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The effect of traffic-borne heavy metal emissions on the quality of road runoff

Theses of the PhD dissertation

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INTRODUCTION

Anthropogenic heavy metal pollution was traditionally dominated by industrial point-source emissions. The evolution of environmental policies in some countries and the decline of metallurgy in others, combined with the continuous development of road transportation have changed the situation. Nowadays, a substantial portion of anthropogenic heavy metal emissions originates from road traffic. In terms of diffuse (non-point) pollution, this source has become of number one importance (Councell et al., 2004; Bergbäck et al., 2001).

The relationship between road traffic and heavy metal pollution already received scientific attention in the past few decades. The measurements of Sternbeck et al. (2002) showed that the Cd, Cu, Pb, Sb and Zn content of road tunnel aerosols are released by vehicles, instead of being of geological origin. They also found that the Cu concentration in the air correlates with traffic volume. Councell et al. (2004) discovered a strong relationship between the accumulation of Zn in lake sediments and the traffic volume of nearby roads. Hjortenkrans et al. (2006) revealed that the Cu and Sb content of roadside topsoil correlates well with traffic volume and its velocity pattern. Additionally, they showed that only a few years of extensive usage as brake pad components increased the topsoil Cu and Sb concentrations so much, that they are several times higher now than the background values. Hulskotte et al. (2007) estimated that in comparison with other sources, the volume of road traffic Cu aerosol emissions are significant in Europe, and verified their model estimations with immission measurements. According to their calculation, road traffic is the major source in Western European countries (reaching 80% of the total emissions in the Netherlands), and an important contributor in Central and Eastern European countries (in the case of Hungary, the share was estimated to be 54%). The serious effects of road traffic originated heavy metal emissions on the quality of rainfall runoff have also been proven by a number of studies (Göbel et al., 2007).

Traffic-borne heavy metals, emitted mainly as solid particles, reach all three environmental compartments, thus causing a complex air, water and soil pollution problem. Due to their persistency, these contaminants tend to accumulate in their recipients: usually in the upper layer of soils and in the sediment of surface waters, or, in the case of urban areas served by combined sewers, in sewage sludge. The bioavailability, and thus, the toxicity of these pollutants depend on several factors, but in the long run they pose certain potential hazards. Studies indicate for example, that the mobility and bioavailability of Zn contained in rubber products is significant (Councell et al., 2004). Von Uexküll et al. (2005) have shown that potentially carcinogenic Sb compounds that can be found in brake wear dust dissolve well both in tartaric acid and in physiological solution. Long-term exposure to heavy metal pollution affects the sediment-dwelling aquatic organisms in water bodies receiving road runoff, as well as the plants grown on roadside agricultural lands. Via the latter pathway, humans may be affected, too. Atmospheric emissions shall not be forgotten, either, since most of the direct human exposure happens in this way.

AIMS OF THE RESEARCH

Water quality problems are increasingly of non-point nature, and in particular, are becoming associated with micro-pollutants: toxic substances that are present in the environment in minute levels. Road traffic emissions are one of the most important among such sources. In spite of its significance, heavy metal pollution by road traffic was hardly investigated in Hungary. Although awareness was raised for a range of heavy metals (Buzás and Somlyódy, 1997), only the lead contamination caused by exhaust fumes managed to gain wider publicity. Other metals originating from road traffic have rarely been studied so far, except for a few studies dealing with air and soil contamination (Salma, 2010; Naszradi, 2007; Kiss and Vidovenyecz, 2008). Water, which is an important pollution carrier medium, was still unexplored from this aspect in Hungary. Hence, four goals were set for my research:

i) thorough analysis of the emission and transmission processes of heavy metals originating from road traffic;

ii) characterization of the heavy metal contamination of road surfaces and of road runoff, based on field measurements;

iii) identification, ranking and quantification of the factors that influence the heavy metal loads of road runoff;

iv) development of robust methods to estimate heavy metal emissions and runoff loads originating from road traffic.

The chosen topics, and in particular the emission and runoff load estimation goals, are a major step forward concerning Hungary, and will serve as a basis for similar studies in the future. Due to the complexity of pollutant transport, most diffuse pollution problems have a special common property: the employed calculation methods are usually empirical, relying heavily on specific field measurements. During my research, I have developed new, efficient sampling procedures, which will also be useful for future investigations.

To reach the goals set above, I employed the research strategy depicted on Fig. 1. During the literature overview, two main topics were highlighted: investigation of the sources and transport processes of heavy metal emissions originating from road traffic, and evaluation of the published results and methods used by a range of foreign road runoff sampling programs. My own sampling programs were set up based on this experience, including the development of two new sample collection methods. Two types of field studies were executed: one concerning the dry weather loads of road surface, and another concerning the loads washed away from there by road runoff. The latter type included two distinct campaigns on various urban and highway sites.

The results of the measurements allowed for the identification of the most important factors influencing the heavy metal loads of road runoff. Detailed information was gathered concerning the spatial distribution of dry weather road surface loads of heavy metals. Based on the measured values, the proportion of brake-emitted particles capable of resuspension following wet deposition was calculated. These, in combination with literature data, were utilized to construct a simple and robust method to estimate long-term emission and road runoff loads of traffic-borne heavy metals on a large spatial scale. Another, completely different runoff load estimation method, based on traffic volume data, was also developed, having the advantage of being applicable on small spatial scales. These methods were evaluated by comparing the various estimated loads against measured values.

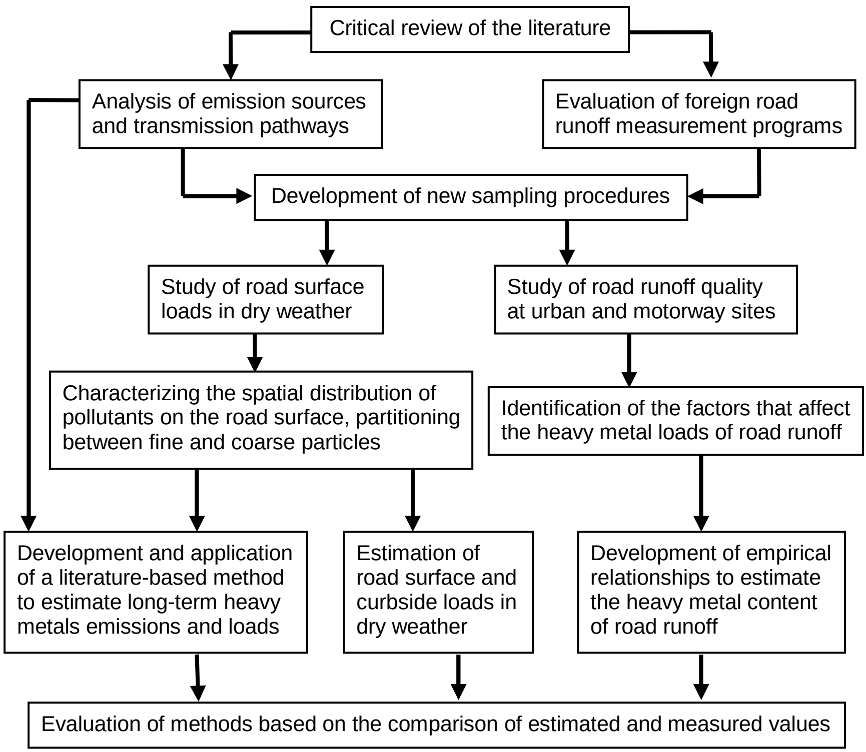

Fig. 1. *Relations between the building blocks of the research.*

SHORT SUMMARY OF RESEARCH RESULTS

Heavy metal emissions of road traffic originate from various sources. The relative strength of these sources varies from one metal to the other, but in general, the wear of brake materials and tire surface are the most important. The amount of worn material depends on a number of factors, among which the most important are: vehicle type (light/heavy); the highly complex and diverse composition of brake pad and tire materials; the rate, duration and frequency of deceleration; the actual condition of tires; the composition and roughness of the road surface; and the frequency of taking turns. To sum up: parent materials, traffic dynamics and driving style all have a significant effect on the rate of brake and tire wear. The remaining portion of road traffic heavy metal emissions is due to the wear of road surface and paint, exhaust fumes, the corrosion of metallic vehicle and road furniture parts, and motor oil leakage.

Heavy metals emitted by road traffic are predominantly in particulate form, ranging in size from submicron dust to a few hundred micrometer pieces. Once emitted, the heavier fraction falls out near to its origin within a short time, while finer particles become part of the aerosol and, staying for a prolonged time in the air, can travel long distances from their sources. In wet weather, both fractions become affected by further transport processes. These include wet deposition from the atmosphere, removal from the road surface by the pressure and suction forces exerted by the wheels of moving vehicles, as well as the wash-off of the accumulated amount of contamination from the roadside curbs and vehicle bodies. Therefore it is essential to study also the dry weather road surface contamination besides the wet weather runoff pollutant loads.

For the dry weather sampling of pollution built up on the road surface, a portable high-pressure washer device was used. This new method has a major advantage over the other commonly utilized techniques (sprinkling and hoovering), because its working principle is identical to the wet weather pollutant mobilization processes caused by rolling vehicle wheels. The other two common methods rather simulate the less efficient wash-off caused by rainfall runoff only, without the amplifying effect of traffic. The new sample collection procedure employing high-pressure washer provides better information about the potentially removable pollutant amounts under real-life circumstances, and functions as a handy method to characterize the spatial distribution of road surface contamination.

Road runoff was sampled using two different methods. One was the obvious option of traditional drainage system sampling. The drawback of this method, also noted by the literature, is that pollutant losses may occur during the transport of runoff in the drainage system. Such samples usually represent decreased loads, which often differ from one site to another and one event to another. As a result, the evaluation, comparison and interpretation of the wide range of values reported in the literature is a rather difficult task. Learning from this experience, part of my research project was to develop a new wet weather sampling approach, targeted at the collection of road runoff very near to the sources of pollutant emissions, with minimal transport losses. This was achieved by mounting an appropriate sampler on a passenger car near the back wheel, which was able to collect part of the splashed road surface sheet flow. The new ”moving vehicle” sampling method was applied to gather road surface runoff from a number of urban roads and motorways, during a number of various precipitation events. The comparison of heavy metal concentrations and their ratios in the runoff samples collected by the different methods proved that the new approach is a good alternative of the traditional drainage system sampling.

Estimation of road traffic heavy metal emissions is a very complicated task. The physically based quantification of the factors influencing the rate of brake pad and tire wear is still an unsolved problem. Furthermore, the partitioning of the generated particles between suspended and settling fractions is also very uncertain. Thus, one must rely on the wide range of empirical emission factors that can be found in the literature. These are usually characterized by great uncertainties, therefore their use is limited to long-term and large-scale applications (for instance, estimating annual emissions on a broad regional scale). During my research project, I developed one such method to estimate annual emissions and road runoff loads in Hungary. The uncertainty of the calculation was successfully decreased by utilizing measurement data. The estimated values illustrate well that urban traffic causes more intensive pollution than motorway traffic. Dry weather surface loads calculated for an urban site matched quite well with the measured values for almost all metals in question.

The results of the dry weather field measurements broadened our knowledge about the spatial distribution of heavy metal contamination on the road surface. Combined with the results of wet weather field measurements, they also helped to quantify the amount of the originally airborne fraction of brake wear emissions that is deposited to the surface by precipitation but gets resuspended as soon as the road dries up. This finding has two important practical consequences. First, it highlights that the timing of street cleaning is a very significant parameter. Second, it proves that most of the road runoff loads of copper, antimony and brake wear emitted lead consist of very fine particles that are easily transported by the runoff flow as a slow-settling suspension, and this way they can travel long distances even in the absence of desorption or dissolution processes.

Using the results of wet weather field measurements, the two most important factors affecting runoff loads could be identified, namely: i) the concurrent traffic and ii) the cumulated traffic of the antecedent dry period. Characteristic concentration ratios of heavy metals in runoff originating from urban roads and motorways have also been identified. Based on these findings, relationships were developed to estimate event mean concentrations, and thus, runoff loads of zinc, copper and lead on urban and motorway sites from traffic data. The main advantage of this new method over the one based on average emission rates is that it can be applied on smaller spatial scales as well. It was demonstrated by means of an example that the new, traffic-based method offers a more reliable alternative for the estimation of road runoff heavy metal loads at specific locations.

THESES

1st Thesis: A robust method, based on specific emission factors, was developed to estimate long-term heavy metal emissions and road runoff loads originating from road traffic. The application of the estimation method was demonstrated for year 2008, using annual Hungarian traffic count data. The wash-off ratio of brake-emitted particles was set to 20-35% according to the Cu/Zn ratios measured in field samples. This interval is much narrower than the range given by the international literature, and thus, it helped to decrease the uncertainty of the estimation. The result of the country-scale estimation underlines the dominant role of urban traffic: although only a quarter of the total traffic takes place in urban environment, about half of the total brake wear and a third of the total tire wear can be attributed to this source. *[1]*

2nd Thesis: Two new sample collection methods have been developed for the study of particle-bound heavy metal pollution originating from road surfaces. The wet weather procedure is based on the collection of sheet flow samples using a vehicle that moves with the traffic. This method is an alternative to the traditional way of taking runoff samples from the road drainage system. One of its advantages is being capable of collecting runoff samples without pollutant losses from practically any kind of site. Another advantage is that multiple sites can be sampled during one rainfall event using the same vehicle. It was shown that the absolute concentrations and the relative ratios of heavy metals are identical to the samples taken with the traditional method. Consequently, it can be said that the ”moving vehicle” approach is a good alternative of the drainage system method. The other new procedure is a dry weather sampling method, based on the collection of road dust samples using a high-pressure washer. The advantage of this method is that it can mimic the wet weather pollutant removal processes exerted by moving vehicles, and thus, the amount and the spatial distribution of potentially detachable heavy metal loads can be characterized. *[2,5]*

3rd Thesis: It was shown with measurements, that traffic-borne heavy metals can be split into two distinct groups based on the distribution of their dry weather loads on the road surface. The different behavior reflects the differences of their emitting sources. The surface loads of entirely or largely brake wear originated copper, antimony, and lead: i) are proportional to the frequency of brake application; ii) get easily mobilized during the dry period; iii) usually build up along the roadside curb; iv) decrease from the curb towards the trafficked part of the road, where they reach background levels; v) the resulting cross-sectional gradient depends on the strength and spatial variability of local winds. The surface loads of mostly tire wear originated zinc and cadmium: i) depend less on the frequency of brake application; ii) do not get easily mobilized during the dry period; iii) usually stick strongly to the road surface; iv) display more even cross-sectional distributions; v) are generally significantly higher than the background levels. *[5]*

4th Thesis: Using measurement results from urban sites, it was shown that the brake wear emitted copper and antimony content of the fine sediment that is washed out to the curbside during rainfall events decreases during the post-rainfall drying up. The remaining fraction was found to be only 27±9% (Cu) and 28±16 % (Sb) of the originally washed off amount. These ratios agree well with the results of the 1st Thesis concerning the wash-off ratio for brake wear emitted particles. The missing portion is made up of very fine particles that get washed out from the air by wet deposition, and are gradually resuspended by winds afterwards, when the sediment dries up. Two important practical consequences can be drawn from this result: i) street cleaning is most effective when applied soon after rainfall events, provided that it is effective at removing fine particles; ii) regarding runoff loads, most of the traffic-borne copper and antimony, and the brake wear emitted portion of other heavy metals make up a stable, slow-settling suspension with the runoff, and can travel long distances, even in the absence of desorption or dissolution. *[5]*

5th Thesis: The analysis of runoff samples collected from motorways and urban roads showed that the heavy metal contamination of urban runoff is higher. The average urban/motorway concentration ratio in the two sample groups varies from one metal to the other: in the case of zinc, copper and lead, these are 1.6±0.2; 2.5±0.4 and 2.7±0.4, respectively. The differences are caused by the differences in traffic dynamics and in the emitting sources of the various metals. In urban traffic, the wear of brake pads, and thus the emissions of copper and lead are more intensive than on motorways. The difference in tire wear is less pronounced, as it happens during undisturbed traffic flows as well. Thus, the zinc emissions of urban and motorway traffic are more identical than in the case of brake-emitted metals. Consequently, the ratio of copper and zinc in road runoff can be used as a good indicator of traffic dynamics. Based on my measurements, the characteristic Cu/Zn ratio in urban road runoff was found to be 0.36 (±0.07), while in motorway runoff it was only 0.23 (±0.05). *[1,2]*

6th Thesis: Based on the findings of the two distinct road runoff sample collection programs, relationships were developed to estimate event mean concentrations of zinc, copper and lead on urban and motorway sites from the two most important factors influencing runoff loads, namely i) the concurrent traffic volume, and ii) the cumulated traffic volume of the antecedent dry period. The equations can be used within their validity interval to predict local road runoff loads of zinc, copper and lead from known or estimated traffic and runoff data. The advantage of this new method is that is can also be applied on a small spatial scale. *[3]*

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