2$-N | 0 |
| TN (total N) | 15 |
| $PO_4$-P | 1.2 |
| Org-P | 0.8 |
| TP (total P) | 2 |
| Cu | 1.6E-03 |
| Zn | 3.4E-02 |

Finally, for the STOIC framework to yield spatiotemporally differentiated LCI of UWS discharges for pollutants p ($kg.y^{-1}$), the required model parameters (which are described in Figure 14) are synthesized in Table 7.

*Table 7. Synthesis of model parameters and estimation procedure leading to spatiotemporally differentiated LCI of UWS discharges for pollutants p*

| *Model parameter* | *Description* | *Unit* |
|---|---|---|
| | ***Pollutant loads inventory sub-model, for pollutants p*** | |
| ***$E_{p,s}$*** | Potential available pollutants (stocks & funds) | $kg.y^{-1}$ |
| ***$SPP_p$*** | Stormwater pollution potential (annual load) from all sources s | $kg.y^{-1}$ |
| ***$rWPP_p$*** | Raw wastewater pollution potential (annual load) | $kg.y^{-1}$ |
| ***$Inf_p$*** | Influent pollution potential (annual load) | $kg.y^{-1}$ |
| | ***UWS pollution distribution sub-model, for pollutants p*** | |
| ***dSS*** | Distribution of SS load (kgSS. $y^{-1}$) in UWS components<br>***dSS*** = SS_DEP + SS_CSO + SS_WTP + SS_SWR | (%) |
| ***dQ*** | Distribution of water flows (Q, $m^3.y^{-1}$) in UWS components<br>***dQ*** = Q_CSO + Q_WTP + Q_SWR | (%) |
| ***$X_{part,p}$ & $X_{diss,p}$*** | Fractionation between particulate and dissolved states for pollutants p in influent | (%) |
| ***$dDEP_p$*** | SS_DEP · $X_{part,p}$ (assuming only particulate fractions are deposited) | |
| ***$dCSO_p$*** | Q_CSO · $X_{diss,p}$ + SS_CSO · $X_{part,p}$ | |
| ***$dWTP_p$*** | Q_WTP · $X_{diss,p}$ + SS_WTP · $X_{part,p}$ | |
| ***$dSWR_p$*** | Q_SWR · $X_{diss,p}$ + SS_SWR · $X_{part,p}$ | |
| ***$DEP_p$*** | Mass vector of pollutants in deposits (annual load)<br>***$DEP_p$*** = ($SPP_p$ + $rWPP_p$) · $dDEP_p$ | $kg.y^{-1}$ |
| ***$CSO_p$*** | Mass vector of pollutants in CSO discharges (annual load)<br>***CSO***$_p$ = ($SPP_p$ + $rWPP_p$) · dCSOp | $kg.y^{-1}$ |
| ***$WTP_p$*** | Mass vector of pollutants in treated WTP effluents (annual load). Removal Efficiency (RE) to specify for pollutants if different. Abatement during wastewater treatment applied is coefficient (1-RE)<br>***WTP***$_p$ = ($SPP_p$ + $rWPP_p$) · $dWTP_p$*(1-RE) | $kg.y^{-1}$ |
| ***$SWR_p$*** | Mass vector of pollutants in stormwater discharges (annual load)<br>***SWR***$_p$ = ($SPP_p$ + $rWPP_p$) · $dSWR_p$ | $kg.y^{-1}$ |

## 3.5 Discussion and conclusions

### 3.5.1 *Model limitations*

The interest of linking human activities, land use and wet-weather flows has been recognized. However, as urban catchments are very diverse there are other potential nonpoint sources which could have major pollutant loads and should be incorporated in the proposed framework when possible. In the proposed framework, we restricted to the three main pathways identified in the European context (cities with old downtowns and modern suburbs). Also, the framework focused on three key stormwater pollutants for which the main nonpoint source-emission pathways were identified in studies based on source flux analyses of real urban catchments (Chèvre et al., 2011; Petrucci et al., 2014). More pollutants could be easily incorporated in the model as principal source-emission pathways are better understood.

The STOIC framework aims at assessing UWS discharges from generic urban catchments, hence specific activities (e.g. industrial, construction/demolition activities and influencing external long-range sources) are not modelled. However these specific activities should be included where necessary, further refining the generic assessment with more site-specific characteristics. Concerning the modeled traffic activity for the generic urban catchments in this framework, the estimation is based on European cities with a decreasing traffic activity with population density. This estimation should be adapted to better reflect metropolitan traffic networks.

Deterministic urban water quality models are complex and difficult to calibrate with typically an extensive number of site-specific data required, and they usually simulate water quality with one or two indicators (often SS). The assumptions on the pollutant fractionation (particular or dissolved states) and the use of SS as a proxy to describe the routing of pollutants within the UWS may be discussed but are acceptable as a first order estimation to compute pollutant loads in IDs.

### 3.5.2 *Data availability and uncertainty*

Model input parameters and the availability of required data (e.g. data available with a global coverage) are presented in Table 8. Some parameters need to be determined and are site-specific, concerning (i) the urban catchment characterization (e.g. total catchment area, metal roof areas, and metal gutter length), (ii) meteorological conditions (e.g. atmospheric deposition, rainfall time series), (iii) a calibrated UWS model describing CSO infrastructures and pollutants dynamics inside the UWS (e.g. with indicator pollutants). These parameters are expected to be significantly variable and the influence of their variability on model outputs (e.g. spatio-temporally differentiated LCI of UWS discharges) should be investigated with a global sensitivity analysis.

In the catchment characterization, emissions from each primary urban nonpoint source are quantified using estimates of activity levels ($\boldsymbol{a_s}$) and their associated emission factors ($\boldsymbol{EF_{p,s}}$) following an estimation procedure. Petrucci et al. (2014) performed an uncertainty analysis for a French residential catchment in the Paris area, which demonstrated that uncertainties around the estimation of $\boldsymbol{a_s}$ generally showed lower uncertainties than for $\boldsymbol{EF_{p,s}}$, except for emissions linked to land cover elements (metal roofs and gutters). Indeed, the main issue for quantifying these land cover elements is the determination of $\boldsymbol{a_s}$. However, for now there are no developed methods for fast characterization of materials used in buildings and infrastructures which would account for small but frequent and highly emitting elements. Also, building materials are often determined by local traditions, regulations, and other specific conditions (Petrucci et al., 2014; Sellami-Kaaniche et al., 2014). As a consequence, it is a challenge to develop generic ratios or average values for urban typologies.

Within the urban catchment traffic volume shows spatial and temporal variability with busier areas experiencing congestion at peak hours through the week (intraday variability), or between work days and weekends. Other transport modes (e.g. buses, trucks or motorcycles) could be included in addition of the traffic estimation with passenger cars in order to increase the accuracy of traffic pollution estimates.

Concerning meteorological conditions, atmospheric deposition is strongly dependent on local conditions which require measurements or statistics, available for most catchments. Rainfall time series with a sufficiently detailed time-step for the rainfall-runoff simulations may be limited to some areas and from certain databases (e.g. Météo France®). However, synthetic rainfall time series can be used instead.

With regard to the UWS model since every urban area has different stormwater management objectives usually tailored to local climate conditions, it is expected that retention capacities of CSO infrastructures are highly variable from site to site. Water quality in deterministic rainfall-runoff models is described by one or several indicator pollutants. The pathways of the indicator pollutant(s) through the catchment and UWS model are usually calibrated using onsite measurements; hence they are also highly site-specific. A runoff model is a simplification of a physical process; therefore, it involves uncertainty to some degree. Rainfall-runoff model uncertainty can come from the observed data, natural uncertainties, parameter estimation, calibration, or model assumptions.

*Table 8. Model input parameters and associated data availability*

| Pollutant loads inventory | Model parameter | | Unit | Value (constant/variable) | Global coverage | Sources |
|---|---|---|---|---|---|---|
| Rainfall | Annual rainfall height | | $mm.y^{-1}$ | Variable (measured) | Yes | Rainfall data for location. Climate average data exist at global scale |
| Catchment description | Total catchment area | | ha | Variable (measured) | No | Catchment area of study to be specified |
| | Distribution of elementary urban areas (roofs, roads, green spaces) | | % | Variable (measured if available, or estimated) | Yes | Proposed range of ratios in LCZ typology |
| Activity level $a_s$ | Atmospheric deposition | Total impervious area in catchment | ha | Variable (measured if available, or estimated) | Yes | GIS, topographic land use maps |
| | Metal corrosion | High Zinc-emitting roof area | ha | Variable (measured if available, or estimated) | No | Classification of roof materials based on satellite images or previous studies in France (Lamprea and Ruban, 2008; Petrucci et al., 2014; Robert-Sainte et al., 2009; Sellami-Kaaniche et al., 2014; Thévenot et al., 2007) |
| | | Low Zinc-emitting roof area | ha | Variable (measured if available, or estimated) | No | |
| | | Copper gutters | m | Variable (measured if available, or estimated) | No | Gutter length estimated for building type in LCZ using a correlation with roof area (See Annex B-3 Table S12) |
| | | Zinc gutters | m | Variable (measured if available, or estimated) | No | |
| | Traffic activity | Traffic volume (daily) | $vkm.d^{-1}$ | Variable (measured if available, or estimated) | European context | Traffic counts available in some cities. Can be estimated for European cities using population density/traffic activity relationship (Salomons and Berghauser Pont, 2012) - for all LCZ classes excepted commercial/industrial zones for which traffic activity is estimated using the surrounding suburban zone activity. |

| Pollutant loads inventory | Model parameter | | Unit | Value (constant/variable) | Global coverage | Sources |
|---|---|---|---|---|---|---|
| Emission factor $EF_{p,s}$ | Atmospheric deposition | Average daily deposition rate on impervious surfaces | $g.(m^2.d)^{-1}$ | Variable (measured if available, or estimated) | No | Measurements in study sites in France (Bressy et al., 2012; Motelay-Massei et al., 2006; Omrani et al., 2017; Percot, 2012; Petrucci et al., 2014) |
| | Metal corrosion | Zn emissions estimated for metallic roofs | $g.(m^2.mm)^{-1}$ | Constant, calculated value | Yes | Proposed values found in Petrucci et al. (2014) are aggregated from a database on experimental corrosion loads measured in Paris (Robert-Sainte, 2010; Robert-Sainte et al., 2009) |
| | | Zn and Cu emissions for Zn and Cu gutters | $g.(m.mm)^{-1}$ | Constant, calculated value | Yes | Proposed values found in Petrucci et al. (2014) are aggregated from a database on experimental corrosion loads measured in Paris (Robert-Sainte, 2010; Robert-Sainte et al., 2009) |
| | Traffic activity | Road, brake and tyre wear estimated for a passenger car | $g.vkm^{-1}$ | Constant, calculated value | Yes | EF for Zn emission estimated in Petrucci et al. (2014) is the same as in ecoinvent v3.4. However EF proposed in ecoinvent v3.4 process "Transport, passenger car, large size, diesel, EURO 3 –GLO" are lower by 1 order of magnitude for Cu and PAH emissions. Choice was made to stay with the most conservative values of Petrucci et al. (2014). Ecoinvent processes' emissions are based on values from Ntziachristos and Boulter (2009) |

| Flows inventory | Model parameter | Unit | Value (constant/variable) | Global coverage | Sources |
|---|---|---|---|---|---|
| Rainfall | 1-year rainfall time series (10-min intervals to ensure numerical stability in rainfall-runoff simulations in SWMM). | mm.min$^{-1}$ | Variable (measured) | No | Detailed rainfall time series can be found in certain databases but not always publicly available. Synthetic rainfall time series can be used instead. |
| Catchment model | Catchment area | ha | Variable (measured) | No | Catchment area of study to be specified |
| | Fraction of impervious areas | % | Variable (measured if available, or estimated) | Yes | GIS, topographic land use maps |
| | Mannings roughness coefficients for surface runoff for pervious and impervious areas | adim. | Constant, calculated value | Yes | Typical Mannings ranges of values are proposed in SWMM User manual (Rossman, 2015) |
| | Depth of depression storage for pervious and impervious areas | mm | Constant, calculated value | Yes | Default values proposed in SWMM User manual (Rossman, 2015) |
| | Average surface slope | % | Variable (measured) | Yes | Average catchment slopes can be calculated by digital elevation models. |
| UWS model | Retention capacity of CSO infrastructures | m$^3$ | Variable (measured) | No | Installed retention capacity is strongly dependent on local climatic conditions, existing sewer networks and management strategies of cities. |
| | Indicator pollutant (some water quality models use TSS, others use COD, BOD, total phosphorus etc) | mg.L$^{-1}$ | Variable | No | Water quality models that describe pollutant dynamics within the UWS are developed for each UWS application hence their site-specificity. |

### *3.5.3 Conclusions*

Results obtained in the present Chapter suggests that (i) the average modelling approach is sufficient for the defined objective (e.g. predicting annual stormwater loads); (ii) it is possible to characterize stormwater in a site-dependent manner (e.g. different loads estimated for compact high rise and open low rise land uses); and (iii) for three pollutants (e.g. Cu, Zn and the PAH class), the major processes of generation can be identified.

In conclusion, although the variability and complexity of the rainfall–runoff response in urban areas remains an area of active research (Fletcher et al., 2013), the STOIC framework provides a method to estimate spatiotemporally differentiated LCIs of UWS discharges from an urban catchment at a sufficient temporal scale to describe climatic variations in typical storm events. Therefore, impacts arising from these intermittent discharges to receiving waters can be assessed using a classical or a dynamic LCIA framework.

Finally, the application of the STOIC framework to a real case study would serve as a proof of concept to demonstrate its feasibility, support the discussion of its implementation and operational results.

# Chapter 4.: Life-cycle perspectives for the management of intermittent discharges in urban wastewater systems: lessons learnt from a French suburban catchment

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Proposed Authors for further journal submission:

Risch, E., Roux, P., Loiseau, E., Maruejouls, T., Sinfort, C.  (Binet, G. : acknowledgements)

This chapter will be submitted for publication in a peer-reviewed, international, scientific journal as the result of fruitful collaborations with urban systems modelling specialists of LEESU and SUEZ LyRE who are pioneers in the development of measurement systems and modelling tools for pollution-related knowledge on wet-weather discharges.

## *Contents of Chapter 4*

*The previous Chapter presented the STOrmwater Inventory Conceptualization (STOIC) framework. This framework develops spatiotemporally differentiated LCI of intermittent discharges from the UWS, which can be used as inputs to a classical LCA to assess wet-weather management strategies. In this Chapter, the STOIC framework is applied to a real case study to evaluate and discuss its applicability. Wet-weather control strategies are compared in order to identify pollution transfers in a life cycle perspective. Finally, elements concerning the LCI and LCIA phases are discussed in relation to the relevance of LCA results. In conclusion, the framework applicability is demonstrated with spatiotemporally differentiated LCI for intermittent discharges. Further work is required to streamline the practical implementation of the STOIC framework to a variety of catchments.*

## 4.1 Introduction

Global stressors, such as population growth, climate change, increasing urbanization, excessive nutrient inputs into surface waters, and water stress place additional pressure on water and wastewater utilities to provide adequate water and sanitation in an energy efficient manner, while protecting human health and the environment (Zimmerman et al., 2008). The ongoing adaptation and development of collection and treatment of wastewater requires an advanced asset management of the whole urban wastewater system (UWS) to meet a growing number of environmental management challenges, including increasing energy demands, greater awareness of greenhouse gases emissions and concerns over contaminants in biosolids and urban discharges.In many older cities, combined UWS are facing increased discharges from the combined sewer overflows (CSO) into receiving waters and overloads of the wastewater treatment plants (WWTPs) downstream due to climate change and increased frequencies of extreme flows. In a combined UWS, although CSO discharges contribute marginally to the total annual water discharge (CSO plus treated plant effluent discharges) from the WWTP, CSO discharges have been shown to contribute up to 90% of the annual load for hormones and micropollutants with high (>90%) wastewater treatment removal efficiency, which can lead to potentially problematic acute concentrations in receiving waters during storm events (Phillips et al., 2012; Weyrauch et al., 2010).

The direct solution to these problems is to increase the capacity of the combined sewer network, which can be done (i) by increasing the dimensions of the pipes; (ii) by adding storage capacity (e.g. retention tanks); (iii) by implementing real time control and weather radars to improve predicted runoff flow pattern and store water in selected parts of the network. Another possible strategy is a complete renovation by constructing a separate sewer system (Hvitved-Jacobsen et al., 2010). The augmented storage capacity with retention tanks (RT) to reduce CSO discharges to receiving waters often incurs high installation costs (Maruejouls, 2012). Moreover, increasing the retention capacity of the UWS with retention tanks may not have an overall positive environmental impact on receiving waters. Lindholm (1985) suggested that there may be a potential degradation in the water quality of WWTP effluents over a long period because retention tanks increase hydraulic and pollutant loads in the WWTP. It is then necessary to use a whole-systems perspective to identify environmental trade-offs across a broad range of environmental issues.

Since the mid-1990s, the life cycle assessment (LCA) method has proven its worth in the evaluation of the environmental sustainability of water and wastewater systems by using a whole-systems approach over their entire life cycle, and by addressing all relevant types of environmental impacts from global to local. For example, the LCA of a large scale, integrated urban water supply and wastewater system showed that wastewater management plays a significant role in its overall environmental burden even when that system is based on very energy intensive water supply technologies (Lane et al., 2015). Some LCA studies have evaluated the environmental implications of low-impact development strategies with green infrastructures such as bio-retention (Byrne et al., 2017a; Flynn and Traver, 2013; O'Sullivan et al., 2015), green roofs (Devkota et al., 2015; Morales-Pinzón et al., 2015; Wang and Zimmerman, 2015), and rainwater harvesting (Cubi et al., 2016; Kosareo and Ries, 2007; Petit-Boix et al., 2018; Saiz et al., 2006). Most LCA research on stormwater technologies focused on the water-management aspect (stormwater runoff reduction in volume) in specific stormwater control measures (Petit-Boix et al., 2017, 2015; Spatari et al., 2011), or in combined sewer overflow (CSO) control strategies (De Sousa et al., 2012). In the context of climate change adaptation, Brudler et al. (2016) compared stormwater management strategies on the primary function of ensuring flood safety, with constant water quality parameters (under the assumption that stormwater is sufficiently cleaned by either infiltration or treatment in a traditional WWTP).

However, only one study compared whole UWS strategies (e.g. green/gray stormwater infrastructure expansion alternatives against a baseline combined sewer system) to reduce CSO discharges to explore environmental trade-offs in the life cycle of UWS between local water quality gains and incremental energy and material costs (Wang et al., 2013). In the latter, water quality gains were determined solely with the freshwater eutrophication potential (kg P-equivalents) of urban discharges, and no other pollutants were included which understates the toxic impacts of many urban pollutants that are routinely discharged. Indeed Chapter 2 identified some priority compounds causing significant freshwater ecotoxicity impacts in intermittent urban discharges at the yearly and the event scale. Despite numerous LCAs on stormwater, these studies focused solely on the water quantity aspect or poorly accounted for water quality in urban discharges. To date, no studies captured sufficient information on the quality and quantity of urban discharges to accurately pinpoint environmental trade-offs in the life cycle of UWS across the broad range of impacts considered within the LCA framework.

The current study explicitly addresses this challenge for a set of wet-weather control strategies in a UWS and enables simultaneous consideration of local (e.g. water quality of discharges) and global impacts (e.g. materials and energy use), to help decision makers balance impacts that directly affect their site-specific human and environmental health with impacts spread over much larger spatial and temporal scales (e.g. greenhouse gas emissions). Previously, the STOIC framework was developed to quantify spatiotemporally differentiated LCIs for pollutant loads in intermittent discharges (IDs) from an urban catchment over a year of rainfall (Chapter 3).

The goals of this Chapter are to (i) quantify the environmental burden of a sub-catchment-scale UWS in a real world case study, where the developed ID loads will be used in the inventory step, and combined to life cycle impact assessment models; (ii) compare global performances of control strategies for IDs in terms of environmental impacts; and (iii) discuss the STOIC framework applicability in LCA of UWS at sub-catchment-scale and accounting for climatic conditions in the context of a real-world case study, in Bordeaux, southwest France.

## 4.2 Material and methods

In this section we describe the steps leading to the extended environmental assessment control strategies for IDs in UWS, by integrating the temporal variability of UWS discharges from an urban sub-catchment. We will follow the recommended scheme of a standardized LCA (ISO, 2006b, 2006a): (i) goal and scope definition (Section 2.2), (ii) inventory analysis (Section 2.3), (iii) impact assessment (Section 2.4), and (iv) interpretation of results (Section 3).

### *4.2.1 Case study description*

The Bordeaux metropolis is located on a 56,000 km2 watershed drained by the Garonne river (south west France) before it reaches the Gironde estuary in the Bay of Biscay, where the climate is oceanic with an average annual rainfall of 820 mm. The mean annual discharge of the Garonne river is 600 $m^3.s^{-1}$, ranging between extreme daily values of 54 and 4720 $m^3.s^{-1}$. The UWS of the metropolis drains an urban surface area of 578 km² and serves a population estimated at 749,595 inhabitants in 2015 with two main WWTPs (Clos de Hilde and Louis Fargue) which continuously discharge treated wastewater in the Garonne river (Lajaunie-Salla et al., 2018). In conclusion of a research initiative in 2014 to reduce its environmental footprint the Bordeaux metropolis drafted a global action plan to restore good biogeochemical water quality in the Garonne river with source reduction strategy for priority pollutants and real-time monitoring of urban flows in the UWS (Polard et al., 2016).

With the deployment of a continuous, real-time monitoring tool on the UWS and its outfalls in the Bordeaux metropolis urban area, the local wastewater authority (SUEZ France) identified one catchment in particular, the Louis Fargue catchment (77 km²) notable for its significant annual CSO discharges through six CSO outfalls. Its UWS is mostly combined (80%) with a network of 1391 km, collecting wastewater from 300,000 inhabitants for treatment (using biofiltration) in the Louis Fargue WWTP whose treatment capacity in wet-weather reaches 476,000 inhabitants. The framework application in the Louis Fargue catchment concerned one of its sub-catchments (Lauzun, 4.9 km²), with an entirely combined UWS collecting domestic wastewater of 9855 inhabitants through a 59 km sewer network. These sub-catchment characteristics are typical in a European context: the area is mainly residential and commercial-light industrial (e.g. warehouses) without any heavy industrial presence. The Lauzun UWS discharges excess flows during wet-weather events to a major CSO outfall on the Garonne river. The land occupation of this sub-catchment can be described as a mixed use development (residential, commercial and industrial uses).

## 4.2.2 Goal and scope definition

### 4.2.2.1 System boundaries

A combined UWS generally consists of three sub-systems including wastewater and stormwater transport network, infrastructures dedicated to storage and the wastewater treatment plant (WWTP). Within the catchment, urban flows are generated continuously from households (wastewater) and more sporadically from rainfall on impervious surfaces (stormwater runoff). In a combined UWS, CSOs are hydraulic structures performing flow separation to capture the first stormwater volumes at the beginning of a storm which may be more polluted. A CSO structure is basically a control chamber with a maximum height of water level, and a retention tank. During dry weather, the flow passes the control chamber and is sent to the WWTP for treatment. During wet weather, the rising level in the control chamber leads to an overflow over a weir to the retention tank. When the tank is full, the inlet of the tank is closed and the water level rises in the control chamber until it reaches the overflow pipe. All the exceeding flow is then routed to the river. Once the spilling has stopped and the conditions in the sewer network allow it, the storm flows stored in the tank are pumped back to the WWTP. Therefore a UWS will deal with continuous flows (e.g. treated effluents from the WWTP) and intermittent flows (e.g. stormwater and untreated CSO discharges)(Figure 18).

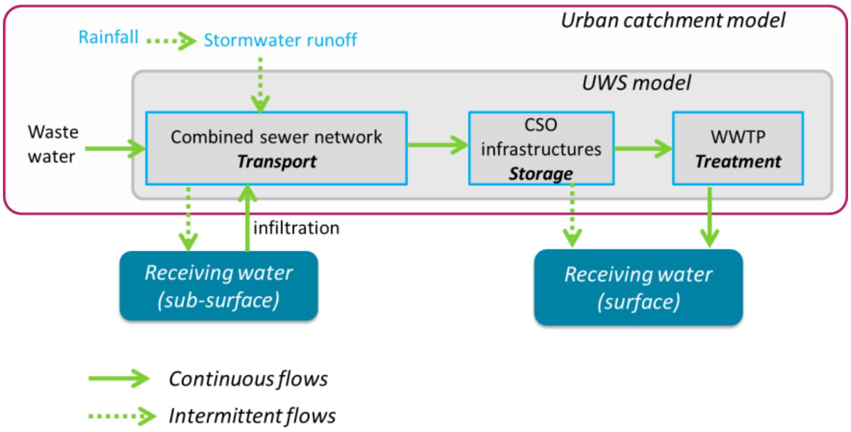

*Figure 18. System representation of the UWS model (adapted from Maruejouls 2012)*

Regarding the scope of this LCA study, all processes of the UWS components included in the perimeter are listed in Figure 19. Given that this study investigates a set of wet-weather control strategies in which some processes occur identically, it becomes possible to exclude these invariant processes from the comparison, based on the ceteris paribus assumption (e.g. the construction stages of the physical infrastructures of both the WWTP and sewer network). The treatment process in the WWTP was modelled using a three-stage conventional wastewater treatment (e.g. mechanical, biological and chemical treatment) which was found in the ecoinvent v3 life cycle databases of processes (Doka, 2009).

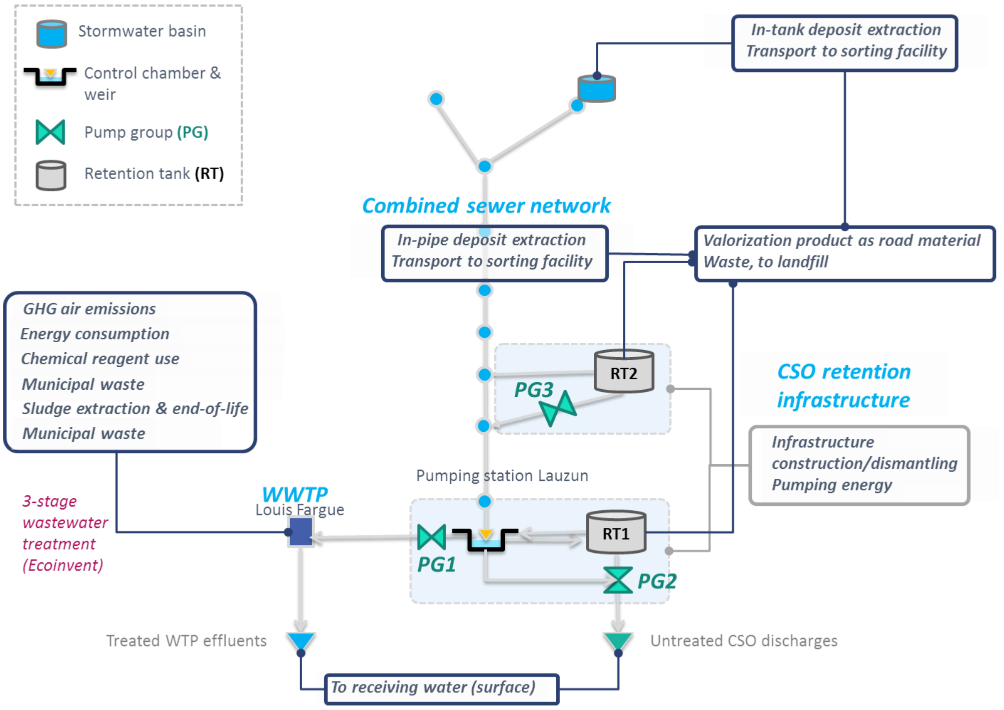

*Figure 19. System boundaries of the Lauzun UWS, showing main process flows through the life cycle stages (construction, operation and end-of-life) of the three UWS sub-systems.*

### 4.2.2.2 Functional unit definition

At the catchment scale, the combined UWS can perform several functions among which: reducing of CSO event numbers; ensuring smooth WWTP return flows during wet-weather conditions; enabling storage in network; reducing overall water pollution in discharges. Therefore a single functional unit (e.g. 1 $m^3$ of wastewater treated) does not capture all of the potential issues of UWS associated with its sub-systems. Applying the territorial LCA approach (Loiseau et al., 2013), we propose to solve the multi-functionality issue using as the reference flow, the studied urban area and UWS under the local climatic conditions. Different scenarios/strategies of wet-weather management of the UWS, for a one-year period, will then be compared on the basis of this reference flow.

### 4.2.2.3 Scenario definition

We investigated possible alternatives for the existing UWS, which included a (non-structural) change in the flow regulation of the overflow structure using a higher weir height, or a structural change with the addition of a new underground retention tank to increase the UWS capacity to store wet-weather flows before returning them to the WWTP for treatment when the storm peak flow has passed. Therefore, in order to meet the objective of this

Chapter, to assess and compare wet-weather control strategies for the UWS in the studied sub-catchment, four classes of scenarios were defined as follows (from least retention capacity to maximum) (Table 9):

i. Baseline: system without any retention capacity to represent the “do nothing” strategy, all wet-weather flows are discharged into the receiving water;
ii. Scenario A: existing system with current strategy (flow regulation in control chamber with weir height set at 0.8 m);
iii. Scenarios B (B0, B1): system with non-structural changes (e.g. with two higher weir heights);
iv. Scenarios C (C0, C1, C2): system with current strategy and structural changes (e.g. additional retention tank)

*Table 9. Scenarios of wet-weather control strategies and associated infrastructures*

| Scenario | Description | Flow regulation strategy in control chamber (weir height) | Pump group PG1 (kW) | Pump group PG2 (kW) | Pump group PG3 (kW) | Retention tank RT1 ($m^3$) | Retention tank RT2 ($m^3$) |
|---|---|---|---|---|---|---|---|
| Baseline | "Do nothing" strategy | 0.8 m | 40 | 400 + 1,200 | No | No | No |
| A | Existing system | 0.8 m | 40 | 400 + 1,200 | No | 980 | No |
| B0 | Non-structural change | 1 m | 40 | 400 + 1,200 | No | 980 | No |
| B1 | Non-structural change | 1.3 m | 40 | 400 + 1,200 | No | 980 | No |
| C0 | Structural change | 0.8 m | 40 | 400 + 1,200 | 40 | 980 | 4,000 |
| C1 | Structural change | 0.8 m | 40 | 400 + 1,200 | 40 | 980 | 10,000 |
| C2 | Structural change | 0.8 m | 40 | 400 + 1,200 | 40 | 980 | 20,000 |

### 4.2.3 LCI models

In this section we present (i) data used to characterize annual pollutant loads generated at the sub-catchment scale, then (ii) data describing the UWS infrastructure and the routing of pollutants within its sub-systems; and finally (iii) a summary of the key primary data sources to determine the life cycle inventories for a UWS during one year of rainfall under the local climatic conditions in Bordeaux. Operational data for the UWS model were compiled from a range of sources combining locally measured data with the best available sources of empirical and literature based information.

#### *4.2.3.1 UWS loads generated from the sub-catchment in a year of rainfall*

Chapter 3 described the framework to estimate annual pollutant loads in intermittent discharges (IDs) from a UWS in a given urban catchment under climatic conditions. This framework provides LCI data to characterize the following IDs for three classes of pollutants (copper, zinc and PAHs): (i) stormwater, estimated using nonpoint source-emissions pathways, and (ii) raw wastewater from the connected person-equivalents in the combined sewer network. CSO discharges result from the mix of the above-mentioned flows. The Lauzun sub-catchment is described using the urban zones typology presented in Chapter 3. Land occupation was determined with visual identification with Google Earth for land topography maps, and expert knowledge on the studied sub-catchment to quantify the surface areas. The climatic data is described in a local rainfall series for the year 2013, representative of the oceanic climate, and with measured rainfall height at every 10-minute intervals. The urban flows sub-model in the framework is based on a stormwater model simulating the quality and quantity of IDs using a full hydrodynamic model (e.g. SWMM software, Rossman, 2015). The water quality model predicts sediment transport and retention processes in the UWS and its sub-systems and is therefore based on the common indicator for particulate pollutants, total suspended solids (SS). In this study, a calibrated water quality model developed by the local wastewater operator of the Bordeaux metropolis (Maruejouls, 2012) was used. In the combined UWS, Pollutants originating from raw domestic wastewater and untreated stormwater are routed to three destination flows (e.g. CSO discharges, inflows returned to WWTP for treatment and deposits accumulated in the sewer and retention tanks). Simulated destination flows for one year of rainfall in the UWS are presented in Table 10.

*Table 10. Average mass repartition of pollutants in destination flows of the UWS for one year of rainfall using the SWMM engine calibrated on total suspended solids (SS). Detailed mass repartition of pollutants is provided in Annex C-2.*

| *Mass repartition of pollutants to destination flows (%)* | Baseline | A | B0 | B1 | C0 | C1 | C2 |
|---|---|---|---|---|---|---|---|
| As CSO discharges | 11% | 8% | 6% | 6% | 6% | 5% | 4% |
| As inflows to WWTP | 86% | 85% | 86% | 86% | 86% | 85% | 86% |
| As deposits in network | 4% | 8% | 8% | 8% | 9% | 10% | 11% |

#### *4.2.3.2 UWS infrastructure*

The UWS modelled in this study is based on the existing UWS infrastructure of the Lauzun sub-catchment. Data for the construction phase inventory of the physical CSO retention infrastructures (e.g. material production and manufacturing, transport to site, and construction) were estimated by scaling up the retention tank inventory of a sub-surface management system (Brudler et al., 2016). In the A B and C scenarios, retention tank RT1 was modelled with 980 $m^3$ and, in the C scenarios, the additional tanks RT2 were modelled with increasing storage capacities (4,000, 10,000 and 20,000 $m^3$) and an additional pumping group RT3 (Table 9). Pumping groups were modelled in terms of infrastructure materials and energy consumption to support water flow circulation in the UWS. The energy required for the water flow circulation between its sub-systems was estimated using the calibrated SWMM model of the Lauzun sub-catchment. Further details on the infrastructure materials and processes are provided in Annex C-1. Table 11 summarizes the inventory flows including the infrastructure, simulated operational inputs and outputs using the stormwater model SWMM for each of the modelled scenarios. Operational inputs and outputs are given for one year of rainfall.

*Table 11. Summary of inventory results for UWS scenarios for the management of one year of rainfall, combining infrastructure and simulated operational inputs/outputs using the SWMM engine. (*) See Table 9.*

| *Infrastructure* | Baseline | A | B0 | B1 | C0 | C1 | C2 |
|---|---|---|---|---|---|---|---|
| Retention tank ($m^3$) | None | 980 | 980 | 980 | 4,980 | 10,980 | 20,980 |
| Pumping groups (*) | 2 | 2 | 2 | 2 | 3 | 3 | 3 |

| *SWMM model results* | Baseline | *Variations from baseline (%)* | | | | | |
|---|---|---|---|---|---|---|---|
| Pump energy (kWh) | 1.72E+05 | +70% | +28% | +66% | +11% | +10% | +4% |
| Deposits to extract ($m^3$) | 2.19E+01 | +55% | +55% | +55% | +60% | +64% | +66% |
| CSO discharges ($m^3$) | 1.88E+05 | -1% | -18% | -32% | -33% | -45% | -102% |
| Inflows to WWTP ($m^3$) | 1.81E+06 | -1% | 1% | 1% | 2% | 2% | 3% |
| Total flows ($m^3$) | 2.00E+06 | -1% | -1% | -1% | -1% | -1% | -2% |

#### 4.2.3.3 Synthesis of inventory flows

Table 12 provides a summary of the key primary data sources to determine the life cycle inventories for the UWS model.

*Table 12. Model elements and information sources required to obtain LCIs for the UWS model (PE: person-equivalents)*

| Model element | Description | Data sources |
|---|---|---|
| *Sewer network (combined)* | Based on existing infrastructure. | Pipe network infrastructure not modelled. Operations data estimated using existing SWMM model. Solid deposits extracted from pipes by vacuum-tanker and sent to valorization (20%) and landfilling (80%). |
| *CSO retention infrastructure* | Based on existing infrastructure. | Infrastructure based on literature data for retention tanks and pump materials (Brudler et al., 2016). Operations data estimated using existing SWMM model. Solid deposits extracted from tanks by vacuum-tanker and sent to valorization (20%) and landfilling (80%). |
| *WWTP treatment* | Matched to the volume of wastewater flows pumped back for treatment. | Operations data based on a WWTP model (ecoinvent v3.4) with an average treatment capacity of 233,000 PE. Average abatement on wastewater pollutants (70%) was assumed to determine the treated effluent quality (Ruel et al., 2012; Thévenot et al., 2007) |
| *Domestic wastewater load* | Matched to the number of connected PE to the UWS. | Number of connected PE given by the wastewater operator. Average raw wastewater composition based on literature data for French households (Deronzier et al., 2001; Gasperi et al., 2014; Mercoiret, 2010; Stricker and Héduit, 2010). |
| *Climate* | Local rainfall series for 2013, representative of the oceanic climate. | Measured in a rain gauge: 1-year rainfall time series with 10-min intervals to ensure numerical stability in rainfall-runoff simulations in SWMM. |
| *Urban zones in the catchment* | Based on the studied sub-catchment. | Visual identification of the urban local climate zones (LCZ) with expert knowledge and Google Earth maps. Surface areas of the LCZ determined from existing catchment decomposition in SWMM. |
| *Urban nonpoint sources* | Matched to LCZ areas in sub-catchment. | Potential available pollutants generated within the catchments: estimated using the framework developed in Chapter 3. |

### 4.2.4 Impact assessment methods

This analysis followed both midpoint and endpoint approaches to Life Cycle Impact Assessment (LCIA). Impact assessment can be carried out at two levels in the cause-effect chain (i.e. environmental mechanism) of an impact category (Bare et al., 2000). First, midpoint indicators (e.g. ozone depletion, global warming, eutrophication etc.) determine a level of potential impact, showing the relative importance of emissions or extractions occurring due to human operations. Next, endpoint indicators are calculated to reflect differences between stressors further in the cause-effect chain, and are of direct societal concern (e.g. human health, ecosystem quality, resources).

The LCA was conducted using Simapro 8.5 software developed by Pré Consultants according to the ISO 14044 standard procedure (ISO, 2006b). The ReCiPe 2016 (v1.02) method was selected as the impact characterization model for its harmonised framework for the calculation of both midpoint and endpoint characterisation factors (M. J. Goedkoop et al., 2009). In ReCiPe 2016, the Hierarchist (H) perspective was selected with 100 year time horizon, which is based on scientific consensus with regard to the time frame and plausibility of impact mechanisms (Huijbregts, 2016). In order to compare and discuss impact assessment results from the toxicity and ecotoxicity model in ReCiPe 2016 (USES–LCA 2.0 model, (van Zelm et al., 2009)), the USEtox 2.01 model (Henderson et al., 2011; Rosenbaum et al., 2008) was also selected to perform a focused analysis of pollutants. Both models share the same substance database which include 3,073 organic chemicals and 20 (essential) metals, developed in the USEtox model (Rosenbaum et al., 2008).

In section 4.3.1, a general analysis of the UWS life cycle for a given scenario (scenario C2) provides an overview of the major contributors. This analysis is performed at two levels (i) first at midpoint level over the breadth of the 18 impact categories, then (ii) at endpoint level across all three areas of protection: human health, ecosystem quality and resources. Finally section 4.3.2 compares scenarios for ID management at endpoint level.

## 4.3 Results

### *4.3.1 Main contributors in a retention scenario (C2)*

Major contributors to the selected impact categories were identified for the C2 scenario with retention tanks for one year of rainfall, to assess global life-cycle contributions with a large retention infrastructure. Contribution analyses of the other scenario groups (e.g. baseline scenario, A and B scenarios) highlighted the same contributors, hence the full results are included in Annex C-3.

#### *4.3.1.1 Overall impact results*

System operations in the UWS subsystems (e.g. wastewater pumping system and the treatment of influent wastewater in WWTP), rather than retention infrastructure construction, are predominant in the majority of the life cycle impacts at midpoint level (Figure 20). We did not include WWTP and sewer network construction since the focus of this study was to investigate the effects of control strategies to manage IDs in an existing UWS. Overall, the wastewater treatment component is the major contributor on most impact categories except local impact categories concerned with water quality. Of the operational inputs required for the UWS operation, electricity and chemical reagents used in the WWTP (e.g. iron (III) chloride and aluminium sulfate) use are the major contributors to global warming (GWP), ozone depletion (OD), ionizing radiation (IR), tropospheric ozone formation (POF), acidification (Ac), terrestrial ecotoxicity (TET), human toxicity (carcinogenic) (HTc), land use (LU), mineral depletion (MD), fossil fuel depletion (FD), and water depletion (WD). The wastewater pumping system has notable impacts (45% of total) on ionizing radiation (IR) due to its electricity origin (French electricity mix) which is predominantly nuclear. Also, due to the background material inputs to manufacture pumping units, the pumping system has a clear impact on metal depletion (MD) due to copper, and on human toxicity, carcinogenic (HTc) due to cast iron. The retention infrastructure only has a significant contribution to human toxicity (carcinogenic) (HTc) resulting from the steel required in the construction phase. Pollutants in UWS discharges (CSO and treated effluents) make the majority contribution to eutrophication (F-Eu and M-Eu) and ecotoxicity (FET and MET), and their contribution on human toxicity (non-carcinogenic) (HTnc) is also marked (73% of total).

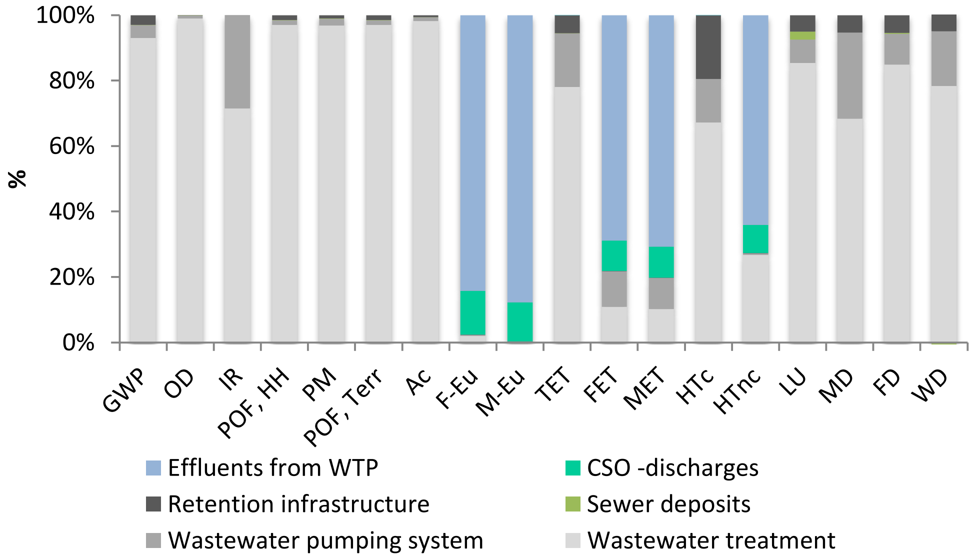

*Figure 20.Main contributors in UWS scenario C2 with retention tank for one year of rainfall (ReCiPe 2016 (Hierarchist) midpoint indicators). Global warming potential (GWP), Ozone depletion (OD), Ionizing radiation (IR), Photochemical ozone formation, human health, Freshwater ecotoxicity (FET), Marine ecotoxicity (MET), Human toxicity cancer (HTc), Human toxicity non cancer (HTnc), Land Use (LU), Metal Depletion (MD), Fossil Depletion (FD), Water Depletion (WD).*

The analysis of midpoint contributions to endpoints (Figure 21) reveals for the three areas of protection (AoP):

- AoP Human Health impacts are dominated by human toxicity (non-carcinogenic), global warming, and fine particulate matter formation.
- AoP Ecosystem quality impacts are led principally by freshwater eutrophication and to a lesser extent by global warming, terrestrial acidification, and ozone formation.
- AoP Resources impacts are driven by fossil depletion.

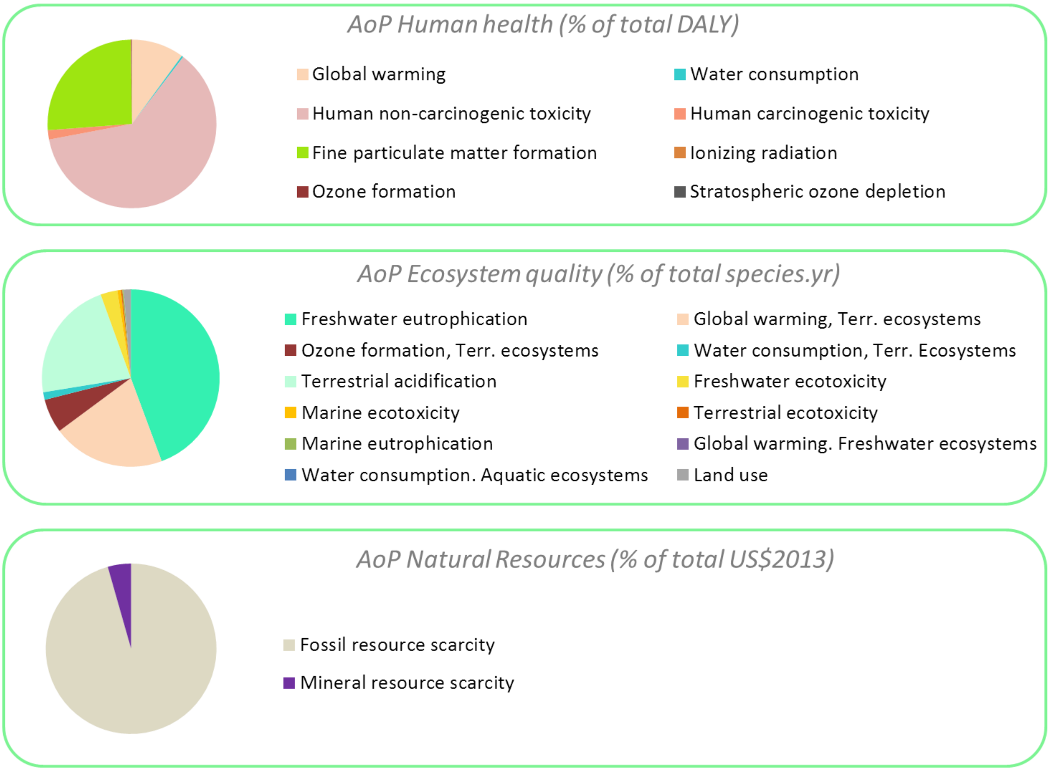

*Figure 21. UWS scenario C2 with retention tank for one year of rainfall: Contributions analysis of midpoints to endpoints - ReCiPe 2016 (Hierarchist) - human health (% of total DALY), ecosystem quality (species.yr) and resource scarcity (US$2013).*

Direct UWS discharges make manifest contributions to local impact categories concerned with water quality which include freshwater and marine eutrophication, freshwater and marine ecotoxicity and human toxicity (non carc.). Yet, the pollutants responsible of these impacts need to be identified in a focused analysis using pollutant characterization models. A first analysis on the above-mentioned categories was performed using the pollutant characterization models in ReCiPe 2016 (toxicity and ecotoxicity model based on the USES-LCA 2.0 model). Then, the toxicity and ecotoxicity impacts were analysed and compared, using the more elaborated USEtox 2.01 model. Indeed, toxicity models are an area of active research in the LCIA community and recent developments implemented in USEtox 2.01 have addressed metal-specific issues such as speciation, bioavailability and freshwater chemistry

(Diamond et al., 2010; Dong et al., 2014; Gandhi et al., 2010), as well as updated ecotoxicological effect data as described in Fantke et al. (2015).

#### 4.3.1.2 Local water quality impacts

Here, we focus on the local impact categories of eutrophication, ecotoxicity and human toxicity (non carc.) to highlight for each of these midpoint categories the most contributing pollutants. Results obtained with the ReCiPe 2016 characterization model indicate that phosphate and ammonia are respectively the main drivers of respectively freshwater eutrophication and marine eutrophication (Table 13). On freshwater ecotoxicity and human toxicity (non carc.), the ReCiPe 2016 model highlights zinc as the single most important toxic substance with a 98-100% contribution on total impacts (Table 14).

*Table 13. UWS scenario C2 with retention tank for one year of rainfall: Contributions of pollutants in UWS discharges to eutrophication impacts (ReCiPe 2016 (Hierarchist) midpoint indicators).*

| **Freshwater eutrophication** | | | **Marine eutrophication** | | |
|---|---|---|---|---|---|
| Substance | Impact score *kg P eq* | Relative contribution to impact % | Substance | Impact score *kg N eq* | Relative contribution to impact % |
| Phosphate | 1.46E+03 | 63% | Ammonia | 3.60E+03 | 75% |
| Phosphorus | 8.49E+02 | 37% | Nitrogen | 1.18E+03 | 25% |
| ***Total impact*** | ***2.31E+03*** | ***100%*** | ***Total impact*** | ***4.78E+03*** | ***100%*** |

*Table 14. UWS scenario C2 with retention tank for one year of rainfall: Contributions of pollutants in UWS discharges to ecotoxicity and toxicity impacts (ReCiPe 2016 (Hierarchist) midpoint indicators). Abbreviations for polycyclic aromatic hydrocarbons: B(a)A, benzo(a)anthracene; B(a)P, benzo(a)pyrene; D(a,h)A, dibenz(a,h)anthracene.*

| **Freshwater ecotoxicity** | | | **Human toxicity, non-cancer** | | |
|---|---|---|---|---|---|
| Substance | Impact score *kg 1,4-DCB* | Relative contribution to impact *%* | Substance | Impact score *kg 1,4-DCB* | Relative contribution to impact *%* |
| Zinc | 1.22E+05 | 98% | Zinc | 4.75E+06 | 100% |
| Copper | 2.04E+03 | 1.6% | Copper | 3.60E+01 | <0.1% |
| Pyrene | 8.55E+01 | <0.1% | Fluoranthene | 8.98E-01 | <0.1% |
| Fluoranthene | 2.68E+01 | <0.1% | Pyrene | 5.58E-01 | <0.1% |
| B(a)A | 1.52E+01 | <0.1% | Fluorene | 2.02E-02 | <0.1% |
| Phenanthrene | 3.26E+00 | <0.1% | Acenaphthene | 5.19E-03 | <0.1% |
| B(a)P | 1.84E+00 | <0.1% | Naphthalene | 4.07E-03 | <0.1% |
| Anthracene | 1.65E+00 | <0.1% | Anthracene | 3.38E-03 | <0.1% |
| Fluorene | 1.11E-01 | <0.1% | | | |
| Acenaphthene | 6.15E-02 | <0.1% | | | |
| D(a,h)A | 1.98E-02 | <0.1% | | | |
| Naphthalene | 9.24E-03 | <0.1% | | | |
| ***Total impact*** | ***1.24E+05*** | ***100%*** | ***Total impact*** | ***4.75E+06*** | ***100%*** |

Results obtained with the USEtox 2.01 model show slightly different outcomes on freshwater ecotoxicity, with copper leading the impact (62% of total), followed by zinc (38% of total) (Table 15). Regarding the human toxicity (non-carcinogenic) impacts, USEtox 2.01 yields similar results to those obtained with ReCiPe.

*Table 15. UWS scenario C2 with retention tank for one year of rainfall: Contributions of pollutants in UWS discharges to ecotoxicity and toxicity impacts (USEtox 2.01 model). Abbreviations for polycyclic aromatic hydrocarbons: B(a)A, benzo(a)anthracene; B(a)P, benzo(a)pyrene; D(a,h)A, dibenz(a,h)anthracene.*

| **Freshwater ecotoxicity** | | | **Human toxicity, non cancer** | | |
|---|---|---|---|---|---|
| Substance | Impact score *PAF.m3.day* | Relative contribution to impact *%* | Substance | Impact score *cases* | Relative contribution to impact *%* |
| Copper | 1.25E+08 | 62% | Zinc | 1.52E-01 | 100% |
| Zinc | 7.66E+07 | 38% | Copper | 1.72E-06 | <0.1% |
| Pyrene | 6.10E+04 | <0.1% | Acenaphthene | 1.09E-09 | <0.1% |
| B(a)A | 1.29E+04 | <0.1% | Fluorene | 5.13E-09 | <0.1% |
| Fluoranthene | 4.94E+03 | <0.1% | Fluoranthene | 1.87E-07 | <0.1% |
| Anthracene | 1.41E+03 | <0.1% | Pyrene | 1.57E-07 | <0.1% |
| Phenanthrene | 9.39E+02 | <0.1% | Naphthalene | 5.49E-10 | <0.1% |
| B(a)P | 2.32E+02 | <0.1% | Anthracene | 7.38E-10 | <0.1% |
| Fluorene | 5.59E+01 | <0.1% | | | |
| D(a,h)A | 2.14E+01 | <0.1% | | | |
| Acenaphthene | 1.89E+01 | <0.1% | | | |
| Naphthalene | 3.58E+00 | <0.1% | | | |
| ***Total impact*** | ***2.01E+08*** | ***100%*** | ***Total impact*** | ***1.52E-01*** | ***100%*** |

In all cases, the contributions of polycyclic aromatic hydrocarbons (PAH) are negligible on the toxicity and freshwater ecotoxicity categories. This contrasts with the research priorities of the water industry in recent decades, in line with the Water Framework Directive 2000/60/EC (WFD) requirements concerning the reduction of selected priority dangerous substances (which include three of these PAH). This difference indicates that improvements in the fidelity of LCIA toxicity models are still required, especially concerning the assessment of metal contribution to the toxic impacts, which may still be overestimated.

### *4.3.2 Scenario comparison*

First global results of the scenario comparison at the endpoint level (i.e. on the areas of protection) are presented in Section 4.3.2.1 to provide a general overview of the ranking order. Then, using a more restricted scope on local impact categories which are sensitive to water quality gains, Section 4.3.2.2 explores the differences between scenarios.

#### 4.3.2.1 On the areas of protection (AoP)

From a whole-system perspective, the comparison of UWS scenarios on the three AoP demonstrates that the scenarios similarly rank on two of them (human health and ecosystem quality) while the scenarios differ more markedly on the AoP Resources (Figure 22). Scenarios Baseline and A both lead equally on the ecosystem quality impact. Regarding the AoP Resources, scenario A closely followed by scenario B1 have the most important impacts due to their relatively high electricity consumption compared to the other alternatives.

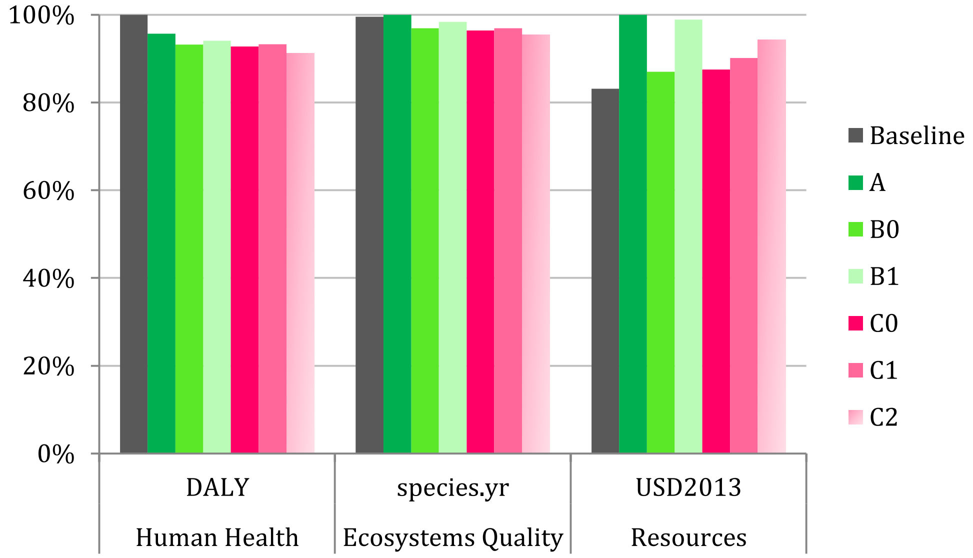

*Figure 22. Comparison of UWS management scenarios for one year of rainfall categories (ReCiPe 2016 (Hierarchist) endpoint indicators).*

#### 4.3.2.2 On local water quality impacts

The focused comparison on the local water quality impacts show that the baseline scenario has the most important contributions on all but two impacts (e.g. terrestrial ecotoxicity (TET) and human toxicity, carcinogenic (HTc)) (Figure 23). Indeed, in this scenario the water quality impacts are not mitigated by the retention infrastructures, hence not a great amount of stored deposits to extract and requires minimal electricity consumption (pumping). As demonstrated in Section 4.3.1.1, terrestrial ecotoxicity (TET) was led principally by electricity use, which is greatest in Scenario A; while human toxicity (carcinogenic) (HTc) was caused by both electricity use and material resources (e.g. copper and cast iron) required in the construction of the retention infrastructure. Results with another toxicity/ecotoxicity model (e.g. USEtox 2.01) are not expected to influence these comparative results.

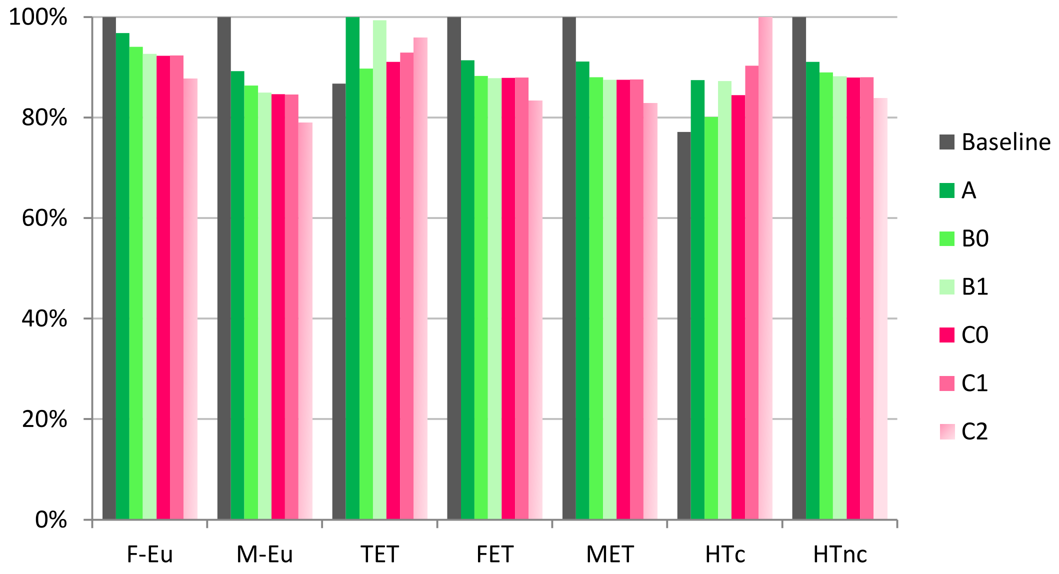

*Figure 23. Comparison of UWS management scenarios for one year of rainfall on local impact categories (ReCiPe 2016 (Hierarchist) midpoint indicators). Abbreviations used for impact categories: Freshwater eutrophication (F-Eu), Marine eutrophication (M-Eu), Terrestrial ecotoxicity (TET), Freshwater ecotoxicity (FET), Marine ecotoxicity (MET), Human toxicity cancer (HTc), Human toxicity non cancer (HTnc).*

## 4.4 Discussion

### *4.4.1 Reliability of LCI models*

#### *4.4.1.1 Wastewater treatment process*

Given the significant impacts of the operational inputs for the wastewater treatment process it is questionable whether the selected Ecoinvent model for wastewater treatment is sufficiently accurate and representative of both treatment technology and infrastructure used in the Louis Fargue WWTP. Indeed, the Ecoinvent model is based on the average Swiss technology and electricity mix for a large WWTP (treatment capacity of 233,000 Swiss person-equivalents, i.e. $4.e^{10}$ $L.y^{-1}$), which may substantially influence results. Results could be further refined using measured input and output data (e.g. electricity, chemicals use and destination of waste/by-products arising from the wastewater treatment). The treated effluent quality is estimated using an average treatment efficiency for PAHs and metal species (Cu and Zn) set at 70% which is expected for these substances in a three-stage wastewater treatment (Choubert et al., 2011). However, if additional substances with different removal efficiencies in a conventional WWTP are added, it would be necessary to define classes of substances according to their elimination rate in WWTP. For instance, a first class could include substances with high elimination rates in WWTP (e.g. Cu, Zn, PAH and wastewater pollutants such as estradiol and ibuprofen); while a second class could comprise substances with low elimination rates in WWTP (e.g. carbamazepine, ethylenediaminetetraacetic acid (EDTA)).

#### *4.4.1.2 Wastewater pumping system*

The wastewater pumping system contributes markedly to global impacts due to the electricity consumption for the pumped flows recirculation and the material inputs to manufacture the pumping groups (cast iron and copper). While the electricity consumption was simulated using the SWMM model (sub-model to assess the cost function of the sewer network developed internally by Suez), the material requirements of the pumping groups could be assessed more accurately based on the actual weights of the pumping groups instead of gross weight estimation from similar pumping groups.

#### *4.4.1.3 Solid deposits in the UWS*

The amount of SS is based on simulations results from the hydrodynamic model SWMM. Yet, data on operational inputs associated with the handling of deposits extracted from the UWS was scarce and warrants a further investigation. Hence, modelling hypotheses (e.g. extraction from the sewer network and retention tanks using a vacuum tanker, and end-of-life destinations) are formulated to give a gross estimate as a first step. For example, the end-of-life of solid deposits in the UWS is based on an average deposit composition, which does not account for the variations in heavy metal levels for example. Drawing a parallel with the bio-solids management (sludge spreading/incineration/landfilling) in wastewater treatment processes, the end-of-life of deposits could be refined by identifying several destination flows using a variable deposit composition. Because heavy metals are trapped in sediments, it is necessary to trace their fate: if they are sent to landfills, there are associated long-term impacts due to metal leaching.

#### *4.4.1.4 Urban catchment characterization*

The proposed framework is intended for the characterization of generic urban catchments and hence considers construction, polluted brownfield or industrial sites as special cases with specificities that need to be described on a case-by-case basis. Classification of land occupation in the studied catchment(s) into LCZ areas can be performed with visual identification based on high spatial resolution images (e.g. Google Earth®) for small catchments, but this interpretation of remote sensing images is time-consuming and prone to errors. Among the available tools for territorial analysis, the Urban Atlas resulting from Earth Observation (EO) Copernicus data is a particularly interesting reference for several reasons. First it provides an easily accessible description of the main urban land uses in major European cities. Second, its potential uses can be either at the local, national level or at the supranational level in order to compare European cities. The EU-funded SATURN project (SATellite applications for Urban Mobility, 2014-2015) delivered a demonstrator for the Bordeaux metropolis combining satellite imagery VHRS Pleiades (Very High Spatial Resolution) with the Urban Atlas with the objective to provide a set of indicators adapted to help urban planners and decision-makers in tackling major urban challenges. These indicators included among others, the urban climate zones based on the land use typology in the proposed framework (Stewart and Oke, 2012), and the impermeable surfaces rates (soil sealing surface rates per cadastral parcel) which can be of interest for flooding issues. The visual identification of land occupation (i.e. identified LCZs) in the Lauzun sub-catchment could be compared with the results of the SATURN demonstrator in the Bordeaux metropolis to ensure consistency. The method developed in the SATURN project based on data from the Urban Atlas allows performing territorial analysis on urban catchments in all European cities covered by the Copernicus territory service. Also, using easily accessible Landsat data and the LCZ framework the World Urban Database and Access Portal Tools (WUDAPT) initiative developed a methodology and support community to digitize urban areas anywhere in the world and disseminate the resulting LCZ results. As a whole, since many cities are already characterized in terms of land use using the LCZ framework, there is potential for comparisons on pollutant generation potentials of cities at a national or supranational level.

#### *4.4.1.5 Climatic conditions*

This framework modelled climatic conditions within the range of usually observed rainfall values in an oceanic climate. The measured rainfall series in 2013 which includes storms with return periods from 1 to 6 months is selected to represent an unremarkable year of UWS management including storm events with CSO discharges. Indeed, the scope of this research is centered on the primary level of service for which UWS infrastructures are designed. This level of service is the management of pollution in urban flows during storm events. Extreme storm events which are characterized by unusually intense rainfall overwhelming the capacity of drainage systems and causing widespread urban flooding belong to the domain of flood protection and their mitigation requires risk assessment approaches. It can be argued that the representativeness of the modelled rainfall time series could be increased by using a longer, historic time series (synthetic or actually measured) describing rainfall rates on a period of at least 5 years, compared to the choice of a single year (e.g. rainfall year of 2013). For a long-term continuous simulation in a hydrodynamic model such as SWMM, the computation time could be in the order of 8 to 10 hours to run 20 years of rainfall data with a time step of 10 minutes.

#### 4.4.1.6 Nonpoint sources

In this framework, the characterization of ID loads in terms of priority pollutants is a clear limitation to be recognized. Indeed, there is still a limited understanding of nonpoint emissions dynamics in relation to the land use and activities in the urban catchment. As our understanding of the most important sources of pollutants progresses with reliable emission factors, more source-emission pathways can be included in the STOIC framework. In particular, pollutants which are routinely found in WWTP effluents (e.g. alkylphenols, polychlorobiphenyls and derivatives) are not included in the proposed STOIC framework due to the multiplicity of sources, potentially including polluted, brownfield or industrial sites while the STOIC framework aims at characterizing relatively “generic” urban catchments. Therefore further studies on emissions and determination of emission factors of organic pollutants are in demand.

### 4.4.2 Consistency of LCIA methods

#### 4.4.2.1 The issue of spatiotemporal consideration in LCIA models

Currently there are several limitations concerning the consistency of LCIA methods which require to be dealt to improve the assessment of global UWS impacts. For example, existing LCIA methods lack sufficient differentiation to reflect spatiotemporal variations in both human-induced stressors and ecological responses from ecosystems.

*Toxicity models*

Most toxicity models focus on including spatial variability in chemical fate and human exposure (e.g. intake fraction). Recent time-dependent toxicity models have been developed using a dynamic fate model within the USEtox model (instead of the steady-state fate model) to provide impact results at any point in time (Shimako, 2017; Shimako et al., 2017). Ecotoxicity and human toxicity models demonstrated critical sensitivity to time step sizes in the dynamic LCI and calculation time span (Shimako et al., 2018). Consequently, it is important to further develop methodological frameworks to link dynamic system modeling with time-dependent impact assessment methods (Beloin-Saint-Pierre et al., 2016; Shimako, 2017). In this regard, the STOIC framework can provide time-differentiated UWS discharges (e.g. at 10-min time steps) during wet-weather events with time-dependent concentrations and volumes of SS. As future LCIA models improve with a more detailed description of pollutant states and effects in the environment drawing on from risk assessment methods, it will become possible to differentiate between acute and chronic effects from time-differentiated UWS discharges.

*Eutrophication models*

In ReCiPe 2016 the spatial differentiation for eutrophication modelling is rather basic, with country-specific characterisation factors. Therefore, Woods et al. (2017) recommends that future eutrophication impact indicators include the characterization of the vulnerability of ecosystems, which results from the sensitivity, adaptive capacity, and recoverability of individual species or communities which can vary over time and space (Zijp et al., 2017).

Consequently the inclusion of spatiotemporal differentiation in eutrophication models would greatly improve the assessment of IDs especially of CSO discharges which carry a significant nutrient load compared to treated wastewater effluents. Indeed, during summers with low river discharge rates and elevated temperatures, an untreated discharge of nutrients and particulate matter could have more serious impacts on the lower than usual dissolved oxygen in riverine ecosystems at this critical period, translating into a greater eutrophication potential.

#### 4.4.2.2 Incomplete pathways in the cause-effect chain

Another limitation concerns missing pathways in the endpoint modelling of existing impact categories due to lack of global information (Huijbregts et al., 2017). This applies to marine eutrophication in the ReCiPe 2016 method, which means that nitrogen loads in the UWS discharges are not currently characterized into damages to ecosystem quality. Inventory results that cannot be assigned to an existing impact category but are assumed to be environmentally relevant, should be reported – in line with ISO clause 4.4.2.5 – separately (e.g. as ‘missing important’ (EC-JRC, 2011)), in addition to the LCIA category indicator results.

#### *4.4.2.3 The issue of metal speciation in LCI and LCIA models*

In an aquatic system, inorganic substances and metals can be found co-existing as different chemical forms (species) with different characteristics depending on external conditions such as pH, redox potential or dissolved organic carbon levels. In general, the speciation of a pollutant influences its effects, transport and treatment. Therefore the speciation of heavy metals requires specific consideration for the following reasons (Jolliet and Fantke, 2015):

(i) The different species of heavy metals show different toxicity in the environment because they possess the ability to form complexes with organic and inorganic substances as ligands. Free metal ions (i.e. as noncomplex bound metal ions) are more potent than metals associated with particles or occurring as metal complexes.

(ii) Removal processes in both natural and treatment systems depend on the nature of the different species (e.g. the charge of a given metal species might influence the sorption on the surfaces of particles).

Recent developments in toxicity models now include speciation modelling in natural waters in the fate, exposure and effect calculations for marine and freshwater ecotoxicity (Dong et al., 2016, 2014, Gandhi et al., 2011, 2010). Concerning removal processes occurring in the technosphere, LCI models are now manifestly required to describe speciation and the equilibrium partitioning of metals between the waste matrix material and the aqueous phase in collection systems (e.g. sewer networks prone to particle sedimentation and biofilms) and treatment systems (land application units, waste piles, landfills, detention basins, aerated tanks etc.). By considering these two aspects, all processes occurring in the technosphere and ecosphere are then captured consistently in either LCI or LCIA. In this regard, boundary overlaps or gaps between LCI and LCIA modelling are prevented, drawing a parallel with the recommendations of van Zelm et al. (2013) concerning toxicological assessments of pesticides.

#### *4.4.2.4 Characterisation of suspended solids (SS) in LCIA*

Suspended solids (SS) are typically fine particles that remain in suspension in water and have a range of detrimental effects on freshwater ecosystems when their concentrations peak (e.g. reduced light penetration due to turbidity, health damages to macroinvertebrate organisms). At the present time in most LCIA methods, SS are not yet included due to immature midpoint-to-endpoint pathways. Hence assumptions regarding the final destinations for SS proposed in this framework cannot be assessed at the moment. Instead, by using SS as proxy for pollutants with a binding affinity for particulates, the framework enables the consideration of possible routes within the UWS for these pollutants. However, future developments in impact assessment methodologies that address SS could easily be incorporated with the current framework structure. For example, Quinteiro et al. (2015) developed a method to assess damages (endpoint level) caused by SS on freshwater species (aquatic invertebrate and macrophyte communities). The resulting method proved that (i) the effects of SS on aquatic biota are comparable with those of phosphorus causing eutrophication in P-limited freshwater ecosystems; and (ii) the persistence of SS is of the same order, if not greater than that of phosphorus. Therefore, the application of this method to the LCI loads of IDs developed in this framework should prove interesting to enhance results of the damage assessment of UWS discharges on the AoP ecosystem quality.

#### *4.4.2.5 How to consider long-term emissions and impacts of metals*

Using two LCIA methods in toxicity and ecotoxicity (e.g. ReCiPe and USEtox v2.01), the contribution analysis of UWS discharges highlighted that two metal species contributed by far compared to other priority pollutants such as PAHs. This “masking” effect can lead to a skewed interpretation of results in decision-making where toxic impacts from organic pollutants appear inconsequential in comparison to those of metals. In fact, toxic impacts depend on a number of driving factors (Rosenbaum, 2015):

1. Emitted quantity (as determined in the LCI)

2. Mobility (as determined by in the fate factor)

3. Persistency (as determined by the fate factor)

4. Exposure patterns and bioavailability (as determined by the exposure factor)

5. Toxicity (as determined by the effect factor)

The substantial overestimation of impacts from metals is a recognized problem in LCIA and results from the lasting occurrence (persistency) and toxic activity (speciation) of metals in the environment over an infinite time horizon, whereas organic pollutants eventually degrade. An infinite time horizon and continuous emission for steady–state condition (solving steady-state mass-balance fate models) is generally used for the calculation of characterization factor for toxicity impact. The consideration of an infinite time horizon may hide the potential impacts occurring over short periods of time in the assessment of a system, because of the different nature of substances considered. Some methods (e.g. ReCiPe and IMPACT World+) allow considering defined time horizons for metals. For instance, in ReCiPe 2016 different time horizons are proposed in scenarios the LCA practitioner can choose from an infinite time horizon (egalitarian scenario), to 20 year (individualist scenario) and 100 year time horizon (hierarchic scenario) (Huijbregts, 2016).

Until the issues raised by the persistency and speciation of metals are resolved, it is arguable that including more organic pollutants in IDs from UWS would change significantly the overall toxicity potentials due to the great "masking" effect of metals. Therefore, we suggest discerning toxic impacts of organic pollutants from those of metals (Figure 24). This would facilitate the interpretation of results and ultimately, decision support concerning priority actions on pollutant reduction as recommended in the Water Framework Directive 2000/60/EC. Indeed, LCIA results provide a screening of the first ten to 30 pollutants significantly contributing to toxic impacts (Rosenbaum et al., 2008).

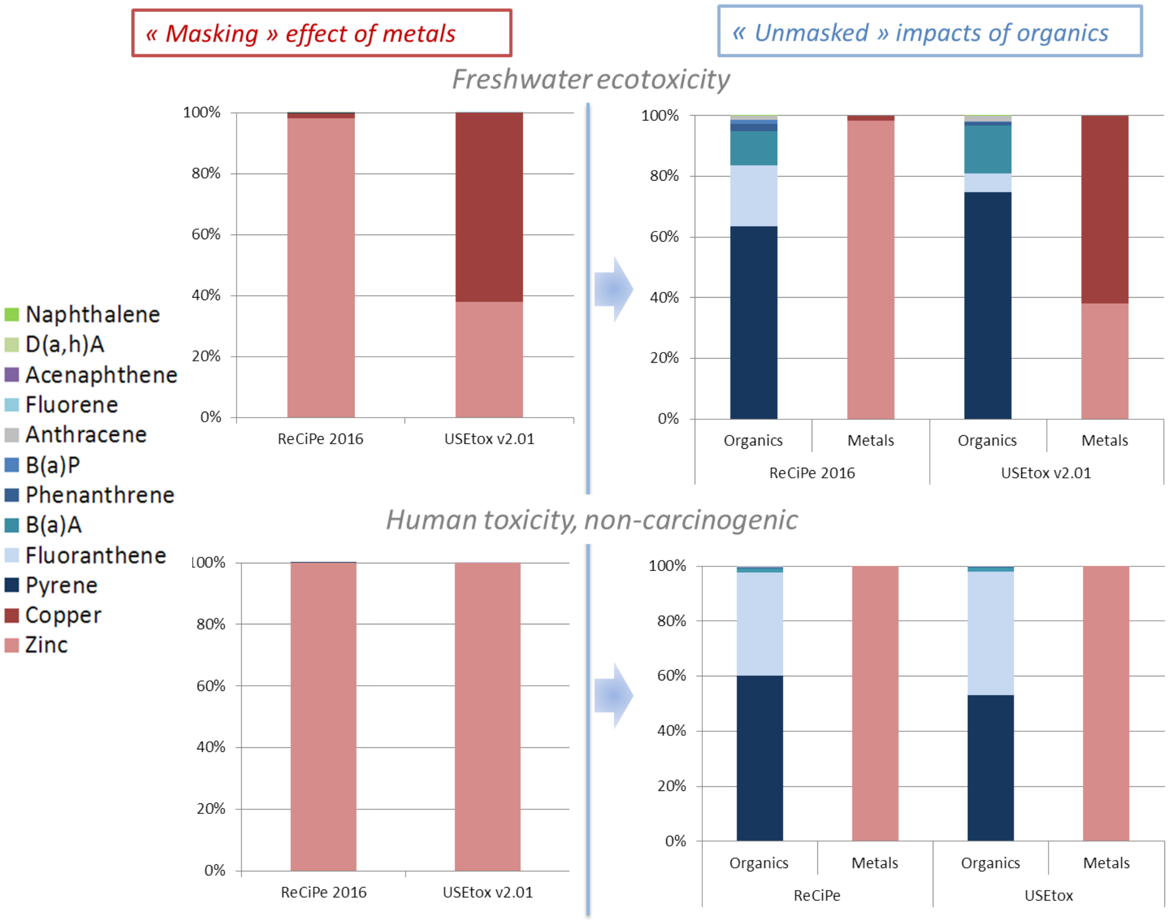

*Figure 24. Proposal to support interpretation of full impact assessment results on toxicity and ecotoxicity for decision-making. The "unmasked" impacts of organics support the identification of a ranking order among priority pollutants.*

Pradinaud et al. (2018) suggests that freshwater pollution impacts should be differentiated between short-term impacts (on ecosystem & human health) and long-term impacts affecting resources. Indeed there is an overlap between existing water deprivation impact models (e.g. AWARE (Boulay et al., 2018)) and toxicity models (e.g. USEtox) concerning the consideration of long-term freshwater pollution and depletion. Water deprivation models consider the issue of local or temporal freshwater unavailability leading to competition for scarce freshwater resources among existing users. In toxicity models, the impacts of a pollutant are calculated over its whole life time (until its degradation), potentially implying several generations of users over the span of several hundred years as for metals. To solve this double counting of environmental issues, Pradinaud (2018) recommends that long-term freshwater pollution and depletion should contribute to impacts assessed on the AoP

Resources, while short-term freshwater pollution concerns the impacts on the AoP Human Health.

#### 4.4.2.6 Including whole-effluent toxicity

In existing LCIA toxicity models, the toxicity of a wastewater or stormwater discharge is assessed by accounting for the sum of toxic impacts of its individual pollutants (simple additivity). It is however a simplification of the aggregate toxicity of a complex effluent given that this assumes (i) no effects of interactions between constituents (e.g., synergism, antagonism) and, (ii) no assessment of the bioavailability of the toxic constituents. Indeed, inorganic nanoparticles can reduce the bioavailability for some priority pollutants in wastewater effluents (Martín-De-Lucía et al., 2017). Integrating whole-effluent toxicity in UWS discharges or in specific industrial/waste treatment discharges would improve toxicity assessment of anthropogenic streams which are commonly modeled in LCAs. To this end, synergistic and antagonistic effects of substances should be explored as well as interactions with the waste matrix materials in treatment systems (e.g. sludge or deposit).

### 4.4.1 Expanding the spatiotemporal scope of LCAs of UWS

#### 4.4.1.1 On the temporal dimension: towards full dynamic LCAs

The STOIC framework describes UWS dynamics and specificities of the urban catchment and climatic conditions leading to dynamic, site-dependent LCIs for intermittent discharges for the foreground processes. Indeed, developing temporal dynamics in LCI for these foreground system processes is relevant for impacts on aquatic ecosystems: the timescales of environmental interventions (emissions or discharges) are of the same order of magnitude or greater than the timescales of impacts (Collet et al., 2014). Nonetheless, system dynamics in background processes such as infrastructure and operation of the UWS components (e.g. sewer network, CSO retention tanks, WWTP) should also be captured to increase the relevance of LCA results. To this end, the STOIC framework could be combined to a computational framework which includes the temporal behaviours of background processes (and their supply-chains), such as the DyPLCA framework (Tiruta-Barna et al., 2016). Since some impact categories are particularly sensitive to dynamics of environmental interventions as demonstrated in Shimako et al. (2018), the integration of temporal dynamics in the LCIA step is advisable. For this complete integration of temporal dynamics in LCA, dynamic LCI results can be used as inputs to dynamic LCIA models as shown in Figure 25.

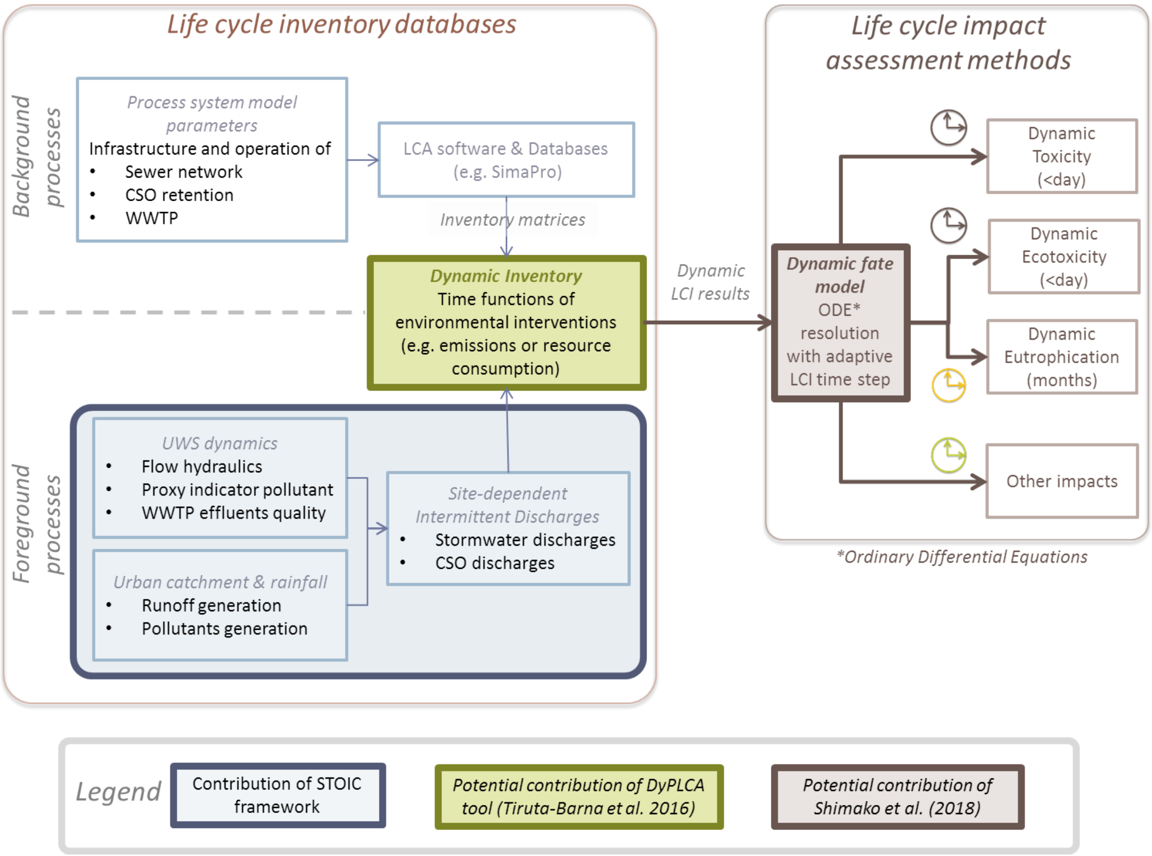

*Figure 25. Integration of the STOIC framework within a DLCA of UWS leading to the integration of dynamic aspects in both foreground and background processes.*

#### 4.4.1.1 On the spatial dimension: bridging the gap in hydrological modelling

The STOIC framework was developed mainly with the aim of improving the characterization of intermittent discharges carrying pollution from nonpoint sources in urban catchments which arises from land occupation and associated human activities. However the quantitative aspect (e.g. flows) of these intermittent discharges was addressed as well. The local urban water cycle is modelled with a description of urban surfaces (LCZ) and hydrologic modelling of urban flows through the catchment and the UWS, thereby adding a spatial dimension at the scale of an urban catchment. Thus, UWS discharges to receiving waters and infiltrated flows through permeable urban surfaces (e.g. sustainable urban stormwater design in blue-green infrastructures, or destination of returned flows as recycled water) are quantified in the STOIC framework. In recent methodological guidelines concerning water consumption impact assessment, the spatial dimension is now addressed with multimedia fate modelling of water flows in natural hydrologic units (Núñez et al., 2018). Hydrological changes at the urban catchment scale could potentially be combined with hydrological processes within the water compartments of the broader watershed and the global water cycle (Figure 26). There is the remaining challenge of defining transparent boundaries between hydrological processes occurring in the urban environment and those taking place in the natural environment, to prevent boundary overlaps between LCI and LCIA modelling. Given the current and future uncertainty in water scarcity, water use and land use change which affect downstream ecosystems considering environmental flows is expected to become increasingly valuable for guiding water-focused decision-making in both foreground and background processes in the UWS.

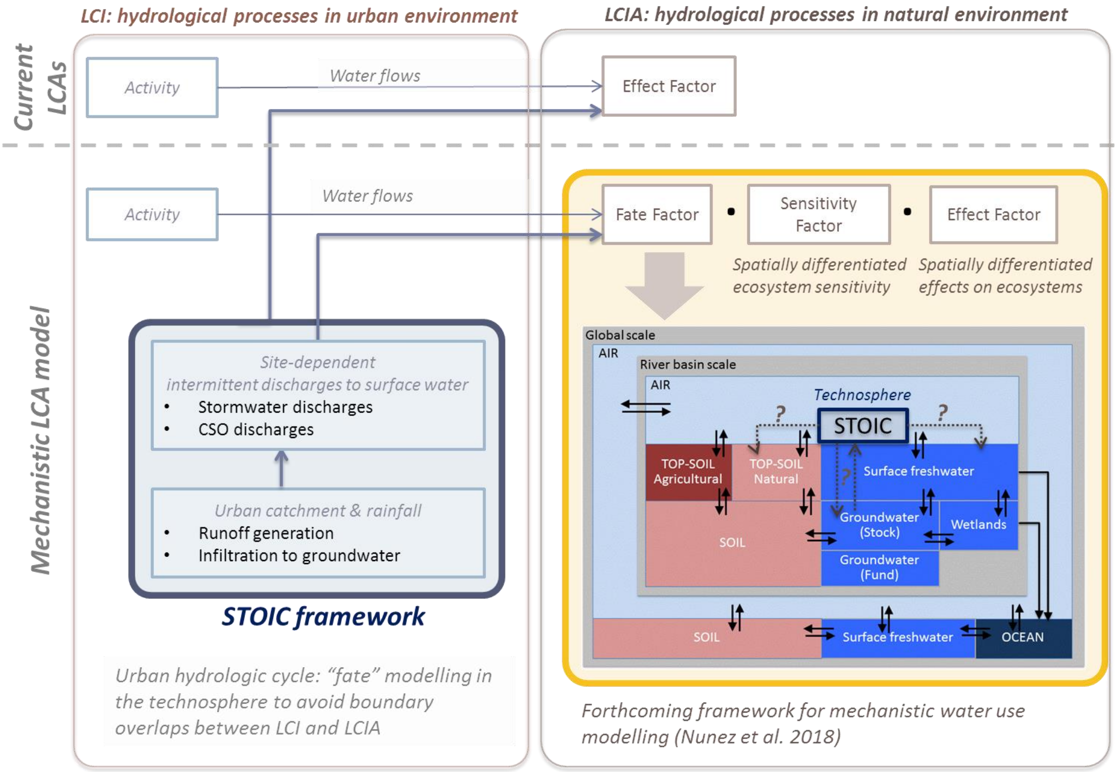

*Figure 26. Modelling impacts from changes in water availability in current LCAs and in a forthcoming framework based on a mechanistic multimedia fate model: opportunities for the integration of the STOIC framework.*

### *4.4.2 STOIC framework for decision support*

The STOIC framework provides spatiotemporally differentiated LCI for UWS discharges; hence the interpretation of full LCA results should be done bearing in mind that LCIA impacts are potential and not actual in contrast to environmental impact assessment (EIA) approaches. First, the LCA framework is based on generic/average ecosystems and impacts (e.g. no site specificity) with simplified effects models due to limited information available on the sensitivity of ecosystems or populations (e.g. the human toxicity model do not account for gender or age of exposed populations in contrast to risk assessment approaches). Second, pollutant flows are inventoried as emitted masses not concentrations, due to unknown baseline environmental conditions or unknown upstream concentrations in the case of freshwater ecosystems. In this context, the potential uses of LCA results concern the eco-design applications for a UWS by:

- Comparing total discharges which are linked to retention capacity of different UWS scenarios involving technological/structural elements (e.g. CSO infrastructures) and/or management strategies. In light of future climatic conditions and urban development, forecasting scenarios can be developed to assess how existing UWS infrastructures perform under altered rainfall patterns (e.g. more intense and grouped storm events) using Intergovernmental Panel on Climate Change (IPCC) scenarios.
- Identifying environmental trade-offs with on the one hand, potential "induced" impacts of CSO infrastructures and energy use to mitigate pollution during storm weather, and on the other hand, potential "avoided" impacts (i.e. avoided pollutant discharges). Another aspect worth exploring concerns the identification of global/local environmental trade-offs for stormwater drainage systems which were originally designed for flow attenuation (e.g., grassed swales and stormwater basins) and are now considered for their water quality benefits. In this case, LCA provides common environmental metrics for both types of potential impacts across the 18 midpoint categories that can be aggregated in three endpoints.

However, LCA results indicate that pressures on local aquatic ecosystems are important, even when considered from the broader life-cycle perspective. The integration of LCA and EIA shows great potential to complement the weaknesses of one approach (e.g. limited consideration of local characteristics in LCA) with strengths of the other approach (e.g. site-specific and local assessment of industrial projects complying with EIA legislation) (Larrey-Lassalle et al., 2017). Focused impact/risk analysis (e.g. local EIA) should therefore remain an important step in UWS decision making, to consider specificities of local receiving waters through the seasons (e.g. time-dependent sensitivity, assimilation capacity and recoverability). Notwithstanding that, LCA does have an important role to play in UWS analysis. Indeed, LCA provides a transparent and robust framework for assessing UWS and its sub-systems in a holistic manner to account for impacts relevant to more "local" categories (e.g. direct discharges negatively affecting the receiving water) along with those "background" processes causing global impacts (e.g. electricity and resource use).

This study did not include the construction of sewer and WWTP infrastructures because the research objective was to investigate and compare in terms of environmental impacts several control strategies for IDs for an existing UWS. However if the research question was to address planning and forecasting scenarios where the sewer network and/or the WWTP facilities are expanded, it would be necessary to include in the systems boundaries the construction of these infrastructures, which have significant environmental impacts (Risch et al., 2015).

## 4.5 Conclusions

The environmental burden of a sub-catchment-scale UWS in a real world case study was assessed using the developed ID loads with the STOIC framework in the inventory step, to enable a complete assessment of the UWS, including wet-weather conditions. This LCA application expanded the scope of the assessment, with the identification of pollution transfers in a life cycle perspective: across sub-systems or across impact categories. Indeed, in order to improve the quality of local water bodies (e.g. eutrophication or ecotoxicity), significant impacts on other categories and in other places can be generated (e.g., climate change associated with energy production).

Global performances of different control strategies for IDs were compared in terms of environmental impacts, revealing small differences among the compared strategies. The study revealed that pressures on local aquatic ecosystems were important, even when considered from the broader life-cycle perspective.

Spatiotemporally differentiated LCIs for IDs were calculated for this case study, which demonstrates the applicability of the STOIC framework to include temporal variability in IDs from UWS in a sub-catchment. However, further work is required to streamline the practical implementation of the STOIC framework to a variety of catchments.

# Chapter 5.: General conclusion

Multi-criteria, quantitative impact assessment of whole systems infrastructure can provide substantial value to guide decision making and policy debate for the urban wastewater industry. However, Chapter 1 pointed out the current limitations in existing environmental assessment methods for UWS impacts. In particular, Chapter 1 underlined the lack of consideration for intermittent discharges from UWS during storm events in LCA to fully capture the impacts of urban systems on the receiving waters.

To address this issue, the first research objective (Chapter 2) evaluated the relative significance of intermittent discharges versus continuous (dry-weather) discharges on impact categories concerned with water quality (e.g. eutrophication and ecotoxicity) using a simple typology for wet-weather event classification. This highlighted a number of challenges associated with measured water quality and flows data on intermittent discharges from a large urban catchment, such as the heterogeneity of measured data to characterize inventory pollutant loads. The proposed choice of wet-weather event classification into categories allowed to better define intermittent discharges and underlined the importance of their associated pollution relative to continuous discharges from UWS at the day and year scales. For the (climatic and infrastructure-related) characteristics of the UWS servicing the Greater Paris, the potential impacts from intermittent discharges were significant despite the important storage capacity. This study identified the major contributors to the freshwater ecotoxicity impact among which metals (Cu, Zn), PAHs and alkylphenols.

Following this first conclusion, the second research objective was to define a parsimonious, site-dependent framework linking urban catchment characteristics to pollutant loads in wet-weather intermittent discharges, and including temporal dynamics where appropriate in the framework (Chapter 3). Chapter 3 therefore provides an answer to this objective, the STOrmwater Inventory Conceptualization (STOIC) framework, through (i) fulfilling inventory data requirements to support sufficiently accurate modelling of intermittent discharges, (ii) conceptualizing stormwater processes from nonpoint source pollutants generated in the catchment (spatial dimension) to the dynamic hydrological transport of pollutants occurring in the UWS of the urban catchment at a sufficient temporal scale to describe climatic variations in typical storm events (temporal dimension). The resulting spatiotemporally differentiated LCI of intermittent discharges from the UWS can then be used as inputs to a classical or dynamic LCA to assess wet-weather management strategies.

The third research objectives (Chapter 4) concerned the evaluation and discussion of the STOIC framework applicability on a real case study. This LCA application expanded the scope of the assessment of UWS, which identified pollution transfers in a life cycle perspective: across sub-systems or across impact categories. Particularly, in order to improve the quality of local water bodies (e.g. eutrophication or ecotoxicity), significant impacts on other categories and in other places can be generated (e.g., climate change associated with energy production).

Finally, the applicability of the STOIC framework is demonstrated by assessing for the studied UWS and climatic conditions, the global performances of wet-weather control strategies in terms of net environmental impacts. In conclusion, the general research question was answered with the main outcomes of the thesis, and a site-dependent framework has been successfully developed to capture temporal variability of IDs from urban catchments, which were not accounted for in previous LCAs of UWS. The STOIC framework is applicable to urban catchments at the national and supra-national levels. This represents a notable step forward in addressing the spatiotemporal impacts of human activities and settlements due to intermittent, nonpoint source pollution reaching aquatic ecosystems. Notwithstanding that, there are numerous perspectives concerning the improvement of this work, which are presented in the Discussion of Chapter 4. Within a short term perspective, this work could be integrated with recent advances in full dynamic LCA frameworks to improve the relevance of water quality impacts of intermittent IDs. While on a longer term perspective with multimedia hydrological fate models, the spatial relevance of water consumption impacts would be improved. Prospective scenarios of UWS with future, altered rainfall patterns, growing urbanization with changes in urban transport modes are a promising area for future research. One area of notable interest concerns further developments to streamline the STOIC framework implementation for UWS stakeholders. Indeed, the majority of LCA studies globally are conducted by users of LCA, rather than methodological researchers, whose LCA skills are strongest in the task of inventory collection.

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# Annexes

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# Annex A – Supplementary information for Chapter 2

## Annex A-1. Description of the sewer networks in the Greater Paris

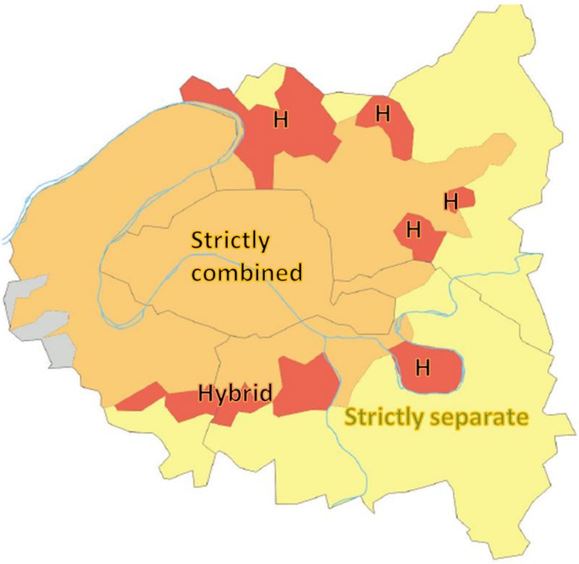


*Figure S1. Typology of sewer networks in the Greater Paris with the inner central area of Paris serviced with strictly combined systems (orange), the outer suburbs with strictly separate systems (yellow) and some areas with both systems (hybrid, in red)*

# Annex A-2. Data required for the statistical treatment to obtain consistent LCI

*Table S1. Concentrations and frequency of occurrences for the 43 priority pollutants included in the study.*

| | | | | S1 | S2 | S3 | S4 | S5 | S6 | S7 | S8 | S9 | S10 | S11 | S12 | S13 | S14 | S15 | S16 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *If no concentrations in treated effluents from Mailler (2015), values measured in raw wastewater (Gasperi et al. 2012) were used with an abatement for activated sludge treatment (Ruel et al. 2010, 2012).* | | | Substance name | Anthracene* | Atrazine | Di-(2-ethylhexyl)-phthalate (DEHP) | Diuron | Fluoranthene | Isoproturon | Lead & compounds | Naphtalene | 4-nonylphenol (N-nonylphenol)* | Benzo(a)pyrene* | Dieldrin | Tetrachloroethylene | Trichloroethylene | Copper | Zinc | Chromium |
| **WTP** | | | | | | | | | | | | | | | | | | | |
| References | | Concentration (µg/L) | | | | | | | | | | | | | | | | | |
| Mailler et al. 2015 | Treated effluent concentrations (Min, Avg, Max and SD) - **when availabl**e (n= 14) | Minimum | | 5.00E-04 | 3.00E-03 | 9.19E-01 | 1.90E-02 | 2.60E-03 | 1.10E-02 | | | 1.43E-01 | 9.00E-04 | 0.00E+00 | | | | | |
| | | Average | | 6.00E-04 | 4.00E-03 | 1.41E+00 | 2.50E-02 | 3.20E-03 | 3.40E-02 | | | 8.41E-01 | 1.40E-03 | 0.00E+00 | | | | | |
| | | Maximum | | 8.00E-04 | 5.00E-03 | 2.70E+00 | 3.00E-02 | 3.90E-03 | 6.20E-02 | | | 1.90E+00 | 1.70E-03 | 0.00E+00 | | | | | |
| | | SD | | 1.53E-04 | 1.00E-03 | 8.62E-01 | 5.00E-03 | 0.00E+00 | 2.20E-02 | | | 6.81E-01 | 0.00E+00 | 0.00E+00 | | | | | |
| | | Occurrence (%) | | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 33.33 | 100.00 | 100.00 | 100.00 | 0.00 | 54.55 | 45.45 | 100.00 | 100.00 | 33.33 |
| Gasperi et al. 2012 | Raw wastewater concentrations (Q10, Q50, Q90) (n=11) | Q10 | | | | | | | | 1.34E+01 | 5.42E-02 | | | | 2.40E+00 | 7.40E-01 | 4.80E+01 | 7.60E+01 | 2.24E+01 |
| | Occurrences in measurements (%) | Q50 | | | | | | | | 1.50E+01 | 6.32E-02 | | | | 4.90E+00 | 1.10E+00 | 8.00E+01 | 1.70E+02 | 2.40E+01 |
| | | Q90 | | | | | | | | 1.50E+01 | 1.66E-01 | | | | 6.15E+00 | 1.36E+00 | 1.44E+02 | 2.36E+02 | 2.64E+01 |
| Ruel et al. 2010, 2012 | Abatement (%) in activated sludge, applied to raw wastewater values in Gasperi et al. (2012) | | Abatement (%) | | | | | | | 73% | 0% | | | | 93% | NA | 83% | 57% | 85% |
| | | | | | | | | | | | | | | | | | | | |
| **SWR** | | | | | | | | | | | | | | | | | | | |
| References | | Concentration ( | Occurrence (%) | 100.00 | 0.00 | 100.00 | 100.00 | 100.00 | 76.92 | 85.71 | 100.00 | 100.00 | 100.00 | 28.57 | 33.33 | 0.00 | 93.00 | 93.00 | 28.57 |
| Gasperi et al. 2012 | Stormwater concentrations in both combined and separate sewers (n=17) | Q10 | | 5.88E-03 | 0.00E+00 | 8.48E+00 | 1.02E-01 | 8.02E-02 | 1.45E-02 | 1.32E+01 | 5.80E-02 | 4.33E-01 | 3.34E-02 | 1.79E-02 | 5.60E-01 | 0.00E+00 | 3.00E+01 | 1.60E+02 | 1.45E+01 |
| Zgheib et al. 2012 | | Q50 | | 1.40E-02 | 0.00E+00 | 1.64E+01 | 3.50E-01 | 1.69E-01 | 4.50E-02 | 2.55E+01 | 9.66E-02 | 7.56E-01 | 6.11E-02 | 6.02E-02 | 8.10E-01 | 0.00E+00 | 5.00E+01 | 2.60E+02 | 2.55E+01 |
| | | Q90 | | 8.60E-02 | 0.00E+00 | 5.00E+01 | 6.48E-01 | 9.11E-01 | 6.30E-02 | 8.60E+01 | 3.47E-01 | 7.10E+00 | 3.04E-01 | 9.22E-02 | 1.19E+00 | 0.00E+00 | 1.46E+02 | 4.18E+02 | 3.44E+01 |
| | | | | | | | | | | | | | | | | | | | |
| **CSO** | | | | | | | | | | | | | | | | | | | |
| References | | Concentration ( | Occurrence (%) | 100.00 | 25.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 | 50.00 | 100.00 | 100.00 | 50.00 |
| Gasperi et al. 2012 | CSO concentrations measured for 4 events with CSO's (n=4) | Campaign 1 | | 4.02E-02 | 0.00E+00 | 1.48E+01 | 4.70E-01 | 3.73E-01 | 4.00E-02 | 1.75E+02 | 2.24E-01 | 1.35E+00 | 1.38E-01 | 5.46E-01 | 9.00E+00 | 1.30E+00 | 1.34E+02 | 1.14E+03 | 1.20E+01 |
| | | Campaign 2 | | 3.25E-02 | 0.00E+00 | 7.99E+00 | 3.70E-01 | 2.63E-01 | 4.00E-02 | 1.22E+02 | 1.05E-01 | 7.89E-01 | 1.03E-01 | 2.40E-01 | 6.50E+00 | 0.00E+00 | 8.60E+01 | 7.70E+02 | 0.00E+00 |
| | | Campaign 3 | | 1.40E-02 | 3.00E-02 | 1.33E+01 | 5.00E-01 | 3.62E-01 | 2.00E-02 | 8.30E+01 | 9.10E-02 | 2.19E+00 | 2.04E-01 | 9.80E-01 | 6.10E+00 | 1.70E+00 | 1.15E+02 | 9.36E+02 | 2.00E+00 |
| | | Campaign 4 | | 7.65E-03 | 0.00E+00 | 3.75E+00 | 1.90E-01 | 1.30E-01 | 2.00E-02 | 4.60E+01 | 1.12E-01 | 6.45E-01 | 6.20E-02 | 2.70E-01 | 2.60E+00 | 0.00E+00 | 9.00E+01 | 6.58E+02 | 0.00E+00 |

| S17 | S18 | S19 | S20 | S21 | S22 | S23 | S24 | S25 | S26 | S27 |
|---|---|---|---|---|---|---|---|---|---|---|
| Fluorene | Phenanthrene | Pyrene | Benzene* | Ethylbenzene | Toluene | Xylenes | Dichloromethane | Chloroform | Pentachlorophenol* | Butylphenol |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| 100.00 | 100.00 | 100.00 | 18.18 | 27.27 | 81.82 | 54.55 | 36.36 | 90.91 | 0.00 | 63.64 |
| 3.00E-04 | 1.30E-02 | 2.10E-03 | 7.13E-02 | 8.40E-01 | 9.90E-01 | 9.20E-01 | 4.86E+00 | 1.90E+00 |  | 1.38E-01 |
| 1.70E-03 | 1.67E-02 | 2.40E-03 | 2.53E-01 | 1.00E+00 | 1.70E+00 | 1.55E+00 | 5.65E+00 | 2.00E+00 |  | 1.80E-01 |
| 4.20E-03 | 1.91E-02 | 2.80E-03 | 4.35E-01 | 2.04E+00 | 4.04E+00 | 6.15E+00 | 6.37E+00 | 2.40E+00 |  | 2.18E-01 |
|  |  |  | NA | NA | NA | NA | 88% | 83% |  | 93% |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| 100.00 | 100.00 | 100.00 | 0.00 | 9.09 | 9.09 | 9.09 | 50.00 | 0.00 | 15.38 | 92.86 |
| 1.89E-02 | 5.63E-02 | 5.75E-02 | 0.00E+00 | 1.00E+00 | 1.00E+00 | 1.00E+00 | 2.00E+00 | 0.00E+00 | 1.06E-01 | 6.20E-02 |
| 3.02E-02 | 1.13E-01 | 1.66E-01 | 0.00E+00 | 1.00E+00 | 1.00E+00 | 1.00E+00 | 2.50E+00 | 0.00E+00 | 1.30E-01 | 1.07E-01 |
| 8.96E-02 | 7.22E-01 | 1.32E+00 | 0.00E+00 | 1.00E+00 | 1.00E+00 | 1.00E+00 | 7.40E+00 | 0.00E+00 | 1.54E-01 | 1.47E-01 |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| 100.00 | 100.00 | 100.00 | 0.00 | 50.00 | 25.00 | 25.00 | 0.00 | 0.00 | 0.00 | 50.00 |
| 3.82E-02 | 2.38E-01 | 3.73E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 1.16E-01 |
| 3.33E-02 | 2.09E-01 | 2.25E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 1.60E-01 |
| 7.00E-03 | 1.31E-01 | 4.10E-01 | 0.00E+00 | 5.80E-01 | 3.20E+00 | 1.20E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| 2.70E-02 | 7.40E-02 | 1.38E-01 | 0.00E+00 | 4.50E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |

| S28 | S29 | S30 | S31 | S32 | S33 | S34 | S35 | S36 | S37 | S38 |
|---|---|---|---|---|---|---|---|---|---|---|
| Simazine* | Glyphosate | Benzo[a]anthracene | Dibenzo(a,h)anthracene | Acenaphtene | PCB28 | PCB52 | PCB101 | PCB118 | PCB138 | PCB153 |
|  |  | 0 | 72.73 |  |  | 0 | 0 | 0 |  |  |
|  |  | 0 | 5.00E-04 |  |  | 0 | 0 | 0 |  |  |
|  |  | 0 | 0.00E+00 |  |  | 0 | 0 | 0 |  |  |
|  |  | 0 | 1.00E-04 |  |  | 0 | 0 | 0 |  |  |
| 0.00 | 36.36 |  |  | 72.73 | 45.45 |  |  |  | 18.18 | 18.18 |
|  | 4.90E-02 |  |  | 2.65E-03 | 2.04E-03 |  |  |  | 1.34E-03 | 1.34E-03 |
|  | 7.50E-02 |  |  | 1.38E-02 | 2.65E-03 |  |  |  | 1.41E-03 | 1.41E-03 |
|  | 1.08E-01 |  |  | 2.97E-02 | 4.38E-03 |  |  |  | 1.48E-03 | 1.48E-03 |
|  | 0% |  | NA | NA | NA |  |  |  | NA | NA |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| 38.46 | 92.31 | 100.00 | 100.00 | 100.00 | 76.47 | 64.71 | 70.59 | 70.59 | 88.24 | 88.24 |
| 2.00E-02 | 2.90E-01 | 2.80E-02 | 8.82E-03 | 1.51E-03 | 1.30E-02 | 1.25E-02 | 3.69E-03 | 7.23E-03 | 1.48E-02 | 1.71E-02 |
| 2.00E-02 | 1.15E+00 | 4.56E-02 | 2.24E-02 | 1.25E-02 | 3.48E-02 | 3.48E-02 | 4.01E-02 | 4.01E-02 | 5.15E-02 | 5.15E-02 |
| 6.60E-02 | 9.29E+00 | 2.55E-01 | 9.50E-02 | 4.24E-02 | 1.03E-01 | 1.03E-01 | 1.04E-01 | 1.04E-01 | 1.03E-01 | 1.03E-01 |
|  |  |  |  |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| 0.00 | 100.00 | 100.00 | 100.00 | 100.00 | 25.00 | 0.00 | 25.00 | 25.00 | 75.00 | 75.00 |
| 0.00E+00 | 2.90E-01 | 1.74E-01 | 3.64E-02 | 2.68E-02 | 3.23E-03 | 0.00E+00 | 3.23E-03 | 2.55E-03 | 5.46E-03 | 6.50E-03 |
| 0.00E+00 | 5.70E-01 | 1.05E-01 | 2.55E-02 | 2.85E-02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 5.10E-03 | 4.93E-03 |
| 0.00E+00 | 1.20E+00 | 1.68E-01 | 9.10E-02 | 6.00E-03 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 9.80E-03 | 7.70E-03 |
| 0.00E+00 | 7.20E-01 | 5.40E-02 | 2.40E-02 | 6.00E-03 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |

| S39 | S40 | S41 | S42 | S43 |
|---|---|---|---|---|
| PCB180 | 4-chloro-3-methylphenol | Desethylatrazine | Aminotriazole | Metaldehyde |
| 0 | | | | |
| 0 | | | | |
| 0 | | | | |
| 0 | | | | |
| | 45.45 | 63.64 | 27.27 | 63.64 |
| | 2.70E-01 | 1.00E-02 | 7.64E-01 | 2.44E-02 |
| | 3.30E-01 | 3.00E-02 | 2.00E+00 | 4.00E-02 |
| | 4.62E-01 | 6.00E-02 | 2.72E+00 | 6.00E-02 |
| | NA | NA | NA | NA |
| | | | | |
| | | | | |
| 70.59 | 0.00 | 23.08 | 76.92 | 61.54 |
| 3.18E-02 | 0.00E+00 | 1.00E-02 | 5.90E-02 | 4.10E-02 |
| 7.16E-02 | 0.00E+00 | 1.00E-02 | 1.95E-01 | 2.35E-01 |
| 1.04E-01 | 0.00E+00 | 1.80E-02 | 9.73E-01 | 4.08E-01 |
| | | | | |
| | | | | |
| 50.00 | 0.00 | 25.00 | 100.00 | 0.00 |
| 3.23E-03 | 0.00E+00 | 3.00E-02 | 2.90E-01 | 0.00E+00 |
| 9.10E-03 | 0.00E+00 | 0.00E+00 | 1.30E-01 | 0.00E+00 |
| 0.00E+00 | 0.00E+00 | 0.00E+00 | 2.84E-01 | 0.00E+00 |
| 0.00E+00 | 0.00E+00 | 0.00E+00 | 4.60E-01 | 0.00E+00 |

*Table S2. Volumes (in million m3) for inventoried flows for the year 2013 and at the scale of wet-weather events. At the yearly scale, values were either measured (effluents and CSO discharges) or estimated from rainfall depth (stormwater). At the event-scale, on a 24-h period, discharged volumes from wastewater treatment plants are assumed to be of an average value of 922/365 = 2,53 million m$^3$*

| | Yearly (total annual volume in 2013) | | | Event-scale | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Effluents - WTP | Stormwater - SWR | Combined sewer overflows - CSO | Effluents - WTP | Stormwater - SWR | | | Combined sewer overflows - CSO | | |
| Volumes discharged ($10^6$ m$^3$) | *measured* | *estimated* | *measured* | *mean* | *d10* | *d50* | *d90* | *d10* | *d50* | *d90* |
| T1 - Dry weather | 922 | | | 2.53 | | | | | | |
| T2 - Wet weather, no overflows | | 27.0 | | | 0.13 | 0.26 | 0.70 | | | |
| T3 - Wet weather, overflows | | 53.7 | 13.7 | | 0.23 | 0.72 | 1.86 | 0.01 | 0.11 | 0.70 |
| | ***Total wet-weather*** | ***94.4*** | | | | | | | | |

Wet-weather flow volumes were estimated with three statistical values: 1st, 5th (median) and 9th deciles. These statistical values divide the sampled event volumes (once ordered) in 10 equal bins of the same number of values. So the 1st decile (d10) represents the value below which there are 10% of event volumes, while the 5th decile (d50) is the median value, and the 9th and uppermost decile (d90) represents the value below which 90% of daily volumes can be found.

# Annex A-3. Inventory results and impact assessment scores on the aquatic categories

The section below presents the detailed results of the calculated mass loads and impact contributions for the three classes of archetypal weather days for the Greater Paris area: dry weather (T1), wet-weather without discharges of untreated sewage from CSOs (T2), and wet-weather with discharges of raw untreated sewage from CSOs (T3).

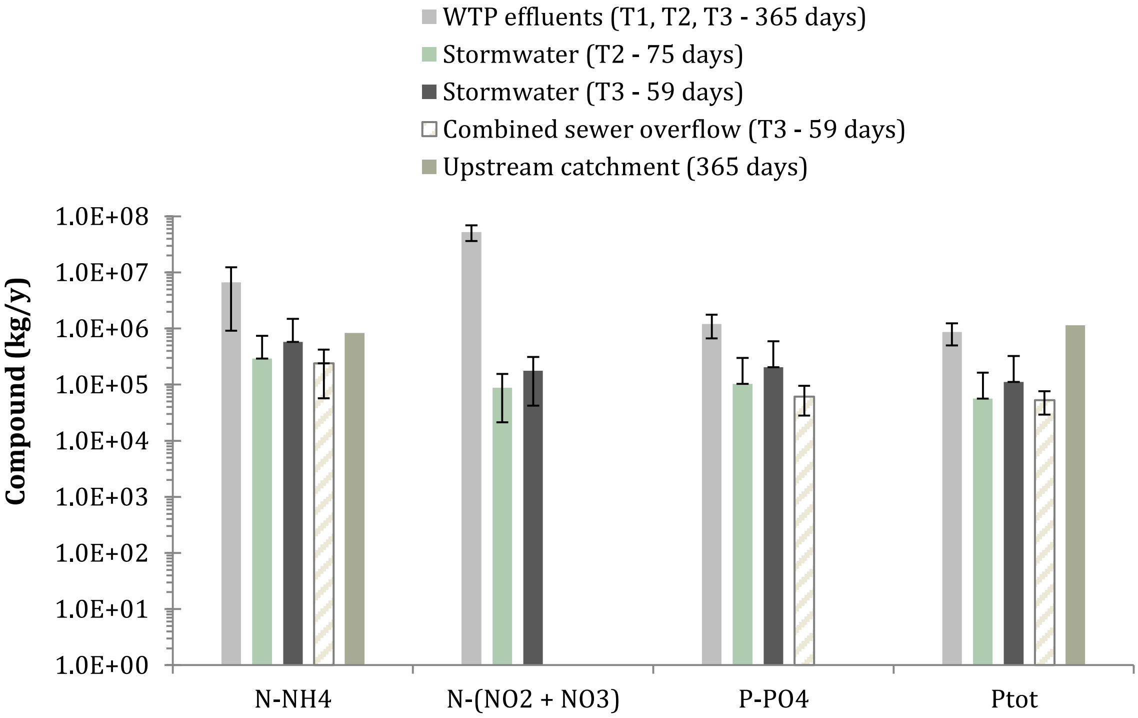


*Figure S2. Inventory results (cumulative mass per year, logarithmic scale for values in kg/y) of the computed mass loads in common pollutants for the different flows over the year 2013.*

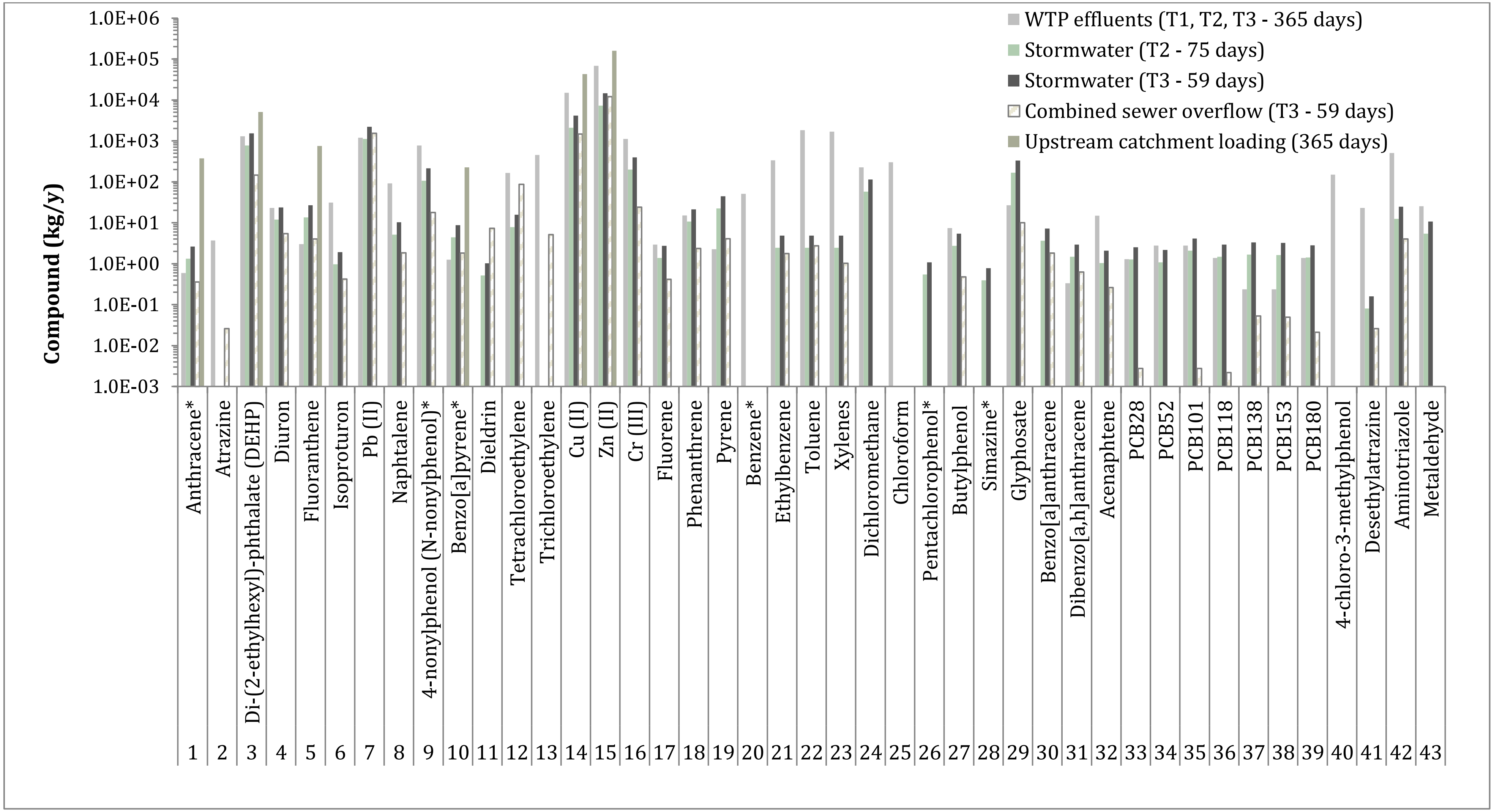


*Figure S3. Inventory results (cumulative mass per year, logarithmic scale for values in kg/y) of the computed mass loads in priority pollutants for the different flows over the year 2013. Pollutant emissions in 2013 which occurred upstream of the UWS in this study (upstream catchment loading) are included despite the scarcity of data.*

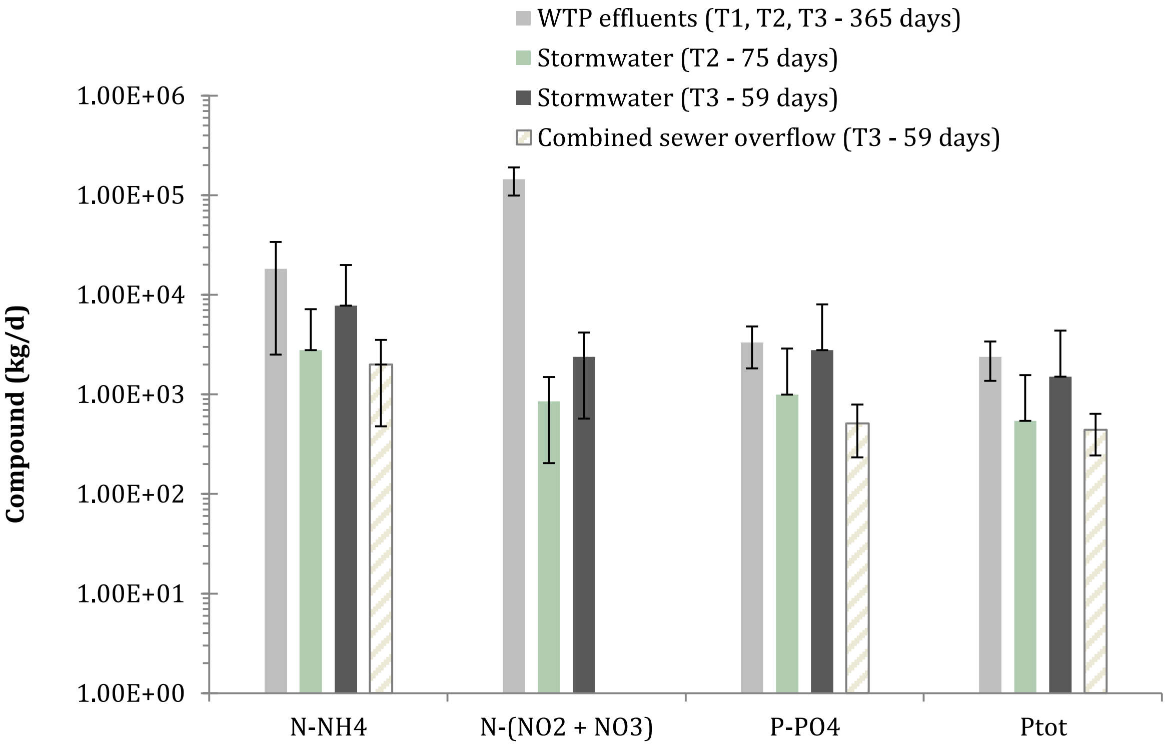


*Figure S4. Inventory results (logarithmic scale for values in kg/d) of the computed mass loads in common pollutants for the different flows on the event scale with median event-volume values (d50).*

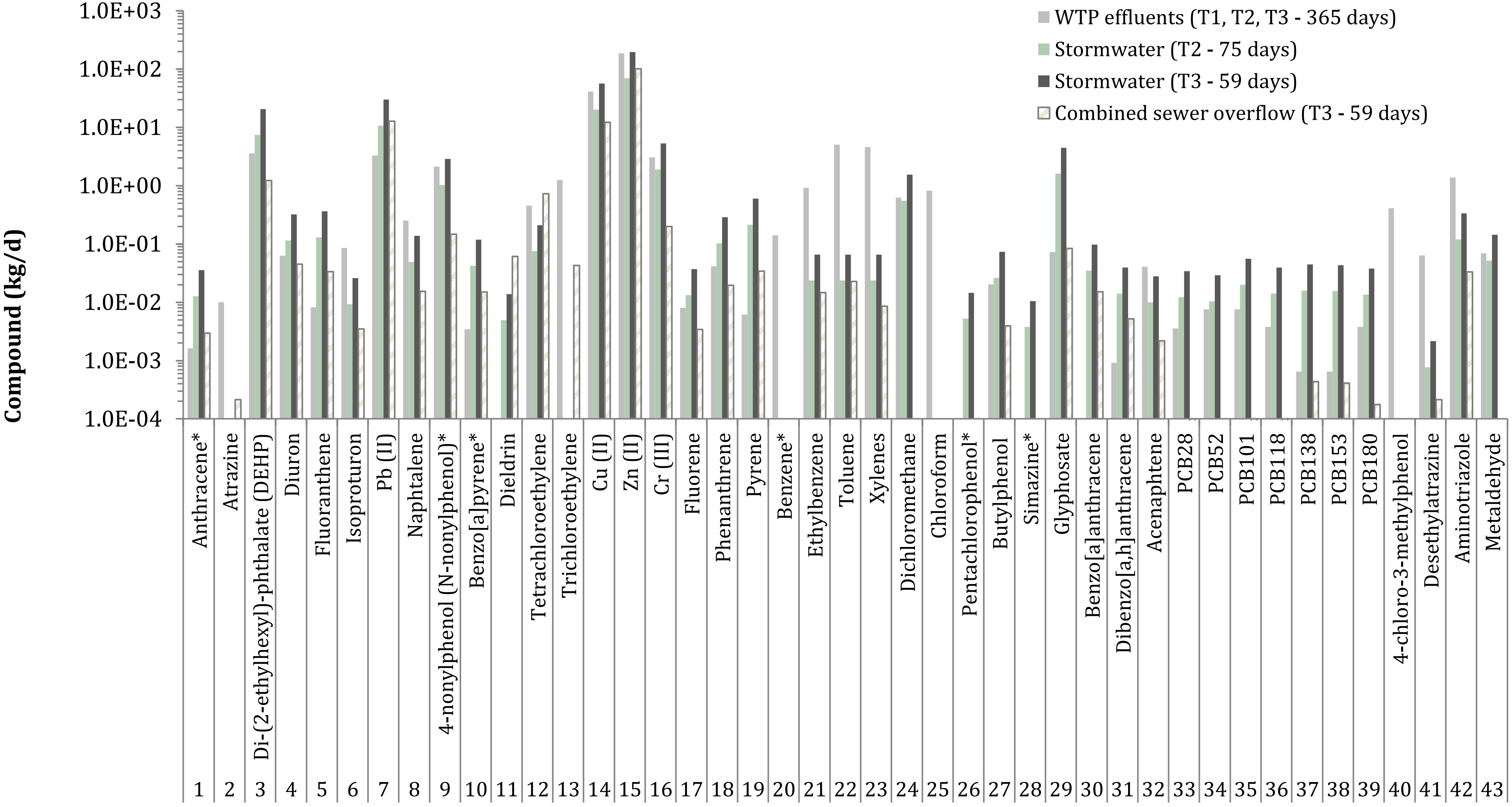


*Figure S5. Inventory results (logarithmic scale for values in kg) of the computed mass loads in priority pollutants for the different flows on the event scale with median event-volume values (d50).*

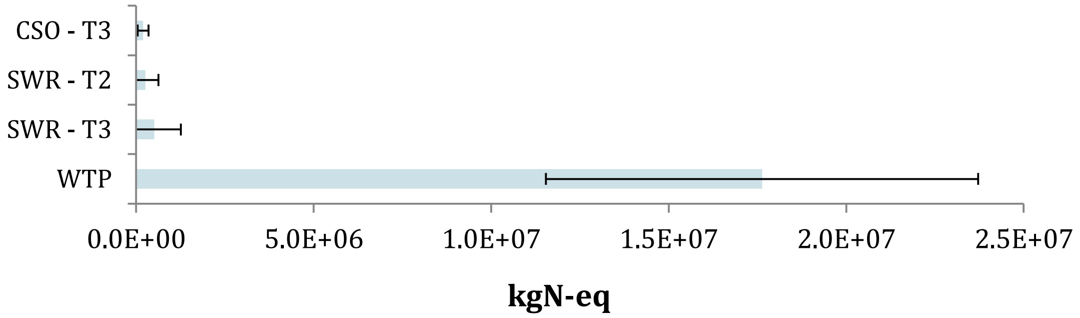
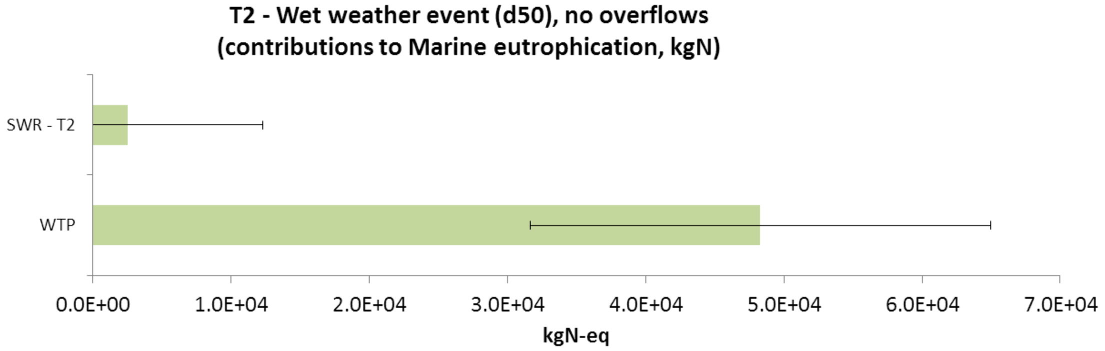


*Figure S6. Contributions of yearly flows to the Marine eutrophication impact (kgN)*



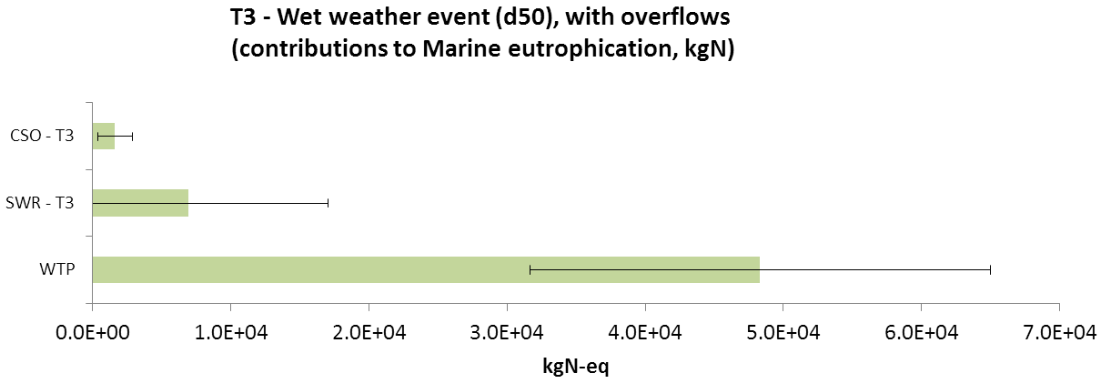
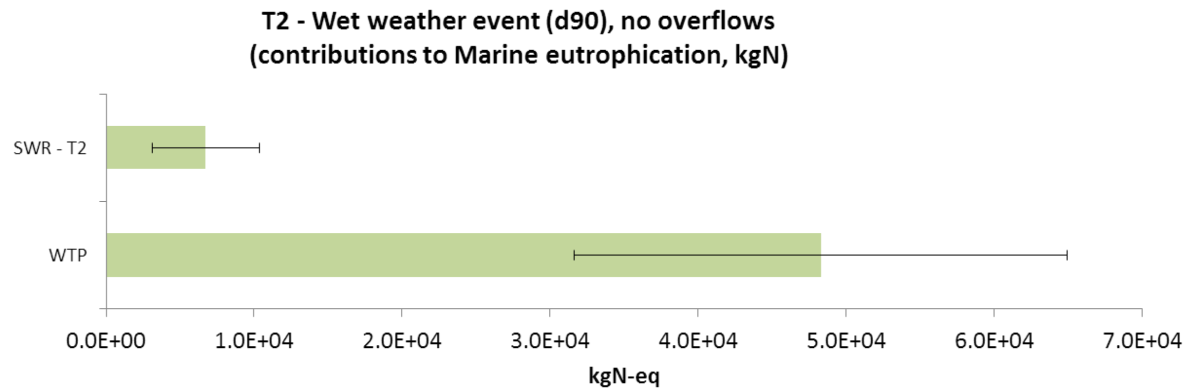


*Figure S7. Contributions to the Marine eutrophication impact (kgN) of urban dry- and wet-weather flows at event scale (d50)*



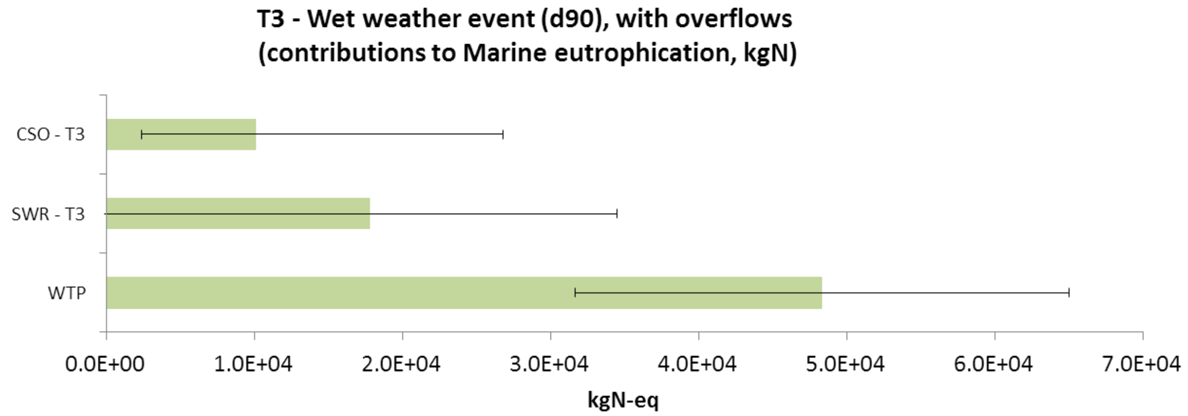


*Figure S8. Contributions to the Marine eutrophication impact (kgN) of urban dry- and wet-weather flows at event scale (d90)*

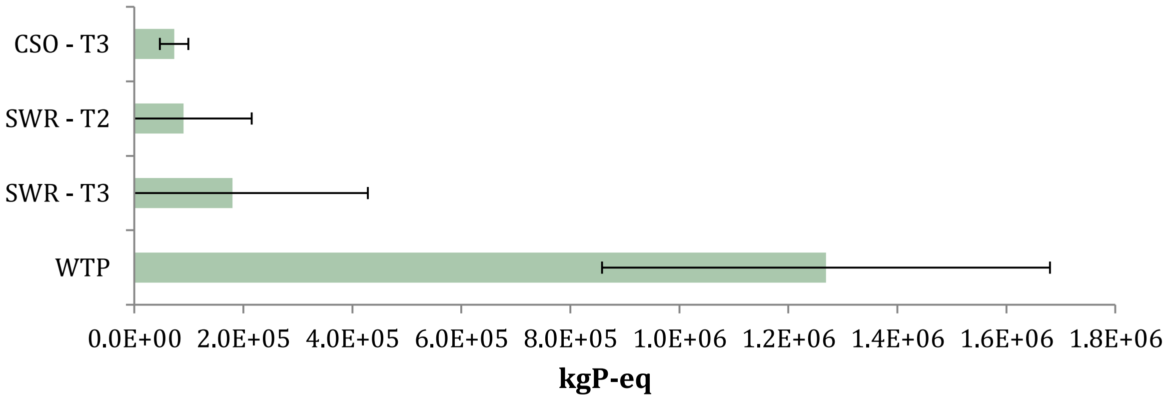


*Figure S9. Contributions of yearly flows to the Freshwater eutrophication impact (kgP)*

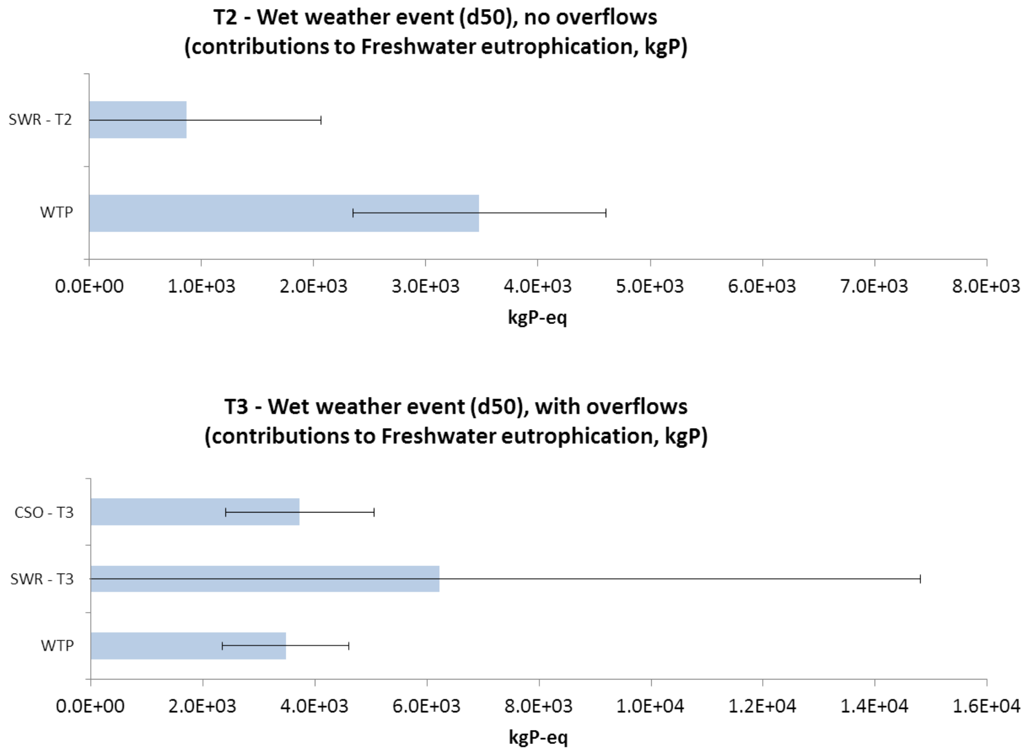


*Figure S10.Contributions to the Freshwater eutrophication impact (kgP) of urban dry- and wet-weather flows at event scale (d50)*

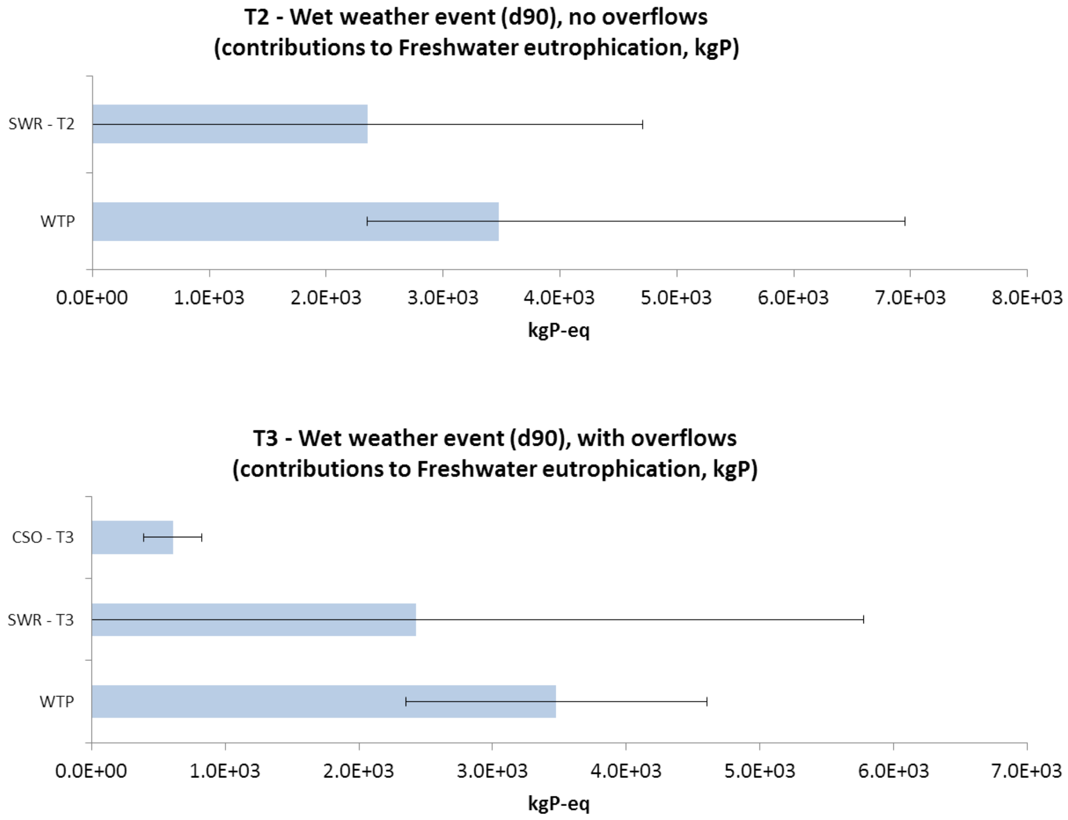


*Figure S11. Contributions to the Freshwater eutrophication impact (kgP) of urban dry- and wet-weather flows at event scale (d90)*

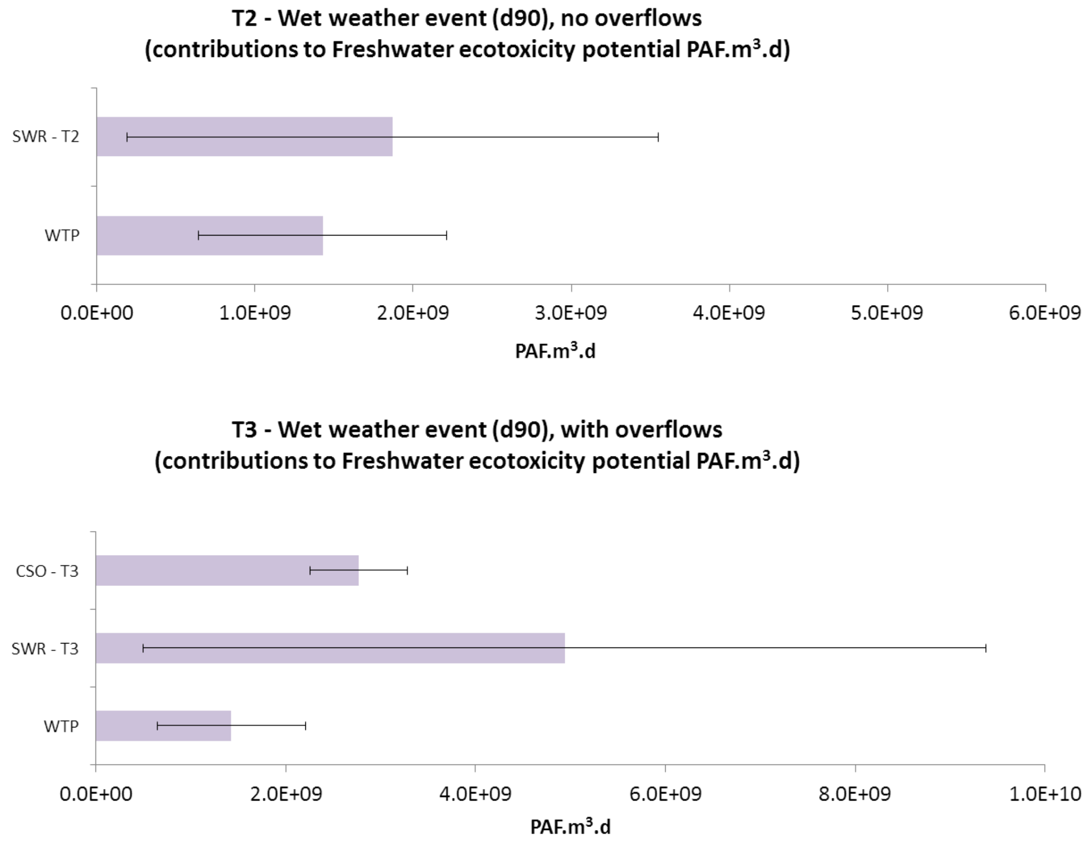


*Figure S12. Contributions to the Freshwater ecotoxicity impacts (PAF.m3.d) of urban dry- and wet-weather flows at event scale (d90)*

*Table S3. Contributing compounds to the aquatic eutrophication categories*

| Contributing compounds in yearly flows to impact categories of Eutrophication (ReCiPe) | Marine eutrophication (kgN-eq) | | Freshwater eutrophication (kgP-eq) | |
|---|---|---|---|---|
| Combined Sewer Overflows (CSO) | $NH_4$ (100%) | - | Ptot (72%) | $PO_4$ (28%) |
| Stormwater (SWR) | $NH_4$ (92%) | NOx (8%) | Ptot (62%) | $PO_4$ (38%) |
| WTP Effluents | $NH_4$ (32%) | NOx (68%) | Ptot (68%) | $PO_4$ (32%) |

*Table S4. Parameter values involved in the calculation of characterization factors for freshwater ecotoxicity potential in USEtox v2.0. Only the most contributing substances have been included.*

| ***USEtox v2.0*** Emission in freshwater compartment | | Fate factor [day] | Available fraction [-] | Ecotoxicity effect factor [PAF.m3.kg-1] | Characterization factor for Ecotoxicity potential [PAF.m3.day.kg-1] |
|---|---|---|---|---|---|
| **Substance** | **Archetype** | **FF** | **BF** | **EF** | **CF** |
| Cu(II) | Europe | 4,79E+01 | 1,88E-02 | 3,56E+07 | 3,21E+07 |
| Dieldrin | Europe | 1,19E+02 | 8,88E-01 | 2,14E+04 | 2,26E+06 |
| Pyrene | Europe | 2,19E+01 | 8,61E-01 | 6,88E+04 | 1,29E+06 |
| Zn(II) | Europe | 4,22E+02 | 5,02E-01 | 2,84E+03 | 6,01E+05 |
| Fluoranthene | Europe | 8,95E+01 | 8,81E-01 | 6,30E+03 | 4,96E+05 |
| 4-nonylphenol | Europe | 4,53E+01 | 7,93E-01 | 4,48E+03 | 1,61E+05 |
| Isoproturon | Europe | 5,07E+01 | 9,99E-01 | 2,99E+03 | 1,51E+05 |
| Cr(III) | Europe | 3,47E+01 | 7,38E-04 | 1,01E+06 | 2,58E+04 |
| Pb(II) | Europe | 3,84E+01 | 7,94E-03 | 7,21E+03 | 2,20E+03 |

# Annex B – Supplementary information for Chapter 3

## Annex B-1. Comparison of dynamic and average modelling approaches

The question of the influence of climatic conditions (especially the temporal succession of dry and wet-weather periods) on stormwater pollution generation is considered and further developed here: *Is stormwater pollution potential dependent on the temporal succession of wet and dry periods?*

Our objective is to develop a framework to predict stormwater pollutant loads for a given catchment at the scale of a year. Hence, we investigated the suitability of integrating deterministic models (such as SWMM) to estimate the stormwater pollutant loads from local urban sources. The ability of accumulation-washoff models to replicate temporal variations of concentrations in stormwater loads was questioned recently (Al Ali et al., 2017; Sage et al., 2015; Shaw et al., 2010). Sage et al. (2015) states that accounting for temporal variability in stormwater loads during rain events may not always be necessary to predict pollutant loads, as these loads are in fact mostly explained by stormwater volumes. Indeed, volume alone explained 85% of the variability in stormwater suspended solids loads in combined sewers (Joannis et al., 2015) and 69% in separate sewers (Shaw et al., 2010). A simpler modelling approach was thus also considered to evaluate the benefits of accumulation-washoff models in the prediction of annual loads for a sub-catchment. This “average approach” is based on a complete transfer of the accumulated pollutants to stormwater (i.e. 100% wash-off).

*Climatic profiles*

Two climatic profiles were included to test the sensitivity of annual exported mass of pollutants to rainfall distribution. Among the 3 pathways studied in the framework, only the leaching of metallic surfaces is dependent on rainfall amount. Annual precipitation data for two climates respectively Mediterranean and Continental central Europe were used, based on measured rainfall series from resp. Montpellier, France and Basel, Switzerland (data spanning 1 year with a time step of 1 hour).

*Material and methods*

The suburban catchment of Sucy-en-Brie in the Greater Paris area was chosen as case study for which a source-flux analysis provided data on nonpoint sources related to three primary activities: traffic, atmospheric deposition and metal corrosion from building elements (Petrucci et al., 2014). The catchment area is 228 ha, impervious cover is 21 %, and the population is about 5,200 inhabitants. Land use is mainly residential, with 97% of buildings represented as individual detached houses.

Traffic and atmospheric deposition generate accumulating “stock” pollutants, while corrosion of metal roof covers or gutters is dependent on rainfall height (e.g. metals leached per mm of rainfall). Leached metals can be assimilated to a “fund” of pollutants which cannot be technically depleted by intense rains. Hence in the following only traffic and atmospheric deposition were included in the quantification of stormwater loads.

*Results for the case study: Quantification of pollutant loads*

Nonpoint sources are quantified following the general estimation procedure:

$$\boldsymbol{E_{p,s}} = a_s \cdot EF_{p,s}$$

The level of activity for traffic and atmospheric deposition is first defined, and EF values for the two primary activities are presented in Table S6:

- ***Traffic***

In the urban catchment of Sucy en Brie, total traffic load was estimated at $21.5 \cdot 10^6$ vkm.y$^{-1}$, or a daily traffic load of $5.90 \cdot 10^6$ vkm.d$^{-1}$.

- ***Atmospheric Deposition***

Impervious areas where deposition occurs, are contributing to stormwater pollution. For the catchment of 228 ha, total impervious area was estimated at 48 ha. Table S5 summarizes the distribution of urban surfaces in the study catchment.

*Table S5. Distribution of urban surfaces for the study catchment*

| | λb Building surface | λi Imperv. Surface | λp Pervious surface |
|---|---|---|---|
| % surfaces | 15 | 6 | 79 |
| *Areas (ha)* | *34.2* | *13.7* | *180.1* |

*Table S6. Summary of EF values for the two primary activities included*

| Source | Source activity factor | | Pollutant | Emission factor, pollutant p, source s | | |
|---|---|---|---|---|---|---|
| *s* | $a_s$ | *Unit* | *p* | $EF_{p,s}$ | *Cv* | *Unit* |
| Atmospheric deposition | Total impervious area in catchment | [m²] | Cu | 8.68E-06 | 30% | $[g.(m^2.d)^{-1}]$ |
| | | | Zn | 2.41E-05 | 39% | $[g.(m^2.d)^{-1}]$ |
| | | | PAH | 6.28E-07 | 89% | $[g.(m^2.d)^{-1}]$ |
| Traffic activity | Traffic volume (daily) | [vkm.d-1] | Cu | 6.03E-07 | 65% | $[kg.vkm^{-1}]$ |
| | | | Zn | 1.46E-06 | 31% | $[kg.vkm^{-1}]$ |
| | | | PAH | 8.12E-09 | 55% | $[kg.vkm^{-1}]$ |

### *Results obtained for the dynamic approach*

We used two contrasted climates with the available rainfall data of 1 year of measurements at a hourly time-step:

- Mediterranean: Fréjorgues, France; total annual rainfall: 556.5 mm - Sep ‘15-Sep’16
- Continental – Central Europe : Basel, Switzerland ; total annual rainfall:738.9 mm - Jan ‘17-Dec ’17

Coefficients for the accumulation-washoff equations in SWMM were determined as follows:

- Dispersion rate D = 0.3 (low value (Borris, 2016))
- Wash-off coefficients for roofs & roads: w1 = 0.12, w2 = 1.5

Results for the dynamic approach are presented in Table S7:

*Table S7. Dynamic approach using SWMM model – total annual loads [kg.y-1]*

| Mass exported, stormwater $[kg.y^{-1}]$ | Continental Europe | | 739 mm | Mediterranean | | 504 mm |
|---|---|---|---|---|---|---|
| | Cu | Zn | PAH | Cu | Zn | PAH |
| Roofs | 3.78E-01 | 1.17E+00 | 2.30E-02 | 2.96E-01 | 9.14E-01 | 2.20E-02 |
| Roads | 3.70E+00 | 8.67E+00 | 5.40E-02 | 2.95E+00 | 6.91E+00 | 4.30E-02 |
| Total export, $kg.y^{-1}$ | 4.08E+00 | 9.84E+00 | 7.70E-02 | 3.24E+00 | 7.83E+00 | 6.50E-02 |

The ratio between total annual loads estimated for the two climates is around 0.8, ranging from 0.8 (Cu and Zn) and 0.84 (PAH). While the ratio between annual rainfall heights is 0.68.

M(Mediterranean)/ M( Continental ) = 0.8-0.84

$H_{Continental}$ /$H_{Mediterranean}$= 0.68

It can be hypothesized that annual pollutant loads are explained by annual rainfall heights (e.g. total stormwater volume). Hence we posit that for our objective of annual loads estimation, a dynamic modelling of pollutant build-up and wash-off is not necessary. However, this proportional relationship should be tested with another climate.

### *Results obtained for the average approach*

This average approach assumes there is a complete transfer of the accumulated pollutants to stormwater (i.e. 100% wash-off). Results are presented in Table S8.

*Table S8. Annual stormwater pollutant loads for the catchment*

| ***Total annual load [kg.y$^{-1}$]*** | | | Cu | Zn | PAH |
|---|---|---|---|---|---|
| LCZ6 | Traffic | | 1.30E+01 | 3.14E+01 | 1.75E-01 |
| | AtD | | 1.52E+00 | 4.22E+00 | 1.10E-01 |
| | Corrosion | Roofs | 0 | 4.65E+01 | 0 |
| | | Gutters | 1.65E+00 | 4.00E+01 | 0 |
| Total impervious surfaces (ha) | 48 | *Total (Kg.y-1)* | *1.62E+01* | *1.22E+02* | *2.84E-01* |

### *Comparison of approaches for the estimation of annual loads*

Results showed that for the two climates, the average approach predicted consistently larger stormwater pollutant loads than in the dynamic approach Table S9, and this was expected as this average approach gives an estimation of the maximum available pollutant (with 100% transfer to stormwater). Hence, when using the same source quantification (emission factors and source extent), estimated stormwater pollutant loads from the two approaches differ by a factor 3 to 4 (Table S10).

*Table S9. Estimated annual loads with dynamic and average approaches*

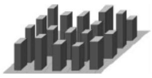
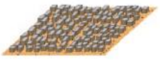
| | ***Dynamic Approach*** | | | ***Average Approach*** | | |
|---|---|---|---|---|---|---|
| | Mediterranean: 556.5mm | | | *Not sensitive to climate* | | |
| | Cu | Zn | PAH | Cu | Zn | PAH |
| *Total (Kg/y)* | *3.24E+00* | *7.83E+00* | *6.50E-02* | *1.45E+01* | *3.56E+01* | *2.84E-01* |
| | Continental: 739mm | | | | | |
| | Cu | Zn | PAH | | | |
| *Total (Kg/y)* | *4.08E+00* | *9.84E+00* | *7.70E-02* | | | |

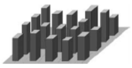
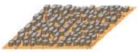
*Table S10. Calculated differences between Average and Dynamic approaches for two climates*

| ***Ratio Average/Dynamic*** | Cu | Zn | PAH |
|---|---|---|---|
| Mediterranean | 4.47 | 4.55 | 4.38 |
| Continental | 3.56 | 3.62 | 3.69 |

# Annex B-2. Description of local climate zones

*Table S11. Full definition of local climate zones (LCZs)including description of settlement structure, location, functions, distribution of building, impervious and pervious surfaces and building materials (Stewart and Oke, 2012)*

| Settlement structure | | Description | Location | Function(s) |
|---|---|---|---|---|
| LCZ1 |  | Compact high rise | Core (central business district, "downtown"), periphery | Commercial (office buildings, hotels), residential (apartment towers) |
| LCZ2 |  | Compact mid rise | Core (old city, inner city, central business district), periphery | Residential (multi-units housings, apartment blocks), Commercial (office bldgs, hotels), Industrial (warehouses, factories) |
| LCZ3 |  | Compact low rise | Old or densely populated cities, towns or villages, core, periphery (high density sprawl) | Residential (single unit housing, high density row housing), Commercial (small retail shops) |
| LCZ4 |  | Open high rise | Periphery. Densely populated cities. | Residential (apartment blocks, high rise housing estates) |
| LCZ5 |  | Open midrise | Periphery | Residential (multi-units housings, apartment blocks), Institutional (research/business parks, campuses), Commercial (office bldgs, hotels) |
| LCZ6 |  | Open low rise | City (medium density); periphery (suburbs), commuter towns, rural towns | Residential (single or multi-unit housing, low density row housing), Commercial (small retail shops) |
| LCZ7 |  | Lightweight low rise | Periphery of large, developing cities. Extended metropolitan regions. Inner city. Rural towns | Residential (shantytowns, squatter settlements, mobile housing), agricultural (small holder lots) |
| LCZ8 |  | Large low rise | Periphery | Light industrial (warehouses), commercial, transportation |
| LCZ9 |  | Sparsely built | Periphery (low density suburbs); Extended metropolitan regions. Newly developed urban tracts. Rural towns, villages. Lightly settled | Residential, institutional (campuses), agricultural (small farms, country estates) |
| LCZ10 |  | Heavy industry | City (periphery) or country | Industrial (factories, refineries, mills, plants) |

| Settlement structure | | Building surface % λb | | | Impervious surface % λi | | | Pervious surface % λp | | | Building height (m) (geometric average) | Construction materials |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | min | max | mean | min | max | mean | min | max | mean | | |
| LCZ1 |  | 40 | 60 | **50** | 40 | 60 | **50** | 0 | 10 | **5** | > 25 | Heavy materials: Steel, concrete, glass |
| LCZ2 |  | 40 | 70 | **55** | 30 | 50 | **40** | 0 | 20 | **10** | 17.5 | Heavy materials: Stone, concrete, tile, brick. Thick roofs and walls |
| LCZ3 |  | 40 | 70 | **55** | 20 | 50 | **35** | 0 | 30 | **15** | 6.5 | Heavy materials: Stone, concrete, tile, brick. Thick roofs and walls |
| LCZ4 |  | 20 | 40 | **30** | 30 | 40 | **35** | 30 | 40 | **35** | > 25 | Concrete, steel, glass |
| LCZ5 |  | 20 | 40 | **30** | 30 | 50 | **40** | 20 | 40 | **30** | 17.5 | Heavy materials: Stone, concrete, tile, brick. Thick roofs and walls |
| LCZ6 |  | 20 | 40 | **30** | 20 | 50 | **35** | 30 | 60 | **45** | 6.5 | Wood, brick, stone, tile |
| LCZ7 |  | 60 | 90 | **75** | 0 | 20 | **10** | 0 | 30 | **15** | 2.8 | Lightweight construction materials (bamboo, thatch, wood, corrugated metal) |
| LCZ8 |  | 30 | 50 | **40** | 40 | 50 | **45** | 0 | 20 | **10** | 5.5 | Steel, cocnrete, metal |
| LCZ9 |  | 10 | 20 | **15** | 0 | 20 | **10** | 60 | 80 | **70** | 6.5 | Wood, brick, concrete, stone, tile |
| LCZ10 |  | 20 | 30 | **25** | 20 | 40 | **30** | 40 | 50 | **45** | 8.6 | Steel, cocnrete, metal |

## Annex B-3. Methodological steps of the STOIC framework based on two sub-models

*Table S12. Pollutant loads inventory sub-model: Methodological steps, modelling hypotheses and associated data and methods used.*

| ***Step 1.*** | Urban area characterization in terms of surfaces contributing to stormwater pollutant generation, and the activity level of identified primary activities. | | | |
|---|---|---|---|---|
| Item | Modelling hypotheses | Data sources | Methods referenced | Comments |
| Identification of land use type of the sub-catchment; description of urban form (building geometry, construction materials, land cover, etc.) | Use of existing generic typology of urban structures: local climate zones (LCZ) | Visual classification on satellite images (Google earth), WUDAPT maps of urban areas worldwide, cartographic data on roads and buildings (BD TOPO® database; IGN 2009) | Stewart and Oke, 2012 | |
| Urban area definition: sub-catchment total area | Use of reported values for the sub-catchment chosen as case study | Reports, previous studies | | |
| LCZ identification | Use of existing generic typology of urban structures: local climate zones (LCZ) | Visual classification on satellite images (Google earth), WUDAPT maps of urban areas worldwide, cartographic data on roads and buildings (BD TOPO® database; IGN 2009) | Stewart and Oke, 2012 | |
| Distribution of elementary urban areas | Proposed range of urban area ratios for a LCZ (λ _i) | Database | Stewart and Oke, 2012 | |
| Identification of metal elements on building roofs (metal gutters and metal roof covers) | Metal gutters on individual housing buildings (open low rise, LCZ6): estimated at 3% of total gutter length. Total gutter length estimated using a correlation coefficient of 0.26 with the roof area. | Residential, open low-rise land use (LCZ6) based on Sucy en Brie, Paris suburban sub-catchment. Gutter_lin = 0.26 m$^2$_roof | Petrucci 2014 | Average ratio obtained from visual classification (average distribution of 2 independent classifications by expert researchers) on gutter materials |

| | | | | |
|---|---|---|---|---|
| | Metal gutters on collective housing buildings (Compact high rise, LCZ1). Buildings with Zn roofing have Zn gutters. Old tiles and slate roofing materials are considered to be associated with gutters made of Zn (95%) and Cu (5%). Total gutter length estimated using a correlation coefficient of 0.24 with the roof area. | Residential, compact high rise based on Le Marais, Paris sub-catchment. Gutter_lin = 0.24 m/m²_roof. | Sellami Kaaniche 2014 | Ratio derived from statistical analyses on collective building classes |
| | No metal gutters on industrial (LCZ8) and residential open mid rise buildings (LCZ5). | Commercial land use LCZ8 based on Chassieu, Lyon sub-catchment. Residential, open mid rise land use LCZ5 based on Pin Sec, Nantes sub-catchment. | Petrucci 2014 (INOGEV, Janv), Lamprea 2009, Sellami-Kaaniche 2014 | |
| | Metal cover: for LCZ1 - high Zn emission (54%), low Zn emission (0%). Other non metal covers to estimate metal gutters: Tiles (23.5%), Slate (18%). | Residential, compact High-rise land use (LCZ1) based on Le Marais, Paris inner centre. Zn roofs account for about 54% of total roof area. In addition, Zn is commonly used for gutters. | Robert-Sainte 2009, Thévenot 2007 | |
| | Metal cover: for LCZ6 - high Zn emission (3.6%) and low Zn emission (4.2%) | Mean value for LCZ6 sub-catchment (Sucy en Brie) | Petrucci 2014 | |
| | Metal cover: for LCZ5 - high Zn emission (1.7%) | Mean value for LCZ5 sub-catchment (Pin Sec, Nantes) | Lamprea 2009 | |
| | Metal cover: for LCZ8 - high Zn emission (27%) | Mean value for LCZ8 sub-catchment (Chassieu, Lyon) | Petrucci 2014 | |

| | | | | |
|---|---|---|---|---|
| Description of traffic activity | Use of population density/traffic activity relationship - for all LCZ classes excepted commercial/industrial zones for which traffic activity is estimated [vkm/d] | Population density values (INSEE in France). Parameter values in equation proposed for European cities. | Salomons 2012 | Method valid in the European cities context. Parameter values in the population density/traffic activity relationship are chosen for European cities. |
| ***Step 2.*** | Linking pollutants to activities: 3 primary pathways for Cu, Zn & PAHs with emission factors (EF) | | | |
| Atmospheric deposition | Average daily deposition rate [g/m²/d] estimated on impervious surfaces | Study sites: Nantes, representative of a relatively unpolluted urban residential area (Omrani 2017). Residential sub-catchment (LCZ6 open low rise) in Sucy en Brie (Petrucci 2014). Le Havre, residential blocks surrounded by a business park without industrial activity (Motelay-Massei 2006) | Petrucci 2014, Percot 2012, Motelay-Massei 2006, Bressy 2012, Omrani 2017 | |
| PAH distribution in atmospheric deposits | PAH fluxes from the atmosphere were calculated and described in detail for 14 PAHs, hence a specific PAH profile for this activity. | Urban watershed of Le Havre, France | Motelay-Massei 2006 | |
| Metal roof corrosion | Zn emissions estimated for galvanized zinc and other metallic roofs [g/m²/mm]. Zn and Cu emissions for Zn and Cu gutters [g/m/mm] | Proposed values found in Petrucci (2014) are aggregated from a database on experimental corrosion loads measured in Paris (Robert-Sainte 2009, 2010). | Petrucci 2014 | |
| Traffic activity | Road, brake and tyre wear estimated for a passenger car [g/vkm] | Literature on Substance flow analyses in Petrucci 2014, ecoinvent database | Petrucci 2014 | EF for Zn emission estimated in Petrucci 2014 is the same as in ecoinvent v3.4. However EF proposed in ecoinvent v3.4 process "Transport, passenger car, large size, diesel, EURO 3 – GLO" are lower by 1 order of magnitude for Cu and PAH emissions. Choice was made to stay with the most conservative values (Petrucci 2014). Ecoinvent |

| | | | | |
|---|---|---|---|---|
| | | | | processes' emissions are based on values from Ntziachristos, L., Boulter, P. (2009) EMEP/EEA air pollutant emissions inventory guidebook 2009. |
| PAH distribution in traffic-related road dust | Traffic-related road dust contains fine and coarse particles with a specific PAH profile for the traffic activity. | PAH loads for 15 PAHs were quantified in road dust samples (mg/kg, in fine & coarse fractions) and the total load was used to characterize the distribution of PAHs estimated with EF and traffic volume. | Ma 2017 | |
| ***Step 3.*** | Quantification of annual stormwater loads from activities on sub-catchment using EF and average rainfall height [kg/y] | | | |
| Atmospheric deposition | Total deposition [kg/y] estimated on impervious surfaces over a year | | | |
| Metal roof corrosion | Average rainfall height [mm/y] | Rainfall data for location | | |
| Traffic activity | Total road deposited sediments in sub-catchment [kg/y] calculated with EF and estimated traffic volume | Traffic volume [vkm/y] | | |

*Table S13. UWS pollutin distribution sub-model: Methodological steps, modelling hypotheses and associated data and methods used.*

| ***Step 1.*** | Determination of influent composition in Combined sewer system (Influent = wastewater + stormwater) | | | |
|---|---|---|---|---|
| Item | Modelling hypotheses | Data sources | Methods referenced | Comments |
| Wastewater composition per person-equivalent (PE) | COD, total | Ecoinvent value per PE | Doka 2009 | |
| | NH4-N | Chosen value within proposed range in technical report | Deronzier, G et al (2001) | Cemagref - FNDAE N°25 - Traitement de l'azote dans les stations d'épuration biologique des petites collectivités |
| | Org-N | | | |
| | PO4-P | Proposed value per PE relative to the Water Framework Directive | Stricker et Héduit, 2010; Mercoiret 2010 | https://epnac.irstea.fr/thematiques-transversales/caracteristiques-des-eaux-usees/ |
| | Org-P | | | |
| | Cu | Measured concentration values in untreated wastewater | Gasperi 2014 | |
| | Zn | | | |
| Affinity of primary pollutants to suspended solids in influent: fraction distribution between dissolved/particulate states | COD, N-species, P-species | Dissolved and particulate fractions given among the influent parameters | Hvitved-Jacobsen 2010 | |
| Affinity of metals and PAHs to suspended solids in influent: Fraction distribution between truly dissolved/colloidal/particulate states | Cu | Truly dissolved fraction in influent estimated at 10% | Hargreaves 2017 | |
| | Zn | Truly dissolved fraction in influent estimated at 8%, rounded to 10% | Hargreaves 2017 | |
| | PAH | Negligible dissolved fraction in influent, compared to particulate PAH fraction | Liu 2016 | |
| Definition of Influent load in a combined sewer system Mt = M-SPP + M-WPP | Sum of wastewater load from connected PE to the sewer system and stormwater load from sub-catchment | Inhabitants connected to sewer system | | |
| ***Step 2.*** | Distribution of pollution load between flows | | | |
| Partition of pollution load between water discharges (treated WTP effluents, CSO discharges) and trapped sediments according to affinity for particulate or dissolved phase: | The particulate fraction of the influent pollution will be assumed to follow the TSS distribution (result of the SWMM model), while the dissolved fraction of the influent pollution will be | | | |

| | | | | |
|---|---|---|---|---|
| | distributed between treated WTP effluents and CSO discharges (using volumes as a proxy). | | | |
| Deposits (trapped sediments) | TSS amount stored in network (pipes and retention tanks) modelled with US EPA rainfall runoff model SWMM | SWMM engine, calibrated TSS sub-model (LyRE, Maruejouls 2018) | | |
| Untreated CSO discharges | Volumes estimated in SWMM simulation. Particulate fractions are assumed to follow TSS distribution as in the SWMM simulation | SWMM engine, calibrated TSS sub-model (LyRE, Maruejouls 2018) | | |
| Treated WTP effluents | Volumes estimated in SWMM simulation. Particulate fractions are assumed to follow TSS distribution as in the SWMM simulation | SWMM engine, calibrated TSS sub-model (LyRE, Maruejouls 2018) | | |

# Annex C – Supplementary information for Chapter 4

# Annex C-1. LCI of infrastructure materials and processes

## *Transport of raw materials for the construction of infrastructure*

| **Materials** | **processes included** | | | | |
|---|---|---|---|---|---|
| | transport, from manufacturing plant to construction site | | transport, from construction site to disposal | End of life | |
| Cast iron | freight train (600 tkm) | freight lorry (50 tkm) | freight lorry (30 tkm) | 95% - recycling into pig iron (using this process: Iron scrap, sorted, pressed {GLO}\| market for \| Alloc Rec, U) | 5% -Waste bulk iron, excluding reinforcement {CH}\| treatment of, sorting plant \| Cut-off, U |
| Steel, unalloyed | freight train (200 tkm) | freight lorry (50 tkm) | freight lorry (30 tkm) | 95% - recycling into pig iron (using this process: Iron scrap, sorted, pressed {GLO}\| market for \| Alloc Rec, U) | 5% - Waste reinforcement steel {CH}\| treatment of, sorting plant \| Alloc Rec, U |
| Steel, chromium steel 18/8 | freight train (200 tkm) | freight lorry (50 tkm) | freight lorry (30 tkm) | 95% - recycling into pig iron (using this process: Iron scrap, sorted, pressed {GLO}\| market for \| Alloc Rec, U) | 5% - Waste reinforcement steel {CH}\| treatment of, sorting plant \| Alloc Rec, U |
| Copper | freight train (200 tkm) | freight lorry (50 tkm) | freight lorry (30 tkm) | 60% - recycling into copper (using this process: Copper {RER}\| treatment of scrap by electrolytic refining \| Alloc Rec, U) | 40% - Scrap copper {CH}\| treatment of, municipal incineration \| Alloc Rec, U |
| Concrete (lean concrete) | | freight lorry (20 tkm) | freight lorry to recycling site (30 tkm), to incinerator (50km) | 100% - Waste concrete, not reinforced {CH}\| treatment of, collection for final disposal \| Alloc Rec, U | |

## *Retention tanks: construction*

| Hypotheses – material estimation for retention tank basins | | | Source | Comments |
|---|---|---|---|---|
| Life time | 100.00 | years | | |
| Material: ratio material amount - basin volume | 0.15 | - | | |
| Material: ratio concrete - material amount | 0.97 | - | | |
| Material: ratio steel - material amount | 0.03 | - | Brudler 2016 | |
| Construction: ratio exc. volume - basin volume | 1.10 | - | | Estimation of excavated soil |
| Construction: ratio disposed soil - exc. volume | 0.90 | - | | Estimation of disposed soil |
| Decommissioning: ratio excavation volume - basin volume | 1.00 | - | | |

| Process | Unit | Storage unit capacity (m3) | | |
|---|---|---|---|---|
| | | 4000 | 10000 | 20000 |
| Material amount estimation | m3 | 600 | 1500 | 3000 |
| Material: concrete production | m3 | 582 | 1455 | 2910 |
| | ton | 1368 | 3419 | 6839 |
| Material: steel production | ton | 180 | 450 | 900 |
| Construction: excavation | m3 | 4.40E+03 | 1.10E+04 | 2.20E+04 |
| Construction: disposal of soil | kg | 5.71E+06 | 1.43E+07 | 2.86E+07 |
| Decommissioning: excavation | m3 | 4000 | 10000 | 20000 |

| **Materials (density)** | | |
|---|---|---|
| soil | 1441.96 | kg/m3 |
| concrete (normal) | 2350.00 | kg/m3 |
| steel | 10000.00 | kg/m3 |
| PE (high density) | 950.03 | kg/m3 |
| gravel | 1522.07002 | kg/m3 |
| granite | 2691.07 | kg/m3 |
| clay | 1600.00 | kg/m3 |
| asphalt | 2243.16 | kg/m3 |
| bitumen | 1000.00 | kg/m3 |
| thermobeton | 275.00 | kg/m3 |

## *Pumping group: construction*

| Hypotheses - Material estimations for a pumping group | |
|---|---|
| Life time (years) | 15.00 |
| Material: ratio cast iron - material amount | 0.50 |
| Material: ratio steel - material amount | 0.15 |
| Material: ratio steel - material amount | 0.15 |
| Material: ratio copper - material amount | 0.20 |

| Material estimation (hypotheses) | Unit | Power (kW) | | |
|---|---|---|---|---|
| | | 40 | 400 | 1200 |
| Cast iron | kg | 7.46E+02 | 1.14E+03 | 3.41E+03 |
| Steel, unalloyed | kg | 2.24E+02 | 3.41E+02 | 1.02E+03 |
| Steel, chromium steel 18/8 | kg | 2.24E+02 | 3.41E+02 | 1.02E+03 |
| Copper | kg | 2.98E+02 | 4.55E+02 | 1.37E+03 |
| ***Total weight*** | ***kg*** | ***1.492E+03*** | ***2.28E+03*** | ***6.83E+03*** |

## *Sewer network: extraction of deposits*

| Deposit removal from sewer network | Ecoinvent material inputs technosphere | |
|---|---|---|
| Vacuum tanker | Lorry, 32 T | 11-16m3 capacity |
| http://www.huwer.com/equipements-huwer/assainissement/hydrocureur-mixteo-32t | Diesel consumption | 15L/h |

| *End of life for sewer deposits* | | |
|---|---|---|
| Transport to sorting facility | 20 | km |
| Valorization product as road sub layer | 20% | |
| Waste, to landfill for inert materials | 80% | |

# Annex C-2. Detailed mass repartition of pollutants in destination flows of the UWS for one year of rainfall using the SWMM engine calibrated on total suspended solids (SS).

*Mass repartition of pollutants in* ***wastewater influent Inf*** *with fractionation (Xp, Xd)*

| | Baseline | A | B0 | B1 | C0 | C1 | C2 |
|---|---|---|---|---|---|---|---|
| In combined sewer overflow discharges, CSOp | | | | | | | |
| NH4-N | 11% | 7% | 6% | 5% | 5% | 5% | 4% |
| Org-N | 11% | 8% | 6% | 6% | 6% | 5% | 4% |
| PO4-P | 9% | 9% | 8% | 7% | 7% | 7% | 5% |
| Org-P | 11% | 8% | 7% | 6% | 6% | 5% | 4% |
| Cu | 11% | 7% | 6% | 5% | 5% | 5% | 4% |
| Zn | 11% | 7% | 6% | 5% | 5% | 5% | 4% |
| PAH | 11% | 7% | 6% | 5% | 5% | 5% | 3% |
| *average* | *11%* | *8%* | *6%* | *6%* | *6%* | *5%* | *4%* |
| In flows returned to treatment in WWTP, WTPp | | | | | | | |
| NH4-N | 85% | 83% | 84% | 85% | 84% | 84% | 84% |
| Org-N | 86% | 85% | 86% | 86% | 86% | 85% | 86% |
| PO4-P | 91% | 91% | 92% | 93% | 93% | 93% | 95% |
| Org-P | 86% | 85% | 86% | 87% | 86% | 86% | 86% |
| Cu | 85% | 83% | 84% | 85% | 84% | 83% | 83% |
| Zn | 85% | 83% | 84% | 85% | 84% | 83% | 83% |
| PAH | 84% | 82% | 83% | 84% | 83% | 82% | 82% |
| *average* | *86%* | *85%* | *86%* | *86%* | *86%* | *85%* | *86%* |
| In deposits within the UWS, DEPp | | | | | | | |
| NH4-N | 4.4% | 9% | 9% | 10% | 11% | 12% | 13% |
| Org-N | 3.7% | 8% | 8% | 8% | 9% | 10% | 11% |
| PO4-P | 0% | 0% | 0% | 0% | 0% | 0% | 0% |
| Org-P | 3.4% | 7% | 7% | 7% | 8% | 9% | 10% |
| Cu | 4.6% | 10% | 10% | 10% | 11% | 12% | 13% |
| Zn | 4.6% | 10% | 10% | 10% | 11% | 12% | 13% |
| PAH | 5.1% | 11% | 11% | 11% | 12% | 13% | 15% |
| *average* | *4%* | *8%* | *8%* | *8%* | *9%* | *10%* | *11%* |

## Annex C-3. Full contribution analyses for Baseline and Scenario A

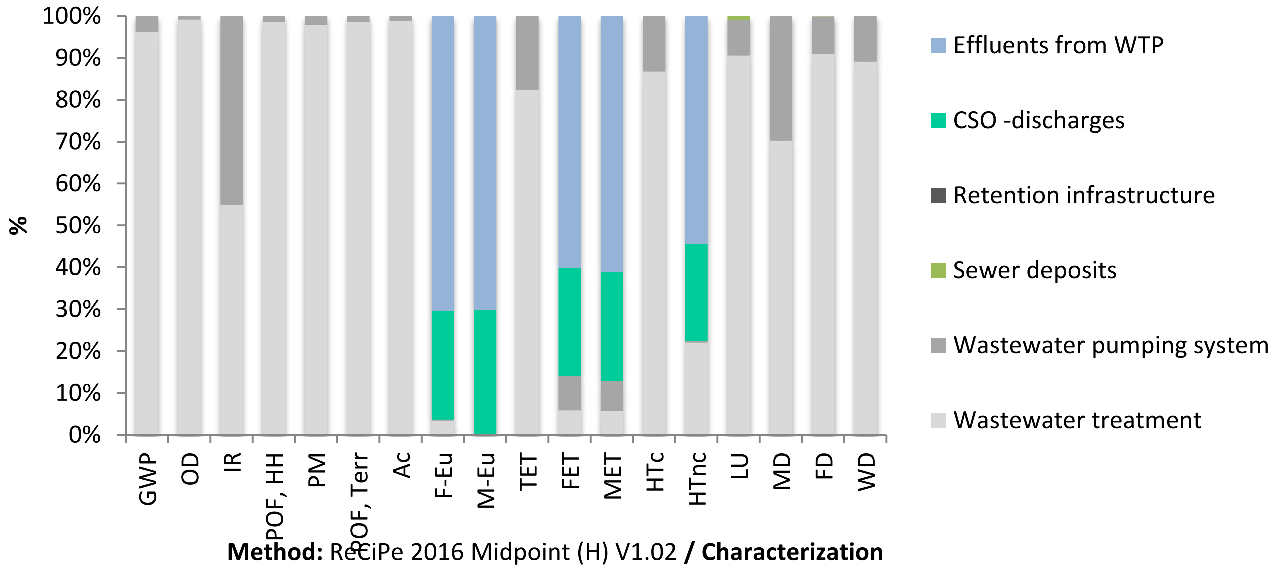


*Figure S13. Baseline scenario midpoint results(ReCiPe 2016 Hierarchist)*

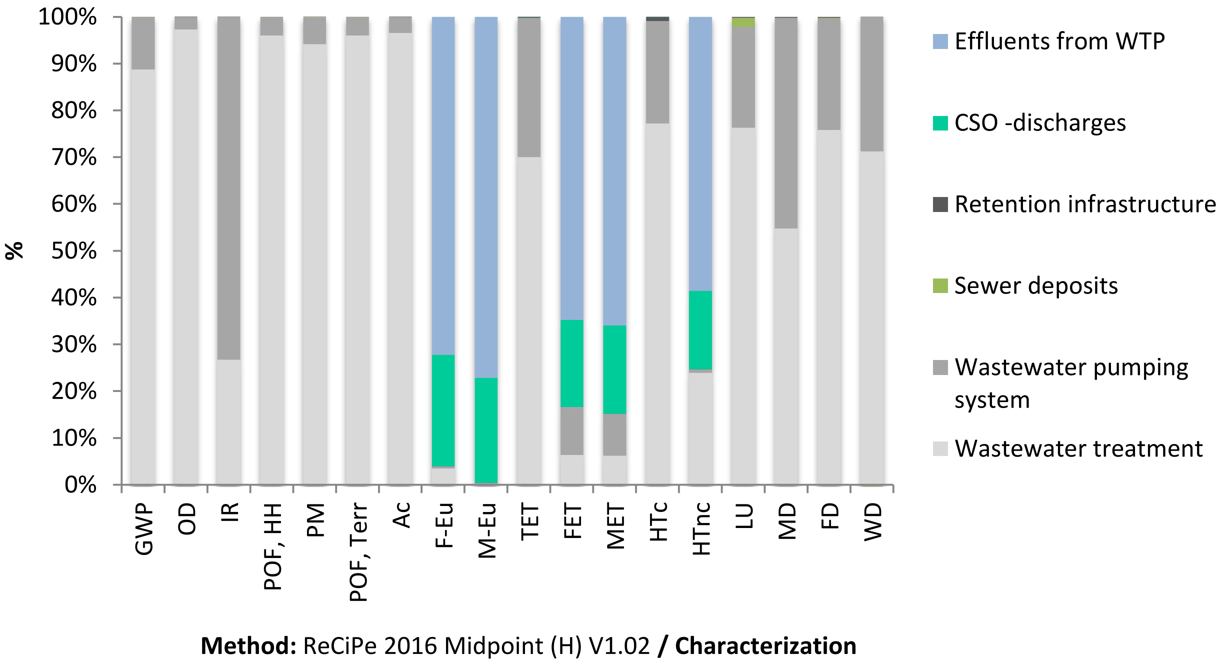


*Figure S14. Scenario A midpoint results(ReCiPe 2016 Hierarchist)*

# French extended abstract - Résumé étendu en Français

## Chapitre 1. Contexte et objectifs généraux

### *Métabolisme des systèmes d'assainissement urbains*

Selon la théorie générale des systèmes, l'essence de la vie est le processus. Par conséquent, les processus nécessitent de l'énergie pour fonctionner. L'énergie est exploitée et manipulée dans des systèmes à diverses échelles, allant des cellules aux écosystèmes, ou des batteries à des industries entières. Au XIXe siècle, en étudiant l'organisation sociale du travail humain, Marx et Engels ont émis l'hypothèse que l'exploitation des ressources de la Terre par les humains pour se nourrir et s'abriter est en soi un processus métabolique qui génère et soutient la civilisation. En 1965, Abel Wolman a inventé le terme métabolisme des villes, apportant une contribution historique au domaine de l'écologie industrielle. Dans son travail de pionnier, une ville moderne est semblable à un organisme vivant avec des besoins métaboliques pour assurer la subsistance de ses habitants à la maison, au travail et dans les loisirs. Dans son modèle métabolique pour une ville américaine les eaux usées sont la composante la plus importante, surpassant de loin les besoins en nourriture, carburant, charbon et gaz naturel. Cependant, il y a un autre produit métabolique des villes : le ruissellement des eaux pluviales, qui est tout aussi important en tant que vecteur de polluants urbains.

En 2018, environ 55% de la population mondiale vit dans des zones urbaines et cette proportion devrait augmenter pour atteindre environ les deux tiers d'ici 2050 (Nations Unies, 2018). En outre, il est certain que les régimes de précipitations changeront à l'avenir en raison du changement climatique anthropique (GIEC 2014). Du fait des effets individuels et combinés de ces deux facteurs, les zones urbaines où les précipitations sont actuellement plus abondantes connaîtront des précipitations de plus en plus intenses et des volumes d'eaux pluviales ruisselées plus élevés. La croissance des zones urbaines et l'intensification de l'utilisation des sols urbains augmentent la conversion des précipitations en ruissellement.

L'urbanisation entraîne également une plus grande abondance de sources de polluants qui, si l'on tient compte de l'augmentation du ruissellement, augmenteront les charges polluantes transportées par le ruissellement des eaux pluviales, ce qui affectera négativement les eaux réceptrices (Goonetilleke et al., 2014 ; Marsalek et al., 2006). En effet, les villes sont des foyers de sources diffuses en raison des activités urbaines (trafic routier et autres processus de combustion) et d'une masse importante et généralement inconnue de substances stockées dans la technosphère sur le long terme (par ex. bâtiments, infrastructures) (Brunner et al., 2001). En outre, de nombreuses zones urbaines sont confrontées à des problèmes causés par le vieillissement des infrastructures de drainage, qui pourraient nécessiter d'importants travaux d'entretien ou de reconstruction dans un proche avenir.

La quantification de tous les impacts environnementaux associés aux bassins versants urbains et à leurs systèmes de traitement des eaux usées urbaines est une condition nécessaire pour réduire leur empreinte. Parmi les outils disponibles pour évaluer les impacts environnementaux de ces systèmes, l'analyse du cycle de vie (ACV) a déjà fait ses preuves. La méthode ACV est expliquée dans la section suivante en soulignant brièvement ses principales forces et ses limites pour l'évaluation des systèmes d'eau urbains.

## *Introduction à l'Analyse du Cycle de Vie*

Pour appréhender l'ampleur des problèmes environnementaux causés par les activités humaines, il est nécessaire de se munir d'outils permettant de quantifier leurs impacts. La méthode ACV permet d'évaluer ces pressions et les impacts environnementaux associés en analysant les ressources extraites et les émissions générées par un produit ou un service sur l'ensemble de son cycle de vie. A la différence d'autres outils d'évaluation environnementale (par exemple, l'empreinte carbone ou le bilan énergétique), l'ACV est une approche multicritère qui prend en compte toutes les étapes du « cycle de vie » d'un bien ou d'un service. Ce caractère holistique de l'ACV permet d'identifier les transferts de pollution entre catégories d'impacts, entre étapes du cycle de vie et/ou entre lieux géographiques. L'ACV est maintenant un outil précieux pour étudier les impacts environnementaux des systèmes d'eau urbains à l'échelle de la ville sur l'ensemble de leur cycle de vie, et pour éclairer les décisions d'éco-conception et sur des aspects opérationnel de ces systèmes.

Les modèles ACV de systèmes d'eau urbains devraient être alignés sur les principaux objectifs de ces systèmes (d'eau urbains), qui sont la gestion optimisée de la quantité et la qualité de l'eau tout en visant la préservation de la santé humaine, la qualité des écosystèmes et les ressources naturelles. Pour développer cet effort, il faudrait envisager de nouvelles approches qui tiennent compte des considérations spatiales, de la quantité d'eau, de la santé humaine et de facteurs économiques et sociaux afin de mieux éclairer la prise de décisions.

### *Limites identifiées*

Bien que l'urbanisation croissante et l'augmentation du ruissellement des eaux pluviales contribuent significativement à la dégradation de la qualité des écosystèmes aquatiques, les recherches dans la communauté ACV sur les impacts des eaux pluviales urbaines sont rares. La majorité des travaux ACV sur les eaux pluviales ont porté sur l'aspect quantitatif (réduction du volume des eaux de ruissellement) de mesures spécifiques de contrôle des eaux pluviales. Il reste donc plusieurs défis à relever dans l'évaluation des impacts des rejets urbains par temps de pluie (RUTP), pour lesquels il n'existe pas encore de flux d'inventaire (ICV). Entre autres, les principaux défis identifiés concernant l'étape d’ICV sont la prise en compte de la variabilité spatiale et temporelle des RUTP, due aux conditions climatiques, aux caractéristiques des bassins versants urbains (par exemple, l'utilisation des sols urbains et les activités humaines) et aux types de système urbain d’eau (définis par une infrastructure et une gestion).

### *Objectifs et organisation de la thèse*

Dans le but d’analyser avec une perspective systémique pour améliorer l’évaluation environnementale des systèmes d’assainissement urbains avec l’ACV, d’un côté en prenant en compte les impacts dus à l’opération et à l’infrastructure de ces systèmes, et de l’autre côté les impacts des polluants rejetés, la problématique générale de recherche est définie comme suit:

« Est-ce qu’il est possible de réaliser l’ACV de systèmes d’assainissement urbains en considérant la variabilité temporelle des rejets aquatiques d’un bassin versant urbain? »

Trois questions de recherche sous-jacentes s’inscrivent dans cette problématique générale. Chacune sera traitée par un chapitre de la thèse faisant référence à une publication scientifique (en cours de publication ou en préparation):

- Question de recherche 1: Est-ce que les RUTP intermittents ont des impacts significatifs par rapport aux impacts des rejets continus (de temps sec), à l’échelle du bassin versant urbain ?

- Question de recherche 2: Comment quantifier, en vue d’établir un inventaire ICV, les rejets intermittents d’un bassin versant urbain en tenant compte des activités urbaines, du climat et de l’occupation des sols?

- Question de recherche 3: Comment les impacts induits par les infrastructures de gestion des eaux pluviales se comparent-ils aux impacts qu’elles évitent grâce au traitement des rejets intermittents?

# Chapitre 2. Impacts des systèmes d’assainissement urbains : Comment prendre en considération les évènements pluvieux marquants en ACV?

## *Contexte et objectifs*

Avec un taux d'urbanisation toujours croissant à l'échelle planétaire, la gestion des eaux pluviales qui sont de plus en plus importantes, polluées et accompagnées de risques d’inondation représente un défi considérable. Les réseaux d’assainissement assurent le drainage, ce qui protège la santé publique, empêche l'inondation des propriétés et protège l'environnement aquatique autour des zones urbaines. Par temps sec, un réseau d’assainissement combiné (ou unitaire) des eaux usées, qui achemine les eaux usées vers la station d'épuration, fonctionne efficacement. Cependant, lors d'un orage violent, les eaux pluviales rejoignent le système de collecte des eaux usées. Cela provoque parfois une surcharge du système de collecte, et une fraction d’eaux usées brutes non traitées débordent au niveau des déversoirs d’orage (DO), avec des répercussions sur les écosystèmes aquatiques récepteurs. Le programme de recherche de l'Observatoire des polluants urbains (OPUR) a abouti à une meilleure compréhension de la contamination par les polluants prioritaires (au titre de la Directive Cadre Eau) des flux urbains dans les eaux réceptrices dans la région Parisienne. L'étude menée dans ce Chapitre a pour but d’évaluer l’importance des impacts associés aux RUTP par rapport aux rejets dans des conditions moyennes « continues » par temps sec, au niveau du bassin versant du Grand Paris.

## *Méthode*

À l'aide de la méthodologie ACV, les polluants rejetés sont quantifiés en termes d’inventaires (charges de polluants) et en termes d’impacts potentiels sur les eaux réceptrices. Les impacts potentiels seront évalués pour trois catégories concernant la qualité des milieux aquatiques, à savoir l'eutrophisation (d'eau douce et marine) et l'écotoxicité en eau douce. Les méthodes d’évaluation des impacts de cycle de vie (EICV) utilisées sont ReCiPe et USEtox v2.0.

La portée de l'étude couvre les rejets générés par un bassin versant urbain avec des réseaux d'assainissement de type unitaires et séparatifs, à deux échelles temporelles, (i) pendant un an, et (ii) pendant les épisodes de temps de pluie. La démarche suivie pour obtenir des inventaires dans les rejets est illustrée à la Figure 27.

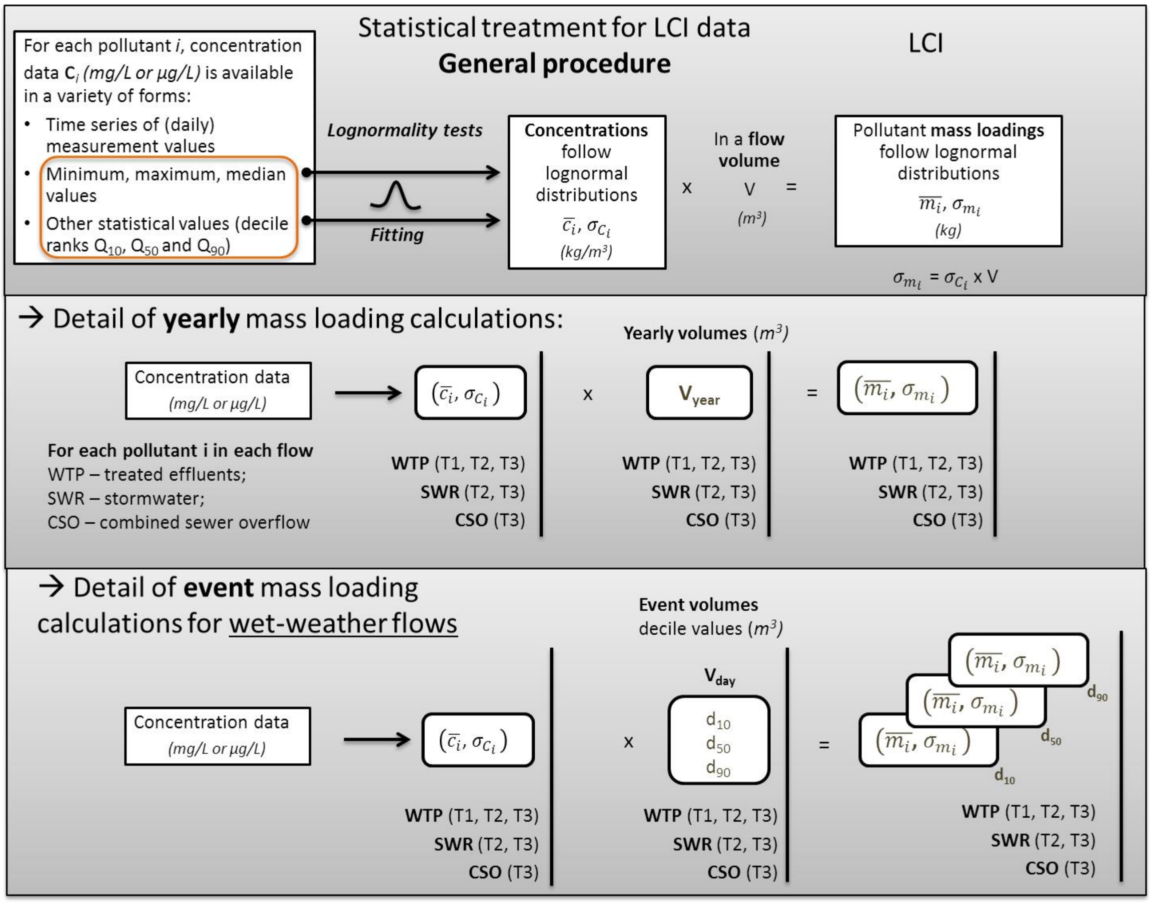


*Figure 27. Traitement statistique des données pour calculer les flux d'inventaire des polluants présents dans les effluents (WTP, eaux usées traitées), les eaux pluviales (SWR) et les rejets des DO (CSO).*

Au total, 43 polluants prioritaires sont inclus dans les inventaires des rejets des systèmes d'assainissement urbains (SAU) sur les 58 pour lesquels des données d'occurrence et des facteurs de caractérisation en écotoxicité en eau douce dans la méthode USEtox v2.01 sont disponibles.

## *Résultats et discussion*

Cette étude a permis de mettre en évidence que la qualité des données sur les rejets urbains est très hétérogène, ce qui a nécéssité une approche probabiliste pour obtenir un inventaire cohérent pour tous les rejets des SAU. Davantage d'efforts de recherche sont nécessaires pour mieux caractériser les rejets des DO (seulement quatre événements ont été suivis pour l'étude de cas) en termes de polluants prioritaires. La classification des événements par temps de pluie en 3 catégories (T1, temps sec; T2, temps de pluie sans rejets de DO; T3, temps de pluie avec rejets de DO) permet de mieux définir les RUTP et souligne l'importance de leurs pollutions par rapport aux rejets par temps sec d'un SAU à l'échelle d'une journée (Figure 28).

A l'échelle annuelle, les impacts potentiels des RUTP sont également significatifs à l'échelle annuelle, malgré la gestion des eaux pluviales (Figure 29).

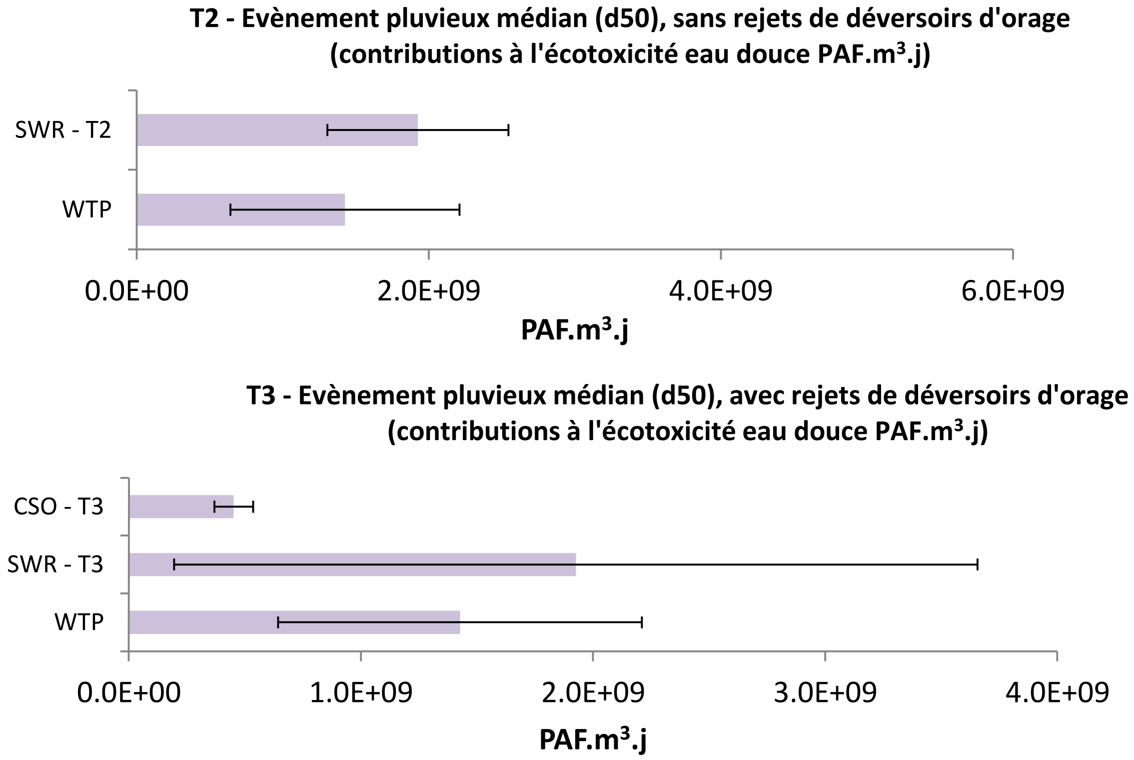


*Figure 28. Contributions à l'écotoxicité en eau douce (PAF.m3.j) des rejets urbains de temps sec et de pluie à l'échelle d'un évènement de pluie (d50, volumes médians). CSO, rejets de déversoirs d'orage; SWR, eaux pluviales non traitées; WTP, effluents de station de traitement des eaux usées.*

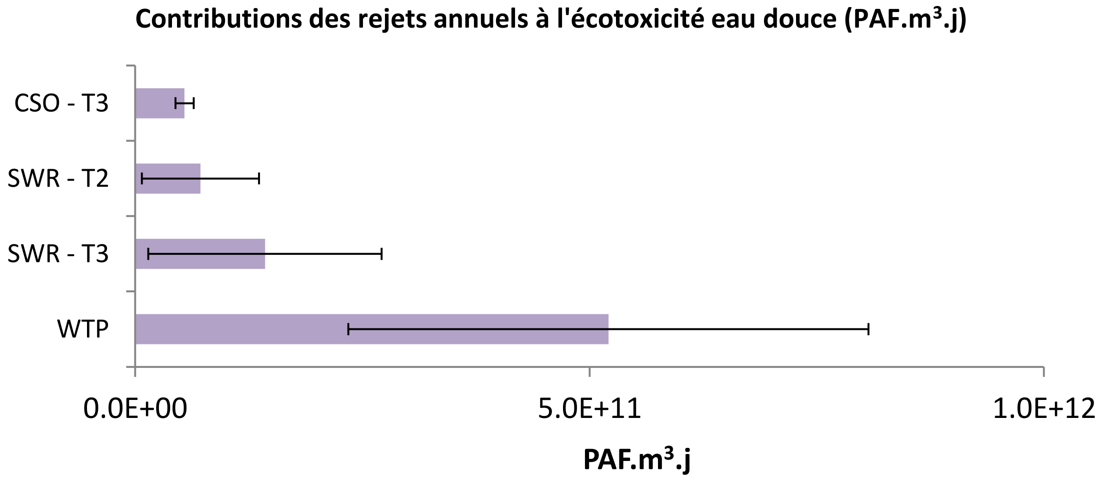


*Figure 29. Contributions à l'écotoxicité en eau douce (PAF.m3.j) des rejets urbains de temps sec et de pluie à l'échelle annuelle. CSO, rejets de déversoirs d'orage; SWR, eaux pluviales non traitées; WTP, effluents de station de traitement des eaux usées.*

La priorisation des contributeurs sur l'écotoxicité montre un effet de masque des deux éléments métalliques (Cu(II) et Zn(II)), qui dominent avec plus de 99 % du score d'écotoxicité, dans tous les rejets considérés. Ensuite, au regard de ces éléments métalliques, le 4-nonylphénol dans les effluents traités, et le pyrène dans les RUTP paraissent avoir des contributions peu importantes.

Compte tenu de l'importance des impacts des RUTP, il faut poursuivre les recherches sur la façon d'inclure la variabilité temporelle de ces rejets dans le cadre de l’ACV afin de mieux comprendre comment la performance d'un SAU affecte le milieu récepteur selon les conditions météorologiques locales. L'approche ACV est bien adaptée à une évaluation comparative des infrastructures par temps de pluie dans les réseaux d'eau urbains, en utilisant des modèles de systèmes de drainage basés sur la physique couplés à un inventaire des RUTP pour évaluer les changements des impacts à l'échelle des événements pour différentes catégories. Toutefois, la considération des effets de toxicité aiguë à la suite d'un pic de pollution au début d'un évènement pluvieux demeure un défi sur le plan méthodologique EICV.

## Chapitre 3. Un modèle parcimonieux des influences des activités humaines, de l’occupation des sols urbains et du climat sur l’estimation des ICV de charges polluantes des rejets intermittents des bassins versants urbains aux milieux aquatiques récepteurs en fonction du site

### *Objectifs du Chapitre*

Le Chapitre 2 a identifié plusieurs défis méthodologiques pour modéliser les rejets intermittents, qui ont une variabilité spatio-temporelle très marquée, en termes de qualité et de quantité dans les études ACV des systèmes d’assainissement urbains (SAU). Le manque de considération de la dynamique temporelle est une limite reconnue de l’ACV, notamment pour la quantification des impacts sur l'écotoxicité et l'eutrophisation qui sont sensibles à la période et au mode d’émission des polluants. Il est donc nécessaire de remédier à cette limite, en permettant la description de la dynamique temporelle des rejets des SAU. Par conséquent, il est fondamental de prendre en compte les rejets urbains de temps de pluie (RUTP) et leur variabilité spatio-temporelle, en complément des rejets continus de temps sec dans l'étape d'inventaire d’analyse de cycle de vie (ICV) afin de comparer en détail les répercussions environnementales des stratégies de gestion actuelles et futures des SAU sur la durée de vie de leurs infrastructures. Le modèle, nommé STOIC pour « STOrmwater Inventory Conceptualization », a pour but de résoudre les questions méthodologiques identifiées dans le Chapitre 2, et vise à permettre l’élaboration d’ACV comparatives pour différents scénarios de gestion des RUTP, avec ou sans infrastructures supplémentaires (par exemple pour augmenter la rétention dans les réseaux combinés). Finalement, ces comparaisons se feront sur la base des impacts environnementaux nets qui résultent (i) des impacts induits par l'exploitation et la construction des infrastructures des SAU et (ii) des impacts évités par le traitement de la pollution dans les RUTP. Ces comparaisons seront des éléments précieux pour établir des recommendations pour l'aide à la prise de décision dans le domaine des SAU

## *Méthode*

L'hétérogénéité des activités urbaines d'un bassin versant est liée aux différents usages (ou occupations) des sols. Pour capturer la dimension spatiale de la variabilité des charges de polluants exportées en aval d'un bassin versant, il est fondamental de relier les charges de polluants dans les eaux pluviales aux activités urbaines qui sont sources diffuses de polluants urbains. Ces sources diffuses de polluants urbains sont ensuite déterminées pour une typologie de zones urbaines, déjà développée et utilisée dans les analyses territoriales urbaines. Quant à la dimension temporelle de la variabilité des charges de polluants, elle peut être capturée par la considération de la dynamique des processus d'accumulation et de transport des polluants dans le bassin versant urbain et son système d'assainissement. Les polluants inclus dans le modèle STOIC sont le cuivre, le zinc et les hydrocarbures aromatiques polycyliques (HAP), qui contribuent à eux seuls à la quasi-totalité des impacts en écotoxicité dans les RUTP (Chapitre 2).

Le modèle STOIC proposé dans ce Chapitre (Figure 30) repose sur deux sous-modèles pour construire des ICV spatio-temporels pour les polluants dans les rejets d'un bassin versant urbain. Le premier sous-modèle (1) décrit les émissions de sources diffuses et leur transfert dans les eaux pluviales, et le second sous-modèle (2) détaille la distribution de la pollution dans les quatre composantes du système d'assainissement urbain et la dynamique temporelle des émissions de polluants. Ces quatres composantes sont les dépôts accumulés dans le réseau (DEP), les rejets de déversoirs d'orage (CSO), les rejets des effluents traités des stations de traitement des eaux usées (WTP), et les rejets des eaux pluviales non traitées (SWR).

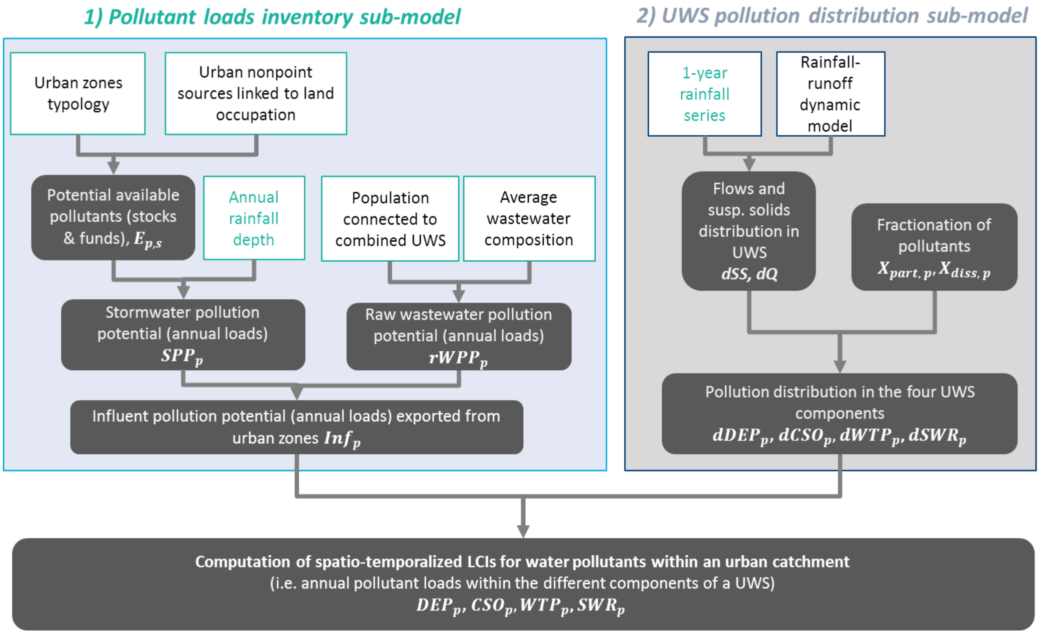


*Figure 30. Modèle STOIC basé sur deux sous-modèles pour construire des ICV spatio-temporels pour les polluants dans les rejets d'un bassin versant urbain, depuis les émissions de sources diffuses jusqu'à l'acheminement et la distribution de la pollution dans les quatre composantes du système d'assainissement urbain (par exemple, dépôts accumulés dans le réseau (DEP), rejets de déversoirs d'orage (CSO), rejets des effluents traités des stations de traitement des eaux usées (WTP), rejets des eaux de pluie non traités (SWR)).*

Pour un bassin versant générique, le sous-modèle de génération de polluants (1) propose de considérer trois principales voies d'entrée pour les polluants retenus (Cu, Zn et HAP), et la question de la pertinence de modéliser la dynamique dans les processus de génération des polluants est soulevée dans ce Chapitre (Figure 31).

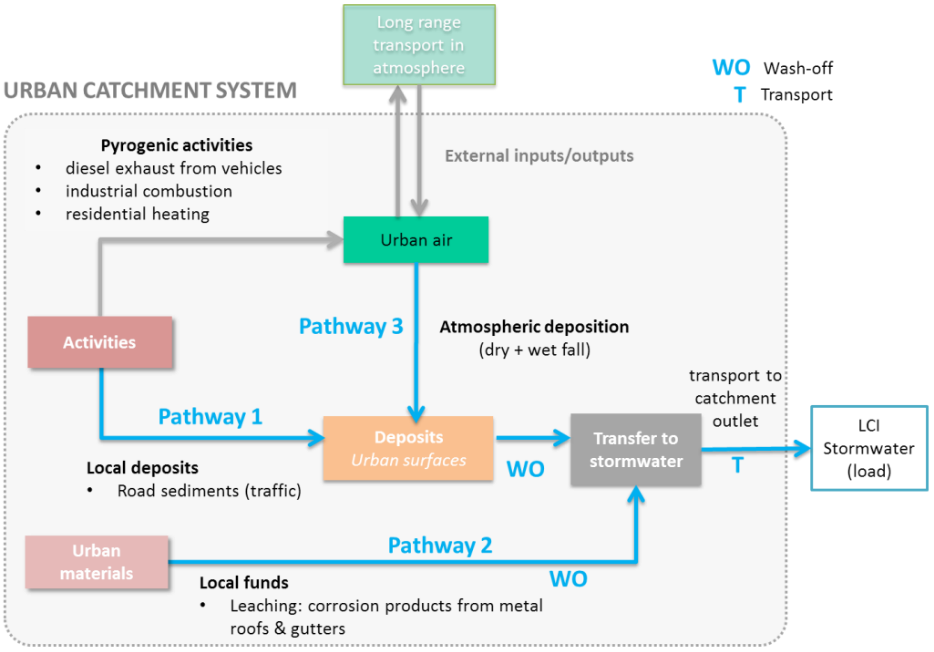


*Figure 31. Modélisation des voies d'entrée pour les polluants des eaux pluviales provenant de sources diffuses urbaines (en bleu, voies considérées dans le cadre conceptuel; en gris, voies non incluses)*

### *Discussion et conclusions*

Le travail réalisé dans ce Chapitre a permis de dégager trois conclusions importantes : (i) la dynamique des processus de génération des polluants sur le bassin versant urbain n’est pas nécessaire pour atteindre l'objectif défini qui est de prévoir les charges annuelles d'eaux pluviales; (ii) il est possible de caractériser les eaux pluviales en fonction du site (par exemple, différentes charges estimées pour les zones denses et compactes du centre-ville, ou les zones péri-urbaines éparses); et (iii) pour trois polluants (Cu, Zn et les HAP), les principaux processus de génération peuvent être identifiés.

En conclusion, bien que la prédiction des pollutions des RUTP d’un bassin versant urbain soit un domaine de recherche actif du fait de leur complexité et variabilité, le modèle STOIC fournit une base solide pour estimer les ICV spatiotemporels des rejets de SAU pour un bassin versant urbain à une échelle temporelle suffisante pour décrire les variations climatiques des événements pluvieux communs (i.e. fréquents). Par conséquent, les impacts causés par ces RUTP aux milieux aquatiques récepteurs peuvent être évalués à l'aide de méthodes d'ÉICV classiques ou dynamiques.

Enfin, l'application du modèle STOIC à une étude de cas réel servirait de preuve de concept pour démontrer sa faisabilité, et enrichir la discussion sur sa mise en œuvre et les résultats opérationnels.

## Chapitre 4. Perspectives de Cycle de vie pour la gestion des rejets intermittents des systèmes d’assainissement urbains : enseignements tirés d’une étude de cas réel sur un sous-bassin versant Français

### *Objectifs du Chapitre*

Le modèle STOIC est appliqué à un cas d’étude: le système d’assainissement urbain (SAU) de Lauzun de type combiné (ou unitaire), situé dans un sous-bassin versant de la Métropole de Bordeaux, en France. Ce cas d’étude vise à vérifier la capacité du modèle à (i) évaluer les impacts environnementaux du SAU; (ii) comparer différentes stratégies de gestion des RUTP en termes d’impacts environnementaux nets ; et (iii) à discuter de l’applicabilité du modèle pour les ACV de SAU au niveau du bassin versant en prenant en compte les conditions climatiques locales.

## *Matériel et méthode*

La portée de cette étude d'ACV comprend les processus des trois sous-systèmes qui composent le SAU : station de traitement des eaux usées, réseau d’assainissement combiné, et infrastructures de rétention (Figure 32).

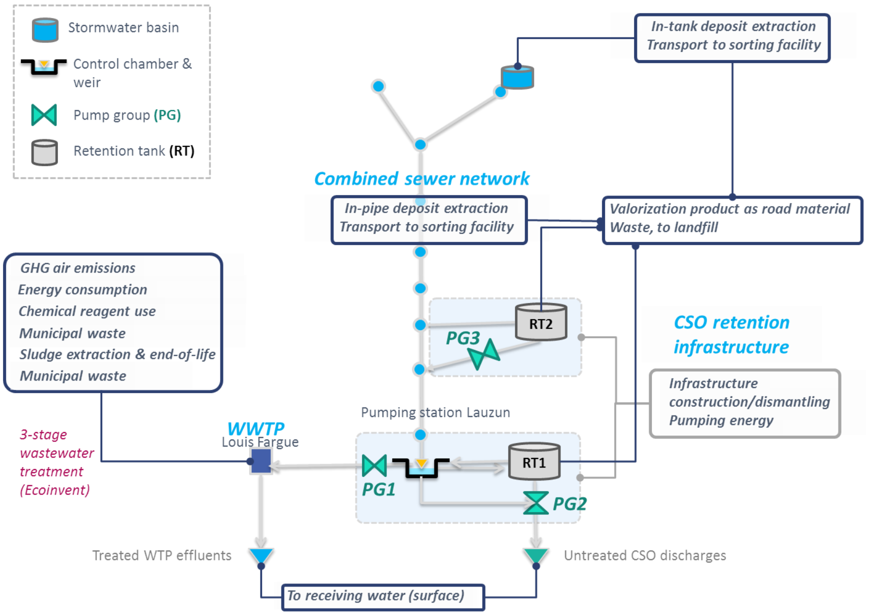


*Figure 32. Limites du système d’assainissement urbain de Lauzun, indiquant les processus modélisés tout au long des étapes du cycle de vie (construction, exploitation et fin de vie) des trois sous-systèmes (station de traitement des eaux usées, réseau d’assainissement combiné, et infrastructures de rétention).*

Des scénarios représentant les stratégies de gestion des RUTP, sont définis pour le SAU étudié et sont comparés sur la base d’un flux de référence, qui est défini comme l’aire urbaine et le SAU dans les conditions climatiques locales pendant une période d’un an. Quatre classes de scénarios sont proposées dont :

i. un scénario de référence où la capacité de rétention du SAU est nulle ;
ii. un scénario représentant la stratégie actuelle;
iii. une classe de scénarios avec des changements non structurels ;
iv. une classe de scénarios avec changements structurels (bassin de rétention supplémentaire).

La méthode ReCiPe 2016 (v1.02) a été choisie comme modèle de caractérisation des impacts pour son cadre harmonisé de calcul des facteurs de caractérisation (midpoint et endpoint). Afin de comparer et de discuter les résultats EICV sur la toxicité et l'écotoxicité de ReCiPe 2016, le modèle USEtox 2.01 est également retenu pour effectuer une analyse ciblée des polluants. Les deux modèles partagent la même base de données sur les substances qui comprend 3 073 produits chimiques organiques et 20 métaux (essentiels), développée dans le modèle USEtox.

## *Résultats et discussion*

Les impacts de l'exploitation du SAU (notamment dûs au traitement des eaux usées en station d'épuration) sont globalement plus importants que ceux de la construction d'infrastructures de rétention, sauf pour les catégories d'impact locales concernant la qualité des eaux. (eutrophisation et écotoxicité). Les deux éléments métalliques (Cu et Zn) contribuent de loin à l'écotoxicité et à la toxicité humaine, devant les HAP. Ces résultats contrastent avec la hiérarchisation des polluants prioritaires de la Directive Cadre Eau, le cuivre et le zinc n'étant pas considérés comme polluants prioritaires dangereux. La fiabilité des methods d'EICV sur l'évaluation de la toxicité et écotoxicité des métaux est discutée

## *Conclusions*

Sur la base de cette étude de cas, les apports et les limites du modèle STOIC sont identifiés. L'évaluation environnementale du SAU d'un basin versant urbain avec une perspective systémique est rendue possible par la consideration des impacts dus à l'opération et à l'infrastructure de ces systèmes, et de l'autre côté les impacts des polluants rejetés, dont les rejets urbains de temps de pluie. Cette application de l'ACV a élargi la portée de l'évaluation, avec l'identification des transferts de pollution dans une perspective de cycle de vie : entre sous-systèmes ou entre catégories d'impact.

Les performances globales des différentes stratégies de contrôle des rejets urbains par temps de pluie ont été comparées en termes d'impacts environnementaux, révélant de petites différences entre les stratégies comparées. L'étude a révélé que les pressions exercées sur les écosystèmes aquatiques locaux étaient importantes, même dans la perspective plus large du cycle de vie. Des inventaires de cycle de vie avec différenciation spatio-temporelle pour les rejets urbains de temps de pluie ont été calculés pour cette étude de cas, ce qui démontre l'applicabilité du cadre STOIC pour inclure la variabilité temporelle des ID d'UWS dans un sous-bassin versant. Toutefois, des travaux supplémentaires sont nécessaires pour rationaliser la mise en œuvre pratique du cadre STOIC pour une variété de bassins versants.

# Chapitre 5. Conclusion générale

Ce travail de thèse a abouti à plusieurs résultats aussi bien d'ordre méthodologique que pratique. Tout d'abord, les principaux challenges méthodologiques liés à la considération de la variabilité spatio-temporelle des rejets intermittents de temps de pluie pour les ACV de systèmes d'assainissement urbains (SAU) ont été identifiés, et des solutions concrètes ont été apportées pour faire face à ces problèmes. En effet, l'importance de la pollution des rejets intermittents a été soulignée par rapport aux rejets continus de temps sec sur les catégories d'impact concernant la qualité de l'eau (eutrophisation et écotoxicité) à l'échelle de l'évènement et à l'échelle annuelle. Les principaux contributeurs à l'écotoxicité ont été identifiés, parmi lesquels les métaux (Cu, Zn), les HAP et les alkylphénols.

Suite à cette première conclusion, le modèle STOIC a été défini pour relier les caractéristiques du bassin versant urbain et conditions climatiques aux charges de polluants des rejets intermittents. Cette conceptualisation propose de considérer la variabilité spatiale des sources de polluants dans le bassin versant, et la variabilité temporelle des processus de génération des polluants et de transport dans le SAU. L'ICV spatio-temporel différencié des rejets intermittents des UWS qui en résulte peut ensuite servir de donnée d'entrée à une ACV classique ou dynamique pour évaluer les stratégies de gestion par temps de pluie. .

Enfin, l'applicabilité du modèle STOIC a été démontrée sur une étude de cas réel, en évaluant pour le SAU sous les conditions climatiques étudiées, les performances globales de différentes stratégies de gestion du temps de pluie en termes d'impacts environnementaux nets. L'objectif général de la thèse, qui était de comprendre comment prendre en compte la variabilité temporelle des rejets par temps de pluie dans l'évaluation des impacts d'un SAU en ACV a donc été réalisé. Néanmoins, les perspectives d'amélioration de ce travail sont encore nombreuses. Dans une perspective à court terme, ces travaux pourraient être intégrés aux progrès récents dans la méthodologie ACV sur l'intégration complète de la dynamique dans l'inventaire et l'impact afin d'améliorer la pertinence des impacts des rejets intermittents sur les milieux aquatiques. Dans une perspective à plus long terme avec des modèles hydrologiques multimédias du devenir hydrologique, la pertinence spatiale des impacts de la consommation d'eau serait améliorée. Les scénarios prospectifs d'UWS avec de futurs modèles de précipitations modifiés et une urbanisation croissante sont un domaine prometteur pour les recherches futures. Il faut aussi noter un qu'il reste à poursuivre des développements pratiques pour rationaliser la mise en œuvre du modèle STOIC pour les parties prenantes et les décideurs.

# Abstract / Résumé

With unprecedented urban growth at the planetary scale, urban wastewater systems (UWS) are faced with the operational challenge of managing ever increasing and polluted stormwater to mitigate impairments of the quality of aquatic ecosystems. However, to which extent do wet-weather control strategies actually help reduce overall environmental impacts? With the general aim of assessing in a whole-systems perspective to improve the environmental assessment of UWS with Life Cycle Assessment (LCA), by including impacts from operation and infrastructure, and impacts from discharged pollutants, the objective of the thesis is: "Is it possible to perform the LCA of Urban Wastewater Systems that captures the temporal variability of water discharges in an urban catchment?". The first step in this work demonstrates the relative importance of the impacts caused by intermittent discharges (ID), generated during storm events, compared to continuous discharges from municipal wastewater treatment plants. Then, the core of this thesis is the development of a site-dependent framework to estimate, for life cycle inventory (LCI) purposes, IDs with consideration for their important spatiotemporal variability at the urban catchment scale. The model (named STOIC) satisfies requirements on inventory data quality to support sufficiently accurate modelling of IDs and conceptualizes stormwater processes occurring within an urban catchment and its UWS. These stormwater processes include the generation of water pollutants from nonpoint source emissions to the routing and distribution of pollution in the UWS components, resulting in spatiotemporally differentiated LCIs for water pollutants within an urban catchment. The STOIC framework is applied to an UWS at sub-catchment-scale to assess global performances of different control strategies for IDs in terms of environmental impacts in a real-world case study in Bordeaux, southwest France. This demonstrates the interest and applicability of an extended environmental assessment which considers the temporal variability of UWS discharges from an urban sub-catchment.

**Key words:** life cycle assessment (LCA), environmental impacts, stormwater, urban catchment

Avec une croissance urbaine sans précédent à l'échelle planétaire, les systèmes d'assainissement urbains (SAU) sont confrontés au défi opérationnel de la gestion d'eaux pluviales toujours plus importantes et polluées afin d'atténuer les dégradations de la qualité des écosystèmes aquatiques. Toutefois, dans quelle mesure les stratégies de contrôle par temps de pluie contribuent-elles réellement à réduire les impacts environnementaux globaux? L'objectif général de cette thèse est d'évaluer avec une vision systémique globale, pour améliorer l'évaluation environnementale des SAU par l'analyse du cycle de vie (ACV), en incluant les impacts opérationnels et liés aux infrastructures, et les impacts des polluants rejetés: "Est-il possible d'effectuer l'ACV des SAU en considérant la variabilité temporelle des rejets d'un bassin versant urbain? La première étape de ce travail démontre l'importance relative des impacts causés par les rejets urbains de temps de pluie (RUTP) intermittents, par rapport aux rejets continus des stations d'épuration municipales. Le cœur de cette thèse est l'élaboration d'un cadre dépendant du site pour estimer, en vue d'établir un inventaire du cycle de vie (ICV), les RUTP en considérant leur importante variabilité spatiotemporelle à l'échelle du bassin versant urbain. Le modèle (appelé STOIC) satisfait aux exigences en matière de qualité des données d'inventaire pour soutenir une modélisation suffisamment précise des RUTP et conceptualiser les processus de pollution des eaux pluviales se produisant dans un bassin versant urbain et dans son SAU. Ces processus de pollution des eaux pluviales comprennent la production des polluants des RUTP à partir d'émissions de sources non ponctuelles jusqu'au transfert et à la distribution de la pollution dans les composantes du SAU. Des ICV différenciés spatio-temporellement pour les polluants des RUTP sont calculés pour un bassin versant urbain. Le modèle STOIC est appliqué à un SAU à l'échelle du sous-bassin versant pour évaluer les performances globales de différentes stratégies de gestion des rejets par temps de pluie en termes d'impacts environnementaux dans une étude de cas réel à Bordeaux, dans le sud-ouest de la France. Cela démontre l'intérêt et l'applicabilité d'une évaluation environnementale approfondie qui tient compte de la variabilité temporelle des rejets de SAU d'un sous-bassin versant urbain.

**Mots-clés:** Analyse du cycle de vie (ACV), impacts environnementaux, eaux pluviales, bassin versant urbain