# Traffic as a source of pollution

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Received 18 December 2007, in revised form 1 February 2008

**Abstract.** Most of the pollution of roadside soils and groundwater originate from traffic emissions, pavement and road furniture. This paper investigates heavy metal contamination, originating from motor vehicle traffic in roadside soils in Tallinn. The investigation was carried out from 2001 to 2007 and it covers heavy metal transportation to the roadside environment by traffic and the relation of pollution and winter road maintenance. As compared to spring, by autumn Pb, Zn and Cd concentrations in the surface of soil are decreased: Zn by 41, Cd by 57 and Pb by 31% on average. A principal scheme for decision-making, concerning immediate protection of soils and groundwater in roadside areas has been recommended. At the same time, the scheme helps to specify sites for monitoring soils and groundwater in the national and local monitoring programmes.

Key words: soil pollution, heavy metals, road maintenance, de-icing salts, quality of groundwater.

### **1. INTRODUCTION**

The rapid motorization and growth of the traffic volume during the last decades requires additional expenditures on road maintenance and environmental protection. According to the EU directive, water protection should be strongly integrated with other domains of environmental strategy, including the area of transportation  $[^1]$ .

Pollution, caused by traffic, appears in different forms: in gaseous (exhaust emissions), liquid (leakages) or in solid form. Solid pollution may, for instance, be caused by the corrosion of metal parts of vehicles, solid particles of emitted gases, wearout of tyres, etc.

In the case of high traffic volume, the quality of soil and groundwater is influenced by the wear of the pavement and the vehicle:

- non-vehicle exhaust emissions, such as tyre abrasion, wear of certain vehicle parts (brakes, engine parts, etc.), influence of studded tyres on the pavement, etc.;
- road and maintenance-related emissions (degradation of the road pavement, corrosion of crash barriers, road and roadside maintenance-related causes).

The constituents of the degradation of the road pavement depend mainly on the type of the pavement. Trace quantities of metals and polycyclic aromatic hydrocarbons are present in bitumen [<sup>2</sup>]. The amounts of PM<sub>10</sub> (concentration of inhalable, smaller than 10  $\mu$ m diameter, particular matter), produced from different pavements, vary to a great extent and they have different toxicological potential in human airway cells [<sup>3,4</sup>]. Dust from pavement wear contaminates the roadside environment and when inhaled is hazardous to human health. According to a Swedish study, the use of studded tyres in winter months leads to extensive road wear [<sup>5</sup>]. The use of studded tyres accelerates road degradation [<sup>6,7</sup>]. Most of the time in winter, when the roads are not slippery, enhanced adhesion properties of tyres remain unused and unnecessary road wear occurs. The wear effect of studded tyres and stud wear increases on damp pavement in Finland annually, is caused by pavement wear from studded tyres and sand, spread on roads [<sup>8,9</sup>].

Pollution from non-vehicle exhaust emissions, pavement wear and road maintenance may exceed significantly that from vehicle exhaust emissions. For instance, the amount of mineral materials and binding particles is about 10–30 times larger than the amount of solid particles from the vechicle exhaust emissions, calculated on the basis of used fuel expense, traffic speed and driving mode [<sup>10</sup>].

The traffic security in northern countries with variable climate is complicated, especially in the winter season. To control and remove snow and ice from the roads, salt (mainly sodium chloride) is widely used. The release of road salts into the environment in very high amounts leads to environmental problems – damage of vegetation and organisms in soil and contamination of the surface and groundwater [ $^{11-13}$ ].

At the same time, traffic is regarded as a source of heavy metal (HM) content (copper, chrome, nickel, volfram, titanium, manganese and zink) in dust [<sup>14</sup>]. Wear and corrosion of vehicle parts (brakes, tyres, radiators, body and engine parts) is one of the potential sources of many HMs [<sup>2</sup>]. Road furniture and vehicle corrosion and relations between salts and pavement condition were studied in Estonia already in 1974 [<sup>15</sup>]. Several studies have focused on soil contamination with HMs [<sup>16–19</sup>]. Sources of pollution, arising from vehicles and roads, are varied and depend on local conditions. Thus it is essential to evaluate the amount of pollution. The environmental impact of traffic may be reflected by the change of groundwater level and chemical composition of water [<sup>20</sup>]. According to Estonian Geology Centre, Cl content in samples taken from drilled wells on roadside areas of high traffic level in Tallinn has a strong rising tendency, giving evidence of negative impact of road salting on the environment [<sup>21</sup>].

The aim of this paper is to describe the investigations, carried out at the Department of Transportation of Tallinn University of Technology (TUT) from 2001 to 2007, covering pollution by traffic, the pollution of roadside areas by HMs (Pb, Cd, Zn, Cu, Cr, Ni, and Co), comparison of pollutant concentrations with recognized target and intervention limits and connections with the increase in chloride content in roadside soils in Tallinn, resulting from road salting in winter maintenance.

### 2. MATERIAL AND METHODS

### 2.1. Study areas and collection of soil samples

During 2001–2007 soil pH, the content of HMs and chlorides was estimated at distances up to 30 m from the pavement at the depths of 3–10 cm. At roads with different traffic volumes, 74 locations were selected for sampling in 2001–2003. Pollution spread was investigated from the pavement up to the distance of 30 m, samples were selected at the distances of 5, 10 and 25–30 m. In 2004 samples were taken at 27 roadside sites at 5 and 15 m from the pavement, seven sites were selected for further continuous monitoring in spring and autumn. Based on the results, obtained from 2005 on, HM contents have been determined only at distances 15 m and soil pH and chloride contents at distances 5 and 15 m from the pavement edge.

To investigate the effect of chlorides on the environmental situation, mostly samples collected since 2002 have been used. In spring, samples have been collected in March or April after soil melt and in autumn, in September and October. In the present article generalizations are based on the data of the sites shown in Table 1. Traffic intensity is characterized by the annual average daily traffic AADT.

All selected road sites were with asphalt surfacing. The requirements for the state of the road on the level 4 must be followed during winter road maintenance [<sup>22</sup>]. Soil samples were collected according to the soil quality requirements in ISO 11464:2004 standard.

## 2.2. Determination of heavy metals, chlorides and soil pH level

Chemical analyses were done in the analytical laboratory of the Institute of Chemistry at TUT. The concentration of HMs was determined by atomic absorption spectrometry (AAS). Using the modifier in AAS simplifies essentially sample pre-treatment [<sup>23</sup>]. For the mineralization of the samples for microwave-assisted acid digestion, a microwave oven with teflon bombs Anton Paar Multiwave 3000 (Graz, Austria, 2004) was used. The Certified Reference Material (CRM) was used for the determination of toxic elements in soil [<sup>24</sup>].

The chloride was titrated amperometrically in a solution, produced by digesting the soil sample by diluted  $(1 \text{ mol/dm}^3)$  nitric acid. The titration was carried

### Table 1. Study site characteristics

| Site       | Description   | ΔΔDΤ   | Number of lanes                    | Speed limit. |
|------------|---|--------|------------------------------------|--------------|
| No.        | Description   | AADT   | and road type                      | km/h         |
| S1         | Highway type road, sampling area up to 27 m flat grassland with a forest  | 49 600 | 2+2                                | 70           |
| S2         | beyond and railway on the other side.<br>No mowing, deciduous trees, shrubs.<br>Road bordered by buildings on one side<br>and urban forest cluttered on the other,<br>mainly pine trees; road bordered by | 37 000 | 4 + 4, separating strip            | 50           |
| <b>S</b> 3 | edgestones, aspnait surface footpath.<br>Highway near crossing a river with steep<br>banks, at the border of the town, high<br>bank river-bed, no mowing.   | 25 700 | 2 + 2, separating strip            | 70           |
| S4         | High pine tree forest area in town, pines,<br>dark topsoil, sampling area about 40 m<br>from crossroad with a roundabout,<br>road with edgestones, asphalt surface<br>footpath.                           | 22 000 | 3 + 2, separating strip            | 50           |
| S5         | Urban park in private houses area, road<br>reconstructed in 2002, edgestones and<br>wide asphalt surface footpath. Mainly<br>pine trees, sandy soil, moss, heather<br>and red bilberry under trees.       | 16 700 | 2 + 2 + 1 (left turn)              | 50           |
| <b>S</b> 6 | Steep incline of the road, open area, quarry at the corner of inroad.   | 14 550 | 1 + 1 + additional lane<br>on rise | 70           |
| S7         | Steep incline of the road, roadside<br>slightly higher and some old trees on<br>one side, new buildings on the other,<br>sample site on the bus traffic side.   | 22 000 | 2 + 3 (1 lane for busses)          | 50           |

out using silver nitrate as the titrant and a rotating platinum-wire electrode. As the reference electrode, a mercury sulphate-mercury electrode was used. This method has been used for a long time in laboratory practice and suits for the analysis of low chloride concentrations. For comparison, the chromatographic HPLC method was used [<sup>25</sup>].

The soil pH level was determined by a conventional potentiometric method.

## **3. RESULTS AND DISCUSSION**

### 3.1. Average concentrations of lead, zinc and cadmium in roadside soils

Table 2 shows summary statistics of HM (Pb, Zn, Cd) concentrations in roadside areas in Tallinn.

Through these years the amount of precipitation and temperatures varied. The number of fluctuations of road surface temperatures over 0°C differed and the ground melted at different times (Fig. 1).

|                                  | Pb     |        |           | Zn     | Cd        |        |
|----------------------------------|--------|--------|-----------|--------|-----------|--------|
|                                  | Spring | Autumn | Spring    | Autumn | Spring    | Autumn |
| Number of samples                | 121    | 65     | 117       | 65     | 117       | 65     |
| Average                          | 48     | 40     | 160       | 100    | 0.72      | 0.68   |
| Standard deviation               | 36     | 28     | 140       | 74     | 0.91      | 0.94   |
| Minimum                          | 2      | 2      | 18        | 12     | 0.05      | 0.07   |
| Maximum                          | 151    | 115    | 895       | 434    | 6.2       | 4.95   |
| Median                           | 40     | 33     | 123       | 87     | 0.39      | 0.31   |
| Dominating natural concentration | 6–26   |        | 13.7–51.6 |        | 0.10-0.50 |        |

**Table 2.** Summary statistics of HM concentrations (mg/kg) in roadside soils within distances 5–15 m from the road pavement in 2001–2007

in humus horizon of soils in

Tallinn and surroundings [<sup>26</sup>]

Results from 2001 to 2003 already showed that the average content of dangerous HMs (Pb, Cd, Ni, Zn, Cu, Co) in the roadside soil of crowded areas and highways exceeds the Estonian averages for several times. Higher HM concentrations were typically found in samples taken at 10–15 m from the road pavement [<sup>27</sup>].

Concentrations of HMs from a total of 103 samples in 2001–2004 were found to be in correlation with the range of traffic volume from 700 to 62700 AADT. The same conclusion follows from earlier investigations [ $^{28,29}$ ].

## 3.2. Average chloride concentrations and soil pH in roadside areas

Tables 3 and 4 show average chloride concentrations in road- and streetside areas at distances L from the road pavement in spring and autumn, depending on the requirements for the state of road in winter conditions. States of road

|                    | Level |       |     |      |     |     |  |  |  |
|--------------------|-------|-------|-----|------|-----|-----|--|--|--|
|                    | 4*    | 4     | 4   | 3    | 3   | 2   |  |  |  |
| Number of samples  | 74    | 56    | 21  | 41   | 41  | 14  |  |  |  |
| <i>L</i> , m       | 5     | 10-20 | 30  | 5    | 15  | 5   |  |  |  |
| Average            | 508   | 248   | 75  | 252  | 133 | 92  |  |  |  |
| Standard deviation | 355   | 247   | 46  | 274  | 137 | 89  |  |  |  |
| Minimum            | 81    | 30    | 18  | 40   | 30  | 26  |  |  |  |
| Maximum            | 1632  | 1249  | 168 | 1368 | 595 | 342 |  |  |  |
| Median             | 350   | 161   | 61  | 153  | 92  | 55  |  |  |  |

Table 3. Summary statistics of chloride concentrations (mg/kg) in roadside soil in spring 2002–2007

\* Calculations of statistical data exclude extremely high values (even values exceeding 5000 mg/kg were obtained), found mainly in samples from 2001 and 2002.

|                    |     | Level |     |    |  |  |  |  |
|--------------------|-----|-------|-----|----|--|--|--|--|
|                    | 4   | 4     | 3   | 2  |  |  |  |  |
| Number of samples  | 18  | 25    | 14  | 1  |  |  |  |  |
| <i>L</i> , m       | 5   | 15    | 5   | 5  |  |  |  |  |
| Average            | 215 | 209   | 197 | 41 |  |  |  |  |
| Standard deviation | 106 | 101   | 76  |    |  |  |  |  |
| Minimum            | 114 | 42    | 109 |    |  |  |  |  |
| Maximum            | 519 | 478   | 286 |    |  |  |  |  |
| Median             | 201 | 208   | 196 | 41 |  |  |  |  |

Table 4. Summary statistics of chloride concentrations (mg/kg) in roadside soil in autumn 2004, 2006, 2007

conditions in winter are determined by slipperiness, amount of snow and evenness. Level 4 of state of road conditions means that the whole road surface must be clear from snow and ice. Level 3 means that wheel tracks on the road must be clear. By level 2, surface is allowed to be covered with snow or ice, but antiskid treatment is required  $[^{22}]$ .

It can be seen that cloride concentrations in roadside soils fluctuate widely in spring but in autumn are practically constant, independently of the road level.

With no regard to road level, the average cloride concentration in roadside soils in Tallinn in spring may be taken as 384 mg/kg within the distance from the pavement 5 m, and 224 mg/kg for the distance from 10 to 20 m. In autumn, from 5 to 15 m from the pavement, the average concentration is 206 mg/kg.

The activity of microorganisms depends on soil acidity, i.e., on the soil pH level. According to literature, cloride concentrations have a positive relation to soil pH and it is obvious that roadside soil in high traffic volume areas is more alkaline (increased pH) [ $^{30}$ ].

In Tallinn, the average pH of roadside soils of 205 samples was 7.36, median 7.45, standard deviation 0.5, minimum value 4.9 and maximum 8.18. The lowest pH values were found at sites S4 and S5. Statistical characteristics of soil pH at sites S1–S6 are shown in Table 5.

Soil  $pH_{KCl}$  values 4.5–6.5 have a favourable effect on plant growth. In non-polluted forests of Lahemaa, soil pH equals 3.

|                    | In spring 2003- | 2007 ( $n = 30$ ) | In autumn 2004, 2006, 2007 ( <i>n</i> = 18) |       |  |
|--------------------|-----------------|-------------------|---|-------|--|
| <i>L</i> , m       | 5               | 15                | 3–5   | 15–30 |  |
| Average            | 7.52            | 6.99              | 7.59  | 6.81  |  |
| Standard deviation | 0.27            | 0.81              | 0.4   | 0.88  |  |
| Minimum            | 6.97            | 4.9               | 6.65  | 5.04  |  |
| Maximum            | 8.18            | 8.03              | 7.92  | 7.86  |  |
| Median             | 7.48            | 7.2               | 7.67  | 7.19  |  |

Table 5. Summary statistics of soil pH in roadside soil at sites S1–S6 in spring and in autumn 2003–2007



Fig. 1. Average monthly ground temperature in Tallinn 2001–2007 (data from the Estonian Meteorological and Hydrological Institute).



**Fig. 2.** Average Zn, Cd and Pb concentrations in soils within 15 m from the pavement at sites S1–S6 in spring and autumn in 2002–2007.



Fig. 3. Chloride concentration in soil at sites S1–S6 in spring and autumn of different years.



Fig. 4. Lead concentration in soil at sites S1–S6. Target value is 50 mg/kg [<sup>33</sup>].



Fig. 5. Zinc concentration in soil at sites S1–S6. Target value is 200 mg/kg [ $^{33}$ ].



Fig. 6. Cadmium concentration in soil at sites S1–S6. Target value is 1 mg/kg [<sup>33</sup>].

# 3.3. Lead, zinc, cadmium and chloride concentration changes with the season

In the analysis of HM concentrations in soil, average values from sites S1-S6 were used. HM concentrations at site S7 were significantly higher than at sites S1-S6 in each year studied. The probable reason for that is a higher proportion of heavy-load vehicles and buses in traffic as well as their steep rise at site S7. Therefore data from site S7 were not taken into account in the calculation of averages. HM concentrations in the soils of sites 1-6 in different years are shown in Fig. 2.

Shown in Fig. 2 HM concentrations for autumn 2006 were measured in October. After a dry summer period, HM concentrations decreased still in November 2006, because of rainfalls in the autumn. Table 6 shows changes in the relative average HM concentrations at sites S1–S6 by the season, calculated as "(spring value–autumn value)/spring value". The least changes by the season are observed in Pb concentrations.

The behaviour of a metal depends on its chemical composition and the soil type. Once emitted, metals can reside in the environment for a long time. In  $[^{31}]$ , the concentrations of Pb, Cu, Cd, Zn, Ni, and Cr were measured in road dust and roadside soils. In general, the city centre and wasteland soils had enhanced heavy metal concentrations to at least 30 cm depth compared to park soils outside the city centre and rural soils in the region. For example, the mean Pb concentration was 104 (max 444) mg/kg at 0 to 5 cm and 135 (max 339) mg/kg at 30 cm depth in the city centre, while the background level was 17 mg/kg. In  $[^{32}]$ , it was found that Cd was the most easily exchangeable element in the case of variations in physical and chemical conditions; a significant risk of mobilization was to be feared in the case of zinc, which is very sensitive to acid pH. Pb and Cu did not appear to be highly mobile.

Figure 2 shows most clearly differences in Zn and Cd concentrations in spring and autumn. In spring 2006, Zn concentrations were higher than in spring 2005. Figure 3 demonstrates that Cl concentrations in soils in spring 2006 were significantly higher than in spring 2005. Average Cl concentration in soils at sites S1–S6 taken at L = 5 m from the pavement is 495 mg/kg. At L = 15 m it is 327 mg/kg in spring and 230 mg/kg in autumn.

|    | Year |      |      |       |                |  |  |  |
|----|------|------|------|-------|----------------|--|--|--|
| _  | 2003 | 2004 | 2006 | 2007* | Average annual |  |  |  |
| Pb | 27   | 40   | 48   | 10    | 31             |  |  |  |
| Zn | 42   | 50   | 47   | 33    | 41             |  |  |  |
| Cd | 28   | 69   | 79   | 68    | 57             |  |  |  |

Table 6. The relative average Pb, Zn and Cd concentrations at sites S1–S6 in different years, %

\* In autumn 2007 samples were taken on 10.09.07; later, after rainfalls, HM concentrations in the 10 cm surface layer decreases until the soil freezes.

The large amount of de-icing salt used in wintertime that accumulates in the roadside soil, also has a considerable influence on the solubility of most HMs. Growth in the quantity of chlorides, used in road maintenance, is accompanied by an increased concentration of zinc, usually also leading to an increase in cadmium. It has been suggested that zinc can be used as the indicator element to demonstrate the traffic pollution in spring [ $^{27}$ ].

Figures 4–6 show Pb, Zn and Cd concentrations in 2002–2007 depending on the traffic volume, i.e. study sites are ranked in decreasing order of AADT.

As Pb is primarily released as fine particles from exhaust emissions, it is prone to long distance transport and hence a smaller proportion is found in the local roadside environment.

Significant fluctuations in HM concentrations in Figs. 4–6 show the influence of weather conditions (amount of precipitation, soil temperature, the number of temperature courses through zero in winter months, etc.) and of the traffic volume on the spread of pollution. In the conditions of increasing traffic volume, the rate of extension in the area of pollution is approximately equal to the rate of increase in the traffic volume [<sup>34</sup>]. To achieve an accurate assessment, samples should be taken in different seasons.

## 3.4. Influence of de-icing chemicals on the groundwater quality

Another consequence of road maintenance in winter is that the soil and groundwater are salinated. The most usual de-icing salt is NaCl, that is extremely soluble. The unpleasant fact is that sodium is an undesirable element for vegetation, displacing other cations such as calcium, magnesium and potassium in soil and rendering these cations unavailable for plant uptake. Large amounts of chloride iones are apt to form mobile chloride–metal compounds. As chlorides are very mobile, metals tend to disperse in soil until a new equilibrium occurs. The speed of flushing out chlorides from soils to a great extent depends on the amount of precipitation.

In Tallinn, springs, opening at the foot of limestone shore or terrace have been preserved, e.g. Varsaallika springs on Kose limestone shore, the springs of the Glehn Park (named Rõõmuallikas) at the slope of Mustamäe Hill and Lepasalu springs in Mustamäe Park. In 2003–2005, in cooperation with the Institute of Geology, we conducted monitoring of spring water in Tallinn. Table 7 shows chloride concentration in spring water.

|                  | Date of sampling |           |            |           |           |           |  |  |  |
|------------------|------------------|-----------|------------|-----------|-----------|-----------|--|--|--|
|                  | 12.05.2003       | 6.10.2003 | 15.04.2004 | 7.09.2004 | 6.06.2005 | 5.09.2005 |  |  |  |
| Varsaallikas     | 105              | 182       | 249        | 78        | 9         | 78        |  |  |  |
| Rõõmuallikas     | 48               | 92        | 53         | 38        | 8         | 66        |  |  |  |
| Lepasalu springs | 42               | 41        | 40         | 21        | 10        | 57        |  |  |  |

Table 7. Chloride concentration (mg/l) in spring water in Tallinn in 2003–2005

Chloride concentrations in Tallinn spring water were compared with average concentration of chlorides from longstanding monitoring of analogous springs. In Pandivere reservation in West-Virumaa that is similar to Varsaallikas, the average Cl concentration was 16.8 mg/l in 1980–1990 and in Lahemaa Muuksi springs it was 7.8 mg/l in 1981–1990. At Koljaku spring in Lahemaa reservation that is analogous to Rõõmuallikas, the average Cl concentration amounted to 7.5 mg/l in 1981–1990. Lepasalu spring water quality can be compared with Viidumäe springs in Saaremaa, the average obtained from monitoring was 9.5 mg/l during 1974–1994. As Table 7 shows, concentrations of chlorides in Tallinn spring water are, as a rule, 4–15 times higher than in analogous springs in Estonia [<sup>10</sup>]. Table 7 reveals that contents of chlorides in samples taken on 6 June 2005 corresponded to the quality of analogous springs in reservations.

Table 8 shows that summer 2004 was extremely rainy, precipitation exceeded over three times the monthly average in July. In water samples, taken on 7 September, chloride concentrations were lower than earlier, but rain still "washed" soil in September and October.

Contents of chloride corresponded to natural spring water in samples taken on 6 June 2005 (Table 7). Movement of chlorides used for de-icing during winter 2004/2005 started in summer 2005, whereas chloride content in spring water had risen again on 5 September 2005. High concentration of chlorides in spring water is caused by de-icing of roads.

In Tallinn, near main road (e.g. Ehitajate tee), after snowmelt chloride content of 500 mg/l in site well 658 in groundwater was found. Though in a neighbouring well with the depth of 12.5 m, opening the confined Quaternary water layer, chloride content in groundwater was found several times lower, it is increasing year by year. That is an evidence of chloride migration through the impermeable layer of clay that allows for relatively low water flow. At the same time, the rate of mineralization is increased [<sup>21</sup>]. Groundwater, closest to surface, is directly affected by precipitation and water from snowmelt. Groundwater resources in Tallinn Quartenary water complex are attached to four water intakes (Männiku, Seevaldi, Ülemiste well field and Külmaallika). Therefore it is important to find solutions to chloride-free winter road maintenance in the recharge areas of water intake in Männiku, Nõmme and Järvevana tee. Undoubtedly, main roads with high traffic volume anywhere else in Estonia are also potential sources of pollution.

**Table 8.** Precipitation in Tallinn in 2003–2005, mm (data from the Estonian Meteorological and Hydrological Institute)

| Year  | Jan  | Feb  | Mar  | Apr  | May  | Jun  | Jul   | Aug   | Sep   | Oct  | Nov  | Dec  | Σ     |
|-------|------|------|------|------|------|------|-------|-------|-------|------|------|------|-------|
| 2003  | 47.3 | 6.4  | 9.4  | 42.7 |      |      |       |       |       | 58.2 | 48.3 | 79.0 |       |
| 2004  | 22.8 | 34.9 | 41.1 | 18.0 | 29.6 | 80.4 | 266.2 | 57.0  | 103.4 | 90.5 | 68.8 | 62.7 | 875.4 |
| 2005  | 81.3 | 23.8 | 6.8  | 5.6  | 36.8 | 61.3 | 61.3  | 153.6 | 25.0  | 44.9 | 67.8 | 37.3 | 605.5 |
| Norm* | 48.6 | 32.8 | 31.7 | 37.3 | 37.8 | 57.9 | 78.8  | 79.6  | 71.9  | 71.3 | 67.9 | 55.6 | 671.2 |

\* Average of years 1961–2003 that could be a monthly norm.

According to long-term investigations, in Finland the evaluation of pollution risk, arising from de-icing salts, is based on monitoring Cl content in ground-water [<sup>35</sup>]. No pollution risk exists if Cl content in groundwater is less than 10 mg/l. With the Cl content in groundwater ranging from 10 to 25 mg/l, pollution risk is present. It is suggested that measures for the protection of groundwater should be implemented if Cl concentration is over 25 mg/l [<sup>35</sup>]. Based on economic considerations and on the density of the road network, the target value for Cl concentration in Holland and Finland is 25 mg/l, maximum permissible being 200 mg/l [<sup>19</sup>].

In  $[^{36}]$  it was found that using of chlorides leads to the increase in HM (Zn, Pb), hydrocarbon (THC) and polycyclic aromatic hydrocarbon concentrations (PAH) in stormwater runoff from roads in winter months. An accumulation of contaminating compounds near the roads can be found to such an extent that mitigation must be considered. Using of chlorides for de-icing increases concentration of elements, hazardous to the environment, in stormwater runoff from roads and in roadside soil, and has a significant effect on the solubility and transport of HM compounds  $[^{37}]$ . It is important to monitor water quality, especially at the beginning of the spring flood period, when the level of road salt and other contaminants are markedly high in urban streams  $[^{38}]$ .

## 3.5. PAHs, chlorides and heavy metals

In spring 2005 concentrations of PAH-compounds in 20 soil samples were determined. According to marginal rates of hazardous substance contents in Estonia [<sup>33</sup>], benzo(a)pyrene concentrations exceeded the target value (0.1 mg/kg) in more than 60% of the samples, whereas half of the values exceeded it by two or three times. Maximum concentrations are found, as a rule, at 15 m from the pavement. As a major contaminant, benzo(a)pyrene is also included in the frame directive of the quality of outside air 96/62EU. Thus benzo(a)pyrene concentration in roadside soil has been recommended as one of the preliminary indicators in the evaluation of environmental conditions in terms of PAH [<sup>39</sup>]. Moreover, a certain degree of toxicity of asphalt pavement, based on benzo(a)pyrene content in oil shale bitumen as well as in oil bitumen, has been reported, whereas pavement surfacing with oil shale bitumen is considered to be somewhat more hazardous to the environment in terms of hygiene [<sup>40</sup>].

It was not possible to determine the relations between PAH-compound concentrations and traffic volume because of insufficient number of samples. To reveal a relationship between determined chlorides, PAHs and HMs in soils, at the sites S1–S7, a correlation matrix was calculated, using the Microsoft Excel environment (Table 9). The correlation exists when the absolute value of the coefficient is greater than 0.5.

Table 9. Correlation matrix of elements determined in roadside soils in spring 2005

|                            | Cl   | pH                                    | Zn                        | Pb       | Cd       | PAH |
|----------------------------|--|---------------------------------------|---------------------------|----------|----------|-----|
| Cl<br>pH<br>Zn<br>Pb<br>Cd | 1<br>0.483132<br>0.874206<br>0.81936<br>0.861003 | 1<br>0.118094<br>0.076346<br>0.200140 | 1<br>0.936151<br>0.964528 | 1        | 1        |     |
| РАН                        | 0.900015   | 0.236744                              | 0.877702                  | 0.825269 | 0.833383 | 1   |

### **3.6.** Possible solutions

Optimum traffic safety and environmental sustainability presume accurate and reasonable use of chemicals for de-icing and improved control of the results. This in its turn assumes real perception of the problem and possible solutions both in technical and economical terms, since pavements are degraded under winter conditions [ $^{41-43}$ ].

The results of the EU POLMIT project [<sup>19</sup>] include main findings of monitoring of 14 field sites in seven countries (Great Britain, Holland, Sweden, Finland, Denmark, France and Portugal), with evaluations of accumulation of single pollutants in soil and groundwater. Results from different countries vary to a great extent, however, it is concluded that metal emissions are in direct dependence on the traffic volume, PAH-compounds depend on the traffic volume and pavement type, and chloride concentrations depend on salt quantities, used in road maintenance during the monitoring period. In the EU POLMIT project, potential pollutant concentrations are compared with target and limit values, established in Holland (which are somewhat different in Estonia). It was concluded that hydrocarbons are pollutants in both groundwater and soil, and chlorides are pollutants in groundwater. Heavy metals (Cd, Cu, Pb, Zn) and PAH are potential pollutants in soil and groundwater [<sup>19</sup>]. In nothern England, Pb concentration was the highest from the four HMs, the concentrations of samples from 35 sites ranged from 25.0 to 1198.0 mg/kg [<sup>44</sup>]. According to the Swedish investigation [<sup>31</sup>], soil samples have to be taken from the surface layer and at depths lower than 30 cm in order to quantify the amount of accumulated metal in urban soil. It is also considered necessary and cost-effective to purify runoff, collected from motor roads and tunnels, starting from 20 000 AADT [<sup>45</sup>].

In [<sup>27,29</sup>], focused on a possible relationship between HM contents and traffic volume, a correlation between Cl and Zn, contained in the soil, was found. The comparison of concentrations with target and limit values, effective in Estonia, indicates that control over the spread of pollution in the case of traffic volume exceeding 15 000 AADT is necessary [<sup>29</sup>]. Regarding increased hazardous HM contents in soil as indicators of risk, the HMs can be ranked according to their risk value as follows: Pb, Zn, Cd, Cu and Cr. In all the samples Ni and Co contents were lower than the corresponding target value [<sup>27</sup>]. However, the test data used to evaluate environmental risks, caused by HM and chlorides originating from the road traffic, were insufficient.

To launch a problem-solving process whether the degradation of pavement, the road-draining system, splash from vehicle wheels, etc. can cause potential pollution risks to the roadside environment and groundwater, the scheme presented in Fig. 7 can be used.

The scheme considers three possible sources of pollution (de-icing salts, road furniture and traffic). In addition to traffic volume, it is necessary to consider all the factors influencing the environment, such as the proportion of heavy vehicle traffic, traffic speed, the year of road construction, pavement material, road draining system, type of soil, topography, size of the catchment, the number of population affected, etc.

To analyse the process on the right side of Fig. 7, an expert group (researchers of soil, chemists, biologists, hydrologists, hydrogeologists, geologists, ecologists, etc.) is required. When the soil types and vulnerability of the area are taken into account, conclusions about mitigation measures to protect soil and groundwater can be drawn. This scheme helps to determine vulnerable points between the traffic and the environment and to establish those, which first of all need continuous intensive national and local monitoring.

## 4. CONCLUSIONS

Heavy metals transportation to roadside environment due to traffic and pollution by chlorides, caused by the winter maintenance of roads, have been investigated in 2001–2007 in Tallinn. The main conclusions of this investigation are the following.

- On the basis of HM monitoring, characterizing the impact of traffic, average concentrations of Pb, Zn and Cd, collected from roadside soils up to 30 m in width and up to 10 cm in depth are as follows: Pb 48 mg/kg in spring and 40 mg/kg in autumn; Zn 160 and 100 mg/kg, respectively; Cd 0.72 and 0.68 mg/kg, respectively.
- Average chloride concentrations in roadside soils after snowmelt in spring were 380 mg/kg at about 5 m from the pavement, and 220 mg/kg at 10–20 m. In autumn the chloride concentration was 206 mg/kg in the 15 m wide area at the roadside.
- Roadside soil in Tallinn is mainly alkaline, average pH being 7.36, the minimum value equals 4.9 and the maximum value 8.18.
- Zn, Cd and Pb contents in soil vary according to the season. In the study sites, in soils at a depth of 10 cm, the HM concentrations, characterizing pollution caused by vehicle traffic, decrease by autumn compared to spring: Zn on average by 41%, Cd 57% and Pb 31%. Thus in the evaluation of the pollution of roadside environment through hazardous HM concentrations in soil, the time of the measurement, the depth of the sample taken and the spread area are important.





• In the conditions of intensive traffic, it is essential to conduct regular investigations to obtain up-to-date information on road and vehicle emissions. The scheme, shown in Fig. 7, can be used to simplify field site selection for the assessment of the impact of traffic on the environment in the national and local monitoring programme and in special municipal planning.

### ACKNOWLEDGEMENTS

This work was supported by the Estonian Ministry of the Environment, Tallinn Municipal Engineering Services Department and Tallinn Environmental Department.

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## Autoliiklus saasteallikana

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Teeäärse pinnase ja põhjavee saastumise peamised põhjustajad võivad olla sõidukite emissioonid ning teekatte ja teepäraldiste kulumine. Aastail 2001–2007 tehtud uurimused käsitlevad teede ja autoliikluse põhjustatud raskmetallide kannet teeäärsesse keskkonda ning saaste seost teede talvehooldusega Tallinnas. On toodud andmed liiklusega kaasnevast raskmetallide saastest linnaalade teede ja tänavate äärses pinnases. Raskmetallide Pb, Zn ja Cd sisaldus pinnase kattekihis väheneb sügiseks kevadega võrreldes: Zn keskmiselt 41, Cd 57 ja Pb 31%. Uurimuse kokkuvõttena on koostatud teedelähedaste alade põhjavee kaitsevajaduse otsuseprotsessi põhimõtteline skeem. See aitab täpsustada pinnase ja põhjavee seirevajadust ning seda saab kasutada planeerimisel riiklikul ja omavalitsuste tasandil.