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Element contents in mountain birch leaves, bark and wood under different anthropogenic and geogenic conditions

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Abstract

Forty samples each of leaves, bark and wood of mountain birch (*Betula pubescens* EHRH.) were collected along a 120 km long south–north transect running through Norway's largest city, Oslo. Concentrations of 26 chemical elements (Ag, As, Au, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Sr, Ti and Zn) as well as loss on ignition for the three sample materials are reported. By far the highest concentrations of most elements appear in the leaves. Prominent exceptions are Au and Pb, both of which are enriched in wood, indicating the importance of root-uptake, and As which is enriched in bark. Bedrock lithology, ore occurrences, soil pH and urban contamination all have a visible influence on the element concentrations in mountain birch leaves, bark and wood. It is often impossible to differentiate between all the factors that can influence element concentrations in the three sample materials. Mountain birch bark shows the strongest anthropogenic impact of the city of Oslo for dust-related elements (Fe, La, Ti) and Sb. Even in mountain birch bark the influence of the city on element concentrations is no longer discernible from the background variation at a distance of less than 20 km from Oslo centre. Compared to terrestrial moss, mountain birch appears to be of little value as a biomonitor for urban contamination.
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1. Introduction

Transects have been used successfully to assess the influence of point sources on the spatial distribution of anthropogenic influence (Wilcke et al., 1996; Reimann et al., 1997; Bonham-Carter et al., 2006), to study the spatial component of large scale natural processes (e.g., Reimann et al., 2000), to locate sources of diffuse PAH-contamination within cities (Wilcke et al., 1999) or to study the influence of a city on PAH-concentrations in forest soils (Jensen et al., 2007). Reimann et al. (2006) have used terrestrial moss (*Hylocomium splendens* (HEDW.) SCHIMP. – Smith, 2004) to study the impact of

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the city of Oslo as a diffuse contamination source on moss chemistry along a 120-km south–north transect. These authors demonstrated that the elements Ag, Al, Au, Bi, Cd, Co, Cr, Cu, Fe, Mo, Ni, Pb, Pt, Sb, Th, Ti and Zn all showed a significant peak in moss samples collected in Oslo and its surroundings as compared to moss sampled along the remaining parts of the transect.

As in any major city, a typical mix of many different emitters exists (or existed) in Oslo, e.g., the chemical and metal industries, tanneries, power plants, garbage incinerators and, in former times, several coke ovens. The inventory of possible contaminating industrial activities in Oslo lists 1798 sites within the city boundaries (see: http://www.helse-og-velferdsetaten.oslo.kommune.no/folkehelse/miljorettet-helsevern/jordforurensning_og_radon/databaser/). A high traffic density must be cited as a further major source of many metals to the urban atmosphere.

The transect (Fig. 1) was chosen such that it crosses a number of different lithologies (Fig. 2). Precambrian gneisses occur at both ends of the transect. Cambro-Silurian sediments, including black shales, are found in the city of Oslo and at the northern end of the transect, in the Randsfjord area. Magmatic rocks of the Oslo Rift occur at the northern fringes of the city and throughout the forest to the north of Oslo. The geochemistry of the rocks and soils along the transect is presented in Reimann et al. (2007).

Here leaves, bark and wood of mountain birch (*B. pubescens* EHRH.) taken at the same sampling sites along the Oslo transect are used to test their suitability as biomonitors of anthropogenic influence, as well as to study the distribution of the elements in different parts of birch under varying natural conditions.

1.1. Location and land use

The study area is located in southern Norway, in the surroundings of, and including, the city of Oslo (Fig. 1). Oslo has about 600,000 inhabitants and is Norway's largest city. Another 1.4 million people, of Norway's total population of 4.5 million, live close to the city. In general the region to the south of Oslo (towards Ås – Fig. 1) is one of the most intensely utilised and richest agricultural areas of Norway with many interspersed suburban developments. Directly north of Oslo the terrain rises abruptly to an elevation of 400–700 m above sea

level (a.s.l.) and land use changes completely. This area, known as "Nordmarka" (Fig. 1), is dominated by forests and many lakes and is extensively utilised for non-motorised recreation purposes by the Oslo population. Forestry is another important activity in the area. Further north, in the Randsfjord area (Fig. 1), agriculture is again the dominant land use. In contrast to the Ås area, animal husbandry is widespread and the area is too far inland from Oslo to feel the pressure of spreading urbanisation.

The annual precipitation in the investigated area is in the range of 700–1000 mm. The yearly average temperature varies between 4 and 6 °C. Due to the higher elevation and higher precipitation, the climate in Nordmarka is markedly colder and wetter than along the rest of the transect. For detailed maps on precipitation, temperature and vegetation, see Moen (1998). The dominant wind direction is towards the north, i.e. from Oslo towards Nordmarka and Randsfjord.

1.2. Geology

The transect was selected so that it crossed different lithologies, typical of the greater Oslo area. The most remarkable geological feature is the Oslo Rift. A late Carboniferous succession of shale, sandstone and conglomerate (the Asker Group) marks the onset of deposition related to the Oslo Rift (Dons and Larsen, 1978). The sedimentary sequence was deposited on a levelled surface and is overlain by basalt followed by a thick sequence of latite lava flows (rhomb porphyry) with thin beds of interlayered sand. Further evolution of this Permian rift is characterised by a number of volcanic centres with basaltic to subordinate rhyolitic lavas preserved in collapsed calderas rimmed by major ring faults and dykes. A variety of plutonic rocks cut the units described above (Dons and Larsen, 1978). Precambrian gneisses occur at the southern- and northernmost ends of the transect. Fig. 2 shows the location of the transect sampling points in relation to the geology.

For simplicity, the sampling sites can be allocated to four large lithological groups (Fig. 2). The soils in the south, the Ås area, and including the southern parts of Oslo have developed on glacial material derived from Precambrian gneisses ($N = 11$). Within the city centre of Oslo in addition to these gneisses, there is a relatively small area underlain by Cambro-Silurian shales and sandstones ($N = 4$). On leaving the city of Oslo towards the north the magmatic
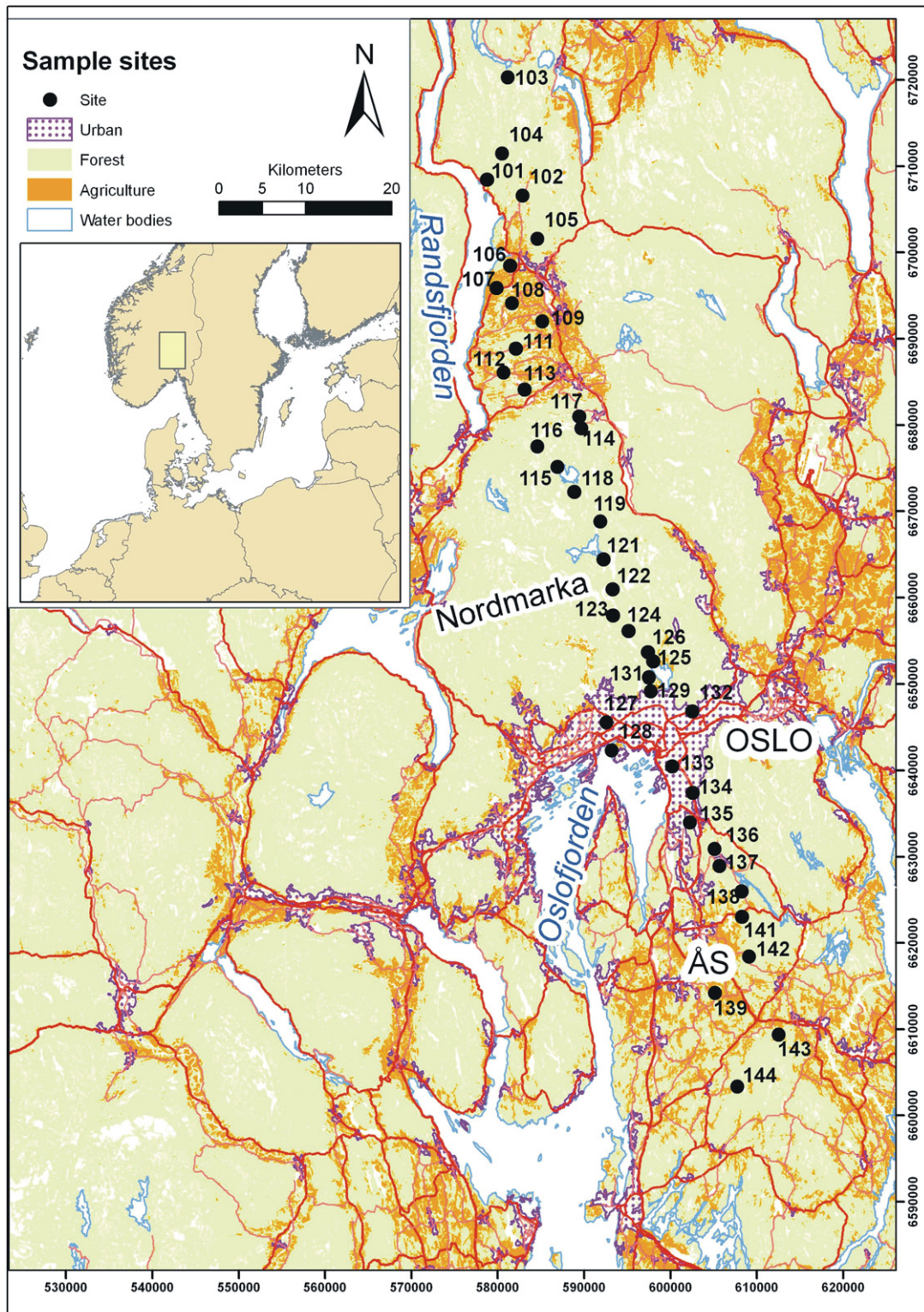
Fig. 1. Topographic map of the survey area showing the location of the south–north transect through Oslo with the sample sites identified by a sample number. Different grey shades differentiate between forests, agricultural areas and urban developments.
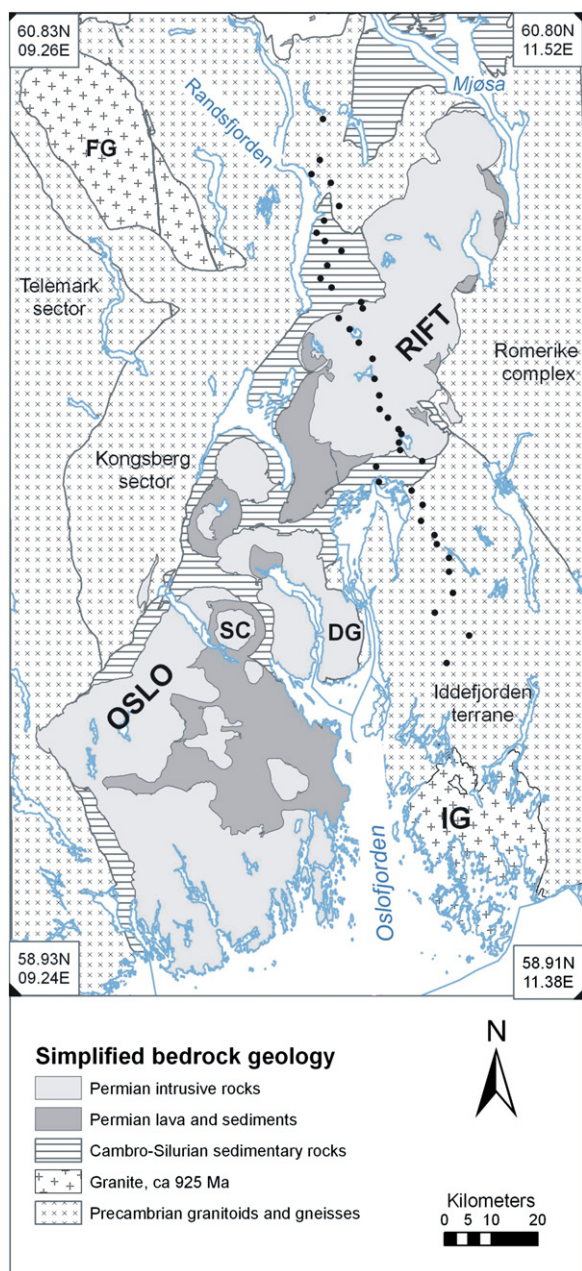
Fig. 2. Simplified bedrock geological map of the Oslo Area (from Lutro and Nordgulen, 2004) sample sites are shown, for site numbers referred to in Fig. 1. Water bodies (see Fig. 1) are in white. FG = Flå granite; IG = Iddefjord granite; DG = Drammen granite; and SC = Sande Caldera.

rocks of the Oslo Rift occur ($N = 13$). Along the transect these mostly have a syenitic composition. North of Nordmarka the Randsfjord area is characterised by the occurrence of the same Cambro-Silurian shales and sandstones that are encountered in Oslo ($N = 7$, total Cambro-Silurian sediments:

$N = 11$). At the northern end of the transect, Precambrian gneisses again form the bedrock ($N = 5$).

During the last ice age the whole study area was ice-covered. The thickness of the ice reached its maximum about 18–20 ka ago, and moraine material was deposited in the study area. Due to the melting of the ice the land rose, and the previous coastline in the area can now be found at 150–200 m above sea level (a.s.l.) (Lundmark, 1986). Below this level, large deposits of marine clays can be found. Today most of these areas are used for agriculture and were avoided during sampling. The same applies to glaciofluvial deposits that can contain material that was transported over long distances, while most of the other moraine material is very local in origin (Haldorsen and Sørensen, 1986). Results presented in Reimann et al. (2007) suggest that the transport distance is less than 3 km – all lithological breaks can be identified via immediate changes in the geochemistry of the C-horizon material. The average transport distance of the moraine material is thus insignificant at the sample spacing used and given the length of the transect. In areas underlain by Cambro-Silurian shales, weathered material (residual soils) can be found at the surface.

The geological map of Oslo and surrounding areas (Lutro and Nordgulen, 2004) provides an excellent and more detailed geological overview of the study area.

2. Methods

2.1. Sampling

Sample sites were selected in accordance with the methods used in the European moss monitoring project (e.g., Rühling, 1994; Zechmeister, 1997). A minimum distance of 300 m to major roads and larger settlements is required, a minimum distance of 100 m to minor roads and houses and a minimum distance of 5 m to forest roads. With two exceptions (site 134, which is only 25 m from a major road (E6) and site 132, which is about 200 m from a major road (E4)) all sample sites, even those in the city of Oslo, fulfil these requirements. Otherwise, at the selected sample density, it was possible to collect the samples at largely "untouched" natural sites in the surrounding forests, which reach into the city, and in large forested parklands. To avoid possible seasonal influences (e.g., Zechmeister et al., 2003) all samples were collected within as short a time

span as possible (14 days) in the autumn of 2005. All sites were located in forest ecosystems with a required minimum number of typical (for the area) plant species (presence of Norwegian spruce (*Picea abies* (L.) KARST.), birch (*Betula pendula* ROTH and *B. pubescens* EHRH.) and European mountain ash (*Sorbus aucuparia* L.) in the tree layer, bracken or other species of fern (*Pteridium aquilinum* (L.) KUHN), cowberry (*Vaccinium vitis-idaéa* L.), blueberry (*Vaccinium myrtillus* L.) and terrestrial moss (*Hylocomium splendens* (HEDW.) SCHIMP.)) in the ground vegetation layer. Even in the city of Oslo it was still possible to find sufficiently ''natural'' forested areas to enable directly comparable sites to be sampled.

Trace element- and powder-free vinyl gloves (one new pair per sample material) were used during sample collection. About 1 L of birch leaves (exclusively from *B. pubescens*) were collected as a composite sample from several trees/locations spread over an area measuring about 50 m × 50 m into clear contamination-free polyethylene (PE)-zip-lock plastic bags. Leaves were collected such that they provided the best possible ''average'' of ''leaf composition'' for birch at the sampling site, i.e. without directional preference and at all locations within the tree that could be reached from the ground or via cutting branches from the tree. To collect birch bark the bark from a minimum of 10 trees was cut from the stem at about 1.5 m over the ground using a carbon steel knife and then peeled from the stems. Again, bark was collected without directional preference to give a good ''average'' composition of birch bark at the site. The bark was collected free of lichen and taken exclusively from large light patches without branches or large black spots. The bark was placed into clear contamination-free polyethylene (PE)-zip-lock plastic bags. To collect wood, large twigs (age > 10a) were cut from the same trees and the bark was completely removed from the twigs using a carbon steel knife. One litre of 2–5 cm long wood cuttings were collected into the same clear contamination-free polyethylene (PE)-zip-lock plastic bags as were used for leaves and bark. Due to the already quite cool fall temperatures there was no problem with condensed water collecting in the bags. In the field the PE bags were marked with the site number and a 3-letter code for the material. UTM coordinates of all sample sites were recorded using a GARMIN GPS system. Each sample site was recorded in a series of at least three field photographs. Every third day the plant samples were posted to the laboratory for immediate drying. After arrival in the laboratory of the Geological Survey of Norway (NGU) all samples were dried to constant weight at $T < 40\,°C$. The plant samples were not washed prior to drying.

2.2. Analyses

All dried samples were shipped to ACME laboratories in Canada by courier. Here the samples were pulverised to pass a −100 mesh screen in a mild steel mill. The method has been thoroughly checked to avoid any contamination to the plant samples by the mill material.

A 0.5 g aliquot of sample material was first leached with concentrated HNO_3 for 1 h and then digested in a hot water bath for an additional hour. After cooling, a modified aqua regia solution of equal parts of concentrated ACS grade HCl and HNO_3 and de-mineralised H_2O was added to each sample (6 mL/g) to leach in a hot (95 °C) water bath for 2 h. After cooling, the solution was made up to a final volume with 5% HCl and then filtered. The sample weight to solution volume ratio is 1 g per 20 mL. The solutions were analysed using a Perkin Elmer Elan 6000 inductively coupled plasma mass spectrometer (ICP-MS) for 37 elements (Ag, Al, As, Au, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Sc, Se, Sr, Te, Th, Ti, Tl, U, V, W and Zn).

In an attempt to improve the detection limits for a number of elements, 15 g of dry plant material underwent additionally controlled ignition to minimise volatilisation at 475 °C. The ash was then digested following the above procedure and analysed for Au, As, Bi, Sb, Se, Te, Th, Tl U, V and Pd and Pt by ICP-MS. Results were recalculated to the dry plant weight.

Loss on ignition (LOI) was determined gravimetrically following the controlled burning of the vegetation samples at 475 °C.

2.3. Quality control

The samples were analysed in one large batch of 400 plant samples (terrestrial moss, birch leaves, wood and bark, spruce needles, wood and twigs, European mountain ash leaves, fern leaves). For quality-control purposes the National Institute of Standard and Technology (NIST) standard reference material 1575a Pine Needles and a laboratory internal reference material, Standard V13 Mountain

Hemlock Needles, were included evenly spread over the whole batch 8 and 13 times respectively. Detailed results of quality control are reported in Reimann et al. (2006). Ashing was not carried out for the standard samples. Thus it was difficult to assess the quality of the analytical results for the ashed samples (there is a high risk that elements are lost during ashing) beyond a direct comparison of the results of ashed and non-ashed samples in XY-diagrams. Due to these problems only results for As and Au are used here. Average precision for the elements reported here is better than 5%. In general analytical results in birch leaves, bark and wood were, for many elements, substantially lower than those found in the moss samples. As a result, for 13 elements more than 50% of the values were below the lower limit of detection: Al (100 mg/kg), Bi (0.02 mg/kg), Ga (0.1 mg/kg), Pd (0.5 µg/kg), Pt (0.2 µg/kg), Sc (0.1 mg/kg), Se (0.1 mg/kg). Te (0.02 mg/kg), Th (0.01 mg/kg), Tl (0.02 mg/kg), U (0.01 mg/kg), V (2 mg/kg) and W (0.1 mg/kg). Thus only the concentrations for 26 elements (Ag, As, Au, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Sr, Ti, and Zn) and the results for LOI are presented here.

2.4. Data analysis

Due to the nature of the data (only 40 samples but 28 parameters), compositional data with closure effects, and expected strong spatial dependencies formal statistics and statistical tests were avoided under data analysis. Rather methods of exploratory data analysis (EDA, Tukey, 1977) and robust measures of central tendency and spread are used. Soil-birch correlations were calculated for the log-transformed data and are Pearson correlation coefficients. All discussed correlations were additionally graphically checked in scattergrams.

3. Results and discussion

Analytical results are summarised in Table 1. Table 2 compares the median values of this investigation with the median values for soils collected at the same sample sites (O- and C-horizon, data from Reimann et al., 2007) and provides the ratios between the different sample materials and of the sample materials to the soils. While elements are sorted alphabetically in Table 1 they are sorted according to decreasing median concentrations in birch leaves in Table 2. Figs. 3 and 4 present the

data in the form of boxplot comparisons for the major (K, Ca, Mg, P and S) and minor nutrients (Mn, Fe, Zn, B, Cu, Ni and Mo) (Fig. 3) and for the trace elements (Fig. 4). Note that in birch leaves and bark the minor nutrient Mn occurs in considerably higher concentrations than the major nutrients P and S (Table 2). This relative enrichment of Mn in birch is well known and documented in the literature (e.g. Perelman, 1975; Reimann et al., 2001a). Results for Cr and Na are provided in the tables but not shown in the figures, because all Cr values in all plant tissues were almost the same (see Table 1) with the exception of one large outlier in birch wood (contaminated sample?). For Na all values in bark and wood were close to the detection limit (10 mg/kg) and in addition reported rounded to 10 mg/kg-values: they are thus of little use for data analysis.

3.1. Element concentration levels in leaves, bark and wood

Fig. 3 shows the large differences in the concentration and variation of major and minor nutrients between leaves, bark and wood. The leaves are strongly enriched in many nutrients when compared to bark and wood (Ca, K, Mg, Mn, P, S, Zn, B, Ni), a long established fact (e.g. Sabinin, 1955). Most of the major nutrients show the sequence leaves ≫ wood > bark (Ca, K, Mg, P). The relative enrichment of the leaves in the macronutrients often covers more than one order of magnitude. A number of nutrients show a constant decrease from leaves over bark to wood (Mn, S, Zn, Fe, Ni, Mo). For Cu, the values in leaves and bark are almost the same, while the values in wood are much lower. The high values of the major nutrients in birch leaves are utilised in forestry to improve the soil quality in coniferous forest.

Fig. 4 shows the boxplots for the trace elements. The patterns for Ba, Cd, Co and Sr follow the same sequence as observed for the major nutrients (leaves > wood > bark), however, the relative enrichment of these elements in the leaves is not as extreme. Mercury, La, Sb and Ti generally follow the pattern displayed by the minor nutrients (leaves > bark > wood). Silver (Ag), Au and Pb are three elements that show an opposite pattern, here the leaves display the lowest concentrations and the wood (Ag: bark) shows the highest concentrations. Kovalevsky (1987) mentions low Pb-concentrations in birch leaves compared to wood and

Table 1
Analytical results for birch leaves, bark and wood

BIL	Unit	DL	Birch leaves					Birch bark					Birch wood				
			Min.	Median	Max.	MAD	CV	Min.	Median	Max.	MAD	CV	Min.	Median	Max.	MAD	CV
Ag	µg/kg	2	2	5	11	1.48	39	5	13.5	27	4.45	32	6	11	30	2.97	45
As_ash	mg/kg	0.02	<0.02	0.06	0.36	0.052	99	0.1	0.245	0.76	0.111	49	<0.02	0.03	0.2	0.03	94
Au	µg/kg	0.2	<0.2	<0.2	2.1		157	<0.2	0.2	2.5	0.148	129	<0.2	0.5	4.4	0.297	95
B	mg/kg	1	16	29	39	5.9	21	2	3	4		24	2	2	4		23
Ba	mg/kg	0.1	24	78	385	56	80	4.3	9.8	56	5.9	75	7.5	16	73	8.0	65
Ca	mg/kg	100	6100	9850	18,800	2002	22	300	500	1500	148	46	500	700	1100	148	23
Cd	mg/kg	0.01	0.2	0.51	1.47	0.23	53	0.08	0.18	0.38	0.074	37	0.08	0.19	0.77	0.059	52
Co	mg/kg	0.01	0.06	0.51	1.93	0.326	62	0.02	0.07	0.3	0.03	71	0.01	0.12	0.37	0.074	61
Cr	mg/kg	0.1	0.7	0.9	1.3	0.148	14	0.7	0.9	1.1	0.148	11	0.8	1.1	22	0.148	203
Cu	mg/kg	0.01	4.5	5.8	9.9	1.09	17	4.3	5.3	7.4	0.689	14	1.1	1.6	2.2	0.297	16
Fe	mg/kg	10	40	70	180	14.8	35	10	20	200	14.8	98	<10	10	1100		452
Hg	µg/kg	1	11	18	28	4.45	21	5	12	26	4.45	37	1	2	5	1.48	45
K	mg/kg	100	5700	8400	12,200	1927	19	100	300	600	148	40	500	700	900	148	14
La	mg/kg	0.01	0.02	0.05	0.65	0.03	130	0.01	0.03	0.37	0.022	132	<0.01	0.01	0.28	0.007	195
LOI	wt%	0.01	93.2	95.3	96.8	0.52	0.71	99.2	99.7	99.8	0.15	0.14	98.9	99.7	99.7		0.13
Mg	mg/kg	10	1440	3650	5200	793	23	100	175	400	59	35	200	260	380	45	18
Mn	mg/kg	1	526	2138	4888	1427	58	192	611	2160	300	53	26	107	408	90.4	63
Mo	mg/kg	0.01	0.02	0.07	0.42	0.045	85	0.01	0.02	0.1	0.015	81	<0.01	0.01	0.13	0.007	128
Na	mg/kg	10	20	30	110	14.8	56	<20	20	40		32	20	20	30		15
Ni	mg/kg	0.1	0.8	2.95	10.3	2	70	0.2	0.4	1	0.148	51	0.1	0.2	0.5	0.148	53
P	mg/kg	10	1220	2065	5070	556	32	150	200	320	30	16	130	210	430	45	28
Pb	mg/kg	0.01	0.25	0.59	7.2	0.297	137	0.21	0.9	8.6	0.63	115	0.16	1.48	52	1.63	239
S	mg/kg	100	1200	1800	2900	445	21	<100	300	600	148	49	<100	200	400	148	52
Sb	mg/kg	0.02	0.02	0.03	0.33	0.015	114	<0.02	0.01	0.09		89	<0.02	<0.02	0.02		16
Sr	mg/kg	0.5	14	30	101	13	51	2.3	3.6	8.8	1.26	39	3.2	4.7	11.2	1.19	36
Ti	mg/kg	1	6	10	29	2.97	37	1	2	27	1.48	122	1	1	2		28
Zn	mg/kg	0.1	94	274	864	106	51	48	79	231	26.1	45	18	44	165	13.9	51

DL, detection limit; Min., minimum value; Max., maximum value; MAD, median absolute deviation; CV, coefficient of variation.

Table 2
Median values for mountain birch leaves (BIL), bark (BBA) and wood (BWO), sorted according to decreasing element concentration in birch leaves

Element	Unit	DL	BIL	BBA	BWO	Ratios				Soil		Soil/plant ratios		
			Median	Median	Median	Max./min.	BIL/BBA	BIL/BWO	BBA/BWO	O-horizon	C-horizon	O-hor/BIL	O-hor/BBA	O-hor/BWO
LOI	wt%	0.1	95.3	99.7	99.7					49	6.6	0.51	0.49	0.49
Ca	mg/kg	100	**9850**	<u>500</u>	700	20	19.7	14.1	0.7	2250	1350	0.23	4.5	3.2
K	mg/kg	100	**8400**	<u>300</u>	700	28	28.0	12.0	0.4	800	850	0.10	2.7	1.1
Mg	mg/kg	10	**3650**	<u>175</u>	260	21	20.9	14.0	0.7	1095	5500	0.30	6.3	4.2
Mn	mg/kg	1	**2138**	611	<u>107</u>	20	3.5	20.0	5.7	353	330	0.17	0.58	3.3
P	mg/kg	10	**2065**	<u>200</u>	210	10	10.3	9.8	1.0	985	525	0.48	4.9	4.7
S	mg/kg	0.01	**1800**	300	<u>200</u>	9	6.0	9.0	1.5	950	<200	0.53	3.2	4.8
Zn	mg/kg	0.1	**274**	79	<u>44</u>	6	3.5	6.2	1.8	100	57	0.36	1.3	2.3
Ba	mg/kg	0.1	**78**	<u>9.8</u>	16	8	8.0	5.0	0.6	104	59	1.3	11	6.6
Fe	mg/kg	10	**70**	20	<u>10</u>	7	3.5	7.0	2.0	13,750	26,200	196	688	1375
Sr	mg/kg	0.5	**30**	<u>3.6</u>	4.7	8	8.5	6.4	0.8	19	8	0.63	5.4	4.0
Na	mg/kg	10	**30**	20	20	2	1.5	1.5	1.0	70	60	2.3	3.5	3.5
B	mg/kg	1	**29**	3	<u>2</u>	15	9.7	14.5	1.5	2	2	0.07	0.67	1.0
Ti	mg/kg	1	**10**	2	<u>1</u>	10	5.0	10.0	2.0	232	530	23	116	232
Cu	mg/kg	0.01	**5.8**	5.3	<u>1.6</u>	4	1.1	3.6	3.3	13	16	2.2	2.4	8.0
Ni	mg/kg	0.1	**3.0**	0.4	<u>0.2</u>	15	7.4	14.8	2.0	9	27	3.1	23	45
Cr	mg/kg	0.1	0.9	0.9	**1.1**	1	1.0	0.8	0.8	12	33	13.3	13	11
Pb	mg/kg	0.01	<u>0.59</u>	0.9	**1.48**	3	0.7	0.4	0.6	89	12	152	99	60
Co	mg/kg	0.01	**0.51**	<u>0.065</u>	0.12	8	7.8	4.3	0.5	3.5	9.2	6.9	54	29
Cd	mg/kg	0.01	**0.51**	<u>0.18</u>	0.19	3	2.8	2.7	0.9	0.69	0.09	1.4	3.8	3.6
Mo	mg/kg	0.01	**0.07**	0.02	<u>0.01</u>	7	3.5	7.0	2.0	1.7	2.0	24	83	166
As_ash	mg/kg	0.02	0.06	**0.25**	<u>0.03</u>	8	0.2	2.0	8.2	4.7	4.8	78	19	157
La	mg/kg	0.01	**0.05**	0.025	<u>0.01</u>	5	2.0	5.0	2.5	15	28	300	600	1500
Sb	mg/kg	0.02	**0.03**	<0.01	<0.01					1.2	0.17	40	240	240
Hg	µg/kg	1	**18**	12	<u>2</u>	9	1.5	9.0	6.0	211	25	12	18	106
Ag	µg/kg	2	<u>5</u>	**13.5**	11	3	0.4	0.5	1.2	216	19	43	16	20
Au	µg/kg	0.2	<u><0.2</u>	0.2	**0.5**	5			0.4	1.5	0.2	15	7.5	3.0
pH		0.1								4.4	5.0			

Median values for the O-horizon (O-hor) and the C-horizon (C-hor) collected at the same sites as the plant leaves are also given (from Reimann et al., 2007). The ratio (median O-horizon/median birch) (O-hor/BIL, O-hor/BBA and O-hor/BWO) provides information about relative uptake/rejection of these elements in mountain birch leaves, bark and wood. Bold indicates highest value and underline indicates lowest value.
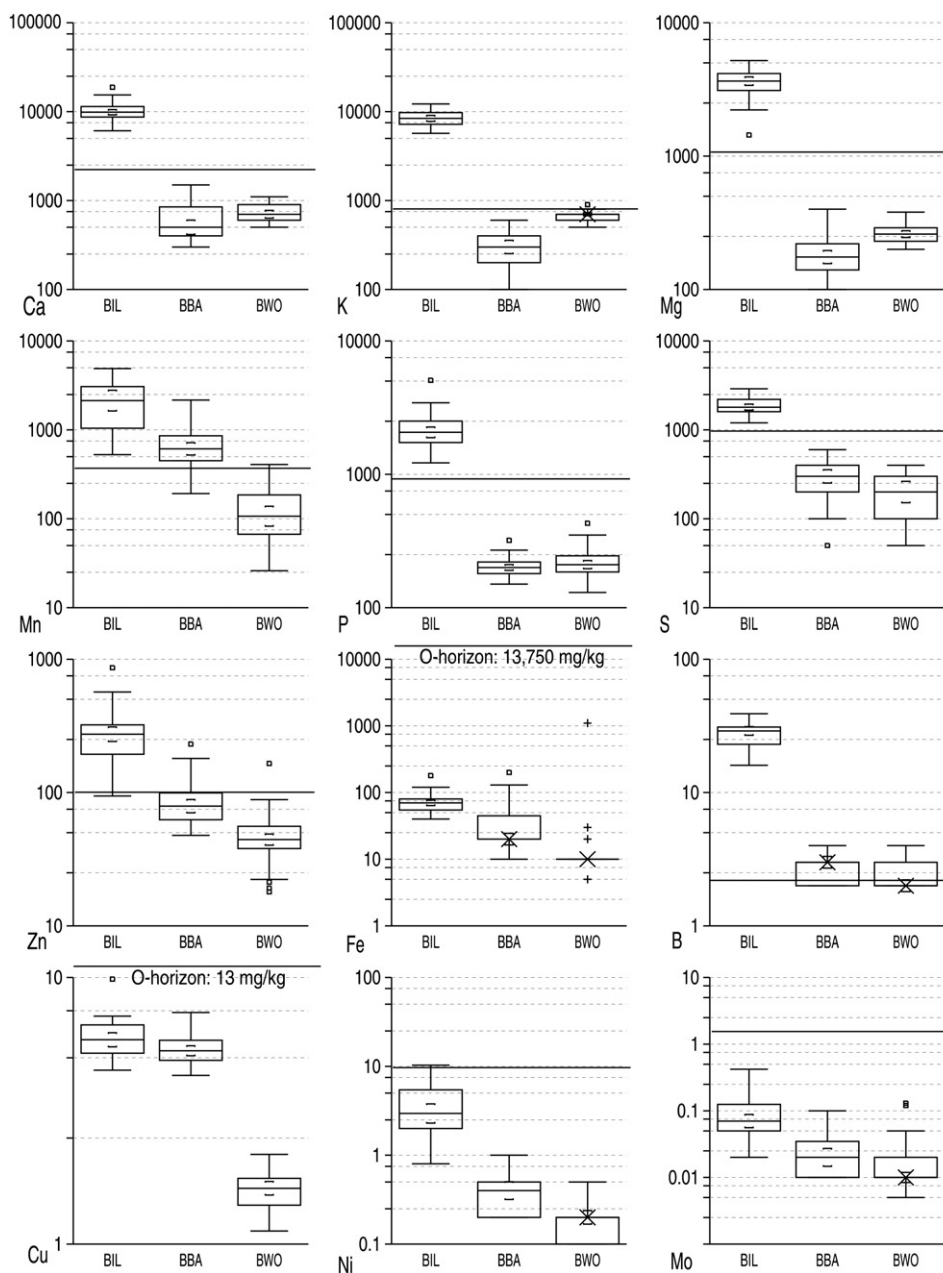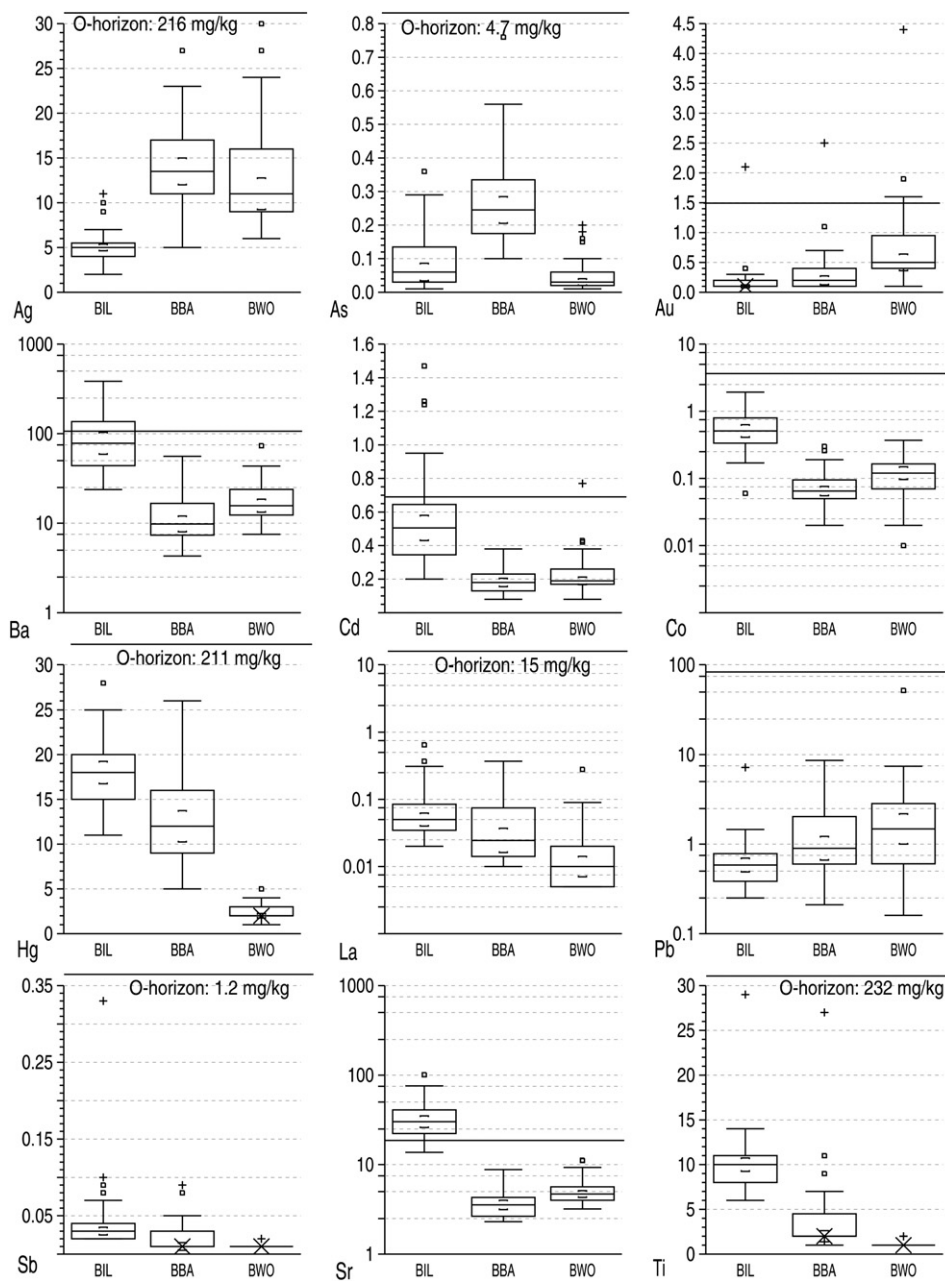
Fig. 3. Tukey boxplot (Tukey, 1977) comparison of element concentration and variation in mountain birch leaves (BIL), bark (BBA) and wood (BWO). Major (K, Ca, Mg, P and S) and minor (Mn, Fe, Zn, B, Cu, Ni, Mo) nutrients. The horizontal line gives the median concentration for the O-horizon (see Table 2). All in mg/kg.

bark. This is a clear indication of the importance of root-uptake of these elements for the concentrations observed in birch. It appears that the low amounts of Ag, Au and Pb that are taken up via the roots are preferably caught in the wood – most likely either via direct adsorption or bound to sulphide-groups. This is probably part of a detoxification strategy in birch. Arsenic behaves especially so, in

that it is strongly enriched in the bark. It is possible that inorganic As taken up from the soil is, for some unknown reason, transferred to organic As in the plant and ends up being strongly bound to the sub-erin in the bark. Some yet unknown positive effect of As in birch bark is, of course, also a possibility that would explain such an enrichment. The orders discussed here are based on the median values of all

Fig. 4. Tukey boxplot (Tukey, 1977) comparison of trace element concentration and variation in mountain birch leaves (BIL), bark (BBA) and wood (BWO), see Fig. 3 for key. Ag, Au and Hg in µg/kg, all others in mg/kg.

samples: when investigating each site in detail other orders are occasionally observed.

3.2. Soil-birch chemistry

Soil acidity and soil quality may also exert a profound influence on the availability of mineral elements to plants (e.g. Tyler and Olsson, 2001; Tyler, 2004). When plotting XY-diagrams of element concentrations in the O-horizon versus element concentrations in birch leaves, bark and wood, there are, however, very few elements that show a correlation. Of course, direct correlation analysis of soil and plant chemistry is always hampered by the fact that the chemical composition of the O-Horizon and the deeper mineral soil horizons

cannot be directly compared (e.g., Reimann et al., 2001b). Birch has deep roots and will take up substantial amounts of its nutrients and other elements from the deeper soil horizons and a high correlation between the chemistry of the O-horizon and birch would thus not be expected. Still, K, Sr and Zn show weak correlations ($r = 0.4–0.5$) between the concentrations found in the O-horizon and those in birch leaves and wood. The only element showing a high correlation between concentrations in the O-horizon and birch leaves, bark and wood is Pb (Fig. 5). Table 2 (soil/plant ratios) demonstrates at the same time that Pb shows some of the highest soil/plant ratios of all elements, i.e. it is strongly enriched in the O-horizon and thus excluded from uptake. Uptake of Pb into the plant is independent of the transport mechanisms of any of the nutrients. The small amounts taken up are either directly caught in the wood as part of the de-toxifying mechanisms of the plant or transported to the leaves to be disposed at the end of the season. Some Pb still reaches the bark, in which the correlation with the O-horizon is lowest because bark will, due to the long exposure time, collect more external dust (e.g., Pb from anthropogenic sources) than the leaves. In general the correlation is probably due to the fact that the Pb in the O-horizon has passed through birch before being deposited in the O-horizon rather than to uptake from the O-horizon. Cadmium, in contrast, is taken up with the important minor nutrient Zn and correlation with the concentrations in the O-horizon are much lower (see Fig. 5, lower left, $r = 0.42$) because here the need of the plant for the nutrients is the more important mechanism determining uptake, rather than the concentration in the substrate. No other elements show any clear relationships between concentrations in the O-horizon and concentration in birch leaves, bark or wood along the transect. The soil/plant ratios in Table 2 show that Fe and La, the elements with the highest soil/plant ratios, are clearly excluded from uptake indicated by increasing soil/plant ratios from leaves (greatest surface area) over bark (smaller surface area) to wood (not exposed). These two elements are thus good dust indicators for leaves and bark. Lead, in contrast, enters the plant via uptake from the deeper mineral soil horizons and not via deposition on the leaves as demonstrated by an increasing ratio from wood over bark to leaves.

When plotting element concentrations in the C-horizon against element concentrations as measured in birch leaves, bark and wood correlations are even weaker than those observed for the O-horizon. Only
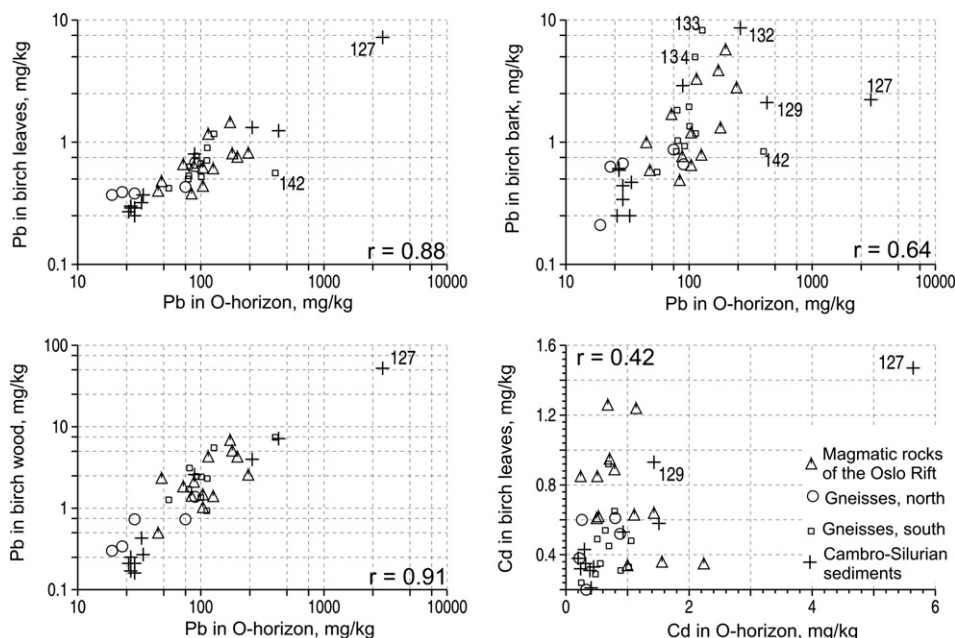

Fig. 5. Scatterplot of Pb concentrations in the O-horizon and in mountain birch leaves, bark and wood. The plot for Cd in the O-horizon versus Cd in mountain birch leaves is added to highlight the difference between a strongly excluded element (Pb) and an element that is involuntarily taken up with a nutrient (Cd with Zn). r = Pearson correlation coefficient for the log-transformed values.

four elements (Ba, Mo, Ni and Zn) show a clear influence of the element concentration as measured in the C-horizon on birch leaves, bark and/or wood. Barium is the most prominent example: concentrations in birch leaves, bark and wood are clearly related to the Ba-values in the C-horizon ($r = 0.57$, 0.44 and 0.57). For Zn there is a clear correlation between concentrations in birch leaves and wood and the C-horizon ($r = 0.47$, 0.53). Birch leaves and bark show such a correlation for Ni ($r = 0.60$, 0.45). Furthermore there is a correlation between Mo concentrations in the C-horizon and Mo concentrations in the birch leaves ($r = 0.46$). Due to the fact that plants are living organisms and can alter their chemical environment such correlation analyses should always be backed by scatterplots in order to be able to identify, for example, the existence of uptake barriers (Kovalevsky, 1987).

3.3. Observations along the transect

Several conditions change along the transect: distance to the coast (from south to north, see Figs. 1 and 2), distance to possible contamination sources (from Oslo towards the south and north – see Fig. 1), altitude (with a strong increase immediately to the north of Oslo on entering Nordmarka – see Fig. 6) and geology (four main lithologies crossed along the transect, see Figs. 2 and 6), which also has a strong influence on the pH in the O-horizon (Fig. 6). To better understand some of the main factors that may influence the element concentrations observed in the birch leaves, bark and wood it is thus important to study element concentration against location along the transect.

3.4. Natural factors

Reimann et al. (in press) have already demonstrated that B and Na-concentrations in the birch leaves are strongly influenced by the input of marine aerosols. Neither bark nor wood shows a relationship between B and Na-concentrations and distance to the coast. Geology is another important factor that will influence element concentrations in the soils. Along the transect the soils developed on top of the Cambro-Silurian sediments show unusually high concentrations for quite a number of elements (Reimann et al., 2007). Some of these patterns are directly reflected in the plant materials.

Plotting the Ni concentrations in birch leaves and bark along the transect reveals the highest Ni concentrations on top of the Cambro-Silurian sediments (Fig. 6). Although the first peak occurs in Oslo, this peak is clearly more related to geology (Cambro-Silurian sediments) than to anthropogenic influences. The location of site 134, the sampling site closest to a major highway demonstrates the anthropogenic influence on the pattern: this site displays by far the highest Ni concentration of any of the samples collected on the gneisses to the south of Oslo (Fig. 6). It appears that birch leaves react most strongly to the additional anthropogenic input. Such a double peak on top of the Cambro-Silurian sediments is observed for Ca (leaves, bark and wood), Cr (leaves and bark), Cu (bark – see Fig. 8), Fe (leaves – see Fig. 7), Mg (leaves, bark and wood), Hg (leaves – but a much stronger anthropogenic Oslo-peak than for Ni), while Ba shows a single strong peak in the Randsfjord area and a very weak peak in Oslo.

Due to the high Ca-concentrations in the rocks the pH values in the soils on top of the Cambro-Silurian sediments are highest (Fig. 6). Some elements are either immobilised by the high pH or their uptake is blocked by the high Ca, the most prominent example being Mn (Fig. 6), for which the values show exactly the opposite pattern to Ni, although Mn-concentrations in the soils are highest on top of the Cambro-Silurian sediments (Reimann et al., 2007). Manganese deficiency on soils developed on carbonaceous rocks has been known for a long time (e.g. Fitter and Hay, 1991). The Cd concentrations in birch leaves reflect this pattern as well, though not nearly as strongly as Mn. The importance of availability of major elements and soil pH for plant uptake is discussed in Tyler and Olsson (2001), Hayati and Proctor (1990) and Kidd and Proctor (2001): the questions arising from the above observations cannot be finally answered without data on the plant availability of these elements.

3.5. Dust as an element source in birch leaves and bark

Iron, La and Ti usually show much higher concentrations in soils than in plant materials (see Table 2, O-horizon/plant ratios). High concentrations of these elements in leaves and bark may thus provide a clear indication of dust playing an important role for the observed chemical composition. Fig. 7 shows analytical results for Fe, La and Ti in birch leaves and bark along the transect.
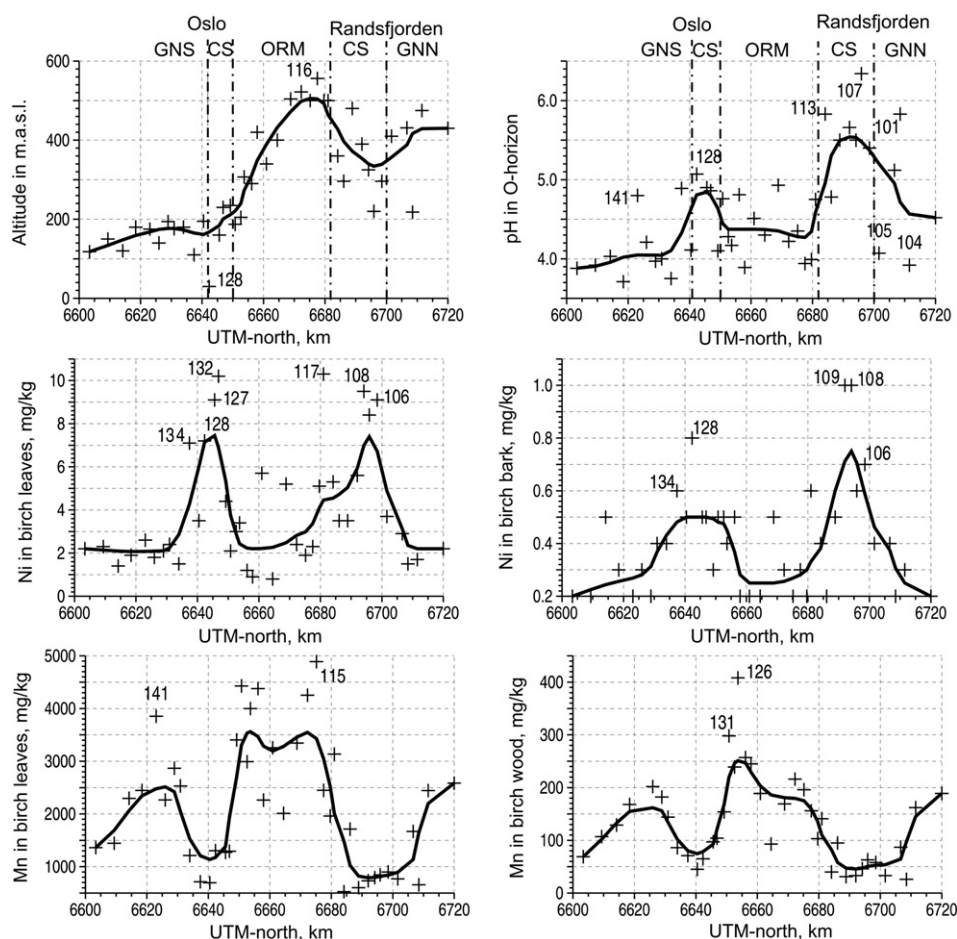
Fig. 6. Altitude and pH in the O-horizon (data from Reimann et al., 2007) along the Oslo transect. The borders between the major lithological units along the transect are indicated by stipulated lines (GNS: gneisses to the south of Oslo, CS: Cambro-Silurian sediments, ORM: Oslo Rift magmatic rocks, GNN: gneisses to the north of the Oslo Rift (upper two diagrams)). Ni in mountain birch leaves and bark (middle two diagrams) and Mn in mountain birch leaves and wood (lower two diagrams) show the importance of bedrock lithology and pH for the element concentrations observed in the plant materials. The smoothed lines shown are calculated using Tukey's running median (Tukey, 1977).

In birch bark a clear Oslo-peak emerges for all three elements. The highest value is observed at site 134, close to the major highway, the E6. This peak clearly marks the enhanced dust levels in the atmosphere around a city like Oslo. Note that for all three elements, background variation is reached at a distance of 15–20 km from the city centre.

For birch leaves such an Oslo peak is only observed for Fe (Fig. 7). However, a second, weaker Fe-peak occurs in the Randsfjord area and demonstrates that geology is quite important for the uptake of Fe into the leaves. Lanthanum shows an unusually high value at site 134 (near E6) but two other sites, in the area underlain by the magmatic rocks of the Oslo Rift, returned even higher values.

The magmatic rocks of the Oslo Rift and the overlying soils show unusually high concentrations of La (Reimann et al., 2007). Thus these high values could be both, an indication of high levels of local dust from forestry roads and/or of the uptake of La into the leaves from the underlying soils. Due to the fact that neither Fe nor Ti shows a peak at these sites, root uptake of La is the most likely source of these two anomalies. Although no biological role is known for La, it is one of the elements that is clearly taken up into plant materials (probably together with Ca) and several ferns are even known to be hyperaccumulators of La (e.g. Ozaki et al., 2000; Tyler, 2004; Reimann et al., in press). Titanium in birch leaves shows a single point anomaly at site
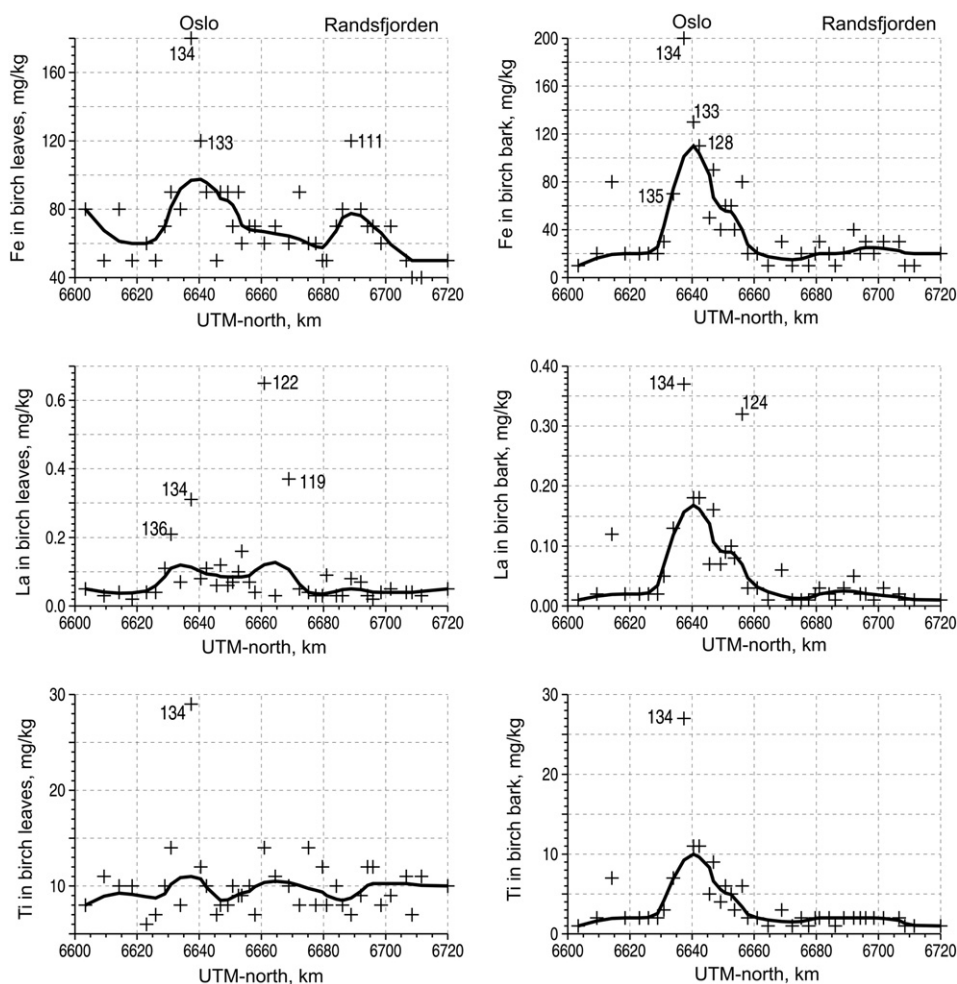
Fig. 7. Concentrations of Fe, La and Ti in mountain birch leaves and bark along the south–north transect through Oslo. For locations of sample sites, see Fig. 1. The smoothed line is calculated using Tukey's running median (Tukey, 1977).

134 (Fig. 7) and highlights the very local impact of the major highway on dust as observed on the birch leaves. Both La and Ti show such a peak at site 134 even in the wood, an indication that uptake possibly plays a more important role for the abnormal concentrations of these elements in the leaves than dust.

3.6. Suitability of birch as a biomonitor of urban contamination

Results presented by Reimann et al. (2006) for moss collected along the Oslo transect showed that the elements Ag, Al, Au, Bi, Cd, Co, Cr, Cu, Fe, Mo, Ni, Pb, Pt, Sb, Th, Ti and Zn all display a significant peak in moss samples collected in Oslo and its surroundings as compared to moss sampled along the remaining parts of the transect. These

authors concluded that moss can be used advantageously as a biomonitor for diffuse urban contamination. They showed that the main reason for moss reacting so strongly to the urban atmosphere lies in its ability to collect fine dust from dry and wet deposition (Reimann et al., 2006). Mosses are especially suitable for accumulating metals because of their large surface area to volume ratio and have long been known to collect metals in particulate form on their whole surface (Brown, 1982). The use of plant materials as biomonitors of anthropogenic impact is a much discussed topic (see Markert, 1993).

When studying element concentrations in birch leaves, bark and wood for elements that are often typically related to urban contamination (e.g. Ag, As, Au, Cd, Co, Cr, Cu, Hg, Mo, Pb, Sb, Zn)

along the transect quite different patterns emerge. Figs. 8 and 9 show the transect for the elements Co, Cu, Pb and Sb as some typical examples. Cobalt (with Cd, Pb and Zn) is one of the few elements that shows an Oslo-peak in all three materials (Fig. 8). The peak is most pronounced in bark (dust-related?): in leaves and wood the peak is less visible and a steady decline of Co-values to the north of Oslo (with increasing altitude – compare with Fig. 6) is a pronounced feature. At site 134 clearly unusual Co concentrations are observed in all three materials. Copper, in contrast, shows a single point anomaly in the leaves collected at site 134. A clear Oslo peak emerges for Cu in bark and site 134 again shows the highest value. However, there is clearly a geogenic influence as well, a second Cu-peak occurs in the Randsfjord area. In birch wood no influence from Oslo is visible in the Cu concentrations in the wood, though the second-highest Cu value occurs at site 134. It would thus appear that the peak in bark is most likely directly dust-related.

On first glance Pb appears to show a clear Oslo peak in all three materials. However, on close scrutiny, it becomes doubtful that urban contamination is the main reason for the peak. Site 134 (E6) is marked by rather low values in leaves and wood. Only bark (dust) shows a high value at this site. Site 127, the location of a geogenic Pb anomaly (Reimann et al., 2007), related to mineralisation in black shales, shows the highest Pb-concentration in both, leaves and wood (uptake from the soil!). In addition the whole peak is somewhat displaced towards the north (Fig. 9). The little depression that can be
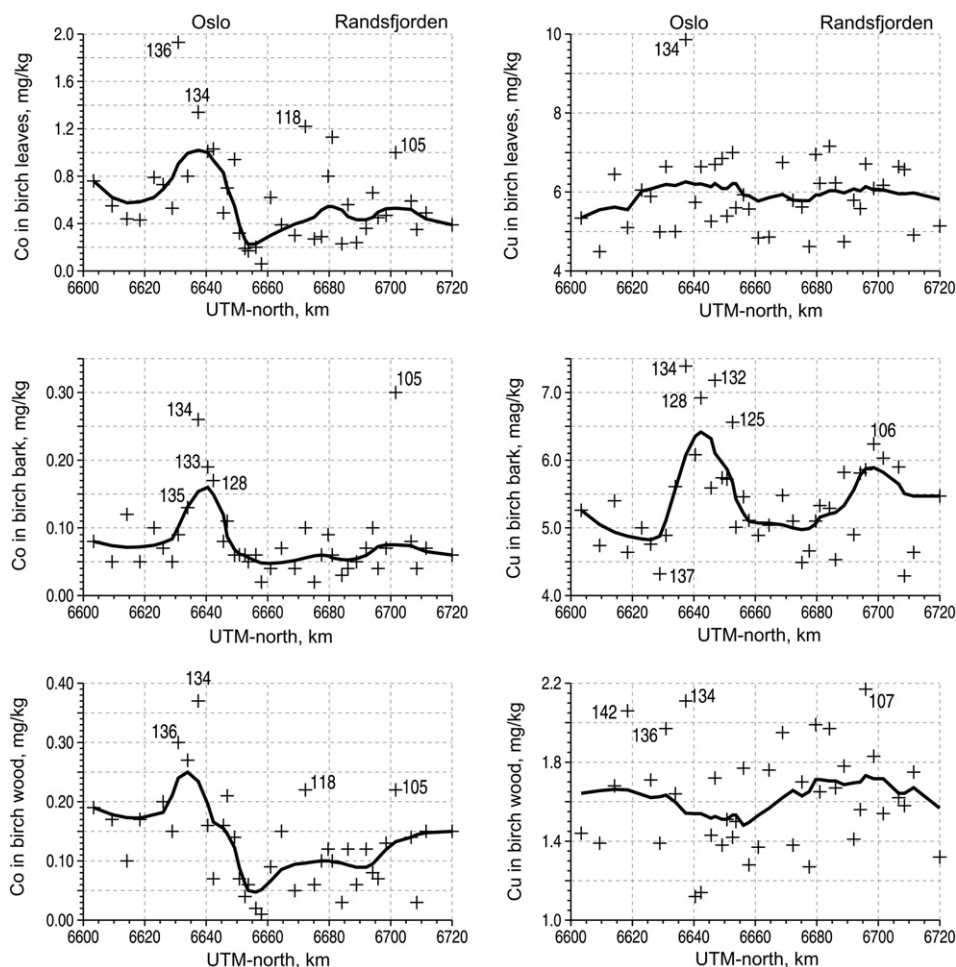

Fig. 8. Concentrations of Co and Cu in mountain birch leaves, bark wood along the south–north transect through Oslo. For locations of sample sites, see Fig. 1. The smoothed line is calculated using Tukey's running median (Tukey, 1977).
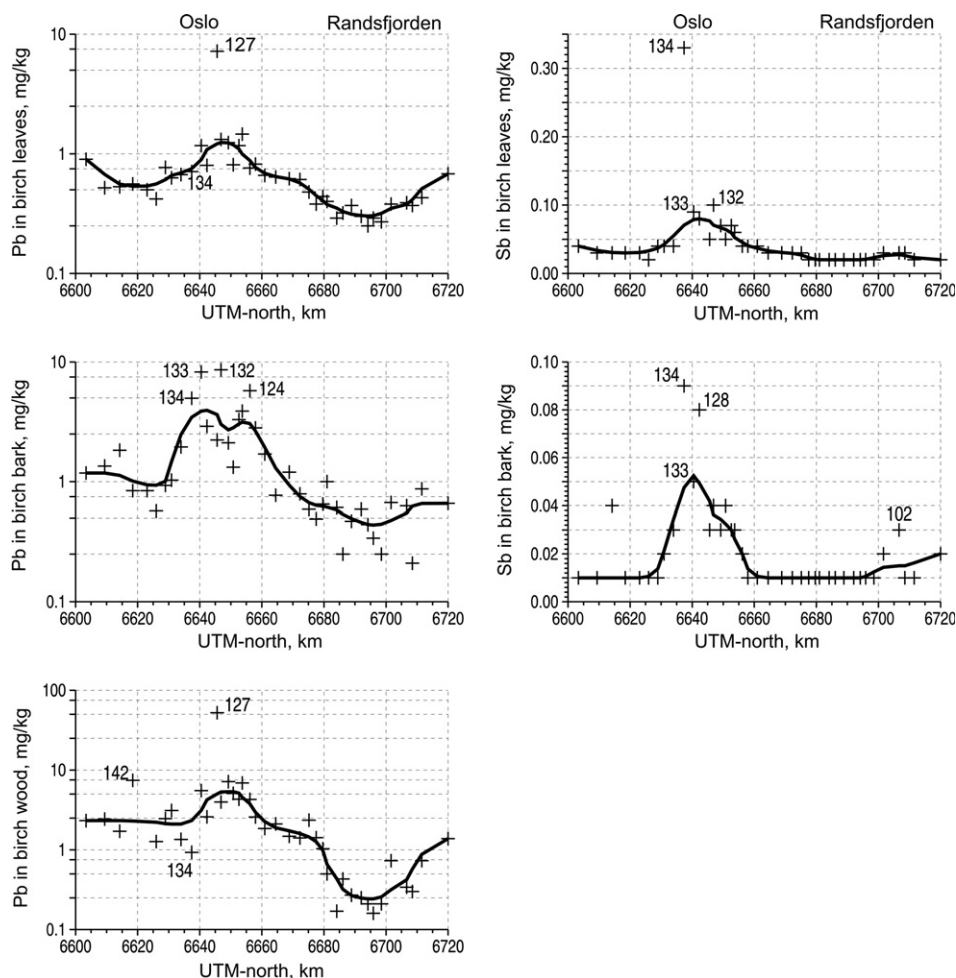
Fig. 9. Concentrations of Pb and Sb in mountain birch leaves, bark wood (wood only Pb because all values for Sb in birch bark are lower than the detection limit) along the south–north transect through Oslo. For locations of sample sites, see Fig. 1. The smoothed line is calculated using Tukey's running median (Tukey, 1977).

noted for birch bark at the northern fringe of Oslo is due to the occurrence of Cambro-Silurian sediments (see Randsfjord area). The transects for Cd, Pb and Zn all show a shift towards the north of Oslo, into Nordmarka (see Fig. 9, Pb). Although this is the main wind direction, it is still surprising that the highest values do not occur directly at the source. It is thus questionable whether the observed peaks are caused by a combination of low-level contamination from Oslo and special local conditions (microclimate) and are thus of anthropogenic origin. They might as well be related to numerous minor mineral showings that occur in this area. The presence of ore showings in the area has resulted in quite typical location names on the topographical map (e.g. "Kopperhaugene" – "copper hills"). The usefulness of plant materials for biogeo-

chemical exploration for ore deposits has a long history (e.g. Brooks, 1972; Kovalevsky, 1987).

Antimony, in contrast, displays a clear peak in Oslo, most pronounced in bark (all values in wood were below the detection limit). In both, leaves and bark, site 134 shows the expected highest concentration. The bark peak most resembles the dust peak as observed for Fe, La and Ti (compare Figs. 7 and 9). Birch leaves and bark also display a clear Oslo-peak for Hg (see also Reimann et al., in press). Moss, in contrast, did not show an Oslo-peak for Hg (Reimann et al., 2006). Birch bark displays a peak for Mo.

All in all, the chemical composition of birch leaves, bark and wood is clearly affected by the urban environment. The impact is predominantly very local (often single site) and even for elements

showing a stronger Oslo-peak all values reach background variation at a distance of less than 20 km from the centre of Oslo. Such a limited impact from anthropogenic contamination and the rapid decline of observed element concentrations with distance to source is in good agreement with many highway studies, usually showing an exponential decrease over the first few to some tens of meters in element concentrations (e.g., Pb, Pd, Pt) with distance to the road (e.g., Caselles, 1998; Jaradat and Momani, 1999; Pagotto et al., 2001; Ayrault et al., 2006) in plant materials or roadside soils. Results from other urban studies (e.g. Tiller et al., 1987; Ho and Tai, 1988) all show the limited impact (up to 50 km) of a city on the element levels observed in plants and soils in their surroundings.

4. Conclusions

Several factors influence the observed element concentrations in birch along the Oslo transect. One important factor is geology, i.e. the chemical composition of the substrate on which the soils supplying the nutrients have developed. High Ca, Mg and Ni-concentrations in soils developed on top of the Cambro-Silurian sediments are directly reflected in birch leaves, bark and wood. Barium concentrations in the three materials are also clearly influenced by the concentrations in the substrate. High Ca-values in the soils, well reflected in the plants (and in the soil pH), result in a reduced Mn-uptake, even if the soils contain high Mn levels. The only other element showing a similar behaviour to Mn is Cd. Mineral occurrences do also have an impact on element concentrations as observed in birch leaves, bark and wood: values for Ag, Cd, Mo, Pb and Zn are affected locally. The transects for Cd, Pb and Zn show a displaced "Oslo-peak", that could be easily misinterpreted as an anthropogenic peak, in leaves, bark and wood due to these mineral occurrences. Because several small mineral showings occur immediately to the north of Oslo it is here difficult to separate geogenic from anthropogenic element sources beyond doubt, though a shift of the peak towards the north provides evidence of the importance of mineralisation for at least some of the observed high values. There appear to be patterns at two different scales: at a very local scale and contamination or mineralisation are typical examples, and at a regional scale, which is dominated by natural factors like geology and climate. To identify contamination or minerali-

sation beyond doubt will often require a geochemical investigation at even higher densities than that chosen in this study. Regional factors (e.g. relation to lithology, climate, distance to coast (sea spray)) influence the geochemical patterns at a much larger scale and would even be detected using much lower sample densities.

The steady input of marine aerosols near the coast can have a pronounced influence on leaf chemistry (e.g. B and Na), but not however, on the chemical composition of birch bark and wood.

The reaction of birch leaf, bark and wood chemistry to the anthropogenic Oslo contamination signal is quite individual for each element and material. The urban environment affects birch-bark chemistry most visibly and the mechanism is the collection of local dust on the birch bark. Birch bark shows the strongest impact of urban pollution due to the long exposure time (years) when compared to leaves (<6 months) even taking the lower surface area of bark into consideration. Iron, La and Ti, the three most prominent dust indicator elements, all display a pronounced Oslo peak. Of the metals, Mo and Sb are the two elements that show a directly comparable Oslo peak in birch bark. Even these peaks disappear in the background variation at a distance of less than 20 km from the centre of Oslo. Many elements show a single high value at site 134, closest to highway E6, in leaves, bark and/or wood. Mercury shows a strong Oslo-peak in birch leaves and bark. Compared to terrestrial moss, birch leaves, bark and wood appear to be of quite limited value as biomonitors of urban contamination.

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