



SAPIENZA  
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# Optimisation of Road Runoff Pollution Tools for use on European Roads

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## ABSTRACT

Environmental concerns are becoming more and more prevalent. One of the main concerns is the preservation of water resources. While the quantitative level of this resource was formerly the main issue, the qualitative level is now equally significant.

Since road runoff is a possible source of diffuse pollution, it has become a growing worry in light of this serious concern and the growth in urbanisation. Road operators and environmental agencies have created new models for road runoff prediction due to the significance of this form of pollution. Four of these models were evaluated in this dissertation: Kayhanian's model (USA), Highways Agency Water Risk Assessment Tool (HAWRAT - UK), PREQUALE (Portugal), and Stochastic Empirical Loading and Dilution Model (SELDL - USA).

Following an assessment of the relevant literature, the study's initial step was gathering monitored data from 20 routes across six European nations. The Site Mean Concentrations (SMC) for total suspended solids (TSS), copper, zinc, lead, and cadmium were determined for each route.

The second stage was to evaluate the four prediction models by comparing monitored data to model results. Along with visual observation, four error indices were produced to determine which model was best suited to the European monitored data. It was determined that none of the models provided sufficiently robust data to be employed as a generic model of application for the entire Europe.

Furthermore, a new prediction equation was developed. In contrast to the four previous models, which were calibrated for a specific country or region, this equation was calibrated using data from all of Europe. The results are consistent with the data because the model was calibrated using the entire dataset. Nonetheless, its usage in actual and diverse roadways should be carefully evaluated.

**Keywords:** Road runoff, PREQUALE, HAWRAT, Kayhanian's model, SELDL, SMC

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## ACRONYMS AND GLOSSARY

AADT	Annual average daily traffic
AADTC	Annual average daily traffic constant
ADP	Antecedent dry period
ANN	Artificial neural network
AR	Annual average rainfall volume with the same duration as the basin concentration
BDF	Basin development factor
C <sub>p</sub>	Estimated concentration for the pollutant p
COD	Chemical oxygen demand
CR	Climate region
CRC	Climate region constant
CRM	Coefficient of residual mass
CSR	Cumulative seasonal rainfall
DA	Drainage area
DL	Drainage length
EMC	Event mean concentration
ENS	Efficiency Nash-Sutcliffe coefficient
GUI	Graphical user interface
HAWRAT	Highways Agency Water Risk Assessment Tool
HRDB	Highway Runoff Database
IDF (curves)	Intensity-duration-frequency (curves)
IF	Impervious fraction
MC	Month constant
MHI	Maximum hourly intensity
MLR	Multiple linear regression
P <sub>annual</sub>	Annual average rainfall
PAH	Polycyclic aromatic hydrocarbon
PC	Pollutant constant
PROPER	Project Road Runoff Pollution Management and Mitigation of Environmental Risks
R <sup>2</sup>	Coefficient of determination
RMSE	Root mean square error
S	Average slope
SELDM	Stochastic Empirical Loading and Dilution Model
SMC	Site mean concentration
T <sub>c</sub>	Concentration time
TER	Total event rainfall
TSS	Total suspended solids
USA	United States of America

# 1 INTRODUCTION

## 1.1 ROAD RUNOFF POLLUTION

The quality of the water bodies is a significant concern in light of the global environmental concerns that are rising. A few management techniques have been used to reduce pollution from both point and nonpoint sources. Road runoff, which may have significantly lower quality than the effluent of some wastewater treatment plants, is one source of nonpoint pollution of these aquatic bodies (Ringler, 2007). Therefore, research and the development of better instruments are required to assist decision makers in managing the quality of these water bodies.

Lack of specific legislation to assess this kind of runoff is one of the most concerning issues at the federal level. Road runoff has very different characteristics from the rejected water from wastewater treatment plants, so it's important to note that the Decree-Law is only used as a reference by the researchers. This is because

- (i) the legislation only applies to punctual pollution, which is not the case for diffuse pollution like road runoff pollution, and
- (ii) seasonality on road runoff is much more evident than in the waters from the wastewater treatment plants.

In response to the European environmental concern, the Water Framework Directive (OJEC, 2000) has emerged, which requires a good understanding of the impacts of pollution sources and also the control of the most relevant to the receiving water bodies. In the case of roads, Barbosa *et al.* (2011) argues that it is important that the assessment of concentrations and pollutant loads take into account the characteristics not only of the road, but also of the climate. The prediction of the quality of a road runoff is a rather challenging issue due to its stochastic and diffuse nature. Winkler (2005) states that, even more complicated is the ability to assess the impact of pollutants on the receptor medium due to the need to analyse the case over a large time scale (*e.g.* due to persistent substances).

In this dissertation, previously collected data of road runoff of six European countries were gathered and compared to the predictions of four models. In order to provide decision makers with the most reliable tools, an assessment of these models was performed. Moreover, a regression model intended to predict total suspended solids (TSS), copper, zinc, lead and cadmium site mean concentrations (SMC) was also developed using the data from six European countries.

## 1.2 SCOPE AND OBJECTIVES

The pollutants generated through the traffic and road construction and maintenance can be automatically deposited in the soil, or emitted into the air and subsequently, some of them, deposited due to gravitational force or precipitation reaching the closest surface water bodies, as indicated in Figure 1. Although the sources of pollutants in these infrastructures are well defined, literature indicates that the prediction of pollutant loads and concentrations is uncertain. This uncertainty is due to the several variables at stake, for instance, the type of pavement of the road, the antecedent climatic conditions and the intensity, frequency and magnitude of rainfall events. These must be viewed as a stochastic phenomenon as it is impossible or not realistic to determine the exact process boundary conditions (Fernandes and Barbosa, 2018).

Tools that apply to the understanding of pollutant sources, their mobilisation, transport to the receiving environment and groundwater, should be seen not in an exact context, but through a statistical or risk assessment (Fernandes and Barbosa, 2018).

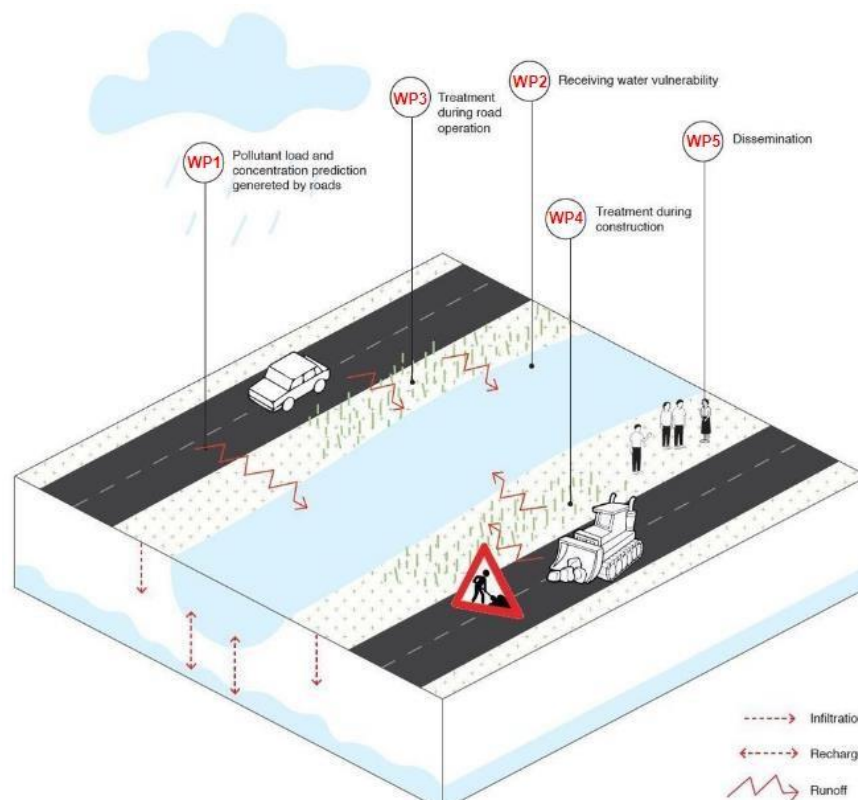


Figure 1 – Representation of road runoff and probable ways of reaching surface waterbodies  
(source: <http://proper-cedr.eu/index.html>)

The current work has the following main objectives:

- (i) Characterisation of the road runoff in and European context;
- (ii) Collection and analysis of available monitored data;
- (iii) The assessment of road runoff predicting tools



- a. (HAWRAT - Highways Agency Water Risk Assessment Tool;
  - b. SELDM - Stochastic Empirical Loading and Dilution Model;
  - c. PREQUALE and
  - d. Kayhanian's multiple linear regression method).
- (iv) Development of a new SMC prediction model.

### 1.3 DISSERTATION STRUCTURE

The dissertation is divided into six distinct chapters, the contents of which are summarised below. The present chapter presents the introduction and the objectives of the dissertation. In the second chapter, a review of the worldwide existing literature is presented, focusing essentially on the European references. This section corresponds to a generic characterisation of road runoff pollution, a description of the pollutants and corresponding sources and the distinction between acute and cumulative impacts. Moreover, specific cases like highways maintenance and accidental spillages, concentrations and loads calculations and a brief view of road runoff model types are presented.

The third chapter comprises the monitoring data collection for each country, its comparison with the legal regulated limits and the description of the predicting models that were assessed in the current work.

The fourth chapter concerns the assessment of the predicting tools. After a sensitivity test of a specific model, the methodology followed for the assessment is presented. Issues like the input data and the easiness of application are presented. Finally, a critical review is presented for each of the models, comparing their predictions with the monitoring data

The fifth chapter concerns the development of a new model. This model is the only model studied in this work that was calibrated with European data from more than one country.

In the final chapter, the discussion of results and conclusions are presented.

## 2 LITERATURE REVIEW

### 2.1 ROAD RUNOFF POLLUTION

#### i. Generic characterization of road runoff pollution

It has been observed that the primary focus of water management has shifted from quantity to quality and quantity. Along with this worry, an effort was made to guarantee the resource's integrated management from a strong sustainability viewpoint, taking into account its technical, economic, social, and ecological elements (Coelho, 2009).

In order to manage and protect the water resources, numerous studies were created. Regarding potential pollutant inputs into receiving bodies, the primary distinction that can be made is between point and nonpoint sources, or diffuse pollution. According to Loague and Corwin (2005), nonpoint source pollution refers to an effluent input from a variety of origins, such as superficial runoff, atmospheric deposition, precipitation, or infiltration, which origin is difficult or nearly impossible to identify. In contrast, the first refers to a direct and easily identifiable input (such as a pipe) of a polluted effluent.

Road runoff is frequently thought of as an effluent with specific properties. Nevertheless, it encompasses a complicated web of contaminants that are mostly influenced by a number of variables, including traffic and the features of the location where they are produced. Barbosa et al. (2011) point out that while examining the possible effects of these pollutants, it is important to consider the features of both the pollutant and the receiving environment. As shown in Table 2.1, the base pollutant matrix of road runoff water is made up of solids, metals, hydrocarbons, and inorganic salts, according to a 2002 report by the Water Research Council (quoted in Higgins, 2006). Depending on their amount and condition, each of these contaminants may be harmful to receiving surface and subsurface water bodies (Barret et al., 1995 in Higgins, 2006). Road signs, as reported by Barbosa et al. (2011), are another major human source contributing to the aforementioned pollutant matrix, along with vehicle circulation and upkeep on highways, as easily proved in the 1994 CIRIA report (in Higgins, 2006).

It is acknowledged that road runoff is a nonpoint source of pollution. The European Union Water Framework Directive has strengthened national laws governing discharges, so management operators and national authorities have an obligation to make sure that these discharges abide by them (Barbosa et al., 2011). Since its introduction, there has been an increased demand for nonpoint source pollution control and for the identification of the source of pollutants influencing the receiving environment (OJEC, 2000 in Higgins, 2006). As a result, strategies for treating water have been continually improved.

Higgins (2006) pointed out the five main categories of factors affecting contaminant concentrations namely:

- (i) Traffic volume and characteristics;
- (ii) Precipitations characteristics and pattern;

- (iii) Surrounding land use;
- (iv) Pavement structure and material used in construction;
- (v) Pollutant characteristics.

Regarding the effects of the traffic volume in the road runoff pollution, at least three different ways of measuring traffic volume can be identified (Irish *et al.*, 1995):

- (i) Vehicles travelling during the storm (VDS);
- (ii) Vehicles travelling in the antecedent dry period (VADP);
- (iii) Annual average daily traffic (AADT).

There are various approaches to classify traffic volume. Although AADT is used by many road agencies to assess whether a highway requires a runoff treatment system, some authors have found no correlation between this indicator and the amount and concentration of heavy metals, oils, lubricants, solid pollutants, or any of these substances (Higgins, 2006). Irish *et al.* (1995) hypothesise that as cars come into close touch with precipitation during a high rainfall event, the quantity of automobiles moving during a storm may have a substantial impact on pollution loads.

Three primary indicators are frequently used in the literature to evaluate the properties of precipitation in relation to road runoff pollution:

- (i) Antecedent dry period (ADP);
- (ii) Rainfall intensity; and
- (iii) Runoff volume.

According to Irish *et al.* (1995), ADP is the amount of time (days or hours) before a rain occurrence that there is no runoff. According to Howell's (1978) early research (used by Irish *et al.*, 1995), the build-up of solids on the highway and the associated pollution loads in the runoff were significantly influenced by the previous dry spell. Because rain is the primary mechanism via which pollutants are removed from the air, cars, and the surface of highways, its intensity is a crucial factor in connection to the loadings of pollutants (UK Transport Research Laboratory, 2002).

The characteristics of precipitation are the most important for the occurrence of the so-called "First-Flush" effect. This phenomenon occurs when a precipitation event is preceded by an ADP of several days. The concentration peak varies for each pollutant during the same rainfall event or in the same watershed during different rainfall events (Wanielista and Yousef, 1993 in Yannopoulos *et al.*, 2013). The occurrence of a first-flush effect in a road runoff event is exemplified in Figure 2.1.

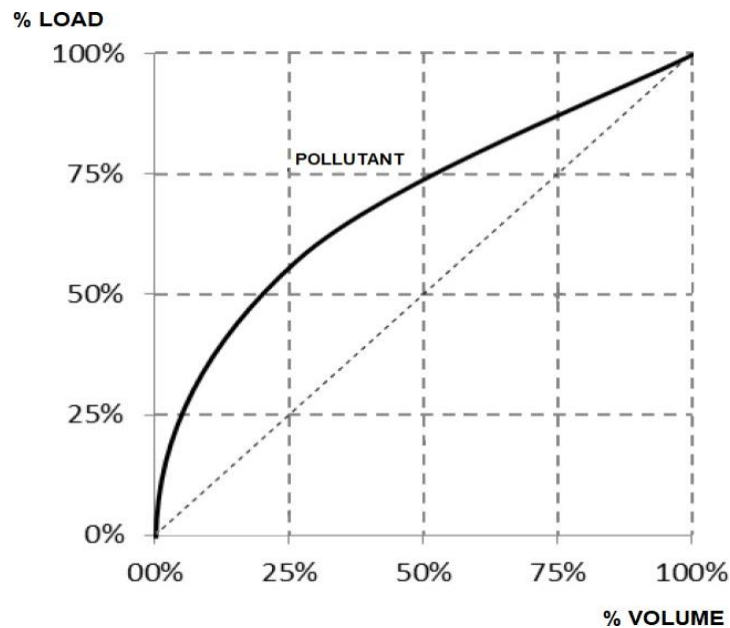


Figure 2.1 – Example of first-flush effect (Adapted from: Antunes, 2014)

During the beginning of the rain event, road runoff water pollution is typically more pollutant concentrated compared to the rest of the storm. As volume increases, the pollutant load begins to converge towards the linear regression line. Volume on the horizontal axis represents the amount of rain that occurred during an event, whilst load on the y axis represents the amount of pollution load contained in percentage volume of precipitation. The first-flush phenomenon is affected by certain parameters like the size of the watershed, rainfall intensity, impermeable area and the antecedent dry weather period.

ii. POLLUTANTS AND SOURCES

Road traffic, weather conditions and the highways maintenance are responsible for the transport of the road runoff to the receiving environment (Piguet, 2007 and Kobriger and Geinepolos, 1984). The main pollutants comprise solids, heavy metals, inorganic salts and hydrocarbons. A list of pollutants and their sources are listed in Table 2.1.

Table 2.1 – Main pollutants and associated sources (Higgins, 2006)

Pollutant	Specific Contaminant	Source
Solids	Carbon	Exhaust, Oil
	Organic Solids	Oil, Exhaust
	Rubber	Tyres
	Plastic	Vehicles
	Grit	Deicing salts, Road structure
	Asbestos	Brakes, Clutches
	Rust	Vehicles
	Metal Filings	Vehicles
Metals	Arsenic	Fuel
	Barium	Paints, Rubber
	Cadmium	Tyres, Oils, Galvanised metals
	Calcium	Oils, Deicing salts
	Chromium	Metal plating, Bearings, Brushings
	Copper	Paints
	Iron	Tyres, Brakes, Oils, Bearings
	Lead	Corrosion
	Magnesium	Fuel, Tyres, Brake linings, Bearings
	Manganese	Cast metal
	Nickel	Tyres, Brakes, Oils, Bearings
	Zinc	Fuel, Paints, Tyres, Lubricants, Corrosion, Brakes
Hydrocarbons	Aliphatic Hydrocarbons	Lubricant, Fuel, Anti-freeze
	Poly Aromatic	Fuel, Lubricants
	Hydrocarbon	Lubricants, Fuel, Anti-freeze
	Phenols	Combustion products
	Carbonyl Compounds	Road surface
	Bitumen	Road surface
	Asphalt	Spillages
	Solvents	Fuel combustion
	Polychlorinated biphenyl (PCB)	Fuel combustion
	Methyl tert-butyl ether (MTBE)	Fuel combustion
Inorganic Salts	Nitrates	Lubricating oil
	Chlorides	Deicing salts
	Phosphates	Lubricating oil
	Herbicides	Road verges (maintenance activities)

As not all the pollutants from runoff presented in Table 2.1 are regularly monitored, Kayhanian *et al.* (2012) suggested a selection of the most important parameters to be monitored to evaluate the road runoff (*cf.* Table 2.2).

Table 2.2 – Road runoff components division (Kayhanian *et al.*, 2012)

Runoff Components	
Conventional and aggregate water quality parameter	TSS; Total dissolved solids; Dissolved organic carbon; Total organic carbon; Chemical oxygen demand (COD); Biochemical oxygen demand; Oil and grease; Hardness as CaCO <sub>3</sub> ; Temperature; pH
Metal constituents	Most frequently: Cadmium; Chromium; Copper; Lead; Nickel; Zinc Less frequently: Aluminium; Arsenic; Iron
Nutrient constituents	*Nitrates; Ammonium; Total Kjeldhal nitrogen; Total nitrogen; Total phosphorus
Infrequently measured water quality parameters	Fecal indicator bacteria; Toxicity; Polycyclic aromatic hydrocarbons (PAHs); Herbicides; Pesticides

"Kayhanian et al. (2012) also points out that the presence of phosphorus and nitrogen as pollutants in the monitoring of runoff water is due not only to pollutant sources related to road traffic, but also due to external factors.

The mentioned pollutants are not only caused by road traffic. They may come from several sources, both anthropogenic and natural. Some are transported long distances by the wind, being deposited later in the most varied places as stated by Fritzer (*in* Winkler, 2005). According to the same source, the most relevant related pollution sources are: the abrasion of road surfaces; the abrasion of tires; drip loss; combustion emission; the abrasion of brake pads and clutch plates.

After considering road traffic and emitted pollutants into the atmosphere as the first and second source of road pollution, Barbosa *et al.* (2011) pointed out the maintenance and construction activities as a third source of pollutants. As far as construction is concerned, the largest pollutants are related to solids and accidental cases, such as situations with fuels, oils and lubricants. Regarding the road maintenance, the main sources of pollutants are the de-icing salts (chlorides) used in some parts of Europe, where snow and ice abound during the colder periods of the year, as well as herbicides, which in high concentrations lead them to be a persistent pollutant in the ecosystem (Mudge and Ellis, 2001 *in* Higgins, 2006).

### iii. RECEIVING WATERS IMPACTS

The potential impacts of each pollutant in the receiving water are presented in Table 2.3.

Table 2.3 – Main impacts per type of pollutants

Pollutant	Impacts
Solids	Reduce light transmission which limits photosynthesis and diminishes aquatic food supply (Goldman, 1986 and Barret et al., 1995 in Winkler, 2005); Lead to an elevated level of insoluble substances with negative impacts on fish eggs and larvae through clogging of the pores between the substrate of the riverbed (Winkler, 2005); Clog fish gills and harm their respiration and the respiration of other aquatic animals (Hill, 2010).
Metals	Can be toxic because metals undergo bioconcentration (Salomon, 2008). The toxicity associated may reduce diversity and abundance of the sensitive aquatic biota and replace them with pollution tolerant species (Hvitved-Jacobsen and Yousef, 1991); Copper, cadmium and zinc could be toxic even in low concentrations (Scheffer and Schachtschabel, 2002 and Hahn, 2004 in Winkler, 2005).
Hydrocarbons	Several PAHs are toxic, mutagenic/carcinogenic. This type of pollutants is highly lipid soluble and thus easily absorbed by human bodies (Abdel-Shafy and Mansour, 2015). Methyl-Tertiary-Butyl-Ether (MTBE) is toxic to several freshwater organisms (Werner et al., 2001).
Inorganic Salts	Like fertilizers and herbicides, used in the maintenance of road shoulders, essentially on the roadside, lead to an increase in phosphorus and nitrogen in the runoff matrix, which contributes to the eutrophication of the receiving environment (Hvitved-Jacobsen and Yousef, 1991).

### iv. ACUTE AND CUMULATIVE IMPACTS

Depending on the pollutant type, concentration, rate of assimilation of organisms, and on its form (dissolved and particulate), the impacts created in the water environment may be acute or cumulative.

Acute effects are associated with accidental spills and/or organic or metallic pollutants entering the composition of road runoff. Other examples of acute effects are the presence of copper in its soluble form, soluble short-chain organic pollutants (*e.g.* herbicides) and runoff of suspended solids (in case of road maintenance campaigns, or after a long period without occurrence of precipitation) as stated by Barbosa *et al.* (2011). Hvitved-Jacobsen and Yousef (1991) defined that the impacts that cause this kind of effects are characterised by short duration events and that the impact declines after the discharge is over; even if the events last for few days it is still considered an acute impact.

Cumulative effects are associated with less soluble metals (although their solubility depends on particle characteristics, water hardness, iron and aluminium oxides content and relative concentration), thus being related to a toxicity that develops due to accumulation pollutants in the tissues of organisms. The most persistent hydrocarbons (as PAHs) are usually considered as the particulate fraction of the pollutants. The physical accumulation of sediments such as silt and clay can change the ecosystem by covering surfaces and choking flora and fauna. Chronic effects may occur when these sediments are

contaminated with PAHs or metals (Barbosa *et al.*, 2011). Besides that, Hvitved-Jacobsen and Yousef (1991) refers that other type of pollutants that may lead to cumulative impacts are nutrients namely to the eutrophication of low hydrodynamic media such as reservoirs.

#### v. SPECIFIC CASES

There are two main types of specific cases in highway runoff pollution, which are not due to the continuous vehicles traffic:

- (i) Highway maintenance;
- (ii) Accidental spillage.

Table 2.1 shows that the most diverse pollutants are related to a highway's maintenance operations. According to Maestri *et al.* (1988), pollutants including herbicides and nutrients are primarily detected in highway runoff as a result of highway maintenance activities and neighbouring land-use impacts. High sediment movement during maintenance operations and fuel, oil, and grease leaks, among other things, are further examples (Barbosa *et al.*, 2011). It is vital to be worried about construction waste and maintenance of treatment systems (for example, sedimentation sludge removal) on heavily travelled roadways where runoff water treatment systems are already in place (Barbosa *et al.*, 2011).

Events like the loss of lubricants and fuels in a car or the leakage of commodities delivered in big goods vehicles greatly increase the likelihood of a leak on a road. According to Barbosa *et al.* (2011), spills typically result in acute pollution when they come into contact with aquatic bodies. Nevertheless, occasionally the spill's byproduct would seep into the earth and contaminate the water.

#### vi. CONCENTRATIONS

In order to evaluate and study road runoffs, it is necessary to define some concepts whose equations units are presented in dimensional analysis. Event mean concentration (EMC) is defined as the pollutant concentration of a composite of multiple samples collected during the course of a storm (Thornburg and Lowe, 2009), as represented in the equation below (*e.g.* Antunes, 2014).

$$EMC = \frac{\sum_{j=1}^n C_j \times V_j}{\sum_{j=1}^n V_j}$$

EMC - Event mean concentration ( $ML^{-3}$ )

j- Number of time intervals analysed by each event

$V_j$ - Volume in each time interval  $j(L^3)$

$C_j$  - concentration of the pollutant in  $V_j$  volume ( $ML^{-3}$ )

This dissertation aims to calculate the SMC *i.e.* the average or the median of the monitored EMC of each site. When the number of monitored events is very low, it is usual to use the average (Barbosa *et al.*, 2011).



$$SMC = \frac{\sum_{k=1}^N EMC_k}{N}$$

SMC - Site mean concentration ( $ML^{-3}$ )

$\sum_{K=1}^N EMC_k$  - Event mean concentration for a storm k ( $ML^{-3}$ )

N - Total number of storms sampled at a given catchment

## 2.1. TOOLS TO PREDICT ROAD RUNOFF

Sitterson et al. (2017) provided an overview of runoff model types. The authors classify the models into three categories:

- i. conceptual,
- ii. physical, and
- iii. empirical.

According to Vaze (2012), conceptual models connect simplified hydrology components and are based on simplified hydrological processes that provide a conceptual view of the catchment area (Sitterson et al., 2017).

Physical models are based on an understanding of the physics involved in hydrological processes. The model is governed by physical equations that describe several components of real hydrologic reactions in the catchment (Vaze, 2012; Sitterson et al., 2017).

Empirical models involve mathematical equations that are derived from observations of the inputs and outputs. In these models, runoff modelling is based in temporal data series (Granata *et al.*, 2016).

Some examples of empirical models are regression analysis, artificial neural networks (ANN) and Monte Carlo methods. Regression analysis could be seen as a set of statistical processes to estimate the relationships between variables. This method allows to understand how the changes in the independent variable influences the dependent variable (Ramana, 2014). Monte Carlo methods simulate random values which give an approximate solution of a mathematical or physical problem (Sobol 1974; Rubenstein, 1981 in Karlovits, 2010).

### 3. DATA AND METHODS

#### 3.1. DATA COLLECTION

The assessment of the models was made considering the comparison between the prediction and monitored data. The former is related to SMC of 20 roads in Europe. These data were collected from direct contacts to the road and research institutes dealing with road runoff. A summary of the main characteristics of the monitored roads is presented in Table 3.1.

Table 3.1 – Road characteristics

Highway	Country	Code	Drainage Area (m <sup>2</sup> )	Impervious fraction (IF) (%)	Annual Precipitation (mm)	AADT
A1	Portugal	P1	22,800	0.412	646	27746
A2	Portugal	P2	1,287	1	528	16344
A6	Portugal	P3	5,580	1	744	2918
A22	Portugal	P4	15,422	0.85	518	24000
A25	Portugal	P5	287	1	1014	15673
IP6	Portugal	P6	7,280	1	709	6539
A27	Netherlands	N1	48,590	0.5	776	63000
A27	Norway	N2	30,510	1	776	63000
E6	Norway	N3	22,000	1	834	42000
A11	France	F1	3,200	1	786	24103
A11	France	F2	3,200	0.5	786	24103
M7	Ireland	I1	14,184	1	731	27500
M7	Ireland	I2	11,368	1	731	27500
M7	Ireland	I3	9,600	1	731	27500
M4	England	E1	8,755	1	745	70000
M4	England	E2	4,348	1	745	35000
M40	England	E3	58,680	1	615	78000
A417	England	E4	20,232	1	843	24000
A34	England	E5	2,760	1	660	64000
A34	England	E6	19,425	0.5	635	36000

In Figure 3.1 the location of the monitored sites in an annual average precipitation map is presented.

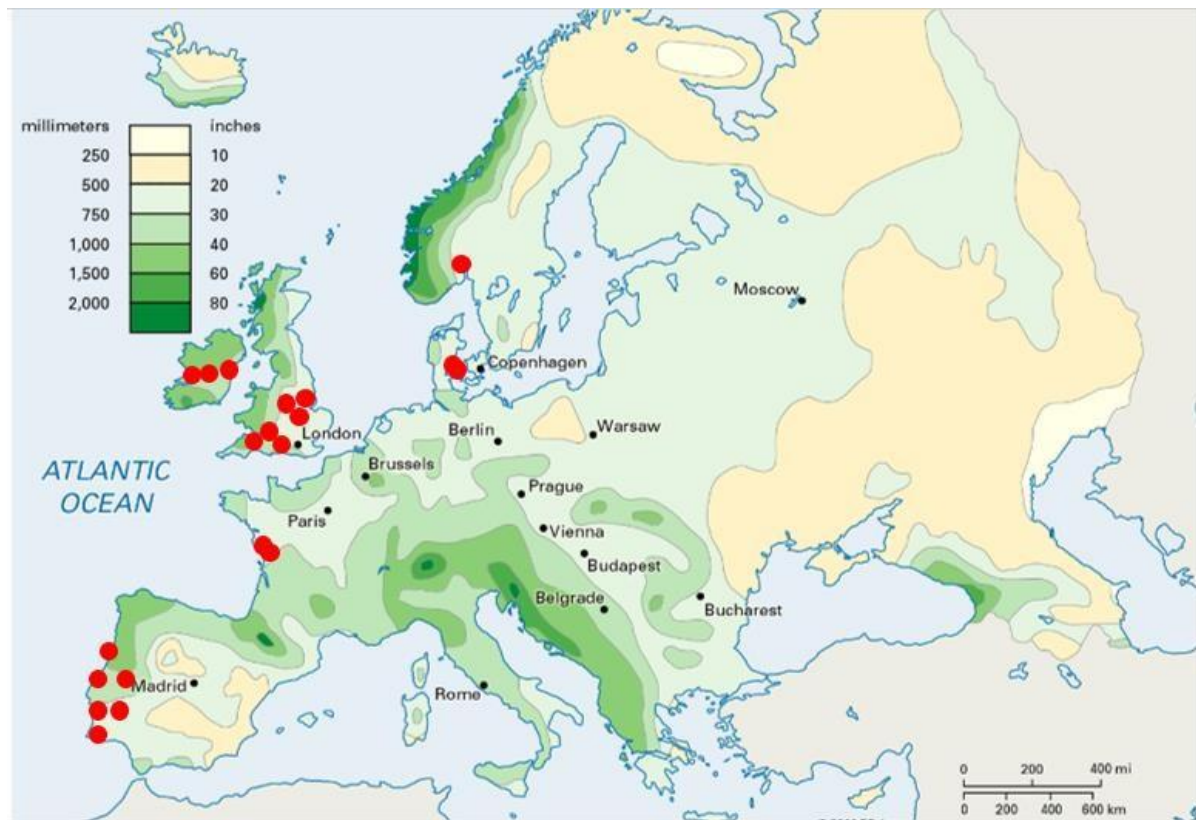


Figure 3.1 – Europe precipitation map with the roads under study. Each dot corresponds to one road

As previously stated, to determine which of the tools under consideration is best suited to the obtained data, the anticipated results for each tool were compared to the monitored data. The monitored data is shown in Annex 1 present at the end of the thesis. To process the monitored data, the EMC values for each site and contaminant were averaged to determine the mean SMC. This analysis is provided in Table 3.2, which confirms that only TSS exceeds the limit (60 mg/L). The project partners collected or made available an hourly precipitation data set.

Table 3.2 – SMC of each site

	Site Mean Concentration of Each Site				
	TSS	Cu (mg/L)	Zn ( $\mu\text{g/L}$ )	Pb ( $\mu\text{g/L}$ )	Cd ( $\mu\text{g/L}$ )
Highways	22,96	19,24	124,07	4,38	0,09
P1	2,50	11,13	69,00	2,10	
P3	19,65	8,10	345,83	1,83	
P4	52,44	24,44		23,33	
P5	57,93	86,86	139,69	28,65	
P6	207,08	31,45	73,17	7,67	1,09
N1		114,71	500,88	29,88	1,60
N2		29,14	118,86	14,43	1,00
N3	227,61	84,09	224,87	14,70	0,21
F1	71,38	45,51	356,08	57,93	1,03
F2	10,89	27,37	160,05	11,64	0,43
I1	856,44	123,29	666,67	139,38	8,70
I2	155,74	48,95	198,25	68,91	4,86
I3	49,52	24,70	82,00	76,90	8,61
E1	88,60	30,00	100,70		
E2	310,87	54,61	221,50	68,98	1,77
E3	50,88	42,65	149,31	15,16	0,43
E4	64,76	23,99	52,60	4,38	0,21
E5	101,13	67,92	219,73	50,45	0,62
E6	82,70	32,46	29,01	16,57	0,25

### 3.2. DESCRIPTION OF THE MODELS

#### 3.2.1. PREQUALE

Between 2002 and 2006, road runoff from multiple roads was monitored as part of the G-Terra research project, which was sponsored by the Portuguese Foundation for Science and Technology and directed by the National Laboratory for Civil Engineering (Barbosa et al., 2011). The PREQUALE tool (Previsão da Qualidade das Águas de Escorrências) was built using data from six routes. This method seeks to directly predict SMC. It is based on the following principles:

- i. input data easily available for designers;
- ii. easiness of calculation;
- iii. clear and transparent model and
- iv. reliable results at national level.

The applicability of the tool is rather simple as it is based on a multiparametric equation (equation 3.1) with the following input variables:

- a. Drainage area (DA in km<sup>2</sup>) – area which contributes with runoff to the discharge point during a rainfall event
- b. Impervious fraction (IF in %) – the percentage of the total drainage area which is impervious;
- c. Average annual rainfall volume with the same duration as the basin time of concentration (AR in mm) – further details on its calculation will be provided below;
- d. Annual average precipitation (P<sub>annual</sub> in mm).

The multiparametric equation takes the following form:

$$SMC = a_i (DA^{\beta^1} \times IF^{\beta^2} \times AR^{\beta^3} \times P_{annual}^{\beta^4}) \quad (3.1)$$

where SMC is the estimated site mean concentration of each pollutant and  $\alpha_i$ ,  $\beta_1$ ,  $\beta_2$ ,  $\beta_3$  and  $\beta_4$  are the corresponding regression coefficients. The AR was calculated in order to denote a representative rainfall event of the region. It was assumed that

$$t_c = 0,0663 \times \frac{DL^{1,155}}{\Delta h^{0,385}}$$

$t_c$  - Concentration time (hours)

DL - "Main river" length (Km) - (in this case is the maximum length of the road in the drainage basin)

$\Delta h$  - Slope(m) – heights difference between the ends of the road

this event is the average precipitation with a duration equal to the time of concentration of the basin and with a return period of two years. To calculate this variable, it is necessary to use auxiliary calculations: firstly, the time of concentration of the drainage basin has to be determined (e.g. equation 3.2, in Lencastre and Franco, 1984).

Secondly, the volume is calculated using intensity-duration-frequency (IDF) curves with a return period of 2 years.

The current version of PREQUALE allows the prediction of SMC for TSS, chemical oxygen demand (COD), Fe, Zn and Cu. This tool was validated for the situations in which the parameters' values were between the values presented in Table 3.3.

Table 3.3 – Intervals for which PREQUALE had been validated (Adapted from: Barbosa *et al.*, 2011)

Parameter	Lower limit	Upper limit
AR (mm)	6,0	7,5
DA (Km <sup>2</sup> )	$2,5 \times 10^{-4}$	$6,5 \times 10^{-2}$
IF (%)	40	100
Pannual (mm)	560	1 200

In Table 3.4 the road characteristics for each road used to calibrate PREQUALE are presented while the regression and correlation coefficients that resulted from the adjusted multiparametric equation of the roads SMC are presented in Table 3.5.

Table 3.4 – PREQUALE roads (Adapted from: Barbosa *et al.*, 2011)

Road	AR (mm)	DA (km <sup>2</sup> )	IF (%)	Pannual(mm)	Observations
A1	7,5	$6,46 \times 10^{-2}$	41,2	1 157,0	Runoff drains to the treatment system
A3 Santo Tirso	6,8	$2,00 \times 10^{-3}$	100,0	782,0	Descending section
A3 Ponte de Lima	6,1	$2,45 \times 10^{-3}$	100,0	1 537,4	Ascending section
A6	6,5	$5,58 \times 10^{-3}$	100,0	761,0	Runoff drains to the treatment system
A25	6,0	$2,50 \times 10^{-4}$	100,0	929,0	Near Aveiro lagoon
IP6	6,0	$7,28 \times 10^{-3}$	100,0	902,0	Runoff drains to the treatment system

Table 3.5 – PREQUALE regression and correlation coefficients (Adapted from: Barbosa *et al.*, 2011)

Parameter	$a_i$	$\beta_1$ (DA)	$\beta_2$ (IF)	$\beta_3$ (AR)	$\beta_4$ (Pannual)	Correlation Coefficient
TSS (mg/L)	$1,22 \times 10^{44}$	0,257	-5,085	-28,797	-2,945	0,9696
COD (mg/L)	$1,91 \times 10^{25}$	0,1644	-3,165	-16,914	-1,064	1,0000
Fe (mg/L)	$9,20 \times 10^{44}$	-0,1491	-6,546	-28,229	-3,371	1,0000
Zn (mg/L)	$1,15 \times 10^{05}$	-0,135	-1,08	-0,323	-1,296	0,8843
Cu (mg/L)	$3,08 \times 10^{01}$	0,036	-0,705	0,396	-0,702	0,9989

### 3.2.2. HIGHWAYS AGENCY WATER RISK ASSESSMENT TOOL (HAWRAT)

HAWRAT was developed by Highways Agency from the United Kingdom as a standalone application aiming at helping highway designers decide if road runoff pollution mitigation measures are needed.

This tool allows the prediction of

- (i) soluble pollutants and
- (ii) sediment related, expressed as EMC for total copper, zinc, cadmium, pyrene, fluoranthene, anthracene, phenanthrene and total PAH. As the model predicts EMC, it is necessary to calculate several EMC (in a time frame) in order to predict the SMC.

Besides the prediction of runoff quality, HAWRAT also incorporates models to predict the impact of the runoff on receiving rivers and streams, as shown in Figure 3.2. HAWRAT comprises three steps:

Step 1 concerns road runoff pollution prediction,

Step 2 is related to the impacts in the receiving water bodies and

Step 3 deals with the selection of mitigation measures.

In the scope of the present work, the results of steps two and three were not analysed.

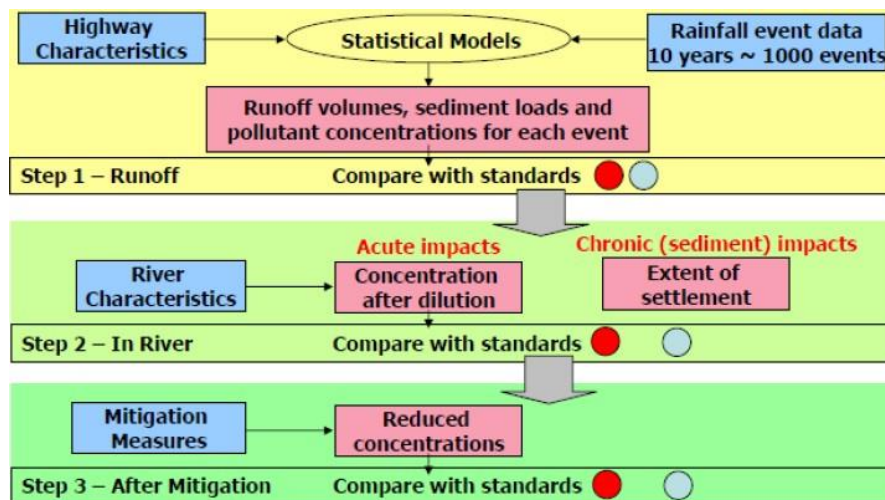


Figure 3.2 – HAWRAT methodological scheme (Jotte *et al.*, 2017)

HAWRAT should not be used in certain cases, such as:

- (i) Urban Highways;
- (ii) Highways with traffic densities outside the range of 11 000 – 159 000 vehicles/day (it can be used for highways with traffic density less than 11 000 vehicles/day but the result may be overestimated) and
- (iii) Highways discharging to receiving watercourse that are tidal and/or saline (Highways Agency, 2009).

The agency also emphasize that the tool can be applied in Wales, Scotland and Northern Ireland, although the basic data was generated in England, and recalls its limited ability to assess the impact on streams where the flow is intermittent or seasonal.

As described by the Highways Agency (2009), in order to use the graphical interface, HAWRAT uses an auxiliary software that stochastically generates hourly rainfall series of the United Kingdom and calculates a main part of the mandatory inputs of the tool.

However, the pollutants that were intended to be studied in this work were not available in the automatic tool. Instead, the equation that was the basis of HAWRAT was used to predict the runoff pollution. This equation (equation 3.3) is a multiple linear regression resulting from a study (Crabtree *et al.* (2008) and Dempsey and Song (2008)). Equation 3.3 allows the user to predict TSS, total copper, total zinc and total cadmium and has the following input variables:

- (i) Pollutant constant (PC) - Fixed to each pollutant;
- (ii) Climate region constant (CRC) – Also fixed to each pollutant;
- (iii) Annual average daily traffic constant (AADTC) – Dependent of the number of cars per day;
- (iv) Month constant (MC) – Fixed and based on the month that the precipitation event occurs;
- (v) Maximum hourly precipitation (MHI in mm/h) – The highest value of hourly precipitation registered in a precipitation event;
- (vi) Antecedent dry period (ADP in hours) – Number of hours without precipitation since the last precipitation event.

$$\log_{10} EMC = PC + CRC + AADTC + MC + \gamma_1 \times MHI + \gamma_2 \times ADP \quad (3.3)$$

Where  $\log_{10} EMC$  is the event mean concentration of the studied pollutant and  $\gamma_1$  and  $\gamma_2$  are the regression coefficients presented in Table 3.6.

CRC is only defined for the region where HAWRAT is applicable (Figure 3.3). In this way, it was defined that the red lines that separate each climatic region will continue indefinitely, so the countries northwest of England will have an "cold/wet" climate, countries to the northeast will have a " cold/dry" climate, countries to the southwest will have a " warm/dry" climate and countries to the southeast will have a "warm/wet" climate.

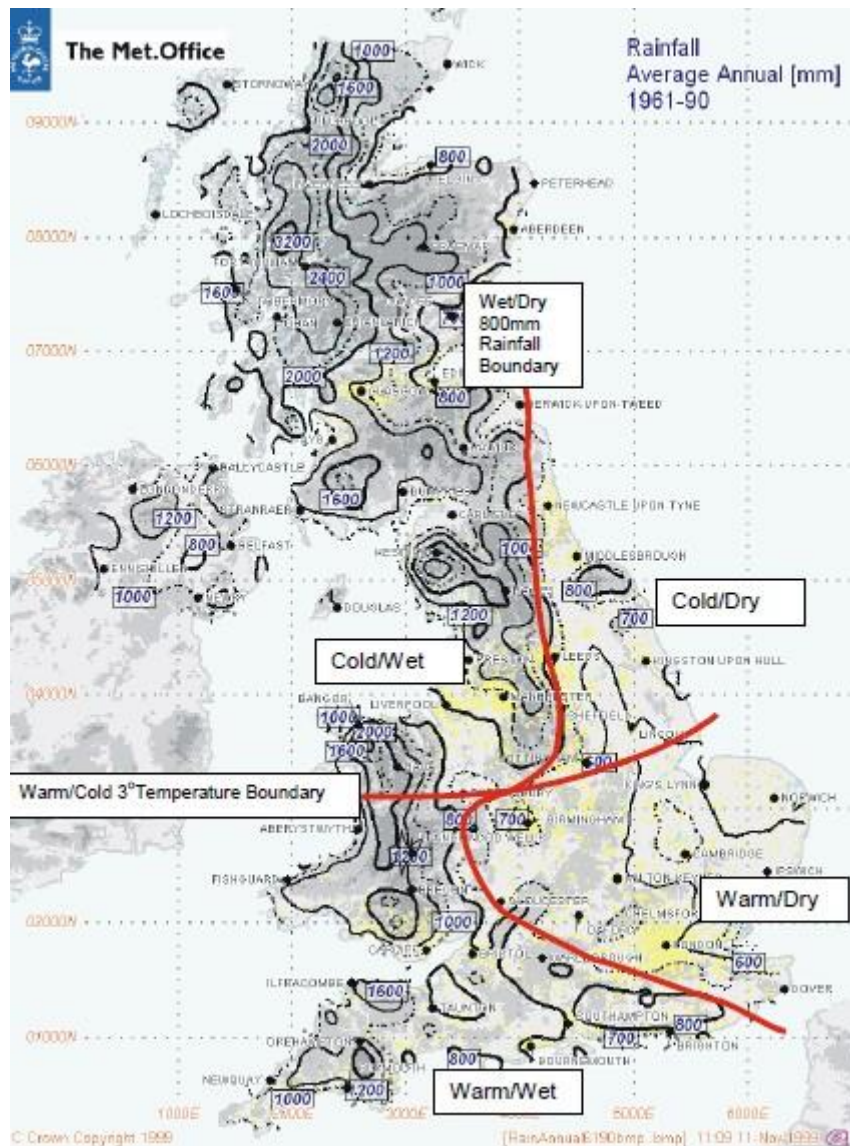


Figure 3.3 – Representative map of the limits/areas used in HAWRAT (Crabtree et al., 2008)

The constants needed for the HAWRAT equation for each combination of region, traffic, month and pollutant are presented in Table 3.6.

Table 3.6 – Constants table in order to predict concentrations trough HAWRAT (Adapted from: Dempsey and Song, 2008)

	Inputs	EMC constants			
		Total Copper	Total Zinc	Total Cadmium	TSS
Site	Constant	1,394	1,91	-0,832	2,1
	Colder/Dry	0	0	0	0
	Colder/Wet	0,042	0	0	-0,217
	Warm/Dry	0,144	0	0	-0,248
	Warm/Wet	0,089	0	0	-0,163



	Inputs	EMC constants			
		Total Copper	Total Zinc	Total Cadmium	TSS
Traffic	AADT<50000	0	0	0	0
	50000= $\leq$ AADT<100000	0,018	0,045	0,093	0
	AADT $\geq$ 100000	0,512	0,502	0,379	0
Months	1	0,402	0,662	0,773	0,535
	2	0,568	0,699	0,565	0,443
	3	0,526	0,704	0,625	0,324
	4	0,427	0,504	0,374	0,193
	5	0,559	0,716	0,579	0,288
	6	0,425	0,32	0,241	0,283
	7	0,258	0,27	0,064	-0,148
	8	-0,064	-0,154	-0,216	-0,108
	9	0,065	-0,098	-0,067	-0,101
	10	0	0	0	0
	11	-0,028	0,068	0,05	0,022
	12	0,085	0,231	0,181	0,491
Extra	MHI	0	0,022	0	0,065
	ADP	0	0	0	0

### 3.2.3. KAYHANIAN'S MULTIPLE LINEAR REGRESSION METHOD

Kayhanian *et al.* (2006) proposed a multiple linear regression (MLR) to predict EMC. This regression was undertaken with the following specific objectives:

- Provide a statistically summary of highway runoff quality in California, United States of America (USA);
- Discuss the impact of selected independent event and site characteristics parameters on highway runoff constituent EMC and
- Evaluate the application of the MLR models as predictive tools to estimate the constituent EMC.

Stormwater runoff data used in Kayhanian *et al.* (2006) were obtained from 34 highway sites in California covering a wide range of annual average daily traffic levels and environmental conditions. These data were obtained, on average, up to eight storm events at each highway site during wet seasons (October to April) over a three years period (2000 to 2003). Some characteristics were recorded in each site, namely surrounding land use (obtained from United States Geological Survey maps, local zoning maps and visits to the sites), catchment area, impervious fraction, latitude and longitude and AADT.

Relationships were established by the authors between highway runoff quality for 24 constituents and the following independent variables:

- Total event rainfall (TER in mm) – height of rain of each precipitation event;
- Antecedent dry period (ADP in days) – the number of days with no rain since the

- lastprecipitation event;
- (iii) Cumulative seasonal rainfall (CSR in mm) – the total precipitation of a known season in aspecific location.
  - (iv) Drainage area (DA in ha) – area which contributes with runoff to the discharge point duringa rainfall event;
  - (v) Annual average daily traffic (AADT in vehicles/day) – number of vehicles that pass each dayin the location under study.

The adapted version of the general equation is presented below (equation 3.4). In Table 3.7, there are the constants used in the equation.

$$\ln EMC = \beta_0 + a \times \ln(TER) + b \times \ln(ADP) + c \times \sqrt[3]{CSR} + d \times \ln(DA) + e \times (AADT \times 10^{-6})$$

Table 3.7 – Constants Kayhanian's model (adapted from: Kayhanian *et al.*, 2006)

	Constituent	$\beta_0$	a	b	c	d	e
Aggregates	TSS	4,28	- 0,124	0,102	- 0,099	—	4,934
	TDS	4,73	- 0,309	0,126	- 0,050	—	2,582
	DOC	4,11	- 0,404	0,123	- 0,129	—	—
	TOC	5,23	- 0,209	0,129	- 0,154	—	—
Metals (total)	Cu	2,9	- 0,161	0,163	- 0,079	—	6,823
	Pb	2,72	—	—	- 0,102	—	9,65
	Ni	2,51	- 0,196	0,141	- 0,075	-0,155	1,013
	Zn	4,83	- 0,227	0,143	- 0,084	—	6,747
Metals (dissolved)	Cu	2,92	- 0,290	0,185	- 0,102	—	3,679
	Pb	2,04	- 0,248	—	- 0,101	—	0,007
	Ni	2,73	- 0,270	0,068	- 0,107	-0,094	—
	Zn	4,74	- 0,343	0,164	- 0,112	—	1,676
Nutrients	NO3-N	1,3	- 0,417	0,092	- 0,090	—	2,87
	P, total	1,2	- 0,143	0,128	- 0,051	—	0,9
	TKN	1,7	- 0,343	0,102	- 0,128	—	1,535

### 3.2.4. STOCHASTIC EMPIRICAL LOADING AND DILUTION MODEL (SELDM)

SELDM was developed by the Federal Highway Administration from the USA and uses analytical approximations to estimate the potential effects of runoff on receiving waters. SELDM aims at predicting EMC, flows and loads in stormwater from a highway site and its upstream catchment. Using input information based on site characteristics, catchment characteristics, rainfall, stormflow, water quality and the performance of mitigation measures, this tool generates statistical distribution of runoff quality in highway runoff and receiving river water (Granato, 2013a).

SELDM uses a highway runoff database which contains data from over 4000 storm events, then uses the Monte Carlo method to generate the distribution of output variables such as EMC (Gardiner *et al.*, 2016).

Novotny *et al.* (1993) as quoted by Santos and Barbosa in 2004 refers that the deterministic nature of most models to represent the variability of a phenomenon has originated some failures. In this case Monte Carlo method is used due to the combination of different variables (such as precipitation, pre-storm flows, runoff coefficients and water quality concentrations).

Granato and Jones (2014) described that SELDM uses Monte Carlo methods to generate a stochastic population of the concentrations, flows and loads needed to implement a mass balance model for a receiving stream and/or lake.

SELDM is not calibrated by changing values of input variables to match a historical record of values. Instead, SELDM's input variables are based on site characteristics and representative statistics for each hydrological variable. The benefit of this method is not to reduce uncertainty in the input statistics, but to represent the different combinations of the values of variables that determine potential risks for water quality (Granato and Jones, 2014).

To estimate the concentrations and loads of water quality constituents in receiving bodies, a mass balance is commonly applied (Granato, 2013a) as shown in Figure 3.4.

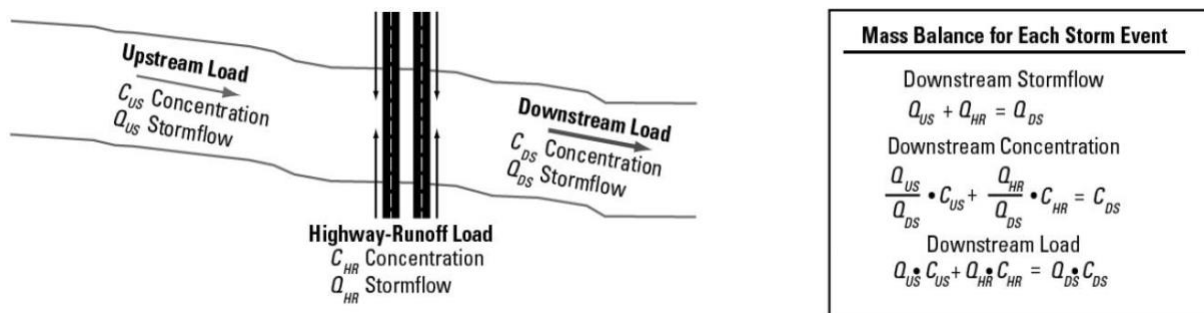


Figure 3.4 – Mass balance for each storm event (Granato, 2013a)

Storm events are commonly defined as independent statistical events characterised by a volume, intensity, duration and time between midpoints of successive storms for the purposes of planning, analysis, and sampling efforts (Driscoll, 1990 in Granato, 2013a). Statistics describing the frequency distributions of component discharges and concentrations are needed to estimate the statistics for downstream discharges, concentrations, and loads (Granato and Jones, 2014). The fact that SELDM was designed to predict road runoff pollution in US areas represents a limitation, which is common to every national based tool. In this case, the USA model defines “Ecoregions” where the parameters are already introduced. Nevertheless, the tool can be used in every region of the world with the manually input information of weather conditions.

The input layout of SELDM is a sequence of graphical user interface (GUI). In total 13 forms need to be completed with inputs information:

- (1) Information about the analyst, project and analysis;
- (2) Highway physical characteristics;
- (3) Ecoregion (when the site under study is in USA);
- (4) Upstream basin characteristics;
- (5) Lake basin characteristics;
- (6) Precipitation statistics (when the ecoregion is settled this form is almost automatically filled, however when the site is out of USA, it is necessary to calculate these data (see Table 3.8) outside the tool);
- (7) Streamflow;
- (8) Runoff coefficient statistics
- (9) Highway runoff quality statistics;
- (10) Upstream water quality statistics;
- (11) Downstream water quality definitions;;
- (12) BMP performance statistics;
- (13) Set of output files and
- (14) Running SELDM form.

As for the road runoff pollution, only two of the 13 outputs are of interest, namely:

- (1) Precipitation event output file and
- (2) Highway runoff quality output file.

SELDM offers seven options for selecting storm-event statistics on the synoptic storm-event-precipitation statistics form as supported by the appendix 4 of SELDM help guide (Granato, 2013b). The default rain zone and ecoregion are automatically selected by entering the latitude and longitude of the highway site. The user, however, can manually select an ecoregion that better represents conditions at a site of interest. The option of entering user-defined statistics can be used to enter site-specific statistics, to do a sensitivity analysis, or evaluate the potential effects of climate change on model results (Granato, 2010).

In order to produce highway runoff quality output file, SELDM uses regional water-quality statistics to facilitate generation of initial planning-level estimates. If necessary, initial estimates can be refined with water-quality statistics based on available data collected at hydrologically similar sites or at the site of interest. SELDM also uses the Highway Runoff Database (HRDB) as source of highway runoff statistics and data, as stated by Granato and Cazenias (2009).

The HRDB application is designed as a data warehouse to document data and information from highway-runoff monitoring studies and as a pre-processor for highway-runoff data for use in SELDM. Available highway runoff data provide the basis for defining runoff quality and quantity at monitored sites and predicting runoff quality and quantity at unmonitored sites. HRDB includes data from 2 650 storms for 39 713 EMC measurements of more than 100 water quality constituents monitored at 103 sites in USA (Granato and Cazenias, 2009).

### 3.2.5. INPUTS AND OUTPUTS SUMMARY TABLE

In the Table 3.8 are presented the inputs which are needed to run each model.

Table 3.8 – Inputs summary table

Inputs	SMC	Predicting tools		
		PREQUALE	HAWRAT	EMC
			Kayhanian's	SELDM
Site characteristics	CR		X	
	DA	X		X
	IF	X		X
	AADT		X	X
	AR	X		
	Pannual	X		X
	Others*	Drainage Length (m); Mean Basin Slope (%)		Location (latitude and longitude); Drainage Length(m); Mean Basin Slope; Basin Development factor
Event Characteristics	Month		X	
	TER			X
	MHI		X	
	ADP		X	X
	CSR		X	
				Average storm event durations; Minimum total storm events; Minimum interevent time;
	Others*			Number of storm events per year

\*As indicated above, SELDM is a tool which needs more inputs than physical and characteristics ones. So, besides those here presented in Table 3.8, SELDM needs the upstream basin characteristics, basin characteristics, streamflow statistics, runoff coefficients and best management practices (BMP) used in the road.

In the Table 3.9 is presented an outputs summary table.

Table 3.9 – Outputs summary table

Predicting Tools					
		SMC		EMC	
		PREQUALE	HAWRAT	Kayhanian's	SELDM *
	TSS	X	X	X	X
	TDS			X	
Aggregates	DOC			X	
	TOC			X	
	COD	X			
	Cu	X	X	X	X
	Pb			X	X
Metals (total)	Ni			X	
	Zn	X	X	X	X
	Cd		X		X
	Fe	X			
	Cu		X	X	
Metals (Dissolved)	Pb			X	
	Ni			X	
	Zn		X	X	
	NO <sup>-3</sup>			X	X
Nutrients (Total)	P			X	X
	KN			X	

\* Besides these outputs, SELDM also has the following outputs: Urban TSS; Ultra Urban TSS; pH; suspended sediment concentration; Total chromium; Total Hardness

\*\* SELDM generates as output Ultra urban TSS; Urban TSS and Non-urban TSS, in this case Non-urban TSS were used as TSS.

## 4. ASSESSMENT OF THE PREDICTING TOOLS

### 4.1. METHODOLOGY

#### STEP 1

An Excel spreadsheet was developed in order to calculate input data of HAWRAT, Kayhanian's model and PREQUALE. Following the recommendations of HAWRAT's help guide: in this spreadsheet it was assumed that a precipitation event is every event above 0,1 mm. The procedure for the development of this spreadsheet was:

- i. Collect the hourly precipitation time series from the closest meteorological stations of each site; Identification of all precipitation events. A precipitation event was considered as every precipitation associated to an hour or several hours with at least 0,1 mm as indicated by HAWRAT's help guide. According to the example in Figure 4.3, this definition lead to the identification of four "precipitation events" in that interval, two of which were only one hour and the other two were two and three hours.
- ii. The calculation of each precipitation event duration was essential for the total event rainfall (TER) calculation, because the calculation of this input requires the number of hours that are needed to be summed.
- iii. Obtain the maximum hourly precipitation (MHI) value from each event. This value was calculated by finding the maximum value of each event in the hourly precipitation series, using as auxiliary calculation the column that identifies an event and the duration of each event.
- iv. Calculation of the antecedent dry period (ADP) of each event by calculating the number of empty cells until the last event.

All the procedure above mentioned is available in Figure 4.3.

AB9												=SE(AA9<>"":10^AA9;"")																											
B				C	D	E				G				H				I				M				Q				T				W					
3				Events				Number of Events				Duration				Event data																							
4				Date and Hours	Month	Year	Precipitation volume (mm)	Number of events with more than one hour				Number of events of one hour				Total number of events				Event Duration				TER (mm)				MHI (mm)				ADP (hrs)							
7				25/01/2001 20:00	01	2001	0																																
8				25/01/2001 21:00	01	2001	0																																
9				25/01/2001 22:00	01	2001	0,1									1				1				1				0,1				0,1				4			
10				25/01/2001 23:00	01	2001	0																																
11				26/01/2001 00:00	01	2001	0																																
12				26/01/2001 01:00	01	2001	0					(i)				(i)				(i)				(ii)				(ii)				(iii)				(iv)			
13				26/01/2001 02:00	01	2001	0																																
14				26/01/2001 03:00	01	2001	0																																
15				26/01/2001 04:00	01	2001	0																																
16				26/01/2001 05:00	01	2001	0																																
17				26/01/2001 06:00	01	2001	0																																
18				26/01/2001 07:00	01	2001	0																																
19				26/01/2001 08:00	01	2001	0,5					1								1				3				1				0,5				9			
20				26/01/2001 09:00	01	2001	0,3																													0			
21				26/01/2001 10:00	01	2001	0,2																													0			
22				26/01/2001 11:00	01	2001	0																																
23				26/01/2001 12:00	01	2001	0																																
24				26/01/2001 13:00	01	2001	0																																
25				26/01/2001 14:00	01	2001	0,3									1				1				1				0,3				0,3				3			
26				26/01/2001 15:00	01	2001	0																																
27				26/01/2001 16:00	01	2001	0																																
28				26/01/2001 17:00	01	2001	0																																
29				26/01/2001 18:00	01	2001	0,9					1								1				2				1,1				0,9				3			
30				26/01/2001 19:00	01	2001	0,2																													0			
31				26/01/2001 20:00	01	2001	0																																

Figure 4.3 – Spreadsheet model to calculate model's inputs. The boxes with numbers refer to the bullets in the steps (i) to (iv) above



- v. After all the event variables were calculated, EMC were calculated for each one precipitation event.
- vi. Calculation of the annual precipitation height. This value was calculated averaging the annual precipitation time series. Due to the lack of data, for Norway the annual precipitation was the average precipitation per year from the hourly precipitation series.
- vii. Calculation of the cumulative seasonal rainfall (CSR). For the calculation of this variable, the year was divided into four seasons:
  - (i) December, January and February;
  - (ii) March, April and May;
  - (iii) June, July and August and
  - (iv) September, October and November.

For each season, it was assigned a characteristic value. This value was obtained by the sum of all the precipitation that had occurred in one of the three months of each season.

- viii. The last variable calculated in the Excel spreadsheet was the AR. This variable is very important as it has great influence in the prediction of PREQUALE.

The explanation of how to calculate the AR was already in the description of PREQUALE (see section 3.1.2). In order to obtain the variable, and after having the IDF curve with two

years of return period, it is only necessary to multiply the value of  $t_c$ , calculated through equation 3.2 to the value in minutes of the IDF curve.

- ix. In order to calculate EMC for each event in the Excel spreadsheet, some physical characteristics of each site were still necessary, such as: DA, AADT, IF, DL, S and CRC.

## STEP 2

Calculation of the predicted SMC. Equation 2.2 was applied for the predicted EMC for each pollutant in the spreadsheet (for HAWRAT and Kayhanian's model) and in the SELDM outputs.

## STEP 3

Comparison of SMC for each highway and pollutant. This comparison was performed considering the four studied tools and the monitored data in each highway. Thus, it was possible to check which tool best predicts SMC at each highway. These results are presented in section 4.4.

## STEP 4

The accuracy of each model was evaluated considering the following indices (Trenouth and Gharabaghi, 2016):

## 4.2. EVALUATION

### 4.2.1. COEFFICIENT OF DETERMINATION

The coefficient of determination ( $R^2$ ) is defined as the squared value of the coefficient of correlation according to Bravais-Pearson. This coefficient estimates the combined dispersion against the single dispersion of the observed and predicted series. The range of this coefficient lies between 0 and 1 which describes how much of the observed dispersion is explained by the prediction. A value of zero means no correlation whereas a value of one means that the dispersion of the prediction is equal to the observation (Krause *et al.*, 2005).

$$R^2 = \frac{[\sum_{i=1}^n (P_i - \bar{P})(O_i - \bar{O})]^2}{\sum_{i=1}^n (P_i - \bar{P})^2 \sum_{i=1}^n (O_i - \bar{O})^2}$$

Where

P<sub>i</sub>= predicted value

P<sub>o</sub>= mean of predicted value

O<sub>i</sub>= observed value

O<sub>o</sub>= mean of observed value

### 4.2.2. ENS COEFFICIENT

The ENS coefficient which was proposed by Nash and Sutcliffe in 1970 (Krause *et al.*, 2005) and assesses the predictive power of the model. Typically, a value of ENS of 0,75 or greater is understood as a result of a good model to predict road runoff. If the value is equal to 1, it is seen as a perfect prediction model (Trenouth and Gharabaghi, 2016). For the case of regression procedures this coefficient is equivalent to  $R^2$ .

$$E_{NS} = 1 - \frac{\sum_{i=1}^n (O_i - P_i)^2}{\sum_{i=1}^n (O_i - \bar{O})^2}$$

where

P<sub>i</sub>= predicted value

O<sub>i</sub>= observed value

N= no of samples

### 4.2.3. ROOT MEAN SQUARE ERROR

The RMSE describes the differences between the observed and predicted values in the units of the variable of study, and is an additional term used to characterise a model performance. This error is always non-negative and the zero value would mean a perfect fit data (Trenouth, 2017).

$$RMSE = \left[ \frac{1}{N} \sum_{i=1}^n (P_i - O_i)^2 \right]^{1/2}$$

where

P<sub>i</sub>= predicted value

O<sub>i</sub>= observed value

N= no of samples

#### 4.3. INPUT DATA

In Table 4.2, the physical characteristics that serve as input of the studied models are presented. In this table, the reference to each one of the highways was made through a code that was firstly presented in Table 3.1.

Table 4.2 – Physical characteristics of road used as inputs in the tools

	CR	DA(m <sup>2</sup> )	DL (m)	S (%)	IF(0-1)	BDF	Pannual (mm)	AR (mm)	AADT (no. veh)
P1	Warm/Wet	22 800	814	2,95	0,412	6	645,95	7,80	27 746
P2	Warm/Wet	1 287	117	7,70 <sup>(c)</sup>	1,000	6	527,98	6,00	16 344
P3	Warm/Wet	5 580	465	3,00 <sup>(c)</sup>	1,000	6	744,43	5,50	2 918
P4	Warm/Wet	15 422	612	3,40 <sup>(c)</sup>	0,850	6	518,33	7,00	24 000
P5	Warm/Wet	287	25	2,50	1,000	6	1013,76	6,00	15 673
P6	Warm/Wet	7 280	520	3,30 <sup>(c)</sup>	1,000	6	708,61	6,00	6 539
N1	Warm/Dry	48 590	1 600	0,20 <sup>(c)</sup>	0,500 <sup>(d)</sup>	6	776,00	3,67	63 000
N2	Warm/Dry	30 510	2 700	0,20 <sup>(c)</sup>	1,000	6	776,00	6,00	63 000
N3	Cold/Dry	22 000	1 630 <sup>(b)</sup>	3,40 <sup>(c)</sup>	1,000	6	834,42 <sup>(e)</sup>	2,50	42 000
F1	Warm/Wet	3 200	275	2,50	1,000	6	786,00	9,00	24 103
F2	Warm/Wet	3 200	275	2,50	0,500 <sup>(d)</sup>	6	786,00	9,00	24 103
I1	Cold/Wet	14 184	1 200	0,94	1,000	6	731,00	3,80	27 500
I2	Cold/Wet	11 368	480	0,50	1,000	6	731,00	3,80	27 500
I3	Cold/Wet	9 600	800	0,50	1,000	6	731,00	3,80	27 500
E1	Warm/Wet	8 755	724	1,10 <sup>(c)</sup>	1,000	6	745,20	2,08	70 000
E2	Warm/Wet	4 348 <sup>(a)</sup>	303	0,66 <sup>(c)</sup>	1,000	6	745,20	1,48	35 000
E3	Warm/Dry	58 680	1 800	2,40 <sup>(c)</sup>	1,000	6	614,80	3,27	78 000
E4	Warm/Wet	20 232	735	3,10 <sup>(c)</sup>	1,000	6	843,40	1,55	24 000
E5	Warm/Wet	2 760	250	0,80 <sup>(c)</sup>	1,000	6	659,70	1,19	64 000

The monitored data is available in: Barbosa and Fernandes, 2012; Leitão *et al.*, 2005; Antunes, 2014; Barbosa, 2007; Brongers, 2011a; Brongers, 2011b; Vollertsen *et al.*, 2007; Mufleh *et al.*, 2010; Higgins, 2006; Moy and Crabtree, 2002a; Moy and Crabtree, 2002b; Moy and Crabtree, 2002c; Moy and Crabtree, 2002d; Moy and Crabtree, 2002e; Moy and Crabtree, 2002f.

The IDF curves used as AR auxiliary calculations are available at: Brandão *et al.*, 2001; Korving *et al.*, 2009; <http://eklima.met.no>; EDF-DTG and Cemagref, 1993 and <https://www.met.ie>.

<sup>(a)</sup> Estimated drainage area by multiplying the length by the section width

<sup>(b)</sup>Estimated drainage length by dividing the available area by the width consulted in Google Earth Pro

<sup>(c)</sup>Estimated slopes through the Google Earth Pro function, elevation profile

<sup>(d)</sup>Assumed impervious fraction

<sup>(e)</sup>Assumed P<sub>annual</sub>

It should be noted that there were some difficulties gathering all the input data. It was necessary to estimate some of the inputs as explained in Table 4.2.

Starting in the first column, it can be verified that a climate region was assigned to each highway studied. This is due to the fact that in this work it was considered that the red lines that limit each climatic region in HAWRAT (only for United Kingdom), were extended in the direction they end to the model (Figure 3.3).

Regarding the drainage area there were not many problems, since the documents referring to each highway, described this characteristic except on the E2, where the determination of the area had to be made by multiplying the length of the section by the width.

In the drainage length column, only N3 did not have the value available to the development of the work. In this case, it was possible to estimate this value, since the area is available as the width was consulted in Google Earth. The same software was used to estimate the missing slopes through the elevation profile function. The estimated slopes are marked in the table with an asterisk.

Regarding the values of impermeable fraction, N1, F2 and I6 have the values of 0,5. This is due to the fact that in the reports in which the study was based, it is only referred that the highways have permeable asphalt. So, the impermeable fraction was considered as 0,5.

In order to have one input that was only needed to run SELDM, it was necessary to estimate a value for BDF. It was decided to consider a value of six for all the highways for two reasons: (i) documents related to the highways characteristics did not have much information in relation to the surrounding lands (forest, bushes, intensive farming, among others) and (ii) in SELDM sensitivity test, several BDF values have been tested, and no great variation was verified.

The annual precipitation was already calculated through the average of annual precipitation data to each site. The only highway which annual precipitation was calculated through the hourly precipitation data was N3 due to the lack of data.

To calculate AR (for PREQUALE), IDF curves were needed. These curves were available for road located in Portugal, Norway, Ireland and Netherlands. For England, it was used an IDF curve which belongs to the Bristol city and for France only one IDF curve for the province of Alps was available.

#### 4.4. RESULTS

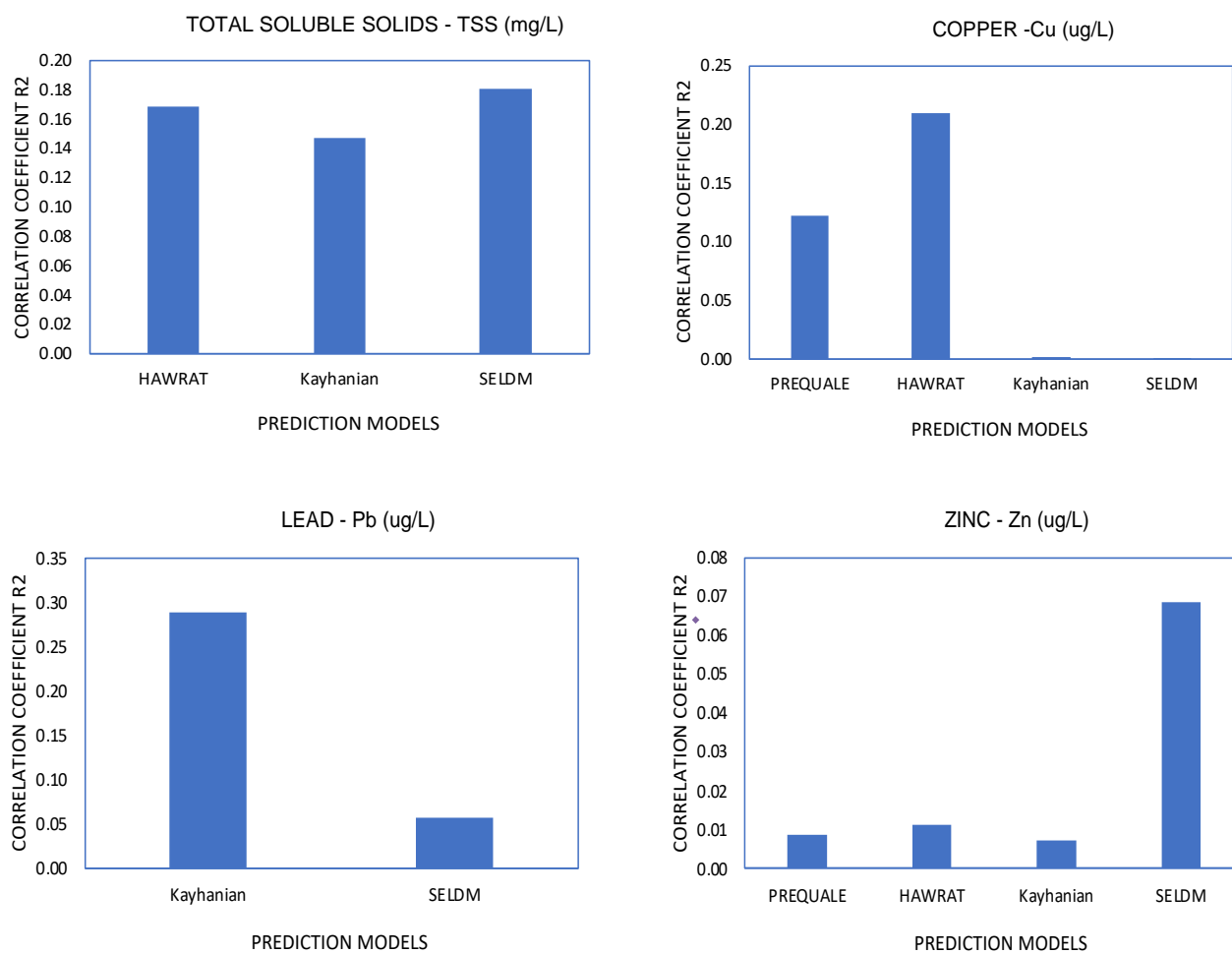


Figure 4.4 – Comparison of correlation coefficients of different predicting tools ( HAWRAT; PREQUALE; SELDM; KAYHANIAN )

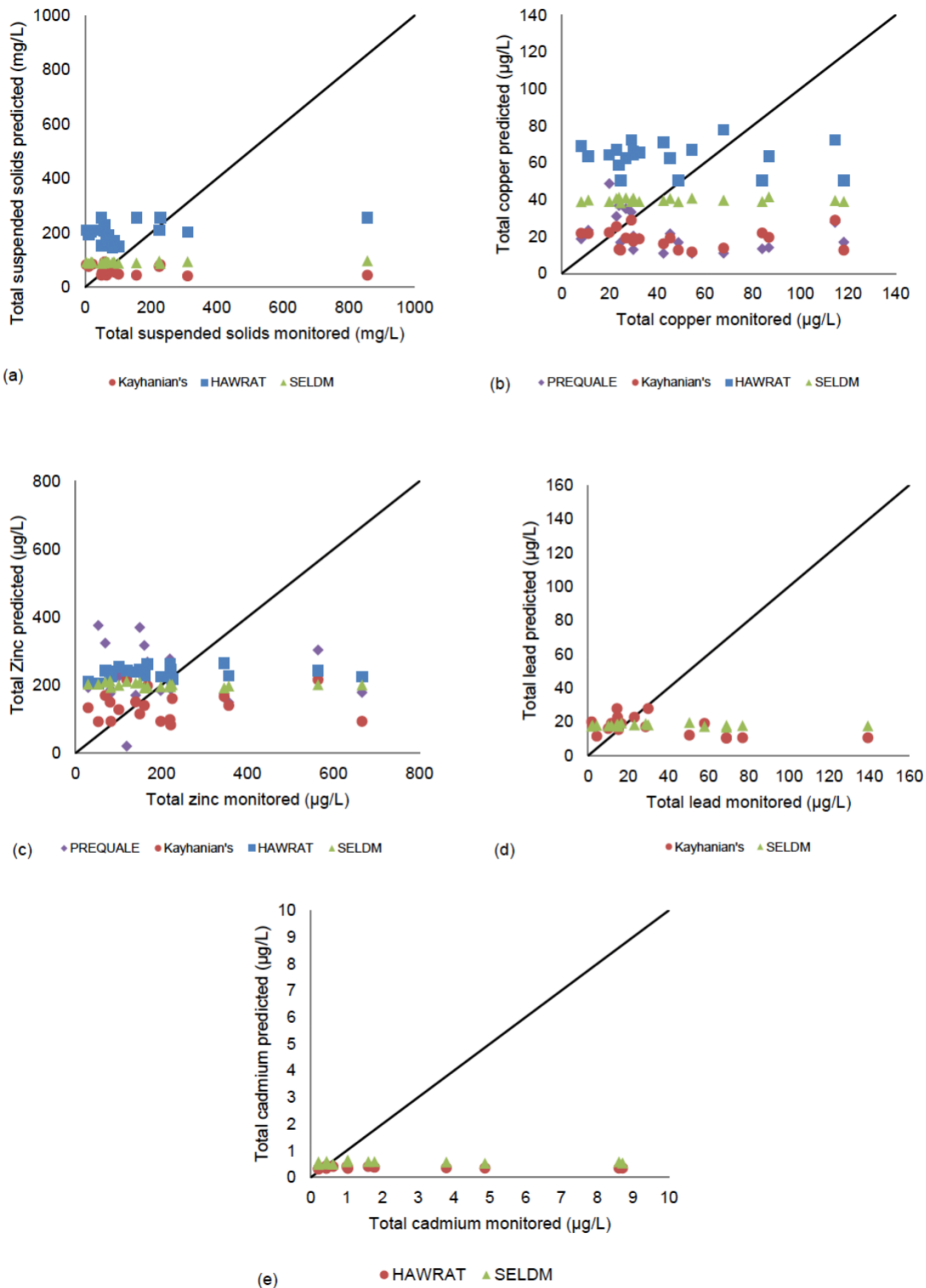


Figure 4.5 – Comparison of observation and predicted data of different predicting tools ( HAWRAT; PREQUALE; SELDM; KAYHANIAN )

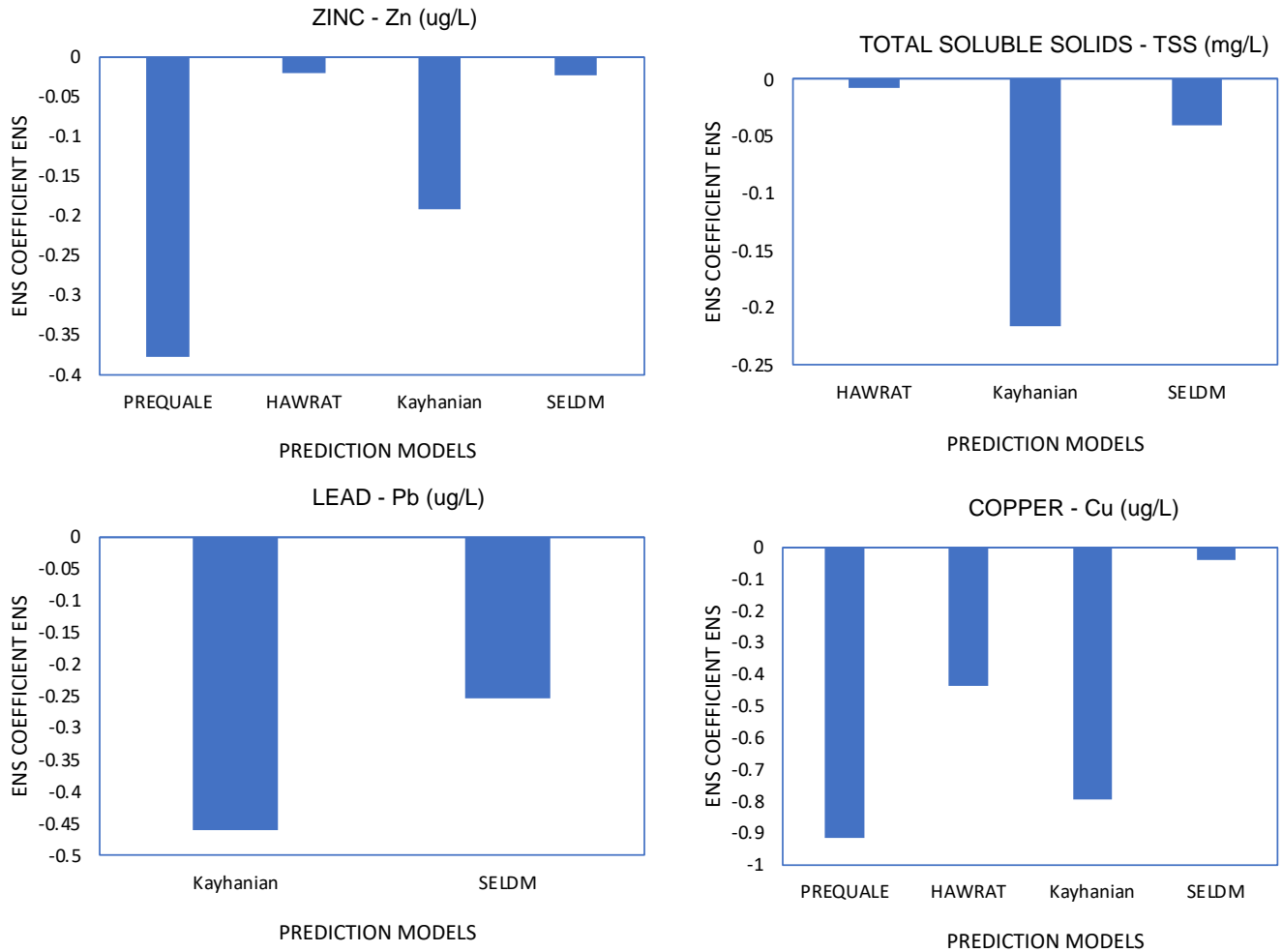


Figure 4.6 – Comparison of ENS coefficient of different predicting tools ( HAWRAT; PREQUALE; SELDM; KAYHANIAN )

At first glance, it is easily observed that any of the models is robust enough to predict road runoff pollution, to the European roads. None of the models predicts the variation of the monitored values. Moreover, none of the models results vary much with great variations of monitored data, which indicates that the models do not possess great sensitivity to the input variations. To corroborate this visual analysis, the error indices were calculated (Table 4.3) and analysed. It is important to notice that although PREQUALE is able to predict TSS, the results of this prediction are not available in the Figure 4.4, because the results are very disperse and very high, thus the graphical representation of these would not allow the visualization of the remaining results.

Table 4.3 – Error indices table

		Error Indices			
		$R^2$	$E_{NS}$	CRM	RMSE
<b>TSS</b>	HAWRAT	$1,682 \times 10^{-1}$	-0,008	-0,500	193,517
	Kayhanian	$1,468 \times 10^{-1}$	-0,216	0,524	212,501
	SELDM	$1,803 \times 10^{-1}$	-0,041	0,329	196,671
<b>Copper</b>	PREQUALE	$1,219 \times 10^{-1}$	-0,916	0,523	43,549
	HAWRAT	$2,090 \times 10^{-2}$	-0,435	-0,377	37,692
	Kayhanian	$1,900 \times 10^{-3}$	-0,792	0,591	42,112
	SELDM	$4,000 \times 10^{-4}$	-0,042	0,137	32,123
<b>Zinc</b>	PREQUALE	$8,700 \times 10^{-3}$	-0,377	-0,077	193,738
	HAWRAT	$1,110 \times 10^{-2}$	-0,021	-0,142	166,839
	Kayhanian	$7,200 \times 10^{-3}$	-0,192	0,330	180,262
	SELDM	$6,850 \times 10^{-2}$	-0,023	0,032	166,973
<b>Lead</b>	Kayhanian	$2,890 \times 10^{-1}$	-0,461	0,509	42,047
	SELDM	$5,750 \times 10^{-2}$	-0,254	0,488	38,961
<b>Cadmium</b>	HAWRAT	$1,270 \times 10^{-2}$	-0,658	0,866	3,778
	SELDM	$3,900 \times 10^{-3}$	-0,5510	0,7957	3,6541

Four error indices were used in order to evaluate the performance of the models. The analysis of the Table 4.3 was made by error index:

- i. The highest  $R^2$  was of 0.28 which is a very low value to guarantee a robust model. However, this information is not enough since it only allows to conclude about a tendency between monitored and predicted data. This means that a high  $R^2$  could be obtained even if the predicted data was very different from the monitored data, as long as there was a linear tendency.
- ii. Although both visual and coefficient of determination analysis show the models were not robust enough, the efficiency coefficient (ENS) was still analysed to assess overall model efficiency and the previous analysis. Since all values were below 0,5, it can be concluded that the model is not robust, as previously suggested by the coefficient of determination analysis.
- iii. Regarding the CRM, most values were not close to zero. This may suggest an under prediction, in the case of positive values, or an over prediction in the case of the negative values. There was one exception: zinc concentration prediction through SELDM which presents and CRM of 0,0321. Still, looking at the graphical representation (Figure 4.4) it is observable that the relation between predicted and monitored data is not satisfactory. Hence, CRM is not enough to determine if a model is well adjusted to the data.
- iv. Regarding RMSE, all of indices calculated are much larger than zero ( $>32 \mu\text{g/L}$  or  $>193 \text{ mg/L}$  in the case of TSS), except for cadmium ( $<4 \mu\text{g/L}$ ). Still, in this case, the error is considered big since the average of the observed data ( $2,74 \mu\text{g/L}$ ) is lower than the error index, which indicates that the error is significant.

#### 4.5. CRITICAL REVIEW OF THE MODELS

PREQUALE is a very simple tool of direct application and does not present great problems. In order



to obtain the necessary data to run the model, almost all the data are also easily obtainable, except one input, the AR. This input requires some data to be obtained for intermediate calculations that are not always available in previously monitored locations such as the length of the drainage section and the variation in height. In addition to these two inputs, the final stage of this input calculation involves the use of an IDF curve of the site, which has become quite complicated to obtain. Indeed, the results may be biased since it was not possible to use the correct IDF curve for UK and France.

HAWRAT is easy to apply having only one input that makes it difficult to apply at European level. For the application of this model it is necessary to indicate the climatic area for each road. However, it only sets out the zones for the United Kingdom, being necessary an adaptation if it is intended to simulate outside UK.

As PREQUALE and HAWRAT, Kayhanian's model only had one input that made it difficult to use. This model uses as input the CSR, which is a variable that is not widely used in Europe, and it was difficult to decide how it should be analysed. It was decided that only four seasons of the year would be considered.

SELDM was the hardest model to work. This model was the only one which used a complex graphical interface. SELDM has several forms to fill with a lot of mandatory inputs and part of them is difficult to obtain, like BDF.

Another feature that makes SELDM more difficult to apply than the other models is its increasing difficulty of use when the model is being applied outside the USA. For the USA, the precipitation data is not needed as input, because by setting the highway location, SELDM automatically fills these data. If the case study is outside of the USA, which is the case, it is needed to complete several precipitation statistics as indicated in the second chapter of the annexes.

Although these models had shown great robustness in previous studies (*e.g.* Barbosa, 2007; Barbosa *et al.* 2011; Dempsey and Song, 2008; Kayhanian *et al.*, 2006 and Granato and Jones, 2015), this was not observed in the present work. The greater robustness in those studies could be possibly explained by the fact that each model was tested in sites that are geographically similar to the ones that were used to calibrate them.

The Table 4.4 is presented below in order to show the pros and cons of each model.

Table 4.4 – Table regarding the pros and cons of each model

MODELS	PROS	CONS
PREQUALE	<p>Simple regression equation</p> <p>Easy to calculate</p> <p>ADP is not used in PREQUALE. It seems to be an advantage since no correlation between road runoff concentration and ADP was noticed in portuguese and international studies.</p> <p>Use IDF curves which are difficult to obtain in UK</p> <p>Predict SMC instead of EMC</p>	<p>The number of monitored roads used to construct the model (six), does not represent a robust quantity and diversity of road characteristics that may be characteristic from all Europe;</p> <p>The model was created from a regression analysis that is only based on four variables.</p> <p>Average annual rainfall volume with the same duration</p> <p>The basin time of concentration (AR), was very hard to find in the monitored data from Europe.</p>
HAWRAT	<p>Easy to calculate;</p> <p>Input data easily available for users;</p> <p>Calibrated with a robust number of monitored roads for UK, which results in good predicting results</p>	<p>Characteristics variability to apply to all Europe is very low;</p> <p>Climate region data of each road is required which is hard to get</p> <p>HAWRAT does not appear to have sensitivity to great variations of SMC, being very constant from site to site.</p>
Kayhanian	<p>Easy to calculate;</p> <p>Input data easily available for users.</p>	<p>One input that's very difficult to obtain in Europe</p> <p>CSR is not widely used in Europe</p> <p>Model made entirely for USA roads, hard to adopt to European studies</p> <p>This method does not appear to have a direct relation between monitored data and predicted data for the highest values of the monitored data.</p>
SELDM	<p>The model defines a range of values where the output should be;</p> <p>The capability to insert precipitation data manually, allows the user to predict road runoff in a climate change scenario.</p>	<p>Hardest model to work due to graphical interface</p> <p>It has a lot of mandatory inputs which are hard to obtain such as BDF</p> <p>Difficult to use due to different rules for different countries. In USA Pannual is not needed as it is calculated automatically, but not the case in other countries</p>

After the analysis of the results of these four models, it was possible to conclude that none of the models was robust enough to be applied to European roads.

## 5. PROPOSED MODEL

### 5.1. DEVELOPMENT OF A NEW MODEL

Considering the gathered monitored data and the poor performance of the predicting models, the development of a new model is proposed.

The proposed model uses a multiparametric equation that allows the user to predict SMC for TSS (mg/L), copper (µg/L), zinc (µg/L), lead (µg/L) and cadmium (µg/L). This model is based on the following principles:

- (i) input data easily available for the user;
- (ii) easiness of calculation and
- (iii) more robust results for Europe as a whole than the previously studied tools.

In order to accommodate the several factors that may influence the pollutant concentrations, the following variables were considered to the model:

- i. Drainage area (DA in m<sup>2</sup>) – Area that contributes to the runoff;
- ii. Drainage length (DL in m) – Length of the road;
- iii. Impervious fraction (IF in 0-1) – The fraction of the DA that is impervious;
- iv. Annual precipitation (P<sub>annual</sub> in mm) – The average of annual precipitations of each site;
- v. Annual average daily traffic (AADT in no of vehicles) – The number of vehicles that pass daily in the site.

The multiparametric equation takes the following forms:

$$SMC = \delta_1 + \delta_2 \times DA + \delta_3 \times DL + \delta_4 \times IF + \delta_5 \times P_{\text{annual}} + \delta_6 \times AADT \quad (5.1)$$

$$LN(SMC) = \delta_1 + \delta_2 \times DA + \delta_3 \times DL + \delta_4 \times IF + \delta_5 \times P_{\text{annual}} + \delta_6 \times AADT \quad (5.2)$$

$$LN(SMC) = \delta_1 + \delta_2 \times LN(DA) + \delta_3 \times LN(DL) + \delta_4 \times LN(IF) + \delta_5 \times LN(P_{\text{annual}}) + \delta_6 \times LN(AADT) \quad (5.3)$$

Where the SMC is given through the form of a natural logarithm. This model was calibrated with the characteristics of the roads that were presented in Table 4.2 and the regression and correlation coefficients that resulted from the adjusted multiparametric equation of the roads SMC are presented in Table 5.1. It is important to emphasize that for the development of this equation it was not took into account which are the variables with more weight, because the main objective was to have an equation with the most easily obtainable parameters.

Table 5.1 – Correlation coefficients

REGRESSION COEFFICIENTS USED BY MODEL NO 2						
	$\beta_0$	a	b	c	d	e
TSS (mg/L)	1.565643	1.3436E-06	-0.000803915	0.000494797	-3.93125e-7	2.829429
Cu(µg/L)	0.483356	0.00000169	-0.000052	0.0008896	0.00000474	-0.018957
Zn(µg/L)	-0.243942	2.8455E-06	-0.000102777	0.163403	0.00611197	0.000013256
Pb(µg/L)	0.897831	6.86E-07	0.000002195	0.483231	0.00173504	0.00000467
Cd(µg/L)	-1.538046	-1.26E-05	0.0002023	2.219996	-0.000526889	-0.0000035

5.2. RESULTS AND PERFORMANCE EVALUATION

In the first part of this section are presented the predictions in Figure 5.1.

5.2.1. TOTAL SUSPENDED SOLIDS RESULTS

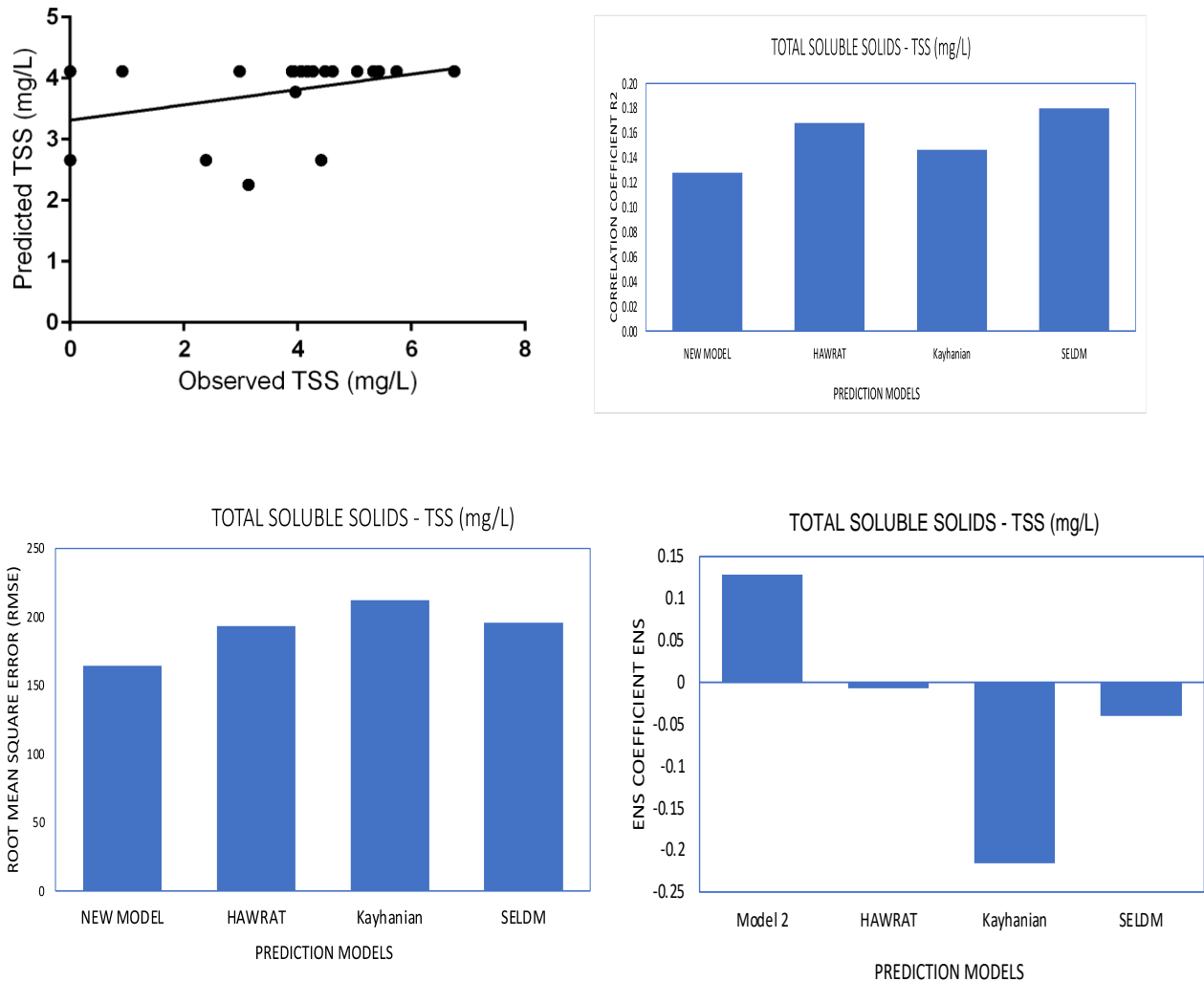


Figure 5.1 – Comparison of Correlation, ENS coefficient and RMSE of TSS (mg/L)

5.2.2. COPPER CU (ug/L) RESULTS

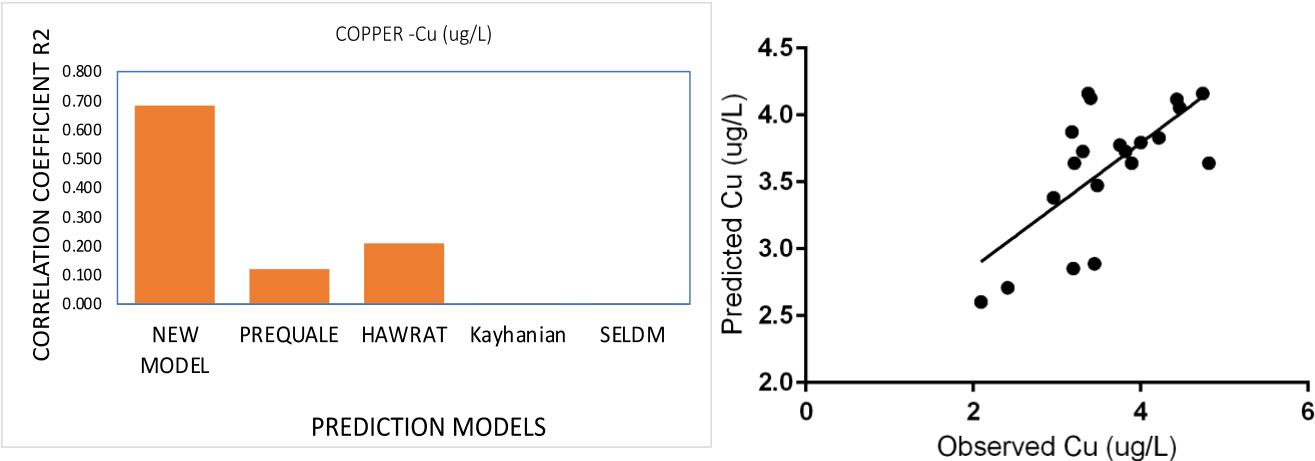




Figure 5.2 – Comparison of Correlation, ENS coefficient and RMSE of Copper Cu(ug/L)

### 5.2.3. ZINC ZN (ug/L) RESULTS

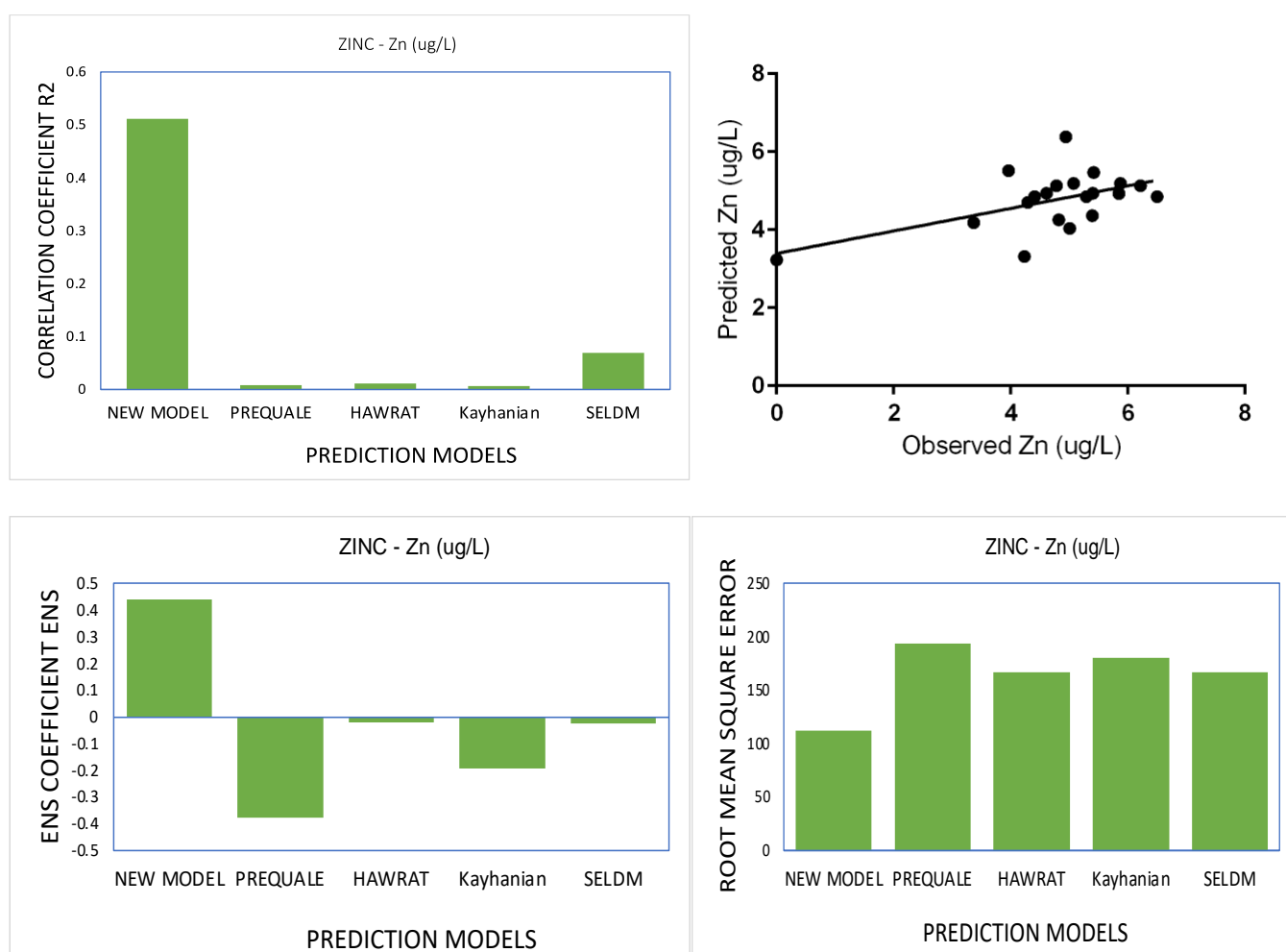


Figure 5.3 – Comparison of Correlation, ENS coefficient and RMSE of Zinc, Zn (ug/L)

### 5.2.4. LEAD Pb (ug/L) RESULTS

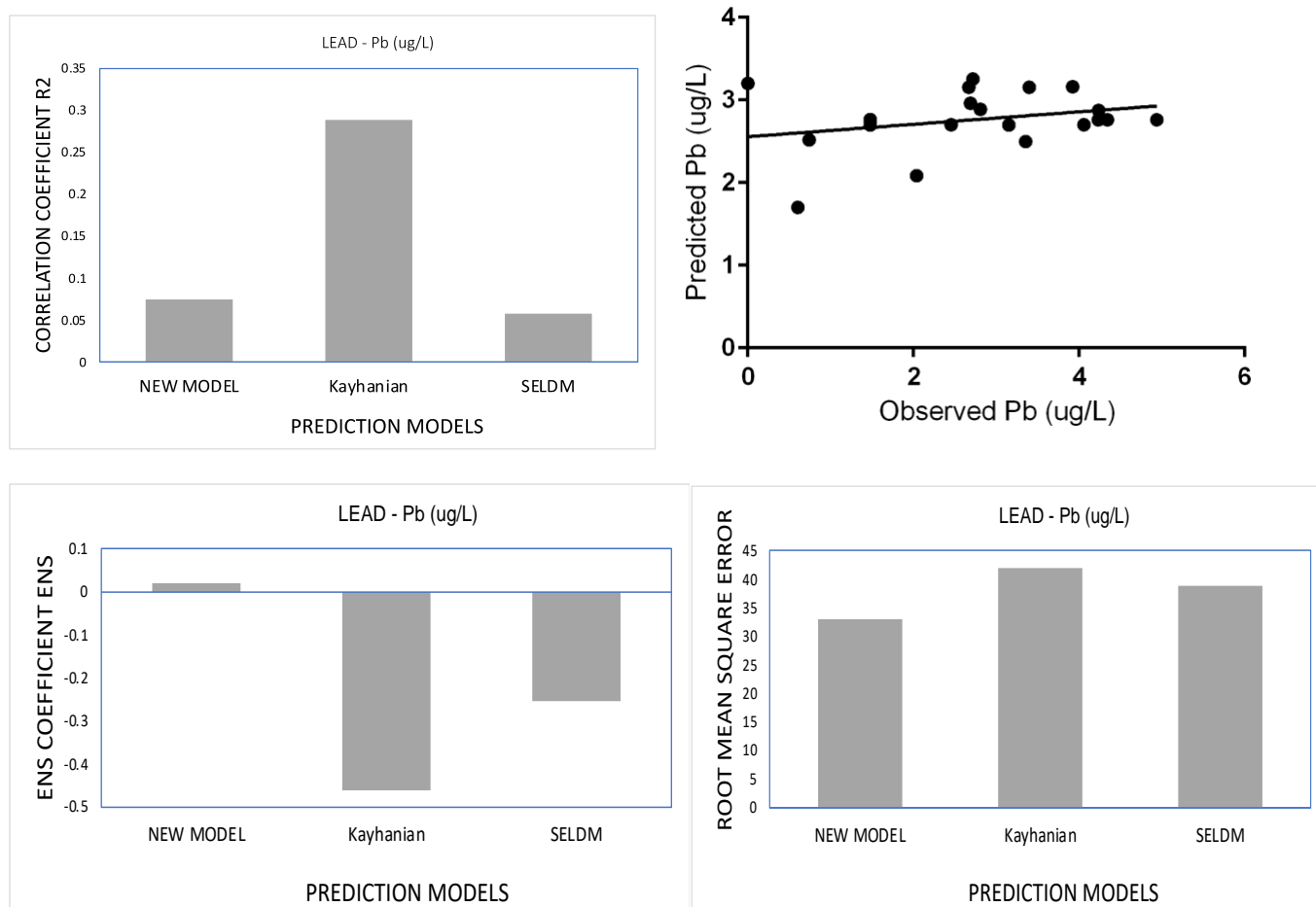


Figure 5.4 – Comparison of Correlation, ENS coefficient and RMSE of Lead, Pb (ug/L)

## 5.2.5. CONCLUSIONS

By the visual analysis of Figure 5.1, it can be observed that the predicted values follow the tendency of the monitored values. The correlation between predicted and monitored seems stronger than the ones obtained for the four models studied previously. Still, there are some differences between pollutants. This correlation seems to be stronger for copper and zinc than for the remaining pollutants.

After the development of the new model it was necessary to check its validity through the calculation of the errors that were used to evaluate the performance of the four tools studied (Table 5.2):

Table 5.2 – New model error indices

		R2	ENS	RMSE
TSS	NEW MODEL	0.13	0.128	164
	HAWRAT	0.17	-0.008	193
	Kayhanian	0.15	-0.216	212
	SELDM	0.18	-0.041	196
Copper	NEW MODEL	0.682	0.548	21
	PREQUALE	0.122	-0.916	44
	HAWRAT	0.209	-0.435	38

	Kayhanian	0.002	-0.792	42
	SELDM	0.000	-0.042	32
Zinc	NEW MODEL	0.511	0.44	112
	PREQUALE	0.009	-0.377	194
	HAWRAT	0.011	-0.021	167
	Kayhanian	0.007	-0.192	180
	SELDM	0.069	-0.023	167
Lead	NEW MODEL	0.075	0.021	33
	Kayhanian	0.29	-0.461	42
	SELDM	0.06	-0.254	39

- i. Regarding the  $R^2$  it was noticed that none of the values were close to 1. However, in comparison to the models studied previously, this model presents much higher  $R^2$ , except for lead and TSS.
- ii. None of the parameters achieved the perfect value of ENS (ENS=1), however two of the indices indicate a reasonably good performance (copper and zinc).
- iii. Concerning RMSE, all indices calculated are much larger than zero. Still, in this case, the error is considered big since the average of the observed data is very close to the error index, which indicates that the error is very large.
- iv. (2,70  $\mu\text{g/L}$ ). Still, in this case, the error is considered big since the average of the observed data (2,74  $\mu\text{g/L}$ ) is very close to the error index, which indicates that the error is very large.

#### 5.2.6. CRITICAL REVIEW OF THE MODEL

The developed model has some characteristics which are critically reviewed in this section:

- i. This new model is a multiple linear regression equation and is based on five variables (DA,DL, IF, Pannual and AADT) that were chosen only based on availability of the variables for each road;
- ii. AADT is one of the input variables. No clear correlation was found by the FHWA (1996) (in Leitão *et al.* 2005) between this variable and the quality of road runoff. In this way, it was concluded by Leitão *et al.* (2005), that every model that is based only in this variable should be carefully evaluated. However, it was already shown during this work that the parameters used to calculate road runoff pollution are still not widely established, as referred in the section 2.1.1, where it is said that Irish *et al.* (1995) consider the AADT as one of the three parameters regarding the volume of traffic

- iii. This tool presents limitations based on the few data that support the construction of the regression equation. This equation was only based on the data studied in the present dissertation.
- iv. The data for which the model was tested were the same as those used for the calibration.

#### 5.2.7. CONCLUSIONS AND FURTHER WORK

The evaluation of the road runoff predicting tools is very important to understand the environmental impact caused by road runoff (diffuse pollution). In a broader view, the control of road runoff pollution may also help the conservation of the receiving water bodies (*e.g.* reduction of runoff pollutant concentration entering to a reservoir, which may lead to an decrease of the cost in the operation of drinking water treatment).

In this work, it was possible to obtain a better understanding of the pollutant characteristics of some European roads through the collection and analysis of several monitored events.

Regarding the study of the predicting tools, it was possible to conclude that none of the models was robust enough to be applied to European roads as a whole. This could be explained by the monitored data used to calibrate each model. Apart from SELDM each model is focused in limited geographical boundaries. Summing up, the following conclusions can be drawn for each model.

- i. PREQUALE is a very interesting model. It is simple to use; all the input data is reasonably easy to get and apply and the regression parameters used were chosen through a principal component analysis. However, this tool was only calibrated for Portugal and even for this country, this model is not complete, as it was assumed by the authors which indicate that it needs to be continuously updated.
- ii. HAWRAT is also a rather user-friendly model with easy application, however this model aims at predicting EMC. The probable reason for this model to not produce robust values in Europe is the same as PREQUALE. This model was only based on monitored roads from UK which may have specific characteristics.
- iii. Kayhanian's model presents the same characteristics of PREQUALE and HAWRAT, mainly since all three models are based in one equation (per pollutant). Kayhanian's model was calibrated with monitored data from California, which could be similar to Portugal in climate conditions but is not similar to the rest of the countries studied.
- iv. SELDM is a quite different model. This model presents a much more complex GUI than HAWRAT. This model is based on a data set of precipitations and previous monitored data. After the selection of one area (the model divides the USA in several *Ecoregions*), it stochastically predicts precipitation event series. Then, each event has several quality and precipitation parameters (*e.g.*



concentrations and rain height) associated. The output series for each pollutant is very similar even with very different roads, which may explain that the model tries to define an approximated SMC. However, the defined value by the model is very different than the averages presented by the monitored data studied throughout this work.

It could be said that the main reason that affects the imprecisions of road runoff prediction in Europe is the fact that the models were calibrated to a defined country or region.

Moreover, it should be recognized that the two more complex models, HAWRAT and SELDM have a broader application than just predict pollutant concentrations. As explained in section 3.2, these two models have different steps and their ultimate objective is the evaluation of the impact of the road runoff in the receiving water bodies and the need for treatment systems. Therefore, the road runoff prediction is a small part of these models.

Since the results were not satisfactory, a new regression model was developed. This model was developed with a regression of several variables, which serve as input of the model. The choice of the variables was made considering essentially their availability to the users. The roads used to construct the regression and to test the model were the same. This model will gain some robustness if more monitored data could be used to calibrate it. In this way, the best way to predict road runoff concentrations could be to take advantage from the knowledge already existent in Europe (in this case from Portugal and UK) to use and construct models for each country instead of trying to creating a general model for all Europe.

Regarding the data of the predicting models, the conclusion is that in the highways runoff the climatic characteristics and land use have as much or more impact than some of the variables usually studied in the models.

If the perspective is to build a model that serves all Europe, some considerations should be taken in the future: (i) Build a SMC regression model for each Köppen-Geiger area or (ii) continue the PREQUALE process, for Portugal and Europe. PREQUALE was developed based on a principle components analysis, and predicts SMC as it is advised by some specialists. Some authors defend that the road runoff concentration out of the urban areas does not vary much with the ADP. In this way, PREQUALE with more monitored data could achieve a determination coefficient for each pollutant, much better than the one presented in this work. After the improvement of these tools or after obtaining the largest number of monitored data possible, it would be possible to define which is the best model to be applied in Europe and start to develop some complementary studies that may allow the European environmental and traffic agencies perform robust environmental evaluation.

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## Annexes

### Annex I – Monitored EMC of each studied highway

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 1	P1	26,200	32,907	276,984	9,000	0,136	0,200	ND
		15,600	20,758	157,625	4,176	0,099	1,900	15,283
		10,300	7,506	73,422	ND	0,059	0,100	ND
		13,800	13,883	92,633	ND	0,048	0,121	0,762
		9,800	13,732	59,899	ND	0,154	0,036	ND
		1,200	10,815	21,078	ND	0,055	0,092	0,477
		100,800	23,540	153,338	3,771	0,095	0,472	0,743
		7,000	30,766	135,082	2,785	0,102	0,005	ND
		36,000	21,713	189,094	5,645	0,073	0,600	ND
		8,900	16,783	81,536	0,914	0,095	0,122	0,312
A 2	P2	5,000	5,400	40,000	ND	ND	0,100	ND
		2,400	14,000	70,000	0,100	ND	0,120	ND
		0,097	14,000	97,000	4,100	ND	0,110	ND
A 6	P3	1,600	2,400	217,000	1,000	ND	0,118	ND
		6,700	9,100	1443,000	1,000	ND	0,211	ND
		3,200	4,800	46,000	1,000	ND	0,080	ND
		60,300	14,000	168,000	4,700	ND	0,672	ND
		19,100	11,000	104,000	1,000	ND	0,766	ND
		27,000	7,300	97,000	2,300	ND	0,273	ND
A 22	P4	40,600	30,000	ND	10,000	ND	2,400	ND
		50,000	30,000	ND	20,000	ND	2,800	ND
		82,400	20,000	ND	20,000	ND	2,200	ND
		88,000	20,000	ND	20,000	ND	1,800	ND
		79,500	30,000	ND	30,000	ND	3,300	ND
		32,100	20,000	ND	20,000	ND	1,200	ND
		47,900	20,000	ND	30,000	ND	2,100	ND
		25,700	20,000	ND	30,000	ND	0,900	ND
		25,800	30,000	ND	30,000	ND	0,700	ND
A 25	P5	26,600	63,600	133,190	28,700	ND	1,269	ND
		101,000	73,000	234,960	55,500	ND	2,723	ND
		47,700	58,900	162,980	35,700	ND	1,855	ND
		28,900	35,500	81,000	33,000	ND	0,898	ND
		43,600	84,300	81,690	43,100	ND	1,909	ND
		96,700	76,600	134,070	41,700	ND	3,712	ND
		159,600	175,900	406,850	69,700	ND	7,352	ND
		31,200	47,400	96,410	20,000	ND	1,600	ND
		109,400	86,100	200,910	49,100	ND	4,972	ND
		78,200	38,800	94,690	66,400	ND	2,653	ND
		96,000	38,800	139,280	21,800	ND	1,997	ND
		207,300	88,000	252,320	35,600	ND	4,882	ND

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 25	P5	139,500	75,000	213,800	25,900	ND	3,001	ND
		25,700	32,000	82,620	18,000	ND	0,748	ND
		16,100	22,400	81,760	18,000	ND	0,774	ND
		87,500	655,800	274,660	46,200	ND	1,307	ND
		31,200	47,900	126,000	18,000	ND	1,230	ND
		9,200	5,500	30,220	6,500	ND	0,195	ND
		53,400	49,800	153,270	20,300	ND	2,400	ND
		17,400	23,700	88,680	18,000	ND	0,701	ND
		30,700	217,500	122,290	21,900	ND	0,695	ND
		52,000	70,400	127,360	18,000	ND	1,322	ND
		32,100	79,900	102,980	18,000	ND	0,653	ND
		20,300	57,800	88,630	18,000	ND	0,774	ND
		50,000	134,800	145,610	18,000	ND	1,376	ND
		8,600	44,900	88,440	18,000	ND	0,315	ND
		2,200	10,500	81,310	18,000	ND	0,035	ND
		15,400	33,800	81,250	18,000	ND	0,120	ND
		45,000	78,300	168,360	18,000	ND	2,414	ND
		75,500	99,000	115,180	22,400	ND	1,859	ND
IP 6	P6	365,399	69,844	147,121	11,865	1,429	ND	9,500
		191,292	43,455	85,091	2,455	1,000	ND	3,000
		23,804	30,565	17,529	23,413	1,000	ND	3,000
		214,455	31,841	55,098	3,841	1,000	ND	3,000
		509,973	46,459	106,622	14,581	1,000	ND	16,973
		60,011	19,123	86,476	1,987	ND	ND	ND
		241,500	7,351	48,350	2,235	ND	ND	ND
		50,218	3,000	39,102	1,000	ND	ND	ND
A 27 - pervious	N1	ND	180,000	1300,000	52,000	2,000	ND	21,000
		ND	88,000	250,000	15,000	1,000	ND	19,000
		ND	11,000	31,000	8,000	2,000	ND	10,000
		ND	15,000	220,000	5,000	1,000	ND	52,000
		ND	420,000	1900,000	130,000	2,000	ND	19,000
		ND	73,000	230,000	13,000	ND	ND	ND
		ND	16,000	59,000	6,000	ND	ND	ND
		ND	ND	17,000	10,000	ND	ND	ND
A 27 - Impervious	N2	ND	31,000	130,000	17,000	1,000	ND	14,000
		ND	17,000	54,000	14,000	1,000	ND	10,000
		ND	21,000	60,000	9,000	1,000	ND	10,000
		ND	30,000	160,000	12,000	1,000	ND	10,000
		ND	77,000	270,000	27,000	1,000	ND	29,000
		ND	11,000	92,000	11,000	1,000	ND	10,000
		ND	17,000	66,000	11,000	1,000	ND	12,000
E 6	N3	99,000	73,000	155,000	5,600	0,080	ND	ND
		181,000	74,200	241,000	12,000	0,130	ND	ND



Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
E 6	N3	388,000	118,000	391,000	29,000	0,340	ND	ND
		522,000	129,000	380,000	28,000	0,350	ND	ND
		96,000	48,600	110,000	8,900	0,120	ND	ND
		107,000	37,300	67,900	1,700	ND	ND	ND
		425,000	133,000	319,000	25,000	0,210	ND	ND
		115,000	25,400	66,800	9,400	0,080	ND	ND
		401,000	84,000	285,000	24,000	0,230	ND	ND
		171,000	60,800	63,300	1,500	ND	ND	ND
		128,000	48,600	175,000	8,600	0,100	ND	ND
		182,000	114,000	189,000	10,000	0,060	ND	ND
		91,000	84,300	124,000	8,900	0,120	ND	ND
		184,000	130,000	96,500	7,600	1,000	ND	ND
		78,000	85,100	140,000	10,000	0,100	ND	ND
		93,000	66,800	149,000	10,000	0,110	ND	ND
		48,000	90,700	128,000	9,800	0,070	ND	ND
		240,000	92,000	323,000	23,000	0,280	ND	ND
		344,000	103,000	371,000	27,000	0,300	ND	ND
		430,000	102,000	408,000	26,000	0,290	ND	ND
		259,000	61,100	235,000	19,000	0,150	ND	ND
		275,000	94,000	266,000	16,000	0,230	ND	ND
		159,000	77,000	211,000	8,400	0,200	ND	ND
		295,000	112,000	355,000	19,000	0,240	ND	ND
		606,000	118,000	544,000	33,000	0,350	ND	ND
		39,000	41,400	66,900	4,200	0,060	ND	ND
		201,000	72,600	170,000	10,000	0,080	ND	ND
		216,000	78,600	266,000	16,000	0,190	ND	ND
A 11 – pervious	F1	65,900	49,000	284,000	60,600	0,610	ND	ND
		94,000	92,500	613,000	92,900	1,010	ND	ND
		47,400	31,200	245,000	28,400	0,490	ND	ND
		46,700	32,400	240,000	27,600	1,640	ND	ND
		36,100	73,700	254,000	33,600	0,630	ND	ND
		61,300	42,600	238,000	49,400	1,600	ND	ND
		31,300	31,400	145,000	35,600	0,470	ND	ND
		27,300	14,300	104,000	21,200	0,460	ND	ND
		117,000	48,000	392,000	70,500	0,740	ND	ND
		25,100	56,600	434,000	21,100	0,370	ND	ND
		32,000	89,700	659,000	43,000	1,870	ND	ND
		220,000	94,200	805,000	126,000	1,030	ND	ND
		238,000	80,000	615,000	138,000	2,130	ND	ND
		20,100	24,800	143,000	21,700	0,510	ND	ND
		24,400	45,800	269,000	17,000	0,860	ND	ND
		20,700	21,600	212,000	18,000	0,280	ND	ND
		92,500	40,500	263,000	62,000	1,020	ND	ND

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 11 - pervious	F1	70,000	146,000	709,000	53,000	1,260	ND	ND
		78,300	42,300	380,000	46,200	0,430	ND	ND
		20,700	35,700	131,000	18,600	0,440	ND	ND
		37,500	27,200	163,000	37,400	0,800	ND	ND
		34,300	22,900	166,000	25,000	1,310	ND	ND
		30,200	26,400	145,000	26,100	0,470	ND	ND
		24,600	25,400	126,000	23,400	0,330	ND	ND
		33,200	23,300	152,000	21,100	0,380	ND	ND
		42,000	24,500	124,000	32,900	0,230	ND	ND
		16,300	50,600	415,000	20,100	1,170	ND	ND
		21,500	28,400	248,000	29,800	0,370	ND	ND
		27,000	31,000	174,000	23,900	0,360	ND	ND
		29,000	11,200	120,000	13,700	0,790	ND	ND
		35,600	25,100	133,000	39,600	0,370	ND	ND
		43,200	28,700	311,000	61,400	2,790	ND	ND
		56,900	31,000	236,000	66,000	1,610	ND	ND
		44,100	27,100	228,000	56,600	0,210	ND	ND
		31,900	31,400	197,000	45,300	0,420	ND	ND
		143,000	36,700	618,000	156,000	1,200	ND	ND
		180,000	83,700	576,000	155,000	1,850	ND	ND
		83,400	39,400	274,000	67,600	0,510	ND	ND
		44,600	24,000	181,000	33,200	0,390	ND	ND
		67,100	29,500	268,000	42,000	1,350	ND	ND
		59,300	26,200	174,000	46,900	2,420	ND	ND
		267,000	63,100	408,000	180,000	0,550	ND	ND
		138,000	73,400	527,000	118,000	1,000	ND	ND
		113,000	73,100	269,000	93,500	0,520	ND	ND
		71,000	32,900	1544,000	44,200	1,680	ND	ND
		125,000	69,000	554,000	95,900	0,450	ND	ND
		70,400	46,900	465,000	71,800	1,240	ND	ND
		211,000	98,600	1322,000	188,000	4,160	ND	ND
		48,500	27,000	195,000	39,600	3,760	ND	ND
A 11 – impervious	F2	3,900	16,900	131,000	3,900	0,340	ND	ND
		48,100	106,400	352,000	33,000	0,150	ND	ND
		14,400	41,500	246,000	10,300	0,120	ND	ND
		22,800	24,500	121,000	17,900	0,040	ND	ND
		10,500	21,500	83,000	8,600	0,140	ND	ND
		63,100	30,300	227,000	19,500	0,230	ND	ND
		8,300	14,100	99,000	8,300	0,120	ND	ND
		5,600	15,500	67,000	7,700	0,050	ND	ND
		4,100	ND	66,000	3,800	0,190	ND	ND
		4,400	11,700	56,000	4,800	0,110	ND	ND
		52,700	150,100	368,000	60,800	0,190	ND	ND

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 11 – impervious	F2	33,800	98,300	198,000	24,900	1,890	ND	ND
		12,600	48,600	130,000	31,200	0,160	ND	ND
		9,900	46,200	134,000	10,100	0,090	ND	ND
		8,100	19,400	80,000	6,200	0,160	ND	ND
		2,700	9,500	44,000	2,200	0,040	ND	ND
		17,000	72,200	183,000	12,800	0,160	ND	ND
		7,700	42,400	133,000	21,000	0,150	ND	ND
		4,200	31,400	110,000	8,300	0,160	ND	ND
		6,000	18,100	50,000	6,900	1,070	ND	ND
		5,500	22,100	63,000	6,800	0,600	ND	ND
		9,700	16,900	51,000	6,700	0,300	ND	ND
		5,900	16,100	78,000	9,300	0,100	ND	ND
		6,900	31,400	125,000	8,800	0,410	ND	ND
		7,700	16,100	65,000	8,500	0,150	ND	ND
		5,700	4,900	44,000	5,100	0,250	ND	ND
		2,300	6,100	58,000	6,500	0,420	ND	ND
		5,000	19,600	86,000	9,500	1,110	ND	ND
		6,900	15,100	78,000	12,300	0,110	ND	ND
		2,400	8,700	98,000	11,700	0,170	ND	ND
		8,900	13,200	89,000	8,100	1,460	ND	ND
		6,200	9,600	67,000	6,700	0,430	ND	ND
		4,500	11,700	110,000	11,700	0,480	ND	ND
		8,000	33,800	631,000	9,900	2,400	ND	ND
		12,700	12,600	93,000	12,000	0,290	ND	ND
		36,900	30,900	633,000	24,900	1,810	ND	ND
		4,400	18,100	410,000	6,000	1,030	ND	ND
		16,500	11,800	133,000	13,800	0,790	ND	ND
		11,400	18,800	128,000	13,500	1,370	ND	ND
		4,100	7,800	42,000	7,200	0,110	ND	ND
		5,100	6,000	48,000	2,000	0,110	ND	ND
		8,400	70,200	1096,000	39,200	0,950	ND	ND
		13,100	31,000	195,000	15,300	0,330	ND	ND
		6,400	21,300	172,000	13,300	0,250	ND	ND
		8,900	52,800	280,000	15,100	0,120	ND	ND
		7,700	22,860	88,000	6,600	0,120	ND	ND
		7,900	12,040	76,000	3,700	0,240	ND	ND
		3,700	9,250	76,000	2,900	0,210	ND	ND
		3,300	15,780	99,000	2,800	0,740	ND	ND
		2,300	11,810	96,000	3,200	0,230	ND	ND
		4,600	9,250	70,000	3,600	0,090	ND	ND
		2,300	9,250	96,000	3,000	0,210	ND	ND
		5,800	25,240	138,000	10,200	0,200	ND	ND
		5,700	9,900	153,000	4,800	0,050	ND	ND

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 11 – impervious	F2	2,000	< 2	160,000	13,200	0,300	ND	ND
M 7 - Kildare	F2	2340,000	207,000	969,000	222,000	18,900	ND	ND
		256,000	39,700	109,000	21,100	4,270	ND	ND
		1350,000	140,000	550,000	156,000	9,000	ND	ND
		519,000	70,800	272,000	59,000	5,000	ND	ND
		181,000	62,000	295,000	43,700	8,290	ND	ND
		208,000	41,300	147,000	14,800	9,000	ND	ND
		ND	89,900	445,000	87,900	10,000	ND	ND
		368,000	94,900	521,000	63,300	9,540	ND	ND
		476,000	ND	ND	ND	ND	ND	ND
	I1	404,000	95,500	554,000	116,000	6,300	ND	ND
		1720,000	259,000	1520,000	319,000	12,200	ND	ND
		704,000	152,000	956,000	188,000	9,160	ND	ND
		2210,000	293,000	1750,000	373,000	8,430	ND	ND
		302,000	77,800	407,000	97,300	5,500	ND	ND
		433,000	73,100	457,000	115,000	0,660	ND	ND
		2020,000	230,000	1400,000	274,000	16,100	ND	ND
		430,000	74,400	393,000	106,000	8,120	ND	ND
		125,000	47,900	205,000	75,800	6,110	ND	ND
		1370,000	171,000	1050,000	177,000	10,000	ND	ND
M 7 - Monasterevin	I2	163,000	59,100	218,000	86,900	6,170	ND	ND
		116,000	41,600	146,000	59,800	2,660	ND	ND
		60,900	43,400	151,000	63,100	5,760	ND	ND
		154,000	40,900	239,000	74,600	5,740	ND	ND
		258,000	69,100	318,000	79,200	7,070	ND	ND
		127,000	42,300	210,000	72,300	2,820	ND	ND
		184,000	51,700	166,000	64,900	5,120	ND	ND
		183,000	43,500	138,000	50,500	3,510	ND	ND
M 7 - Portlaoise	I3	25,400	23,000	106,000	87,000	9,400	ND	ND
		44,400	43,000	32,000	89,000	4,000	ND	ND
		59,800	6,000	18,000	79,000	2,000	ND	ND
		116,000	26,200	154,000	59,400	7,230	ND	ND
		14,500	33,000	150,000	95,000	24,000	ND	ND
		37,000	17,000	32,000	52,000	5,000	ND	ND
M 4 – Brinkworth	E1	235,670	67,000	246,000	ND	ND	ND	ND
		41,230	29,000	160,000	ND	ND	ND	ND
		246,500	13,000	84,000	ND	ND	ND	ND
		29,230	ND	32,000	ND	ND	ND	ND
		47,610	25,000	81,000	ND	ND	ND	ND
		21,770	31,000	85,000	ND	ND	ND	ND
		96,460	28,000	54,000	ND	ND	ND	ND
		15,150	23,000	94,000	ND	ND	ND	ND

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
M 4 - Brinkworth	E1	87,940	ND	66,000	ND	ND	ND	ND
		64,480	24,000	105,000	ND	ND	ND	ND
M 4 - River Ray	E2	124,740	33,000	115,000	60,000	ND	ND	ND
		89,000	17,000	56,000	ND	ND	ND	ND
		663,000	16,000	294,000	ND	ND	ND	ND
		86,000	15,000	51,000	ND	ND	ND	ND
		62,000	13,000	90,000	ND	ND	ND	ND
		384,000	242,000	688,000	178,000	0,960	ND	49,900
		95,000	39,900	250,000	23,400	5,400	ND	3,300
		1350,000	36,400	143,000	29,500	0,800	ND	5,700
		193,000	93,400	384,000	88,800	1,300	ND	21,100
		62,000	40,400	144,000	34,200	0,400	ND	9,600
		45,960	26,800	94,400	5,860	0,540	ND	3,100
M 40	E3	53,360	26,100	81,900	9,290	0,340	ND	4,600
		60,560	93,800	316,000	27,800	0,740	ND	9,300
		61,020	37,100	108,000	13,300	0,500	ND	2,100
		ND	35,200	140,000	16,200	0,340	ND	4,500
		31,880	22,000	21,100	4,830	0,140	ND	6,700
		29,270	20,900	68,700	6,520	0,240	ND	1,900
		30,580	35,600	123,000	12,000	0,350	ND	2,800
		87,410	86,300	379,000	36,000	0,790	ND	9,500
		57,850	42,700	161,000	19,800	0,360	ND	3,700
		44,400	25,400	72,200	4,500	0,290	ND	4,300
A 417	E4	44,900	49,700	45,200	0,200	0,100	ND	2,900
		32,800	15,400	53,600	5,200	0,230	ND	2,500
		21,900	16,600	61,900	6,600	0,220	ND	4,300
		82,900	10,200	41,700	6,300	0,100	ND	1,700
		45,200	32,800	52,400	3,610	0,170	ND	2,600
		16,300	23,200	55,700	3,290	0,120	ND	0,900
		54,100	30,400	69,100	5,650	0,240	ND	3,400
		120,300	22,700	53,700	7,440	0,190	ND	2,800
		184,800	13,500	20,500	1,000	0,400	ND	1,800
		181,940	108,000	390,000	88,900	ND	ND	12,000
A 34 - Gallos Brook	E5	27,770	31,500	140,000	15,200	0,190	ND	4,640
		99,500	79,400	207,000	61,500	1,000	ND	15,800
		37,710	42,600	73,700	21,800	0,580	ND	5,900
		20,310	20,200	41,000	3,500	0,160	ND	5,900
		18,420	24,900	48,600	8,100	0,160	ND	6,000
		135,130	82,600	291,000	46,800	0,910	ND	6,600
		231,000	83,000	329,000	63,800	0,820	ND	6,500
		179,000	104,000	397,000	95,900	1,110	ND	13,200
		80,550	103,000	280,000	99,000	0,620	ND	14,300
		129,920	13,000	60,000	2,000	0,400	ND	3,800

Highways	Code	TSS (mg/L)	Cu (µg/L)	Zn (µg/L)	Pb (µg/L)	Cd (µg/L)	Fe (mg/L)	Cr (µg/L)
A 34 - River Enborne	E6	38,510	63,500	30,400	39,100	0,640	ND	8,800
		114,950	27,100	15,800	12,600	0,130	ND	3,200
		66,210	75,900	46,000	48,400	0,370	ND	13,400
		136,560	24,500	8,500	13,000	0,280	ND	2,900
		73,550	49,900	26,000	33,500	0,220	ND	4,400
		130,180	23,500	14,900	3,570	0,090	ND	2,700
		50,800	15,500	38,700	2,040	0,100	ND	30,000
		40,010	11,000	20,500	4,810	0,130	ND	6,600
		46,280	20,700	29,300	6,710	0,160	ND	1,500

\*ND means no monitored data available