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Critical remarks on the use of terrestrial moss (*Hylocomium splendens* and *Pleurozium schreberi)* for monitoring of airborne pollution

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"Capsule": *Moss species can be combined for regional mapping purposes.*

Abstract

Duplicate samples of the two terrestrial moss species *Hylocomium splendens* and *Pleurozium schreberi*, which are widely used to monitor airborne heavy metal pollution, have been collected from eight catchments spread over a 1,500,000 km^2 area in northern Europe. These were analysed for a total of 38 elements by inductively coupled plasma-mass spectrometry, inductively coupled plasma-atomic emission spectrometry and cold vapour-atomic absorption spectometry techniques. Results show that the moss species can be combined without interspecies calibration for regional mapping purposes. For the majority of elements the observed within-catchment variation is large — big composite samples over a large area should thus be collected when moss is to be used for monitoring purposes. For the majority of elements the input of dust governs moss chemistry. For a reliable 'contamination' signal over a sizeable area a major source is needed. Some elements show a dependence on climate/vegetation zone. In coastal areas the input of marine aerosols will alter the chemical signal obtained from moss samples. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Terrestrial moss; Environmental monitoring; Northern Europe; Airborne pollution; Heavy metals

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1. Introduction

Terrestrial moss (especially *Hylocomium splendens* and *Pleurozium schreberi*) has been used successfully to map and monitor airborne heavy metal pollution in northern European countries for more than 20 years (e.g. Rühling and Tyler, 1973; Niskavaara and Äyräs, 1991; Rühling, 1994; Takala et al., 1994; Nash and Gries, 1995; Steinnes, 1995; Äyräs et al., 1997; Reimann et al., 1997; Rühling and Steinnes, 1998). *H. splendens* and *P. schreberi* have been used as biomonitors in regional mapping of airborne pollution in northern Europe at 5-year intervals since 1985 (Rühling and Steinnes, 1998). For this purpose terrestrial mosses have several advantages when compared to higher plants: they lack a developed root system; they lack the variability of morphology through the growing season that many other plants have; they lack cuticula; they have a high surface-to-volume ratio; and they exhibit ion exchange properties (e.g. Gjengedal and Steinnes, 1990). It is assumed that they receive their nutrients directly from the atmosphere. Berg et al. (1995) and Berg and Steinnes (1997) found significant correlation between wet deposition of many elements and their concentration in

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moss (*H. splendens*). Steinnes (1995) and Caritat et al. (2000) discussed problems associated with using mosses as biomonitors of atmospheric pollution and concluded that local dust and interaction of the moss with local litter and soil should not be underestimated as element sources.

In northern Europe the two carpet-forming moss species *H. splendens* and *P. schreberi* are very common. Still, to give regional cover, both species had to be combined in all regional mapping projects so far. Thus, estimates of interspecies variability, especially between *H. splendens* and *P. schreberi* are given in several studies (e.g. Ramenskaya, 1974; Folkeson, 1979; Rinne and Mäkinen, 1988; Ross, 1990; Wolterbeck et al., 1995; Thöni et al., 1996; Berg and Steinnes, 1997; Halleraker et al., 1998). With the exception of Halleraker et al. (1998) these studies cover a limited number of elements only. Halleraker et al. (1998) compared the results of all previous studies and came to the conclusion that the results presented in all these studies do not agree very well. Some authors suggest calibration between the results obtained for *Hylocomium* and *Pleurozium*. Others advise that although there are some differences in chemical composition between the two species calibration should be avoided because differences in analytical results from these two species differ with deposition conditions (Halleraker et al., 1998). One reason for these differences in the results of these previous studies could be that they were never carried out systematically over a large enough geographical region.

A total of 140 duplicate samples of *H. splendens* and *P. schreberi* were taken in eight catchments spread over a 1.5 million km^2 area. This area includes north-western Russia from St. Petersburg to the northern end of the Ural Mountains, northern Norway and Finland (Fig. 1). A major geochemical mapping project is planned to cover this whole area in the years 2000–2003, the "Barents Ecogeochemistry Project". During 1999 nine catchments located at widely spaced intervals throughout the 1,500,000 km^2 area were visited. A minimum of 11 moss samples were collected well spread over each of eight (out of nine) of the catchments. These varied in size between 14 and 50 km^2 (Table 1). Depending on availability duplicate samples of *Hylocomium* and *Pleurozium* were taken from 4 to 11 sites per catchment. Table 1 summarises important information about the visited catchments (location, landscape, climate, vegetation zone, geology, geochemical characteristics of the parent soil material). Note that no moss could be collected in C6, Monchegorsk, which is in the immediate vicinity of the Monchegorsk nickel refinery. Due to extreme levels of pollution almost all vegetation is destroyed there.

In this paper analytical results for 38 chemical elements of duplicate samples of *Hylocomium* and *Pleurozium* from eight catchments will be used to present an interspecies comparison. Results for both species combined are then compared between the catchments to allow a judgement of the relative importance of local versus regional variability. Finally the results are used to discuss the processes influencing moss chemistry in these catchments.

2. Methods

2.1. Sampling

All samples were collected as described by Äyräs and Reimann (1995) in accordance with the methods used in the European moss monitoring project (Rühling, 1994). Every effort, starting in the field, was taken to collect comparable samples of each species. Trace element-free vinyl gloves were used during sample collection and smoking was strictly prohibited. To avoid the influence of throughfall precipitation and stemflow, the field manual (Äyräs and Reimann, 1995) requires that samples should be taken at least 5 m away from any trees and larger bushes. This is not always easy to achieve in densely forested areas, especially when collecting plant species that tend to grow in the shade. In any case, no samples that could be influenced by stemflow were collected. About 1 l of the required cleaned (from other species and litter) moss species was collected at each site as a composite sample from an area measuring at least 50×50 m. Samples were placed in white plastic-lined, trace element-free bags and marked with catchment and site number. The bags had to be opened in the evening in the field camp to allow the moss to air dry. PE gloves were worn whenever the bags were opened. After arrival in the laboratory of the Geological Survey of Finland (GTK) all samples were dried to constant weight at 40°C.

2.2. Sample analyses

The dried samples were checked again for any contamination by other plant species or litter prior to milling in a centrifugal mill to pass through a 1-mm sieve. Tests were run to prove that neither milling nor sieving could contaminate the samples with any of the 38 elements on the analytical program. A 0.5-g aliquot of sample material was dissolved in 10 ml of concentrated nitric acid (Merck suprapure) in a microwave oven. The samples were then diluted to 50 ml with de-ionised water before being analysed for 38 elements by inductively coupled plasma-mass spectrometry, inductively coupled plasma-atomic emission spectrometry and cold vapour-atomic absorption spectroscopy (CV-AAS) techniques (CV-AAS for Hg only). The procedure closely followed the US-EPA-standard 3051. GTK's laboratory is accredited to meet the requirements of the EN-standard 45001 and ISO-guide 25. Table 2

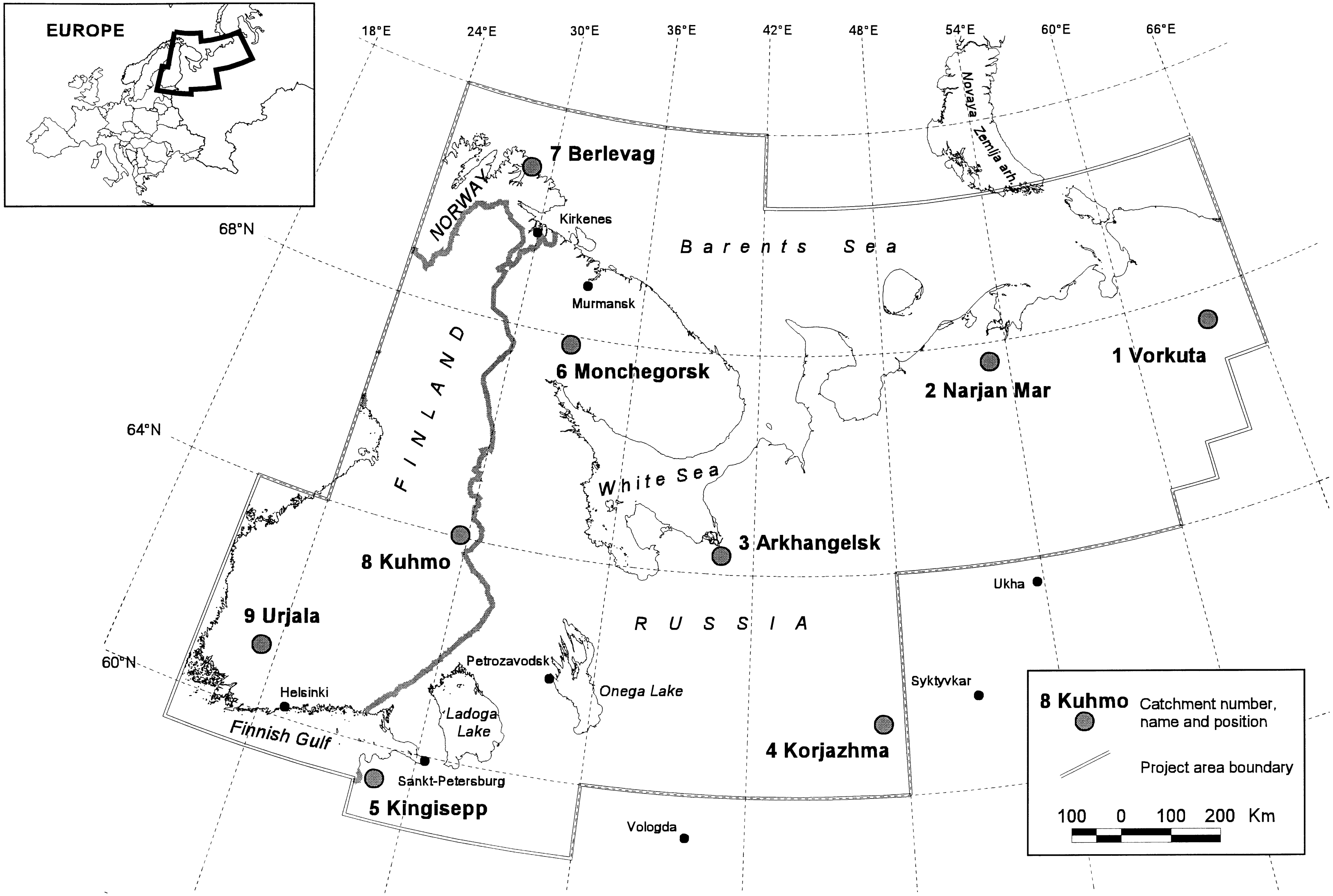

Fig. 1. The Barents Project area and location of the eight catchments discussed here within the project area.

Table 1
General characteristics of the eight catchments where moss samples were collected[a]

Catchment No.	Name	Outlet coordinates	Area (km^2)	Altitude (m a.s.l.)	Annual precipitation (mm)	Vegetation	Bedrock	Quaternary deposits	Special geochemical characteristics of the C-horizon
Russia									
C1	Vorkuta	67°27′9″ N 63°49′38″ E	19.7	140–220	600	Moss–lichen shrub tundra with birch and willow	Sandstone, siltstone, argillite, coal seams	Alluvial, fluvioglacial, glacial and marine glacial deposits	High: As, Be, Cd, Sb, V
C2	Narijan-Mar	67°39′18″ N 53°03′39″ E	16.8	5–15	580	Southern tundra with birch and shrub	Clay, sandstone, siltstone and brown coal	Lake-glacial and lacustrine deposits	Low: Ag, Al, Ba, Ca, Cd, Cr, Cu, Mg, S, Sr, V, Zn
C3	Arkhangelsk	64°22′06″ N 40°37′55″ E	24.4	24–64	650	Northern taiga with coniferous forest	Sandstone, siltstone dolomite, limestone	Glacial-marine, lake-alluvial, lake-bog sediments	High: B, Ba
C4	Korjazhma	61°18′39″ N 47°08′50″ E	40.3	40–80	780	Middle taiga	Siltstone, limestone, clay	Modern alluvial, lake-glacial deposits	Low: Ag, As, Be, Bi, Cd, Co, Cr, Cu, Fe, K, P, Sb, Sc, Sr, V, Zn
C5	Kingisepp	59°24′15″ N 28°34′59″ E	36.6	10–60	710	Southern taiga	Marl, limestone, dolomite, siltstone, sandstone, clay	Modern alluvial, modern lake, lake-glacial, fluvioglacial and glacial deposits	High: Ag, B
C6	Monchegorsk	67°48′29″ N 32°47′08″ E	13.6	150–400	391	Northern taiga. Almost completely destroyed vegetation cover	Metagabbroid, tuff, tufflava, gabbro, gabbronorite, gabbrolabradorite	Modern alluvial, lake-glacial, fluvioglacial deposits, till, exposed bedrock	High: Al, Ca, Cr, Cu, Na, Ni, V low: As, Sb
Norway									
C7	Berlevåg	70°51′32″ N 28°55′37″ E	46.7	20–320	850	Arctic tundra	Sandstone, phyllite, siltstone	Till, marine shore deposits, fluvial deposits, till, exposed bedrock, bog	High: Al, As, Bi, Co, Cu, Fe, Li, Mg, Ni, P, Pb, Sb, Th, Zn
Finland									
C8	Kuhmo	64°03′17″ N 29°46′33″ E	36.0	170–270	750	Mixed forest with spruce, pine and birch	Leucocratic granitegranodiorite, mafic metavolcanite, tonalite, trondhjemite and migmatite	Till, peat, exposed bedrock	Low: As, Be, Bi, S, Sr
C9	Urjala	61°06′58″ N 23°25′56″ E	40.1	100–140	650	Mixed forest with spruce, pine and birch	Granodiorites, tonalites, quartz diorites, mica shist, mica gneiss	Till, peat, exposed bedrock	High: Ag, As, K, Mo, S, Se, Th, Ti

[a] Note that no moss could be collected in C6, Monchegorsk due to the high pollution level.

Table 2
Analytical program, methods used for the different elements and detection limits[a]

	Element	Techn.[a]	Accr.	DL	Unit
1	Ag	ICP-MS	YES	0.005	mg/kg
2	Al	ICP-AES	NO	15	mg/kg
3	As	ICP-MS	YES	0.02	mg/kg
4	B	ICP-MS	YES	0.5	mg/kg
5	Ba	ICP-MS	YES	0.05	mg/kg
6	Be	ICP-MS	YES	0.01	mg/kg
7	Bi	ICP-MS	YES	0.002	mg/kg
8	Ca	ICP-AES	NO	20	mg/kg
9	Cd	ICP-MS	YES	0.005	mg/kg
10	Co	ICP-MS	YES	0.02	mg/kg
11	Cr	ICP-MS	YES	0.2	mg/kg
12	Cu	ICP-MS	YES	0.02	mg/kg
13	Fe	ICP-AES	NO	5	mg/kg
14	Hg	CV-AAS	YES	0.04	mg/kg
15	K	ICP-AES	NO	30	mg/kg
16	Li	ICP-MS	YES	0.03	mg/kg
17	Mg	ICP-AES	NO	10	mg/kg
18	Mn	ICP-AES	NO	0.5	mg/kg
19	Mo	ICP-MS	YES	0.01	mg/kg
20	Na	ICP-AES	NO	20	mg/kg
21	Ni	ICP-MS	YES	0.3	mg/kg
22	P	ICP-AES	NO	10	mg/kg
23	Pb	ICP-MS	YES	0.02	mg/kg
24	Rb	ICP-MS	YES	0.01	mg/kg
25	S	ICP-AES	NO	20	mg/kg
26	Sb	ICP-MS	YES	0.01	mg/kg
27	Sc	ICP-AES	NO	0.03	mg/kg
28	Se	ICP-MS	YES	0.5	mg/kg
29	Si	ICP-AES	NO	20	mg/kg
30	Sn	ICP-MS	YES	0.01	mg/kg
31	Sr	ICP-MS	YES	0.01	mg/kg
32	Th	ICP-MS	YES	0.02	mg/kg
33	Tl	ICP-MS	YES	0.005	mg/kg
34	U	ICP-MS	YES	0.005	mg/kg
35	V	ICP-MS	YES	0.02	mg/kg
36	Y	ICP-MS	YES	0.001	mg/kg
37	Zn	ICP-AES	NO	0.4	mg/kg
38	Zr	ICP-MS	YES	0.01	mg/kg

[a] Techn., decomposition in conc. HNO_3 in a microwave oven and finish by: ICP-AES, inductively coupled plasma-atomic emission spectrometry; ICP-MS, inductively coupled plasma-mass spectrometry; CV-AAS, cold vapour-atomic absorption spectroscopy. Accr., the technique is covered by the laboratories accreditation YES/NO. DL, detection limit.

summarises the analytical program, the method used to determine the different elements and the detection limit for each element. Analytical precision as judged from frequent duplicate analyses was generally better than 5%. A microwave digestion in concentrated nitric acid completely dissolves all organic material. Mineral particles (e.g. silicates) which might be incorporated in the moss will usually not dissolve completely.

2.3. Data analysis

Due to the limited number of samples and locations, exploratory data analysis (EDA) methods (Tukey, 1977; Velleman and Hoaglin, 1981) were used to study and present our data. Kürzl (1988), Rock (1988) and O'Connor and Reimann (1993) give an introduction to the advantages of using exploratory data analysis methods when dealing with geochemical data. The construction of the box plot used in the graphics follows Tukey's (1977) original definition (whiskers to last real data point up to 1.5×hinge spread), data outliers are marked by squares ($>1.5\times$hinge spread) or plusses (far outliers $>3\times$hinge spread). Coincidence of the median and one (or both) of the hinges is marked by a cross. The notches (square brackets), placed at 1.58×(hinge spread/$\sqrt{(n)}$) on either side of the median, are a test of the significance of medians from different populations or boxes. Values below detection were set to one half the detection limit for the purpose of graphical data analysis. Note that the construction of the boxplot requires a minimum of five data points. In cases where less than five analyses are available, a line in the graphics marks the individual results. To detect inter-element relations in the whole data set a robust factor analysis (Filzmoser, 1999; Pison et al., 2000) with varimax-rotation was used after checking all data graphically in a draftman's display for artificial high or low correlation.

3. Results and discussion

Analytical results for both species are summarised in Table 3. This table also shows the correlation coefficient between *Hylocomium* and *Pleurozium*, and the median ratio *Hylocomium*/*Pleurozium*. Table 4 gives the results for *Hylocomium* and *Pleurozium* combined (moss). The table also shows the relative variation for each element based on the median absolute deviation and the (maximum value/minimum value)-ratio. For comparison the median and maximum results from the Kola Ecogeochemistry project, a regional geochemical mapping project in which 598 samples of terrestrial moss were collected over a 188,000 km^2-area in the European Arctic (Reimann et al., 1998) are also included. Ratios for the median and maximum concentrations found are also given.

At first glance analytical results for *Hylocomium* and *Pleurozium* are quite comparable (Table 3). In both species K, Ca, P, Mg and S show the highest concentration, Be and Bi the lowest. Fig. 2 shows all analytical results for *Hylocomium* and the variation of each element in a boxplot comparison sorted according to decreasing median concentration. For *Pleurozium* the sequence of elements is exactly the same with the one exception that Mg and S exchange place (Mg > S in *Pleurozium*). Fig. 2 shows that there are elements with a low variability (tight box). These are primarily the major and minor plant nutrients (e.g. K, Ca, P, S, Mg,

Table 3
Analytical results in mg/kg (median [MED], median absolute deviation [MAD], and minimum [MIN] and maximum [MAX] values) for *Hylocomium splendens* ($n = 70$) and *Pleurozium schreberi* ($n = 70$) as collected in all catchments from the project area[a]

Element	*Hylocomium*				*Pleurozium*				Correlation	MEDIAN
	MIN	MAX	MAD	MED	MED	MAD	MIN	MAX	HYL/PLE	HYL/PLE
Ag	0.012	0.056	0.006	0.025	0.025	0.006	0.013	0.064	0.68	1.0
Al	87.9	2380	191	359	350	177	88.8	1550	0.78	1.0
As	0.058	1.11	0.120	0.278	0.194	0.070	0.054	0.911	0.84	1.4
B	< 0.5	46.62	1.9	3.92	2.78	1.57	< 0.5	10.6	0.87	1.4
Ba	8.11	75.76	7.3	22	21	6.80	7.33	83.0	0.76	1.1
Be	< 0.01	0.087	0.012	0.017	0.015	0.010	< 0.01	0.122	0.81	1.1
Bi	0.007	0.029	0.005	0.017	0.015	0.003	0.007	0.030	0.67	1.2
Ca	1800	10900	870	3110	3025	805	1690	7170	0.87	1.0
Cd	0.032	0.536	0.031	0.134	0.129	0.031	0.035	0.267	0.71	1.0
Co	0.051	1.702	0.124	0.293	0.285	0.114	0.056	2.13	0.77	1.0
Cr	0.230	5.84	0.600	1.16	0.99	0.51	0.22	5.01	0.65	1.2
Cu	3.11	8.73	0.960	5.36	4.31	0.700	2.45	17.48	0.73	1.2
Fe	107	2390	220	462	411	205	103	1860	0.79	1.1
Hg	< 0.04	0.477	0.010	0.060	0.051	0.010	< 0.04	0.765	0.63	1.2
K	2970	8140	780	4765	5575	680	3780	9260	0.73	0.9
Li	0.032	1.09	0.088	0.210	0.184	0.095	< 0.03	0.931	0.76	1.1
Mg	544	1650	216	948	1060	235	565	2020	0.90	0.9
Mn	33.3	848	106	385	365	127	47.4	1080	0.75	1.1
Mo	0.055	0.307	0.045	0.125	0.079	0.029	0.035	0.253	0.95	1.6
Na	27.2	244	21.3	65	58	18.4	25.6	348	0.96	1.1
Ni	0.71	5.76	0.950	2.39	2.16	0.770	0.730	6.16	0.90	1.1
P	760	2830	260	1475	1325	220	700	2780	0.83	1.1
Pb	1.65	11.9	0.790	4.14	4.11	0.680	2.18	9.86	0.77	1.0
Rb	1.33	42.2	5.69	12.9	12.5	5.43	2.18	47.9	0.96	1.0
S	610	1710	19.6	1020	960	192	613	2020	0.90	1.1
Sb	0.023	0.196	0.027	0.071	0.055	0.019	0.015	0.180	0.86	1.3
Sc	< 0.03	0.510	0.050	0.090	0.080	0.040	< 0.03	0.480	0.79	1.1
Se	< 0.5	< 0.5		< 0.5	< 0.5		< 0.5	< 0.5		1.0
Si	102	627	78	335	305	104	101	551	0.89	1.1
Sn	0.034	0.148	0.019	0.075	0.065	0.013	0.030	0.129	0.49	1.2
Sr	3.48	37.8	3.99	11.9	11.0	3.10	4.23	33.75	0.89	1.1
Th	< 0.02	0.582	0.031	0.059	0.051	0.029	< 0.02	0.636	0.81	1.2
Tl	0.011	0.214	0.027	0.054	0.037	0.022	0.007	0.161	0.89	1.4
U	0.007	0.157	0.02	0.034	0.026	0.011	0.006	0.16	0.90	1.3
V	0.67	9.66	1.28	2.41	2.33	1.19	0.6	8.94	0.91	1.0
Y	0.036	1.82	0.119	0.177	0.166	0.100	0.036	1.36	0.92	1.1
Zn	17	63.1	5.95	33.4	34.4	6.1	19	54.3	0.61	1.0
Zr	0.068	1.57	0.205	0.373	0.369	0.171	0.080	1.81	0.72	1.0

[a] The correlation coefficient between the two species is also given. For methods and detection limits see Table 2. HYL/PE, *Hylocomium*/*Pleurozium*.

Zn, Cu). It would appear that mosses regulate their concentration range to an optimum level with little deviation. Some other elements show a very high variation (wide box). Many of these are probably indicative of dust input to the moss (e.g. Fe, Al, B, Rb, Cr, Zr, Li, Y, Sc, Th, U).

When comparing the median of the moss samples collected in the eight catchments discussed here to that given for the Kola data set, a number of elements show a lower median in the Kola data. Aluminium (Al), B, Fe, Si, Th, Tl, and U are prominent examples. It appears that the majority of these elements indicate a higher proportion of minerogenic material (dust particles) in the samples collected for this study. Very few elements show a considerably higher concentration in the Kola data set: Ni, followed by Cu and Co are the most prominent examples. These three elements are the major elements emitted by the Kola smelters (see also maximum-ratio). The difference is even more dramatic when the maximum-ratio is studied. The maximum value for Ni from the Kola Project is more than 60 times higher than the maximum observed in this study. Much higher values for Cu, Bi and Ag were also reported from Kola. The higher maximum values for Be and Sr in the Kola data set have another source: mining and processing of alkaline rocks near Apatity.

Table 4
Analytical results for 'moss' (*Hylocomium splendens* and *Pleurozium schreberi* combined)[a]

Element	BARENTS-MOSS (*Hylocomium* + *Pleurozium*)						KOLA		MED Kola/ MED Barents	MAX Kola/ MAX Barents
	MIN	MAX	MED	MAD	Rel. var.%	MAX/MIN	MED	MAX		
Ag	0.012	0.064	0.025	0.006	24	5	0.033	0.824	1.3	12.9
Al	88	2380	355	184	52	27	193	4850	0.5	2.0
As	0.054	1.11	0.218	0.096	44	20	0.17	3.42	0.8	3.1
B	<0.5	47	3.31	1.67	51	186	1.76	21.6	0.5	0.5
Ba	7.33	83	22	6.95	32	11	19	175	0.9	2.1
Be	<0.01	0.122	0.016	0.011	69	24	<0.03	1.51	–	12.4
Bi	0.007	0.03	0.016	0.004	25	4	0.018	0.544	1.1	18.1
Ca	1690	10900	3050	820	27	6	2620	9320	0.9	0.9
Cd	0.032	0.536	0.132	0.031	23	17	0.089	1.23	0.7	2.3
Co	0.051	2.13	0.287	0.119	41	42	0.395	13.2	1.4	6.2
Cr	0.22	5.84	1.06	0.545	52	27	0.6	14.4	0.6	2.5
Cu	2.45	17.5	4.91	0.875	18	7	7.2	355	1.5	20.3
Fe	103	2390	443	218	49	23	212	5140	0.5	2.2
Hg	<0.04	0.765	0.055	0.010	18	38	0.05	0.155	0.9	0.2
K	2970	9260	5250	755	14	3	4220	8590	0.8	0.9
Li	<0.03	1.09	0.192	0.100	52	73	<0.05	NA	–	–
Mg	544	2020	973	217	22	4	1090	2380	1.1	1.2
Mn	33	1080	371	106	29	32	433	1170	1.2	1.1
Mo	0.035	0.307	0.098	0.039	39	9	0.08	1.08	0.8	3.5
Na	26	348	62	19.5	31	14	72	918	1.2	2.6
Ni	0.71	6.16	2.24	0.80	36	9	5.4	396	2.4	64.3
P	700	2830	1380	245	18	4	1270	3800	0.9	1.3
Pb	1.65	11.9	4.13	0.745	18	7	2.98	29.4	0.7	2.5
Rb	1.33	48	12.7	5.58	44	36	11.5	33.5	0.9	0.7
S	610	2020	965	197	20	3	863	2090	0.9	1.0
Sb	0.015	0.196	0.061	0.022	36	13	0.041	0.623	0.7	3.2
Sc	<0.03	0.510	0.090	0.050	56	34	<0.1	1.1	–	2.2
Se	<0.5	<0.5	<0.5	–	–	1	<0.8	1.23	–	4.9
Si	101	627	321	93	29	6	197	983	0.6	1.6
Sn	0.030	0.148	0.070	0.016	22	5	NA	NA	–	–
Sr	3.48	38	11.4	3.625	32	11	9.4	435	0.8	11.5
Th	<0.02	0.636	0.055	0.030	55	64	0.023	1.14	0.4	1.8
Tl	0.007	0.214	0.046	0.026	56	31	0.023	0.35	0.5	1.6
U	0.006	0.16	0.029	0.015	51	27	0.011	0.451	0.4	2.8
V	0.6	9.66	2.38	1.24	52	16	1.6	83.8	0.7	8.7
Y	0.036	1.82	0.174	0.111	64	51	<0.1	5.9	–	3.2
Zn	17	63	34	6.25	19	4	32.2	81.9	1.0	1.3
Zr	0.068	1.81	0.369	0.179	48	27	NA	NA	–	–

[a] Median absolute deviation (MAD), relative variation (rel. var.%; (MAD/MED)×100) and the ratio of the maximum to the minimum value reported are also given. For comparison median and maximum values from the Kola project were added (data from Reimann et al., 1998). The ratios of median (MED) and maximum (MAX) value Kola to median and maximum value as observed in moss from this project were added for easier comparison. To calculate the ratios for elements where one of the values was below detection a value of one-half of the detection limit was

3.1. Inter-species comparison

For inter-species comparison *xy*-diagrams have been drawn for 37 elements (all Se values were below detection — Table 3) and correlation coefficients were calculated. The 1:1 line in these diagrams allows the reader to grasp at one glance whether one of the two species shows generally higher or lower values than the other. Fig. 3 shows selected examples of these *xy*-diagrams. The majority of elements shows a high inter-species correlation (see Table 3 for correlation coefficients for all elements). Note that three data outliers were removed before correlation was calculated because inclusion of these samples resulted in unrealistically high (Hg) or too low (all other elements) correlation coefficients. Na and Rb show the highest correlation, Sn the lowest. There is no obvious connection between function (e.g. essential vs. toxic), solubility, and/or ion-size of elements and correlation. All coefficients are statistically significant at the 99% (Sn) or 99.9%-level (all others). It is thus possible to combine both species for a regional survey.

The need for a calibration between the two species is more difficult to answer. To be able to calibrate, one species has always to show higher (or lower) values than the other over the whole concentration range. The

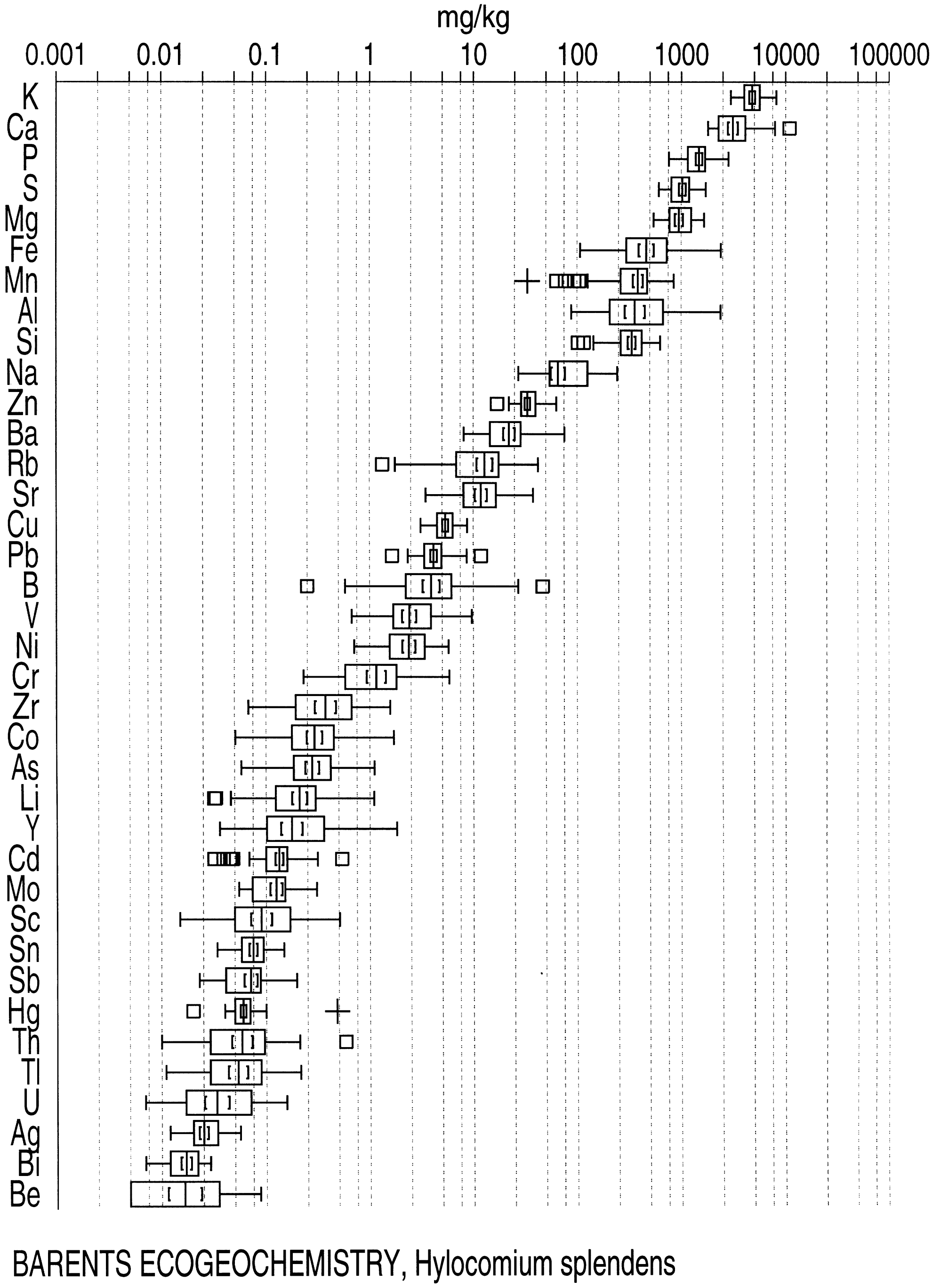

Fig. 2. Boxplot comparison of concentration range and total variability of 37 elements as determined in the moss species *Hylocomium splendens*. Note that for Se all values were below detection.

xy-diagrams show that there is a rather high variability between results even within one catchment (Fig. 3). This may be another indication that particulate input (dust) plays an important role for the observed chemical composition. To avoid such a problem the moss samples could of course be washed prior to analyses. It has been proven, however, that these moss species have ion-exchange properties (Gjengedal and Steinnes, 1990) and washing the moss might significantly distort the results. The median ratio between *Hylocomium* and *Pleurozium* (Table 3) suggests that *Hylocomium* generally returns slightly or clearly (As, B, Mo, Tl) higher results than *Pleurozium* (with the exception of K and Mg). This may be due to the much higher surface area of *Hylocomium* making this species more vulnerable to dust contamination. When studying the *xy*-diagrams in detail, however, it is obvious that for all elements there are always some samples that plot on the opposite side of

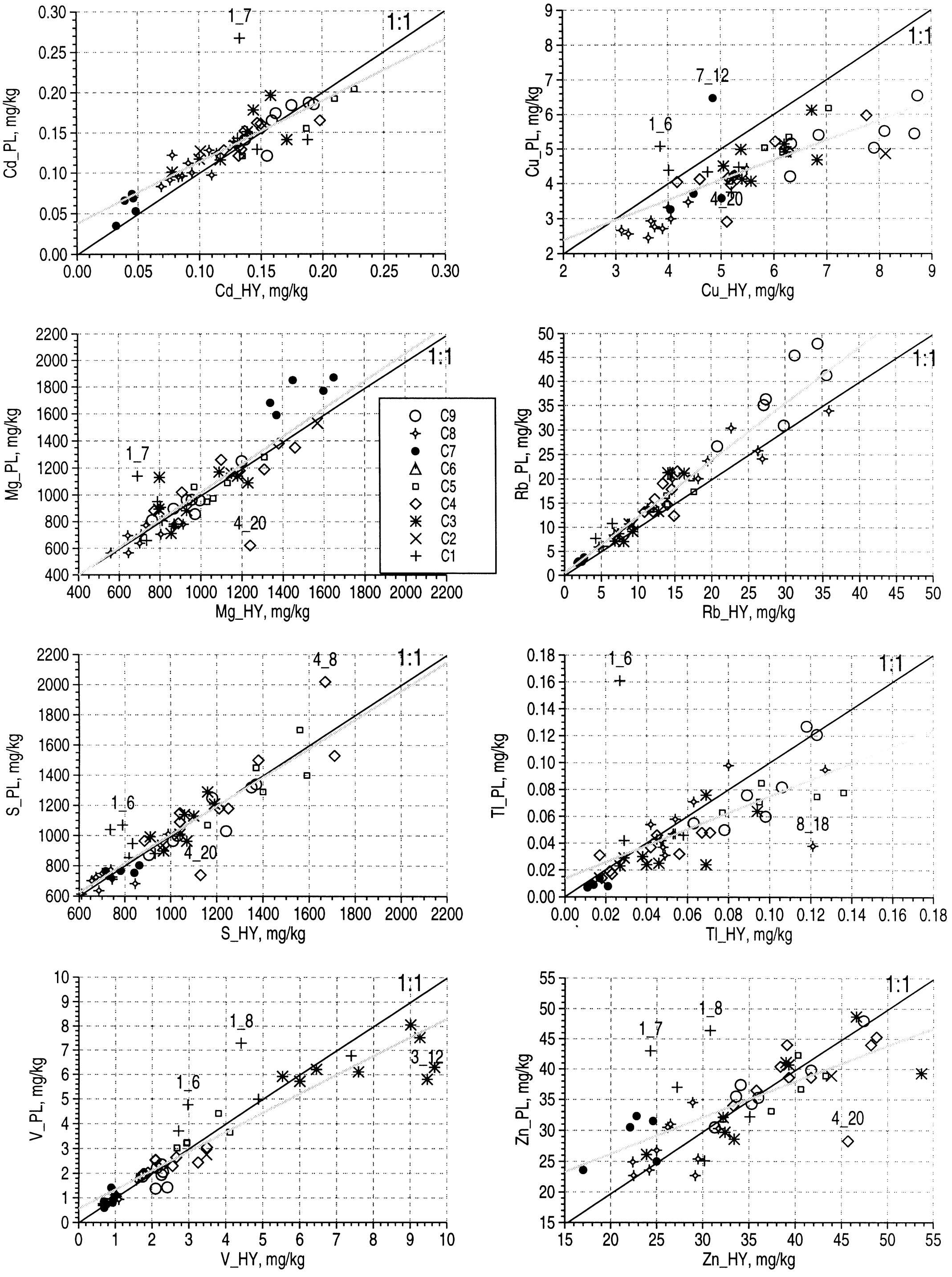

Fig. 3. Selected *xy*-plots showing results of the interspecies comparison *Hylocomium splendens* (HY) versus *Pleurozium schreberi* (PL). The grey line is the regression line (see Table 2 for the correlation coefficient). Note that no moss samples could be collected in C6.

the 1:1 line than the majority of samples (Fig. 3). This suggests that local site conditions can considerably alter the analytical results received (see also Zechmeister, 1995). A general calibration of the results would thus give an increased overall error. Calibration between the species does not thus seem advisable. This is in agreement with the results presented by Thöni et al. (1996) and Halleraker et al. (1998). Regional maps for elements like Mo, Tl, As, B, Sb, U and Cu, for which the inter-species differences judged by the median ratio given in Table 2 is > 20%, will thus be less reliable than all other maps. The relative importance of this problem will depend on the total variation observed for these elements in any one region.

3.2. Factor analysis

Fig. 4 shows the results of robust factor analysis in a simple plot of factor loadings against cumulative explained variability. The four factors explain 73% of the total variability.

Factor 1, explaining 40% of the total variability has a clear 'geogenic dust' signature (Al, Cr, Fe, Li, Sc, Si, Y, Zr all with high loading). These typically dust-related elements are accompanied by many other elements like Co, Mo, Sb, Ni, V, Ag, and Bi. This indicates that the concentration of these elements in moss is seriously influenced by the amount of dust particles in each sample. It appears that deposition of dust is one of the most important factors influencing moss chemistry. In a biplot (Fig. 5) one can discern that this influence is very strong in catchments 2, 1 and 5. The interesting question whether this is just local soil dust or fine, long-range transported atmospheric dust remains open.

Factor 2 combines many highly volatile elements that will be part of atmospheric aerosols, e.g. Ag, Bi, Cd, Hg, Pb, S, Sb, Sn. These are the elements that the moss may take up directly from the local air or which are associated with very fine particulates. The biplot (Fig. 5) suggests that this is an important pathway in catchments 1, 9 and 3.

The loading of factor 3 is dominated by many important plant nutrients, e.g. Ca, K, P, S, Zn, accompanied by some unexpected elements like Be, Cd, Hg and Pb. The biplot reveals a differentiation between the northern (C1, C7, C3, C8) and southern (C5, C9, C4) catchments. A probable explanation is that climate and vegetation zones influence concentrations of these elements in moss (Reimann et al., 2000).

Factor 4 shows a positive loading for several important elements related to marine aerosols (Mg, Na, Sr, B) while Rb, Tl, and Cd show a negative loading. This indicates that the input of major ions from an important source like the sea can prohibit the uptake of other elements. The biplot (Fig. 5) shows that all samples from C7, Berlevåg, which is immediately adjacent to the Barents Sea (but also the northernmost catchment), are strongly influenced. C4 plots in the same direction as C7 due to emissions of Na from the cardboard factory.

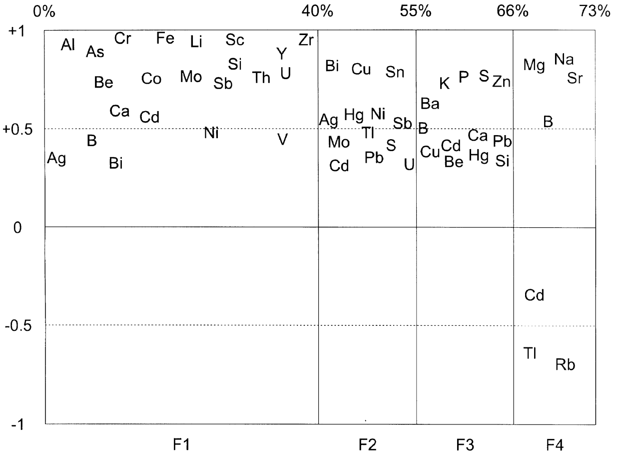

Fig. 4. Graphical display of the results of a robust factor analysis (FA) performed with all moss samples — the x-axis gives the cumulative explained variance, the y-axis represents the factor loadings for each element (only elements with loadings >0.4 are shown).

In general the biplots suggest that all four element-sources play an important role in determining the chemical composition of moss in space but that every catchment has its own special signature.

3.3. Local versus regional variability and element sources

Boxplots have been drawn for all elements, comparing the results between the eight catchments. Fig. 6 shows selected examples. In addition, *xy*-diagrams were drawn for all possible element combinations and were also used for the interpretation of the features seen in the boxplots. Fig. 7 shows selected examples. Although most elements display significant differences between the catchments (see median values and brackets in boxplots) there is still a very high local variation for many elements in many catchments. There are elements that show a comparatively high local variation in all catchments (e.g. Ag, Ba, Bi, Cu, Hg, Mg, Mn, Zn). Other elements display a high variation only in some few catchments (e.g. As, Ca, Cd, Cr, Fe, Li, Mo, Na, Rb, Sb, U, V, Y, Zr). This indicates that in many cases a single moss sample will not be very representative for a large area. Local, site-specific conditions (e.g. dust level, relative distance to important emission sources, composition of the local vegetation) can have a major influence on the observed element concentration in the moss samples.

Near catchment 1 (C1), Vorkuta, coal mining is the most important industrial activity. Due to the arctic climate and permafrost conditions, vegetation is rather sparse. Moss collected in this catchment shows comparatively high[1] concentrations (see Fig. 6 for examples) of: **Ag**, (Al), **As**, (Ba), Be, **Bi**, (Ca), ((Cd)), (Co), Cr, Fe, **Li**, **Mo**, Ni, **Sb**, Sc, (Si), (Sr), (Th), U, (V), (Y) and **Zr**. Almost all of these elements occur in high concentrations in the local soil parent material (compare Table 1). The combination of elements like Al, Fe, Li, Sc, Si, Th, U, Y and Zr argues for local dust being the most important supplier of elements to moss in this catchment.

An *xy*-plot of Al/Fe shows the very high correlation of these two 'geogenic dust' related elements in all catchments (Fig. 7). In contrast, many of the important

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[1] No brackets, highest median of all catchments; (), second highest; (()), third highest; bold, remarkable enrichment in comparison to all other catchments; italics, remarkable depletion.

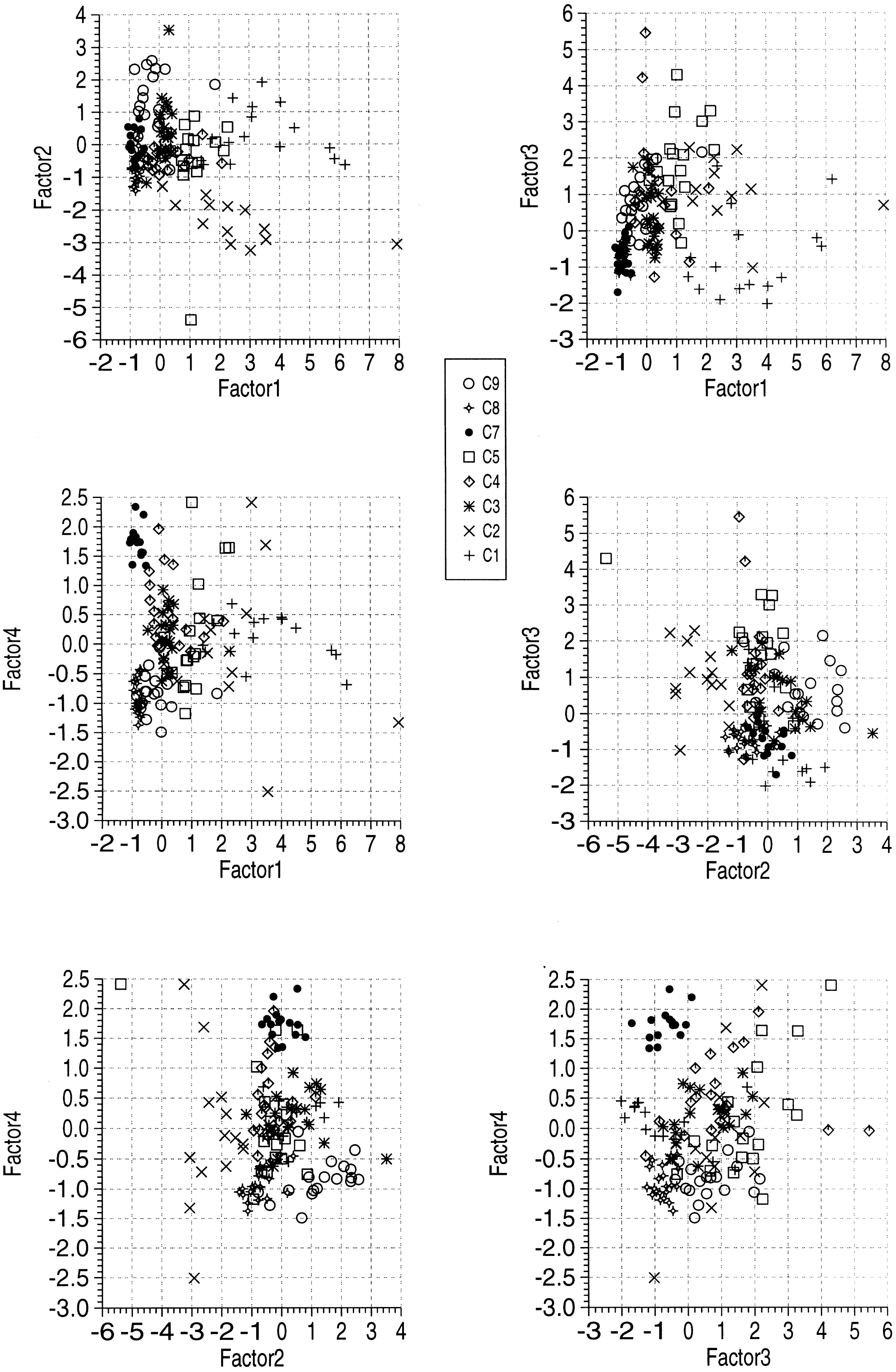

Fig. 5. Biplots showing the results of the robust factor analysis for the samples from each catchment.

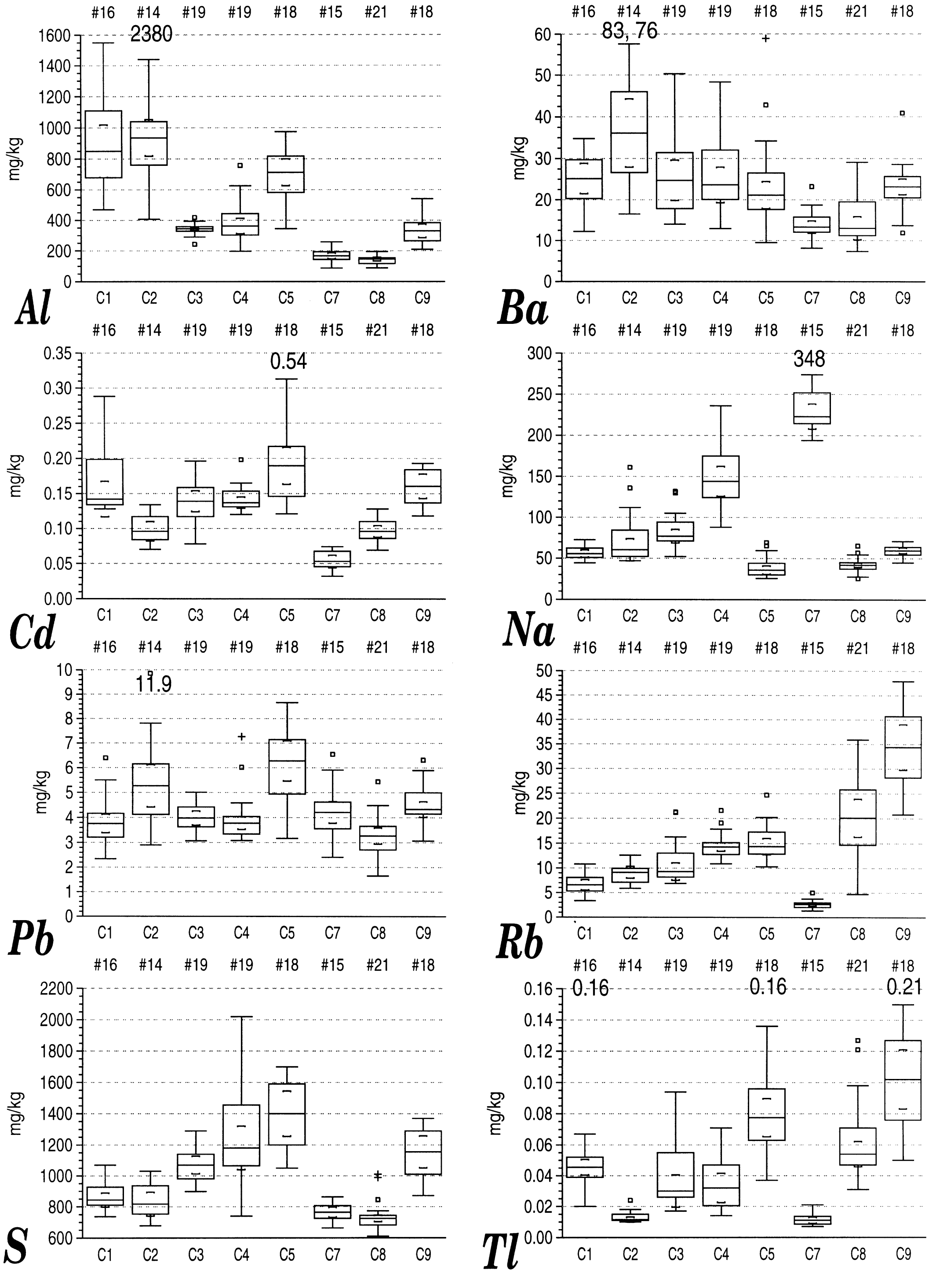

Fig. 6. Selected boxplots allowing to directly graphically compare concentration range and variation of the moss samples from each catchment.

plant nutrients show especially low values here: ***K***, Mg, P, S and Zn, accompanied by B, Cu, Rb, Sn and Tl. If coal burning or coal dust were an important ingredient of the atmospheric input in C1 one would expect clearly elevated concentrations of elements like B, Sn and Tl which all occur in high concentrations in coal (Reimann and Caritat, 1998). An *xy*-plot of Mg/Sr (Fig. 7) suggests that there must be an unusual Sr-source in C1 (or that Mg is much lower than should be expected). When comparing all catchments, Rb shows a very clear N-S trend — the more northern the location of the catchment the lower Rb values are observed in the moss (Fig. 6, Rb). Some other elements show a similar tendency (e.g. Cd, Mn, S, Tl, V) but for these the trend is

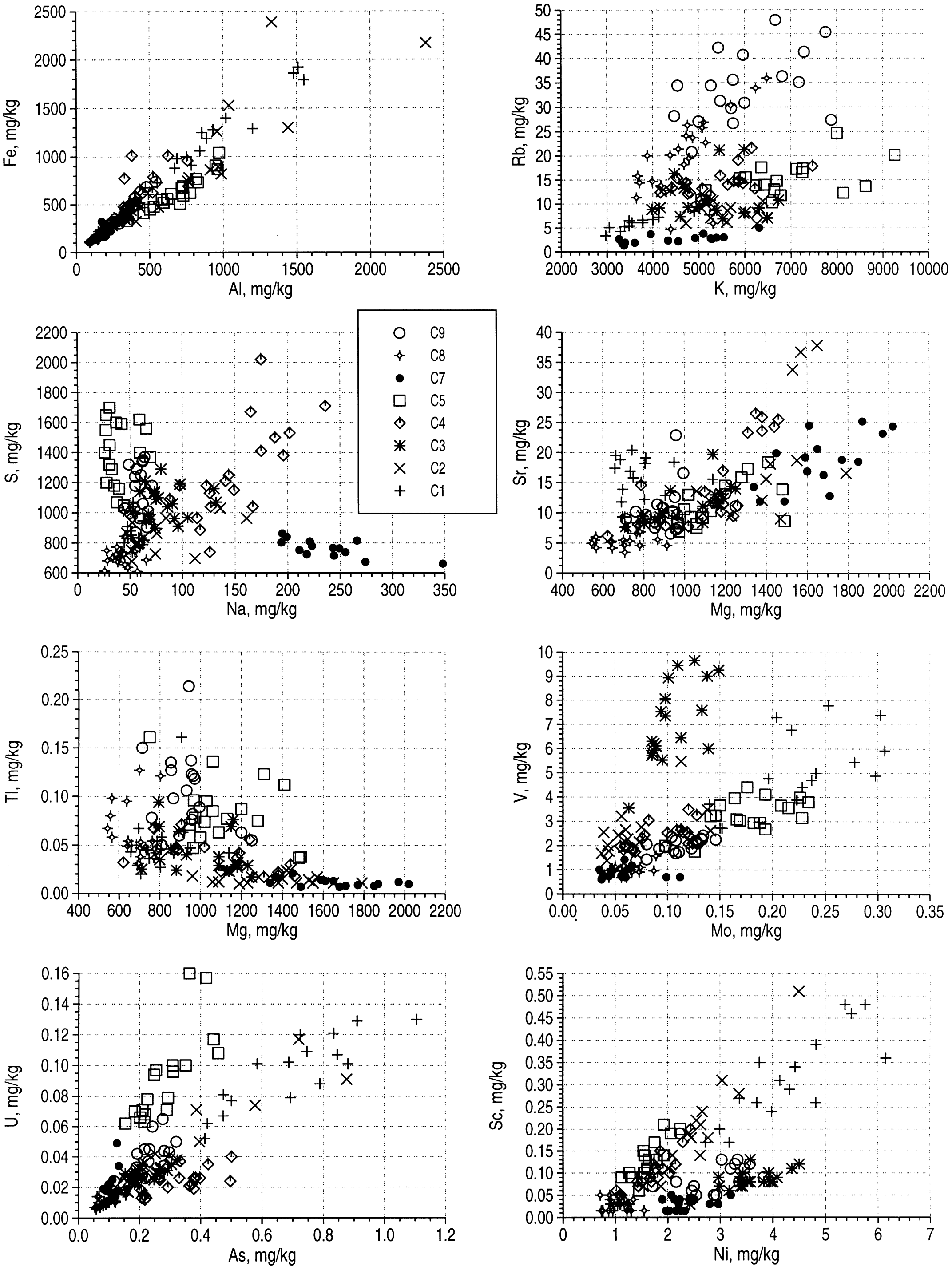

Fig. 7. Selected *xy*-plots showing inter-element relations in the total moss data set.

often disturbed by some local element source. The reason that Rb shows this trend so very clearly may be that it is one of the few elements without any major emissions due to human activities.

Catchment 2 (C2), Narjan Mar, is located adjacent to the local airport. Otherwise this is a very remote catchment near the mouth of the Pechora River. One might expect some influence from marine aerosols on the element concentration observed in moss here. Large swamps and sand dunes dominate the landscape. The elements Al, Ba, (Be), Co, (Cr), (Fe), (Li), (Mg), Mn, (P), (Pb), (Sc), Si, ((Sr)), Th, ((U)), Y, Zn, ((Zr)) all appear in comparatively high concentration here. Cd, Rb, Sb, Sn and Tl show very low concentration levels. Again, local dust appears to be the major source of high concentrations of some elements observed in moss from

C2. Input from marine aerosols may be the main reason for the high values of Mg and Sr observed here (compare also Fig. 7 — Mg/Sr). Mg is very low in the C-horizon of C2 (Table 1). None of the other elements indicative of marine input (especially Na, S) follows suit. It is also interesting to note that Tl shows by far the lowest values in the two northernmost catchments (C2 and C7 — see discussion below).

Catchment 3 (C3), Archangelsk, is located at the outskirts of one of the largest towns in the survey area. There is a lot of industrial activity here, including pulp mills, military industries, and even a rocket launching pad. ((Ag)), ((Cu)), (Mn), ((Na)), (Ni), ((Sn)), V, and ((Zn)) show high concentrations here — probably indicative of the power plants burning heavy oil in the nearby town (see Fig. 7 — Mo/V). The *xy*-diagrams suggest clearly that Ni is added from other than geogenic sources (Fig. 7, Sc/Ni). The elements Al, Li, Sc, Si, and Zr not only show low levels here but also a very low variability — indicative of dust input playing only a minor role for the chemical composition of moss in this catchment.

Catchment 4 (C4), Korjazhma, is located near one of Russia's largest cardboard factories. (As), (B), ((Ca)), ((Cr)), Hg, ((Mg)), (**Na**), (S), and (Zn) are comparatively high here. In addition Na, S, and Sr show an unusually high variability in C4. In contrast to the 'sea' catchment C7 (see below), the Na/S-diagram (Fig. 7) demonstrates that in C4 both Na and S are high, pointing to different sources resulting in different uptake mechanisms. Ca, Na, Mg and S would normally be indicative of the input of marine aerosols — C4 is, however, one of the most 'inland' catchments of our study and a marine influence can be excluded. Hg shows the highest median in C4 but in general contrast between catchments is low. Moss is probably not an ideal sample medium for monitoring input of Hg.

Catchment 5 (C5), Kingisepp, is the southernmost catchment near the border between Russia and Estonia. It is relatively close to St. Petersburg and a major phosphate producer is located near the outlet of the catchment. Large parts of this catchment are used for agriculture. Many elements show comparatively high concentrations in the moss collected here: ((Al)), B, ((Be)), ((Bi)), **Ca**, Cd, (Cu), ((Fe)), ((Hg)), **K**, (**Mo**), P, Pb, **S**, (Sb), ((Sc)), ((Si)), (Sn), ((Th)), (Tl), (**U**), ((V)), ((Y)), and (Zr). All of these, with the exception of Cu, Fe, Hg, Mo, and V, show also an exceptionally high variability in C5. Here enhanced concentrations are due to several different sources/processes. Input of local soil dust (Al, Fe, Mo, Pb, Sc, Si, Th, U, V, Y, and Zr are highly correlated in C5) is one source. A second group of elements showing high correlation is Ca, Cd, Cu, Mg, (P), S, Sr and Zn. These elements probably originate from the local phosphate producer. K does not correlate with any of the other elements and shows higher values in samples from this catchment than in all other catchments. With C9, C4 and C8 also showing rather high K concentrations, while the northernmost catchments C1 and C7 showing very low levels, one can speculate whether these high K-levels in the moss samples from C5 are related to climate/vegetation zones. Mn and Na concentrations in moss are unusually low in C5. In Fig. 7 several of the *xy*-diagrams demonstrate the special processes determining moss composition in C5.

No moss could be collected in catchment 6 (C6), Monchegorsk. Close (5 km) to the smelter most vegetation is dead and mosses and lichen are completely absent. Data presented in Reimann et al. (1998) show that moss will reach maximum concentrations for many of the elements in the vicinity of Monchegorsk. Table 4 compares the maximum values observed in moss in this study, excluding Monchegorsk, and the maximum values obtained by Reimann et al., who collected moss at a somewhat greater distance (ca. 20 km) from Monchegorsk. Results show that many elements can reach higher maximum concentrations by one to two orders of magnitude in the vicinity of a major pollution source. These values decline rapidly with distance from source (Reimann et al., 1997) — probably an indication of particulate input governing the chemical signal observed in the moss samples.

The Norwegian catchment 7 (C7), Berlevåg, is special in so far as it is immediately adjacent to the coast of the Barents Sea. The importance of the input of marine aerosols for moss chemistry can be seen in the very high concentrations of Mg, Na and Sr. Other sea-salt-related elements like B, Ca, Li and S are not enriched in the moss samples in C7. This proves that moss is quite selective as to which elements are taken up and concentrated. At the same time a large suite of elements shows unusually low concentrations, and, more importantly very low variability in the samples from this catchment. Ag, Al, As, Ba, Be, Bi, Ca, ***Cd***, Cr, Fe, K, Li, Mn, Mo, ***Rb***, S, Sb, Sc, Si, Sn, Th, ***Tl***, U, ***V***, Y, Zn, Zr all belong into this group. Rb and Tl have been mentioned above (C1, C2) as showing low values in the 'northern' catchments. Cd and V join these two elements in C7 with exceptionally low values.

There exist a number of possible explanations for this observation:

1. A large distance from urbanisation with its many pollution sources and a very low traffic and population density in the surroundings of this catchment are reflected in very low element concentrations and a low local variability. It should, however, be kept in mind that Nikel and Zapoljarnij, two of the world's largest point source emitters of sulphur dioxide and metals are located less than 250 km to the south-east of Berlevåg.
2. Low local dust levels due to the relatively 'wet' climate result in low concentrations of most

elements (low values of Al and Zr as an indication). Low dust levels are a bit surprising, however, because there is a very sparse vegetation cover here due to the northern location. No trees and hardly any bushes grow in C7. The scarceness of vegetation might be expected to actually increase dust levels. Many of the open areas here, however, are bedrock or boulder fields and not soil.

3. Heavy rainfall hitting the moss directly due to missing higher vegetation will wash off any dust particles — the moss samples are thus 'cleaner' than in other catchments.
4. The moss chemistry is much less influenced by higher vegetation than in all other catchments (Mn, K, Zn!) — there is not much chance that moss takes up elements from litter and soil in this catchment, the moss chemistry is hardly influenced by throughfall from higher vegetation here.
5. The high input of Na and Mg due to marine aerosols prevents the uptake of the other elements or may even replace other elements from ion-exchange positions.

When studying the geochemical characteristics of the soil parent material in the different catchments C7 actually shows high concentrations of Al, As, Bi, Cd, Co, Cu, Fe, Li, Mg, Mn, Mo, P, Pb, Sb, Th, Y and Zn in comparison to the other catchments. Of these elements Al, As, Bi, Cd, Fe, Li, Mn, Mo, Sb, Th, Y and Zn were all named above as having low concentration and variation in moss from C7. This observation is an indication that local dust has no influence on the element concentrations in moss from this catchment. It would thus appear that moss chemistry in C7 is governed by the input of marine aerosols, with Mg, Na and Sr preventing other ions from being taken up by the moss. The fact that other 'marine' elements like B, Ca and S do not show very high concentrations in this coastal catchment demonstrates that moss is selective in which elements are taken up if there is an 'oversupply' of more suitable elements. The very low concentrations of elements like Cd, Rb, Tl and V suggest that these elements are not only not taken up but might even be removed from the moss by sea-salt input. Depending on the total available element spectrum at any place moss can thus alter the atmospheric input signal drastically.

Catchment 8 (C8) near Kuhmo in Finland is located in a densely forested part of Finland, distant from any important pollution source. The only industrial activity in this area is forestry. Of all the elements analysed here only Rb and Mn show high values. Litter is in rich supply and is the most likely source of these two elements (and K) in the moss. Many elements show exceptionally low concentrations and also very low variation in C8: ***Al, As, B***, Ba, Be, ***Bi, Ca***, (Cd), ***Co***, Cr, ***Cu***, Fe, Hg, Li, ***Mg***, Mo, Na, ***Ni***, P, Pb, S, Sb, Sc, ***Si***, ***Sr***, Th, ***U***, V, Y, Zn, and Zr. There are no large open areas that could result in enhanced dust levels due to wind erosion in this area. C8 also does not show any geochemical peculiarities in the soil parent material (Table 1). Experience from the Kola Project (Reimann et al., 1998) shows that a low variation in element concentrations is one of the best indicators of a minimal influence from pollution.

Catchment 9 (C9), Urjala, is located in south-west Finland in a predominantly agricultural area. The soil parent material shows unusually high values of Ag, As, Cu, K, Mo, S, and Th here (Table 1). Moss shows high concentrations of (Ag), (**Bi**), (Cd), Cu, ((Hg)), (K), ((Ni)), ((Pb)), **Rb**, ((S)), ((Sb)), Sn and **Tl** in C9. Some, but not all can be explained by the special geogenic signature of the soils in this area. K, Mn and Rb may again be high due to the southern location of this catchment (see C5). Local dust levels appear to be low (little variation in Al, Zr) even with large agricultural fields spread all over the catchment. One explanation may be that there are a lot of small forests interspersed with these fields. Elements like Ca, Li, Mg and Na, which may be able to replace other ions from the moss (see C7) are all very low. The reason for the comparatively high concentrations of Bi, Pb, Sn and Tl are probably the relatively high population density in this area and diffuse emissions from traffic and power plants affecting a rather large area.

4. Conclusions

The two moss species *H. splendens* and *P. schreberi* can be combined for regional bio-geochemical mapping. Although interspecies variability will increase the total survey variability for a number of elements (especially Mo, Tl, As, B, Sb, U and Cu), results obtained for all single pairs of moss show that it appears inadvisable to attempt interspecies calibration.

Local variability is high when compared to total variability for the majority of elements and for regional mapping it appears desirable to collect as large composite samples over as large an area as practically possible. For reliable large-scale low-density (e.g. one site per 300–1000 km^2) regional bio-geochemical mapping using moss it might even appear advisable to collect several sub-samples spread over a whole catchment of 30–100 km^2 to give one representative composite sample. The observed high between-site variability places some doubt on the suitability of terrestrial moss for regional monitoring purposes. It may be possible to estimate time trends on a European or country-based scale but probably not for smaller areas without taking very large composite samples.

In many catchments moss chemistry appears largely dominated by input of local dust or airborne particular matter. It thus primarily reflects local site conditions and not major differences in atmospheric chemistry (which will, of course, also be influenced by local site conditions). It appears doubtful that moss is a sensitive enough indicator to reliably detect subtle differences in atmospheric chemistry due to long-range transport of air pollutants. By now it is well established that major local sources give a major local signal, that declines rapidly with distance from source (see also Reimann et al., 1997, 1998, and maps in, for example, Rühling and Steinnes, 1998, or Siewers and Herpin, 1998). All regional maps show that this signal usually disappears in the high local 'noise' levels at a distance of 100–200 km from source. Using other sample media, e.g. rain, snow or ice cores, transport of many metals over much larger distances has been demonstrated (e.g. Boutron et al., 1994, 1995; Hong et al., 1994; Candelone et al., 1995; Ferrari et al., 2000). It would thus appear that moss best reflects the travel distance of (fine) particulates emitted by large industrial sources.

A very low regional variation of the major nutrients in the moss suggests that uptake of these elements is regulated by the plant. Input of marine aerosols to moss near coastal sites has a major influence on the observed concentration of many other elements in the moss. Moss preferentially takes up Na and Mg (maybe Sr), and does not then accept other elements, among them S. Element input from litter to moss is reflected in unusually high concentrations of elements like Mn and Rb in the southernmost catchments. In spite of all these difficulties there is no doubt that regional bio-geochemical mapping using the moss technique is able to provide valuable insights to environmental processes, important element sources and relative ecosystem health for large areas though without giving a true reflection of atmospheric chemistry.

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