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Can the contamination of urban street sediment be used as an indicator for traffic density? A case-study in the city of Leuven, Belgium.

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Abstract

The concentration, in urban street sediment, of 13 chemical elements was measured in the city of Leuven, Belgium. A total of 77 locations were sampled in pedestrian zones, streets with limited traffic, streets with much traffic, and on the ring road that has very busy traffic. The data were compared to NO₂ concentrations measured by the Curieuzeneuzen-Vlaanderen project, a large-scale study with 20,000 sampling points all over Flanders (the northern part of Belgium). NO₂ is a frequently used indicator for traffic pollution. In Leuven the highest enrichment (strongest pollution) was measured for Cu, Sb, Pb and Zn. These elements could be related to brake wear and tire wear, respectively. For Cu and Zn the concentrations in Leuven exceed those in most of the other cities investigated in the literature. Moderate enrichment was measured for Cd, Cr, Ni and S. The other elements (As, Co, Fe, Mn and V) showed concentrations close to the background value in unpolluted soil. The Integrated Pollution Index (IPI) for Leuven is 8.15, which, according to criteria proposed by the literature on street sediment, classifies Leuven, on average over its surface area, as a "very highly polluted" city. As expected, for Cu and Sb the highest contamination is found on the ring road and the busy traffic circulation loops. For Zn and Pb, on the other hand, the highest contamination occurs in the city center, in the pedestrian zone where no traffic is allowed except for buses and taxis. We hypothesize that this is a result of historic accumulation of these elements at the time traffic was still allowed in this zone. In Leuven the chemical composition of street sediment did not correlate to the NO₂ concentrations.. This study shows that measurements of current pollution by traffic are not sufficient to determine the health risk because much exposure to toxic substances may be caused by resuspension, by traffic or wind, of substances that have accumulated in the city over time, sometimes decades ago, when regulations were much less stringent than today.

Key words: street sediment, dust, heavy metals, pollution, traffic

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Highlights

- A total of 13 chemical elements were measured in urban street samples
- Strongest pollution can occur in dense traffic or in traffic-free pedestrian zones
- Pedestrian zones can be heavily polluted by historic accumulation of contaminants
- Pollution of urban street sediment was not found a good indicator for traffic density

1. Introduction

Cities are important sources of dust production. Two types of dust are produced in cities: (1) gases and particles produced by combustion processes, and (2) mineral particles (Lu et al., 2009; Al-Awadhi and Aldhafiri, 2016). The first group includes vehicle exhaust and other combustion gases such as carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO₂) and volatile organic compounds (VOCs), and fine solid particles produced during combustion such as diesel soot. The second group consists of mineral particles originating from street works, building and infrastructural works, parks, vacant lots vulnerable to wind or water erosion, and similar sources. These mineral particles are much coarser than combustion dust. At roadside locations, most traffic exhaust particles are 10-30 nm in diameter (Gidhagen et al., 2004) whereas the mineral particles are often coarser than 100 μm and can sometimes even be coarser than 2 mm (El-Hasan et al., 2006). The term “street dust”, which is abundantly used in the literature, is somewhat misleading because in most studies it is applied to any size of loose mineral particles on the road or sidewalk. Dynamically, dust refers to particles smaller than approximately 60 μm in diameter, which are usually transported in true suspension (Pye and Tsoar, 1990). This study uses the term street (or road) sediment to describe the loose mineral particles on the road surface.

If not too coarse, both combustion-generated dust and mineral particles are easily resuspended by traffic (Amato et al., 2011). In contrast to combustion dust, which can easily be evacuated by wind due to its very small size (but which is continuously re-produced by traffic or other human activities), the coarser mineral particles will settle rather quickly and may remain in the city for a long time. Mineral road sediment has been identified as one of the most significant sources of fugitive dust in cities. In Stockholm, road sediment was found to account for up to 90% of PM₁₀ in the city during winter (Meister et al., 2012). In Berlin, about 45% of the local traffic contributions to roadside PM₁₀ concentrations were due to suspended soil material, the remaining 55% being the result of vehicle exhaust and tire abrasion (Lenschow et al., 2001). Other studies also concluded that in cities, the importance of the non-exhaust emissions is comparable to, or even higher than that of emissions from vehicle exhaust systems (for a detailed literature review, see Amato et al., 2011).

As outlined above, street sediment originates from a variety of sources, among which road works, infrastructural works, building constructions, and vacant lots are the most prominent visible examples. However, degradation of pavements, especially asphalt concrete, is also an important source of street sediment as asphalt concrete usually contains, besides gravel fragments and bitumen, also finer sand and/or silt-sized sediment. A similar less visible source is sand used to fill the space between sidewalk tiles. Sand and dust washed from cars and their tires are other sources of street sediment. Once on the road, all these sediments become polluted by heavy metals originating mainly from car tires, brakes, and engines (Kupiainen et al., 2011). Loose sediment on streets and sidewalks, and in the gutter, is usually substantially enriched in heavy metals.

The sampling and analysis (especially chemically) of street, gutter and sidewalk sediment is not new. Analysis of street sediment has been performed for decades although most studies were carried out after the year 2000. Reviews and lists of studies can be found in, among others, the works by Garnaud (1999), Bris et al. (1999), Tamrakar and Shakya (2011), Shabbaj et al. (2018) and Zglobicki et al. (2018). Detailed samplings of an entire city are very rare however. The sampled cities are usually metropolises, and too large for a detailed sampling (e.g. Paris: Bris et

123 al., 1999; Kathmandu: Tamrakar and Shakya, 2011; Mersin: Arslan and Gizir, 2006; Jeddah:
124 Shabbaj et al., 2018; and many more), or the aim of the study is to compare cities by sampling
125 a limited number of strategic locations (e.g. Amato et al., 2011; Wang et al., 1998; and others).
126 Zhang et al. (2014) performed a sampling of a large part of the city of Xi'an, but their samples
127 included, besides road sediment, also building construction dust, cement samples (from
128 construction sites) and fresh soil sediment collected during excavations at construction sites.
129 Deocampo et al. (2012) sampled two portions of the city of Atlanta, but studied only the
130 distribution of lead; not of other heavy metals.

131

132 This study describes the analysis of street sediment in the city of Leuven, Belgium. The whole
133 historic (medieval) city was sampled in detail. This was possible because its surface area is
134 rather small. The city center forms an almost perfect circle with a diameter of only 2.4 km (Fig.
135 1). Its surface area is only 4.5 km². In addition, the city has many streets because of its medieval
136 origin. There is considerable variation in traffic density: there are pedestrian zones, streets with
137 limited traffic, streets with much traffic, and the ring road forming the border of the city has
138 very intense traffic. Clear differences in pollution of street sediment can therefore be expected
139 over the city because traffic has been determined to be the most important factor causing
140 pollution of street sediment (Robra, 2010).

141

142 A total of 77 locations were sampled, distributed over the whole city. We only sampled the
143 central (medieval) city itself, without the suburbs of Heverlee, Kessel-Lo and Wilsele. This
144 article describes and discusses the results for a large number of heavy metals, reconstructs the
145 origin of the polluting elements, and investigates the relationship between traffic density and
146 the degree of pollution of the road sediment. A health risk analysis was also performed, but
147 these results will be presented in a separate study.

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150 2. Procedure

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153 2.1 Sampling

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156 Leuven is a historic, early medieval city. The road pattern is therefore typical radial-concentric,
157 with an almost perfectly circular ring road following the trajectory of the second medieval city
158 wall (14th century). Several streets in the center also follow the trajectory of the first city wall
159 (12th century). The large number of streets on a limited surface, and the great variation in traffic
160 density, make the town an ideal place for a detailed sampling. Currently there is no more heavy
161 industry in Leuven or its surroundings. Landuse near the city mainly consists, apart from rural
162 land, of private habitation, research buildings and tertiary sector shops. The only industrial
163 complex is a brewery but it does not release heavy metals into the atmosphere. This means that
164 traffic is the major factor causing pollution of road sediment in the city. Renovation works are
165 currently being carried out in several parts of the town, but these do not release heavy metals
166 although they locally result in denser heavy traffic. Leuven has about 30,000 permanent
167 inhabitants in the central town itself plus another 60,000 inhabitants in the suburbs of Heverlee,
168 Kessel-Lo and Wilsele-Dorp. From late September to early July an additional 40,000 students
169 live in the agglomeration, many of them in the medieval city center itself.

170

171 A total of 77 locations were sampled (Fig. 1). When selecting the sampling sites we made sure
172 to cover the whole city center and to include both pedestrian zones, streets with little traffic,

173 streets with much traffic and streets with very busy traffic. We also made sure to include enough
174 (and a comparable amount of) sampling locations for each of these four traffic classes. We
175 avoided sampling large open spaces such as parks and squares because in these areas there is
176 more wind and the local pollution source is less easy to determine, which makes it difficult to
177 investigate a possible correlation between traffic emissions and the pollution of street sediment.
178 . We also opted for sampling the road surface itself and not the gutter or the sidewalk. We
179 avoided sampling the gutter because it serves as a transport channel for water and sediment
180 washed away upstream, sometimes far away from the sampling area. This is especially
181 important in the western part of Leuven, where the city has been built on the western slope of
182 the Dijle valley and where many steep slopes and roads occur. We also opted for not sampling
183 the sidewalk because previous studies have shown that the sediment on sidewalks is often quite
184 coarse (substantially coarser than on the road itself, see for example Artières, 1987) and also
185 because the sidewalk is located farther away from the traffic than the road surface itself,
186 resulting in less resuspension of loose sediment on it compared to the road surface.

187
188 Road sediment was collected using a plastic brush and a plastic dustpan, similar to most road
189 dust studies (Yongming et al., 2006; Shabbaj et al., 2018; Zglobicki et al., 2018). We recognize
190 that this can result in a loss of some fine dust (Amato et al., 2011). However, the goal of this
191 study was not to determine the absolute amount of sediment on the roads, as this can vary
192 greatly over even short distances (see, for example, Deocampo et al., 2012). For optimal
193 sampling, collecting the sediment by dry vacuuming (e.g. Butler et al., 1992; Amato et al.,
194 2011) or wet vacuuming (e.g. Bris et al., 1999) is recommended. We had no permission in this
195 study to use these techniques in the streets of Leuven.

196
197 Sweeping was performed at about 1 m from the sidewalk. This can be considered the area where
198 most car tires are in contact with the street surface and, thus, where most street dust
199 contamination can be expected to occur. Both sides of the road were sampled, each over a length
200 of 10 m. After each sampling the brush and dustpan were carefully cleaned to avoid cross-
201 contamination of the samples. Before sampling, possible street sweep actions by city services
202 were carefully checked on each sampling location because comparing cleaned with uncleaned
203 streets will give an incorrect picture of the spatial distribution of the contamination, and there
204 is also a risk of cross-contamination.

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206 After collection, the sediment was stored in clean, closed plastic bags. Samplings were carried
207 out in the first week of July 2018, after a 3-week long dry spell.

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210 *2.2 Sediment analysis*

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213 *2.2.1 Grain size*

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216 There is inconsistency in the literature with regard to the size of sediment analyzed in street
217 sediment studies. Very different size fractions have been used, varying from very fine ($<10\ \mu\text{m}$,
218 for example Amato et al., 2011) to very coarse ($<2\ \text{mm}$, for example El-Hasan et al., 2006).
219 Because the concentration of heavy metals may substantially vary with particle size (Sutherland
220 et al., 2012) direct comparisons between studies are often very difficult. In this study we opted
221 for analyzing the fraction $<60\ \mu\text{m}$ for several reasons. First, these particles are easily
222 (re)suspended and may remain airborne for long time periods (Shilton et al., 2005; Soltani et

223 al., 2015). Second, heavy metal concentrations generally decrease with increasing particle size
 224 (Fergusson and Ryan, 1984; Sutherland et al., 2012). Third, small particles have been proven
 225 to cause the largest threat to human health (Zhou et al., 2003; Liu et al., 2014). As a consequence
 226 many studies on street sediment focus on the particles smaller than 63 μm (Saeedi et al., 2012;
 227 Shabbaj et al., 2018). Sieving was done in two steps: the collected sediment was first sieved at
 228 125 μm to remove impurities such as leaves, branches, rocks, glass, etc.; then the remaining
 229 sediment was sieved at 60 μm . Plastic sieves were used to avoid any contamination of the
 230 samples.

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2.2.2 Geochemical analysis

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236 For each sample a total of 100 mg was transferred to a glass tube and 1 ml of 65% HNO_3
 237 solution was added, followed by 3 ml of 37% HCl solution. The samples were left to dissolve
 238 for one night. The next day the tubes were heated on a hot plate for 3 hours to ensure all metals
 239 were fully dissolved. The plate was first heated to 90 $^\circ\text{C}$; after 30 min the temperature was
 240 increased to 120 $^\circ\text{C}$, and then the temperature was increased by 20 $^\circ\text{C}$ every 30 min ending with
 241 a 30-min period at 200 $^\circ\text{C}$. The tubes were then left to cool down, after which the solutions
 242 were diluted to 20 ml with ultrapure Milli-Q water. The tubes were then thoroughly shaken to
 243 ensure a homogeneous solution. Samples were then left to allow for settling of the insoluble
 244 particles. Finally 7 ml of the solution was transferred to small plastic flasks using a pipette.
 245 Flasks were stored in a fridge until analysis.

246

247 Inductively coupled plasma-optical emission spectrometry (ICP-OES) was used to determine
 248 the concentration of the following elements: As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, S, Sb, V, and
 249 Zn. The instrument used was a Agilent Varian 720 ES analyzer with a detection limit varying
 250 between 1 and 100 ppb (Agilent, 2012). For quality control and assessment each batch of
 251 samples also contained two reagent blank samples, two reference materials, and at least two
 252 duplicates for each sample. BCR-143R (Sewage sludge amended soil) and QCM-31 standard
 253 material were used as standard reference materials.

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2.3 Quantification of pollution

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259 Several indices have been used in the literature to quantify the pollution of urban street
 260 sediment. The most frequently used ones are the Enrichment Factor (EF), the Geoaccumulation
 261 Index (Igeo), and the Pollution Index (PI). They are also used in the present study.

262

Enrichment Factor (EF)

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265 This factor compares the concentration of a heavy metal in the street sediment sample to the
 266 background value in local unpolluted soil (Yongming et al., 2006; Saeedi et al., 2012). It is
 267 defined as $EF = \frac{C/C_{ref}}{B/B_{ref}}$, where C and C_{ref} are the measured concentrations of the heavy metal

268 and a reference element in the street sample and B and B_{ref} the concentrations of the heavy metal
 269 and the reference element in local unpolluted soil. In this study Mn was used as the reference
 270 element since its concentration in the street sediment was close to the background value in the
 271 local soil. Mn has also been used as a reference element in other studies (Saeedi et al., 2012).

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Geoaccumulation Index (I_{geo})

This index, introduced by Müller (1969), compares heavy metal concentrations in urban soils to background concentrations in local unpolluted soils. It is defined as $I_{geo} = \log_2 (C/1.5B)$, where C is the measured concentration in the street sample and B the background value in the unpolluted soil. The factor 1.5 minimizes the effect of possible variations in the background value (Sutherland, 2000).

Pollution Index (PI)

This index, defined as $PI = C/B$ (Lu et al., 2009), is the ratio of the heavy metal concentration in street sediment (C) to the background value in unpolluted soil (B). To assess the integrated pollution level of all heavy metals present in street sediment the Integrated Pollution Index (IPI) can be used: $IPI = (PI_1 + PI_2 + PI_3 + \dots + PI_n)/n$, where PI_1 is the Pollution Index for heavy metal 1, PI_2 the Pollution Index for heavy metal 2, etc. and n is the number of heavy metals considered.

Table 1 shows the degree of pollution for each of the contamination indices used.

3. Results

3.1 Elemental concentrations

Table 2 gives an overview of the statistics for the 13 chemical elements analyzed in Leuven. Recall that 77 locations were sampled in total. Background values for Flanders (last column on the right) are from Tariku et al. (2005) and Tariku et al. (2006). For Co, Fe, Mn, S, Sb and V no data were provided by these authors; for these elements background values for European soils (Salminen et al., 2006) were used. The table shows that substantially elevated concentrations occur in the street sediment in Leuven for several elements, although comparable concentrations have been measured in other cities (Table 3). Note that to eliminate the effect of particle size, only studies that investigated a similar particle size fraction as in Leuven are shown in Table 3. No data are given in the table for As, S and Sb since many street sediment studies do not consider these elements.

Table 4 shows that, in Leuven, highest enrichment (strongest pollution) was measured for Cu, Sb, Pb and Zn. For Cu and Zn the concentrations in Leuven clearly exceed those in most of the other cities in Table 3. Moderate enrichment was measured for Cd, Cr, Ni and S. The other elements (As, Co, Fe, Mn and V) showed concentrations close to the background value in unpolluted soil. The Integrated Pollution Index (IPI) for Leuven is 8.15, which classifies Leuven, on average over its surface area, as a "very highly polluted" city (Table 1).

3.2 Multivariate analysis

321 Principal Component Analysis (PCA) and Cluster Analysis (CA) were used to group the
322 elements in distinct classes based on their intensity of contamination. Fig. 2 shows the PCA
323 loadings in a 3D-plot, for all 13 elements analyzed in this study. A total of 4 groups can be
324 selected: S, Co, Mn and V (group 1), As and Fe (group 2), Pb, Ni, Zn and Cd (group 3; note
325 that Cd is located somewhat eccentric in this group), and Sb, Cu and Cr (group 4). Cluster
326 analysis based on Ward's (1963) clustering method allows to separate Cd from group 3 so that
327 5 groups were finally retained, as shown in Fig. 3.

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330 *3.3 Source identification*

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333 Heavy metals that belong to a same group (as defined before) can be expected to have a similar
334 source. The lowest degree of contamination was found for S, Co, Mn and V (group 1) and As
335 and Fe (group 2). Analysis of the pollution indices for these elements (Table 4) shows that the
336 concentration of these elements in Leuven street sediment is similar to the concentration in
337 uncontaminated soil, which suggests that their source is natural soil. The elements in groups 3,
338 4 and 5, on the other hand, are clearly enriched, suggesting that these elements can be associated
339 with one or more types of vehicle traffic emissions (Sutherland, 2000; Sternbeck et al., 2002;
340 Duong and Lee, 2011). For Cd (group 5), likely sources are combustion engines and motor oils
341 (Sutherland, 2000). Brake wear is a major pathway for Cu and Sb emissions (Sternbeck et al.,
342 2002; Adachi and Tainosho, 2004; Lijima et al., 2007); therefore group 4 (Cu, Sb and Cr) is
343 likely associated with brake wear. Zn and Pb are often associated with tire wear (Apegyei et
344 al., 2011; Duong and Lee, 2011; Sutherland, 2000); therefore tire wear is a likely source for the
345 enhanced concentration of the elements of group 3 (Zn, Pb and Ni). Pb has also been related to
346 historic contamination caused by leaded gasoline (De Miguel et al., 1997; Charlesworth et al.,
347 2003) but leaded gasoline has already been forbidden in Belgium since the year 2000.

348

349 A schematic overview of these potential sources is shown in Fig. 3. One should realize that
350 pinpointing one particular source for each group is difficult as street sediment can be (and
351 usually is) contaminated by a variety of processes; but the distinction between the natural and
352 anthropogenic sources (groups 1 and 2 versus groups 3, 4 and 5) is obvious from the data
353 collected.

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356 *3.4 Spatial distribution*

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359 The spatial distribution of pollution for the most enriched elements (Cu, Sb, Pb and Zn) is
360 shown in Fig. 4. For Cu and Sb the highest contamination is found on the ring road and the busy
361 traffic circulation loops. This could be expected since Cu and Sb are associated with brake wear.
362 For Zn and Pb, however, the highest contamination occurs in the city center, in the pedestrian
363 zone where no traffic is allowed except for buses and taxis. One would expect that for these
364 elements, which are most likely associated with tire wear, concentrations would be highest
365 where the traffic volume is also highest, such as on the ring road. A possible explanation is that
366 the current pattern still reflects the historic accumulation of these elements at the time traffic
367 was still allowed in the current pedestrian zones. Many streets in these zones are quite narrow
368 and behave like street canyons, hampering the evacuation of pollution. Several of them are also
369 covered with cobblestones and the space between the stones is filled with soil material; it could
370 thus be that these surfaces still contain much more historic contamination than other streets with

371 more (and faster) traffic where the surface material is more likely to be moved or resuspended
372 by traffic movements.

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4. Discussion

378 The spatial pollution pattern for especially Zn and Pb shows that the strongest pollution of urban
379 street sediment does not necessarily occur in the zones with the highest traffic density. The
380 question can therefore be asked whether or not the contamination of urban street can be
381 considered a reliable tool to quantify the current traffic density and reverse. As suggested by
382 Fig. 4, the role of historic pollution should not be neglected and could even become dominant.

383
384 To additionally test the hypothesis that the unexpected preferential concentration of Zn and Pb
385 in the pedestrian area is a result of historic accumulation we compared the concentrations with
386 information on the current traffic density and pollution. For the latter we used data collected by
387 the project CurieuzeNeuzen Vlaanderen (De Craemer et al., 2020; Weichenthal et al., 2020). In
388 that project, airborne NO₂ concentrations were measured at 20,000 locations all over Flanders
389 (the northern part of Belgium). NO₂ is a very good proxy to quantify traffic pollution (Vlaamse
390 Milieumaatschappij, 2017). Road transport is the largest contributor to NO_x (NO and NO₂)
391 pollution, ahead of the energy, commercial, institutional and household sectors (Degraeuwe et
392 al., 2019). The CurieuzeNeuzen field campaign took place from April 28, 2018 to May 26,
393 2018, very close to the period where the Leuven street sediment samples were collected.
394 Technical information on the CurieuzeNeuzen project and the analysis methods used can be
395 found in Meysman et al. (submitted). In the area where our street samples were collected, NO₂
396 was measured at a total of 114 locations. Several of these coincided with places where our street
397 sediment samples were collected. Others did not because of logistic or other reasons. For the
398 latter spots we estimated the NO₂ concentrations based on the data of neighboring measuring
399 points, in the same (where possible) or in comparable adjacent streets, and a careful check of
400 the local traffic intensity during field observations. In 8% of the cases the samplings for street
401 sediment and NO₂ could be performed at exactly the same place, and in 53% of the cases the
402 points were less than 100 m from each other, many of them even less than 50 m. The average
403 distance between a sediment sampling point and the corresponding NO₂ sampling point was
404 116 m. For several neighborhoods we could also use information collected by the projects
405 Straatvinken (<http://www.straatvinken.be>) and Straten Vol Leuven (<http://stratenvolleuven.be>).
406 Straatvinken counts traffic movements in Leuven, and Straten Vol Leuven performed
407 measurements of black carbon emitted by traffic in the city and some of its suburbs.

408
409 If our hypothesis that the unexpected high concentrations of Zn and Pb in the pedestrian area
410 results from historic accumulation is correct, then no correlation should be seen between the Zn
411 and Pb concentrations and the NO₂ concentrations since both are mainly produced by traffic.
412 To make the analysis more complete and check whether the chemical composition of urban
413 street sediment can be considered a good indicator for traffic density at all we extended the
414 analysis to Cu and Sb, which are produced by car brakes, and to Cd, which occurs in car exhaust
415 and in motor oil.

416
417 The correlations are shown in Fig. 5. The IPI values in the graphs are for the combination of
418 the metals in question (Cu and Sb, Zn and Pb, and Cd). For the three types of car pollution we
419 checked the correlations for the entire city center and for the pedestrian and car-allowed area
420 separately. Out of these nine tests, seven did not show any correlation (slope of the regression

421 line not significantly different from zero). The two remaining cases are (1) brake pollution, all
422 areas (significant at the 0.05 level but not at the 0.01 level), and (2) combustion / motor oil
423 pollution in the pedestrian area (significant at the 0.01 level). The latter relationship may look
424 a little strange since we are talking of the pedestrian area, but recall that in Leuven buses and
425 taxis are still allowed in this zone. We conclude that the preferential concentration of Zn and
426 Pb in the pedestrian zone of the city of Leuven is not related to the current traffic, and because
427 there are no other important sources for these two elements (there are, and have been, no Zn or
428 Pb related industries or manufacturing activities in what is now the pedestrian zone in Leuven)
429 the preferential concentration in the pedestrian zone can only be the result of historic
430 accumulation of these elements at times where cars were still allowed in this zone.

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432

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5. Conclusions

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436 The results show that, at least for the city of Leuven, both the spatial and the quantitative
437 pollution patterns of street sediment generally did not match with the pattern of NO₂
438 concentration. Since it is well known that in sediments the concentrations of chemical elements
439 are the highest in the finest particle fractions and our study considered the fraction <60 μm,
440 which is the finest fraction in street sediment, we consider it likely that our conclusions will
441 also apply to other cities worldwide although explicit confirmation by future studies will be
442 necessary.

443

444 In Leuven the highest enrichment (strongest pollution) was measured for Cu, Sb, Pb and Zn.
445 For Cu and Zn the concentrations in Leuven exceed those in most of the other cities investigated
446 in the literature. Moderate enrichment was measured for Cd, Cr, Ni and S. The other elements
447 (As, Co, Fe, Mn and V) showed concentrations close to the background value in unpolluted
448 soil. The Integrated Pollution Index (IPI) for Leuven is 8.15, which, according to the criteria
449 proposed by Wan et al. (2016a) and Chen et al. (2005), classifies Leuven, on average over its
450 surface area, as a "very highly polluted" city.

451

452 For Cu and Sb the highest contamination is found on the ring road and the busy traffic
453 circulation loops. For Zn and Pb the highest contamination occurs in the city center, in the
454 pedestrian zone where no traffic is allowed except for buses and taxis. We hypothesize that this
455 is a result of historic accumulation of these elements at the time traffic was still allowed in this
456 zone. Many streets in this zone are quite narrow and behave like street canyons, hampering the
457 evacuation of pollution. Several of them are also covered with cobblestones and the space
458 between the stones is filled with soil material; it could thus be that these surfaces still contain
459 much more historic contamination than other streets with more (and faster) traffic where the
460 surface material is more likely to be moved or resuspended by traffic movements.

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462 This study shows that measurements of current pollution by traffic are not sufficient to
463 determine the health risk because much exposure to toxic substances may be caused by
464 resuspension, by traffic or by wind, of substances that have accumulated in the city over time,
465 sometimes decades ago, when regulations were much less stringent than today. Adequate
466 environmental management should consider such aspects.

467

468

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479 Supervision

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481 **References**

- 482
- 483
- 484 Adachi, K., Tainosho, Y., 2004. Characterization of heavy metal particles embedded in tire
485 dust. *Environment International* 30, 1009-1017.
- 486
- 487 Agilent, 2012. Agilent 720/725 ICP-OES. Agilent Technologies, Santa Clara, CA, USA.
- 488
- 489 Al-Awadhi, J.M., Aldhafiri, B.T., 2016. Heavy metal concentrations in roadside-deposited
490 sediments in Kuwait city. *Arabian Journal of Geosciences* 9, 535.
- 491
- 492 Amato, F., Pandolfi, M., Moreno, T., Furger, M., Pey, J., Alastuey, A., Bukowiecki, N.,
493 Prevot, A.S.H., Baltensperger, U., Querol, X., 2011. Sources and variability of inhalable road
494 dust particles in three European cities. *Atmospheric Environment* 45, 6777-6787.
- 495
- 496 Apeagyei, E., Bank, M.S., Spengler, J.D., 2011. Distribution of heavy metals in road dust
497 along an urban-rural gradient in Massachusetts. *Atmospheric Environment* 45, 2310-2323.
- 498
- 499 Arslan, H., Gizir, A., 2006. Heavy metal content of roadside soil in Mersin, Turkey. *Fresenius*
500 *Environmental Bulletin* 15, 15-20.
- 501
- 502 Artières, O., 1987. Les dépôts en réseau d'assainissement unitaire: origine, caractéristiques,
503 pollution, transport. Ph.D. Thesis Université Louis Pasteur Strasbourg, 209 pp.
- 504
- 505 Bris, F.-J., Garnaud, S., Apperry, N., Gonzalez, A., Mouchel, J.-M., Chebbo, G., Thévenot,
506 D.R., 1999. A street deposit sampling method for metal and hydrocarbon contamination
507 assessment. *Science of the Total Environment* 235, 211-220.
- 508
- 509 Butler, D., Thedchanamoorthy, S., Payne, J.A., 1992. Aspects of surface sediment
510 characteristics on an urban catchment in London. *Water Sci Technol* 25, 13-19.
- 511
- 512 Charlesworth, S., Everett, M., McCarthy, R., Ordonez, A., De Miguel, E., 2003. A
513 comparative study of heavy metal concentration and distribution in deposited street dusts in a
514 large and a small urban area: Birmingham and Coventry, West Midlands, UK. *Environment*
515 *International* 29, 563-573.
- 516
- 517 Chen, T.B., Zheng, Y.M., Lei, M., Huang, Z.C., Wu, H.T., Chen, H., Fan, K.K., Yu, K., Wu,
518 X., Tian, Q.Z., 2005. Assessment of heavy metal pollution in surface soils of urban parks in
519 Beijing, China. *Chemosphere* 60, 542-551.
- 520
- 521 Christoforidis, A., Stamatis, N., 2009. Heavy metal contamination in street dust and roadside
522 soil along the major national road in Kavala's region, Greece. *Geoderma* 151, 257-263.
- 523
- 524 De Craemer, S., Vercauteren, J., Fierens, F., Lefebvre, W., Meysman, F.J.R., 2020. Using
525 large-scale NO₂ data from citizen science for air-quality compliance and policy support. *Env.*
526 *Sci. Technol.* 54, 11070-11078.
- 527
- 528 De Miguel, E., Llamas, J.F., Chacón, E., Berg, T., Larssen, S., Røyset, O., Vadset, M., 1997.
529 Origin and patterns of distribution of trace elements in street dust: unleaded petrol and urban
530 lead. *Atmospheric Environment* 31, 2733-2740.

- 531
532 Degraeuwe, B., Pisoni, E., Peduzzi, E., De Meij, A., Monforti-Ferrario, F., Bodis, K.,
533 Mascherpa, A., Astorga-Llorens, M., Thunis, P., Vignati, E., 2019. Urban NO₂ Atlas. EUR
534 29943 EN, Publications Office of the European Union, Luxembourg.
- 535
536 Deocampo, D.M., Reed, J., Kalenuik, A.P., 2012. Road dust lead (Pb) in two neighborhoods
537 of urban Atlanta, (GA, USA). *Int. J. Environ. Res. Public Health* 9, 2020-2030.
- 538
539 Duong, T.T., Lee, B.K., 2011. Determining contamination level of heavy metals in road dust
540 from busy traffic areas with different characteristics. *Journal of Environmental Management*
541 92, 554-562.
- 542
543 El-Hasan, T., Batarseh, M., Al-Omari, H., Ziadat, A., El-Alafi, A., Al-Naser, F., Berdanier,
544 B.W., Jiries, A., 2006. The distribution of heavy metals in urban street dusts of Karak City,
545 Jordan. *Soil & Sediment Contamination* 15, 357–365.
- 546
547 Fergusson, J., Ryan, D., 1984. The elemental composition of street dust from large and small
548 urban areas related to city type, source and particle size. *Science of the Total Environment* 34,
549 101-116.
- 550
551 Ferreira-Baptista, L., De Miguel, E., 2005. Geochemistry and risk assessment of street dust in
552 Luanda, Angola: A tropical urban environment. *Atmospheric Environment* 39, 4501-4512.
- 553
554 Garnaud, S., 1999. Transfer et evolution géochimique de la pollution métallique en bassin
555 versant urbain. Thesis, Ecole Nationale de Ponts et Chaussées, Paris Institute of Technology.
- 556
557 Gidhagen, L., Johansson, C., Olivares, G., 2004. Simulation of NO_x and ultrafine particles in
558 a street canyon in Stockholm, Sweden. *Atm. Env.* 38, 2029-2044.
- 559
560 Kupiainen, K., Pirjola, L., Ritola, R., Väkevä, O., Viinanen, J., Stojiljkovic, A., Malinen, A.,
561 2011. Street dust emissions in Finnish cities – summary of results from 2006–2010. *City of
562 Helsinki Environment Centre* 5/2011.
- 563
564 Lenschow, P., Abraham, H-J., Kutzner, K., Lutz, M., Preuss, J-D., Reichenbacher, W., 2001.
565 Some ideas about the sources of PM₁₀. *Atm. Env.* 35, Suppl. 1, S23-S33.
- 566
567 Lijima, A., Sato, K., Yano, K., Tago, H., Kato, M., Kimura, H., Furuta, N., 2007. Particle size
568 and composition distribution analysis of automotive brake abrasion dusts for the evaluation of
569 antimony sources of airborne particulate matter. *Atmospheric Environment* 41, 4908-4919.
- 570
571 Liu, E., Yan, T., Birch, G., Zhu, Y., 2014. Pollution and health risk of potentially toxic metals
572 in urban road dust in Nanjing, a mega-city of China. *Science of the Total Environment* 476,
573 522-531.
- 574
575 Lu, X., Wang, L., Lei, K., Huang, J., Zhai, Y., 2009. Contamination assessment of copper,
576 lead, zinc, manganese and nickel in street dust of Baoji, NW China. *Journal of Hazardous
577 Materials* 161, 1058-1062.
- 578
579 Meister, K., Johansson, C., Forsberg, B., 2012. Estimated short-term effects of coarse
580 particles on daily mortality in Stockholm, Sweden. *Environm. Health Perspect.* 120, 431-436.

- 581
582 Meysman, F.J.R., De Craemer, S., Lefebvre, W., Vercauteren, J., Sluydts, V., Dons, E.,
583 Hooyberghs, H., Van den Bossche, J., Trimpeneers, E., Fierens, F., Huyse, H. (submitted).
584 Citizen science reveals the population exposure to air pollution. Preprint doi:
585 10.31223/osf.io/bryje.
586
- 587 Müller, G., 1969. Index of geoaccumulation in sediments of the Rhine River. *Geology Journal*
588 2, 109–118.
589
- 590 Pye, K., Tsoar, H., 1990. *Aeolian Sand and Sand Dunes*. Unwin Hyman, London.
591
- 592 Robra, J.P., 2010. An emissions inventory of air pollutants for the city of Bogotá, Colombia.
593 Thesis, Swiss Federal Institute of Technology Lausanne.
594
- 595 Saeedi, M., Li, L.Y., Salmanzadeh, M., 2012. Heavy metals and polycyclic aromatic
596 hydrocarbons: Pollution and ecological risk assessment in street dust of Tehran. *Journal of*
597 *Hazardous Materials* 227-228, 9-17.
598
- 599 Salminen, R., De Vos, W., Tarvainen, T., 2006. *Geochemical Atlas of Europe*. Geological
600 Survey of Finland.
601
- 602 Shabbaj, I., Alghamdi, M., Shamy, M., Hassan, S., Alsharif, M., Khoder, M., 2018. Risk
603 assessment and implication of human exposure to road dust heavy metals in Jeddah, Saudi
604 Arabia. *International Journal of Environmental Research and Public Health* 15, 36.
605
- 606 Shilton, V.F., Booth, C.A., Smith, J.P., Giess, P., Mitchell, D.J., Williams, C.D., 2005.
607 Magnetic properties of urban street dust and their relationship with organic matter content in
608 the West Midlands, UK. *Atmospheric Environment* 39, 3651-3659.
609
- 610 Soltani, N., Keshavarzi, B., Moore, F., Tavakol, T., Lahijanzadeh, A.R., Jaafarzadeh, N.,
611 Kermani, M., 2015. Ecological and human health hazards of heavy metals and polycyclic
612 aromatic hydrocarbons (PAHs) in road dust of Isfahan metropolis, Iran. *Science of the Total*
613 *Environment* 505, 712-723.
614
- 615 Sternbeck, J., Sjodin, A., Andréasson, K., 2002. Metal emissions from road traffic and the
616 influence of resuspension - results from two tunnel studies. *Atmospheric Environment* 36,
617 4735-4744.
618
- 619 Sutherland, R.A., 2000. A comparison of geochemical information obtained from two fluvial
620 bed sediment fractions. *Environmental Geology* 39, 330-341.
621
- 622 Sutherland, R.A., Tack, F.M.G., Ziegler, A.D., 2012. Road-deposited sediments in an urban
623 environment: A first look at sequentially extracted element loads in grain size fractions.
624 *Journal of Hazardous Materials* 225-226, 54- 62.
625
- 626 Tamrakar, C.S., Shakya, P.R., 2011. Assessment of heavy metals in street dust in Kathmandu
627 Metropolitan City and their possible impacts on the environment. *Pak. J. Anal. Environ.*
628 *Chem.* 12, 32-41.
629

- 630 Tariku, M., Van Meirvenne, M., Tack, F., 2005. Geostatistische analyse en kartering van
631 cadmium, zink, lood en arseen in de bodems van Vlaanderen. Technical report OVAM,
632 Mechelen.
633
- 634 Tariku, M., Van Meirvenne, M., Tack, F., 2006. Geostatistische analyse en kartering van
635 chroom, koper, kwik en nikkel in de bodems van Vlaanderen. Technical report OVAM,
636 Mechelen.
637
- 638 Vlaamse Milieumaatschappij, 2017. Luchtkwaliteit in het Vlaamse Gewest (Air Quality in the
639 Flemish Region). Jaarverslag Immissiemeetresultaten – 2016. Vlaamse Milieumaatschappij,
640 Aalst.
641
- 642 Wan, D., Han, Z., Yang, J., Yang, G., Liu, X., 2016a. Heavy metal pollution in settled dust
643 associated with different urban functional areas in a heavily air-polluted city in North China.
644 *International Journal of Environmental Research and Public Health* 13, 1119.
645
- 646 Wan, D., Zhan, C., Yang, G., Liu, X., Yang, J., 2016b. Preliminary assessment of health risks
647 of potentially toxic elements in settled dust over Beijing urban area. *International Journal of*
648 *Environmental Research and Public Health* 13, 491.
649
- 650 Wang, W.H., Wong, M.H., Leharne, S., Fisher, B., 1998. Fractionation and biotoxicity of
651 heavy metals in urban dusts collected from Hong Kong and London. *Environmental*
652 *Geochemistry and Health* 20, 185-198.
653
- 654 Ward, J.H., Jr., 1963. Hierarchical grouping to optimize an objective function. *Journal of the*
655 *American Statistical Association* 58, 236–244.
656
- 657 Weichenthal, S., Dons, E., Hong, K.Y., Pinheiro, P.O., Meysman, F.J.R. (2020). Combining
658 citizen science and deep learning for large-scale estimation of outdoor nitrogen dioxide
659 concentrations. *Environmental Research*, <https://doi.org/10.1016/j.envres.2020.110389>.
660
- 661 Yongming, H., Peixuan, D., Junji, C., Posmentier, R., 2006. Multivariate analysis of heavy
662 metal contamination in urban dusts of Xi'an, Central China. *Science of the Total Environment*
663 355, 176-186.
664
- 665 Zglobicki, W., Telecka, M., Skupinski, S., Pasierbinska, A., Koziel, M., 2018. Assessment of
666 heavy metal contamination levels of street dust in the city of Lublin, E. Poland.
667 *Environmental Earth Sciences* 77, 774.
668
- 669 Zhang, Q., Shen, Z., Cao, J., Ho, K., Zhang, R., Bie, Z., Chang, H., Liu, S., 2014. Chemical
670 profiles of urban fugitive dust over Xi'an in the south margin of the Loess Plateau, China.
671 *Atmospheric Pollution Research* 5, 421–430.
672
- 673 Zhou, Y., Levy, J.I., Hammit, J.K., Evans, J.S., 2003. Estimating population exposure to
674 power plant emissions using CALPUFF: a case study in Beijing, China. *Atmospheric*
675 *Environment* 37, 815-826.
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678 **Figure captions**

679

680

681 Fig. 1: City of Leuven with location of the 77 sampling sites. The suburbs of Heverlee,
682 Kessel-Lo and Wilsele-Dorp were not sampled and are therefore not shown.

683

684 Fig. 2: 3D plot of PCA loadings, for the 13 chemical elements investigated

685

686 Fig. 3: Cluster dendrogram using Ward's (1963) hierarchical clustering method, for the 13
687 chemical elements investigated. The 5 identified groups are indicated by the colored boxes;
688 the most likely source for each group appears below each box. Height in the ordinate is a
689 measure for the dissimilarity between clusters, calculated by minimizing the within-cluster
690 error sum of squares.

691

692 Fig. 4: Spatial distribution of the enrichment factor for Cu, Sb, Zn and Pb

693

694 Fig. 5: Relationship between airborne NO₂ concentration and the concentration of the
695 analyzed chemical elements that can be related to traffic. NO₂ data are from the
696 CurieuzeNeuzen Vlaanderen project, Universiteit Antwerpen.

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698

699 **Table captions**

700

701

702 Table 1: Contamination indices used in this study

703

704 Table 2: Elemental concentrations (mg/kg) in the street sediment in Leuven

705

706 Table 3: Average elemental concentrations (mg/kg) in street sediment of different cities
707 around the world. Only studies that investigated a similar particle size (PS) to that analyzed in
708 Leuven are shown. Numbers in *italic* are the standard deviations for the Leuven data.

709

710 Table 4: Mean values for Leuven of the contamination indices used in this study. Numbers in
711 *italic* are the standard deviations.

712