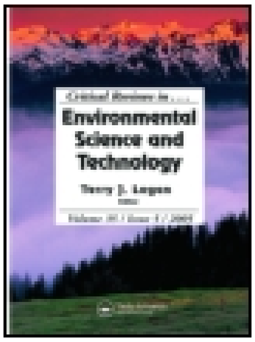

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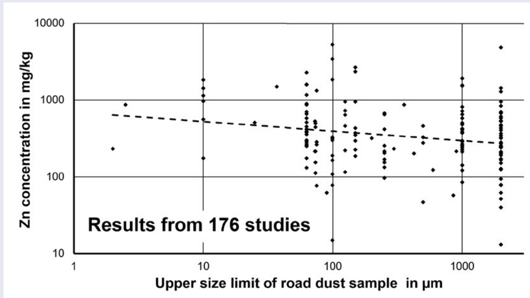
Toward more intercomparable road dust studies

Christof Lanzerstorfer

School of Engineering/Environmental Sciences, University of Applied Sciences Upper Austria, Wels, Austria

ABSTRACT



Road dust (RDS) often contains elevated concentrations of pollutants, especially metals. Numerous studies were performed in the last two decades investigating the concentrations of metals like Cu, Pb, and Zn in RDS. In a literature search 177 studies were found where RDS bulk samples were analyzed. In another 49 studies the RDS samples were split into a number of size fractions to consider also the size dependence of the metal concentrations. In RDS bulk sample studies, the upper size limit (USL) of the RDS samples ranged from 2 µm to more than 2000 µm. This is partly a result of the different aims of the studies. However, the concentrations of metals in RDS particles are quite size-dependent. Consequently, comparing of results from different studies makes little sense. Based on the available literature, a standardized sample preparation sequence is proposed in this work. To serve the various aims of RDS studies the separation of the samples into four size fractions is suggested: <10 µm, 10–63 µm, 63–250 µm, and 250–2000 µm. The determination of the mass fractions of the size fractions parallel to the chemical analysis then allows calculation of the concentrations for RDS bulk samples with the four different USLs.

KEYWORDS Road dust; size fractions; upper size limit

1. Introduction

In the last two decades, a large number of studies were carried out all over the world to determine the concentrations of metals in road dust or road-deposited sediments (RDS) with respect to the sources and transport pathways of the metals, their spatial distribution and their bio-availability and resulting risks to humans (Loganathan, Vigneswaran, & Kandasamy, 2013). Also the transformation processes in the RDS occurring in the environment

CONTACT Christof Lanzerstorfer c.lanzerstorfer@fh-wels.at School of Engineering/Environmental Sciences, University of Applied Sciences Upper Austria, Stelzhamerstraße 23, A-4600 Wels, Austria.

were investigated (Jayarathne, Egodawatta, Ayoko, & Goonetilleke, 2019). A major source of metals like Cu, Pb, and Zn in the environment is road transport. The considerable technological improvements made in recent years helped to reduce the particulate matter emissions from combustion engine exhausts. Thus, the nonexhaust emissions of road transport such as those from RDS resuspension, brake wear, tyre wear, and road wear contribute to the ambient air particulate matter concentrations in cities to a similar extent than the tailpipe exhaust emissions (Amato et al., 2014), and RDS resuspension seems to be dominant in terms of particulate mass. This has increased the environmental concern related to RDS.

In the literature search covering the last 20 years, a total of 177 studies published in English were found reporting the concentration data of bulk RDS samples for metals like Cu, Pb, and Zn. Another 14 papers found were based on the same RDS concentration data as published in one of the studies. Studies investigating roadside topsoils where the upper few centimeters of the roadside soil were sampled were not included. A further 49 studies reported concentrations of metals for size-fractionated RDS samples. Close to half of these studiers (21 papers) also reported metal concentration data for the bulk RDS samples. In another 8 papers data of concentrations by particle size from previous studies were used.

The concentration of metals in RDS depends also on the size of the particles. Typically, the highest metal concentrations are found in the finest size fractions of the RDS. In a study by Al-Rajhi, Al-Shayeb, Seaward, and Eswareds (1996), where the RDS samples were sieved into eight size fractions between 1.0–2.0 mm and <40 µm the maximum concentrations were two to six times the minimum concentrations depending on the metal. In a recent study the significant influence of the upper size limit (USL) for bulk RDS samples applied in the sample collection and preparation procedure on the concentration results was demonstrated (Lanzerstorfer & Logiewa, 2019). Depending on the metal the concentration of an RDS semple with an USL of 37 µm was found to be up to eight times higher compared to the same RDS sample with an USL of 2000 µm.

In many RDS studies, tables are presented comparing the concentration results with data from other studies. Thereby, results for RDS samples with quite different USLs—sometimes from 2 µm to 2000 µm—are often shown in the same table. In most of such tables the USL of the RDS samples is not even presented. Thus, comparison of the results makes little sense.

The aim of this work is to evaluate the RDS studies performed in the last two decades in order to gain a better understanding of the effect of the size dependence of the metal concentrations and to come up with feasible suggestions for future RDS studies which allow comparison of the results. Figure 1 gives an overview of the studies included in the evaluation.

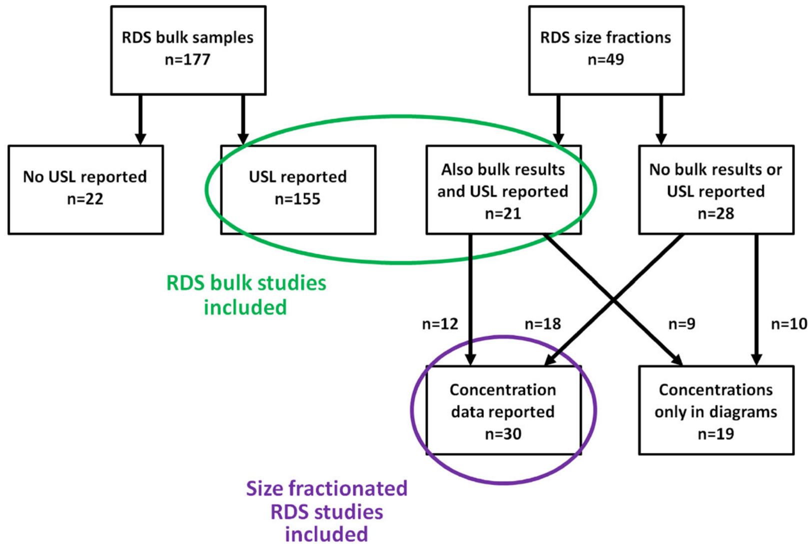


Figure 1. RDS studies included in the evaluation.

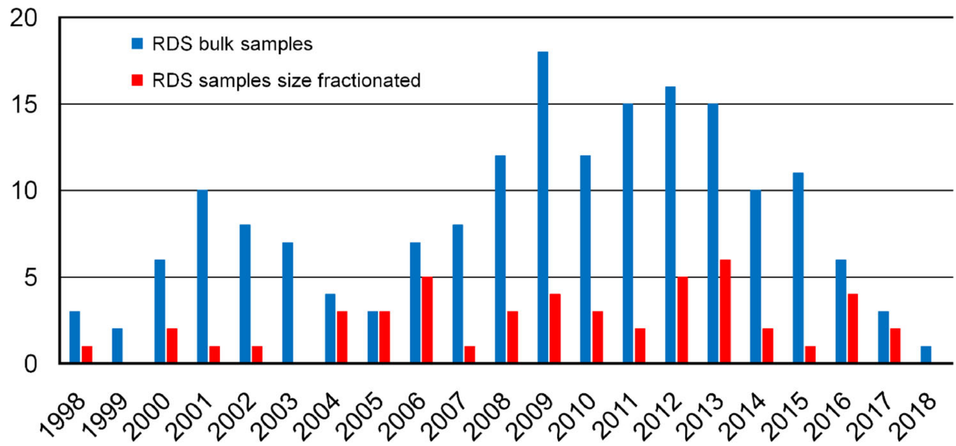


Figure 2. Annual number of studies investigating RDS metal concentrations.

2. Overview RDS studies

In this evaluation, 177 studies performed in the last two decades investigating the metal content of RDS bulk samples and 49 studies investigationg size-fractionated RDS samples were included. Figure 2 shows the number of studies performed each year. The year refers to the end of the RDS sampling period. If the sampling period was not reported in the study the date of submission for publication was used. Therefore, the numbers are quite low for the last few years. However, it can be assumed that some studies with samples collected in these years are ongoing or in submission status since in the evaluated studies the average time period between RDS sampling and publication was approximately 3 years.

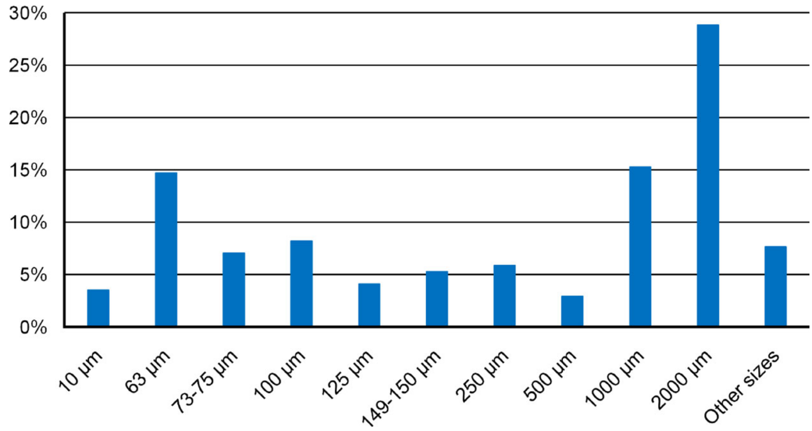


Figure 3. Frequency of use of various upper size limits (USLs) for RDS bulk samples.

The majority of the RDS bulk sample investigations were performed in Asia (67%), especially in China (36%). In contrast, studies investigating size-fractionated RDS were performed mainly in Europe (29%) and in China (27%).

In the literature two main methods are described for collecting RDS samples: on the one hand sweeping with a brush or a broom and collecting the sample with a dust pan and on the other hand vacuuming the road surface. Vacuum sampling is applied in different ways. For sampling of RDS from small areas dry or wet vacuum cleaners are used, while the discharge from street-sweeping vehicles gives an average sample for a large area. In approximately 90% of the studies the RDS sample collection method was descibed. In the RDS bulk studies the brush and dust pan method was mostly used (88%), while small-scale vacuum sampling was used in 9% and samples collected by street-sweeping vehicles were used in 3%. For the studies with sice-fractionated RDS samples the distribution of the used sampling methods was quite different: 53% small-scale vacuum sampling, 40% brush and dust pan sampling, and 7% samples from street-sweeping vehicles.

3. RDS bulk samples

Despite the importance of the USL, 22 out of 177 RDS bulk sample studies did not report a defined USL of the RDS samples. Some of these studies reported the manual removal of stones and other coarse debris. In the remaining 155 studies the USL applied was between 2 µm and 2000 µm. Figure 3 shows the frequency of USL use. The most frequently used USL was 2000 µm followed by 1000 µm and 63 µm. In most cases the USL was ensured by sieving with a mesh with the respective size. Only for achieving a USL of 10 µm or less the collected RDS samples were redespersed into air and then collected with a PM10 or PM2.5 sampling device. In one study the RDS was suspended in a sedimentation device and collected with a pipette (Nazzal, Rosen, & Al-Rawabdeh, 2013). On average, a slight trend to lower values of the USL can be observed over the years.

Most of the USLs used for RDS samples represent an upper limit of a soil size class according to Folk (1974): 1000–2000 µm (very coarse sand), 500–1000 µm (coarse sand), 250–500 µm (medium sand), 125–250 µm (fine sand), 63–125 µm (very fine sand), 54–63 µm (silt and clay), and <54 µm (clay).

In several studies the USL chosen resulted from the aim of the study. In studies investigating the relation between air quality and RDS a USL of 10 µm (PM10) was usualy chosen (Amato et al., 2009; Chow, Watson, Ashbaugh, & Magliano, 2003; Kong et al., 2011). In many other studies the selected USL was also decided by the aim of the study. However, the choice of the USL is not always reasonable. The majority of the studies reported metal pollution in more general terms. The applied USL ranged from 2 µm (Nazzal et al., 2013) to 2000 µm. In several of the studies the spatial distribution of metals in an larger area was invetigated, Quite different USLs were applied, for example 2000 µm, 500 µm, and 125 µm (Ordonez, Loredo, DeMiguel, & Charlesworth, 2003; Shi et al., 2008; Tang, Ma, Zhang, & Mao, 2013). Also in studies dealing with the seasonal variation of metal concentrations in RDS, various values for the USL were used, ranging from 150 µm (Lin, Gui, Wang, & Peng, 2017) to 1000 µm (Robertson & Taylor, 2007).

Many studies dealt with the assesment of human health risk by RDS. The typical USL used in these studies was 63 µm (Hu et al., 2011; Li, Qian, Hu, Wang, & Gao, 2013; Saeedi, Li, & Salmanzadeh, 2012). However, USLs of 100 µm (Ferreira-Baptista & de Miguel, 2005), 125 µm (Shi et al., 2010), 850 µm (Ma & Singhirunnusorn, 2012), 1000 µm (Wang, Lu, Ren, Li, & Chen, 2014), and even 2000 mm (Kamani et al., 2017) were also used.

In studies investigating the relation between metal concentrations in RDS and stormwater runoff, USLs from 75 µm (Aryal et al., 2017) to 2000 µm (Brown & Peake, 2006) were used. Also in studies on source appointment quite different USLs, for example 37 µm, 125 µm, and 1000 µm (Pan, Lu, & Lei, 2017; Valotto et al., 2015; Žibret, Van Tonder, & Žibret, 2013) were applied. The same was found in studies comparing the metal concentrations in indoor and outdoor dust. The USLs ranged from 125 µm (Žibret & Rokavec, 2010) to 595 µm (Yaghi and Abdul-Wahab, 2004).

Table 1 summarizes concentration results for Cu, Pb, and Zn from the 155 RDS bulk studies reporting a USL and the 21 studies with size-fractionated RDS reporting also results for the bulk RDS samples, which represent more than 9000 individual RDS samples. For each study the calculated average concentrations for all RDS samples were used. Figure 4 shows the concentrations

Table 1. Average RDS bulk sample concentrations of metals in 176 studies reporting the USL of the sample.

	End of sampling period	Upper size limit (USL)	Number of individual RDS samples	Cu in mg/kg	Pb in mg/kg	Zn in mg/kg
Abbasi et al. (2017)	2014	2000	24	118	95	283
Acosta, Faz, Kalbitz, Jansen, and Martinez-Martinez (2014)	2011	2000	15	91	189	246
Acosta et al. (2015)	2014	2000	18	93	93	152
Addo, Darko, Gordon, Nyarko, and Gbadago (2012)	2011	250	50	61	23	134
Ahmed and Ishiga (2006)	2003	1000	70	38	49	121
Ali et al. (2017)	2016	74	90	43	–	154
Al-Khashman (2004)	2003	2000	15	11	11	13
Al-Khashman (2007a)	2004	2000	100	281	844	318
Al-Khashman (2007b)	2004	2000	127	32	145	125
Al-Khashman (2013)	2011	2000	175	20	55	165
Al-Momani (2009)	2005	2000	74	139	162	402
Amato et al. (2009)	2007	10	13	1153	227	1425
Amato et al. (2011)	2008	10	23	2030	216	1834
Aminiyan et al. (2018)	2015	63	200	791	123	253
Andrews and Sutherland (2004)	2002	63	15	235	445	902
Apeagyei, Bank, and Spengler (2011)	2008	2000	85	105	73	240
Arslan (2001)	2000	90	10	—	230	62
Aryal et al. (2017)	2012	150	9	182	216	950
Atiemo, Ofosu, Aboh, and Oppon (2012)	2009	100	60	71	112	327
Bada and Oyegbami (2012)	2012	100	4	—	185	15
Banerjee (2003)	1999	500	4	224	249	330
Benhaddya, Boukhelkhal, Halis, and Hadjel (2016)	2015	2000	13	84	288	257
Bian and Zhu (2009)	2006	2000	88	81	282	538
Bourliva, Papadopoulou, and Aidona (2016)	2014	250	30	526	191	671
Bourliva et al. (2017)	2009	63	31	662	209	453
Brown and Peake (2006)	2001	2000	3	129	289	528
Cai and Li (2019)	2018	74	234	91	155	496
Cao et al. (2018)	2014	25	32	62	122	509
Cesari et al. (2012)	2011	2000	5	70	36	205
Chang et al. (2009)	2006	63	28	121	189	1181
Charlesworth, Everett, McCarthy, Ordonez, and de Miguel (2003)	2002	1000	149	388	48	485
Chow et al. (2003)	2002	10	3	168	109	965
Christoforidis and Stamatis (2009)	2003	63	88	131	322	287
Dehghani, Moore, Keshavarzi, and Hale (2017)	2015	63	30	275	213	666
Deocampo, Reed, and Kalenuik (2012)	2011	250	74	69	82	214
Divrikli, Soylak, Elci, and Dogan (2003)	1999	600	42	30	53	—
Duan, Wang, Zhang, and Xuan (2017)	2017	149	79	130	68	186
Duong and Lee (2009)*	2006	2000	9	127	164	128
Duong and Lee (2011)	2009	2000	10	129	106	170
Darán and Gonzalez (2009)	2006	75	14	3768	—	—
Duzgoren-Aydin et al. (2006)	2003	2000	30	176	240	586
El-Hasan et al. (2006)	2004	2000	23	24	51	95
Elik (2003)	2000	297	36	94	223	231
Ewen, Anagnostopoulou, and Ward (2009)	2006	125	75	269	207	463
Faiz, Tufail, Javed, Chaudhry, and Naila (2009)	2009	125	13	52	104	116
Ferreira-Baptista and de Miguel (2005)	2002	100	92	42	351	317

(*continued*)

Table 1. Continued.

	End of sampling period	Upper size limit (USL)	Number of individual RDS samples	Cu in mg/kg	Pb in mg/kg	Zn in mg/kg
Gabarrón, Faz, and Acosta (2017)	2016	2000	63	133	155	462
German and Svensson (2002)	2000	250	18	282	45	257
Gunawardana, Goonetilleke, Egodawatta, Dawes, and Kokot (2012a)	2010	425	8	92	31	200
Guney, Onay, and Copty (2010)	2008	2000	20	111	177	245
Han et al. (2008)	2001	1000	65	95	231	421
Han et al. (2014)*	2013	250	16	131	86	205
Han, Lu, Zhang, Wuyuntana, and Pan (2016)	2012	1000	121	29	58	86
Harb et al. (2015)	2014	500	80	40	26	47
Hu et al. (2011)	2009	63	35	130	104	394
Hu et al. (2016)	2012	63	144	101	61	—
Huang et al. (2014)	2010	100	30	163	218	1850
Huang et al. (2016)	2013	73	51	44	67	215
Jiries (2003)	2001	2000	12	157	640	323
Jordanova, Jordanova, and Petrov (2014)	2012	63	15	127	105	267
Joshi et al. (2009)	2007	355	18	3138	173	863
Kamani et al. (2015)	2013	2000	60	32	32	191
Kamani et al. (2017)	2015	2000	60	286	81	695
Kantor et al. (2018)	2017	63	21	98	37	379
Karanasiou et al. (2014)	2009	10	72	444	121	1135
Karmacharya and Shakya (2012)*	2012	2000	21	30	22	62
Kartal, Aydın, and Tokalıoğlu (2006)	2000	74	33	84	415	443
Khanal et al. (2014)*	2012	2000	10	254	95	1296
Kim, Doh, Park, and Yun (2007)	2000	1000	50	461	150	816
Kim, Kim, Kang, and Ko (2016)*	2013	63	25	466	297	2285
Kong et al. (2011)	2007	10	12	115	51	174
Kong et al. (2012)*	2010	100	15	85	45	188
Kumar, Furumai, Kurisu, and Kasuga (2013)	2008	2000	2	551	132	847
Lanzerstorfer and Logiewa (2019)*	2017	2000	1	20	12	40
Lanzerstorfer (2018)*	2016	200	4	122	37	320
Lee, Yu, Yun, and Mayer (2005)	2002	150	633	446	214	2665
Li, Poon, and Liu (2001)	2001	2000	45	173	181	1450
Li, Qian, et al. (2013)	2012	63	40	102	72	298
Li, Feng, et al. (2013)	2010	149	55	139	956	2379
Li et al. (2014)	2013	2000	16	83	77	271
Li, Shi, and Zhang (2015)	2013	2000	8	126	511	51
Li, Liang, Wang, and Yang (2015)	2014	100	23	36	184	299
Li, Zhang, et al. (2016)	2013	74	51	44	67	215
Li, Yu, et al. (2016)	2012	500	40	92	73	278
Li et al. (2017)	2015	63	75	102	83	298
Lin et al. (2017)	2015	150	276	28	45	225
Liu, Zeng, Yang, Qiu, and Chan (2009)	2009	1000	42	53	81	255
Liu, Yan, Birch, and Zhu (2014)	2011	100	48	262	116	314
Logiewa et al. (2020)*	2017	1000	2	218	112	505
Lu et al. (2010)	2006	1000	38	123	433	715
Lu et al. (2014)	2013	1000	42	33	75	142
Ma and Singhirunnusorn (2012)	2011	850	8	13	10	58
Ma, Chen, Li, Bi, and Huang (2016)	2012	149	43	105	262	377
Manasreh (2010)	2008	63	24	69	143	132
Mashi, Yaro, and Eyong (2005)	2003	2000	75	97	210	79
Rastegari Mehr et al. (2016)	2015	63	31	66	132	174
Men, Liu, Wang, Guo, and Shen (2018)	2016	1000	72	77	65	290

(*continued*)

Table 1. Continued.

	End of sampling period	Upper size limit (USL)	Number of individual RDS samples	Cu in mg/kg	Pb in mg/kg	Zn in mg/kg
Nazzal et al. (2013)	2011	2	42	162	183	233
Okorie et al. (2012)	2011	250	9	132	992	421
Ordonez et al. (2003)	2002	2000	112	183	514	4892
Ozaki, Watanabe, and Kuno (2004)	2001	2000	17	—	—	—
Padoan et al. (2017)*	2016	2000	29	181	74	200
Pal, Wallis, and Arthur (2011)	2010	1000	65	68	77	212
Pan et al. (2017)	2015	1000	90	55	125	269
Pathak, Yadav, Kumar, and Kumar (2013)	2012	2000	63	216	55	707
Perez, Lopez-Mesas, and Valiente (2008)	2007	2000	13	216	283	542
Poleto, Bortoluzzi, Charlesworth, and Merten (2009)	2008	63	20	114	52	256
Praveena and Aris (2018)	2016	73	51	425	593	527
Qiang, Yang, Jingshuang, Quanying, and Zou (2015)	2013	2000	38	43	71	171
Qiao, Schmidt, Tang, Xu, and Zhang (2014)	2009	2000	132	80	77	343
Rajaram, Suryawanshi, Bhanarkar, and Rao (2014)	2009	75	7	169	129	264
Rijkenberg and Depree (2010)	2009	1000	2	53	235	385
Robertson and Taylor (2007)*	2001	1000	72	204	258	288
Robertson, Taylor, and Hoon (2003)	2001	1000	19	113	265	653
Saeedi et al. (2012)	2010	63	50	222	254	864
Sager, Chon, and Marton (2015)	2013	75	8	322	263	1330
Sezgin et al. (2004)	2002	500	22	115	189	459
Shabbaj et al. (2017)	2016	63	24	146	147	510
Shen, Liu, Aini, and Gong (2016)	2014	1000	20	106	74	256
Shi and Wang (2013)	2008	1000	30	132	77	375
Shi et al. (2008)	2007	125	273	197	295	734
Shi et al. (2010)	2009	125	43	218	210	641
Shinggu, Ogugbuaja, Barminas, and Toma (2007)	2006	250	10	18	71	154
Shinggu, Ogugbuaja, Toma, and Barminas (2010)	2007	2000	10	60	339	616
Singh (2011)	2006	2000	13	26	48	78
Soltani et al. (2015)	2012	63	24	182	393	707
Suryawanshi, Rajaram, Bhanarkar, and Chalapati Rao (2016)	2009	75	9	192	121	285
Sutherland and Tolosa (2000)	2000	2000	13	167	106	434
Sutherland, Tack, and Ziegler (2012)	2012	2000	20	409	537	671
Taiwo et al. (2017)	2015	2000	39	14	32	94
Tamrakar and Shakya (2011)	2011	250	15	—	19	96
Tang et al. (2013)	2009	500	220	64	50	—
Tang et al. (2017)	2014	100	70	39	45	—
Tanner, Ma, and Yu (2008)	2008	63	25	235	150	1608
Tokalıoğlu and Kartal (2006)	2003	74	29	37	75	112
Trujillo-González, Torres-Mora, Keesstra, Brevik, and Jiménez-Ballesta (2016)	2014	2000	15	213	468	210
Turner and Hefzi (2010)	2008	63	4	105	40	356
Tüzen (2003)	2001	75	6	32	164	76
Urrutia-Goyes, Hernandez, Carrillo-Gamboa, Nigam, and Ornelas-Soto (2018)	2017	250	44	167	227	649
Valdez Cerda, Hinojosa Reyes, Alfaro Barbosa, Elizondo-Martinez, and Acuña-Askar (2011)	2001	2000	30	—	300	475

(*continued*)

Table 1. Continued.

	End of sampling period	Upper size limit (USL)	Number of individual RDS samples	Cu in mg/kg	Pb in mg/kg	Zn in mg/kg
Valotto et al. (2015)	2014	37	16	1814	678	1495
Vega et al. (2001)	1998	25	3	150	380	870
Wang et al. (2012)	2006	1000	71	73	63	297
Wang et al. (2014)	2010	1000	38	178	1586	1919
Wang, Li, et al. (2016)	2010	63	60	141	119	585
Wang, Lu, and Pan (2016)	2015	100	94	49	98	165
Watson and Chow (2001)	2000	10	11	86	189	554
Wei, Jiang, Li, and Mu (2009)	2007	149	169	95	54	294
Wei, Jiang, Li, and Mu (2010)	2007	149	42	101	57	349
Wei, Gao, Wang, Zhou, and Lu (2015)	2010	125	152	70	105	223
Xiang, Li, Yang, and Shi (2010)	2009	1000	9	108	72	239
Xie, Dearing, Boyle, Bloemendal, and Morse (2001)	1998	1000	76	—	985	1531
Xu, Lu, Han, and Zhao (2015)	2012	100	116	32	60	78
Yaghi and Abdul-Wahab (2004)	2003	595	119	47	65	124
Yang, Liu, Li, Zeng, and Chan (2010)	2002	1000	31	62	103	224
Yekeen et al. (2016)	2013	2000	44	—	393	—
Yetimoğlu, Ercan, and Tosyali (2007)	2004	149	65	191	368	431
Yeung, Kwok, and Yu (2003)	2001	100	7	98	110	3479
Yongming, Peixuan, Junji, and Posmentier (2006)	2001	1000	65	95	231	421
Yu et al. (2014)	2013	63	87	55	61	—
Yuen et al. (2012)	2011	63	30	614	241	1596
Yun, Choi, and Lee (2000)	1998	2000	28	269	144	532
Zafra-Mejía et al. (2019)	2011	250	230	108	92	168
Zanders (2005)*	2002	2000	6	124	249	962
Zhang and Wang (2009)	2005	2000	20	134	235	367
Zhang, Qiao, Appel, and Huang (2012)	2008	1000	69	140	228	583
Zhang, Deng, Wang, Chen, and Xu (2013)	2012	63	38	129	136	421
Zhang, Hua, and Krebs, (2015)*	2012	1000	12	200	—	410
Zhang, Lu, Chen, Gao, and Fu (2015)	2013	1000	42	32	75	142
Zhao, Lu, Chao, and Xu (2015)	2012	100	204	41	53	109
Zheng and Zhang (2008)	2005	900	63	42	61	214
Zheng, Liu, Wang, and Liang (2010)	2009	100	35	264	533	5271
Zhou et al. (2015)	2011	100	72	918	1559	—
Zhu, Li, Bi, Han, and Yu (2013)	2012	1000	91	305	477	1552
Žibret and Rokavec (2010)	2009	125	6	149	146	961
Žibret et al. (2013)	2012	125	46	73	89	222

*Studies investigating size fractionated RDS reporting also the concentrations for the bulk samples and the USL.

of Cu, Pb, and Zn as a function of the USL applied in sample preparation. Especially for Cu and Zn the measured concentrations tend to be lower for larger values of the USL.

4. Size fractionated RDS samples

In 49 studies the metal concentration of RDS was reported for at least two different size fractions. The maximum number of size fractions was 11 and the average number was 4.9 ± 2.0. Figure 5 shows the size ranges of the RDS size fractions investigated in the various studies. An x-symbol at

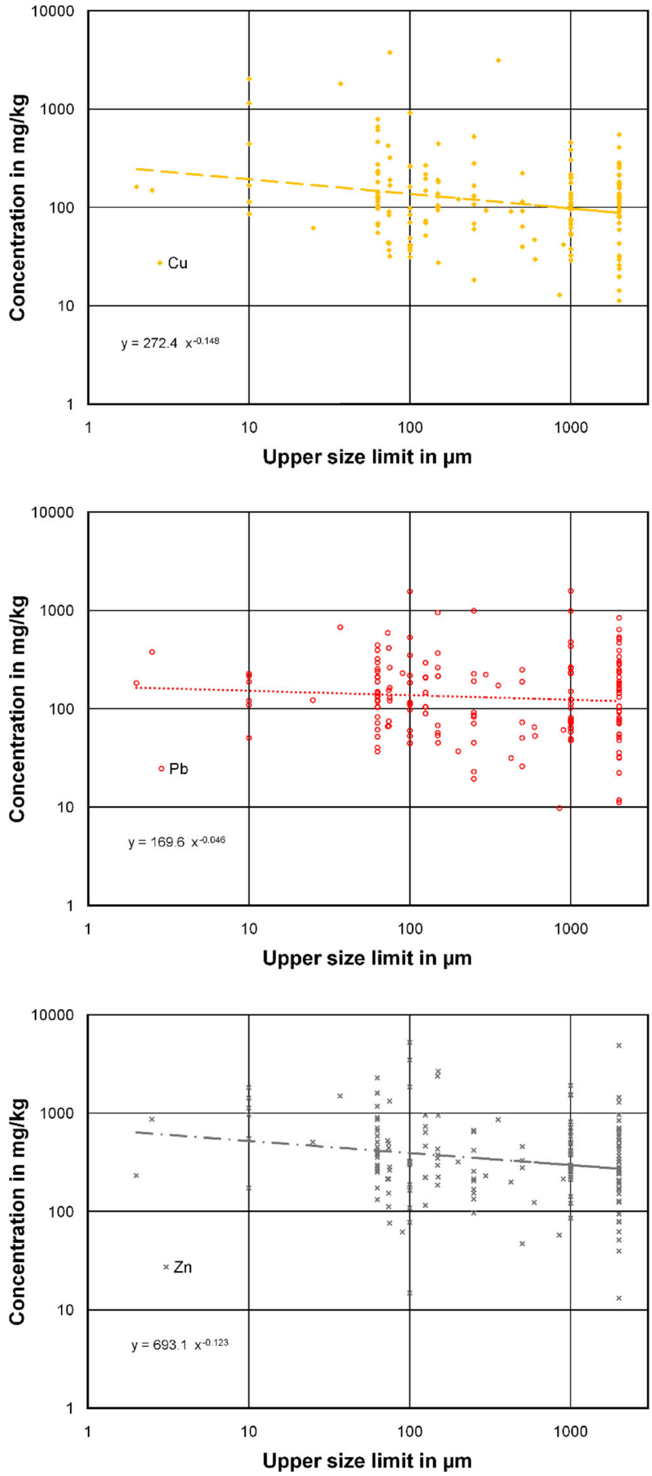


Figure 4. Concentrations of Cu, Pb and Zn as a function of the USL applied in sample preparation.

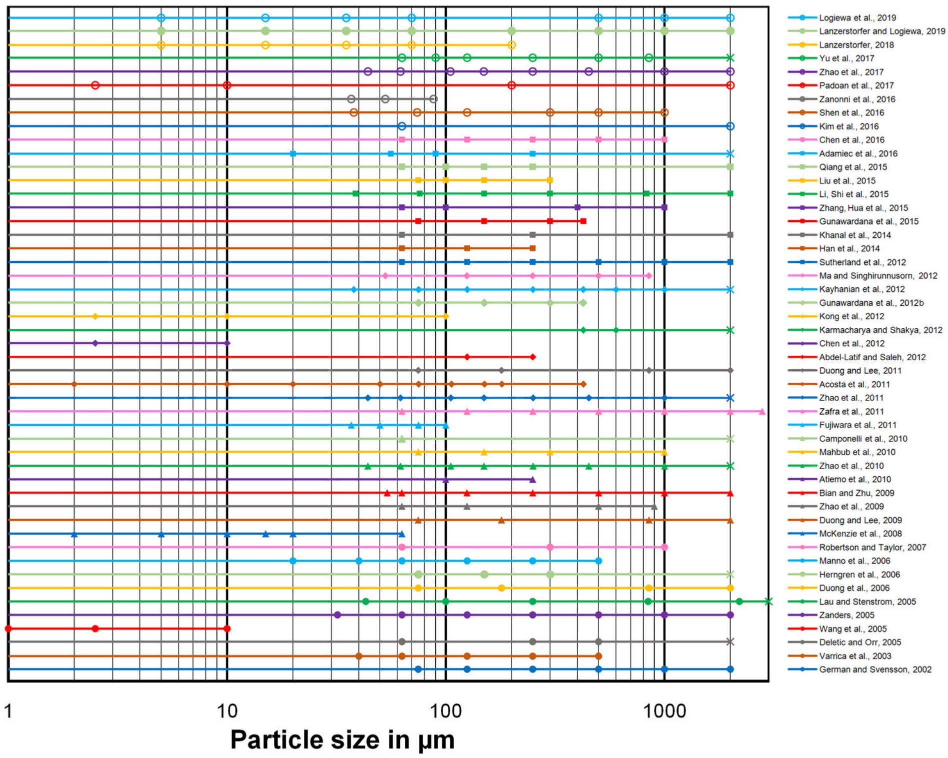


Figure 5. Size ranges of the RDS size fractions investigated.

the upper limit of the largest size fraction indicates an estimate because in the study only the lower limit of the largest size fraction was reported. A wide variation of size limits for the various size fractions ranging from 1 µm to 3000 µm was found.

Size fractionation was performed in most cases by sieving, using meshes with the respective sizes. In some studies RDS samples were redespersed into air and the dust was collected, for example with a PM10, PM2.5, or PM1 sampling device (Chen, Wang, Liu, & Ren, 2012; Kong et al., 2012; Wang, Chang, Tsai, & Chiang, 2005) for achieving a separation at very small paricler sizes. In a few other studies air classification was applied (Lanzerstorfer, 2018; Lanzerstorfer & Logiewa, 2019; Logiewa, Miazgowicz, Krennhuber, & Lanzerstorfer, 2020) and in one study the RDS was suspended in a liquid for size fractionation (Padoan, Romè, & Ajmone-Marsan, 2017).

The reported concentrations are always for the size ranges between the limits except for the studies by Chen et al. (2012) and by Kong et al. (2012), where the data are for the whole size range from zero up to the maximum size.

In 30 studies the metal concentration data were presented in tables, while in 19 studies the data were shown in diagrams only. In both groups, the metal concentrations were higher in the finer size fractions of the RDS. However, investigation of the size dependence of the concentrations only the data from the tables could be used. The geometric mean of the size limits was used as a representative particle size for each size fraction except for the finest size fraction with the lower limit of zero, where the arithmetic mean was used. For each study the average concentrations for each size fractions were calculated. Generally, the concentrations are higher in the finer size fractions. However, the gradient of this trend is different in the various studies. The Cu concentration frequently shows deviations from the general trend of the size-dependence. Especially in the coarser size fractions $>100\,\mu m$ higher concentrations can be found. Such fluctuations are less frequently found for Pb and Zn. The gradients of the size dependence have a significantly higher negative value in comparison to the gradients in Figure 4. This is because the data in Figure 4 show the average concentration for the dust samples from zero up to the applied USL.

To make results of the various studies comparable, relative concentrations were calculated, where the concentration was related to the concentration for a particle size of 63 µm. The theoretical concentration for a particle size of 63 µm was obtained by linear interpolation in a double-logarithmic scale using the nearest two data points. Figure 6 shows the results for Cu, Pb, and Zn. The exponent b of the regression function $c_i = A_i/x^{b_i}$ is a measure for the size dependence of the concentration. It was highest for Zn (0.38), followed by Pb (0.34), and was lowest for Cu (0.29). The highest correlation coefficient r^2 was found for Zn (0.76), followed by Pb (0.65). The low correlation coefficient of 0.48 for Cu reflects the more frequent fluctuations of the Cu concentration as can be seen in Figure 5.

5. Toward a standard for size limits in RDS sample preparation

In the comparison of RDS metal concentrations measured at different locations, results from studies were frequently used where different USLs were applied during sample preparation typically ranging from 63 µm to 2000 µm (Ferreira-Baptista & de Miguel, 2005; Manasreh, 2010; Saeedi et al., 2012). In several cases the values of USLs are even not shown in the tables (Christoforidis & Stamatis, 2009; Joshi, Vijayaraghavan, & Balasubramanian, 2009; Yu, Wang, & Zhou, 2014). In fact, in some studies average concentrations were calculated for samples with different USLs, for example USLs ranging from 125 µm to 2000 µm (Wei & Yang, 2010) or even from 10 µm to 2000 µm (Okorie, Entwistle, & Dean, 2012; Zafra-Mejía, Gutiérrez-Malaxechebarria, & Hernández-Peña, 2019).

However, the comparison of data of RDS samples or the calculation of an average from RDS samples with a different USL are quite meaningless.

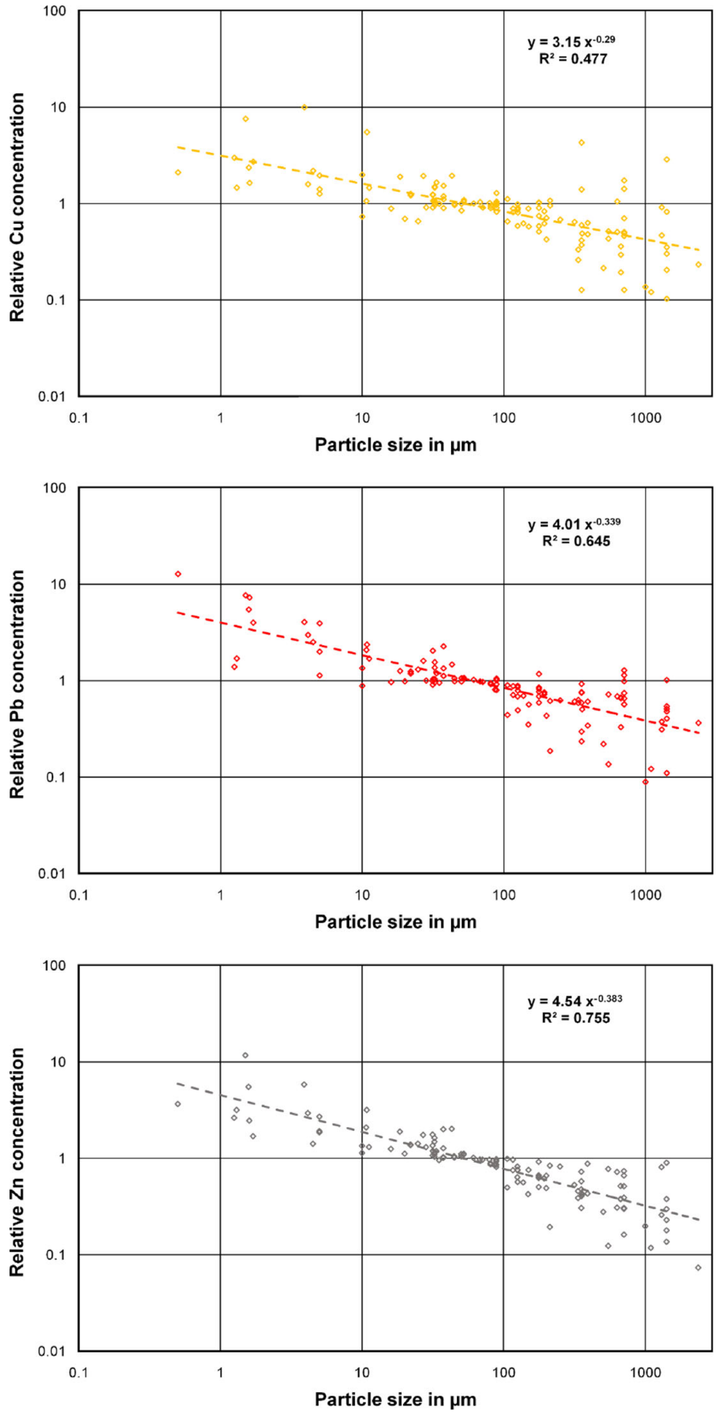


Figure 6. Relative concentration of Cu, Pb, and Zn based on the calculated concentrations for a particle size of 63 µm.

Table 2. Suggested size fractions for RDS studies.

Suggested USL of the size fraction	Fractionation method	Mesh size	Representative particle size in μm	Number of size fraction k
2000 μm	Dry sieving	10	700[a]	4
250 μm	Dry sieving	60	125[a]	3
63 μm	Dry sieving	230	25[a]	2
10 μm	Wet sieving or resuspension and PM10 sampling or air classification	850	5[b]	1

[a]Geometric mean of the upper and lower sieve size.
[b]Arithmetic mean for the finest size fraction.

To make such investigations reliable a more standardized sample preparation would be useful. The following items have to be considered:

- The produced data should be usable for assessments of various kinde (resuspension into the atmosphere, stormwater runoff, hazards, and risk)
- The laboratory work should remain limited.
- For sieving, similar mesh sizes according to US and ISO standards should be available

Because of the different requirements with respect to particle size resulting from the various aims of RDS studies the application of size fractionation of the RDS samples would be preferable instead of the investigation of RDS bulk samples only. Minimizing the number of size fractions produced for chemical analysis is recommended because of the related costs. The suggested limits for the RDS size fractions are summarized in Table 2.

The most reasonable USL for the whole RDS sample and therefore for the largest size fraction is 2000 μm because this would be in accordance with the USL used in many RDS studies performed in the past.

In air quality studies PM10 is widely used (Amato et al., 2009; Chow et al., 2003; Kong et al., 2011). Therefore, 10 μm would be the most feasible upper limit for the finest size fraction. Additionally, experiments by Kuhns et al. (2001) and Hussein, Johansson, Karlsson, and Hansson (2008), where the increase of the airborne dust concentration behind the wheels of a driving car was measured for various size fractions, showed that resuspension of dust from the road surface into the air is mainly limited to particle sizes <10 μm. Therefore, this size fraction is representative for dust resuspension.

The mobility of RDS particles with stormwater runoff also strongly depends on the particle size. In a study by Kim and Sansalone (2008) 65–99% of the particles in the effluent after the local particle separator were <75 μm. Therefore, studies investigating stormwater runoff usually concentrate on fine size fractions like <38 μm (Kayhanian, McKenzie, Leatherbarrow, & Young, 2012), <63 μm (Camponelli, Lev, Snodgrass, Landa, & Casey, 2010), or <75 µm (Li & Zuo, 2013; Liu, Liu, Li, & Guan, 2015). Particle size distribution measurements of the suspended solids in stormwater runoff showed that the particles are smaller than approximately 250 µm (Aryal et al., 2017). Thus, two further size fractions would be recommended: firstly 10–63 µm and secondly 63–250 µm. The 63 µm limit would serve for runoff studies and is also the USL of many road dust studies performed in the past. The 250 µm limit also fits well into the sequence since the representative particle size of the four size fractions are nearly equidistant on the logarithmic size scale.

With the exception of the separation of the finest size fraction <10 µm the proposed size limits were already used in a RDS study by Khanal, Furumai, and Nakajima (2014). Additionally, the proposed size limits 63 µm, 250 µm, and 2000 µm were frequently used in studies with size fractionation of RDS samples (Figure 5).

In a study the mass fractions of the four resulting size fractions 0–10 µm, 10–63 µm, 63–250 µm, and 250–2000 µm also have to be determined. Then the mass concentration of a component n can be calculated for the RDS samples for all three USLs. For the USL x_k the concentration can be calculated from the concentrations in the size fractions smaller than this USL and the corresponding mass fractions of these size fractions using the following equation:

$$c_n(x \le x_k) = \frac{\sum_{i=1}^{k} c_{n,i} \cdot x_{m,i}}{\sum_{i=1}^{k} x_{m,i}}.$$

$c_{n,i}$ is the mass concentration of the component in the size fraction i and $x_{m,i}$ is the mass fraction of size fraction i.

If there would be a limitation in a study to have only one size fraction of RDS to be analyzed, maybe, it would be best to use an USL of 63 µm. On the one hand, this USL covers the requirements of stormwater runoff studies and on the other hand, it is the USL in the fine particle size range which has been used most frequently in the past. However, separating the RDS into the four size fractions mentioned above would always be preferable.

6. Conclusion

Because of concerns about metal pollution, RDS studies were performed all over the world. In most of the studies only RDS bulk samples were analyzed, while in some of the studies the RDS samples were split into a number of size fractions. As shown in many studies, the metal concentrations are substantially higher in the finest size fractions and lower in the coarse size fractions. As a result, the concentrations measured for RDS bulk samples depend on the USL applied during sample preparation. The USLs used in various studies range from 2 µm to 2000 µm. This is partly a result of the different aims of the studies, for example the influence of RDS resuspension on air quality or the transport of RDS with stormwater runoff. Under these circumstances, the comparison of results from different studies is quite limited. Therefore, a standardized sample preparation procedure is proposed. To serve the various aims of RDS studies separation of the RDS samples into four size fractions would be required: <10 µm, 10–63 µm, 63–250 µm, and 250–2000 µm. The chemical analysis of the size fractions together with the determination of the mass fractions allows the calculation of the concentrations for RDS bulk samples with the different USLs.

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