MŰEGYETEM 1782

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IMPACT OF VEHICULAR TRAFFIC ON HYDROCARBON CONTAMINATION OF RAINWATER RUNOFF

PhD Thesis

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1. Introduction, background of the research

The research of traffic driven water pollution has a long history. In the last three decades of the past century research results have been published *(Sartor and Boyd, 1972, Shaheen, 1975, Kobriger, 1984, Kramme, 1985, Hahn, 1990, Novotny, 1995)*, in which the pollution of urban areas and highways, as well as their impact as emission sources were discussed. The first regulation experiment against such pollution was executed in this period as well. *(Council Directive, 05/04/1976 and 12/17/1979)*. Already these initial researches demonstrated that the typical pollutants of urban roads differ that of periphery, mainly highways. Due to the more complex land use in urban areas than it is highways, a set of pollutants is not detectable in case of latter, *(McElroy et al., 1989)*.

The vehicles in the traffic emit numerous, mainly toxic organic and inorganic micro pollutants to the environment, which reach the surface and ground waters in different transmission paths. The Water Framework Directive is classifying some of the emitted pollutants as priority (hazardous) substances and permits only a low immission concentration of them in the recipients. Some polycyclic aromatic hydrocarbon species subject to the dissertation belong here too. The other discussed pollutant, the group of aliphatic hydrocarbons has a notoriously adverse impact on the aquatic ecosystem and restricts human water uses.

In the past decades hundreds of kilometres of highways have been built, crossing watercourses and vulnerable subsurface aquifers. However, there is no law available for the practice environmental authorities, in which the permissible limit values of the pollutants transported with stormwater runoff are based on scientifically established research. The permissible TPH concentration in stormwater running off the highways is determined by the certain regional authorities in different ways due to the lack of knowledge concerning the applicable laws and the probable pollution of the runoff. The basis of the prescriptions is Decree no. 28 of 2004 (December 25) of the Ministry of Environment and Water on the limit values concerning the emission of water pollutants and on the certain rules of their application. The decree is dealing with the activities characterized with continuous emission. The application of the decree is questionable in many respects in case of emissions related to rain events because of the large scale variability of the occurrence and extent of the pollution as well as the difficulties of controllability. The overall conditions are similar in case of soil-ground water recipients. The traffic related PAH emission is not regulated in the national practice.

Due to the above and in absence of the expected traffic-dependent values of pollution, and design parameters of technical solutions, the designers of highway drainage are facing a task difficult to solve. In practice, in all cases when the licensing authority prescribed a limit value for an inlet into surface water, the designer drafted one of the oil removers available in the market. However, the removal efficacy of these for extremely altering hydraulic and pollution conditions was not analyzed by any of the parties. As a result of this, expensive but questionable solutions evolved from the point of view of environmental efficacy.

2. Subjects and objectives of the dissertation

Waters running off the road surface contain a wide range of chemical and biological pollutants besides inert suspended matter, in a concentration and i depending on traffic and precipitation conditions. Nowadays the authorisation practice of the environment protection authority gives prescriptions among these for the concentration of mineral oil (TPH – Total Petroleum Hydrocarbon). The dissertation, taking into account also the application of the results is discussing the vehicular source pollution of storm water runoff and the removal of the pollutants primarily with regard to hydrocarbons (TPH and PAHs).

The environmental impact of highways, within this the pollution affecting surface and groundwaters is one from of the diffuse pollution regarded by the experts and environmental authorities as of key importance on international level, on which however there have been no systemized research carried out until now. The authorities' licensing practice has no scientifically established law, while there have been and there are new highways being built in a quick range. There are no available sizing principles for the drainage system designers, including the pollution reduction.

The practical aim of our research was the development of planning method of water quality protection constructions and support of the authorization process. It needs the development of a method, of which one parameter is the basic technical parameters of road planning, namely the traffic with taking into account its probability nature, and, which can also treat the probability nature of factors affecting the relationship among traffic and the regulation parameters of water quality protection, TPH concentration and/or TPH emission.

Development of targeted tasks as objective of the dissertation was as follows:

- Development and application of a sampling method appropriate for the exploration of the water quality features of stromwaters in the designated pilot areas;
- Analysis of the process of washing down with experimental (measuring) and model calculation;
- Exploration of the relationship between the quality of runoff and the pollutants and their dependence on the characteristics of rain events and traffic. Assessment of the experimental results on the basis of national and international experiences;
- Calculation of the traffic induced hydrocarbon pollution of surface runoff, determination of the influencing factors;

- Evaluation of the removal technologies of pollutants arising from washing down; testing and comparison of the efficacy of the treatment methods (settling, filtration) with laboratory experiments;
- Development of the planning bases of storage constructions for highway runoff treatment.

3. Research methodology

The literature is reporting of pollutant's concentrations variable within a wide range, occasionally reaching one or two orders of magnitude for all pollutants, indicating the dominant role of the complex processes and the sampling and analytical methods in the reliability of the recoverable information *(Kayhanian et al, 2001, Barrett, 1998, Zakharova and Wheatley, 2007).* It is known only of a small part of the published results, where and among what traffic and rain conditions the samples have been taken within the drainage system.

The complex environmental pollution process discussed in the dissertation can be divided into two main phases: (i) accumulation of traffic and other sources emitted pollutants and their accumulation on the road surface in dry periods, and (ii) spread of the emitted pollutants by the traffic taking place during precipitation and by the polluted road surface as emission sources on the transmission route of rainwater runoff. If we wanted to specify the role of all otherwise essential parameters building upon on-site measurements, we would need a very widespread measuring programme executed for a long period of time in numerous sites. The analytical methods serving the determination of the components in question (TPH and PAHs) are considerably expensive. It is obvious that realistically it is not possible to carry out the required amount of measurements due to expenditure and time demand, and this way to gain statistically sufficient data to assess. Therefore such methodology considerations are required which enable the acceptable engineer analysis of the phenomenon with less information. In our dissertation we were trying to simplify the basic processes of the hydrocarbon load of vehicular traffic origin on planning parameter level (Figure 1).

A sampling programme was carried out from 2005 to 2007 in two sections of highways M0 and M7 for the study of the pollution of the runoff. We developed a device suitable for the collection of sequenced sample series for the continuous sampling of runoff stormwaters. The meteorological features (rain, wind, temperature and humidity) were continuously registered with our automatic station installed in the sampling site. For the measuring period the data of the automatic traffic counting station of highway M0 of the Állami Autópálya Kezelő Zrt. at Diósd were available.

The sampling took place from the water just running off the road surface on the embankment side. This way the impact of the special conditions of the local drainage system on pollution could be excluded and the results can be generalized as the emission of the road surface.

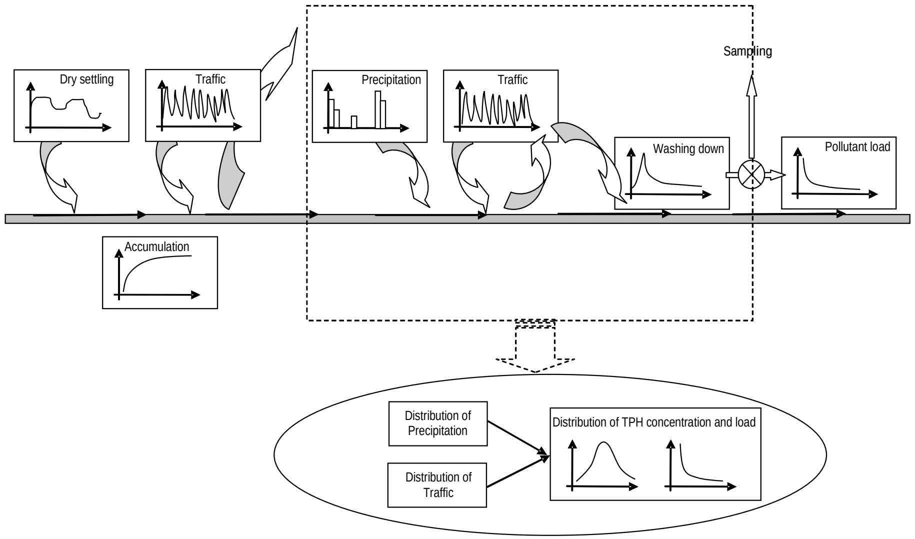


Figure 1: Basic processes of the diffuse load of vehicular traffic and its simplification on planning parameter level

The following parameters from the water samples were analysed: conductivity and pH value, total suspended matter, chemical oxygen demand (COD), total petroleum hydrocarbon (TPH), and poly-aromatic hydrocarbon (PAH). Compounds demonstrated among PAHs: Naphthalene, 2-methyl- naphthalene, 1-methyl-naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenantrene, Anthracene, Fluoranthene, Pyrene, Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene+Benzo(k)fluoranthene, Benzo(e)pyrene, Benzo(a)pyrene, Indeno(1,2,3-cd)pyrene, Dibenzo(a,h)anthracene, Benzo(g,h,i)perylene). The analysis of TPH and PAH components was executed in an accredited laboratory, the others were carried out in the laboratory of the Department according to the Hungarian standards.

One of the important specialities of diffuse pollution is that this load is stochastic, namely it is changing in time and space. Therefore a single sampling at any time, like a snapshot, is not suitable for the characterization of the contaminated runoff. Instead of it, the literature is applying the event mean concentration in order to compare the measurements executed in different areas.

The time series of flow rates of every sampled runoff events were calculated with the EPA SWMM (USA) applying the data of precipitation and the data of catchment area. In view of the filling up time of the sampling bottles it was possible to specify the pollution of the parts of the runoff wave represented by the content of the certain bottles. The mean concentration of the entire runoff, the event mean concentration (TPH_E) could be calculated of these data.

The main topics discussed in the dissertation and their relationships are illustrated in Figure 2.

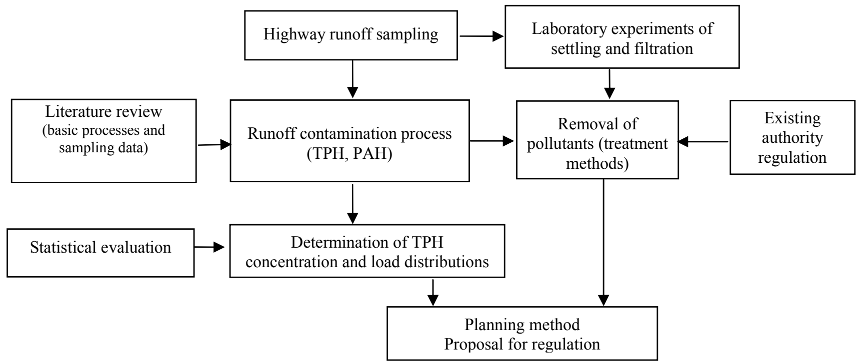


Figure 2. Main topics and their relationships

On the basis of the explored relationships, besides the distribution parameters of TPH_E in case of different traffic conditions (characterized also by a set of distribution parameters) the specific emission (emission released from the unit area of a given highway surface, gTPH/ha) and the annual specific emission (kgTPH/ha, year) became also calculable. Furthermore, in view of the concentrations we determined the specific storage capacities required for water quality protection. Statistical methods were applied for the processing of the sampling data: (i) linear regression calculations with two variables; (ii) distribution assessment with X^2 test; (iii) Monte Carlo simulation for the determination of common probability and for the examination of sensitivity; (iv) completion of deficient traffic data time series using fuzzy method. The subsequent steps of data processing gained in sampling program are shown in Figure 3.

A proposal for the environmental authority was developed (i) on the acceptable technical solutions that provide sufficient protection for the recipient against pollutions released by the highways, and (ii) on the emission parameters that must meet the constructions for the approval of the plan with.

In line with the on-site sampling we carried out laboratory experiments for the removal of TPH, PAH and suspended matter. We examined two treatment methods, namely settling and filtration. In the filtration programme we applied three types of filtering media: (i) olephylic expanded perlite, (ii) coalescent filter and (iii) sand. Mixed the runoff samples were applied; hereby we gained a sample compound polluted on the average representing the composition of the waters running off the road surface.

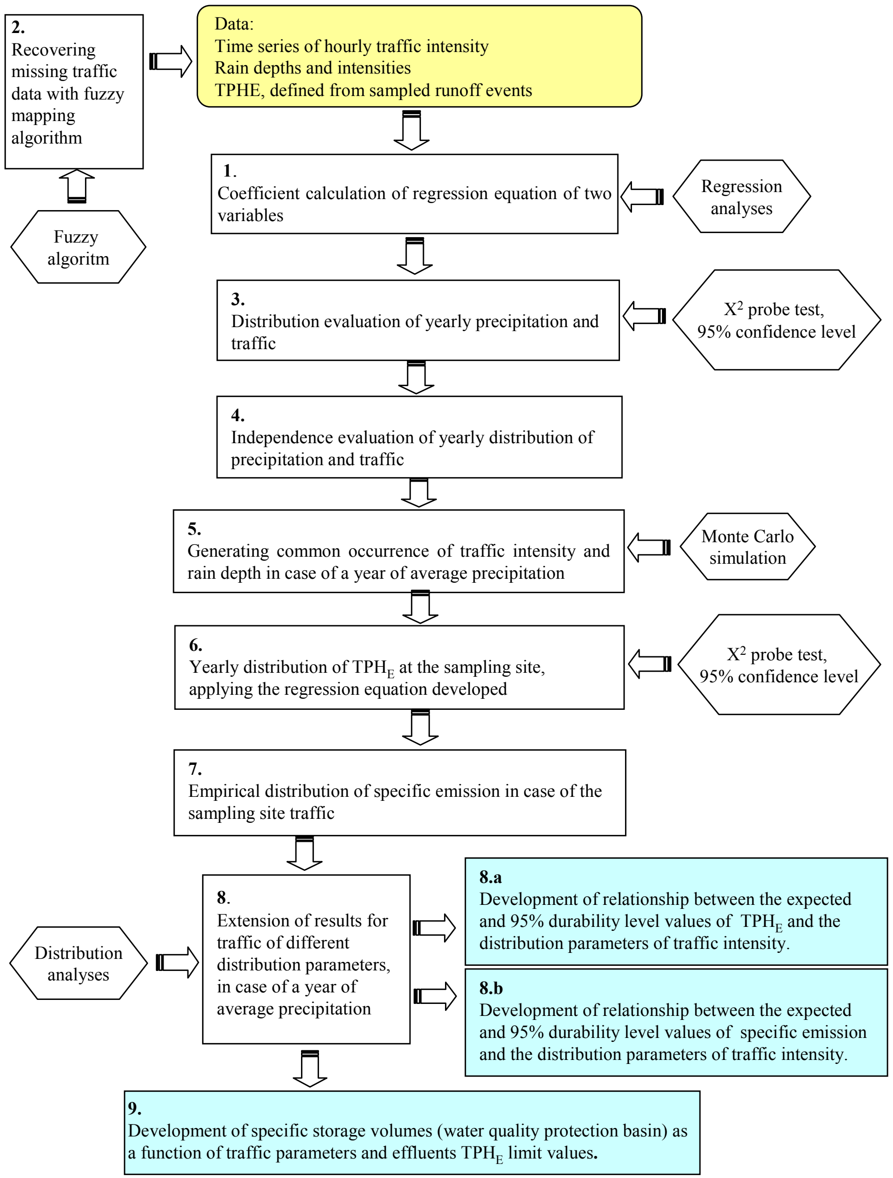


Figure 3. The main steps of data processing (precipitation, traffic, and sampling analyses)

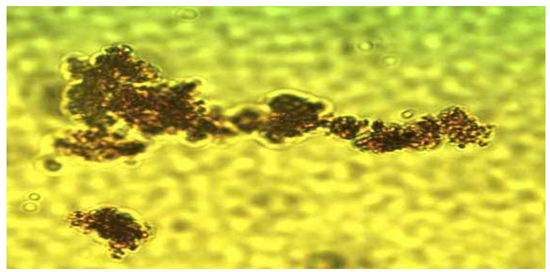
4. Brief summary of the results

Our measurements showed - in accordance with the information of the literature - that there can be found significant amounts of PAHs and heavy metals beside TPH in the water running off the road surface (the latter ones were not subject to the dissertation). Our sampling method enabled us to take into account the change of pollution within one runoff too. The concentrations changed in the course of the individual rain events, but a definite initial concentration increment, the so called „first-flush" phenomena – with the exception of suspended matter – was typical neither of TPH, nor PAHs in contrast to urban runoffs.

4.1 Characterization of the TPH contamination of the runoff

As the source of the TPH accumulated on the road surface and getting into the runoff is (engine oil) lube oil. The GC analyses of the samples refer to this fact, which show the presence of carbon C_{28} component to the biggest proportion. In the course of the dry period traffic TPH emission of vehicles and their accumulation on the surface is continuous. At the same time there is a removal process taking place: while rolling, tyres of the vehicles generate an air-flow characterized by high pressure change and speed between the road surface and the slot system of the tyre, which is able to tear down these oily particles from the road and to emit them to the atmospheric environment.

So that these granules are washed down from surface, a higher energy is required than that generated by the rain drops and surface runoff. This high energy is generated by the tyres rolling with high speed during the storm. In this condition a huge pressure difference occurs in the ducts of the tyre pattern, specifically between the front and the back side. The ram at the front is transformed into atmospheric at the back side. In the meantime the surface water film is streaming with high speed in the ducts. The high flow velocity and the pressure drop adsorb and hereby remove the oily pollution stuck to the surface and then raise and spray it into the air. This way they get into the runoff. The oil washing down this way does not form an emulsion.



The shot in the picture was taken with a 400-fold enlargement. The length of the bigger granule is ≈15µm, while that of the smaller one is ≈ 2 µm. The other pattern seen in the picture appeared due to the fraction evolving because of the not perfectly plain slide. The apolar colouring matter (safranin) indicated the location of the oil that solves it. It is visible that the oil drops of a micron range stick to the surface of the even smaller solid, probably tyre and asphalt crumb-granules, as well as to the surface of the aerosol granule sized PAH particles, respectively they form flocculate agglomerate. Therefore the TPH removal effect of oleophylic substances does not appear, since oil does not touch their surface. Similarly, the efficacy of coalescent filters is also slight, because in the lack of oil drops the phenomenon of coalescence does not occur.

To sum up, we can draw the conclusion that the traffic taking place during rain events has a decisive role in the washing down of the fine suspended matter with big adsorbing surface containing TPH (quasi-colloidal range).

4.2 PAH species and their sources occurring in the runoff

In the case of polycyclic aromatic hydrocarbons the low proportion of the less stable PAH species of smaller molecule weight (2-3 rings) is typical. This pollutant may have two possible sources: combustion processes and PAH containing matters such as dispersing aliphatic hydrocarbon, tires and asphalt containing crumbs *(Boonyatumanond et al, 2007, Kim et al, 2005, Yunker et al, 1996)*.

The identification of the processes causing the PAH pollution of the runoff is possible in a similar way as that of sediment. We assessed the proportions of the typical PAH species in all samples. Figure 4. shows that combustion process is determinative. The role of the dispersed lube oil and other background pollution is negligible. It can be stated that the PAH content of the runoff was determined by the soot content of exhaust gases emitted by the running vehicles. This explains the weak correlation relationship between the TPH and PAH concentration of the runoff and the higher correlation factor between the suspended matters and PAH concentrations.

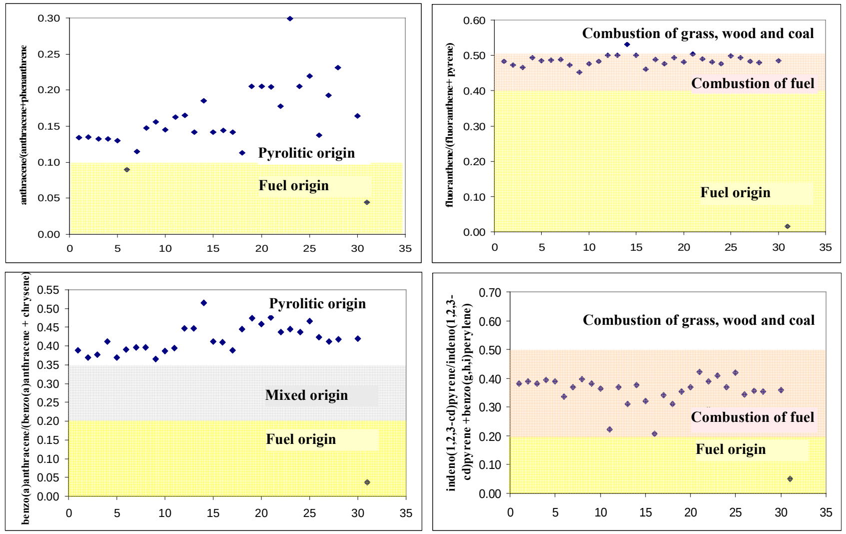


Figure 4. PAH sources detected in highway runoff

4.3Annual distribution of TPH concentration and emission, determination of the capacity of the water quality protection aquifer

Among the factors influencing the TPH_E, event mean concentration of the runoff events rain depth and the volume of traffic are dominant. Neither the length of the dry period preceding the rain event nor rain intensity had a demonstrable impact – at least in the examined case.

From the measurement results we set up an empiric relationship with a two variable linear regression to determine the TPH_E (mg/l) concentration belonging to the runoff event depending on the rain depth (H, mm) generating the runoff and the intensity of traffic expressed with a unit vehicle per hour number (uv) during the rain event (J, uv/h). The traffic time series measured by ÁAK ZRt. was also missing. Due to the limited data for statistical analyses, completion of data series had a great significance.

We used the fuzzy mapping algorithm *(Buzás, 2001)* formerly successfully applied for stop the gap of the missing data of the water quality time sequence. We achieved the determination of the annual distribution of the concentrations and emissions characterising the runoff with the application of a calculation method consisting of numerous steps. We developed a graphical device (planning nomograms) (figures 5-7) applicable. The nomogram of Figure 8. presents the necessary storage capacity of the water quality protection detention basin (the minimal storage volume, which is however big enough to receive all runoffs with a concentration exceeding the limit value).

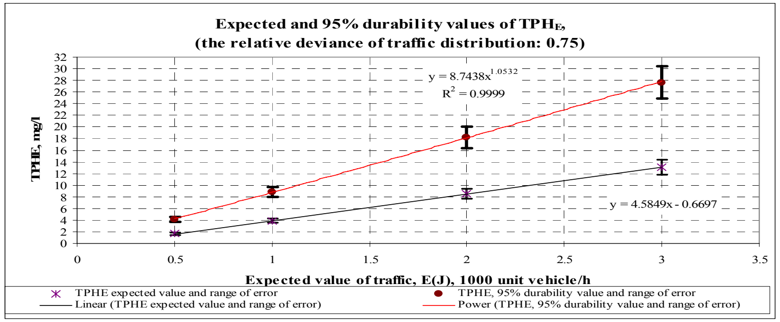


Figure 5. The mean and 95% probability concentrations of $\mathbf{TPH_E}$ as a function of expected value of traffic, in case of the traffic relative deviation 0.75

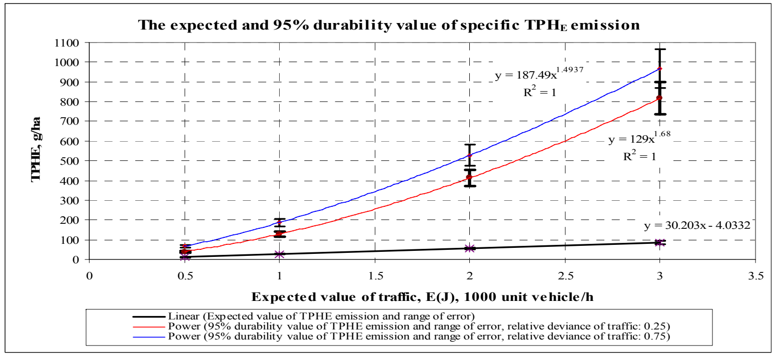


Figure 6. The mean and 95% probability concentrations of ***l*** (specific emissions, gTPH/ha) as a function of expected value of traffic

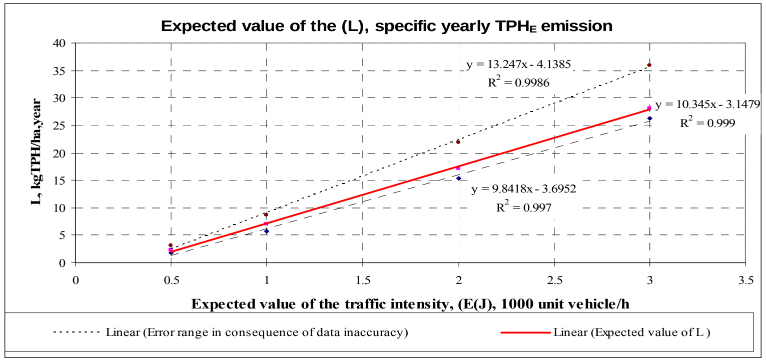


Figure 7. The expected values of **L**, (kgTPH/ha,year) as a function of expected value of traffic

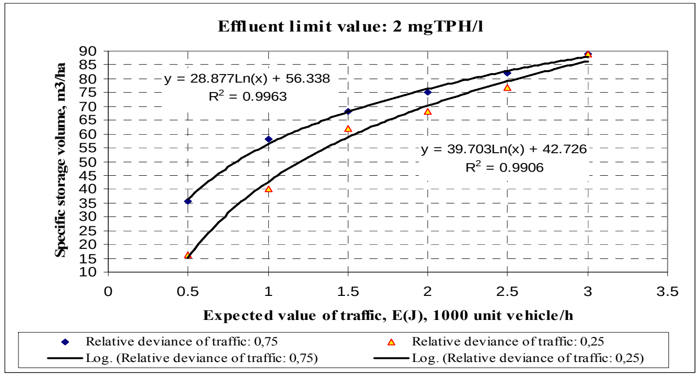


Figure 8. Specific storage capacity demand to keep the limit concentration of effluent on a 95% probability level, as a function of distribution parameters of traffic intensity

5. Practical use of the results

On the basis of our results we suggest that the emission limit value of the authority regulation for highway runoffs, in the case of surface recipients shall refer to the 95 % probability value of the event mean concentration (figure 5). The designer should be also allowed to take into account the TPH retaining capacity of the drainage system, which reduces the extent of pollution coming from the road surface according to Figure 5. In the case of a well sized grass-protected ditch the extent of reduction is 60%, while in the case of a coated ditch system it is maximum 20%.

If the concentration exceeds the limit value despite the utilization of the reduction possibility, the nomograms of figure 8 can help the designer in the determination of the required storage capacity, and to create a technical solution meeting the requirements of the authority. The detention basin sized this way is able to ensure the

observance of the prescriptions in an average pracipitation year with an occurrence probability of 95%.

If the technical content of the plan is a settling-filtration basin for example, the minimal capacity required from water quality protection point of view can be determined also from the nomograms of figure 8. We emphasize that this capacity is minimal one, and its determination does not allow neglecting the analysis of the water balance of basin, involving evaporation and infiltration features of the region. As long as the latter one requires a larger capacity, this fact must be taken into account.

The practical utilization of the experimental results can be summarized in three planning and operation aimed statements:

(i) The infiltration basins supplied with an at least 20 cm deep sand filtering layer provide a reliable and sufficient protection against the TPH and PAH pollution of the soil and groundwater. This type of basins located in regions having a sandy or sandy silt soil with sufficient permeability do not pollute the geological layers seated deeper than 20 cm below bottom level. The protection of groundwater is also implemented regarding that its level must be at least 1 meter lower below the bottom level of the basin due to safety reasons.

(ii) In order to increase the lifespan of the filtering layer a settling basin coated with impermeable overlay is recommended in front of the inflow. According to the result of the settling experiments the required retention time for the design hydraulic discharge (yearly frequency) in the settling basin shall be between a half and one our.

(iii) The expiration of filtering capacity due to colmation can be restored by the partial removal and replacement of the upper layer, as long as it is technically possible to remove (10 cm thick). Since the results show that this layer will be heavily polluted, the environment protection authority will probably classify the exploited sand as hazardous waste. In consideration of the high transportation and placement costs it is an important statement that it is not needed to replace and place the whole filtering layer.

6. Thesis

Thesis 1

Sampling runoffs from highway stretches used for continuous traffic I demonstrated that TPH drops of some micrometer size are confined in agglomerates in size of 10-100 µm. The oil content of the runoff therefore does not form an emulsion with stormwater („oil-in-water" type emulsion). Such a formation of oil has two substantial consequences:

(i) The energy of raindrops and surface runoff is not high enough to remove the agglomerates stuck to the road surface. However, the energy of water with high flow rate evolving in the water film between the tyres of the moving vehicles and the road surface is able to remove the pollutants of the road surface and to pass them into the surface runoff in the form of spray evolving around the vehicles. Therefore the extent of traffic intensity during rain events can be regarded as the determining factor of TPH contamination of runoff.

(ii) For the removal of such form of aliphatic hydrocarbon content of the runoff, the applications of the oleophylic adsorbents and the devices based on the coalescence principle are capable only of a low efficiency.

Papers: [1,6]

Thesis 2

Analysing the occurrence proportion of the PAH species of different molecule weights in runoffs, (PAH profiles), I demonstrated that the significant proportion of the PAH is arising from a combustion process and probably are bound to the to the soot content of the exhaust gases of running vehicles. The role of other potential PAH sources (atmospheric deposition, tyre and asphalt crumbs, as well as spilling lube oils) are negligible.

Papers: [1,6]

Thesis 3

In practice, the infiltration basins and sand filter basins are efficiently applicable for the protection surface and groundwaters against pollution impact of highway runoffs. I justified the statement with laboratory experiments executed with highway runoff samples. I demonstrated that:

- Using sand filtration combined with pre-settling TPH and PAH removal efficacy over 90% is achievable. Retention of pollutants takes place in the 2.5 cm deep upper layer of the filtering media placed on the reservoir bottom. The colmation speed of this upper layer determines how often the layer should be changed.
- The oleophylic perlite filters media works as a simple granulated filter media. In the removal of aliphatic hydrocarbon enclosed in the flocculated structure (i.e. in absence of free oil surface), the oleophylic nature does not playing a role.
- The settling as a single treatment has lower TPH removal efficiency that that of for suspended solids, because TPH adsorbs primarily to the surface of finer, quasi-colloidal sized granules, which are scarcely to settle.

Papers: [3,7]

Thesis 4

I justified that the missing data of the traffic (unit vehicle/hour) can be recovered with application of a fuzzy mapping algorithm trainable with the known data. This algorithm creates a fuzzy rule system from numerical data. The process was successfully applied for completion of the incomplete traffic time series of Diósd

measuring station of highway M0. The dynamics of the data sequence completed with this method shows a good correspondence with that of the measured data sequence. The most important feature of the process is that with the recovered missing traffic data the distribution parameters (the expected value and deviation) of the whole traffic time series are not modified. Consequently, the distribution of the calculated event mean concentrations depending on the distribution features of the traffic and that of TPH emissions does not alter either. Therefore the complemented data sequence is applicable also for the capacity sizing of the water quality protection basin.

Papers: [2]

Thesis 5

I worked out a calculation process for the determination of the annual distribution of event mean concentration (TPH_E, mgTPH/l - F(TPH_E)), and of specific TPH emission (*l*, gTPH/ha - F(*l*)), and of yearly specific TPH emission (L, kgTPH/ha, year – F(L)). All are depending on the distribution parameters of traffic, and are valid in case of condition of an average rainy year. The results enable the designer to determine the expected emission and in view of this the authority to consider the expected environmental impact. The basis of the process is a linear regression equation with two variables, which I determined for the runoff event mean concentration TPH_E= f(traffic, rain depth). I demonstrated that the traffic characterized with normal annual distribution and the rain depths characterized with annual exponential distribution are not independent of each other. Therefore I created the common occurrence probability with Monte Carlo simulation. With these conditions the values of F(TPH_E) were extended to the probable national traffic intensity ranges. I gave the calculation method of the expected values of TPH_E and the values of 95% durability. I worked out also the nomograms for the specific emissions based on similar methods.

Papers: [3,7,8]

Thesis 6

Knowing the statistical features of the traffic and precipitation I worked out a calculation method for the determination of the lowest storage capacity of water quality protection basin that is sufficient to receive all runoffs with a concentration exceeding a given TPH_E limit value. I created planning nomograms, where the specific basin volume required storing runoffs when the (assumed) regulation limit values are TPH_E = 2, 3, 5 and 10 mgTPH/l, is suit on the 95 %-security level. These storage volumes are delineated as function of the distribution parameters of the traffic.

Papers: [3,7,8]

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