



Metals concentrations in road dust from high traffic and low traffic area: a size dependent comparison

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Abstract

Metals in road dust are increasingly investigated. The fine size fractions are of particular importance because of their higher mobility and the increased concentrations of pollutants. In this study, two average road dust samples from areas with high- and low-traffic density were investigated. Prior to chemical analysis, a combined sieving and air classification procedure was applied where the road dust samples were split into seven size fractions. The metal concentrations in the road dust from the low-traffic area were only up to twice the background concentrations. An exception was Cu where the concentration was 15 time higher. In the sample from the high-traffic area, the respective concentrations of Zn, Mn, Fe, Pb, Ba, Sb and Cu were 5.4, 3.2, 3.0, 2.7, 2.7, 2.5 and 1.2 times higher compared to the low-traffic area. The ratio of the concentration in high- and low-traffic road dust was 0.8–2.0 for the fine size fractions and 2.0–6.0 for the coarse size fractions for several metals, including traffic-related metals like Fe and Ba. An explanation for this surprising result might be the atmospheric transport of re-suspended particles from high-traffic to low-traffic areas, which is feasible only for smaller particles because of their low sedimentation velocity. However, for the highly traffic-related metals Pb and Zn, the ratio was more size independent. For Cu, the concentration ratio was fluctuating. Some particles with a high Cu content in the analysed samples might be the reason.

Keywords Road dust · Metals · Particle size · Traffic density

Introduction

In the years since 2000, the composition of road dust has been studied intensively. Thereby, the focus of these studies has been mainly on the metal content (Adamiec et al. 2016; Aryal et al. 2017; Kamani et al. 2017; Aminiyan et al. 2018). Besides the local soil and long-range transport, the emissions from industry as well as traffic emissions contribute to the composition (Žibret et al. 2013). Thus, road dust can be seen as general indicator for environmental pollution in an area. In several studies, the important contribution of traffic emissions to the metal content of road dust has been demonstrated (Fujiwara et al. 2011; Budai

and Clement 2018). Traffic-related particulate matter (PM) emissions originate from vehicle tailpipe exhausts as well as from wear of brake pads and discs, tires and the road surface (Loganathan et al. 2013). A recent study performed in Krakow (Bogacki et al. 2018) demonstrated the importance of the re-entrained road dust on PM10, which can be responsible for up to 25% of the total PM10 concentration near the roads during winter and even up to 50% in summer.

The size of the road dust particles is crucial for their fate in the environment, especially for re-suspension and atmospheric transport. Therefore, the composition of road dust in dependence on the particle size has been studied (Fujiwara et al. 2011; Adamiec et al. 2016). A distinct size dependence of the concentration of several metals was found in these studies. Typically, the concentrations were higher in fine particles, which can be attributed to the increased surface area of fine particles (Loganathan et al. 2013). Dry sieving was used in these studies for the separation of the road dust into various size fractions. However, dry sieving is limited with respect to the finest particle size. Thus, the finest size fraction of road dust in these studies was typically < 40 µm or larger. Recently, a wet sedimentation procedure (Padoan et al. 2017) and air classification (Lanzerstorfer

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2018) were used to separate also very fine size fractions with a median diameter of a few micrometre from road dust samples.

In this study, the distribution of metals in various size fractions of road dust collected from a busy road and from a low-traffic downtown area in Krakow, Poland, was investigated. To extend, the investigation into the very fine size fractions most relevant with respect to re-suspension into the atmosphere, classification with a rotating wheel air classifier was applied for size separation in addition to sieving.

Materials and methods

Sample collection

The road dust samples were collected on April 11th and 12th, 2017, in different parts of Krakow, Poland. The samples were taken from the discharge of vacuum street sweeping vehicles. By this collection method, an average road dust sample for a large street area is obtained whereby local fluctuations in the composition are eliminated. The first sample was collected downtown in the area of the main square of the Old Town of Krakow. There the street sweeping vehicle was cleaning the narrow, short streets surrounding the main square, which are mostly a pedestrian zone. The sampling area is indicated in Fig. 1. This sample representing road dust with little traffic was labelled RDS LT. The second sample was collected in the Azory district of Krakow. There the street sweeping vehicle was cleaning Joseph Conrad Street (European route E77, approximately at km 664; Road no. 7 in Fig. 1) and a short section of Wojciech Weiss Street. This sample representing high-traffic density was labelled RDS HT. The average traffic data for 2015 are available for the close-by E77 section KRAKÓW-RZAŚKA

(km 667.9–669.7) (Generalna Dyrekcja Dróg Krajowych i Autostrad 2015). For the categories motorbikes, cars and mini buses, light trucks, heavy trucks, trucks with a trailer and buses, the respective numbers were 157 per day, 35,983 per day, 2436 per day, 806 per day, 1291 per day and 610 per day.

A volume of approximately 10 dm³ road dust was collected for each site by taking several smaller amounts from various points of the dust heap discharged from the street sweeping vehicle during unloading. In the laboratory, the material was dried at 110 °C for 24 h.

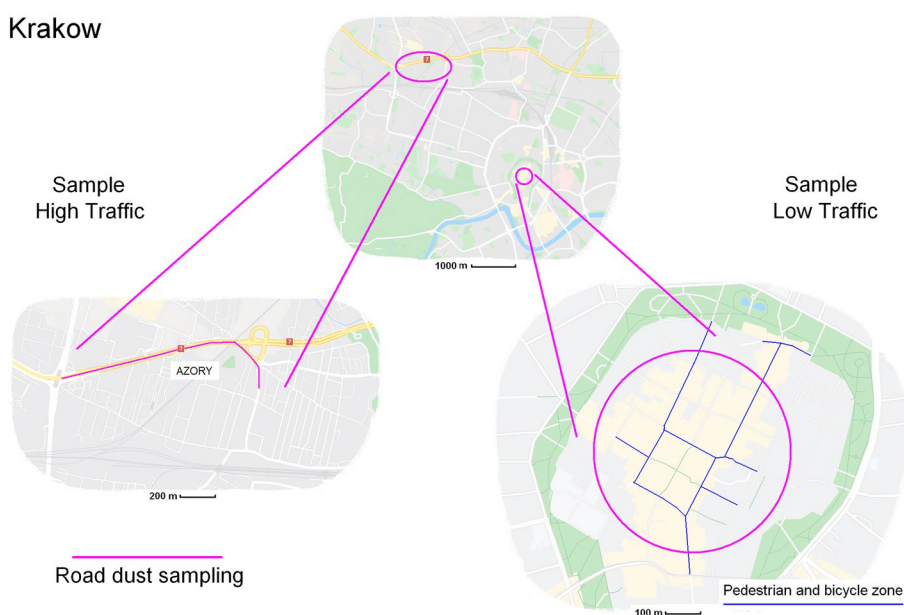
Classification and chemical analysis

In a combined sieving and air classification procedure, the road dust samples < 2 mm were classified into seven size fractions (F1–F7), while the fraction > 2 mm was discarded. Details of such a classification procedure can be found elsewhere (Lanzerstorfer 2015a; Lanzerstorfer and Logiewa 2019). The mass median diameter of the size fractions was determined with a Sympatec HELOS/RODOS laser diffraction instrument with dry sample dispersion. A Sympatec SiC-P600⁰⁶ standard was used to check the calibration of the instrument.

For chemical analysis, dust samples were dissolved by a standard aqua regia digestion. Concentrations of the metals Al, As, Ba, Co, Cu, Fe, Mn, Pb, Sb, Sr, V and Zn were measured by inductively coupled plasma atomic emission spectroscopy (ICP AES) with a Thermo Scientific iCAP7200 Duo system. External calibration was performed using MERCK CertiPur[®] ICP multi-element standards.

In the classified dust, the concentrations of chromium and nickel were increased because of erosion of some stainless steel material (classifier wheel), which makes evaluation of these metals impossible (Lanzerstorfer 2015b).

Fig. 1 Location of road dust sampling areas



Total carbon content (TC) was analysed with a liquiTOC system with a manual solid material injector from Elementar Analysensysteme. The external calibration used a soil standard with 4.1% TOC/TC from Elementar Analysensysteme. In the results, the average values ($n=2$) are presented.

For the whole road dust sample <2.0 mm, the concentration of a component can be calculated according Lanzerstorfer and Logiewa (2019).

The method validation for ICP AES and TC is based on the norms ISO 17025, ISO 5275-1, ISO guide 32 and 33. The statistic values linearity ($n=2*3$), accuracy (trueness and precision $n=3*3$), LOD (level of determination) and LOQ (level of quantification) from mean method standard deviation ($n=3*3$) were determined. Table 1 shows these method validation parameters for the ICP AES analysis.

Table 1 ICP AES method parameters for elemental distribution determination: optical emission wavelength, mean relative standard deviation (RSD), level of detection (LOD) and level of quantification (LOQ) for screened elements

Element	Wavelength in nm	Mean RSD in %	LOD in mg/kg	LOQ in mg/kg
Al	396.152	4.6	1.7	5.0
As	189.042	4.7	1.7	5.0
Ba	455.403	2.3	0.3	1.0
Co	228.616	3.8	0.2	0.5
Cu	324.754	4.5	1.7	5.0
Fe	238.204	2.9	1.7	5.0
Mn	257.610	3.8	0.2	0.5
Pb	220.353	4.8	0.8	2.5
Sb	206.833	3.5	0.3	1.0
Sr	407.771	3.0	0.2	0.5
V	292.402	3.5	0.8	2.5
Zn	213.856	0.9	0.2	0.5

Table 2 Size fractions produced by classification

Size fraction	F1	F2	F3	F4	F5	F6	F7
<i>RDS low traffic</i>							
Mass fraction in %	1.0	1.7	2.0	5.6	43.1	28.2	18.5
Mass mean diameter in μm	2.0	4.5	11	25	120	–	–
Geometric mean diameter ^a in μm	–	–	–	–	–	710	1400
Maximum size in μm	4.5 ^b	8.5 ^b	21 ^b	50 ^b	500	1000	2000
<i>RDS high traffic</i>							
Mass fraction in %	0.4	1.6	6.1	13.3	47.5	20.6	10.6
Mass mean diameter in μm	1.9	4.3	8.8	26	94	–	–
Geometric mean diameter ^a in μm	–	–	–	–	–	710	1400
Maximum size in μm	4.0 ^b	9.0 ^b	17 ^b	45 ^b	500	1000	2000

^aCalculated from the upper and lower sieve size

^bApproximated by the x_{90} of the size distribution

The various elements have different quantitative and linear ranges.

Results and discussion

Classification

The size characteristics and the mass fractions of the size fractions F1–F7 are summarized in Table 2. The size distribution of the RDS LT sample was coarser. In particular, the two coarse size fractions F6 and F7 had a larger share. The reason for the lower share of coarse size fractions in RDS HT could be the more effective grinding of road dust particles by tires in high-traffic areas. For the finest size fraction F1, a higher share was found in the RDS LT. Such fine particles are usually not produced mechanically by grinding but by formation from the gas phase. The higher share might be explained by less re-suspension of deposited particles in the low-traffic area.

In both samples, the most common size fraction with almost 50% was F5. The finer particle size fractions were present only in small proportions. The sum of F1–F4 which represents the road dust smaller than approximately 50 μm was in the range of 10–20%. Similar results were reported by Bian and Zhu (2009) and Bourliva et al. (2017). In these studies, the finest sieving fractions of <54 μm and <63 μm had a proportion in the range of 5–14% and 8–25%, respectively.

Road dust composition

In Table 3, the calculated concentrations of various elements for the total road dust samples <2.0 mm are shown. Generally, the contamination of the RDS LT sample with metals was fairly low in comparison with the concentrations of the local

soil (Lis and Pasieczna 1995). The metal concentrations were up to approximately two times the background concentration with the exception of Cu, which was increased by a factor of approximately 15. In the RDS HT sample, the measured concentrations were significant. The only exception from this was found for TC. For most metals, the concentration in the RDS HT sample was two to three times higher compared to the RDS LT sample. For Zn, this ratio was highest (5.3) while for Cu it was lower (1.2).

In the literature, the concentration ratio of copper to antimony is used as an indicator for brake dust (Thorpe and Harrison 2008). The typical ratio for brake-related particles is around 5:1, while the crustal ratio is around 125:1. In this study, the ratio was approximately 27 for RDS LT and 12 for RDS HT, which confirms the higher influence of brake dust on the composition of the RDS HT.

Composition of road dust size fractions

The composition of F1–F7 is depicted in Fig. 2. The left side shows the concentrations in the RSD LT sample, and the right side shows the concentrations for RDS HT. All elements except TC were present at higher concentrations in respective size fractions of the RDS HT sample. Generally, the concentrations were higher in the fine size fractions. Increased concentrations were found especially in the three finest size fractions F1–F3, which represent the road dust smaller than approximately 20 μm . The concentration minimum was found in size fraction F5 for most elements.

Figure 3 shows the concentration of Ba, Cu, Mn, Pb, Sb and Zn as a function of the particle size in comparison with the available literature data for Beijing (CN), Katowice (PL), Turin (IT) and Wels (AT) (Zhao et al. 2011; Adamiec et al. 2016; Padoan et al. 2017; Lanzerstorfer 2018). For the two coarse

size fractions produced by sieving, the geometric mean of the upper and lower sieve size was used as the representative particle size since no mass mean diameter for these size fractions was available. The same was done with the size limits of the literature data. For the finest size fractions starting at zero, the arithmetic mean was used.

Although the concentrations of the metals in the various road dust samples differed substantially, similarities with respect to the size dependence of the concentrations are evident.

The concentration of Cu shows a tendency to lower values for coarser road dust fractions and quite high fluctuations in all studies with the exception of the study of Padoan et al. (2017). However, a clear common trend of the size dependence obtained in the different studies is not visible. Traffic-related sources for Cu in road dust are brake wear, tire tread wear and exhaust emissions (Loganathan et al. 2013). These sources are especially relevant for the fine size fractions (Gietl et al. 2010). Since the fluctuations of the Cu concentration concern especially the coarser size fractions, pieces of litz wire or other particles with a high Cu content might be a possible reason. When a particle consists of nearly 100% Cu, even a small single piece would have a high impact on the measured Cu concentration.

The Zn concentration decreases with increasing particle size. This size dependence is similar in all studies though the concentration levels were quite different. Only few data points of the sample RDS LT from this study showed a deviation from this trend. The generally higher Zn concentrations in the study by Adamiec et al. (2016) might reflect additional emissions from industry. Currently four steel mills are located in Katowice which process mainly scrap in contrast to the integrated steel mill in Krakow which processes iron ore. Thus, dust emissions from these steel mills are likely to have an increased Zn content (Yatkin and Bayram 2008).

Table 3 Calculated concentrations for the whole RDS samples <2.0 mm and geochemical background of Krakow

Element	Unit	RDS LT <2.0 mm	RDS HT <2.0 mm	RDS HT/ RDS LT	Geochemical background of Krakow (Lis and Pasieczna 1995)
TC	g/kg	24.3	15.3	0.63	–
Al	g/kg	3.0	6.1	2.0	4.1
As	mg/kg	8.8	20	2.3	5
Ba	mg/kg	28	75	2.7	50
Co	mg/kg	3.2	10	3.1	3.4
Cu	mg/kg	166	192	1.2	11
Fe	g/kg	7.9	24	3.0	7.7
Mn	mg/kg	174	564	3.2	257
Pb	mg/kg	19	51	2.7	23
Sb	mg/kg	6.5	16	2.5	–
Sr	mg/kg	25	69	2.8	13
V	mg/kg	11	37	3.4	10
Zn	mg/kg	80	428	5.4	73

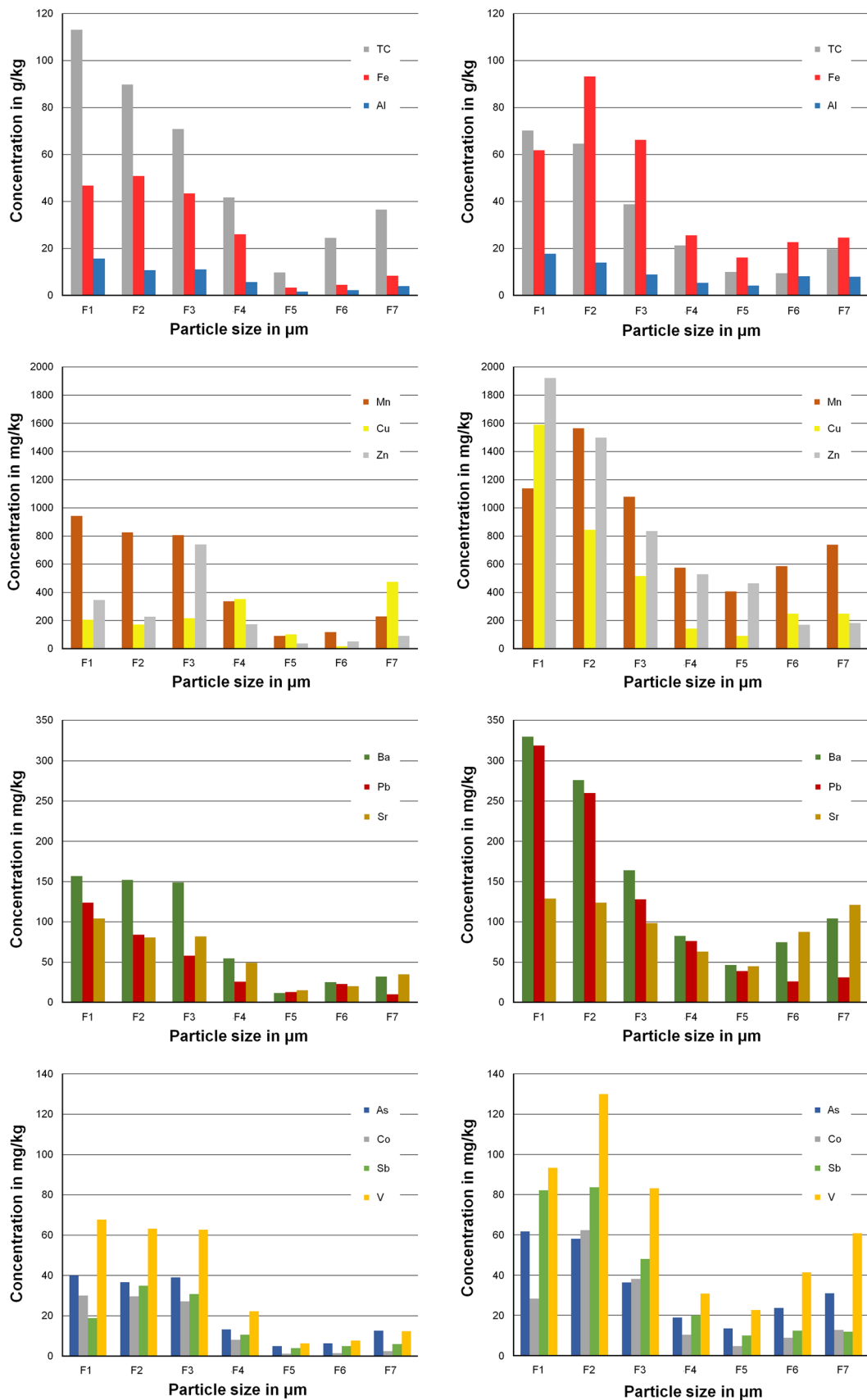


Fig. 2 Concentration of the elements in the size fractions F1–F7 (left side: RDS LT; right side: RDS HT)

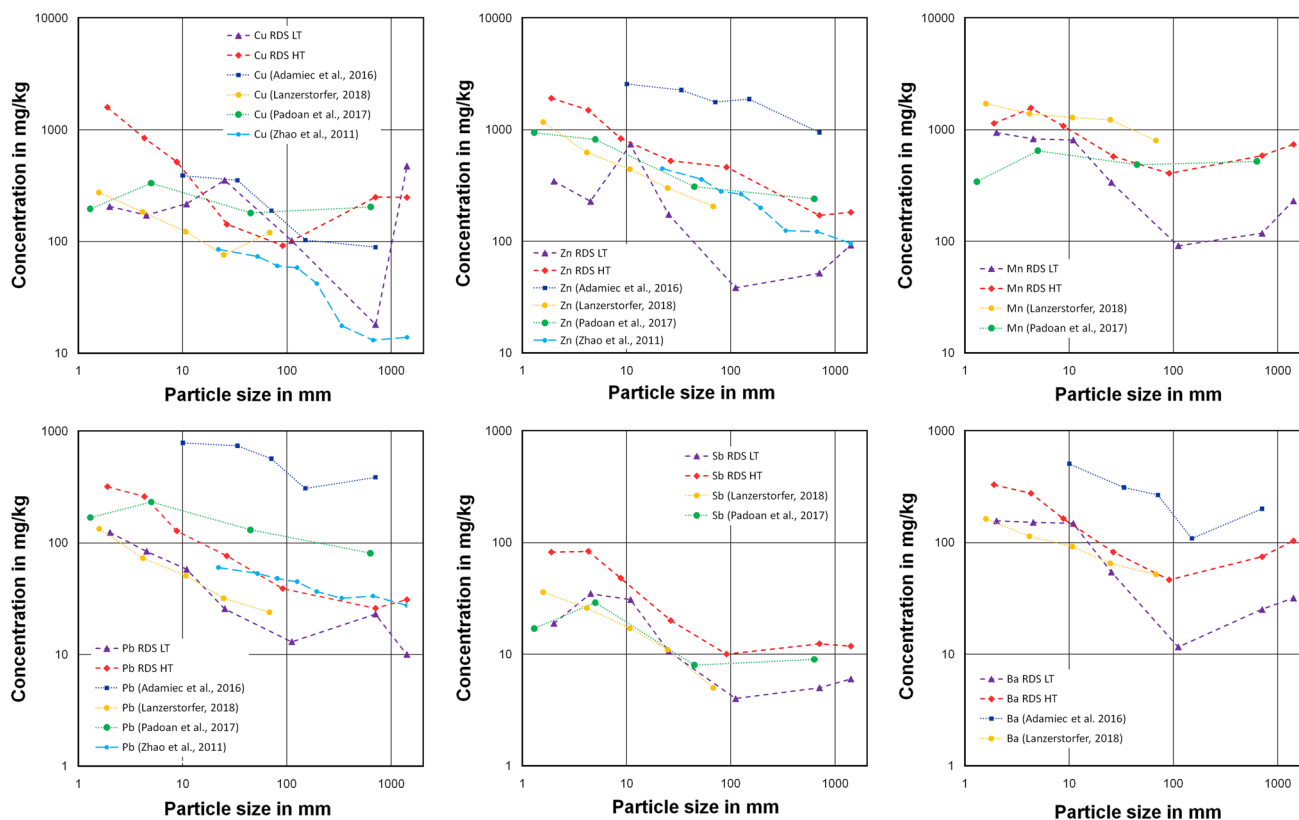


Fig. 3 Concentration of various metals as a function of the particle size

Generally, the concentration differences between the various studies were smaller for Mn than for Cu and Zn. A slight size dependence of the Mn concentration is visible. In all studies, the concentration decreases with the particle size. Substantially, lower Mn concentrations in the coarse size fraction were found only in the sample RDS LT of this study.

The dependence of the Pb concentration on the particle size was very similar in all studies, whereas the concentrations differed substantially. The size dependence of the concentration was quite similar to those of the Zn concentration. The higher Pb concentration of the road dust sample from Katowice might also reflect the industrial background of Katowice with four steel mills in operation (Yatkin and Bayram 2008). After the phase-out of leaded gasoline, Pb is still present at trace levels as a natural component in unleaded automotive fuels (Harris and Davidson 2005). Additionally, vehicular Pb emissions are caused by loss of Pb wheel weights (Hwang et al. 2016). This may be reflected in the increased Pb concentration of some of the coarse size fractions.

The Sb concentration shows the smallest differences between the various studies. The size dependence of the Sb concentration is very similar in all studies. The Sb concentration in the finest size fractions $< 2.5 \mu\text{m}$ is typically lower than in the $5 \mu\text{m}$ size fraction. This corresponds to the size distribution of brake wear dust, which has the maximum in

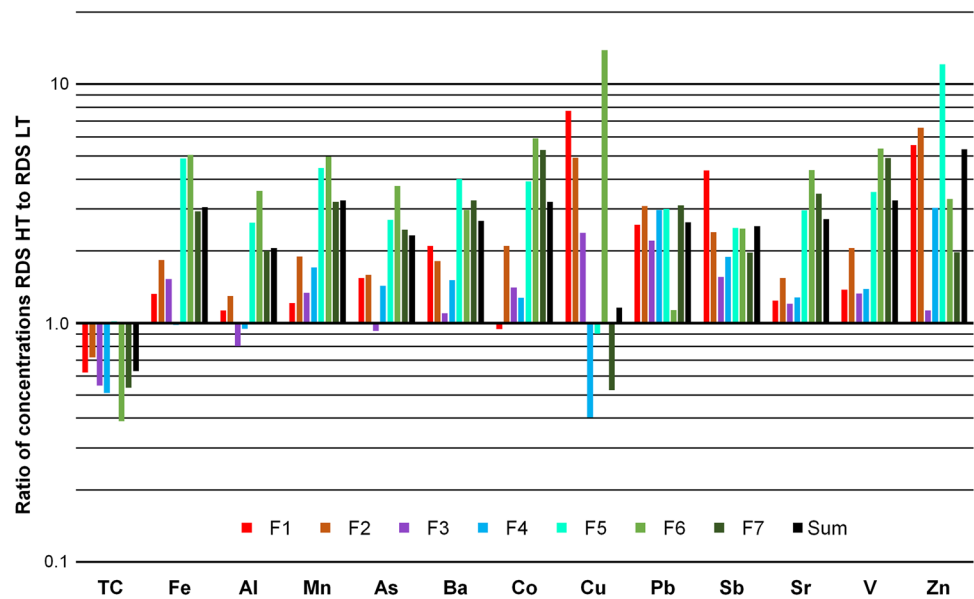
the mass-based size distribution in the range of $4\text{--}6 \mu\text{m}$ (Iijima et al. 2007). The corresponding maxima and the similar course reflect that the main source of Sb in road dust is brake wear.

The dependence of the Ba concentration on the particle size was also very similar in all studies. Besides Cu, Fe and Sb, Ba is also a common tracer element for brake wear (Wahid 2018). Gietl et al. (2010) even suggested using Ba as a quantitative tracer for brake wear emissions, because brake wear is the predominant source for Ba in the urban environment. This is supported by the substantially lower concentrations in the coarse size fractions.

Concentration ratios by size fraction

Figure 4 shows the concentration ratios RDS HT to RDS LT for the analysed elements by size fraction. For most elements, a certain trend in the size dependence of this ratio is visible. An exception from this observation is Cu for which the ratio fluctuates from > 1.0 to < 1.0 several times. This is mainly a result of the high variation of the Cu concentration in the RDS LT sample. Some metallic Cu particles or particles with a high Cu content in the RDS LT might be the reason for this. Brake wear dust, another source of Cu in road dust, can be excluded as the source of these fluctuations, because its particle size is mostly smaller than $10 \mu\text{m}$ (Gietl et al. 2010). Therefore, it

Fig. 4 Concentration ratio RDS HT to RDS LT in the size fractions F1–F7



should be found mainly in the size fractions F1–F3 and not in the coarse size fractions.

Surprisingly, for most of the metals (Fe, Al, As, Ba, Co, Mn, Sr and V) the concentration ratio RDS HT to RDS LT was higher in the coarse size fractions. However, these metals are usually attributed to different sources: Fe and Ba are attributed to brake wear (Gietl et al. 2010), while Al, As, Fe, Mn and V are used as tracers for re-suspended road dust (Pant and Harrison 2013). There is little information available about Co in road dust. Yıldırım and Tokaloğlu (2016) reported that Co is in a cluster with Ni and Cr, elements, which are found in brake dust and tire tread wear (Loganathan et al. 2013). In contrast, Sr is not especially related to traffic emissions. A possible explanation for this finding could be that the contamination of the RDS LT sample is strongly influenced by atmospheric transport of road dust re-suspended into the atmosphere in the surrounding areas with higher traffic and subsequent deposition of this dust in the low-traffic area. Smaller particles can be transported longer distances, thus contaminating areas more distant from the source. In contrast, re-suspended larger particles redeposit mostly nearby because of a higher settling velocity. The relevance of re-suspension of road dust by the traffic is well known. In a study by Patra et al. (2008), the determined transfer into the atmosphere of the PM10 fraction of road dust was 40%. Similar values were found in a study by Bogacki et al. (2018). Additionally, re-suspension of road dust is significantly reduced in the low-traffic area because of the low-traffic density. Thus, a net flow of fine road dust would result from high-traffic areas to low-traffic areas.

In contrast to the findings for the ratios of Fe, Al, As, Ba, Co, Mn, Sr and V, the ratios for Pb, Sb and Zn were rather size independent. Since Pb, Sb and Zn are also attributed to traffic emissions, a reasonable explanation for this difference is currently not available.

Conclusion

Generally, the enrichment of metals in the RDS LT sample <2.0 mm in comparison to the concentrations in local soil was not very high. With the exception of Cu, the concentrations were up to twice the background concentrations, while for Cu the concentration was 15 times the background concentration.

For the metals, the concentrations were two to five times higher in the road dust sample from the high-traffic area compared to the low-traffic area. For TC, an opposite trend was observed.

The concentration of all metals decreased from the finest size fractions to the coarser size fractions. The minimum concentrations were typically found in F5, while in F6 and F7 the concentrations were somewhat higher.

The comparison of the size dependence of the concentration of Ba, Cu, Mn, Pb, Sb and Zn with the literature data showed quite similar trends in the few studies available although the concentration levels were quite different. For Cu, the differences were somewhat higher especially for the coarser size fractions.

The study shows for the first time the ratio of the concentration in RDS HT and RDS LT by particle size. For several metals, this ratio was quite different for fine and coarse size fractions. For the metals Al, As, Co, Mn, Sr, V and the highly traffic-related metals Ba and Fe the ratio was in the range of 0.8–2 for F1–F4, while it was typically in the range of 2–6 for F5–F7. This result might be explained by the different settling velocities of fine and coarse particles. In contrast to coarser particles, the smaller particles can be transported longer distances and, therefore, be deposited also in more distant areas with low-traffic emissions. Therefore, a reduction in traffic metal emissions would also lead to reduced metal concentrations in such areas. However, for the metals Pb, Sb and Zn which largely

come from traffic emissions, this trend is not evident. Very high fluctuations in the ratio of the concentration in RDS HT and RDS LT were found for Cu, which can be attributed to the high variation of the Cu concentration in the coarse size fractions. These fluctuations are caused mainly by the variations in the Cu concentration of the RDS LT sample and might be the result of the presence or absence of small pieces of Cu or Cu-rich particles in the analysed road dust sample.

A measure for reduction in unwanted metals in road dust could be the reduction in the content of these metals in brake linings and tires (Balasubramanian et al. 2017). Regenerative braking which does not rely on frictional wear of brake materials could also be a method to reduce brake wear emissions.

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