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A Survey of Heavy Metal Contents of Rural and Urban Roadside Dusts: Comparisons at Low, Medium and High Traffic Sites in Central Scotland

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Abstract

Roadside dust can contain particulates enriched with potentially toxic elements (PTEs) as a result of the degradation of mechanical vehicular parts, tyre wear and combustion processes. To assess the potential accumulation of these metals in roadside areas, a snapshot study was carried out, investigating metal content at rural and urban locations in central Scotland. Samples of road dust were collected at six sites representing low, medium and high traffic intensity at rural and urban locations. The samples were separated based on particle size, and analysed for heavy metal content using inductively coupled plasma optical emission spectroscopy (ICP-OES) after acid digestion. The metals analysed were aluminium (Al), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), magnesium (Mg), manganese (Mn), lead (Pb) and zinc (Zn). The rural area measurements were carried out in West Lothian, approximately 13 to 17 miles west of the city of Edinburgh (UK). The urban area measurements were carried out in the southern part of the Edinburgh city district (UK). Concentrations of Cu, Cr and Zn were found to correlate with traffic intensity, although only Cu and Zn concentrations exceed recommended EC directive 86/278/EEC guidelines for urban run-off materials. The metal concentrations of small particles (0.45 – 20 μm) were exceedingly high in both Cu and Zn at areas of high traffic intensity, indicating potential areas of concern regarding health impacts for pedestrians and cyclists who are exposed to roadside dust on a regular basis.

Introduction

The mass utilisation of mechanised vehicles used to transport people and goods in modern day society has become a common part of everyday modern life; however, it brings with it serious consequences for both human health and the wider environment. As well as harmful air pollution emitted as the direct result of the combustion of fossil fuels (i.e. nitrogen oxides (NO_x), volatile organic compounds (VOCs) and particulate matter (PM) in the form of soot (DEFRA, 2019)), the degradation of vehicles via operational wear and tear results in the production of PM enriched with a variety of potentially toxic elements (PTEs) (DEFRA, 2019; Duong & Lee, 2011; Pagotto et al., 2010; Wei & Yang., 2010). As a consequence, the PTE content of roadside dust can increase considerably in areas with high traffic flow. Human exposure then occurs via inhalation or ingestion of such dusts (Du et al., 2013; Li et al., 2013; Soltani et al., 2015).

Since the Industrial Revolution in the mid-19th Century, smelting of metal ores, fossil fuel combustion and, from the early 20th Century, combustion of leaded petrol, released large quantities of PTEs to the atmosphere which were then transferred to UK soils and waters via both wet and dry deposition (Nriagu 1996; Renberg et al., 2001). Since the 1980s, however, atmospheric emissions of PTEs have fallen dramatically due to introduction of industrial regulations established to protect human health (DEFRA, 2019) (e.g. the 1982 EC Directive 82/884/EEC and the 1985 EC Directive 85/210/EEC which limited lead in the air and introduced unleaded petrol). Although the acute toxicity of exposure to many of these elements is well understood, the long-term impacts of low and sustained exposure to these compounds is less defined (Fortoul et al., 2015; Kampa et al., 2008; Singh et al., 2011). There are many diseases and illnesses, e.g. Alzheimers, Parkinson's, cancer, numerous cardiovascular diseases, etc.., that are believed to be linked to elevated heavy metal concentrations in the body (Ashley et al., 2012; Järup, 2003; Rehman et al., 2017; Tchounwou et al., 2012). As the number of

vehicles in use continues to rise at a global scale in an increasingly urbanised world (especially in Asia, e.g. Arora et al., 2011; Huo et al., 2007), human exposure to heavy metals in road dust is likely to increase as a result.

In environments where the PTE content of PM is elevated, exposure to fine particulates (< 2.5 µm) is of greater concern as these particles are not so easily cleared from the lungs via the mucociliary response (the natural process which removes PM from the body (Kreyling et al., 2002)). Smaller particles are more likely to be fully ingested and thus transfer heavy metals into vital organs and the blood stream where PTEs may cause harm in a number of ways. Smaller particles are also more likely to stick to skin, clothes and other surfaces than larger particles, resulting in increased ingestion of particulate matter in foods and especially by young children who frequently put their hands in their mouths (Bekö et al., 2013; Ferreira-Baptista et al., 2005) and tend to ingest higher quantities of soil (US-EPA, 2017). The human bioaccessibility and potential harm posed by particulates in the < 20 µm size fraction is still poorly understood. Traditionally, inhalation is believed to be the primary route of ingestion of road dust, with focus typically limited to the < 2.5 and < 10 µm particulate fractions (Khan & Strand, 2018). These small size fractions are considered to cause respiratory and cardiovascular health problems; however this simplistic assessment has been criticised as it does not account for deposition of particles in the lungs and in-vivo availability (Kastury et al., 2017). Ingestion of larger particles via the stomach is also well documented with variables such as pH and organic matter content both thought to be important in determining bioaccessibility (Poggio et al., 2009; Broadway et al., 2010).

The most significant contributors of PTE pollution from vehicles are considered to be brake wear, tyre erosion, exhaust emissions and oil losses (Napier et al., 2008), although there are also a large variety of other sources. Brakes pads, which degrade over time, are often classed as semi-metallic materials since they contain ~30-65% metal by weight, e.g. including

77 iron (Fe) and copper (Cu) together with lead (Pb), chromium (Cr), zinc (Zn) and antimony (Sb)
78 compounds as friction modifiers (Garg et al., 2000; Thorpe & Harrison, 2008). One reason that
79 brake wear can have considerable health effects is that the brake pad materials often degrade
80 into very small (< 2.5 μm) high-density particles which can be easily inhaled deep into our
81 lower respiratory system. Therefore their toxicity has to be considered with more attention than
82 larger particles (> 20 μm) (Garg et al., 2000).

83 Tyre erosion is also a significant source of heavy metal traffic pollution. Tyre tread
84 contains heavy metals such as manganese (Mn), Fe, cobalt (Co), nickel (Ni), Cu, Zn, cadmium
85 (Cd), and Pb (Adachi et al., 2004; Ellis & Revitt, 1982; Johansson et al., 2009). The vast
86 quantities of tyre rubber that is released on to the roads each year (estimated at 0.23 to 4.7
87 kg/year per capita in Kole et al., 2017) is cause for concern as it increases the mass of metal
88 rich roadside dust, and thus human exposure to inhalable and ingestible particulate matter.
89 Exhaust emissions from traffic contain harmful heavy metals such as mercury, nickel, cadmium
90 and lead from the burning of fossil fuels, and catalytic converters also degrade with time,
91 resulting in losses of platinum, palladium and rhodium, released onto the roadside in the form
92 of small high density particulate metallic dust (Johansson et al., 2009; Zereini et al., 1997).

93 Other sources of heavy metal pollution from traffic include the many potentially toxic
94 liquids used in vehicle maintenance. Antifreeze, oil, transmission fluid, power steering fluid,
95 brake fluid, and windscreen wash all leak from traffic sources onto roads. Metals present in
96 these fluids include chromium from coolants, nickel and zinc in motor oil and lead from
97 lubricating oil (Fox and Cramer, 1997). The general corrosion of automobiles also contributes
98 to heavy metal pollution. Aluminium (Al), Fe, Cr, magnesium (Mg), Ni, Zn, Cu and titanium
99 (Ti) are all used in car parts that degrade over time. Corrosion of the road itself can release
100 particulates containing heavy metals as asphalt contains bitumen which is known to contain
101 Ni, vanadium (V), Pb, Cr, mercury (Hg), and other potentially toxic elements (Legret et al.,

2005). Road works, abrasion of surfaces, weathering and frost damage loosens asphalt materials which in turn releases particulate matter containing heavy metals into the environment.

The potential increase in concentrations of heavy metal content in roadside areas presents with it several long-term environmental and health problems (Namdeo and Bell, 2005). As PM is mobile in the environment in both the atmosphere and aquatic systems, it can potentially transport heavy metal materials in road dust over long distances in the air (after initial release and re-suspension) and in moving water bodies via colloidal interactions (Frimmel et al., 2007; Karathanasis, 1999; Lee et al., 2018). These routes of transportation allow PTEs to disperse far from the roadsides given time, and contaminate nearby agricultural and residential areas where ingestion of heavy metals is likely to increase as a result of contamination of water supplies and crop uptake.

Major hosts for contaminant trace metals in roadside dusts and sediments are iron-rich spherical glass grains derived from high temperature combustion processes and iron oxide particles derived from the corrosion of steel (reported as > 25 % of the total mass of road dust in Taylor and Robertson, 2009). These materials have a high binding affinity for heavy metals and are easily mobilised into water sources by rain water. PTEs bound to particulates are cause for concern in drainage from urban areas as they can pollute drinking water and damage ecological systems (Davis and Birch, 2009). Once the heavy metal contaminant particles have deposited in sediment it is also very difficult to remove them without incurring large economic costs, therefore it is inevitable that most roadside areas will eventually end up with harmful concentrations of heavy metals if left long enough without remediation efforts (Mulligan et al., 2001).

This small scale study aims to examine a snapshot of the heavy metal content of roadside dust at six locations of varying traffic intensity in central Scotland, identifying the

concentrations of nine common heavy metals, chosen to represent the mechanical degradation of metallic parts, as well as tyre dust and combustion sources associated with traffic (aluminium (Al), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), magnesium (Mg), manganese (Mn), lead (Pb) and zinc (Zn)). The study also aims to quantify the metal content in varying particle size fractions, identifying where heavy metal content of smaller size fractions is higher as a result of the influence of traffic. Due to the uncertainty in current literature about the true risk of exposure to PTEs in regards to particle size, we use the < 20 µm size fractions in this study to identify potential human exposure to PTEs in terms of both inhalation and ingestion.

Methodology

Data in this study was analysed using the statistical software 'R' (R Core Team, 2017), and presented using the 'ggplot2' package (Wickham, 2016).

Sample collection

Approximately 500 g of roadside dust was taken from the surface 2 cm using a small trowel from 6 different sampling sites with varying environmental characteristics (Figure 1, Table 1). Samples were collected from each site (within a 5m radius) on a weekly basis (Fridays) for five weeks (n = 5), from 1st to the 29th of October in 2010. Protective gloves and face masks were worn during collection to prevent exposure to contaminated dust. Three of the sites chosen were in rural areas and three were chosen in urban areas. In both areas, samples were taken at low, medium (med) and heavy traffic sites, using local knowledge and Department for Transport: Road Traffic Statistics (for 2010: https://roadtraffic.dft.gov.uk) to distinguish these characteristics. The rural sites were located in West Lothian, approximately 13 to 17 miles west of the city of Edinburgh (UK). The urban area measurements were carried out in the southern part of the Edinburgh city district (UK). Post codes are provided to exclude exact measurement

sites for the privacy of local residents. Samples were collected approximately one metre from the kerbside of the road. None of the sites chosen in the study shared proximity (within 10 miles) to local sources of potential metal contamination other than traffic (i.e. incinerators or refineries, etc…), thus, in this study we assume that long term metal deposition in the roadside dust is not influenced significantly by nearby industry.

[Figure 1 near here]

[Table 1 near here]

Sample Preparation

Sub-samples of 400 g were weighed into aluminium trays and placed into a drying oven held at a constant 105 ℃. Dry sample mass was recorded after drying for 24 hours. The dried samples were then passed through a sequential sieve-stack (Endecotts Ltd. Stainless steel laboratory test sieve) with gradually decreasing pore diameters of 2 mm, 125 µm, 53 µm and 20 µm. The samples were dry-sieved until the majority of particles had been separated. Two litres of deionised water were passed through all stacked sieves to aid separation. The water and particulates passing through the 20 µm sieve was collected in a 2 L flask. The < 20 µm fraction was then passed through a 0.45 µm filter (Whatman Cellulose Nitrate Membrane Filters 0.45 µm) paper and the 0.45-20 µm particulate fraction was retained on the filter. After separation, each size fraction was put in an aluminium tray and placed in a drying oven overnight at a constant 105℃. The dry mass of each size fraction was recorded after it was dried. The dissolved fraction that had passed through the 0.45 µm filters was evaporated down to about 1 L. Aliquots (25 mL) of the solution were transferred into 3 replicate labelled 30 mL plastic tubes (Sterilin universal container, Scientific Laboratory Supplies Ltd). The volume of the remaining solution was measured before being discarded.

Approximately 1 g of each size fraction was ground into a fine powder using a mortar and pestle. Approximately 0.25 g of the ground sample was accurately weighed into a 50 ml

pyrex beaker. The beakers were covered with a watch glass and put in a furnace at 450℃ for 4 hours. The samples were allowed to cool prior to weighing. Loss on ignition and then percentage organic content were calculated to establish the ratio of mineral to organic material in the sample, and to investigate relationship with PTEs.

Microwave Digestion

Microwave digestion was carried out via MARS5 (CEM) microwave digestion apparatus using the methodology and variables as follows. Ashed samples were transferred into a clean microwave vessel (HP-500, CEM) and the remaining residue washed into the vessel from the glass beaker using 10 ml of concentrated HNO_3 (ARISTAR, 69% w/v, VWR Ltd.). In order to dissolve silicate materials in the sample, 1 ml of hydrofluoric acid (HF) was also added to the vessel (used as supplied, ARISTAR grade, 48% w/v, VWR Ltd.). Twelve soil samples, a blank and a reference material vessel were incorporated into each microwave run. The EPA3052H - HP500 method (#3052) was used to microwave the samples.

After the apparatus had cooled the vessels were removed from the carousel and vented with care. The solutions were transferred into clean Teflon beakers and the vessels rinsed with 1 - 2 ml of 2% ARISTAR HNO_3 to ensure complete sample transfer. The Teflon beakers were heated on a hot plate until around 1- 2 ml of solution remained. The flasks were made up to the 25 ml mark using 2 % (v/v) ARISTAR HNO_3. The contents of the flasks were transferred into labelled 30 ml plastic tubes and stored at room temperature for further analysis.

ICP-OES Analysis

The concentrations of Al, Cd, Cr, Cu, Fe, Mg, Mn, Pb and Zn in the digestates were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) using a Perkin Elmer Optima 5300 DV, employing an RF forward power of 1400 W, with argon gas flows of 15, 0.2

and 0.75 L min^{-1} for plasma, auxiliary, and nebuliser flows, respectively. Using a peristaltic pump, sample solutions were taken up into a Gem Tip cross-flow nebuliser and Scotts spray chamber at a rate of 1.50 ml min^{-1}. The selected wavelengths for each element were analysed in fully quantitative mode (three points per peak). After the analysis was completed the wavelengths which recorded the most accurate results for the reference materials and which gave the most consistent data for each element were selected for reporting results (Table 2).

[Table 2 near here]

A range of calibration standards were prepared using single element 1000 mg L^{-1} stock solutions (Al, Cd, Cu, Fe, Mg, Mn, Pb and Zn: Fisher Scientific UK LTD; Cr: Spectrosol Standard Solution, BDH Lab Supplies) diluted with 2% v/v HNO_3 (Aristar). Standard solutions of 0.1, 1, 10, 20 and 50 mg L^{-1} were prepared for each Al, Fe, Mg and Mn. Standard solutions of 0.01, 0.1, 0.25, 1 and 10 mg L^{-1} were prepared for each Cd, Cr, Cu, Pb and Zn. Using these standards, calibration lines were calculated to cover the range of concentrations found in the digestates.

Results

Particle size distribution

The particle size distribution of the individual samples from each location was dominated by the presence of larger particles (i.e. greater than 125 µm in diameter) (Figure 2). The 125 µm – 2 mm fraction contributes to the majority of the dry mass of most of the samples, with larger contributions from the > 2mm fractions in the rural areas when compared to urban. The mean ratio of the 0.45 – 20 µm fraction at the different sites ranges from 1.6 to 5.5 % of the dry mass, with the largest fraction at the high traffic urban location. The high traffic urban location had a

greater proportion of PM with diameter less than 125 µm than any other site, exceeding a quarter of the total mass of the samples.

[Figure 2 near here]

Organic matter content

The organic matter content (OM) at the different sites (bulk sample) was broadly similar (5.0 ± 2.4 % OM), with the exception of the urban low traffic location (19.7 ± 8.0 % OM), where samples were collected near a tree line at the edge of the road (Figure 3). With the exception of the urban low traffic location, a similar trend in OM within the different particle size ranges is observed, with OM approximately 5 times higher in the smallest particle size fraction when compared to the largest particle size fraction. Excluding the urban low traffic location, OM ranged between 15 to 25 % for the 0.45 – 20 µm size fraction and between 3 – 5 % for the > 2mm size fraction (0.25 g samples for all measurements).

[Figure 3 near here]

Metal content by size fraction

Nine metals in total were measured after microwave digestion of the road dust samples (Al, Cd, Cr, Cu, Fe, Mg, Mn, Pb and Zn) (Figure 4). Four of these (Al, Fe, Mg and Mn) are major soil constituents which we know to be present in soils (especially the clay fraction) in naturally high concentrations. In the case of Fe and to some extent Mg, a fairly uniform distribution of metal content is observed across the different measurement sites and throughout the particle size ranges. But, elevated concentrations in the 0.45 – 20 µm size fraction are evident for these metals when compared to the larger size fractions. Concentrations of Al are very high in the 0.45 – 20 µm size fraction, averaging 31.8 ± 5.6 g kg^{-1} when compared to the mean

concentration of the other size fractions 11.8 ± 4.1 g kg^{-1}. There is little observable difference between the metal content of Al, Fe and Mg at the sites with different traffic intensities or the urban/rural divide, suggesting that traffic and other local human activity is unlikely to contribute significantly to the total metal content at the sites.

Concentrations of Cd in the size fractions greater than 20 µm vary little between the sites, with a mean metal content of 0.38 ± 0.8 mg kg^{-1} across all sites. However, the Cd content of the 0.45 – 20 µm size fraction varies more significantly, with mean values ranging from 4.3 to 24.3 mg kg^{-1}. The highest Cd concentrations are observed at in the 0.45 – 20 µm size fraction at the low traffic rural location, suggesting a local specific source.

There seems to be a large source of Pb contamination at the medium traffic rural site, which skews the comparison between the different sites. Although this appears to be an outlier of sorts, repeated measurements from the same location (samples taken within 1 m of each other) show that the Pb content of the samples were consistently high. With the exception of the medium traffic rural site, samples from the other rural locations appear to contain less Pb than those measured in the urban locations. Concentrations of Pb were generally highest in the 0.45 – 20 µm size fraction, decreasing in concentration with increasing particle size, although there was some variability in the Pb content of the samples in the larger size fractions.

The Cu, Cr and Zn content of the samples follow the trend with traffic intensity more than the other metals, especially in the 0.45 – 20 µm size fraction (see Figure 4). For each of these metals, concentrations were much higher in the smaller size fractions, following a negative trend as particle size increased.

[Figure 4 near here]

277 Metal concentrations in the dissolved fractions of the samples were very low in
278 comparison with those in the solid size fractions (Figure 5). This can be attributed to the low
279 solubility of the metals in their various oxidation states and the near to neutral pH conditions
280 (see Table 1). The concentrations in the dissolved fraction do not correlate with the metal
281 content in the solid size fractions in the corresponding samples, with the exception of Cu. Cu
282 in the dissolved fraction appears to correlate with traffic intensity, as does the metal content of
283 the solid size fractions.
284
285 [Figure 5 near here]
286
287 ***Total metal content by location***
288 No temporal patterns were observed in the metal content of individual samples taken over the
289 five week period at each site (Figure 6). Although metal concentrations were generally highest
290 in the 0.45 – 20 µm size fraction, in most cases these particles contributed only a small fraction
291 to the overall metal content of the road dust as samples primarily consisted of particulates
292 greater than 125 µm in diameter (see Figure 2). The dissolved fraction contribution to total
293 metal content of the samples was negligible at all sites. In the cases of the high abundance
294 metals (Al, Fe, Mg and Mn) there were no observable trends between metal content and traffic
295 intensity, with differences in elemental composition most likely dominated by natural variation
296 between the sites.
297 Total Cd content was similar across the sites, with slightly higher concentrations at the
298 urban locations (rural mean = 1.98 mg kg^{-1} and urban mean = 2.46 mg kg^{-1}). The Pb
299 contamination at the medium traffic rural location is clearly observable in the results, with a
300 mean concentration of 250 mg kg^{-1} at this site when compared to a mean of 49 mg kg^{-1} at all
301 other sites (see Figure 6). Excluding this site, the concentrations of Pb in the urban sites were

higher than those at the remaining two rural locations (rural mean = 26.8 mg kg^{-1} and urban mean = 64.2 mg kg^{-1}, t-test p=0.09); however, there appears to be no direct correlation with traffic intensity.

Concentrations of Cu, Cr and Zn in the road dust correlates well with traffic intensity at the sites, with large differences between low and high traffic locations for each element. Concentrations of Cu exceeding 150 mg kg of Cu are measured on several occasions, with means of 55.0, 83.1 and 145.5 mg kg^{-1} for all low, medium and high traffic sites, respectively. This pattern is similar in the observed Zn concentrations, but there appears to be an additional effect caused by the rural/urban divide with concentrations at the high traffic intensity rural site being similar in magnitude to the low traffic urban site. Concentrations of Cr are higher at the medium and high intensity traffic sites than the low traffic locations; however there is little difference between high and medium sites.

[Figure 6 near here]

Relationship between OM and metal content

For several of the metals there are consistent trends between metal content and OM in the road dust (Figure 7). This relationship differs to some extent with the traffic intensity at each of the sites, with a larger metal to OM ratio at the high traffic sites. Due to the very high OM at the low traffic urban site and the relatively similar OM at the other sites, it is difficult to assess the direct effect of traffic intensity on this relationship; however, differences are most notable for Cu, Cr and Zn. The OM % is largest in the 0.45 – 20 μm size fraction, decreasing with increasing particle size (see Figure 3). Our previous results show that metal concentrations increase as particle size decreases at our sites (see Figure 4), thus OM % and metal content correlate well for most of the metals. However, our results cannot determine if high OM results

in accumulation of metals in smaller particles or if the two variables are correlated for other reasons (i.e. increased surface area interactions).

[Figure 7 near here]

Discussion

The metal concentrations of the road dust reported in this study are of the same order of magnitude of those reported in previous studies, although measurements carried out in large cities can be considerably higher than what was observed in the Edinburgh district. A summary of 15 studies reported by Charlesworth et al. (2003) showed that in large cities, Cd content varied from 1 to 6250 (but generally remained below 10) mg kg^{-1}, Cu varied from 69 to 1987 mg kg^{-1}, Pb varied from 60 to 2583 mg kg^{-1} and Zn varied from 152 to 3358 mg kg^{-1}. When comparing our results with these studies, and multiple more recent studies (e.g. Duong & Lee, 2011; Wei & Yang, 2010) we find that our observations are within the range of reported results in studies from locations with similar traffic intensities. As Edinburgh is a fairly small city (population < 0.5 million) and is not considered a heavy industrial polluter in comparison to the mega-cities of the world, it stands to reason that metal concentrations would be closer to the lower end of the scale when compared directly (Table 3).

[Table 3 near here]

The metals that appeared to be most influenced by traffic intensity in this study were Cu, Cr and Zn. This agrees with what we might expect from previous studies in which the highest concentrations of heavy metals in road dust were found at busy junctions where traffic was often stopped by traffic lights and crossings (Charlesworth et al., 2003). This is because brake

and tyre wear in these locations is much higher than open road, and thus deposition increases considerably. There will also be additional combustion emissions while the cars are stationary or moving slowly. In our study, the medium and high intensity traffic locations were all at junctions, which would explain the higher metal contents observed in comparison to the lower intensity site. An estimated 90 % of Cu emissions from traffic are as a result of brake wear (Johansson et al., 2009) which explains why copper correlated so well with traffic intensity in this study. Cu has been correlated with Zn and Cr in previous studies, highlighting the importance of brake wear on the deposition of these metals as well (Johansson et al., 2009).

Concentrations of Cd and Pb in our study did not correlate well with traffic intensity; however, there were differences observed between rural and urban locations (when excluding the Pb contamination at the medium traffic rural site). It has previously been reported that Pb concentrations are higher in urban areas when compared to rural locations (Aelion et al., 2012). These differences can be attributed to a number of factors, particularly legacy deposition (as unlike other pollutants, metals cannot degrade), and disturbance of soils that were affected prior to the banning of Pb additives in fuel in the year 2000, thus construction activity can result in re-release of Pb in dust and run-off. The larger number of construction projects in and around cities are a likely cause of elevated Pb in urban areas. The cause for the Pb contamination at the medium traffic rural site has not been identified in this study, but highlights that significant heavy metal contamination is possible in seemingly unexpected locations. As Cd contamination is typically associated with industrial activities and fossil fuel burning (Hutton, 1983) and in some cases mineral fertilisers (Roberts, 2014), it is not surprising that there was little difference in Cd content at the sites. Cd is known to be emitted from combustion sources (DEFRA, 2018); however, heavy metals released in this manner may disperse further from the roadside than the metal particulates associated with vehicle wear. This study does not

conclusively show a difference in Cd content of roadside dust as a result of traffic or of an urban/rural divide.

The metal content was highest in the 0.45 – 20 µm size fraction for all metals. This was also the case for the metals with a naturally high abundance (Al, Fe, Mg & Mn), suggesting that the high concentrations of metal in the small size fractions for the metals more influenced by traffic intensity (Cu, Cr, Zn) is not solely due to anthropogenic factors. The correlation between organic matter and heavy metal content is well documented in previous studies (Lin & Chen, 1998; Quenea et al., 2009; Yang et al., 2010), which have detailed the affinity for different metals in different sediment types and resultant changes in microbial carbon cycling. Due to the similarity of the organic matter size distributions at our sites (with the exception of the low traffic urban site), measurements suggest that the concentrations of Cu and Zn in the 0.45 – 20 µm size fraction are especially high as a result of traffic activity and not due to organic matter accumulation. This agrees with previous studies that suggest brake wear can result in large amounts of fine metallic dust gathering at busy traffic junctions (Charlesworth et al., 2003; Johansson et al., 2009).

Other factors to take into consideration that we cannot quantify in this study are the physical barriers/interactions at the different locations of the sampling. Road side areas are often lined with trees, buildings or other infrastructure such as barriers and bridges. These barriers (especially trees) may filter airborne particulates, altering deposition from traffic and surrounding areas (McDonald et al., 2007). The impact of fallen (deciduous) leaves are also likely to have impacted road dust abundance, absorbing particulates on the surface of the leaves which may be removed via street sweeping which would also have a major impact. Of the six sites sampled from, the most affected by these physical interactions would be the low traffic urban site with the nearby treeline and regular street sweeping; however, these conditions are

not abnormal and the road dust present is still an accurate assessment of the area during sampling.

Based on the EC Directive 86/278/EEC, the guideline threshold metal concentrations of sewage sludge (urban run-off) deemed safe for regular human exposure are presented in Table 4. Although not specifically used to determine the risks posed by the metal content of roadside dusts, this directive has been derived specifically to prevent harmful effects on soil, vegetation, animals and people (via ingestion of food grown on sludge-amended soil). We therefore use the guidelines as a proxy for environmental and health risks in this study as we do not have data to provide either the atmospheric concentrations or occupational exposure to the metals in question. The only metals in exceedance of these guidelines are Cu and Zn, the two metals which correlated strongly with traffic intensity. At the low traffic sites, metals fall well within the guideline values. The exceedance of these guidelines, paired with the high metal content in the smallest particle sizes highlights a potential health hazard at areas of high traffic intensity in urban and rural areas. The general public in Edinburgh (and the UK as a whole) are very unlikely to wear face masks (which may only offer limited protection anyway, Shakya et al., 2016) due to the relatively low atmospheric pollution in the city when compared to other global mega cities. This combined with the relatively large number of road cyclists in the city, likely means that heavy metal pollution is having some degree of negative health impact, especially when exposure to Cu and Zn is concerned. This exposure may also be higher for other metals not investigated in this study that are also present in tyres and brake pads (i.e. cobalt and nickel).

However, the seriousness of these health impacts, especially that of inhaled metal elements, remains poorly understood. Oral exposure to ingested metals such as copper, zinc and nickel is typically not considered a human health concern, and few studies have set out to explicitly identify the direct impacts on human health of low exposure to particular elements

425 that are not considered acutely toxic (such as arsenic, lead and cadmium) (Noonan et al., 2003;
426 Taylor et al., 2019; Genchi et al., 2020). As this study was limited in scale, further wide scale
427 work would be required to better identify the dangers that heavy metal pollution from vehicles
428 presents to traffic heavy populated areas in the UK. Future studies should also investigate
429 smaller size fractions where resources allow, such as PM_{10} and $PM_{2.5}$ that are more traditionally
430 associated with human health effects.

431 [Table 4 near here]

432

Conclusions

This snapshot study shows that the heavy metal content of road dust in the Edinburgh and West Lothian regions is generally below thresholds considered harmful to human health, although hotspots of contamination exist where traffic intensity is high and brake and tyre wear is greater than normal. Concentrations of Cu, Cr and Zn were found to correlate with traffic intensity in this study, with contamination considerably higher in both rural and urban environments where traffic intensity increased. Both Cu and Zn concentrations exceed recommended guidelines and are dangerously high in small particles (0.45 – 20 μm) which humans can ingest through inhalation or consumption, indicating areas of concern for pedestrians and cyclists. These concentrations are not as high as reported in many other larger cities with serious traffic pollution issues; however, as the long term effects of exposure to these metals is not fully understood, it is difficult to assess the true impact of this pollution on human health even in low doses. Future studies in this field should attempt to quantify human exposure more clearly, as measuring dust concentrations may not be the best way to directly quantify this. Further research identifying the risk of exposure to different particle sizes > PM_{10} would also clarify the true risk posed by elevated concentrations of PTEs in road dust.

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Declarations

Ethics approval and consent to participate

Not applicable.

Consent to participate and publication

Not applicable.

Author Contributions

N. Cowan carried out the field work, sample collection, data analysis and writing of the manuscript. D. Blair assisted in sample preparation and carried out analysis of metal concentrations using ICP-MS instrumentation. H. Malcolm assisted with writing of the manuscript, and contributed to the discussion section of the study. M. Graham supervised the logistics and scope of the study, analysis laboratory and manuscript preparation. All co-authors assisted in the production of the manuscript.

Availability of data and materials

Data will be submitted to the Environmental Information Data Centre (EIDC) upon publication of the study, and will subsequently be freely available in the public domain.

Competing Interests

The authors declare that they have no competing interests.

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667

668 **Table 1** Samples of roadside dust were taken from three rural and three urban locations in the
669 Edinburgh and Lothians area in October 2010. Average daily flow of traffic is presented where
670 nearby manual count point data is available (from https://roadtraffic.dft.gov.uk).

Location/ Traffic	No. of Vehicles per day	Soil pH (H_2O)	Postcode	Description
Rural				
Low	< 1000	6.7 ± 0.1	EH52	Small country B-road surrounded by arable fields
Med	4836	6.8 ± 0.1	EH49	Busy rural B-road junction near cottages and arable fields
High	17511	6.1 ± 0.1	EH52	Busy A-road junction with traffic lights, nearby houses
Urban				
Low	< 1000	6.0 ± 0.1	EH16	Small backstreet in residential area with tree line next to road
Med	19807	6.3 ± 0.1	EH16	Busy urban junction in town near residences
High	65535	6.5 ± 0.1	EH20	Very busy motorway interchange

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672

673 **Table 2** Selected ICP-OES Analysis Wavelengths used for concentration determination

Al 394.401 nm	Cu 327.393 nm	Mn 259.372 nm
Cd 228.802 nm	Fe 238.204 nm	Pb 220.353 nm
Cr 267.716 nm	Mg 285.213 nm	Zn 213.857 nm

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675

676 **Table 3** Concentrations (mg /kg) of metal content in road dust reported in previous studies

Study	Source	Cd	Cu	Pb	Zn
This study	Edinburgh, UK (Rural)	3.3	81.5	101.4	112
This study	Edinburgh, UK (Urban)	4.1	107.6	64.2	268
Miazgowicz et al., 2020	Krakow, Poland (Urban)		192	51	428
Nakajima et al., 2018	Tokyo, Japan (Urban)		259	271	1827
Adamiec et al., 2016	Katowice, Poland (Urban)	0.35	239	430	2030
Suryawanshi et al., 2016	Delhi, India (Urban)	2.65	191.7	120.7	285.5
Li et al., 2001	Hong Kong (Urban)	3.8	173	181	1450
Fergusson et al., 1984	London, UK (Urban)	6.5	197	3030	1174
Miguel et al., 1997	Madrid, Spain (Urban)		188	192.7	476
Fergusson et al., 1984	New York, US (Urban)	8.0	335	2583	1811
Chon et al., 1995	Seoul, Korea (Urban)	3.0	101	245	296

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679 **Table 4** The range of metal contents observed in this study are compared to EC Directive
680 86/278/EEC guideline thresholds for sewage sludge and urban run-off deemed safe for regular
681 human exposure. The range of mean values at each site are used (n = 5). Thresholds vary by
682 pH for some metals, thus we compare our observations to the pH range that best matches our
683 measurements.

Metal	EC Directive 86/278/EEC threshold (mg kg^{-1})	Range of observations in this study (site means) (mg kg^{-1})
Cd	3.0	1.9 – 2.7
Cr	400	16 – 50
Pb	300	17 – 250
Cu (at pH 6 – 7)	135	37 – 152
Zn (at pH 6 – 7)	300	48 – 388

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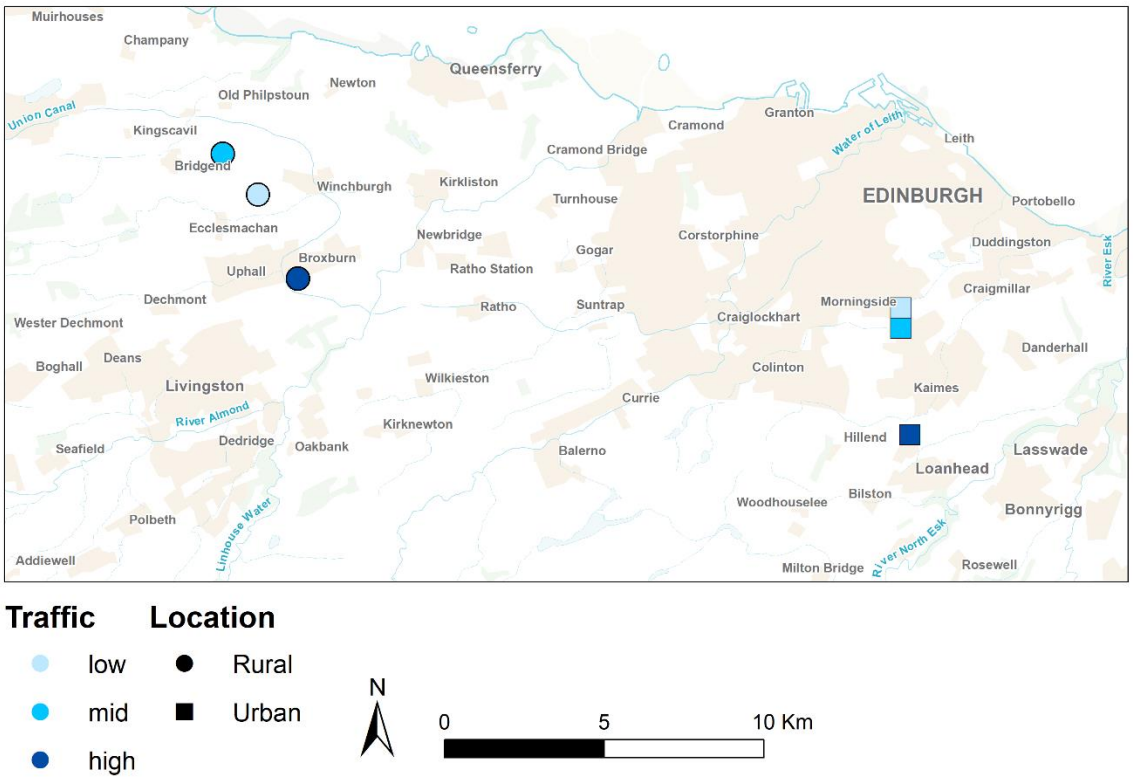
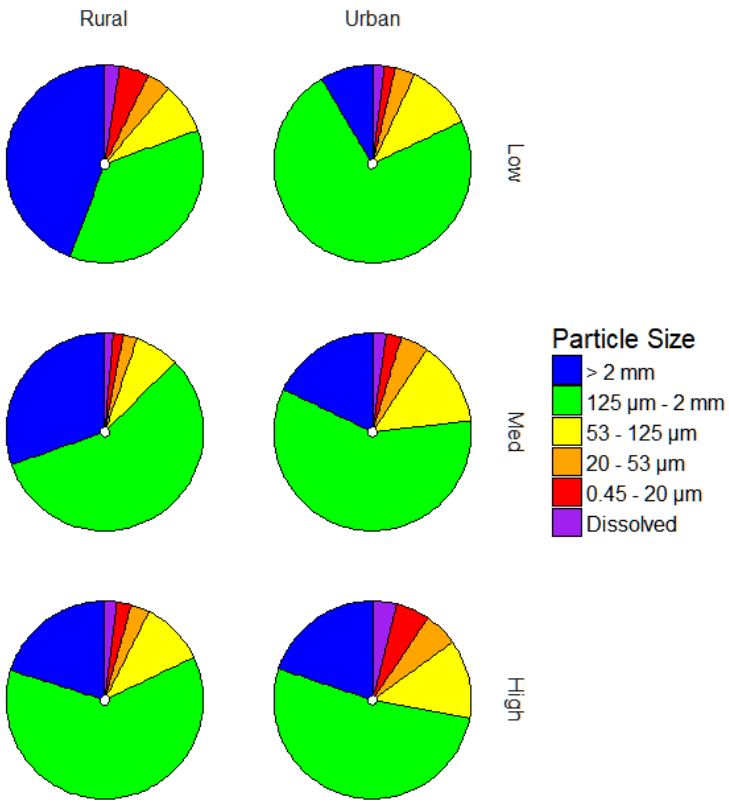


688

689 **Figure 1** Samples were collected from six sites. The samples sites classed as Urban were
690 situated in the south of the city of Edinburgh, while the Rural sites were sampled from the West
691 Lothian region to the west of the city. Locations classed as low, medium and high traffic were
692 chosen for both Urban and Rural areas based on local knowledge and traffic data.

693



694

695 **Figure 2** The mean particle size distribution of road dust collected at six sites in central
696 Scotland, separated manually by sieve (n samples = 5). Three sites were chosen in each rural
697 and urban locations, with low, medium and high traffic intensity.

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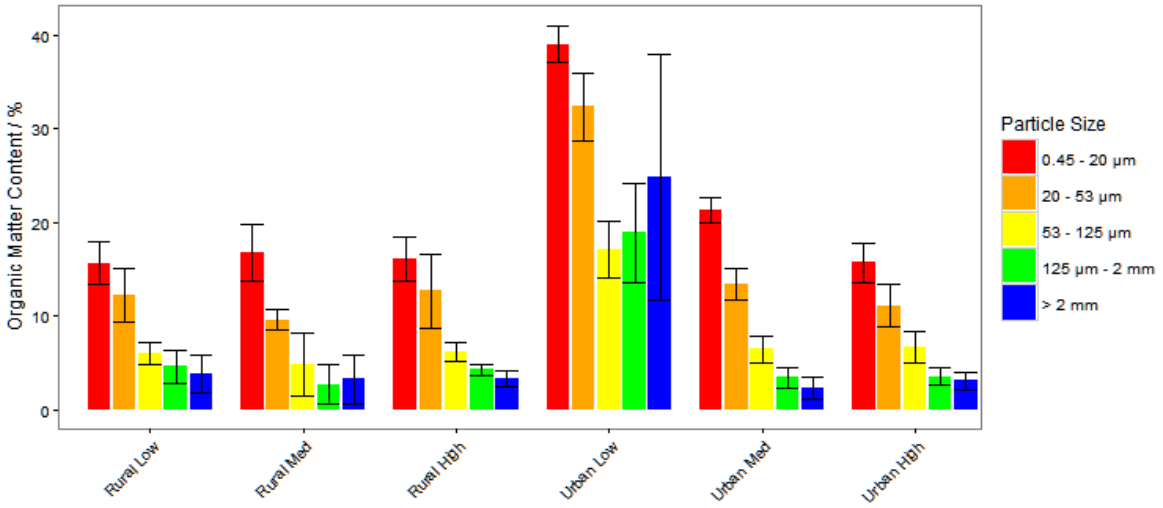

699

700 **Figure 3** Mean organic matter content of road dust samples after particle size separation,

701 calculated by loss on ignition (Error bars denote standard deviation of samples, n = 5).

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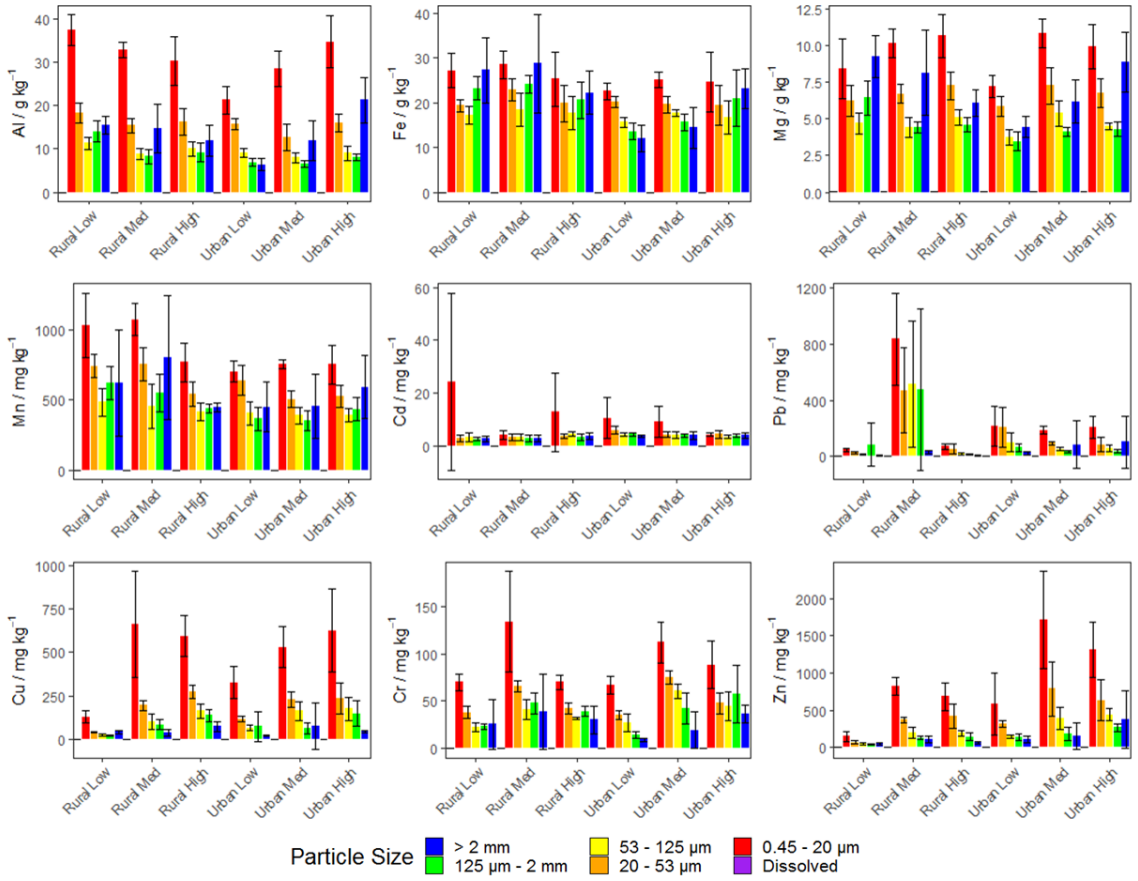

704

705 **Figure 4** Mean metal content of each of the size fractions at the six measurement sites. Standard

706 deviation of n = 5 samples.

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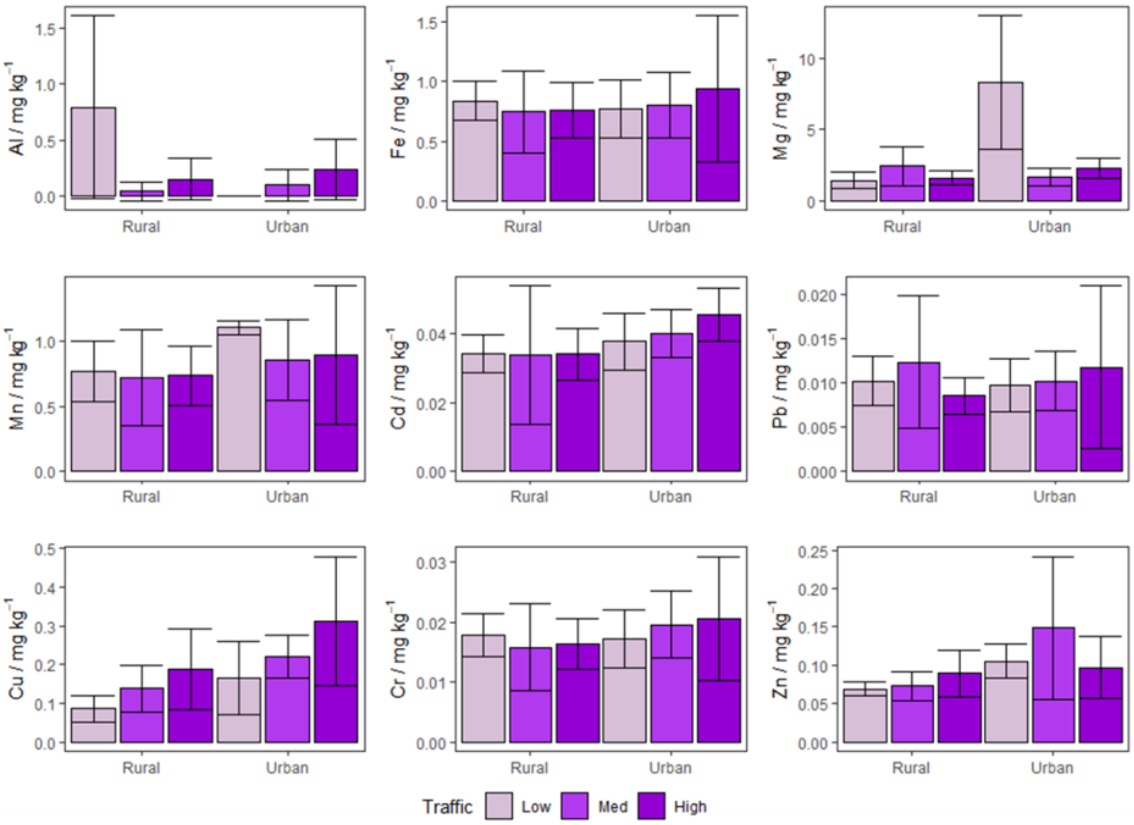
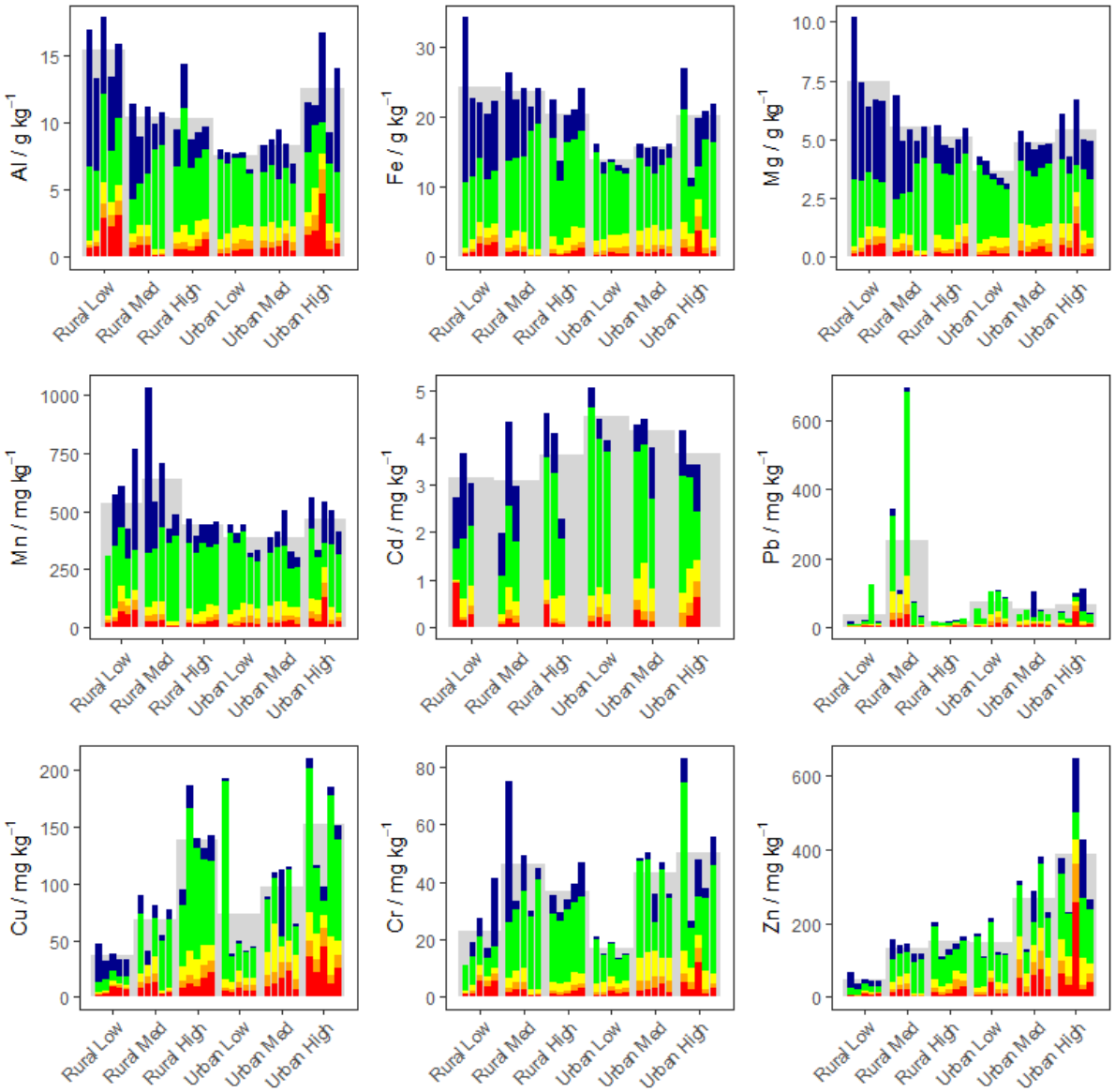


708

709 **Figure 5** Metal content of the dissolved fraction of road dust samples, extracted using deionised
710 water only (2 litres), with concentration expressed as mg of metal present per kg of sample
711 sieved. Standard deviation of n= 5 samples included.

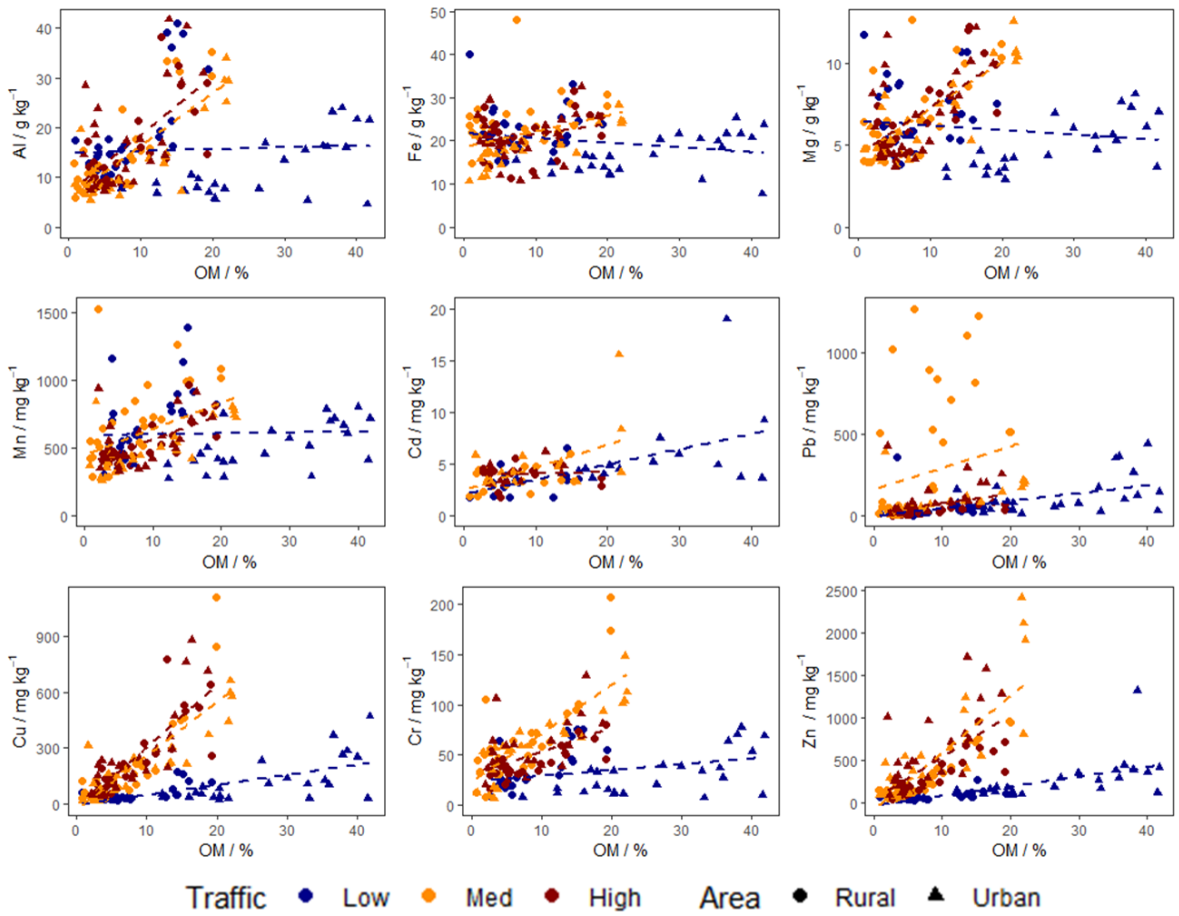
712



713

714 **Figure 6** The total metal content of each sample collected from six locations. Contributions
715 from each size fraction are separated, with mean metal content for each site included (grey
716 shaded).

717



718

719 **Figure 7** Correlation between metal content and organic matter content (OM) of each separated
720 size fraction from each road dust sample. Linear regression is fit between combined urban and
721 rural site data, grouped by traffic intensity (dashed line).