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Contaminated urban road deposited sediment (RDS), Greater Manchester, UK: a spatial assessment of potential surface water impacts

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Abstract

This paper presents data on spatial variability in urban road-deposited sediment (RDS) composition (HCl-digestable Mn, Cu, Pb and Zn, H₂O-leachable sulphate, nitrate and chloride) across the centre of the Greater Manchester conurbation. Given the importance of RDS to surface water quality within the urban environment, an understanding of the spatial variability in RDS composition is an important component of managing RDS and associated pollution.

Road-deposited sediment (RDS) was sampled on a five-day period in June 2003 over a 5km by 5km area of Manchester and Salford City centres, at a sampling density of 0.25km² (100 samples in total). The highest levels of accumulated sediment were recorded away from the city centres, probably as a result of regular removal of RDS by street sweeping in the city centres. HCl-extractable Fe, Mn, Zn, Cu and Pb, and H₂O-leachable chloride, sulphate and ammonium, all displayed high levels of variability across the sampled area. In general, Pb and Zn displayed high levels in the centres of Manchester and Salford, probably reflecting increased traffic density, with lower values outside these areas. Iron and Mn were highest away from the city centre, possibly suggesting derivation from soils and other geogenic sources that are rare in the city centres. Cu distribution was distinctive from both Pb and Zn, and Fe and Mn. Whilst Cu has previously been sourced to vehicle activity, the data for Manchester RDS suggest additional, unidentified, sources. H₂O-leachable chloride, sulphate and ammonium all showed similar distribution patterns, with high values in the centres of Manchester and Salford, and low values outside the centres. Sulphate and ammonium probably derive from wet and dry deposition, ultimately derived from vehicular and industrial processes. Chloride is presumed to be derived from road-salting. The presence of high levels of these soluble components indicates that RDS should be expected to have an impact upon surface water quality, and their chemical characteristics should be considered in surface water quality management. The recognition of spatial variability in the RDS in this city suggests that such data should be routinely collected to aid in the effective implementation of urban pollution management strategies based upon contaminant source control.

Keywords

Manchester, Salford, Road deposited sediments, Pollution, Urban, Metals

Introduction

Road-deposited sediments (RDS – also commonly referred to as ‘street dust’ or ‘road dust’) are an accumulation of particulates on surfaces such as pavements and road surfaces in urban environments. RDS are a complex mix of particulates and contaminants derived from an extensive range of urban and industrial sources and processes. Sources are either anthropogenic in nature (including vehicle exhaust emissions, vehicle tyre and body wear, brake-lining material, building and construction material, road salt, atmospheric deposition), or naturally-derived (soil material, plant and leaf litter, and atmospheric deposition) (Hopke *et al.* 1980; Beckwith *et al.* 1986; Xie *et al.* 1999; Lecoanet *et al.* 2003; Robertson *et al.* 2003).

RDS has been documented to carry a high loading of contaminants including metals (Fe, Cu, Zn, Pb, Mn, Cr, Cd) and organic pollutants (Farmer and Lyon 1977; Harrison 1979; Beckwith *et al.* 1986; Stone and Marsalek 1996; Kim *et al.* 1998; Yunker *et al.* 2002). RDS, and their associated contaminant load, are ubiquitous in urban and suburban drainage basins and are typically available for mobilisation and transportation to subsurface drainage systems by stormwater runoff, thus playing a critical role in degrading receiving water bodies.

Given the importance of RDS to surface water quality within the urban environment, an understanding of the spatial variability in RDS composition is an important component of managing RDS and associated

pollution. A limited number of studies (Linton *et al.* 1980) have documented the small scale variability of metal concentrations on street surfaces that may be present, whilst it has also been documented that Pb levels are commonly lower in outer city locations compared to inner city sites, indicating the role that vehicle traffic density plays in determining the spatial pattern of this contaminant (Duggan and Williams 1977; Massadeh & Snook 2002; Robertson *et al.* 2003). Such inner and outer city patterns have also been reported for urban soils (Madrid *et al.* 2000). There has been a paucity, however, of systematic spatial analysis of RDS composition over the scale of a city. A recent spatial survey of Birmingham and Coventry, UK (Charlesworth *et al.* 2003) documented significant city-scale variability, with contamination hotspots evident. An understanding of the city-scale spatial variability of RDS composition is a critical requirement for two reasons. First, urban pollution management strategies consider the city-scale as the most appropriate unit for pollution control. Second, with respect to surface water quality the river basin-scale (generally equal to or larger than the city-scale) is considered the most appropriate scale to consider (Owens *et al.* 2004). Therefore, an increased understanding of the variability of RDS composition, and its sources, is a pre-requisite for urban pollution management; data that are largely lacking.

The aim of the research presented here was to determine the spatial variability in RDS composition (HCl-digestible Mn, Cu, Pb and Zn, H₂O-soluble sulphate, nitrate and chloride) across the centre of the Greater Manchester

conurbation in north west of England. The study here focuses on the coarser fraction of the RDS, which is likely to impact surface water quality, (in contrast to finer particulates less than 10µm in size which have an impact upon air quality). The number of major waterways running through Greater Manchester means that such data are required to inform surface water quality management.

Methods

Study area and sample locations

The conurbation of Greater Manchester is extensive, contains major rail and road routes, and impacts heavily upon river catchments in the area. The location of sample sites and sampling density were predetermined using a random stratified sampling strategy approach. The area studied covered the central Manchester and Salford; a sampling density of four samples per 1km² in a 25 square km section was chosen, resulting in 100 sample sites (Fig. 1). The area sampled is characterised by a range of urban environments, including relatively quiet to busy central city streets, busy commercial shopping streets, major urban thoroughfares, quiet residential areas close to busy thoroughfares, light industrial areas and busy, main thoroughfare junctions. Within this paper we consider "city centre" to apply to those areas where commercial retail activity is concentrated, and traffic is slow flowing. The areas outside these are dominated by light commercial, light industrial and residential areas with relatively fast flowing traffic.

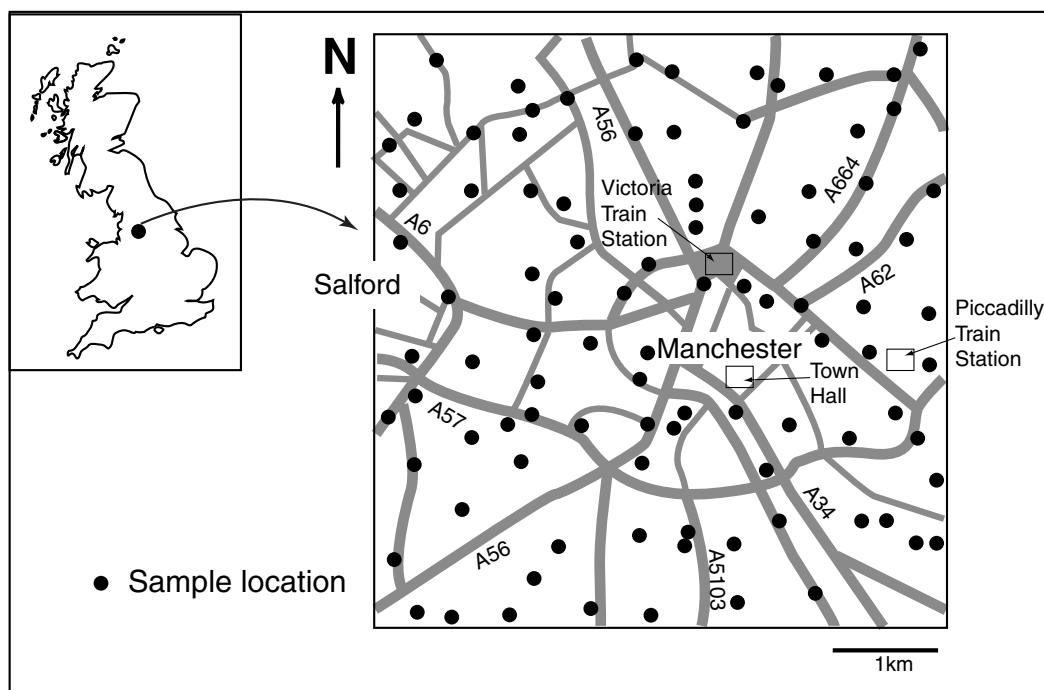


Figure 1: Location map of the study site, showing the location of individual samples taken in the study.

Sample collection

Sample collection took place over five consecutive days in June 2003 after a two-week dry spell of weather to enable adequate quantities of accumulated sediment for a multi-analytical investigation. This allowed for an analysis of the spatial variability, but does not allow for an assessment of the temporal variability. Several methods have been successfully applied by researchers to collect urban street dust samples including battery-powered vacuum cleaners (Grottker, 1987), small vacuum sweeper trucks (Rogge *et al.* 1993; Yang & Baumann 1995), and by using a clean polyethylene dustpan and brush (Kim *et al.* 1998; Xie *et al.* 1999; Vermette *et al.* 1991). Although this method may bias against smaller particles compared to the use of a vacuum cleaner (Bris *et al.* 1999), previous observations have shown that RDS collected from Manchester are made up primarily of medium sand-sized particles (Robertson *et al.* 2003). Consequently, the utilisation of a dustpan and brush was chosen as a suitable sampling method for the research presented here. Samples were taken from a constant area (1 m²) of the curbside of the road surface.

Sample analysis

Air-dried samples were sieved to remove the greater than 2mm size fraction and homogenised. Sub-samples of all <2mm samples (n=100) were digested for metal analysis. All samples were digested in 0.5M HCl for one hour at room temperature (methods described in Sutherland *et al.* 2004), which has been shown to be useful in determining the bioavailable portion of metals in sediments (Luoma and Bryan 1982; Sutherland 2001). In addition to <2mm sediment samples, samples were dry-sieved to obtain the <63µm size fraction. This size fraction has been shown to be a major carrier of contaminants in sediments, due to the increased surface area (Horowitz 1991), and it was separated here to test for the level of contaminant loadings in the fine-grained fraction of these RDS. Due to the coarse nature of the sediment, only 50 samples provided enough <63µm sample for metal analysis. These <63µm were digested in 0.5M

HCl in the same manner as for the <2mm samples. Metal contents (Fe, Mn, Cu, Pb, Zn) for all digests were determined on resulting solutions using a Unicam 929 atomic absorption spectrometer (AAS). The accuracy of the extraction procedure was determined by analysing certified reference material (stream sediment GBW07311), and was found to be within 10%. H₂O-leachable chloride, sulphate and ammonium were determined using ion chromatography to approximate to that component soluble in surface water runoff. Organic matter content was estimated by loss on ignition at 450°C (Charlesworth and Lees 1999; Xie *et al.* 1999; Robertson *et al.* 2003). Low field magnetic susceptibility (χ_{lf}) was measured on all samples (<2mm fraction). Magnetic susceptibility has been shown to aid in determining source apportionment (Robertson *et al.* 2003), and has also been proposed as a rapid proxy method for metal levels in sediments (Berry and Plater 1998). Spatial distribution maps were generated for geospatially-referenced data using SurferTM. We stress here that these maps are presented solely to display spatial patterns in compositional variability, and are not used for interpolation and prediction of unknown values between sample points.

Results

RDS composition

The physical characteristics and composition of the RDS are summarised in Table 1. The mass of RDS recovered from the street surfaces was highly variable (7.3 to 740 gm⁻²), as was percent mass <63µm size fraction in the RDS (0.5-20.8%). Loss-on-ignition values range from 3.8-22.6%. The metal contents removed by the 0.5M HCl leach all display high ranges, with means for Cu, Fe, Mn, Pb and Zn of 88, 2005, 170, 164 and 268 µg/g respectively. H₂O-leachable sulphate, chloride and ammonium display similar wide ranges in values (means of 152, 66 and 76 µg/g⁻¹). Levels of Fe, Mn, Pb, Cu and Zn are significantly higher in the <63µm than in the <2mm size fraction (Table 2; significant to 0.01 level in Wilcoxon ranked pairs tests).

Table 1: Summary data for RDS <2mm sample composition.

	Mass (g/m ²)	% mass <63µm	Loss on ignition (%)	Cu (µg/g)	Fe (µg/g)	Mn (µg/g)	Pb (µg/g)	Zn (µg/g)	H ₂ O-leachable Cl (µg/g)	H ₂ O-leachable SO ₄ ²⁻ (µg/g)	H ₂ O-leachable NH ₄ ⁺ (µg/g)	χ_{lf} (10 ⁻⁶ m ³ kg ⁻¹)
Mean	96.3	6.0	7.8	88	2005	170	164	268	66	152	76	4.25
Range	7.3-740	0.5-20.8	3.8-22.6	14-342	438-5251	63-856	45-1461	65-990	5-776	19-2139	20-412	1.19-18.34

Table 2: 0.5M HCl leach metal concentration ($\mu\text{g g}^{-1}$) for RDS $<63\mu\text{m}$ fraction.

Metal concentration	Cu ($\mu\text{g/g}$)	Fe ($\mu\text{g/g}$)	Mn ($\mu\text{g/g}$)	Pb ($\mu\text{g/g}$)	Zn ($\mu\text{g/g}$)
Mean	181	3438	293	263	394
Range	25-1532	772-7398	128-495	25-1496	106-1212

RDS Spatial Variability

The spatial variability in RDS composition across the city of Manchester is shown in Figures 2, 3 and 4. Both mass of sediment recovered per m^2 of road surface and percent of the sediment $<63\mu\text{m}$ in size show a similar distribution pattern, with generally higher values outside Manchester and Salford City centres, and low values in the centres (Fig 2). Neither loss-on-ignition or magnetic susceptibility data display systematic patterns of distribution (Fig. 3).

Of the metals, only Zn and Pb possess significant correlation ($r = 0.454, P < 0.001$), with weaker, but significant,

relationships between Cu and Zn ($r = 0.329, P < 0.01$), and Fe and Mn ($r = 0.224, P < 0.05$). As a consequence, Pb and Zn display similar distribution patterns, with generally higher levels in the city centres (Fig. 3). Iron, and to a lesser extent Mn, tends to display higher levels outside the centres, whilst Cu displays highest levels to the south of Manchester City centre (Fig. 3). H_2O -leachable chloride, sulphate and ammonium all display high values in the city centres, with low values away from the centres (Fig. 4), and all possess highly significant correlations with each other.

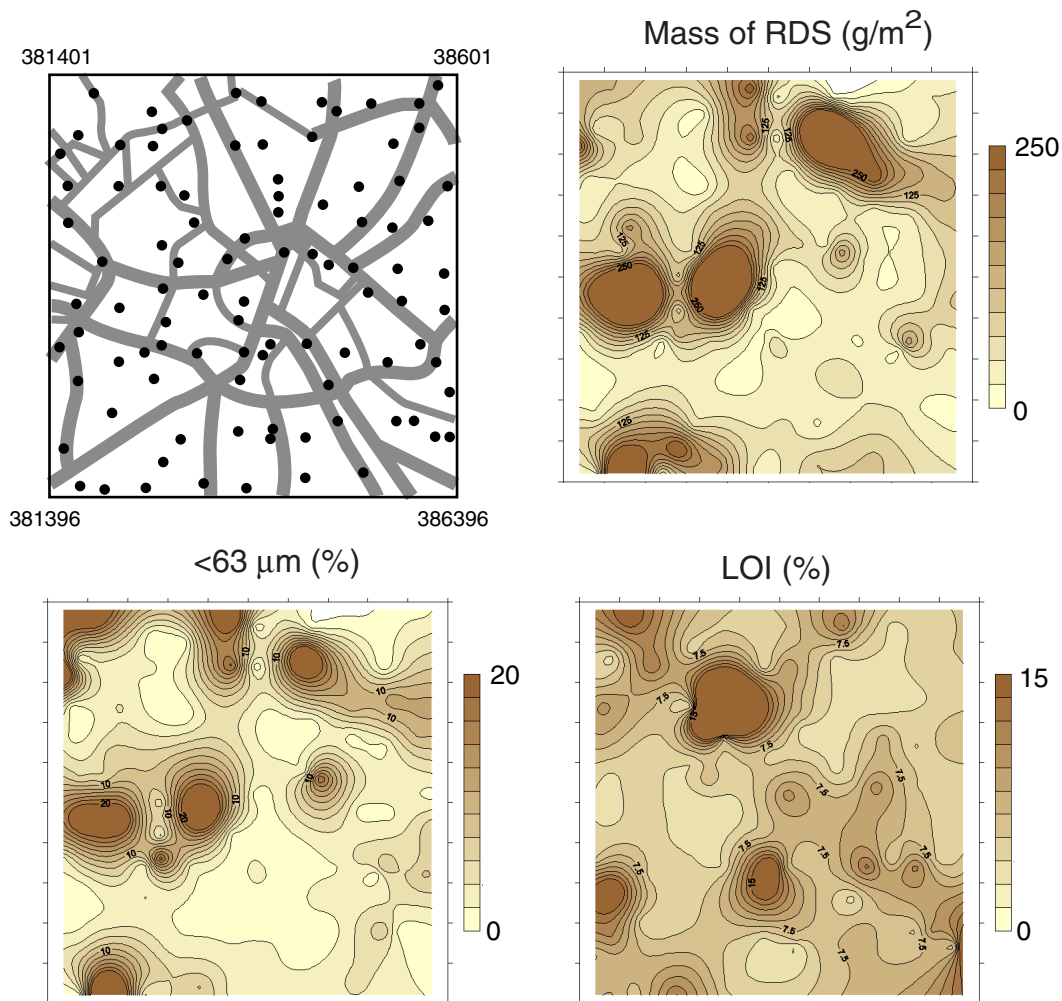


Figure 2: Spatial distribution of RDS sample mass recovered, proportion $<63\mu\text{m}$, and loss-on-ignition values.

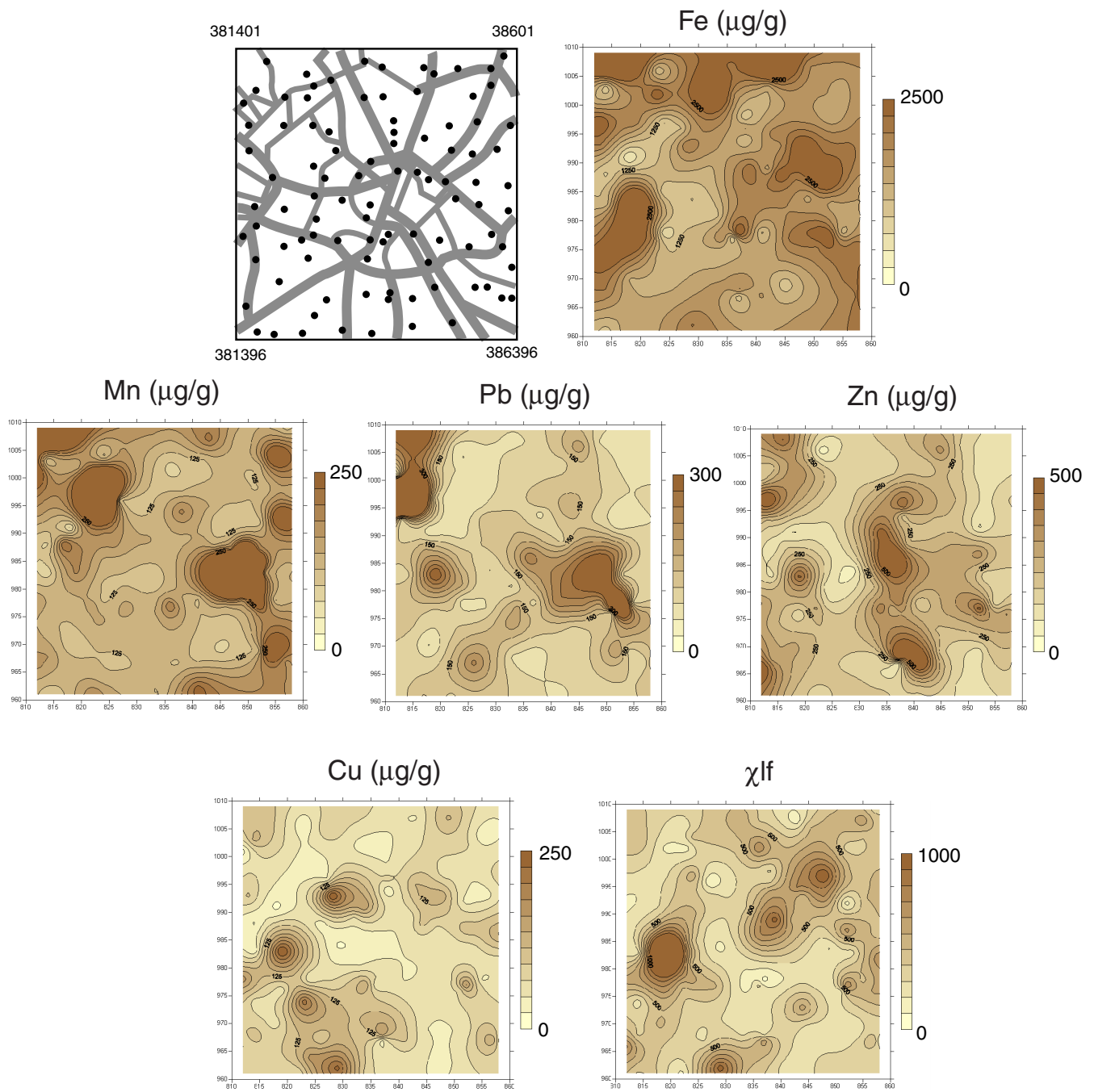


Figure 3: Spatial distribution of Fe, Mn, Pb, Zn, Cu and magnetic susceptibility for RDS samples.

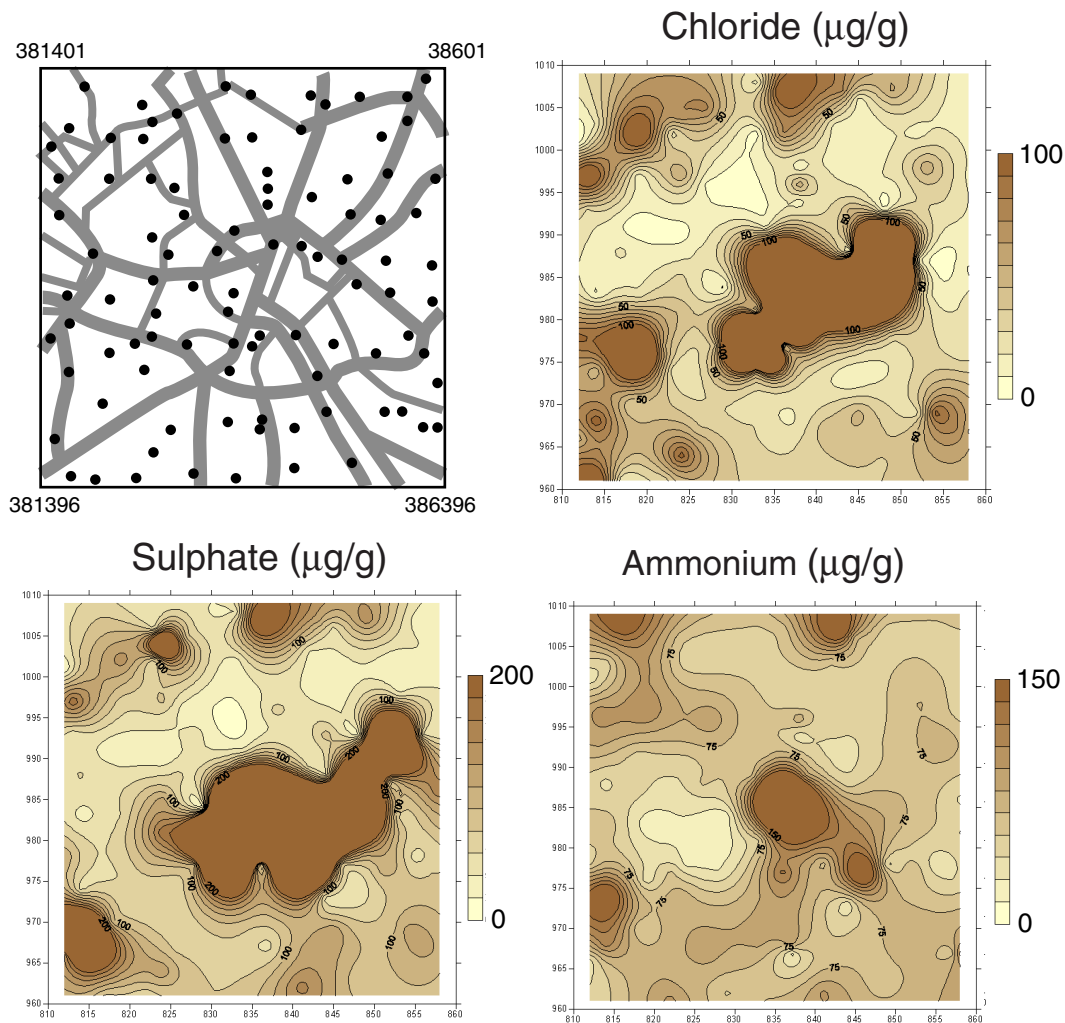


Figure 4: Spatial distribution of H₂O-leachable chloride, sulphate and ammonium for RDS samples.

Discussion

Physical Characteristics

There is a high degree of variability in the mass of sediment recovered at different sample sites, from 7.3 to 740 gm⁻². These variations will represent an interplay between those processes responsible for accumulation and removal of particulates. Within the urban environment there are several mechanisms that will remove RDS from street surfaces, including wind, surface water runoff and street sweeping. Given similar climatic conditions across Greater Manchester, the most likely controls on RDS accumulation are extent of sediment sources and frequency of street sweeping. The former of these is hard to assess. For aesthetic reasons street sweeping takes place much more regularly in the city centres and, therefore, significant sediment accumulation can only take place away from these areas. We, therefore,

conclude that this is the most important control on this data set. Loss-on-ignition values display a high range (3.8–22.6%). The lower end of this range is similar to values reported in previous studies of RDS (Fergusson and Ryan 1984; Al-Chalabi and Hawker 1996; Xie *et al.* 1999; Robertson *et al.* 2003), but the higher end of the range is significantly higher than in these published studies. Organic matter is generally higher in the city centre (Fig. 2), suggesting an anthropogenic component, but the specific sources are undetermined.

Metal levels and distributions

Whilst numerous data sets have been published on metals in RDS, different studies have used different digestion methods, thereby limiting the direct comparisons that can be made to this study. This is particularly true for the HCl leach method utilised here. Sutherland (2002), using a single

step 0.5M HCl digest on soils and RDS in an urban catchment in Hawaii, reported similar levels of Pb, Zn and Cu to this study, but higher levels of Mn and Fe. However, that study included soils, which would be expected to have higher levels of Fe and Mn than in this study, which did not include soils. The finding that metal levels are significantly higher in the <63 μ m size fraction is consistent with other published studies on urban RDS (Sutherland 2003). However, there is no significant paired correlation between metal levels in the <2mm samples and the weight % of 63 μ m size fraction in the samples. Therefore, spatial variations in contaminant concentrations of the samples (see below) are not a result of variations in grain-size, but real variations in contaminant levels.

Previous study of the Manchester RDS (Robertson *et al.* 2003) documented that Pb levels were highest in inner city sites, an observation reinforced by the larger data set presented here (Fig. 3). Such patterns of Pb distribution have been explained by the increased density of traffic in city centres (Farmer and Lyons 1977; Duggan and Williams 1977; Hamilton 1984), and we interpret the patterns in Manchester to be explained similarly. In the UK, the reduction, and then discontinuation, of leaded-petrol has led to reductions of RDS-Pb loads in recent years (Massadeh and Snook 2002). The presence of significant Pb levels in the RDS samples in this study suggest that there are other sources of Pb than fuel combustion, possibly redistributed historical soil pollution. The distribution of Zn in this study (Fig. 4), similar to that of Pb, suggests similar sources. Zn has been suggested to have been derived from tyre wear (Hopke *et al.* 1980; Kim *et al.* 1998), consistent with this interpretation. Cu distribution is distinctive from Pb and Zn (Fig. 3). Whilst Cu has been sourced to vehicle activity, coming from corroded car bodywork (Beckwith *et al.* 1986), the data for Manchester RDS suggest additional, unidentified, sources. Iron and Mn display similar spatial distribution to each other, but distinct from other metals (Fig. 3). Iron and Mn have been documented to have been derived from a large number of sources, including geogenic background sources (Hopke *et al.* 1980; Linton *et al.* 1980). In this study, they are highest away from the city centre, possibly suggesting derivation from soils and other geogenic sources that are rare in the city centres.

Some studies have suggested that magnetic susceptibility may be a quick and easy proxy for metal contamination in aquatic sediment (Berry and Plater 1998; Chan *et al.* 2001; Perry and Taylor 2004). Magnetic susceptibility in this study, however, displays a spatial distribution that contrasts to metals, suggesting that it is not a good measure of metal

contamination in these sediments. This is probably due to the mineralogy of RDS. Robertson *et al.* (2003) concluded that the magnetic mineralogy of RDS is derived from combustion processes, rather than from soils. Furthermore, Robertson and Taylor (in review) document that a significant proportion of the metals in RDS are hosted in industrial glass grains and iron oxide fragments derived from vehicle wear. Therefore, it is unsurprising that mineral magnetic measurements in the RDS do not act as a proxy for metal levels, as the metals and mineral-magnetic components are derived from different sources.

H₂O-leachable chemical species.

High levels of H₂O-leachable chloride, sulphate and ammonium have been observed in this study. Such components have not been reported from RDS previously, and so comparison to published data is not possible. However, surface water quality in urban areas has been studied extensively, and the effect of readily mobilised chemical species has been clearly documented; the 'first-flush' associated with rainfall events (Morrison *et al.* 1984; Lee and Bang 2000). The high levels of H₂O-leachable Cl⁻, SO₄²⁻ and NH₄⁺, clearly indicate the presence of soluble phases within the Manchester RDS. The application of de-icing salt (NaCl) is the likely source of soluble chloride to RDS (Norrstrom and Jacks 1998). Sulphate and ammonium are most likely derived from atmospheric deposition onto street surfaces, and is related to vehicular and industrial emissions (Samrani *et al.* 2004). The same authors also report barium sulphate particles in urban runoff sourced from automobile brake material and road paint. The clear spatial association to the city centres for all soluble species in this study suggests a vehicular source.

The presence of soluble chloride mineral phases in RDS has been previously documented, and these may also play a role in metal complexing (Lumsden *et al.* 1995; Serrano-Belles and Leharne 1997). The significant levels of leachable components in the RDS suggest that these sediments can have significant impacts upon surface water chemistry. The management, through removal and minimisation of these sediments on street surfaces should, therefore, form a significant component of urban pollution management. Further mineralogical and chemical study is needed to understand the nature of soluble species in RDS.

This study has illustrated that there is significant spatial variability in RDS contaminant levels across the centre of Manchester. This data set was taken from a single 5-day period in time, which minimised temporal variation. Seasonal variations in RDS have been documented by

Vermette *et al.* (1991), the high seasonal variability being associated with the anthropogenic source materials contributing to RDS. Seasonal variability has been suggested to be controlled by climate (Vermette *et al.* 1991; Fergusson and Kim 1991) specifically, the number of dry days, rainfall wash off and prevailing wind direction, all of which contribute to the extent of washing of sediments and the dominant source materials. These studies documented that concentrations of Pb fell due to the washing of sediments and concentrations of Zn rose due to the washing of galvanised roofing. Monitoring data, both spatial (which have been monitored in the present study) and seasonal, are required in order to gain a better representation of major polluting episodes that remain unaccounted for within spot sampling regimes. Such monitoring data would lead to a better understanding of RDS quality and associated urban pollution.

Conclusions

First, road-deposited sediment (RDS) was sampled on a five-day period over a 5km by 5km area of the centres of Manchester and Salford, at a sampling density of 0.25km² (100 samples in total). The highest levels of accumulated sediment were recorded away from the city centres, probably as a result of regular removal of RDS by street sweeping in the city centres.

Second, for the RDS samples, HCl-extractable Fe, Mn, Zn, Cu and Pb, and H₂O-leachable chloride, sulphate and ammonium, all displayed high levels of variability across the sampled area. In general, Pb and Zn displayed high levels in the centres of Manchester and Salford, probably reflecting increased traffic density, with lower values outside these areas. Iron and Mn were highest away from the city centre, possibly suggesting derivation from soils and other geogenic sources that are rare in the city centres. Cu distribution was distinctive from both Pb and Zn, and Fe and Mn. Whilst Cu has previously been sourced to vehicle activity, the data for Manchester RDS suggest additional, unidentified, sources.

Third, magnetic susceptibility displays a spatial distribution that contrasts to metals, and, therefore, does not serve as a good proxy for metal contamination in these sediments.

Fourth, H₂O-leachable chloride, sulphate and ammonium all show similar distribution patterns, with high values in the centres of Manchester and Salford, and low values outside the centres. Sulphate and ammonium probably derive from wet and dry deposition, ultimately derived from vehicular and industrial processes. Chloride is presumed to be derived from road-salting. The presence of high levels of these soluble components indicates that RDS should be expected to have an impact upon surface water quality, and their chemical characteristics should be considered in surface water quality management.

Fifth, the recognition of spatial variability in the RDS in this city suggests that such data should be routinely collected to aid in the effective implementation of urban pollution management strategies based upon contaminant source control. A small-scale sampling regime, within one grid would also supplement these findings. This study sampled over a single period of five days but temporal (seasonal, yearly) variability also needs to be considered in future assessments.

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