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Comparison of anthropogenic metal deposition rates with excess soil loading from coal, oil and gas industries in the Usa River Basin, NW Russia

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Abstract: Trace metal composition of snowpack, snow-melt filter residues and top-soils were determined along transects through industrial towns in the Usa River Basin: Inta, Usinsk and Vorkuta. Elevated concentrations of deposition elements and pH in snow and soils associated with alkaline coal ash within 25–40 km of Vorkuta and Inta were found. Atmospheric deposition in the vicinity of Vorkuta and Inta, added significantly to the soil contaminant loading as a result of ash fallout. The element concentrations in soils within 20–30 km of Vorkuta do not reflect current deposition rates, but instead, reflect an historical pollution legacy, when coal mining activity peaked in the 1960s. There is little evidence of anthropogenic metal deposition around the gas and oil town of Usinsk.

Key words: Russian Arctic, Usa River Basin, soil and snow chemistry, soil loading, coal combustion, ash fallout.

Introduction

The continuing exploitation of non-renewable resources in Russia has created some large Arctic towns (Ziegler 1987), and increases the risk of localised environmental pollution (Pryde 1991). Long-range pollutants are also transported from industrial regions in the middle latitudes to the Arctic due to global atmospheric circulation patterns (Rahn 1982). The Usa River Basin in north-western Russia has a total surface area of 93,000 km^2 and has been subject to a range of industrial impacts, producing variation in land use from pristine uninhabited regions to densely populated areas of industrialization including oil and gas fields, coal and ore mining. Coal combustion (associated with power generation) has historically been the principal source of SO_2 and heavy metal pollution in the Usa River Basin (Solovieva *et al.*

2002; Walker *et al.* 2003a). Other sources include gas and oil extraction, cement production, pulp and paper production, construction and oil refineries (State of the Environment of the Komi Republic, 1992–1998). Exploitation of Usa coal is in decline due to its poor quality and associated high transportation costs (Lausala and Valkonen 1999). Recently, however, the oil and gas industries have boomed and are expected to expand further, bringing about significant risks of environmental pollution, *e.g.* from gas flaring and oil spills (Vilcheck and Tishkov 1997; Walker *et al.* 2005). Impacts of acid pollution have been seen close to emission sources around the city of Vorkuta where there are examples of environmental damage due to acidification and eutrophication (Getsen *et al.* 1994; Virtanen *et al.* 2002; Walker *et al.* 2003b). However, it should be emphasised that vast areas of the Russian Arctic appear close to pristine (Rovinsky *et al.* 1995). A regional geochemical mapping study by Salminen *et al.* (2004) demonstrated that in highly polluted areas, such as parts of the Kola Peninsula, physical and chemical properties of top-soils can become radically modified when compared to some of the most pristine areas in Russia. Likewise, the objective of this study was to compare the current rates of deposition from emissions due to fossil fuel combustion against excess soil loading concentrations within the Usa River Basin, between latitudes 64° and 68°N. The data provides a useful baseline against which the extent of pollution in the past and future can be gauged, and was achieved by quantifying the chemical status of snow and top-soil. The analysis of the chemistry of snow provides useful information on the distribution patterns of anthropogenic substances emitted into the atmosphere (Shaw *et al.* 1993). Concentrations of dissolved elements in melted and filtered snow were used to estimate wet deposition during winter and the sum of these values produces estimates of total element concentrations in snow (Reimann *et al.* 1996), and the analysis of the underlying soil, can be used to evaluate long term accumulation of conserved pollutants (Reimann *et al.* 2000; Salminen *et al.* 2004). Soil contamination due to alkaline emissions from coal combustion has been widely reported (Larssen *et al.* 2000; Walker *et al.* 2003a).

Materials and methods

Transects through point sources were undertaken to assess the distribution of pollutants from coal mining and burning in Inta and Vorkuta and from oil and gas extraction and use in Usinsk (Table 1, Fig. 1). The transects were established through the principal towns of Vorkuta (67°30'N, 64°05'E), Inta (66°03'N, 60°10'E) and Usinsk (66°01'N, 57°30'E). Between 4–6 sampling locations were selected along each transect at logarithmically increasing distances from each town. At each location, three sites were selected, approximately 1 km apart, and at each site 6 replicate samples of snow and soil were collected >10 m apart in open areas subject to minimal tree canopy effects (Reimann *et al.* 1999). Snowpack was

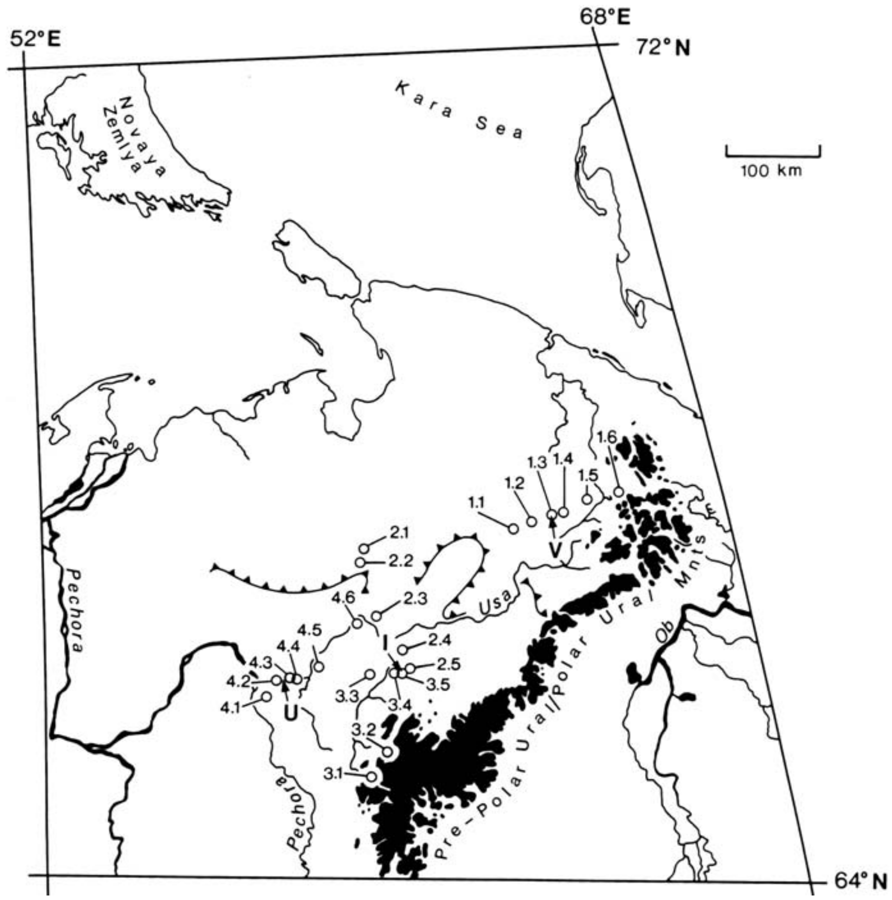

Fig.1. Map showing sampling sites on transects through the towns of Vorkuta (V), Inta (I), and Usinsk (U) in the Usa River Basin, NW Russian Federation. Linked triangles indicate the approximate position of the treeline across the Usa River Basin. Land above 500 m is also indicated. Map reproduced from Walker *et al.* (2003a).

sampled between March and April, prior to the onset of the spring thaw; as most of the total solute load can be lost with the first melt (Tranter *et al.* 1988). Snow pits were excavated at each site and snow collections and analysis were made according to Walker *et al.* (2003a). The trace metals Ag, Al, As, Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sr and Zn were determined on pooled samples composed of equal aliquots of filtered meltwater taken from each of the six replicate samples from each site, and trace elements were analyzed by ICP-MS. Analysis of Ca and K was by Flame-AAS. Cellulose acetate membrane filters used to filter the snow-melt and suspended solids were determined gravimetrically and photographed. Only results for Ba, Ca, K and Sr are given as they demonstrate the highest elevated concentrations around the towns. Six cellulose acetate membrane filters from each site were digested together in 10 mL of concentrated HNO_3 and element analysis of Al, Ba,

Table 1

Summary of main sampling sites along 4 transects within the Usa River Basin study area.

Site No.	Name of site	Co-ordinates	Elevation (m.a.s.l.)	Distance from nearest town
Transect 1				
1.1	Lake Padvaty	67°27'N, 63°05'E	160	45 km W of Vorkuta
1.2	Malen'kiy arvozh	67°30'N, 63°30'E	150	30 km SW of Vorkuta
1.3	Vorkuta	67°34'N, 64°09'E	180	2 km NE of Vorkuta
1.4	Lake Ngayats'yakha	67°35'N, 64°15'E	160	12 km NE of Vorkuta
1.5	Lake Mutnoye	67°44'N, 64°52'E	260	40 km NE of Vorkuta
1.6	Lake Protochnoye	67°47'N, 65°37'E	180	70 km NE of Vorkuta
Transect 2				
2.1	Khosedayu River	67°15'N, 59°37'E	70	130 km N of Inta
2.2	Lake Tumbulovaty	67°07'N, 59°34'E	110	110 km N of Inta
2.3	Two Lakes near Adak	66°35'N, 59°45'E	75	62 km N of Inta
2.4	Lake Lyz'vad	66°16'N, 60°13'E	52	20 km N of Inta
2.5	Lake Swan (Lebedinoye)	66°06'N, 60°15'E	21	7 km N of Inta
Transect 3				
3.1	Vangyr River	64°59'N, 59°12'E	270	110 km SW of Inta
3.2	Mezhgornyye lakes	65°15'N, 59°40'E	510	90 km SSW of Inta
3.3	Lake Van'kavad	65°59'N, 59°27'E	60	32 km SSW of Inta
3.4	'Fox Lake'	65°59'N, 60°01'E	60	7 km SSW of Inta
3.5	Inta	66°03'N, 60°10'E	60	2 km SSW of Inta
Transect 4				
4.1	Lake Bosmanvad	65°48'N, 57°03'E	40	30 km SW of Usinsk
4.2	Lake Chakty	65°58'N, 57°16'E	50	15 km W of Usinsk
4.3	Usinsk	65°59'N, 57°35'E	60	2 km E of Usinsk
4.4	Lake Kankur'ya	65°57'N, 57°42'E	40	10 km NE of Usinsk
4.5	Isaak-Ty/Shar'yu	66°07'N, 58°18'E	50	35 km NE of Usinsk
4.6	Adz'vavom	66°33'N, 59°16'E	50	100 km NE of Usinsk

Ca, Cd, Cu, K, Mg, Mn, Ni, Pb, Sr and Zn was undertaken by GF-AAS and F-AAS; concentrations were recalculated in relation to the volume of filtered melt-water to allow direct comparison with the solution data (Reimann *et al.* 1996). Again, only the elements with the highest elevated concentrations around the towns are presented which were: Al, Ba, Ca, K, Pb and Sr. Net acidity or alkalinity was determined by granplot acid titrations following the method of Legrand *et al.* (1982). Soil samples were collected over the 0–5 cm depth horizon and included organic and mineral soil, between July and August from the same sub-sites as snow samples. Details of sampling and analysis are given in Walker *et al.* (2003b). Frozen soil samples were allowed to thaw overnight at room temperature and a representative sub-sample (5–10 g) was oven dried (105°C) in acid washed silica dishes. Samples were ashed in a muffle furnace at 450°C overnight, allowed to cool in a dessicator and weighed for loss on ignition (LOI). A sub-sample (1 g) of the soil ash was digested in 10 ml of concentrated HNO_3 (Aristar). The digest residue (~1 mL) was diluted and filtered through Whatman No. 42 ashless filters then made up to 100 mL using ultra-pure water to give a final matrix of 1% HNO_3. Analysis of Ba, Cu, Mg, Mn, Na, Ni, Pb, Sr and Zn was undertaken using GF-AAS and F-AAS, with data presented for Ba, Sr and Zn. Soil pH was measured on moist samples, suspended in DI water at a solid: solution ratio of 1:2.5 following equilibration for 1 hour. Approximately 20 km beyond the northern-most site at Khosedayu River, 2.1 (see Fig. 1) on transect 2/3, an additional set of top-soil samples were collected along the Upper Kolva River (Table 2).

Table 2

Concentrations of metal elements within soil ash taken at the Upper Kolva River.

Element analysed	Element concentrations in soil ash (mg kg^{-1})	± Standard Error (n = 6)
Ba	28	3
Ca	172	19
Cd	0.1	0.02
Cu	12	1
K	1326	176
Mg	1593	314
Na	200	36
Ni	103	31
Pb	35	1
Sr	2	0.3
Zn	40	1

Element concentrations in filtered snow-melt snow (including suspended solids from digested cellulose acetate filters) and soil for Ba, Cu, Pb and Sr were considered together in an attempt to explain the large concentrations of elements found in top-soil samples around the cities in relation to current atmospheric deposition rates (Equation 1) (Walker *et al.* 2003a).

$$T_d = \frac{(C_d - C_p) \times S_d}{A_d} \qquad (1)$$

where

T_d – Time required to accumulate excess element concentration (y)

C_d – Concentration of element in soil ash within deposition area (g kg^{-1})

C_p – Concentration of element in soil ash at pristine site (g kg^{-1})

S_d – Mass of soil in 5 cm slice in deposition area (kg m^{-2})

A_d – Current element deposition rate (g m^{-2} y^{-1}). Values of A_d were derived by combining element concentrations in snow-melt and filter residues (g m^{-3} liquid) then multiplying by the mean annual precipitation (mm y^{-1}) for each town.

This approach assumes that:

– the lowest concentrations of elements found in soil (C_p) occur at sites at the transect extremes, furthest away from the emission sources (*e.g.* Transect 1 through Vorkuta – sites 1.1 and 1.6; Transects 2/3 through Inta – sites 2.1 and 3.1);
– highest concentrations (C_d) occur close to emission sources (*e.g.* Vorkuta: sites 1.3 and 1.4, Inta: sites 2.5 and 3.5);
– all historical deposition is contained within the top 5 cm of soil (S_d);
– the mean annual precipitation is the same at all sites on a transect;
– element concentrations in soils with low mineral contents (*i.e.* from peaty sites) can be compared against element concentrations in soils with high mineral contents by correction through the ashing process.

Results and discussion

The results of granplot titrations shows high alkalinity associated with ash deposition around Vorkuta (see Fig. 2), and was reflected in measurements of snow pH (see Walker *et al.* 2003b). By contrast, net acidity was recorded along the Usinsk transect. Along the Inta transect alkaline ash deposition around the city has effectively neutralised any local acidity and the background acidity shown in pristine snow further to the north. Fig. 3a, b, shows dried filters showing particulates collected from snow samples taken along transects through the industrial towns of Vorkuta and Usinsk. The darker filters close to Vorkuta are a result of the ash fallout from coal combustion which is reflected in Fig. 4, which shows the mass of suspended solids collected on cellulose acetate filters following filtration of thawed snow. The trend along each transect strongly suggests a substantial contribution from coal combustion in Inta and Vorkuta. The amount of solids emitted in the Vorkuta area from 1992 to 1998 were between 5–8 times greater than for Inta, which in turn was approximately 3 times greater than for Usinsk. The mean value of solids emitted from Vorkuta between 1992 to 1998 was 84,000 t y^{-1} (State of the environment of the Komi Republic, 1992–1998). The greater part of emissions from Vorkuta comprised hydrocarbons followed by solid particles of fly ash originating from coal mining and a cement factory. In contrast, the majority of emissions from Usinsk were CO_2 from the oil and gas industry based there. Along transects through Vorkuta and Inta filtered snow-melt collected at sites closest to the towns contained elevated concentrations of Ba and Sr (Fig. 5). In addition elevated concentrations of Ca and K in snow were found around Vorkuta, which may suggest that combustion ash was the source (Reimann *et al.* 1996). Arsenic concentrations were greatest at the urban sites around Vorkuta and lower, respectively, in Inta and Usinsk and concentrations of the trace metals Cd, Cu, Ni, Pb and Zn were uniformly low at all sites (data not shown). However, the absence of a pro-

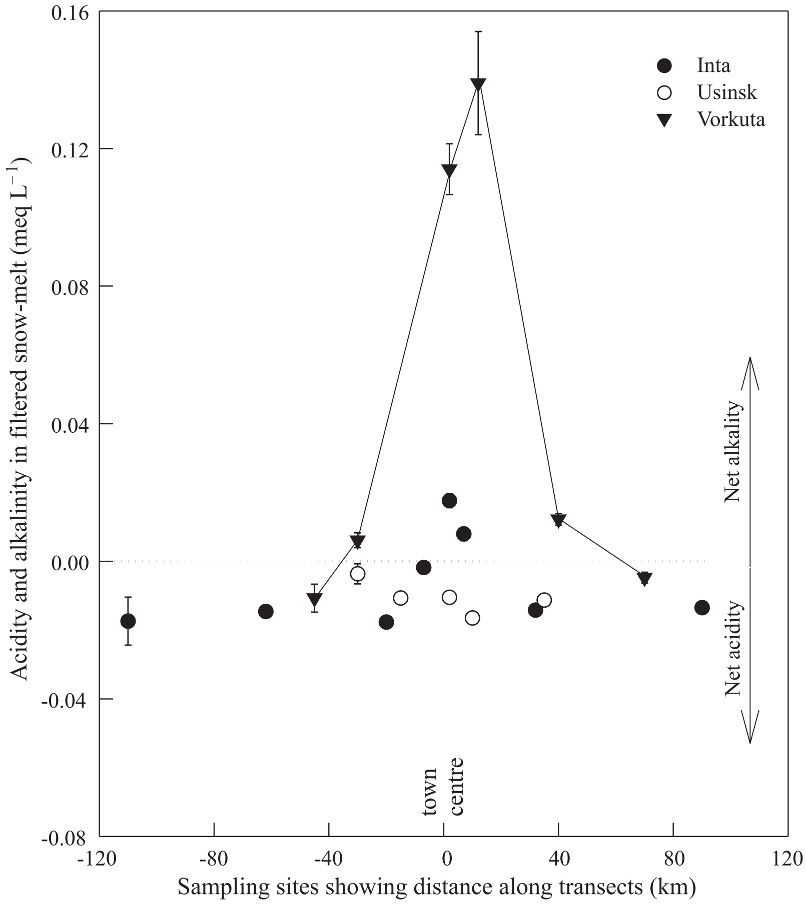

Fig. 2. Values of granplot acid titrations performed in filtered snow-melt samples taken along transects through the industrial towns of Vorkuta, (running W-E), Inta, (running N-S) and Usinsk, (running SW-NE) during Spring 1998 and 1999. Plotted values are means ±SE ($n = 18$). From Walker *et al.* (2003a).

nounced urban peak in solution concentration may have been a result of adsorption on alkaline ash. In addition, concentrations of Ba, Co, Cu, Sr and Zn were slightly greater in Inta than around Vorkuta, possibly because the town's coal-fired power plant was closer to those sampling sites. In the immediate vicinity of Vorkuta and Inta there were elevated concentrations in suspended solids in snow-melt of the major combustion ash constituents: Al, Ba, Ca, K and Sr (Fig. 6). Urban plumes for Pb are evident from analysis of snow-melt particulates around Vorkuta and Inta. The cationic composition of the suspended solids was dominated by Ca, and if it is assumed that the Ca is present as $CaCO_3$ this would constitute approximately 12% of the suspended solids around Vorkuta. The entire transect through Usinsk generally reflected background concentrations similar to those observed in pristine areas at both ends of the Inta and Vorkuta transects. Concentrations of ions associ-

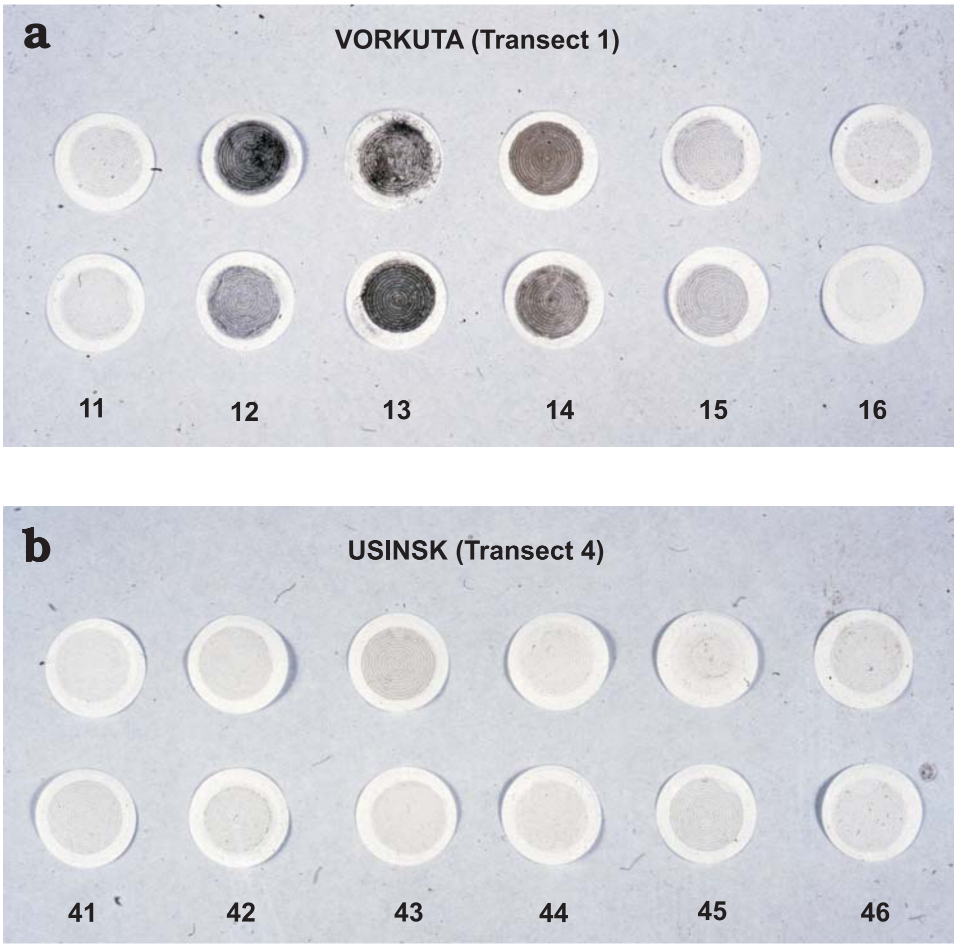

Fig. 3. Dried filters showing particulates collected from snow samples taken along transects through the industrial towns of Vorkuta (a) and Usinsk (b).

ated with particulates and those in free solution were strongly positively correlated in snow collected on the Vorkuta transect *e.g.* Ca ($r^2 = 0.97$), Sr ($r^2 = 0.97$) and Ba ($r^2 = 0.86$) (Walker *et al.* 2003a). The slope of correlations between element concentrations in suspended solids and filtered snow showed that the majority of the major constituents of combustion ash were in particulate form. Mean solid: solution ratios for the total quantities of Ca, Ba and Sr were 12, 17 and 10 respectively. This confirms that estimates of deposition from filtered snow melt only, can seriously underestimate deposition rates by up to 90% for certain elements where there is significant particulate pollution (Reimann *et al.* 1996).

The depth of the surface organic layer in top-soil from each quadrat varied between 0.5–>5 cm. Elevated soil pH was found only in the Vorkuta area (Fig. 7), possibly a result of local deposition of alkaline ash material from coal combustion

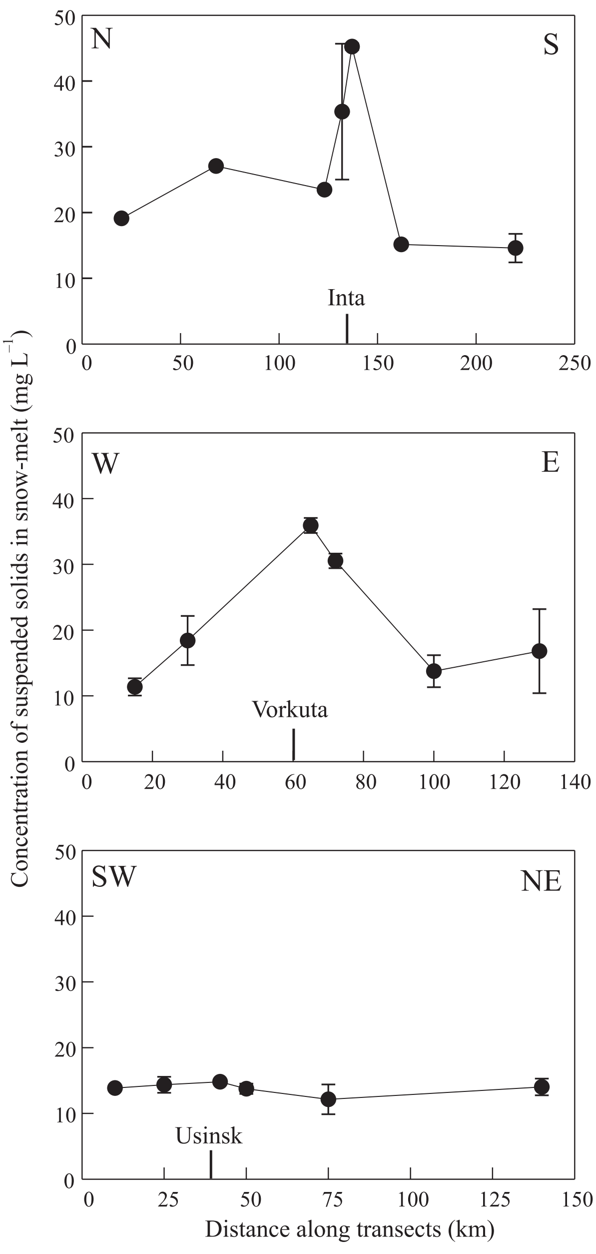

Fig. 4. Plots showing the concentration of suspended solids in filtered snow-melt samples taken along transects through Vorkuta, Inta and Usinsk, during Spring 1998 and 1999. Plotted values are means showing standard errors (n = 3). Adapted from Walker *et al.* (2003a).

and cement factory. In mineral soil (0–5 cm depth) Rusanova (1995) measured pH values of 7.2 within the town, whereas the pH fell to 4.8 at 15 km SW of the town limits; these figures compare favourably with the present data. Soil pH along

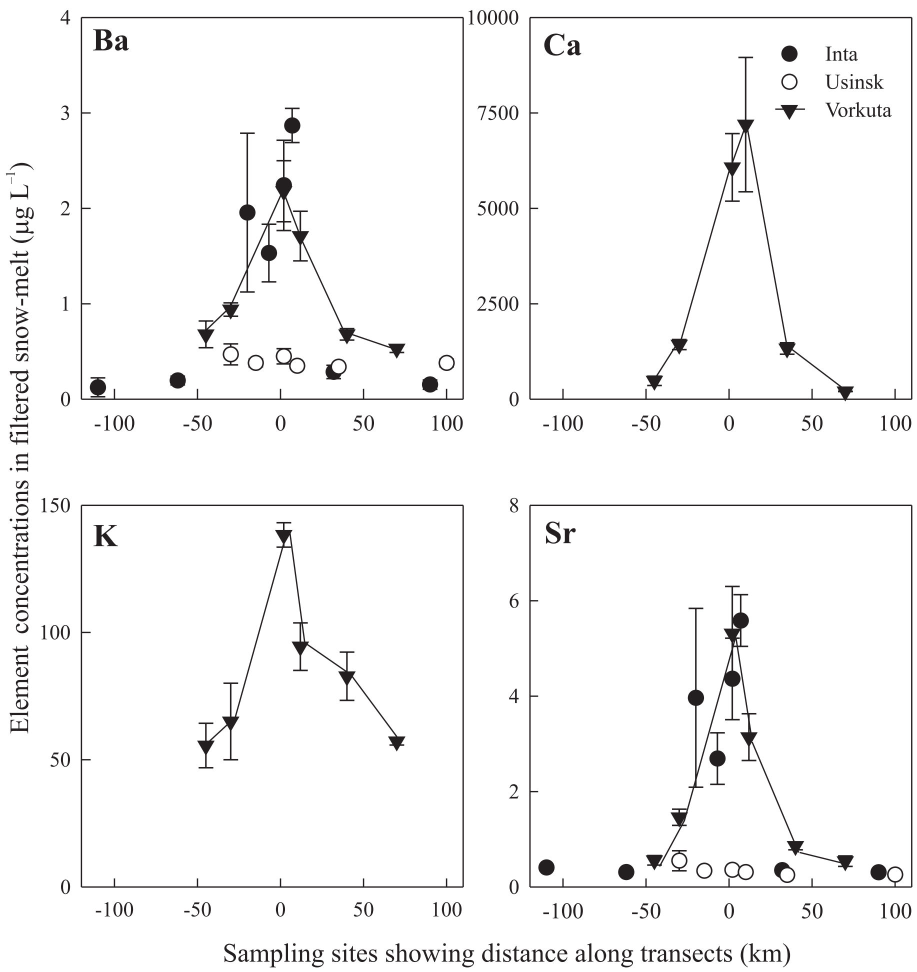

Fig. 5. Concentrations of elements in filtered snow-melt samples taken along transects through Vorkuta, Inta and Usinsk during Spring 1998 and 1999 measured by ICP-MS. Concentrations of Ca and K were measured by F-AAS along the Vorkuta transect only. Plotted values are means showing standard errors (n = 3). Adapted from Walker *et al.* (2003a).

transects through Inta and Usinsk were uniformly low, as were the values recorded at the extremes of the Vorkuta transect. For the less common soil constituents found in combustion ash *e.g.* Ba and Sr (Vassilev and Vassileva 1997) there was clear evidence of increased loading close to Vorkuta. There may also be soil enrichment of Zn around Vorkuta. The spatial extent of the local alkalisation of top-soil at sites around Vorkuta corresponds with vegetation changes evident in satellite images which have been confirmed by ground plot field observations (Virtanen *et al.* 2002). The distribution of the pH within top-soil shows the effects of the emission sources of Vorkuta and Inta; the most acidic samples were collected at remote sites. The higher soil pH values around Vorkuta correlate well

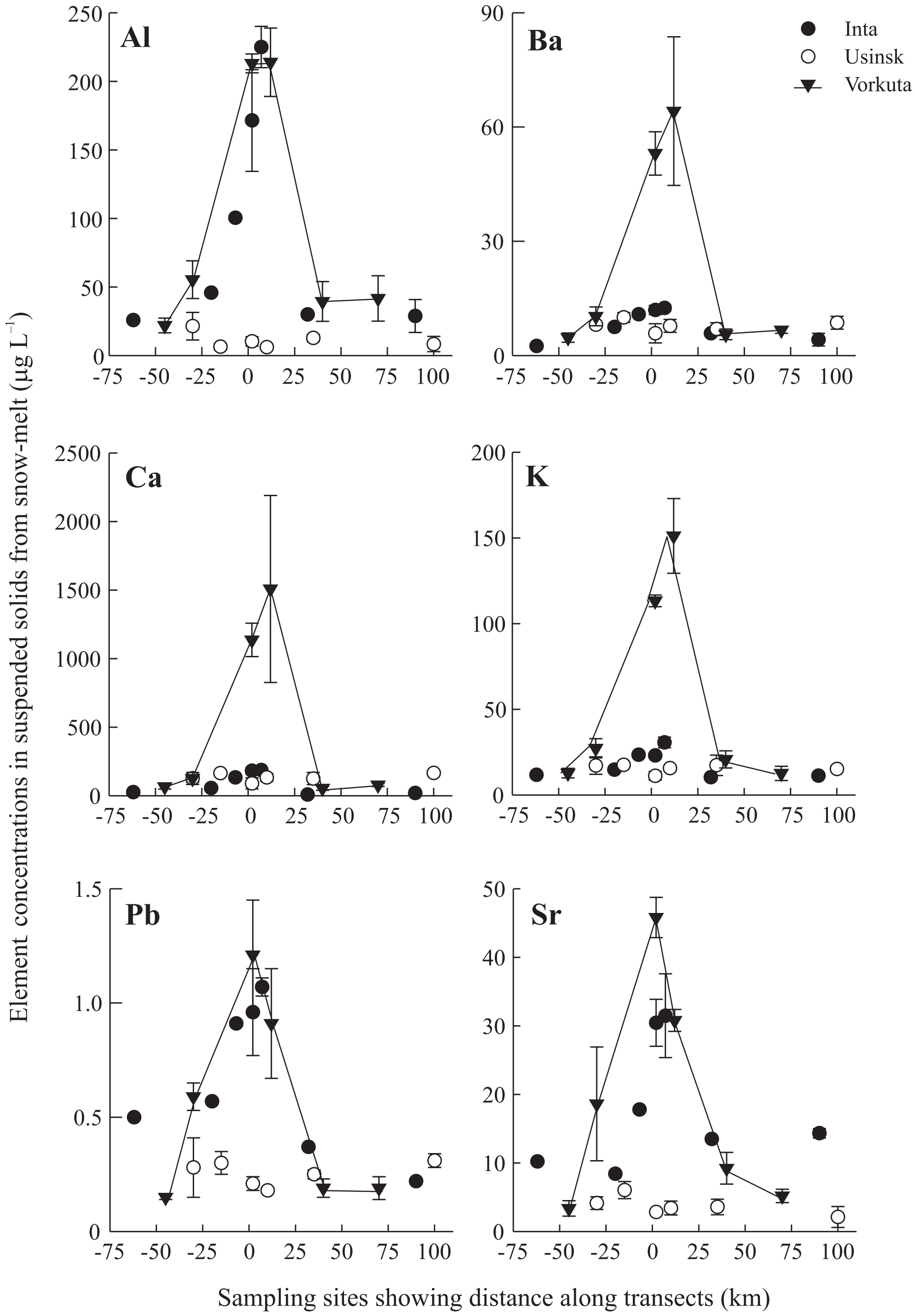

Fig. 6. Concentrations of elements within filter residue digest samples taken along transects through Vorkuta, Inta and Usinsk, during Spring 1998 and 1999. Plotted values are means showing standard errors ($n = 3$). Adapted from Walker *et al.* (2003a).

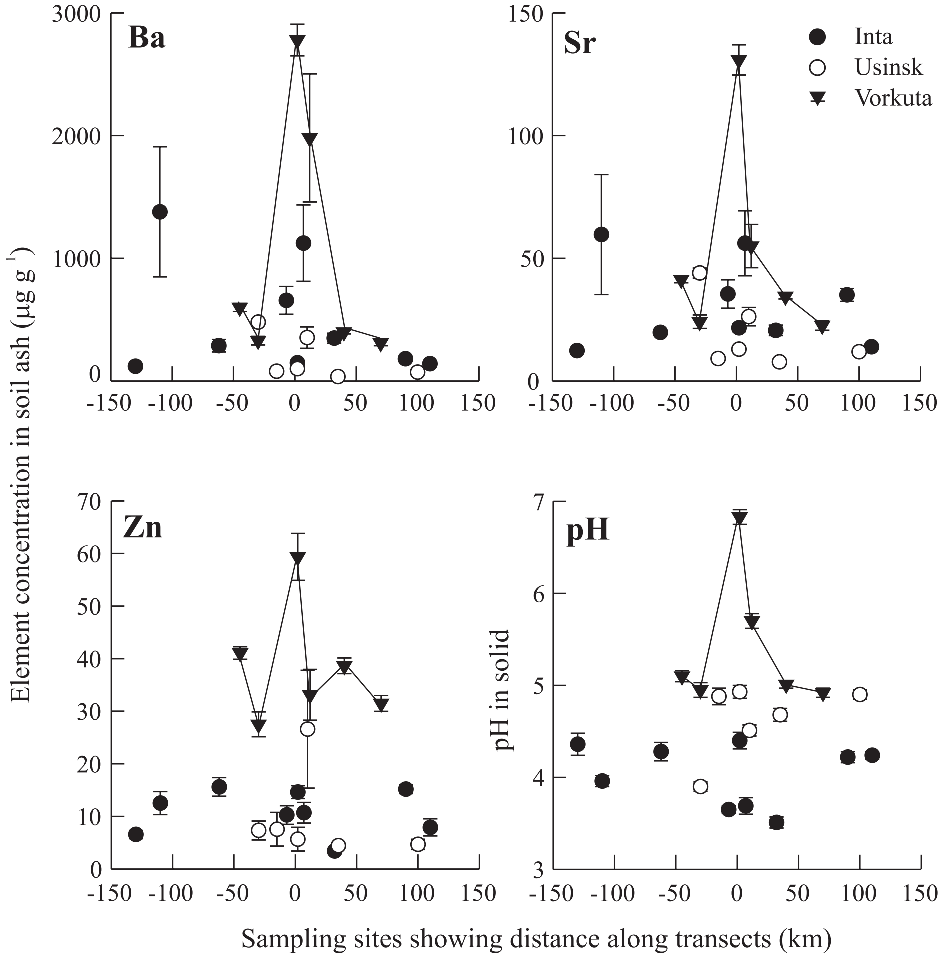

Fig. 7. Concentrations of of soil pH and elements in soil ash digest samples taken along transects through Vorkuta, Inta and Usinsk, during Summer 1998 and 1999. Plotted values are means showing standard errors (n = 18). Adapted from Walker *et al.* (2003a).

with the Ca concentrations in snow and suspended solids. This indicates an important role for ash input from coal combustion and cement dust from the nearby cement factory in regulating snow and soil pH. Potentially, a reduction in fly ash and cement dust emissions could mean solubilization of accumulated heavy metals due to a progressive drop in soil pH (Reimann *et al.* 1996). Robb and Young (1999) found that large additions of calcareous, metal-rich, fly ash to acidic soils increased the soil pH, to the extent that the metal ion concentration in the soil solution was reduced despite the increased total metal loading to the soil. A small pollution signature was also detected in concentrations of metal elements within soil ash at an additional site at the Upper Kolva River where oil and gas operations produces gas flares (Table 2). For both major and minor elements T_d was considerably

Table 3

Comparison between pristine and contaminated sites along transects showing measured element concentrations within soil and the amount of time required for accumulation of these elements from current (apparent) deposition rates. The time in years required to accumulate excess element concentration in soil was determined by equation 1.

Element	Transect	Site No.	Conc. in soil ash (mg kg^{-1})	Mass of mineral (kg m^{-2})	Expected conc. in ash (mg kg^{-1})	Expected conc. (g m^{-2})	Actual conc. (g m^{-2})	Change (g m^{-2})	Deposition rate (g m^{-2} y^{-1})	Time required to accumulate (y)
Ba	1	1.6	3.1×10^5	35.70	3.1×10^5					
		1.3	2.8×10^6	37.60	3.1×10^5	11.630	104.500	92.870	2.9×10^{-2}	3247
	2/3	3.5	1.5×10^5	41.03	1.5×10^5					
		3.4	1.1×10^6	8.51	1.5×10^5	1.260	9.540	8.280	7.3×10^{-3}	1139
	4	4.5	3.6×10^4	47.77	3.6×10^4					
		4.4	3.5×10^5	45.13	3.6×10^4	1.610	1.700	0.090	4.0×10^{-3}	23
Cu	1	1.2	2.9×10^3	40.10	2.9×10^3					
		1.3	9.4×10^3	37.60	2.9×10^3	0.109	0.354	0.245	8.1×10^{-4}	304
	2/3	2.3	1.2×10^3	29.53	1.2×10^3					
		2.5	2.7×10^3	4.45	1.2×10^3	0.005	0.012	0.007	2.5×10^{-3}	3
	4	4.5	1.7×10^2	47.77	1.7×10^2					
		4.4	1.5×10^3	45.13	1.7×10^2	0.008	0.070	0.062	1.2×10^{-3}	51
Pb	1	1.2	1.6×10^4	40.10	1.6×10^4					
		1.4	2.6×10^4	37.60	1.6×10^4	0.579	0.940	0.361	9.0×10^{-4}	400
	2/3	2.3	1.3×10^4	12.89	1.3×10^4					
		3.4	2.2×10^4	8.51	1.3×10^4	0.110	0.191	0.081	5.4×10^{-4}	149
	4	4.5	4.7×10^3	47.77	4.7×10^3					
		4.4	1.3×10^4	45.13	4.7×10^3	0.214	0.586	0.372	3.7×10^{-4}	989
Sr	1	1.6	2.3×10^4	35.71	2.3×10^4					
		1.3	1.3×10^5	37.60	2.3×10^4	0.850	4.920	4.070	2.6×10^{-2}	154
	2/3	2.3	2.0×10^4	12.89	2.0×10^4					
		3.4	5.6×10^4	8.51	2.0×10^4	0.168	0.477	0.309	1.8×10^{-2}	18
	4	4.5	7.8×10^3	47.77	7.8×10^3					
		4.4	2.6×10^4	45.13	7.8×10^3	0.350	1.180	0.830	1.8×10^{-3}	450

in excess of the life span of the city of Vorkuta (Table 3), suggesting current rates of metal deposition in precipitation (A_d) cannot explain the apparent accumulation in soil. For example the values of T_d for the trace element Cu was 304 y; for the alkali earth elements Ba and Sr T_d was 3250 and 154 y. The high soil metal loadings found locally around Vorkuta provide supporting evidence for such a historical change in Vorkuta's pollution climate because the quantities of metals present are too great to be explained by current deposition. However, these anomaly patterns may be strongly controlled by element distributions in the bed rock (Salminen *et al.* 2004). This discrepancy between measured element concentration within soil and the amount of time required to accumulate this from current deposition rates may also arise for a number of other reasons. Firstly, there has been a substantial decline in coal mining in Vorkuta, since the peak of activity in the 1960s and 1970s. Within the last decade, the thirteen coal mines operating in 1990 were reduced to just seven by 1999 (State of the environment of the Komi Republic, 1992–1998). Coal production at the largest mine, Vorgashorskaya, has fallen by half between 1991 and 1999, from nearly 21 x 10^6 tonnes to 13 x 10^6 tonnes (State of the environment of the Komi Republic, 1992–1998). This trend is consistent with data on spherical carbonaceous particles (SCPs) found in lake sediment profiles showing that a peak in SCPs is found at 1.5 cm below the sediment surface and suggesting that SCP deposition rate has since been in sharp decline (Solovieva *et al.* 2002) and changes in tundra vegetation (Virtanen *et al.* 2002). Secondly, Vorkuta has a total population of around 200 000 inhabitants and urban development on this scale within the tundra ecosystem requires a great deal of construction activity, which would import considerable quantities of cement dust and other particles, resulting in elevated deposition of alkali-earth elements (Jalkanen *et al.* 2000). It is therefore likely that element concentrations in soils within 20–30 km of Vorkuta do not reflect current deposition rates but are a historical legacy of greater construction and industrial activity in the recent past.

In conclusion, results show considerable local alkalisation in both the top-soil and snow-pack around Vorkuta; in the vicinity of Inta alkalization was only apparent in snow-pack. Deposition of coal ash around both towns is mainly responsible, with possible contributions from a cement factory located in Vorkuta. Near pristine areas located more than 30 km from Vorkuta and Inta appear to be unaffected by ash deposition, and are characterised by acidity in both snow-pack and top-soils. Sites along the entire transect through the town of Usinsk, which is supplied by a gas-fired power station, were near pristine. There were elevated concentrations of Al, Ba, Ca, K and Sr in suspended solids and snow-pack around Vorkuta and Inta. Most of the deposition of elements commonly associated with combustion ash, occurred in particulate form. There was evidence that trace element concentrations of Pb, Cu and Zn in snow-pack were reduced around Vorkuta, due to adsorption onto alkaline ash particles. Extremely high element concentrations of Ba and Sr in top-soils around Vorkuta could not be explained by current deposition rates. This discrepancy may be the result of greater coal mining and construction activity in previous decades.

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