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# SOIL POLLUTION BY HEAVY METALS AND POLYCYCLIC AROMATIC HYDROCARBONS IN THE TOWN OF BRNO (CZECH REPUBLIC)

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The authors present the heavy metal and PAH content in the upper 20 cm soil level in the town of Brno (Czech Republic). These results correspond to a network of 54 sampling sites in which the main soil properties, traffic intensity data and microclimatic conditions have also been determined. The average content of Cd, Cr, Cu, Ni, Pb and Zn in the 2M HNO<sub>3</sub> extract was 0.49, 6.7, 27.5, 8.0, 50 and 80 mg/kg. The total amount of Hg was 0.35 mg/kg and the sum of 16 PAHs obtained by dichloromethane extraction was 3.8 mg/kg. A soil metal index has been calculated characterising the total inorganic soil pollution relative to limit values. Areas influenced mainly by traffic and those affected by other small sources (mainly local heating) were distinguished by means of correlation analysis and the graphic processing of results (GIS). No remarkable industrial point source of pollution was found.

**KEY WORDS:** Heavy metals, PAH, soil, pollution, sources, traffic.

## INTRODUCTION

The pollution of urban soil by lead from traffic is a well known problem and is often documented. The fuel combustion in motor vehicles is responsible for the emissions of 273.10<sup>9</sup> g lead a year of the total world lead emissions amount of 449.10<sup>9</sup> g<sup>1</sup>. But recently, more and more works have been dealing with the pollution of urban soil by other risk elements and organic compounds. This is due to (a) reduction of the Pb content in fuels (b) determination of other pollution sources in towns and cities and (c) the effort for improving the environment in towns. The risk of soil pollution by cadmium in towns was described by Mielke *et al.*<sup>2</sup> and by lead and cadmium by Hernandez *et al.*<sup>3</sup>. The whole spectrum of toxic elements in these environments was studied by Thornton and Jones<sup>4</sup>; Bridges<sup>5</sup>; Wong<sup>6</sup>; Ho and Tai<sup>7</sup>; Lux *et al.*<sup>8</sup>; Tam *et al.*<sup>9</sup>; Leblová *et al.*<sup>10</sup>.

The PAHs are another important pollutant group in urban soils because of its mutagenic and cancerogenic activity<sup>11</sup>. Compared with lead, the proportion of traffic on air deposition of PAHs is much lower, as they originate by pyrolytic reactions during any combustion of

organic matter. Their principal global source is the coal industry and production of energy, whereas traffic participates in PAH deposition only by about 1%<sup>12</sup>. However, in developed countries the distribution of PAH sources is quite different: for example using the data of Grimmer<sup>13</sup> we can estimate the proportion of traffic on PAH air depositions in Federal Rep. Germany to be about 13%. In towns with a higher density of transport, the importance of the traffic among other PAH sources must be even greater. So many papers dealing with the measurement of PAHs in urban air, soils or street dust are published every year (according to Chemical Abstracts 40 in 1989, 60 in 1990, 98 in 1991). Such studies on metal and organic pollution in towns are of great importance not only from the scientific point of view, but also for the elucidation of regional pollution conditions, different from town to town due to the structure and the intensity of pollution sources, climate, landscape and soil properties. Therefore, the results can be of particular interest to (city-)health organizations, urban planners, traffic divisions, environmental scientists, and the general public.

The Czech Republic is, unfortunately, well known as a country with a very bad environment, but the subject of this article is situated in the center of a relatively clean southeast region with average SO<sub>2</sub> aerial concentrations about 20 µg/m<sup>3</sup>, reaching 50 µg/m<sup>3</sup> in the limits of the town. Brno, with its nearly 4.10<sup>5</sup> inhabitants, is the second largest town in the Czech Republic and a very important center of commercial, industrial, and transport activities. The main part of the town is situated in a vast valley closed from three sides by the slopes of the woody headlands of the Bohemian massif with altitudes up to 300 m. Towards the south the landscape is open to the plains of the common basin of the rivers Svatka and Svitava. However, the very bottom of the valley (altitude about 200 m) is dominated by two rocky cliffs, the one where the city castle is situated and the other with the cathedral, both forming the original town center. The environment in the town is supposed to be strongly affected by the traffic, the intensity measured reaching 3.09.10<sup>5</sup> vehicles a day in the very center of the town, mainly due to the north-south axis.

## MATERIALS AND METHODS

54 sampling sites were chosen in the town (see Figure 1). The sites were proposed by mutual collaboration between urban planners, soil scientists and representatives of the town hall in order to reach the following criteria: (a) to include all main traffic networks and crossroads, (b) to have the possibility of assessing frequently visited public places (children's playgrounds, parks, schoolyards etc.), (c) to allow comparison with the background localities, (d) to be overgrown by grass to allow subsequent plant analysis and (e) to cover the whole town area. The samples were taken during one week with stable weather conditions at the beginning of July 1991. Each sampling point was represented by a rectangular area of 2 × 5 m from which about 25 individual samples were mixed to give a representative average sample. Along the roads, rectangles were close to the edge only. The surface layer was sampled to the depth of 20 cm.

The samples were dried at room temperature, sieved and homogenized. Since most places were situated on anthropogenic soils, pH<sub>KCl</sub>, cation exchange capacity (CEC), humus and clay content were measured to characterize the soil. Then, contents of Cd, Cr, Cu, Ni, Pb,

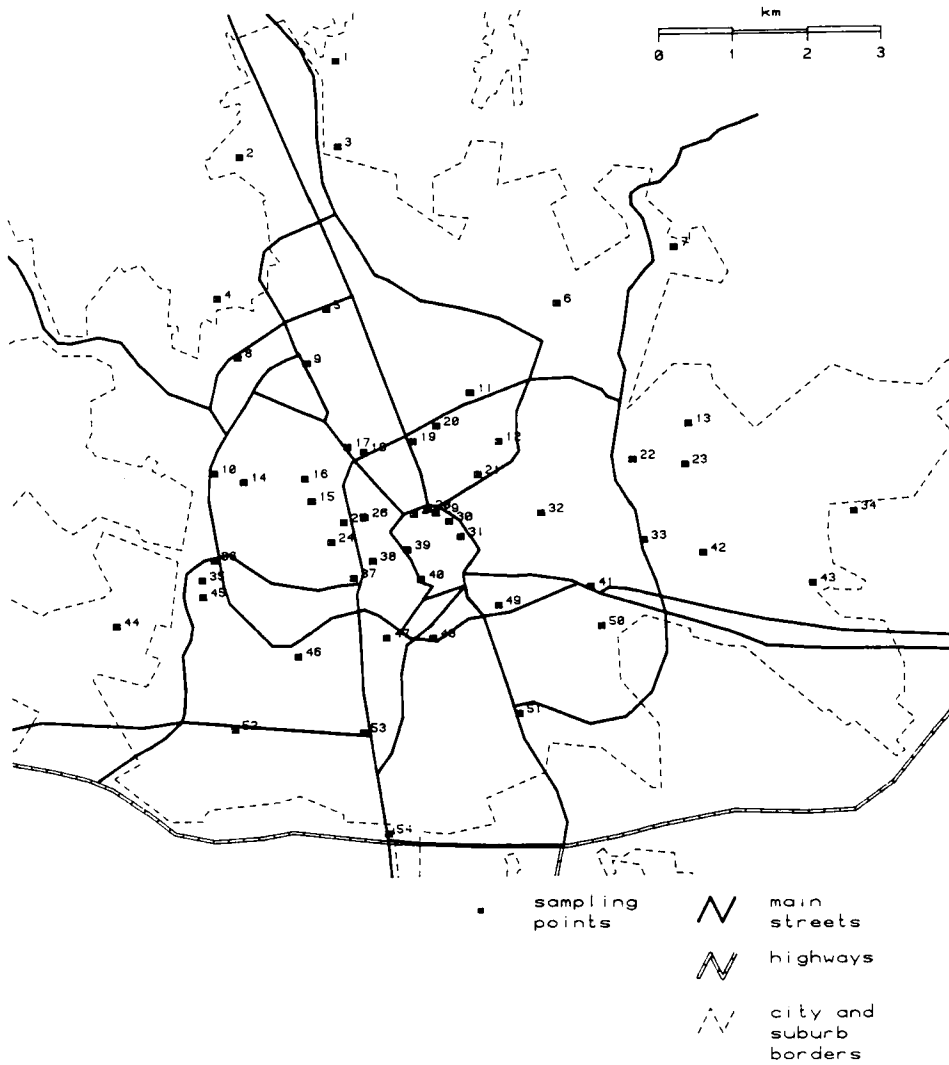


Figure 1 Soil sampling locations in the inner town.

and Zn were analyzed in an extract of 2M  $\text{HNO}_3$  by means of an atomic absorption spectrometer (Perkin-Elmer 5100 PC). The total Hg content was analyzed on a trace mercury analyzer (Labora TMA 254). The polyaromatic hydrocarbons were analyzed by the following procedure<sup>14</sup>: the soil was extracted for 10 hours with dichloromethane in a Soxhlet extractor, after the extraction the volume was reduced to 10 ml, this volume (or its exact part—according to expected concentration) was mixed with 0.5 g of activated silica. Then the solvent was carefully evaporated and the sample was dried at ambient temperature in a dessiccator. This extract was adsorbed on silica and placed on a glass column with  $15 \times 1$

cm of activated silica and the column was then gradually eluted by n-hexane and dichloromethane (DCM), the flow of the DCM fraction (in which PAHs are dissolved) through the column being watched in UV light.

This PAH fraction was concentrated at a temperature of 40 °C in a gentle stream of nitrogen to the volume of 0.2 to 0.8 ml and subsequently analyzed by a HRGC. The analysis was carried out by capillary GC-FID on a Hewlett-Packard HP 5890A with a HP 5895A GC Workstation. The gas chromatograph was equipped with a fused silica capillary column DB-5 (J&W Scientific, length 30 m, inner diameter 0.25 mm). The samples were injected in the splitless method, hydrogen was used as a carrier gas. The temperature program was as follows: initial temperature 40 °C, rate 5 °C/min up to the final temperature of 300 °C, final isotherm 23 minutes. The limits of detection of individual hydrocarbons at these conditions were about 1 ng/g. The following 16 individual hydrocarbons were determined (according to EPA list): naphthalene, acenaphthene, acenaphthylene, fluorene, anthracene, phenanthrene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b) and benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene. The total amount of PAHs mentioned further in the text is reflected to the sum of these 16 individual hydrocarbons. The analytical standards of all 16 PAHs were obtained from Supelco SA.

The measured soil characteristics, metal content and the polyaromatic hydrocarbons were then processed by means of regression analysis with special regard to the influence of traffic. For that purpose, indexes of soil exposition to influences of traffic (TII = traffic influence index) were calculated for each sampling point using the formula:

$$TII = MC \cdot TD$$

where: MC = microclimatic conditions of a locality given by its building profile. This index was evaluated according to the height of buildings (vegetation) and their distance from roads:

Where: MC = 0.25—open localities without any buildings or vegetation along roadsides  
 MC = 0.50—low buildings on one side of the road or on both sides but very low and at some distance

MC = 0.75—high buildings along one side of the road or low buildings on both sides

MC = 1.00—high buildings, very near along both sides of the road

For localities without any influence of traffic, the TII was set to 0.01.

TD = traffic density given by the number of all vehicles passing during 24 hours (in tens of thousands).

Because the TD numbers were known individually for lorries (mostly diesel) and benzine cars, the TIIs were calculated also for these two groups:

TIIb = MC · TD<sub>b</sub> for vehicles with a benzine engine

TII<sub>d</sub> = MC · TD<sub>d</sub> for vehicles with a diesel engine

Plants were also sampled at each sampling point and analyzed. It will make possible to evaluate the role of soil on plant contamination in comparison with air pollution.

For the whole soil heavy metal burden, the special index SMI (soil metal index) was calculated for each locality. The relative values of element contents with regard to the limit

(= 100%) were calculated at first. These values were summed up and expressed again as an average of relative limit values (= 700% because of 7 elements)

$$SMI = 1/n \cdot \sum_{i=1}^n 100 \cdot V_i/L_i$$

where:  $n$  = number of elements

$V$  = content of an element in the soil (mg/kg)

$L$  = limit value of an element content in the soil (mg/kg)

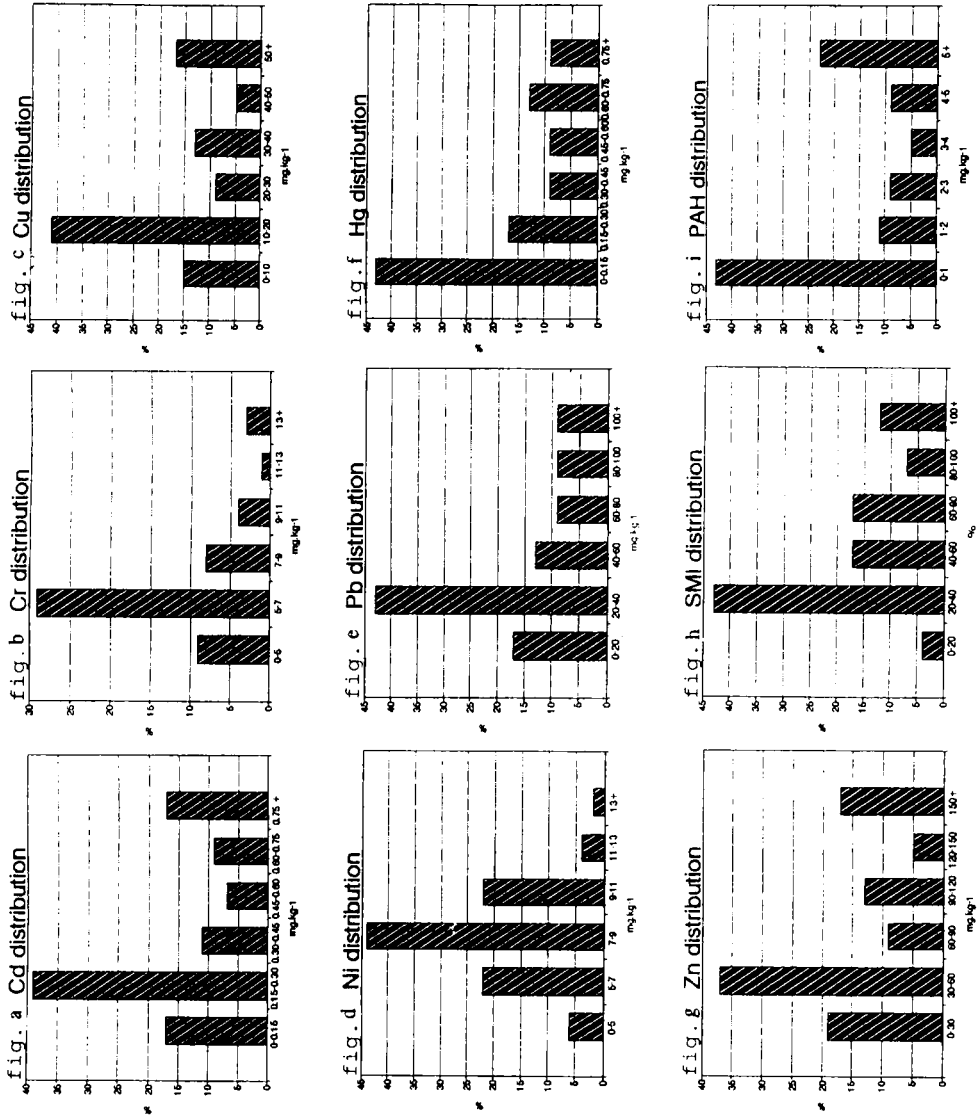
## RESULTS AND DISCUSSION

The average values of metals and polyaromatic hydrocarbons in the soils of Brno, shown in Table 1, seem to be not so serious although an increase with respect to the background values is evident<sup>15</sup>: Pb and Cd  $2.5 \times$ ; Hg  $10 \times$ ; Zn  $1.5 \times$ , PAHs  $3.8 \times$ . On the other hand, in comparison with similar studies<sup>2,4,7,16,17</sup>, which have analyzed the total metal content, the results are in most cases several times lower, partly due to the weaker extraction procedure of elements by nitric acid of this study. The frequency distribution of these heavy metals and polyaromatic hydrocarbons contents is shown in Figures 2, their superficial distribution in Figures 3–6.

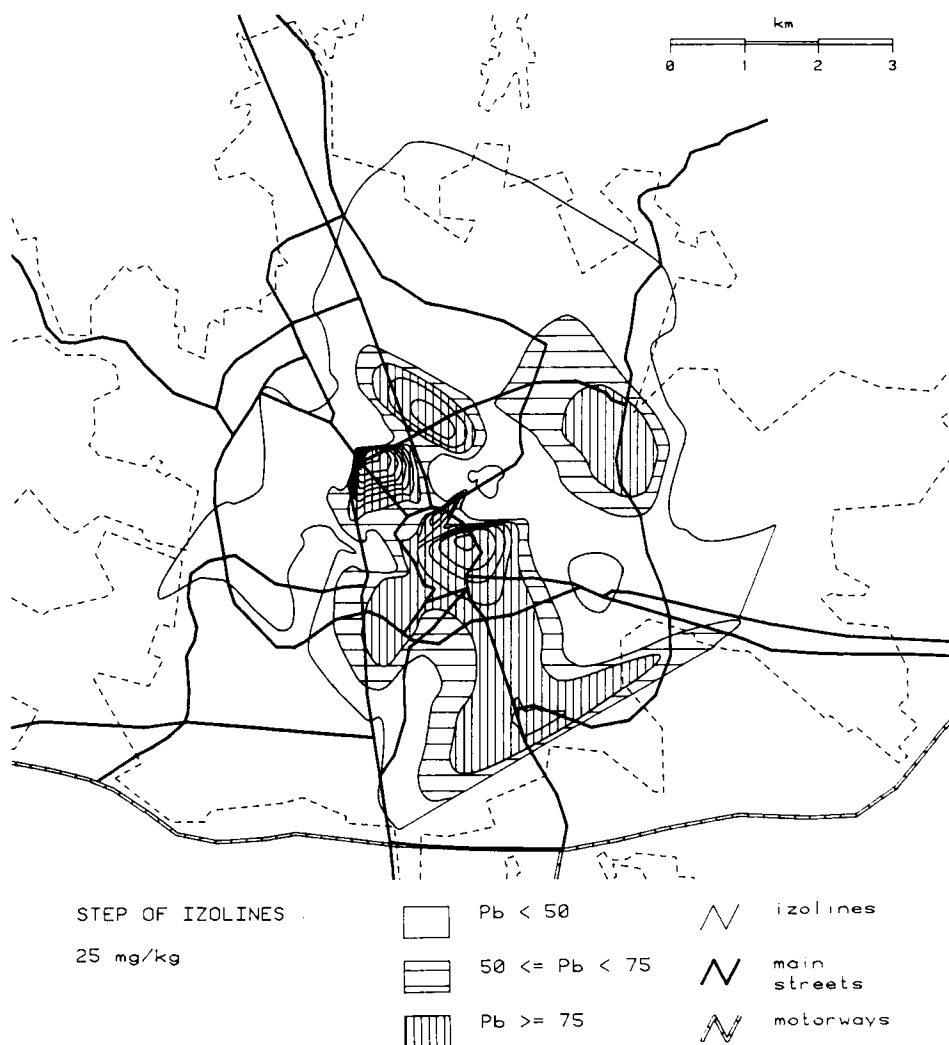
The dependence of soil pollution on soil characteristics was tested by means of regression analysis. As it can be seen in Table 2, no significant relationship was found for  $pH_{KCl}$  (the critical value for 54 couples and the level of confidence  $P = 0.01$  is 0.35). Only the humus content (Cox) seems to have a statistically important influence on the content of some elements (Cd, Cr, Cu, Hg, Zn) in the soil. The distribution of these elements depends only very slightly on the traffic and that is probably why their bounding on organic matter could be displayed. The CEC factor is statistically important for Cr and Ni, clay content influences Ni only. More significant relationships between soil characteristics and heavy metal content, which were found in some studies concerning agricultural soils, were restrained by dominant anthropogenic factors here. Soils in towns are mostly of anthropogenic origin and moreover they are exposed to decisive airborne deposition impact. The illustrative average characteristic of soil pollution by heavy metals could be represented by the soil metal index (SMI). Its distribution in the Brno soils is given in Figure 2. Although it cannot precisely reflect the

**Table 1** The average values and ranges of heavy metal and PAH contents in soils (mg/kg).

component	average	geom. mean	range
Pb	50.1	38.2	13.9 – 257.4
Cd	0.49	0.31	0.01 – 3.01
Cr	6.7	6.5	4.23 – 15.8
Hg	0.35	0.21	0.023 – 1.28
Ni	8.0	7.8	3.8 – 15.7
Zn	79.9	60.1	11.05 – 310.0
Cu	27.5	21.2	4.9 – 95.0
PAHs	3.78	1.67	0.175 – 45.75



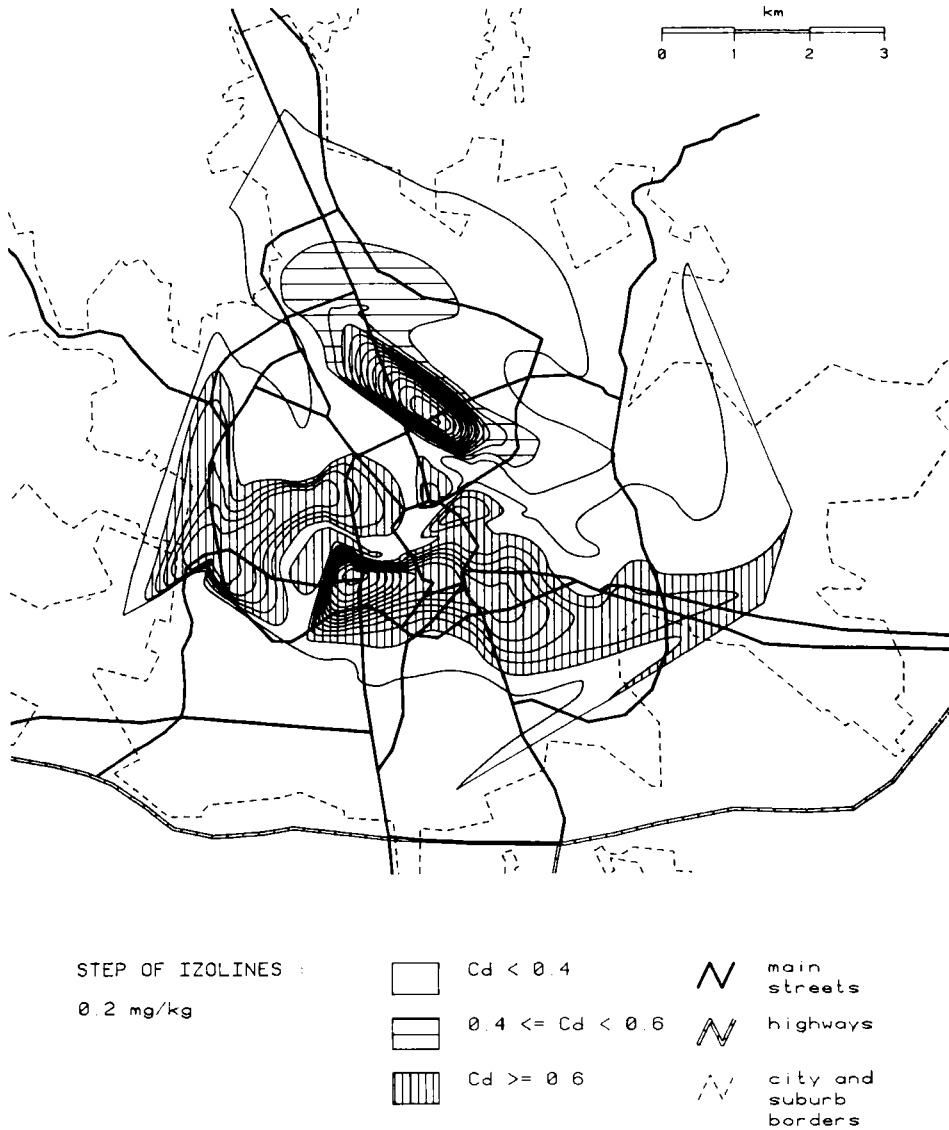
**Figure 2** Distribution of heavy metals, PAHs and SMI indexes in the soil. Values on the Y axis are percentage from the number of localities.



**Figure 3** Level of soil pollution by lead [mg/kg].

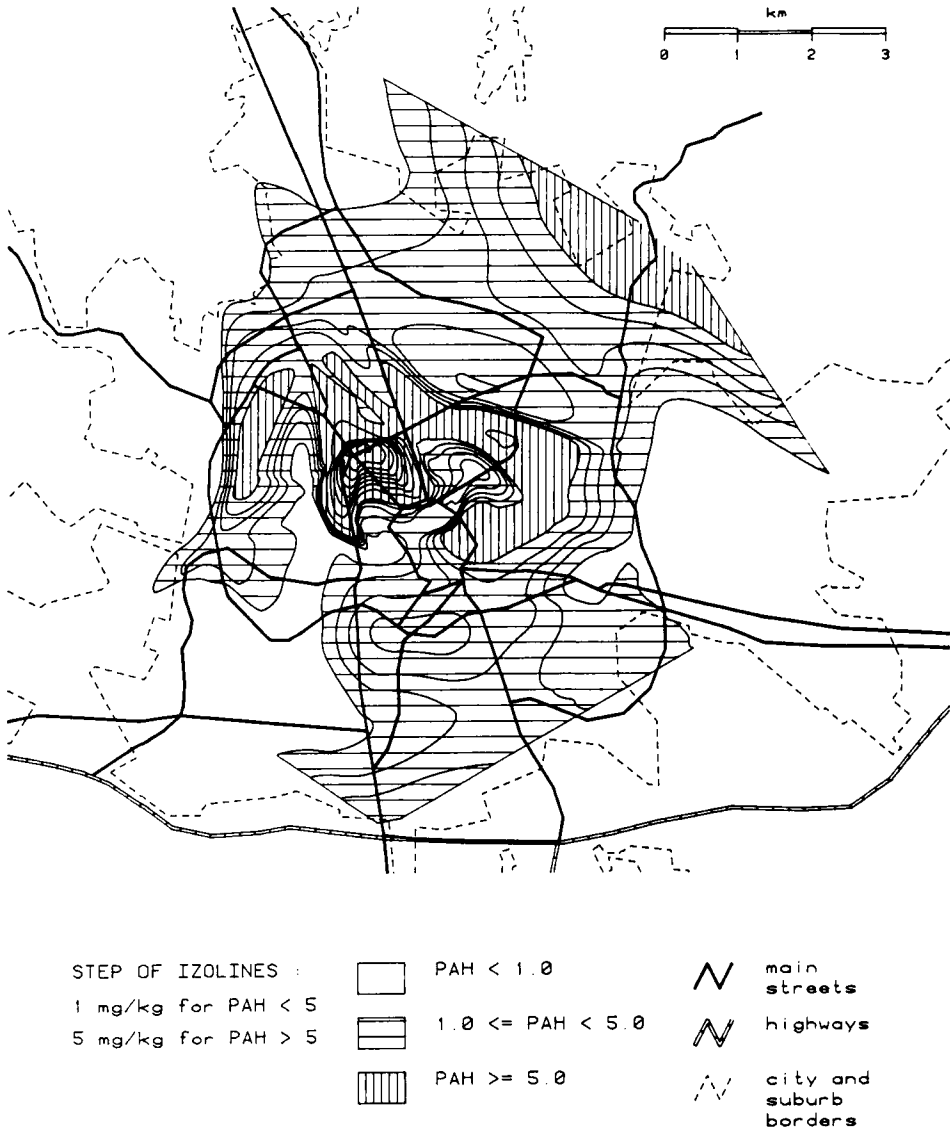
excessive pollution by one or two individual metals, it characterizes the whole burden of any locality. It can be seen from the correlation coefficients between the SMI and the individual metal contents (Table 2) that the main pollutants influencing the whole burden are, in decreasing order, Zn, Pb, Cd, Hg and Cr. It should be taken into account that the SMI values can be different due to the limit values of heavy metals in soils which vary according to the country. Since the SMI is a relative value, exceeding the limit 100 means an inadmissible concentration of observed elements in the soil. Figure 2 shows that 12 percent of the examined localities have a SMI value higher than 100. All of them are situated in places with a high traffic density, especially near crossroads.





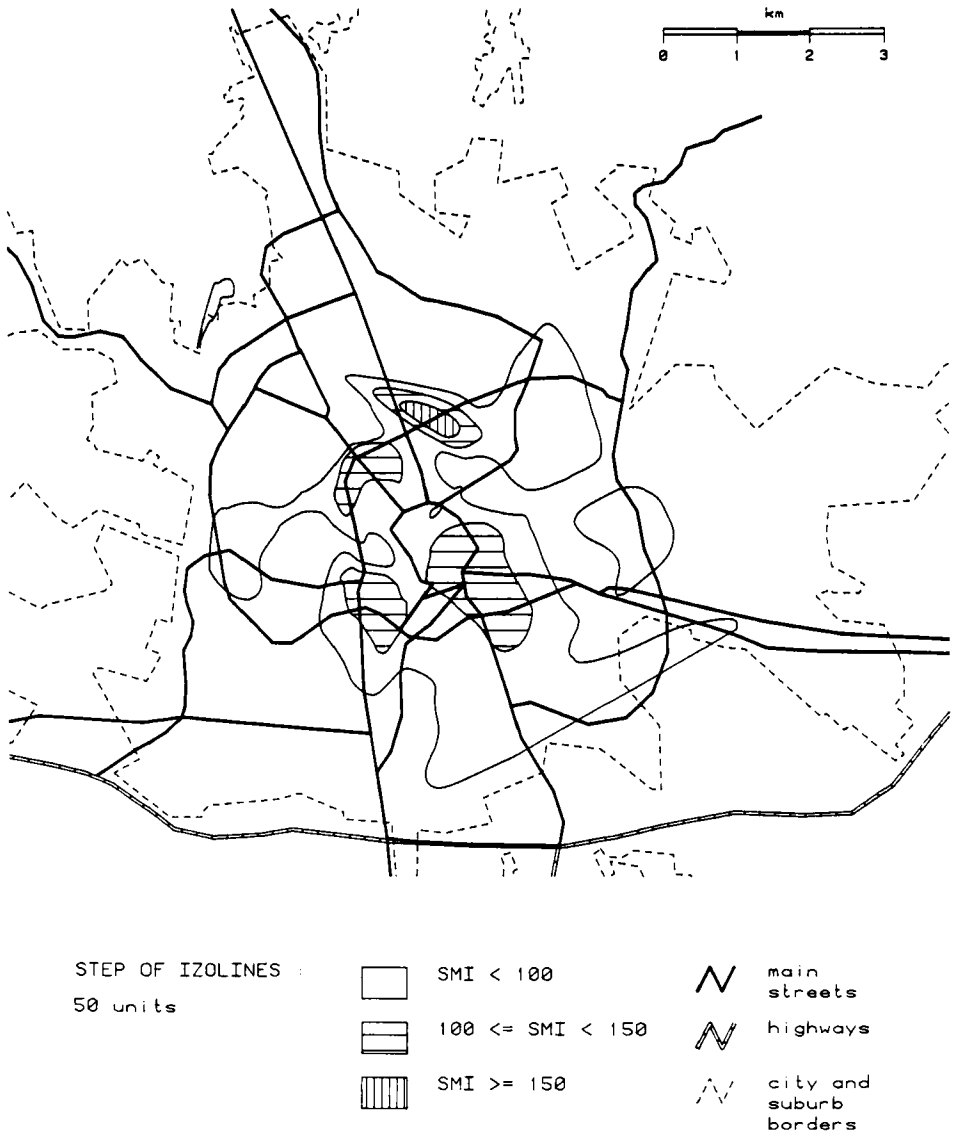
**Figure 4** Level of soil pollution by cadmium [mg/kg].

Based on the traffic influence index (TII), the importance of traffic on the soil pollution was calculated by means of regression analysis (Table 3). A significant relation was only found for lead, SMI, and PAHs. TIIb (benzene) has a stronger relation with Pb than TIIc (diesel), which corresponds to the content of lead in benzene. In the case of PAHs, stronger dependence was found with TIIb than TIIc although the difference is not so big at the level of significance. It is evident that traffic density and the dispersion of exhaust fumes, which



**Figure 5** Levels of soil pollution by PAH [mg/kg].

are dependent on the microclimatic conditions, strongly influence the content of lead in the soil. Nevertheless, for better accuracy, other factors should be included in the TII calculation, notably the speed of vehicles and the time of soil exposition to traffic influence. These factors are confusing especially in towns since the traffic speed is different in various sections and due to construction activities the soil can be turned, removed or dumped.



**Figure 6** Soil metal index [units].

These statistically important correlations were treated by multiple regression statistical analysis. The graphic representation of component and residual values (Figure 7) gives following pictures:

The couple of PAH and TII values shows significant positive residuals at 8 localities (points 7, 10, 12, 14, 17, 18, 25, and 32). Six of them are accompanied by high positive

**Table 2** Correlation coefficients between soil characteristics (analyzed and calculated) and contents of pollutants (heavy metals and PAHs).

<i>component</i>	<i>pHKCl</i>	<i>Cox</i>	<i>CEC</i>	<i>clay cont.</i>	<i>SMI</i>
Pb	0.21	0.22	0.06	0.22	0.82*
Cd	0.07	0.55*	0.34	0.11	0.67*
Cu	0.14	0.44*	0.19	0.21	0.19
Cr	0.21	0.58*	0.42*	0.24	0.38*
Hg	0.19	0.39*	0.23	0.26	0.57*
Ni	0.03	0.33	0.42*	0.36*	0.30
Zn	0.17	0.43*	0.19	0.22	0.91*
PAHs	0.10	0.01	0.16	0.24	0.48*

\*correlation coefficients higher than the critical value 0.35

residuals for couples PAHs/Pb. This result supports the hypothesis that at these points we can see a very strong influence of other PAH sources than traffic, probably of local heating or the incineration of garden wastes. This idea is corroborated by the fact, that points 7, 12, 14, and 32 are situated on calm streets in residential areas with low TII values. The elevated PAH contents at points 17 and 25 on busy crossings seem to be supplementarily influenced by other PAH sources, probably by local heating of old houses in this area. The elevated PAH concentration at point 10 corresponds to its high intensity of traffic, but the whole effect of multiple regression is affected by the low Pb value, which is due to relatively recent construction of the crossing when huge amounts of clean soil were removed here. The concentration of Pb in the soil is normally a result of a long-term aerial pollution, which is not this case. The worst polluted point 18 is a badly aerated crossing where an important proportion of vehicles turn to the left in all four directions and wait for a long time inside or in the vicinity of the crossing (see also Figures 3 and 5). Therefore the levels of Pb and PAH pollution here exceed proportions observed anywhere else in the town.

Significant negative residuals on this plot can be seen at 8 localities (points 20, 22, 29, 30, 31, 39, 40, 49 and 51). With the exception of point 22 they are all situated on straight sections or crossings of green-wave principal roads with negligible or no turning to the left. Relatively lower than predicted proportion of PAHs here may reflect the fact, that the whole regression takes into account all PAH sources together and the sites with only one highly dominant source—the traffic in this case—seem to be less contaminated. This explanation is supported by the coincidence with positive residuals on Pb/TII plot (Figure 7) and negative residuals on PAH/Pb plot. The exception of point 22 with very high Pb pollution (92.5 mg/kg) and the almost background PAH value (1.5 mg/kg) could be explained by its special

**Table 3** Correlation coefficients between indexes TII and soil pollution characteristics.

	<i>Pb</i>	<i>SMI</i>	<i>PAH</i>
TII	0.61	0.61	0.56
TIIb	0.62	0.61	0.57
TIIc	0.51	0.57	0.47

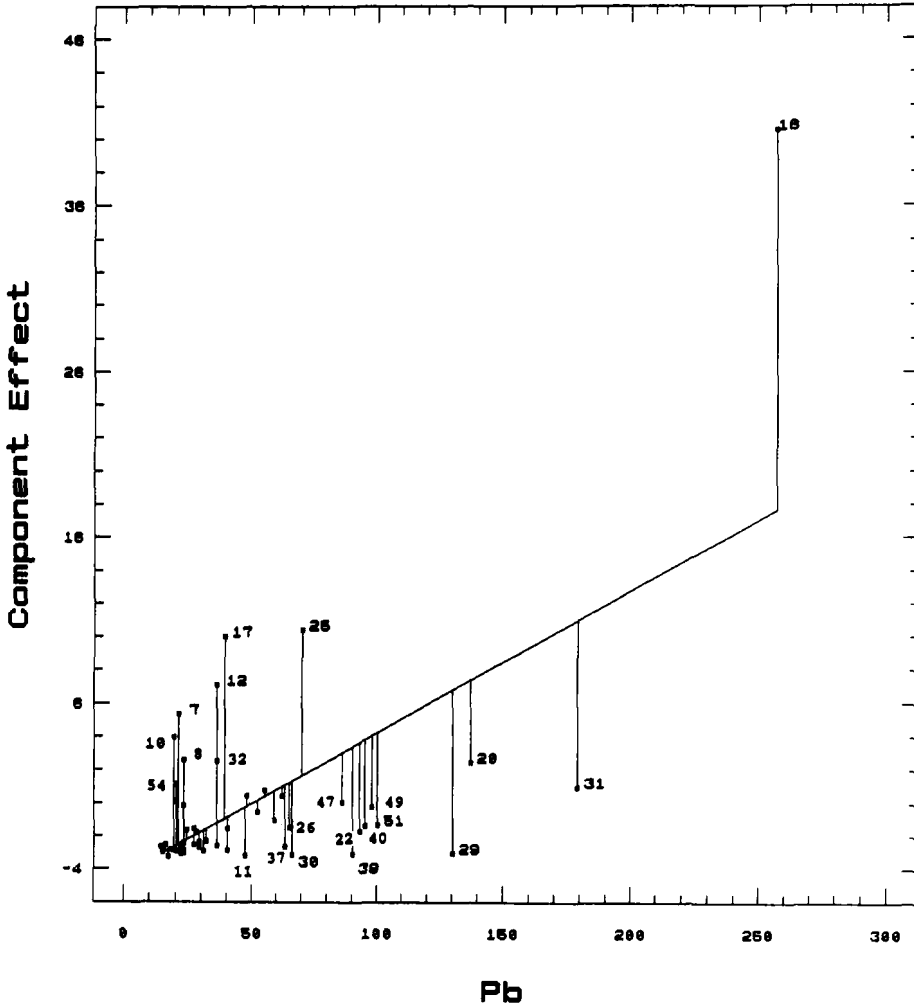


Figure 7a Component + residual plot for total amount of PAHs.

position and different lifetimes of these two pollutants. This point is situated on a calm street in a residential area, which is limited at a distance of about 150 m by two heavily frequented principal roads and their crossing. The low altitude (213 m) of the area causes very frequent temperature inversions mainly in winter, when the stream of smog from heavy traffic inundates the almost adjacent area. Compared with Pb, the halftimes of PAHs decomposition in soil are relatively low (weeks or months<sup>18-20</sup>) and therefore the missing quantity of PAHs may have disappeared by the time of sampling (June).

Positive residuals on the Pb/TII plot have been partly mentioned as the sites with lowered PAH/TII values on busy streets and crossings. There are 3 other points in this group: points

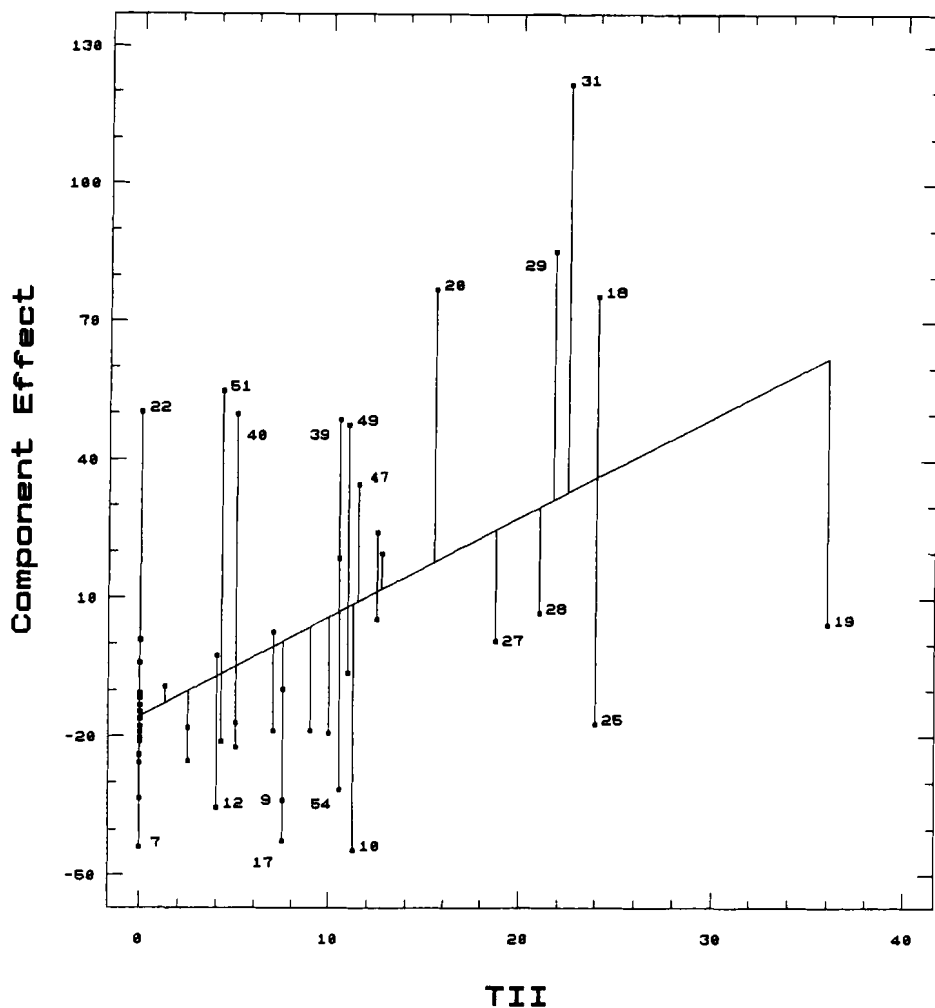


Figure 7b Component + residual plot for total amount of Pb.

11 and 49, a very busy street and crossing with predominant traffic influence and the extremely polluted point 18. This extreme suggests that our calculation of TII should involve a factor of speed, which would take into account the duration of the pollution event. A car driving with the speed of 10 km/h pollutes the crossing approximately three times more than the identical vehicle at 50 km/h (not calculating the lower efficiency of engines at lower speeds) and a waiting car pollutes its neighbourhood even more.

Significant negative residuals can be seen at 10 points of this graph. Points 7, 12, 17 and 25 have been mentioned as examples of a principal or additional non-traffic PAH pollution, point 9 seems to be a similar case. The observed content of Pb does not correspond to elevated

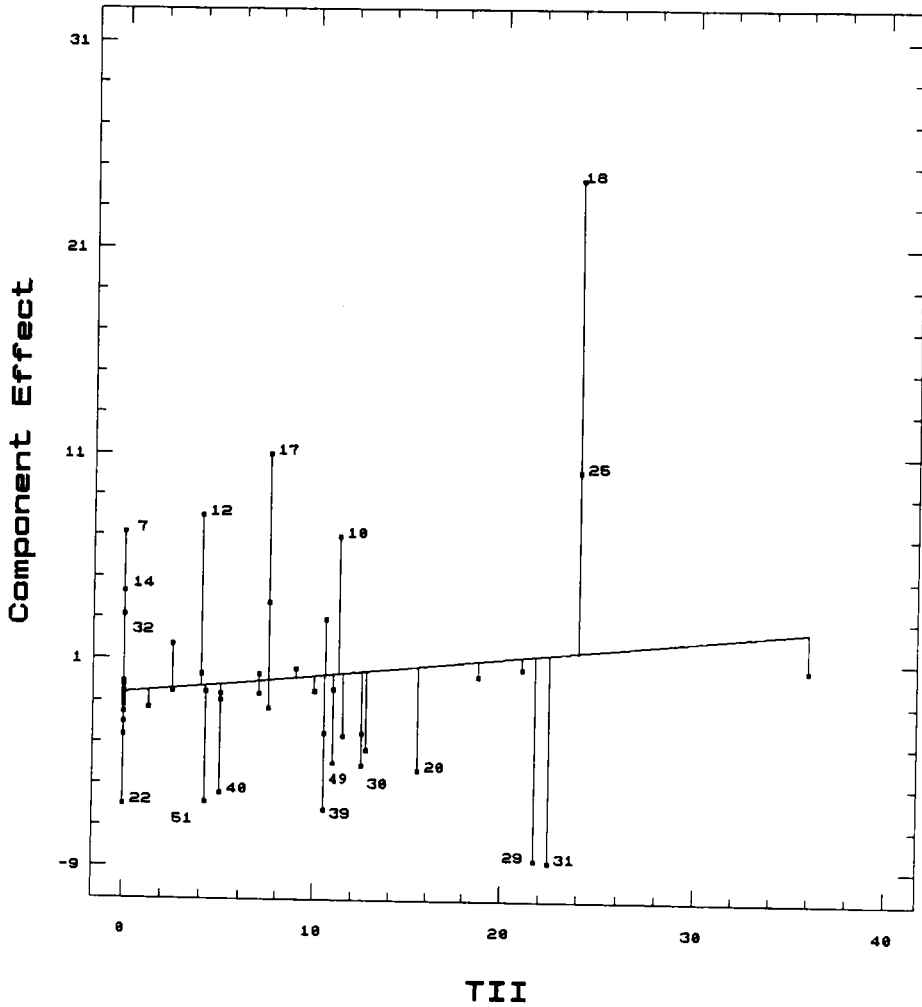


Figure 7c Component + residual plot for total amount of PAHs.

PAH values with respect to the close correlation PAHs/Pb. Points 10 and 54 reflect recent construction works on these crossings and represent the true lack of Pb content. The lowered Pb values on very busy crossings 19, 27 and 28 are probably connected with the high proportion of heavy diesel vehicles at these sites.

The highest correlation coefficient has been found between PAHs and Pb content (Figure 7). The positive residuals of multiple regression coincide with the already mentioned cases of elevated PAH/TII values. Point 8 is the only one which has not been mentioned before: its location in a residential area suggests that the relatively elevated PAHs quantity (3.2 mg/kg) and the almost normal Pb value (22.9 mg/kg) can be explained by the influence

of small domestic sources. All positive PAH residuals of assumed non-traffic origin are situated partly around the local northwest maximum and, the other group, in the centers of northeast and east local maxima of PAHs concentration (Figure 5).

Nine important negative residuals on this plot correspond to elevated Pb/TII values. With the exception of point 22 all of them are situated on busy streets and crossings with traffic as the predominant source of pollution. The same holds for points 26, 30 and 37. The reason for low PAHs (0.967 mg/kg) and the relatively higher Pb content (47.2 mg/kg) on site 11 could be similar as for point 22—originally higher PAH values from winter have already decomposed at the moment of summer sampling. This probable decomposition of PAHs may also explain our problems with the interpretation of pattern distribution of individual hydrocarbons.

We have also analysed the correlation between all mentioned values and the altitude to examine the influence of general landscape. As expected, all correlation coefficients have negative values. That of PAHs has a negligible significance level 0.39, Pb is correlated at the significant level of 0.03, and the highest one is the correlation between TII and an altitude with the significance level of 0.003, which has a very trivial meaning: important streets are constructed rather in basins than on knolls.

To elucidate some causes and relations of different pollutants within the town soils we have chosen the most typical examples of Pb, Cd, PAHs and SMI for designing their spatial distribution by means of ARC-INFO geographical information system (Figures 1, 3–6). Only one couple of maxima on graphs 3 and 4 (Pb and Cd) coincide: the north ones. With this exception, which is difficult to explain, we can see totally different distributions of these two elements: a well-marked NW-SE oriented zone of elevated Pb values and a perpendicular belt of elevated Cd values. PAHs distribution in Figure 5 differ totally from that of Cd, but there is a NW-SE oriented zone of elevated PAH values similar to that of Pb. We can see three additional PAH source areas on graph 5: one on the NW and the other on the NE borders and the east one near the center. All of them represent relatively old residential areas. The SMI isolines (Figure 6) evidently show areas with the highest total soil metal burden and reflect especially the pollution by Pb, Cd and Zn (see Table 2). On the other hand, the possible excess of an individual element could be suppressed in this display.

## CONCLUSIONS

The results presented on the determination of heavy metals and polyaromatic hydrocarbons in town soils have revealed that this environmental compartment is seriously disturbed, mainly in the center of the town. The concentrations of PAHs in the soil are higher than the limit background value (1 mg/kg) at 57 % of measured points, the worst polluted point 18 reaching the value 45 mg/kg, Pb concentrations are higher than the background value (20 mg/kg) at 83 % measured points with the highest value 257 mg/kg also at point 18. Since no industrial point source has been found, the traffic is presumed to be the main source of pollution. This idea is supported by the results of correlation analysis and the multiple regression of all experimental data, where the most significant correlations are those between



Pb, PAHs and the traffic influence index. Houses with a high percentage of solid fuel heating are another important pollution source in residential areas, contributing especially to elevated PAH values.

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