Temporal trends in the concentration of arsenic, chromium, copper, iron, nickel, vanadium and zinc in mosses across Europe between 1990 and 2000

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Abstract

The European heavy metals in mosses biomonitoring network provides data on the concentration of ten heavy metals in naturally growing mosses and is currently coordinated by the UNECE ICP Vegetation (United Nations Economic Commission for Europe International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops). The technique of moss analysis provides a surrogate, time-integrated measure of metal deposition from the atmosphere to terrestrial systems. It is easier and cheaper, less prone to contamination and allows a much higher sampling density than conventional precipitation analysis. Moss surveys have been repeated at five-yearly intervals and in this paper we report on the temporal trends in the concentration of arsenic, chromium, copper, iron, nickel, vanadium and zinc between 1990 and 2000. Maps were produced of the metal concentration in mosses for 1990, 1995 and 2000, showing the mean concentration per metal per 50 km x 50 km EMEP grid square. Metal- and country-specific temporal trends were observed. Although the metal concentration in mosses generally decreased with time for all metals, only the decreases for arsenic, copper, vanadium and zinc were statistically significant. The observed temporal trends were compared with emission trends for Europe reported by EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutant in Europe).

Keywords: biomonitoring, EMEP maps, heavy metal, metal deposition, moss

1. Introduction

The heavy metals in mosses biomonitoring network was originally established as a Swedish initiative (Rühling and Skärby 1979; Tyler, 1970). The idea of using mosses to measure atmospheric heavy metal deposition is based on the fact that ectohydric mosses obtain most trace elements and nutrients directly from precipitation and dry deposition; there is little uptake of metals from the substrate. The technique of moss analysis provides a surrogate, time-integrated measure of metal deposition from the atmosphere to terrestrial systems. It is easier and cheaper than conventional precipitation analysis as it avoids the need for deploying large numbers of precipitation collectors with an associated long-term programme of routine sample collection and analysis. Therefore, a much higher sampling density can be achieved than with conventional precipitation analysis. The higher trace element concentrations in mosses compared to rain water makes analysis more straightforward and less prone to contamination. In addition, heavy metal measurement data from precipitation analysis can be very uncertain if the detection limits of the applied analytical technique are high (Ilyin et al., 2006). Despite improvement of the analytical techniques the latter remains a problem due to the general decline in anthropogenic emissions and subsequent deposition of heavy metals in recent decades. Although the heavy metal concentration in mosses provides no direct quantitative measurement of deposition, this information can be derived by using regression approaches relating the results from moss surveys to precipitation monitoring data (e.g. Berg and Steinnes, 1997; Berg et al., 2003).

The moss survey has been repeated at five-yearly intervals and the number of participating European countries has expanded greatly since 1990 (Buse et al., 2003; Rühling, 1994; Rühling and Steinnes, 1998). Currently, the 2005/2006 moss survey is being conducted in 32 countries, analysing moss samples from over 7 000 sites across Europe. For the first time the majority of countries (18) will also determine the nitrogen concentration in mosses (ca. 3 200 sites), as a pilot study for selected Scandinavian countries has shown that there was a good linear relationship between the total nitrogen concentration in mosses and atmospheric nitrogen deposition rates (Harmens et al., 2005). During 2001, responsibility for the coordination of the moss survey was handed over from the Nordic Working Group on Monitoring and Data, Nordic Council of Ministers, to the UNECE ICP Vegetation Coordination Centre at the Centre for Ecology and Hydrology (CEH) Bangor, UK.

The UNECE ICP Vegetation was established in the late 1980s to consider the science for quantifying damage to plants by air pollutants. It is one of seven ICPs and Task Forces that report to the Working Group on Effects of the Long-Range Transboundary Air Pollution (LRTAP) Convention on the effects of atmospheric pollutants on different components of the environment (e.g. forests, fresh waters, buildings) and human health (Working Group on Effects, 2004). The objectives of the ICP Vegetation (Harmens et al., 2006) are designed to meet the requirements of the LRTAP Convention, particularly at present the need to provide information for the review of the 1999 Gothenburg Protocol to abate acidification, eutrophication and ground-level ozone and the 1998 Aarhus Protocol on heavy metals. The latter was the first Protocol for the control of emissions of heavy metals; cadmium, lead and mercury emissions were targeted as they are the most toxic.

The European moss survey provides data on concentrations of ten heavy metals (As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, V, Zn) in naturally growing mosses (Buse et al., 2003; Rühling, 1994; Rühling and Steinnes, 1998). The main purpose of the survey is (a) to provide, in the form of maps, spatial information on the distribution of heavy metal concentrations in mosses in Europe, (b) identify main polluted areas, and (c) develop the understanding of long-range transboundary pollution. In general, there was a clear east/west decrease in the concentration of heavy metals in mosses, related in particular to industry. Former industrial or historic sites of heavy metal pollution (e.g. mines) accounted for the location of some high concentrations of heavy metals in mosses in areas without contemporary industries. Long-range transboundary transport appears to account for elevated concentrations of heavy metals in areas without emission sources (e.g. in Scandinavia). Many contributors to the survey have reported their national data in greater detail elsewhere.

In this paper, we report on the temporal trends (1990 – 2000) of arsenic, chromium, copper, iron, nickel, vanadium and zinc concentrations in mosses and these trends were compared with trends in anthropogenic emission data reported to EMEP (Ilyin et al., 2006; Task Force on Heavy metals, 2006). In a previous paper we reported on the temporal trends of cadmium, lead and mercury concentrations in mosses and the comparison with modelled deposition data reported by EMEP (Harmens et al., in press).

2. Materials and methods

Moss samples were collected across Europe in 1990/1991 (Rühling, 1994), 1995/1996 (Rühling and Steinnes, 1998) and 2000/2001 (Buse et al., 2003; Harmens et al., 2004). Throughout the paper we refer to the survey years as 1990, 1995 and 2000 respectively. The carpet-forming mosses *Pleurozium schreberi* and *Hylocomium splendens* were the preferred species for analysis. However, since the mosses were collected in a range of habitats from the sub-arctic climate of northern Sweden to the hot and dry climate of parts of southern Italy, it was inevitable that a wide range of moss species was sampled (Buse et al., 2003; Harmens et al., 2004). The moss sampling procedure was according to the guidelines described in Rühling (1994) and Rühling and Steinnes (1998) and was described in more detail in the protocol for the 2000 survey (ICP Vegetation, 2001). Only the last three years' growth of moss material was used for the analyses. The concentration of arsenic, chromium, copper, iron, nickel, vanadium and zinc were determined by a range of analytical techniques, under the broad headings of atomic absorption spectrometry, inductively coupled plasma spectrometry (both

ICP optical emission spectrometry and ICP mass spectrometry), atomic fluorescence spectrometry and neutron activation analysis. All metal concentrations were expressed as μg g^{-1} dry weight at 40 °C. For further details on the methods and quality control procedures we refer to the reports of the individual surveys (Buse et al., 2003; Rühling, 1994; Rühling and Steinnes, 1998).

For each survey year EMEP maps were produced according to the method described by Buse et al. (2003); they show the mean concentration of each metal within individual EMEP grid squares (50 km x 50 km). Please note that the designations employed and the presentation of material in this paper do not imply the expression of any opinion whatsoever on the part of the United Nations concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries.

Statistical analysis of the temporal trends (1990 – 2000) across Europe was performed by calculating the geometric mean values per metal per survey year for each country. Subsequently, a general linear model ANOVA (Minitab version 14) was applied to each metal using only the geometric mean values for the countries which had determined the heavy metal concentration in mosses in all survey years for that metal. The geometric mean values were analysed with country as a factor, year as covariate and the number of samples as weights. Weighting was applied to take into account the accuracy of the calculated geometric means (i.e. the density of sampling varied between countries) and to give more weight to larger countries and less to smaller ones.

3. Results

The concentration of heavy metals in mosses showed country- and metal-specific temporal trends between 1990 and 2000 (Tables 1-5, Figures 1-7).

Arsenic (As)

Only five countries determined the As concentration in mosses in all three survey years. For these countries, the As concentration declined significantly (P = 0.026) between 1990 and 2000 with the biggest decline between 1990 and 1995 (Figure 1, Tables 1, 4, 5). For these five countries, the highest As concentrations were found in mosses in the Czech Republic in all survey years. However, 17 countries determined the As concentration in both 1995 and 2000 and for those countries the As concentration did not change significantly (P = 0.30) between 1995 and 2000 (Tables 4, 5). In the central European countries the mean As concentration in mosses generally decreased with time (Figure 1).

Chromium (Cr)

As for As, the mean Cr concentrations in mosses generally decreased with time in central European countries (Figure 2). However, the average median Cr concentration across Europe declined by only 8% between 1990 and 2000 (Table 4) and no significant trend was found in the average geometric mean values for countries that analysed Cr in all survey years (Table 5). For some countries (Iceland, Italy, Lithuania, Slovakia, Spain and UK) the median Cr concentration in mosses increased between 1990 and 2000 (Table 1). The highest median Cr concentrations were found in Romania and Slovakia.

Copper (Cu)

Although the year of survey significantly affected the average geometric mean Cu concentration in mosses (P = 0.026; Table 5), this was primarily due to a decline between 1990 and 1995. The decline in the average median value across Europe was only 16% between 1990 and 2000 (Table 4). For some countries the decline was highest between 1990 and 1995, for others between 1995 and 2000, whereas some countries showed no change or a small, steady decline between 1990 and 2000 (Figure 3; Table 1). In quite a number of countries (Austria, Faroe Islands, France, Hungary, Italy, Romania, Switzerland and Ukraine) the median Cu concentration in mosses increased between 1990 (or 1995) and 2000 (Table 1). The highest median Cu concentrations were found in Bulgaria, The Netherlands, Romania and Slovakia.

Iron (Fe)

The average geometric mean Fe concentration in mosses decreased between 1990 and 1995, but increased again between 1995 and 2000, resulting in no significant change with time (P = 0.099; Table 5). The decrease between 1990 and 1995 was particularly observed in most of central and eastern Europe (Figure 4; Table 3). The high Fe concentrations in mosses in Iceland are due to drift of volcanic ash and windblown soil dust. Extremely high Fe concentrations were also observed in Romania (due to local industry) and Spain (possibly due to soil contamination by windblown dust) in 1990; the 1990 data for Spain are based on sampling from only 8 sites. Overall, the decrease in the median Fe concentration in mosses was 44% between 1990 and 2000 (Table 4).

Nickel (Ni)

Despite a steady decline in the average geometric mean Ni concentration in mosses across Europe between 1990 and 2000, the decline was not significant (P = 0.074; Table 5). The overall decline in the median value was 30% (Table 4). For some countries the decline in the median value was highest between 1990 and 1995, for others between 1995 and 2000 (Table 2). In quite a number of countries (Bulgaria, Faroe Islands, France, Hungary, Iceland, Italy, Slovakia and Spain) the median value increased between 1990 (or 1995) and 2000. Unusually high median Ni concentrations were found in The Netherlands (due to the presence of industry and possibly analytical bias) and Portugal (due to the presence of industry and a dense motorway network) in 1995. Only Germany and the area of St. Petersburg in the Russian Federation showed a steady decline in the Ni concentration in mosses with time (Figure 5; Table 2).

Vanadium (V)

The average geometric mean V concentration in mosses declined steadily and significantly between 1990 and 2000 ($P = 0.000$; Table 5), with an overall decline in the median value of 32% (Table 4). Despite the steady decline with time across Europe, country-specific changes in the median values between 1990 and 1995 or 1995 and 2000 were observed, with decreases being found in one time period but not the other (Figure 6; Table 2). In Poland (France, Italy and Slovakia) the median V concentration in mosses actually increased between 1990 (1995 respectively) and 2000, with no change being observed in Iceland and Lithuania.

Zinc (Zn)

The average geometric mean Zn concentration in mosses declined significantly with time and the highest decline occurred between 1990 and 1995 ($P = 0.021$; Table 5). The overall decline in the median value was 19% between 1990 and 2000 (Table 4). Nevertheless, country-specific temporal trends were observed between the survey years, with even an increase in the median Zn concentration being observed for Bulgaria, France, Hungary, Iceland, Italy and Romania between 1990 (or 1995) and 2000 (Table 2). The maps show a clear decline in the Zn concentration in eastern Germany and an increase in France between 1995 and 2000 (Figure 7).

4. Discussion

Whereas the arsenic, copper, vanadium and zinc concentration in mosses decreased significantly ($P \leq 0.05$) across Europe between 1990 and 2000, the decreases for chromium ($P = 0.180$), iron ($P = 0.099$) and nickel ($P = 0.074$) were not significant. The observed decrease for arsenic was based on data for five countries only; no change in the arsenic concentration in mosses was observed between 1995 and 2000 when comparing the data for 17 countries. Country-specific trends were observed for all the metals with decreases, no changes or even increases being observed between 1990 and 1995 or 2000 and between 1995 and 2000.

The temporal trends for cadmium, lead and mercury were reported previously (Harmens et al., in press): the cadmium and lead concentrations in mosses decreased significantly between 1990 and 2000, but no significant change was observed for the mercury concentration in mosses between 1995 and 2000. The temporal trends for cadmium, lead and mercury were similar to the temporal trends reported by EMEP regarding the modelled total deposition of these metals in Europe (Ilyin et al., 2005). Currently, no data are available for the modelled total deposition of other heavy metals across Europe. However, decreases in the anthropogenic emission of metals according to official data combined with experts estimates were ca. 40, 25 and 55% for arsenic, chromium and nickel, respectively (Ilyin et al., 2006). Between 1995 and 2000 the decrease in the anthropogenic emission of arsenic was about 17%. In addition, the Task Force on Heavy Metals (2006) reported the following decreases in anthropogenic emissions of copper and zinc between 1990 and 2000 for 17 European countries: ca. 24 and 27% respectively.

For Europe as a whole total emission (including anthropogenic, natural and historical) and deposition trends should be of a similar magnitude. At a smaller scale (regions, country, provinces etc.) the trends can be different, depending on local emissions, depositions from long-range transport, meteorological peculiarities, site specific characteristics (e.g. Schröder et al., in press) etc. However, when comparing deposition trends or the heavy metal concentrations in mosses with the trends in anthropogenic emissions, the latter should be steeper as the annual natural plus historical emissions are almost the same from year to year according to EMEP parameterizations (Ilyin, pers. comm.). This was indeed the case when comparing the temporal trends in the concentrations in mosses with the temporal trends in the anthropogenic emissions reported by EMEP for As (between 1995 and 2000), Cr and Ni. For plant essential trace elements such as copper and zinc the difference in temporal trends between the concentrations in mosses and anthropogenic emissions would be expected to be even bigger since mosses recycle these essential elements within the plant and have a background level for essential trace elements. However, no big difference were observed at the European scale in the current study: 16 – 19% decrease in the concentration in mosses compared to a 24 – 27% decrease in anthropogenic emissions for copper and zinc reported by the Task Force on Heavy Metals (2006).

Other studies have reported in more detail on the temporal trends of the heavy metal concentrations in mosses at the national level, showing a decline for the majority of metals in the final decade(s) of the 20th century (e.g. Nikodemus et al., 2004; Poikolainen et al., 2004; Rühling and Tyler, 2004; Schröder and Pesch, 2005; Steinnes et al., 2003; Suchara and Sucharová, 2004). This decline can mainly be attributed to cleaner industries and road transport, but also to a decrease in domestic emissions (e.g. Poikolainen et al., 2004), resulting in a significant decline in the deposition of heavy metals from long-range atmospheric transport (Poikolainen et al., 2004; Rühling and Tyler, 2004; Steinnes et al., 2003).

For 2003, the following anthropogenic sources were identified as the main contributors to anthropogenic emissions for nine countries (Belgium, Finland, France, Hungary, the Netherlands, Slovakia, Spain and the UK) across Europe (Task Force on Heavy Metals, 2006):

- Arsenic: Other, manufacturing industries and contructions (29%); Non-ferrous metals (22%);
- Chromium: Metal production (23%); Other, manufacturing industries and construction (22%);
- Copper: Road transportation (45%); Non-ferrous metals (15%);
- Nickel: Petrol refining (33%); Public electricity and heat production (22%);
- Zinc: Road transportation (42%); Metal production (21%).

The anthropogenic emission sources for cadmium, lead and mercury were described in detail elsewhere (Harmens et al., in press). Although both nickel and vanadium are thought to derive from crude oil combustion, the moss maps for these metals were quite different for the early European surveys; for the most recent survey in 2000 the maps for nickel and vanadium appear to be more similar. Nevertheless, for the whole of Europe the vanadium to nickel ratio in mosses did not change, based on the average median values for countries that determined the metals in both 1990 and 2000: 1.66 and 1.63 in 1990 and 2000 respectively. Differences in nickel and vanadium deposition maps have also been reported at the national scale (e.g. Fowler et al., 2006).

When examining the results of the moss surveys it should be kept in mind that the heavy metal concentrations in mosses do not directly reflect the total deposition of heavy metals. There are differences in the accumulation of individual heavy metals in mosses and the heavy metal concentrations in mosses are also affected by factors other than atmospheric pollution. These factors were discussed in more detail by Harmens et al. (in press). However, the similarity in temporal trends reported for the data of the European moss survey and the modelled total depositions of cadmium, lead and mercury suggests that at the European scale the reported temporal trends for these metals were not affected by any potential confounding factors (Harmens et al., in press).

5. Conclusions

Mosses provide a cheap and effective method for monitoring temporal trends in heavy metal pollution in Europe. Temporal trends in the concentrations of arsenic, chromium, copper, nickel and zinc in mosses were in agreement with those reported for anthropogenic emissions of these metals in Europe. Reductions in anthropogenic emissions of heavy metals between 1990 and 2000 have resulted in a significant reduction of the accumulation of arsenic, copper, vanadium and zinc in mosses. Decreases in the concentrations of chromium, iron and nickel in mosses with time were not significant. Therefore, the observed temporal trends in the concentration of heavy metals in mosses were metal-specific. In addition, many temporal trends were country-specific.

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(1994), Rühling and Steinnes (1998) and Buse et al. (2003)). We thank Tim Sparks (CEH Monks Wood) for statistical advice.

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Figure captions

Figure 1. The mean concentration of arsenic in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 2. The mean concentration of chromium in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 3. The mean concentration of copper in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 4. The mean concentration of iron in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 5. The mean concentration of nickel in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 6. The mean concentration of vanadium in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Figure 7. The mean concentration of zinc in moss per EMEP grid square (50 km x 50 km) for 1990 (a), 1995 (b) and 2000 (c).

Table 1. Median values of arsenic (As), chromium (Cr) and copper (Cu) concentrations in mosses across Europe in 1990, 1995 and 2000; - = not determined.

	As ($\mu g\ g^{-1}$)			Cr ($\mu g\ g^{-1}$)			Cu ($\mu g\ g^{-1}$)		
Country	1990	1995	2000	1990	1995	2000	1990	1995	2000
Austria	0.56	0.13	0.10	1.85	0.70	0.73	5.85	5.35	6.13
Bulgaria	-	-	0.21	-	2.30	2.41	-	14.70	14.51
Czech Republic	1.70	0.50	0.29	1.90	1.37	1.88	8.40	7.15	6.52
Denmark	-	0.27	-	1.22	0.65	-	6.41	4.73	-
- Faroe Islands	-	-	0.15	-	0.68	0.68	-	5.47	6.84
Estonia	-	0.23	-	1.63	0.77	1.01	5.48	3.64	3.39
Finland	-	0.23	0.16	1.47	1.43	1.06	5.07	4.46	3.38
France	-	0.30	0.23	-	3.16	1.69	-	5.30	6.40
Germany	0.34	0.25	0.16	1.83	1.39	0.91	9.13	9.57	7.14
Hungary	-	-	-	-	3.61	6.40	-	5.77	7.65
Iceland	-	0.07	0.14	2.33	2.38	2.61	8.42	8.09	8.36
Italy	-	0.29	0.40	2.16	2.47	3.80	8.90	8.90	9.10
Latvia	-	-	0.06	1.46	1.13	0.95	6.03	3.79	5.10
Lithuania	-	0.40	0.32	1.17	1.31	1.27	6.55	5.87	6.45
Netherlands	0.39	0.41	-	2.45	4.23	-	13.21	23.96	-
Norway	0.27	0.21	0.13	0.90	1.05	0.69	5.22	5.21	4.26
Poland	-	-	-	2.34	1.50	0.89	9.30	7.60	8.03
Portugal	-	-	0.33	1.40	2.17	1.08	7.00	7.37	6.16
Romania	-	0.96	1.56	10.85	9.15	8.46	18.42	11.30	21.56
Russian Fed.	-	0.24	0.21	-	1.27	1.43	-	7.12	5.84
- St. Petersburg	-	-	0.17	-	1.99	1.42	4.90	4.58	5.19
Slovakia	-	-	0.71	3.55	13.21	6.45	18.60	16.35	8.76
Slovenia	-	0.38	0.33	-	4.29	2.59	-	8.40	-
Spain	-	0.19	0.21	4.89	2.71	5.73	7.78	6.07	4.24
Sweden	-	0.15	0.16	1.28	0.60	0.68	5.47	4.58	4.36
Switzerland	0.33	0.12	0.12	2.40	0.76	0.89	3.90	4.30	4.35
Ukraine	-	0.10	0.24	-	1.70	1.50	-	6.20	7.31
United Kingdom	-	0.37	0.16	0.60	1.40	1.47	6.10	5.43	4.32

Table 2. Median values of nickel (Ni), vanadium (V) and zinc (Zn) concentrations in mosses across Europe in 1990, 1995 and 2000; - = not determined.

	Ni (μg g^{-1})			V (μg g^{-1})			Zn (μg g^{-1})		
Country	1990	1995	2000	1990	1995	2000	1990	1995	2000
Austria	2.50	1.30	1.26	2.00	1.30	1.27	36.6	30.0	31.5
Bulgaria	-	3.06	3.33	-	4.90	4.95	-	30.5	32.6
Czech Republic	3.40	1.95	1.95	5.40	2.00	1.52	45.5	41.9	35.0
Denmark	1.32	1.38	-	2.66	2.51	-	36.0	41.8	-
- Faroe Islands	-	1.56	1.73	-	4.36	3.34	-	14.6	14.4
Estonia	2.07	1.21	1.01	2.88	3.90	1.72	30.8	32.8	31.5
Finland	1.70	1.65	1.38	3.36	2.18	1.24	35.9	37.5	27.6
France	-	1.94	2.30	-	2.46	2.89	-	32.4	40.4
Germany	2.38	1.64	1.13	2.87	1.71	1.06	50.2	54.0	41.0
Hungary	-	4.00	5.35	-	4.62	4.20	-	27.6	30.0
Iceland	2.59	2.96	3.32	12.15	11.30	11.95	18.2	17.2	27.7
Italy	1.47	2.28	3.80	-	3.10	5.89	31.3	45.0	48.3
Latvia	1.40	1.07	0.98	3.19	3.05	1.80	41.7	30.2	31.0
Lithuania	1.75	1.78	1.36	3.34	4.58	3.44	42.0	40.0	34.5
Netherlands	2.64	15.00	-	4.71	4.53	-	47.5	68.6	-
Norway	1.56	1.63	1.11	2.36	2.27	1.36	36.4	37.7	29.5
Poland	2.21	1.44	1.57	4.80	4.00	5.84	53.1	43.0	41.5
Portugal	1.80	10.75	1.21	-		2.72	29.0	40.4	28.1
Romania	8.41	2.19	3.35	12.53	6.40	7.99	69.1	43.9	79.6
Russian Fed.	-	4.98	2.01	-	3.03	2.79	-	38.0	35.3
- St. Petersburg	6.70	2.70	2.05	5.10	4.13	2.18	42.0	48.1	36.2
Slovakia	1.70	1.99	3.15	-	0.12	5.70	162.5	49.1	55.0
Slovenia	-	2.76	-	-	4.00	-	-	38.8	34.5
Spain	3.86	1.95	4.16	9.60	-	-	35.4	40.7	30.0
Sweden	1.50	1.11	1.41	2.36	2.19	1.31	43.7	40.0	38.8
Switzerland	3.00	1.25	1.22	2.03	1.40	0.88	29.8	30.8	29.7
Ukraine	-	2.69	2.06	-	1.80	1.29		31.0	29.3
United Kingdom	1.60	1.52	0.83	1.40	1.55	0.99	29.2	34.2	22.7

Table 3. Median values of iron (Fe) concentrations in mosses across Europe in 1990, 1995 and 2000; - = not determined.

	Fe (μg g^{-1})				Fe (μg g^{-1})		
Country	1990	1995	2000	Country	1990	1995	2000
Austria	544	340	409	Netherlands	590	645	-
Bulgaria	-	1587	1412	Norway	466	332	365
Czech Republic	747	401	401	Poland	1190	362	429
Denmark	427	375	-	Portugal	812	1116	561
- Faroe Islands	-	457	754	Romania	5114	1937	2518
Estonia	619	372	289	Russian Fed.	-	436	537
Finland	357	275	210	- St. Petersburg	1050	645	422
France	-	549	654	Slovakia	1555	1483	1561
Germany	561	443	343	Slovenia	-	1007	713
Hungary	-	953	1760	Spain	3475	497	243
Iceland	3187	2877	4073	Sweden	298	184	228
Italy	709	663	1408	Switzerland	312	265	337
Latvia	466	363	134	Ukraine	-	333	313
Lithuania	555	580	623	United Kingdom	145	347	-

Table 4. Average median values of metal concentrations in mosses for countries that determined the metals both in 1990 and 2000, and their decrease with time.

Metal	Median 1990 ($\mu g\ g^{-1}$)	Median 2000 ($\mu g\ g^{-1}$)	Decrease with time (%)	Number of countries
As[1]	0.64	0.16	75	5
Cr	2.44	2.25	8	18
Cu	7.92	6.67	16	19
Fe	1223	809	44	18
Ni	2.72	1.91	30	19
V	4.38	2.97	32	15
Zn	45.4	36.8	19	19

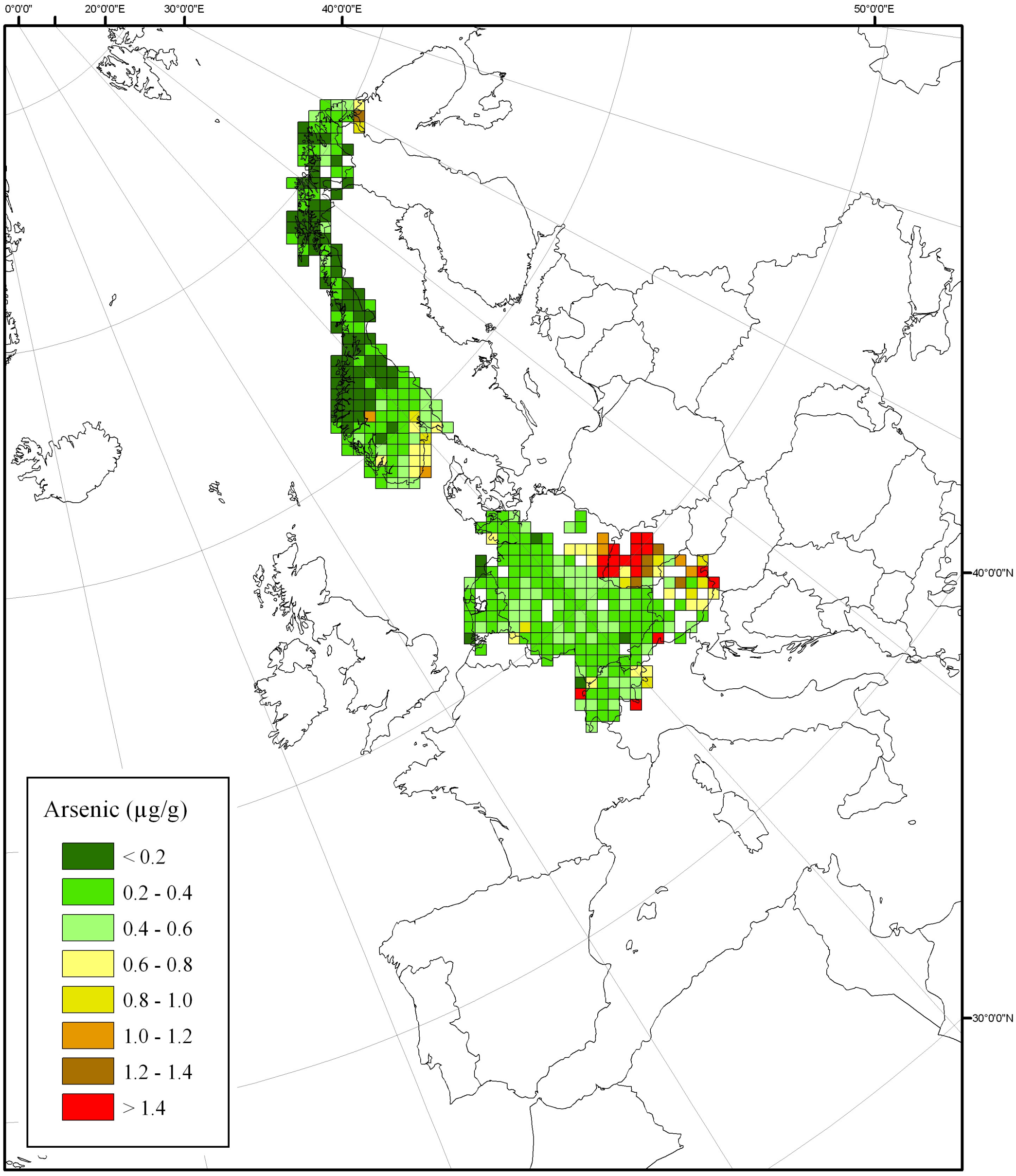
[1] For arsenic the values are based on data from 5 countries only (see Table 1). The median value of arsenic concentrations in mosses for countries (n = 17) that analysed arsenic both in 1995 and 2000 is 0.29 for both years, indicating that arsenic concentrations in mosses primarily decreased between 1990 and 1995.

Table 5. Average geometric mean values of metal concentrations in mosses for countries that determined these metals in all three surveys; n = the number of countries. The statistical significance (P-value) of country and year of the survey are also shown.

Metal	Average geometric mean (µg g^{-1})				P-value	
	1990	1995	2000	n	Country	Year
As[1]	0.66	0.26	0.17	5	0.205	0.026
Cr	2.47	2.63	2.36	18	0.000	0.180
Cu	8.04	7.25	7.29	19	0.000	0.026
Fe	1262	765	852	18	0.000	0.099
Ni	2.76	2.30	2.00	19	0.049	0.074
V	4.32	3.59	2.96	15	0.000	0.000
Zn	46.8	40.2	38.9	19	0.000	0.021

[1] For arsenic the values are based on data from 5 countries only (see Table 1). The geometric mean values of arsenic concentrations in mosses for countries (n = 17) that analysed arsenic both in 1995 and 2000 are 0.32 and 0.31 respectively; therefore, the arsenic concentrations in mosses did not change significantly (P = 0.30) between 1995 and 2000 for those countries.

1990



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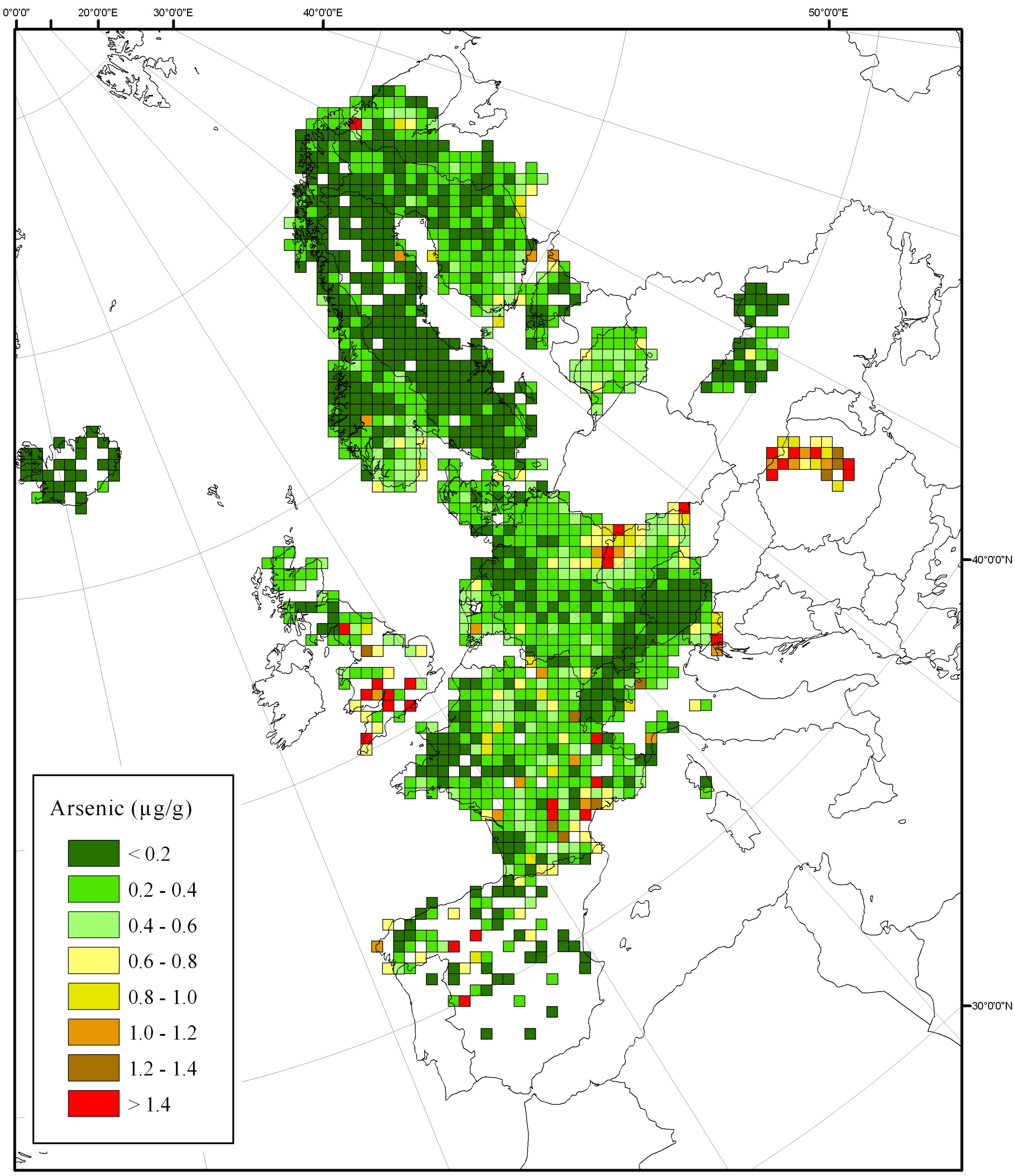
20°0'0"E
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Arsenic (µg/g)
< 0.2
0.2 - 0.4
0.4 - 0.6
0.6 - 0.8
0.8 - 1.0
1.0 - 1.2
1.2 - 1.4
> 1.4

1995



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30°0'0"E
40°0'0"E
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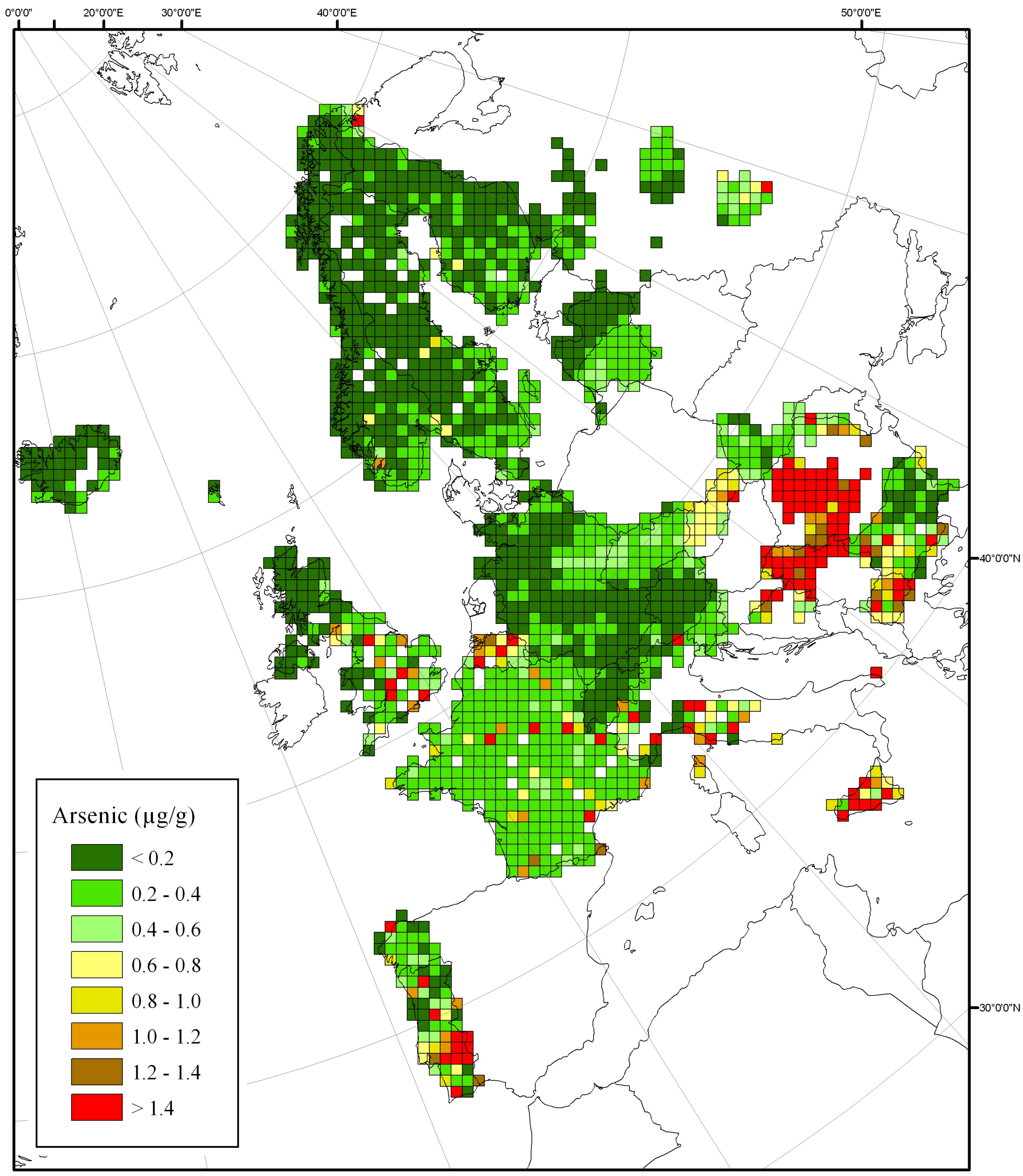
40°0'0"N
30°0'0"N
Arsenic (µg/g)
< 0.2
0.2 - 0.4
0.4 - 0.6
0.6 - 0.8
0.8 - 1.0
1.0 - 1.2
1.2 - 1.4
> 1.4

2000



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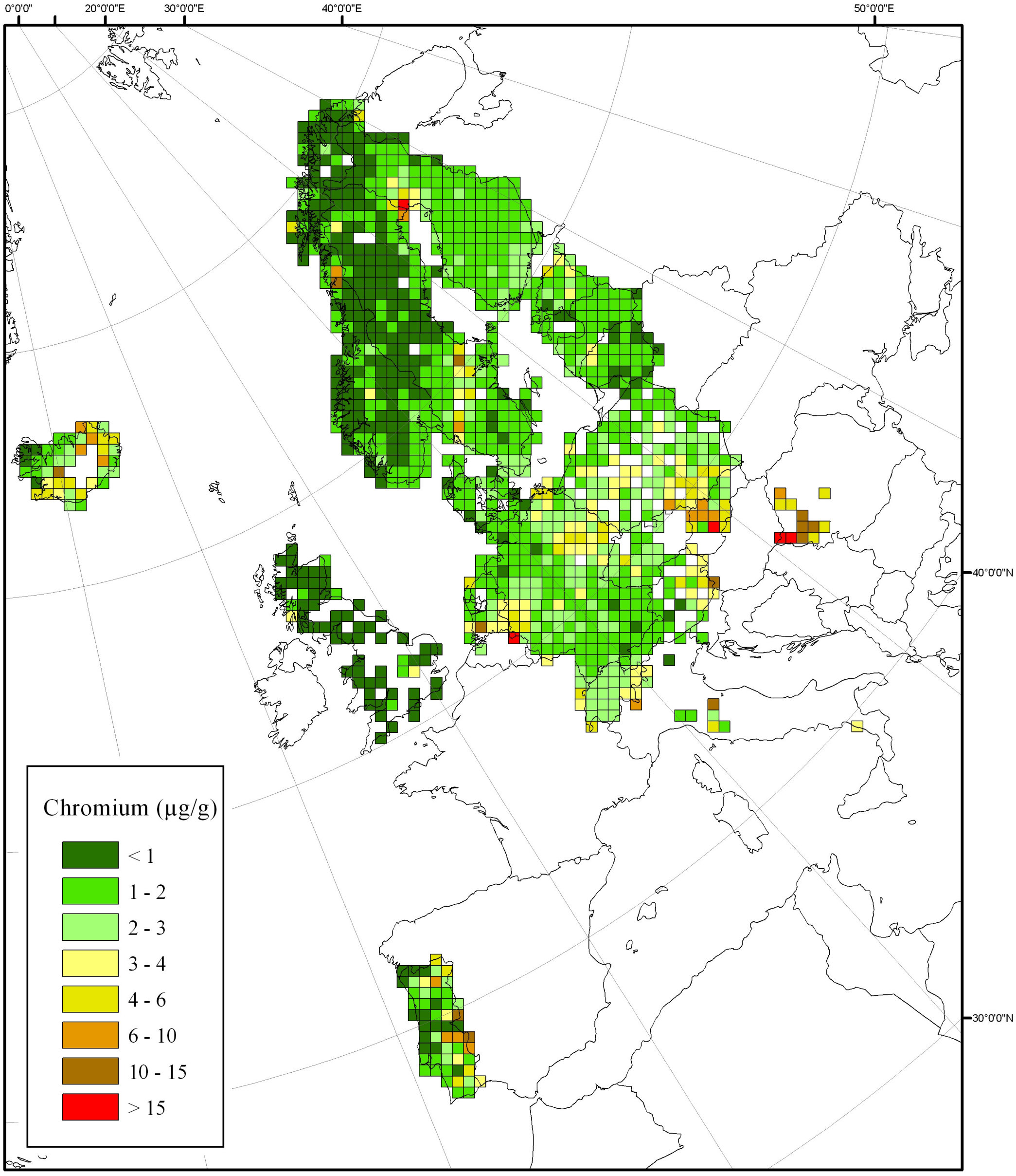
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Arsenic (µg/g)
< 0.2
0.2 - 0.4
0.4 - 0.6
0.6 - 0.8
0.8 - 1.0
1.0 - 1.2
1.2 - 1.4
> 1.4

1990



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40°0'0"E
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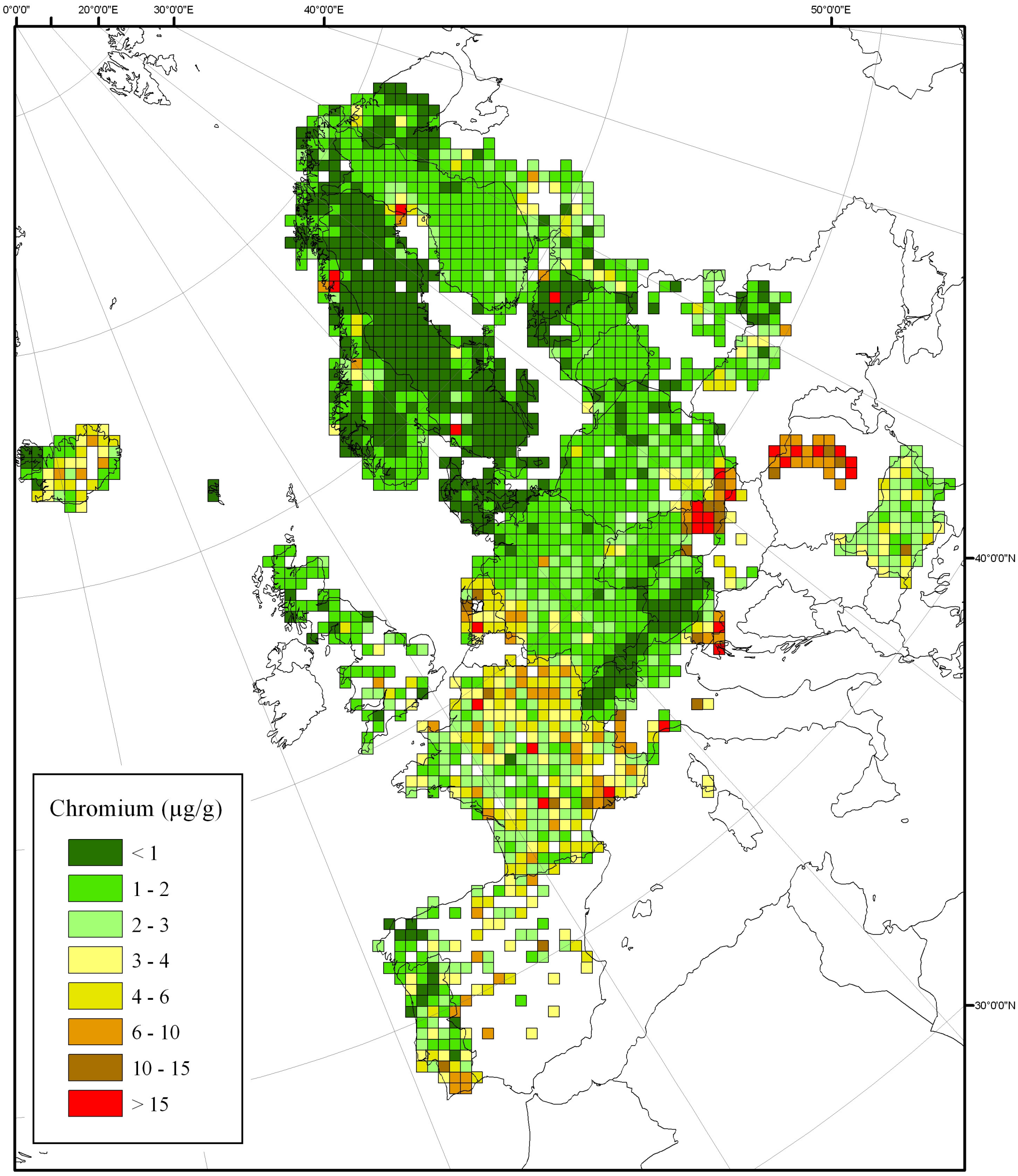
Chromium (µg/g)
< 1
1 - 2
2 - 3
3 - 4
4 - 6
6 - 10
10 - 15
> 15

1995



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40°0'0"N
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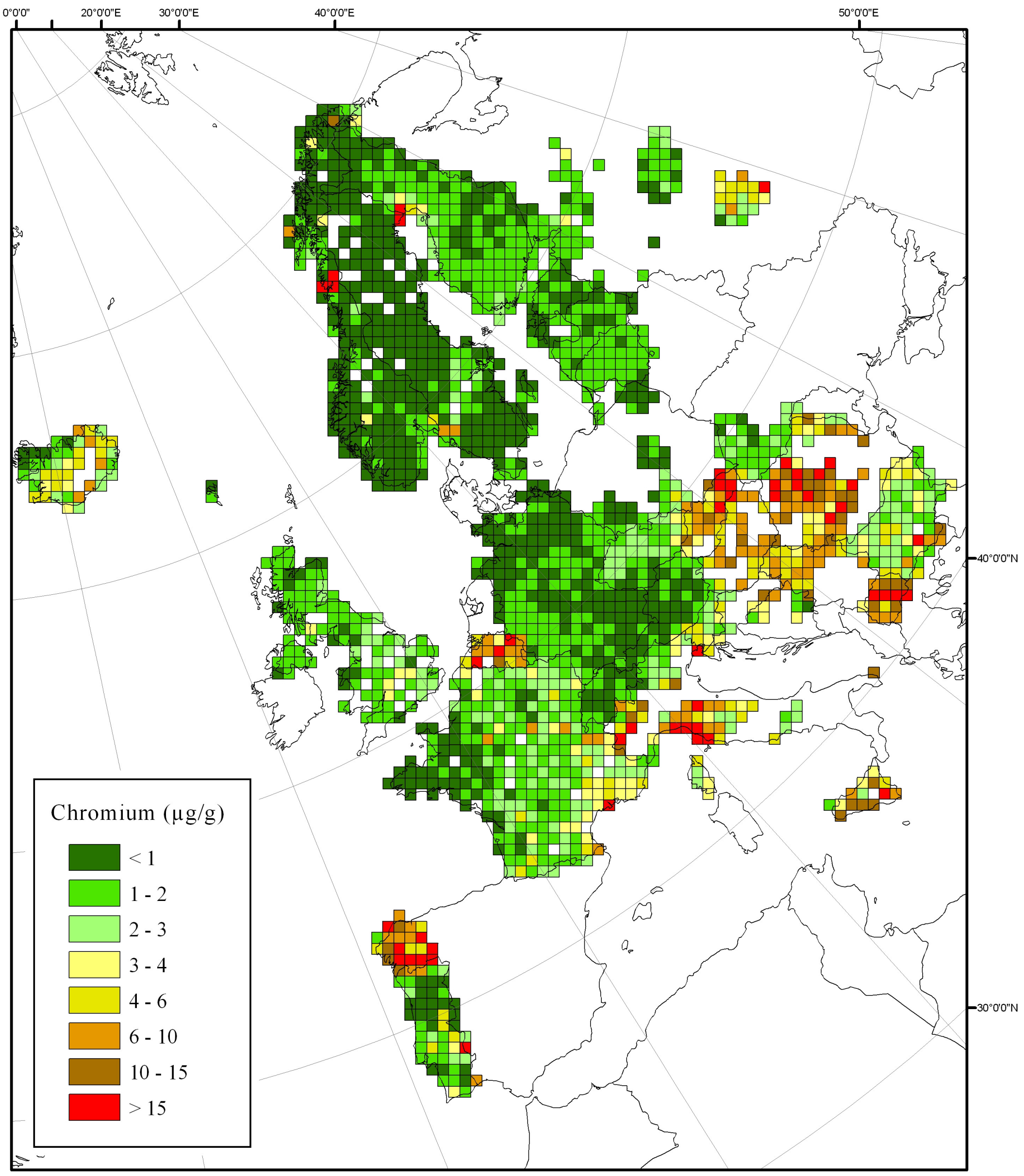
Chromium (µg/g)
< 1
1 - 2
2 - 3
3 - 4
4 - 6
6 - 10
10 - 15
> 15

2000



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40°0'0"N
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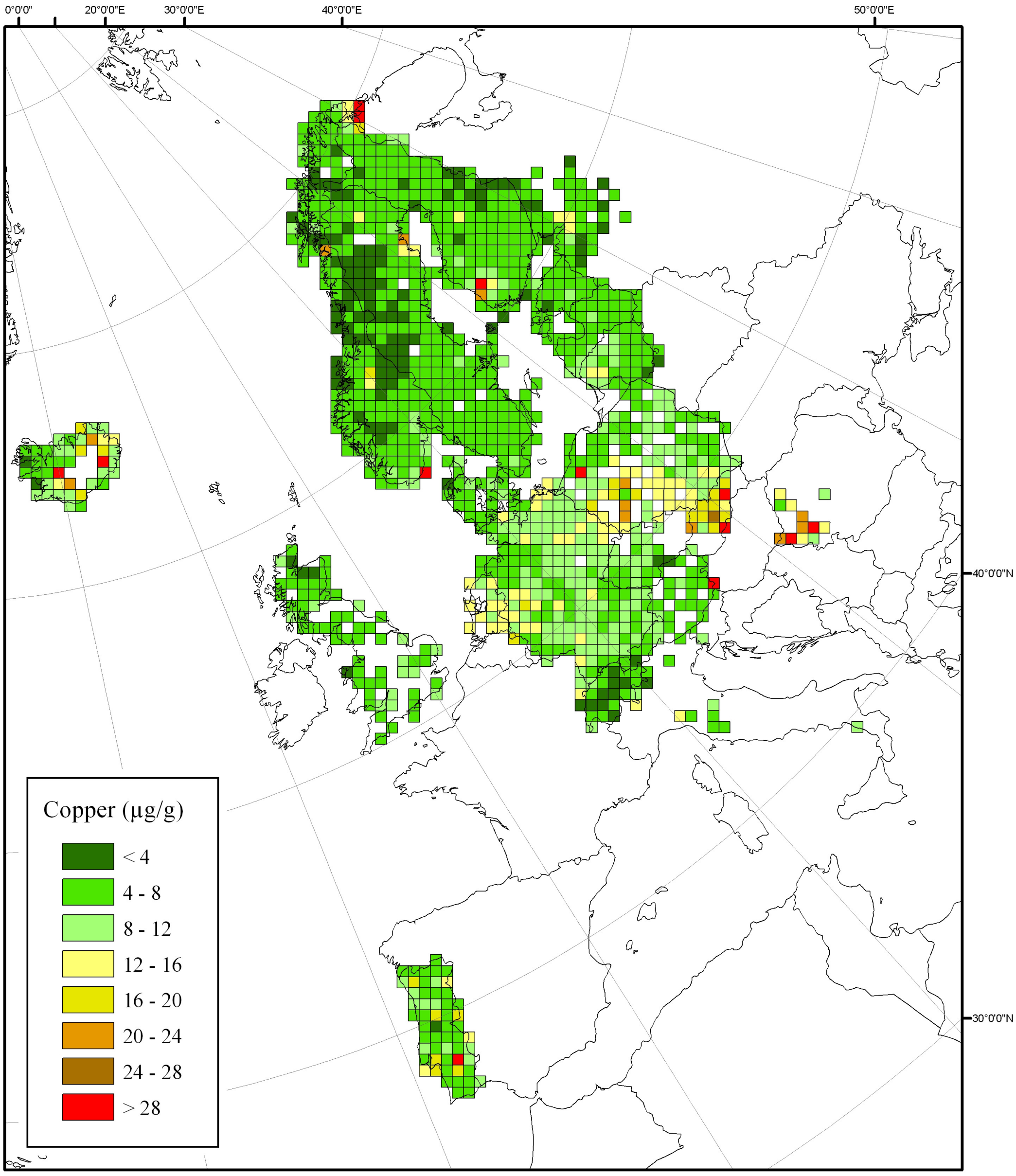
Chromium (µg/g)
< 1
1 - 2
2 - 3
3 - 4
4 - 6
6 - 10
10 - 15
> 15

1990



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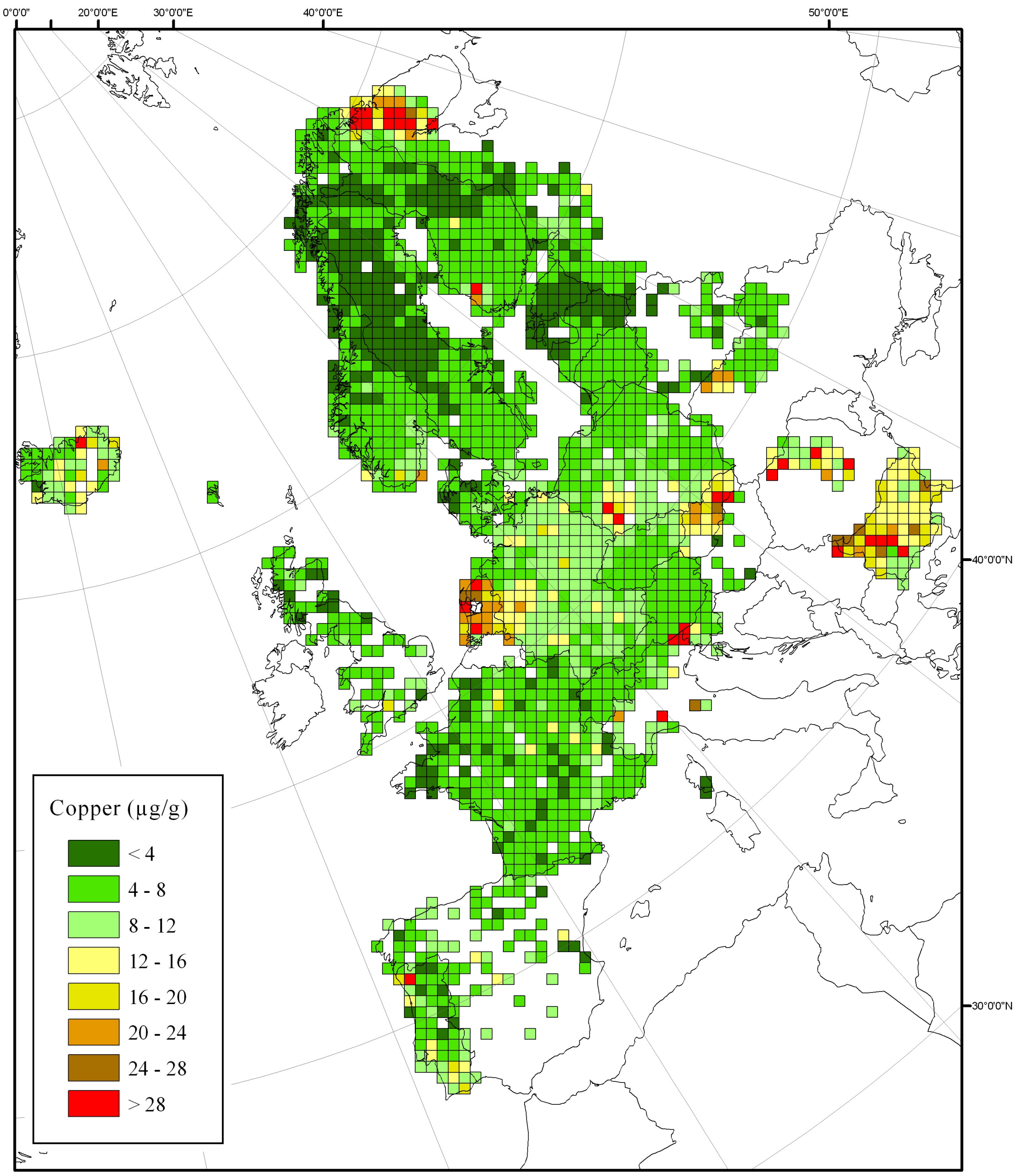
Copper (µg/g)
< 4
4 - 8
8 - 12
12 - 16
16 - 20
20 - 24
24 - 28
> 28

1995



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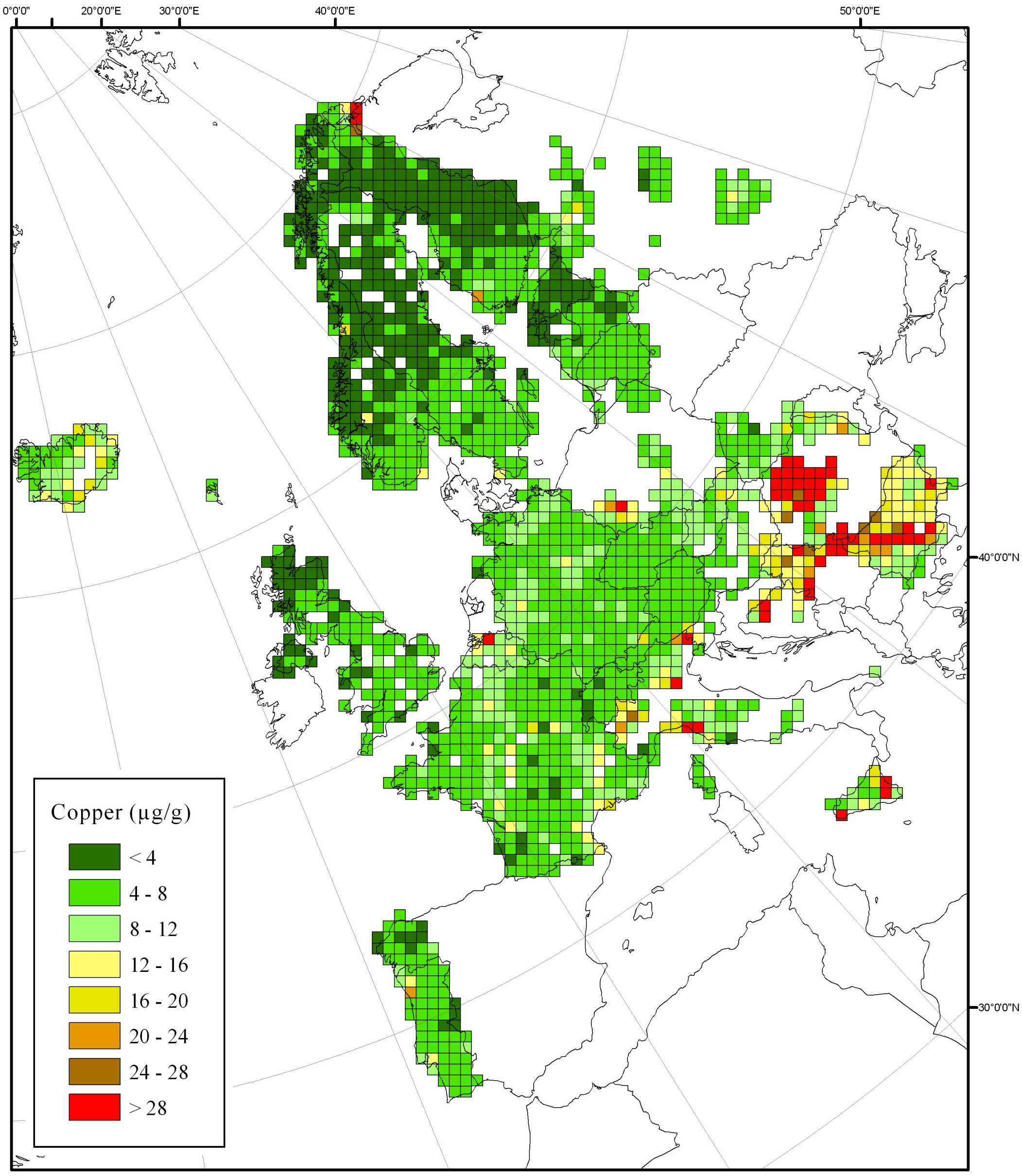
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Copper (µg/g)
< 4
4 - 8
8 - 12
12 - 16
16 - 20
20 - 24
24 - 28
> 28

2000



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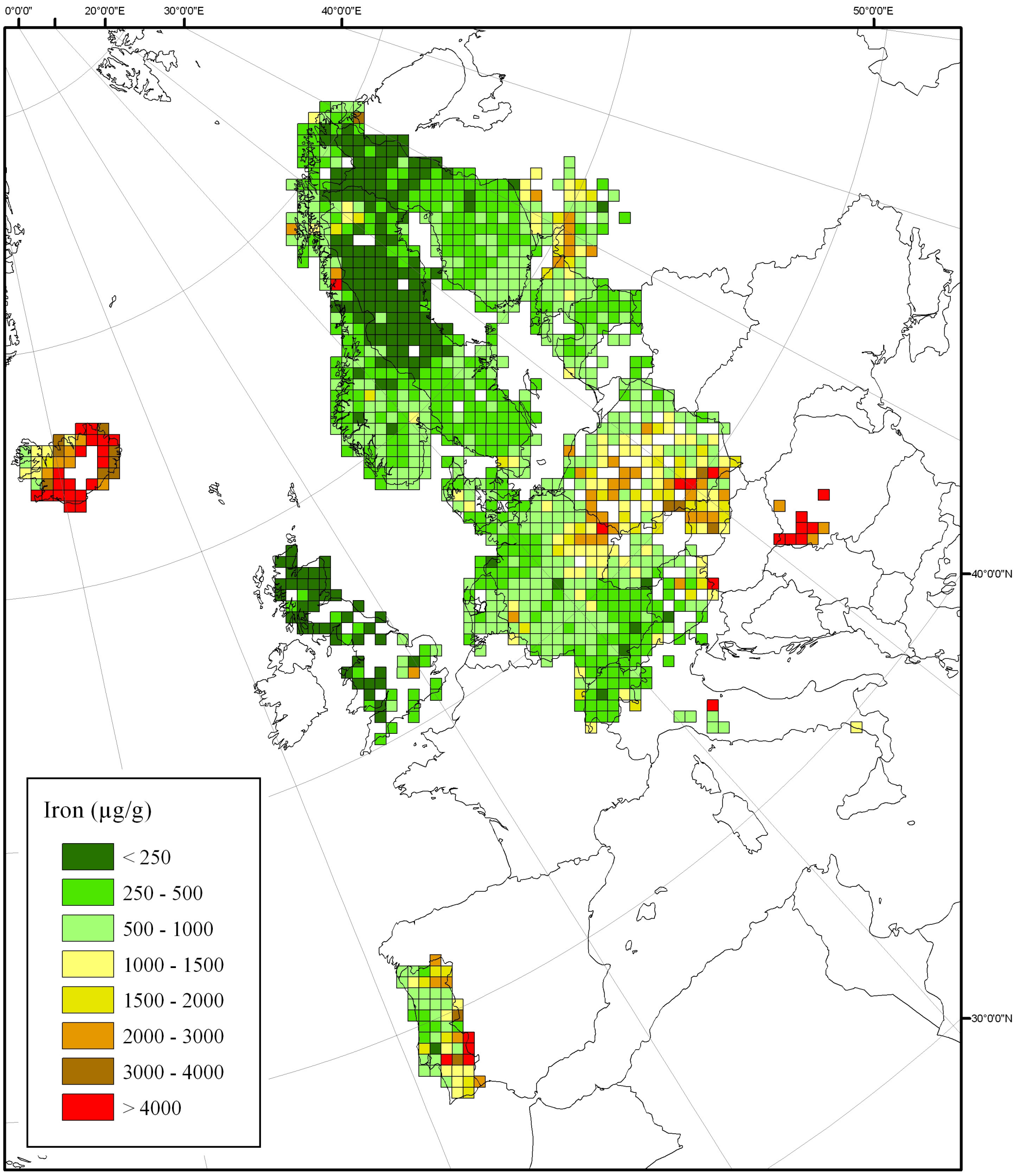
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Copper (µg/g)
< 4
4 - 8
8 - 12
12 - 16
16 - 20
20 - 24
24 - 28
> 28

1990



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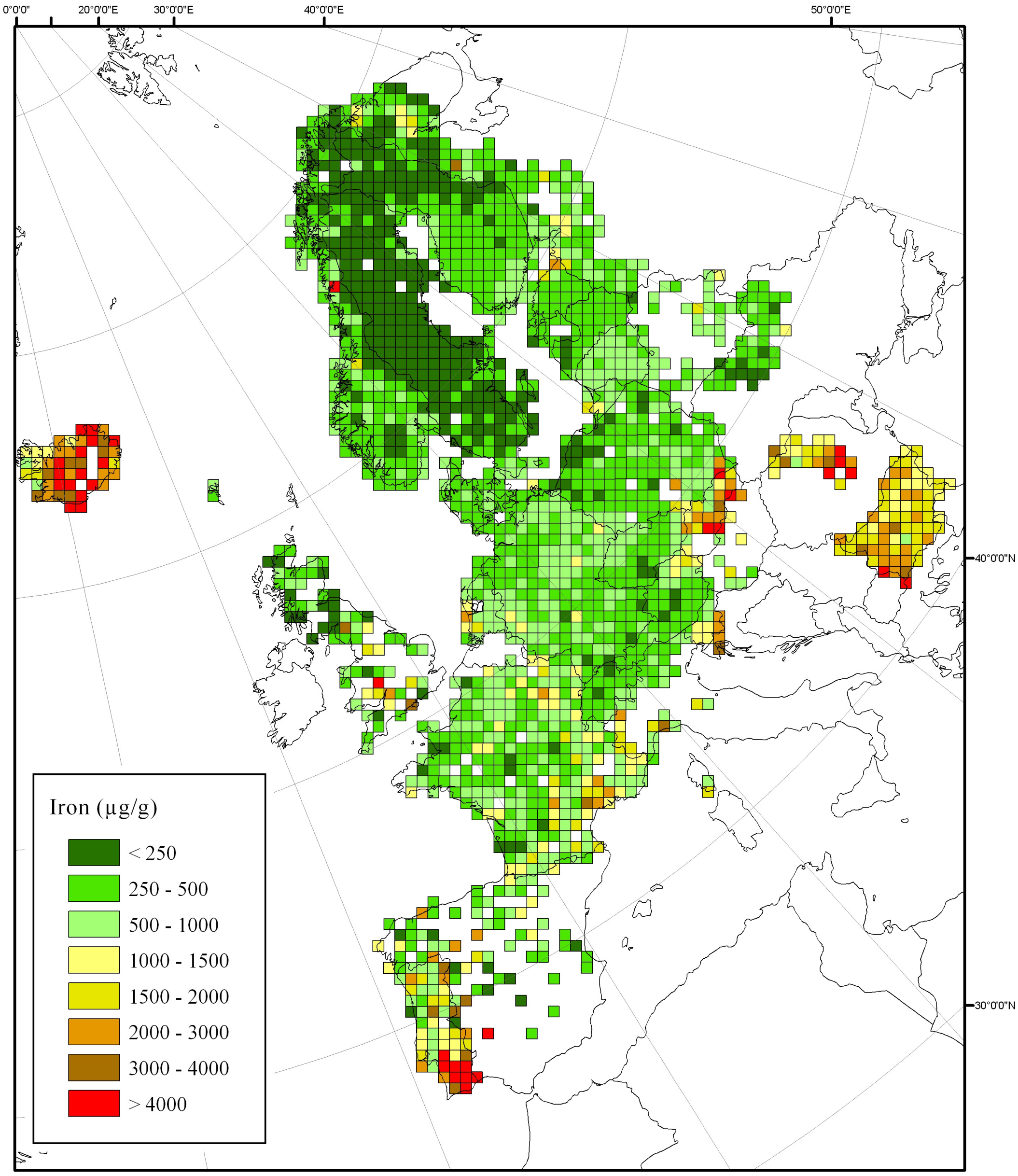
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Iron (µg/g)
< 250
250 - 500
500 - 1000
1000 - 1500
1500 - 2000
2000 - 3000
3000 - 4000
> 4000

1995



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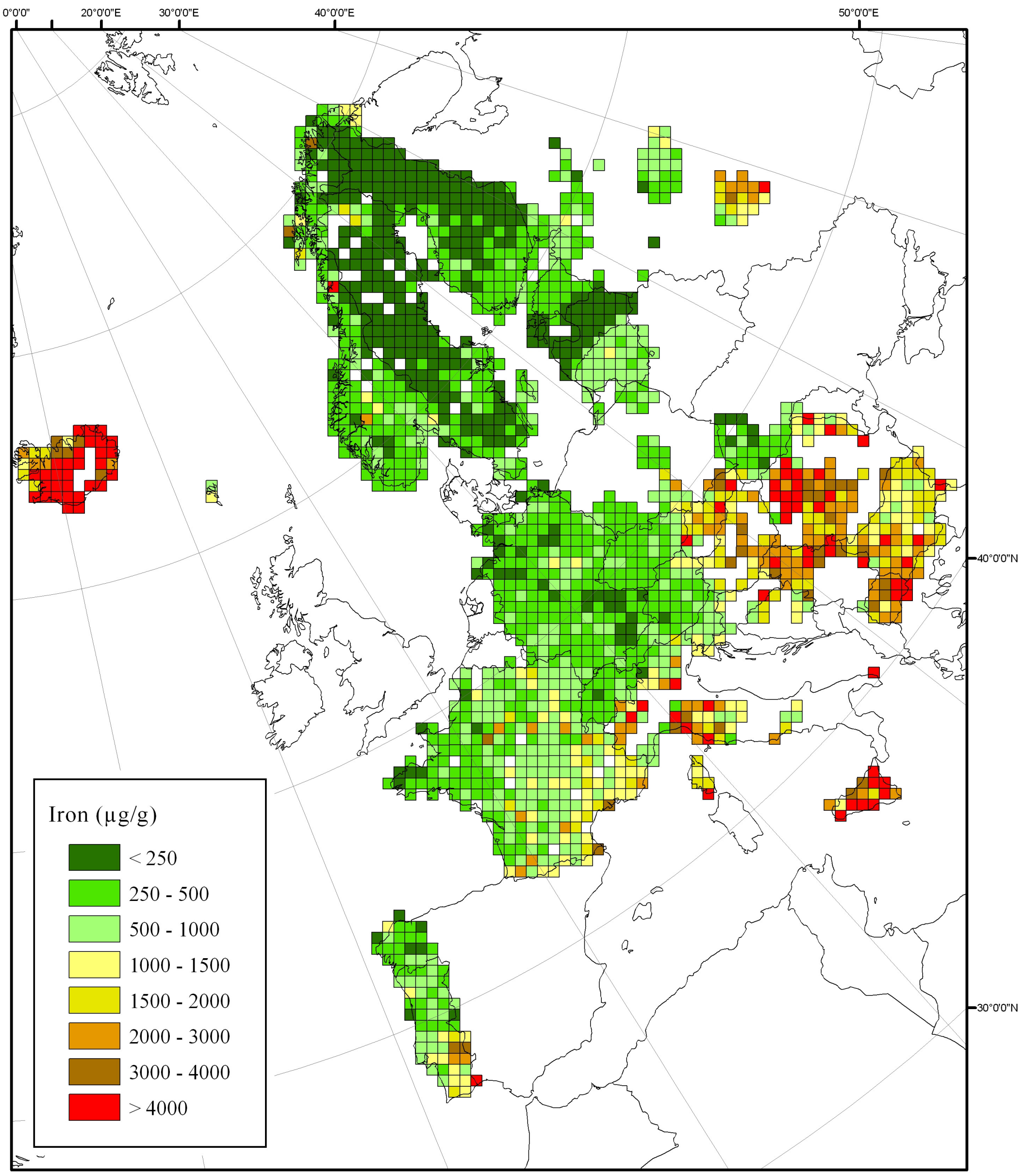
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Iron (µg/g)
< 250
250 - 500
500 - 1000
1000 - 1500
1500 - 2000
2000 - 3000
3000 - 4000
> 4000

2000



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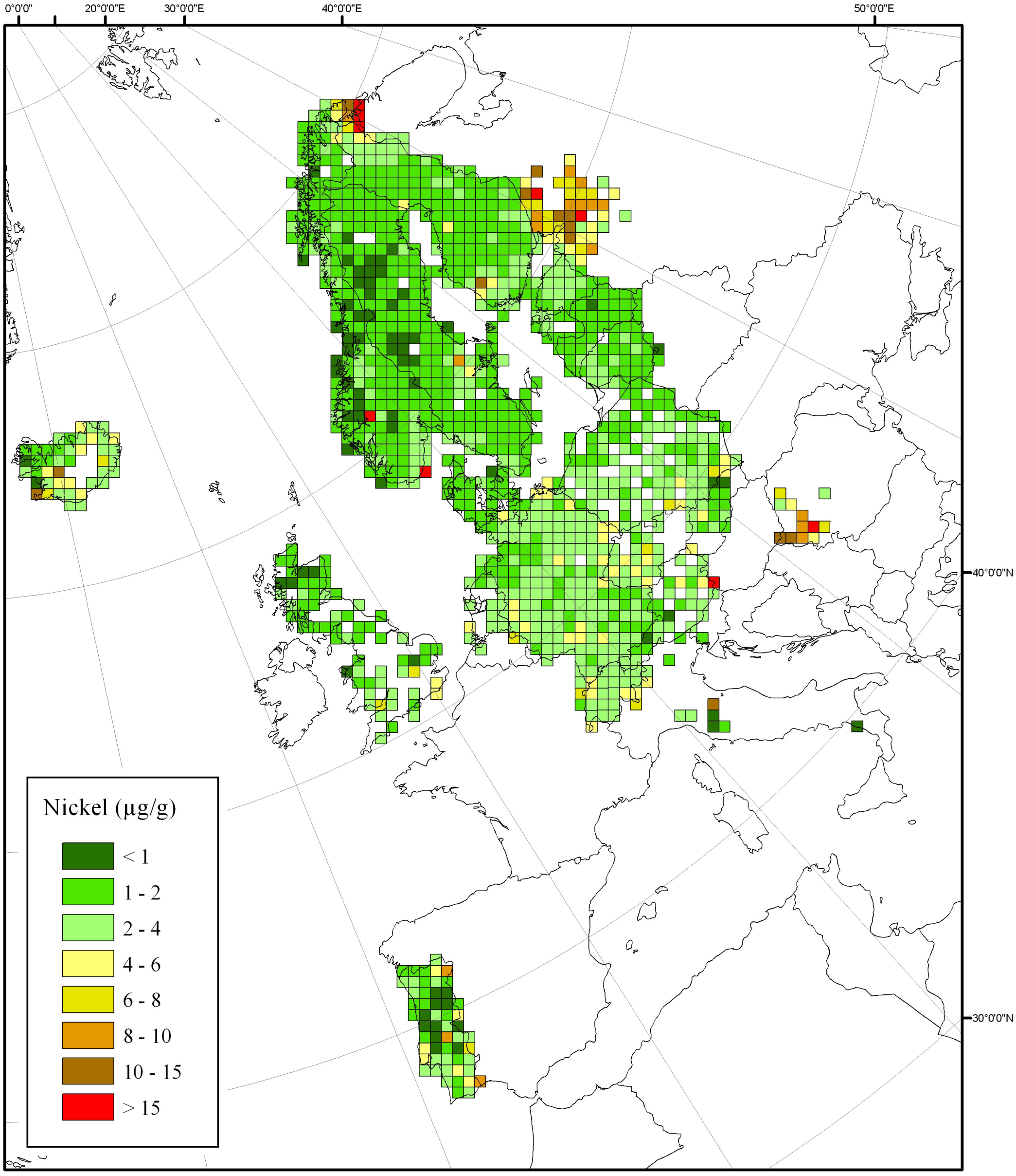
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Iron (µg/g)
< 250
250 - 500
500 - 1000
1000 - 1500
1500 - 2000
2000 - 3000
3000 - 4000
> 4000

1990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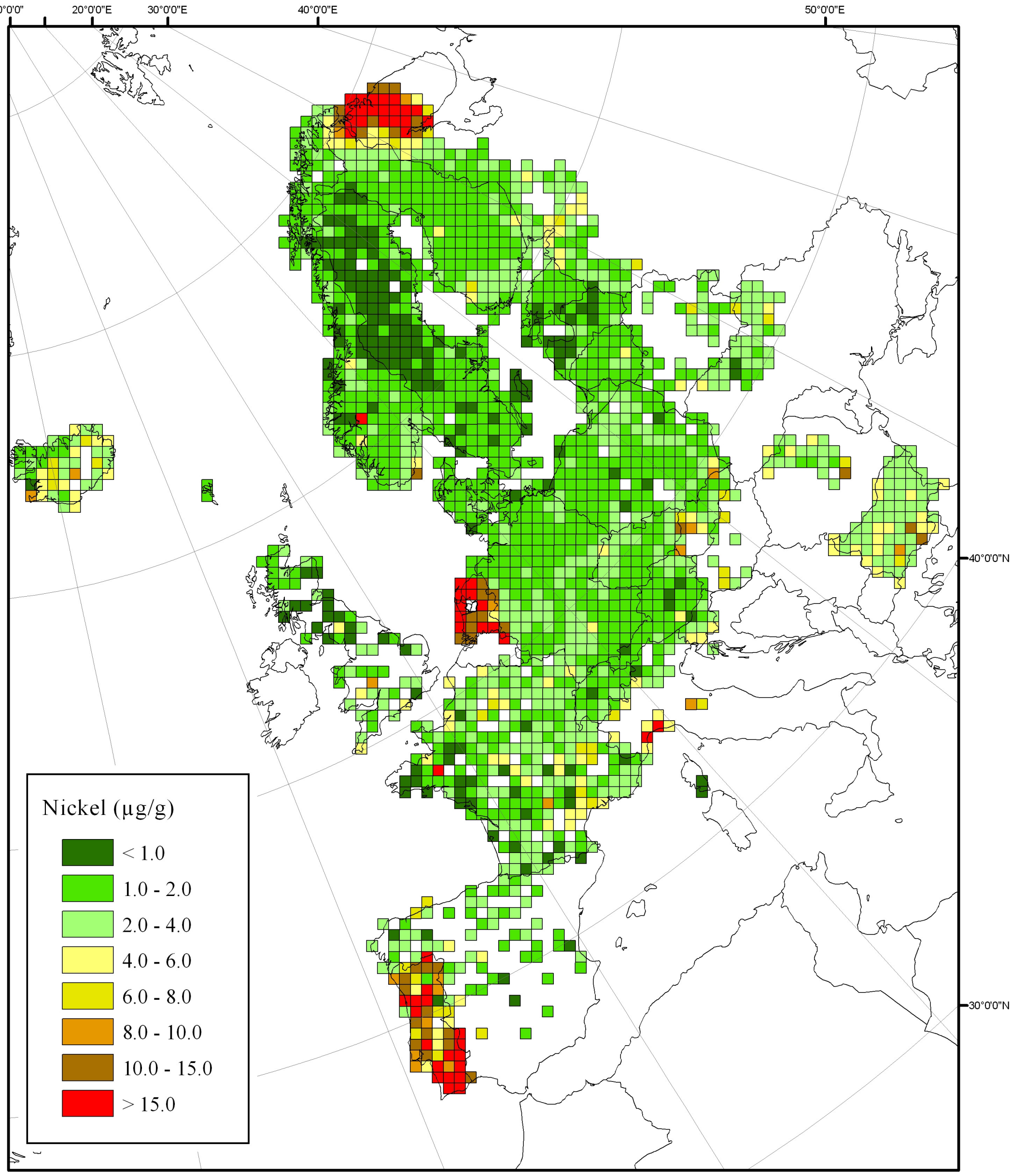
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Nickel (µg/g)
< 1
1 - 2
2 - 4
4 - 6
6 - 8
8 - 10
10 - 15
> 15

1995



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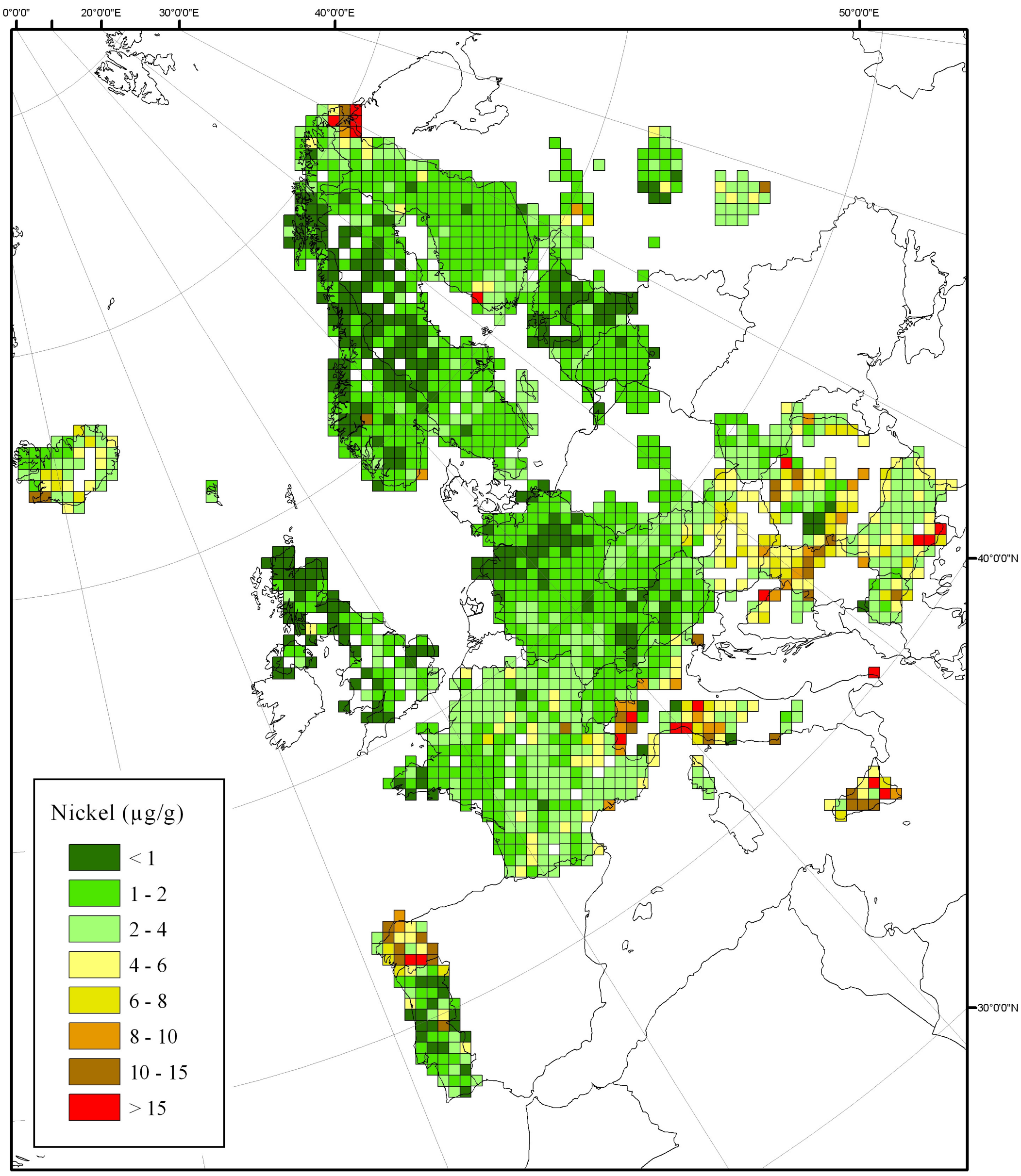
20°0'0"E
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Nickel (µg/g)
< 1.0
1.0 - 2.0
2.0 - 4.0
4.0 - 6.0
6.0 - 8.0
8.0 - 10.0
10.0 - 15.0
> 15.0

2000



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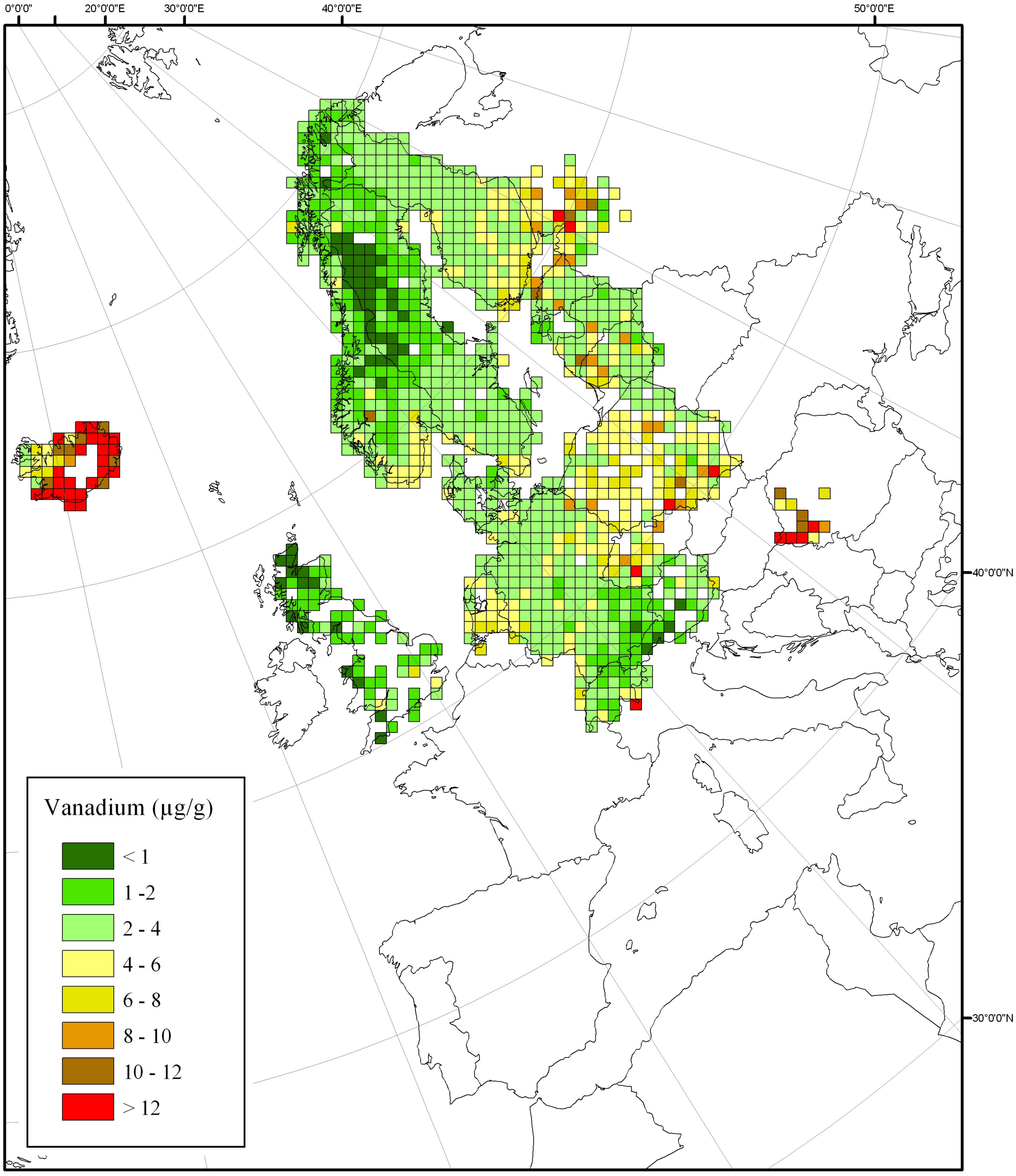
50°0'0"E
40°0'0"N
30°0'0"N
Nickel (µg/g)
< 1
1 - 2
2 - 4
4 - 6
6 - 8
8 - 10
10 - 15
> 15

1990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40°0'0"N
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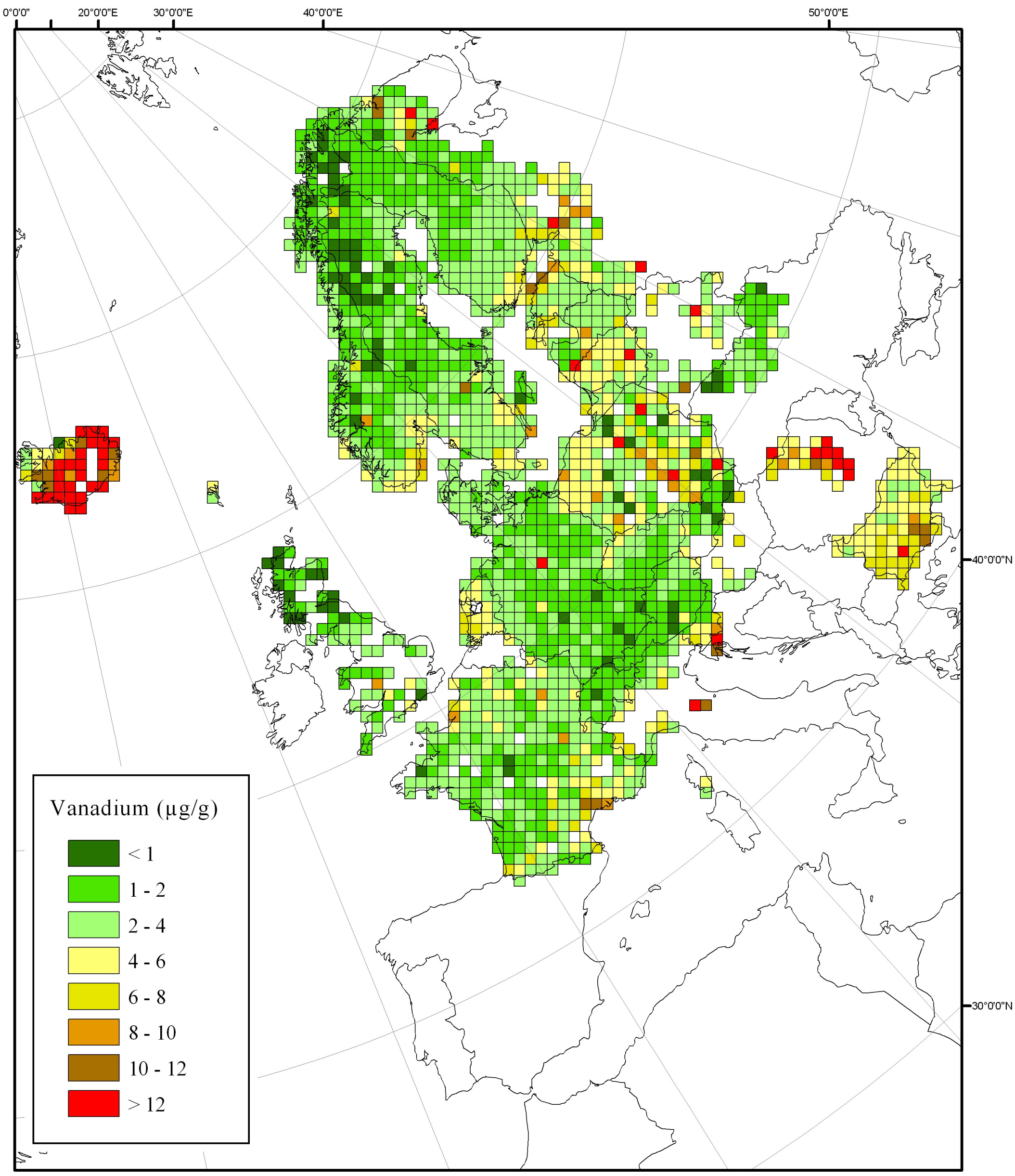
Vanadium (µg/g)
< 1
1 -2
2 - 4
4 - 6
6 - 8
8 - 10
10 - 12
> 12

1995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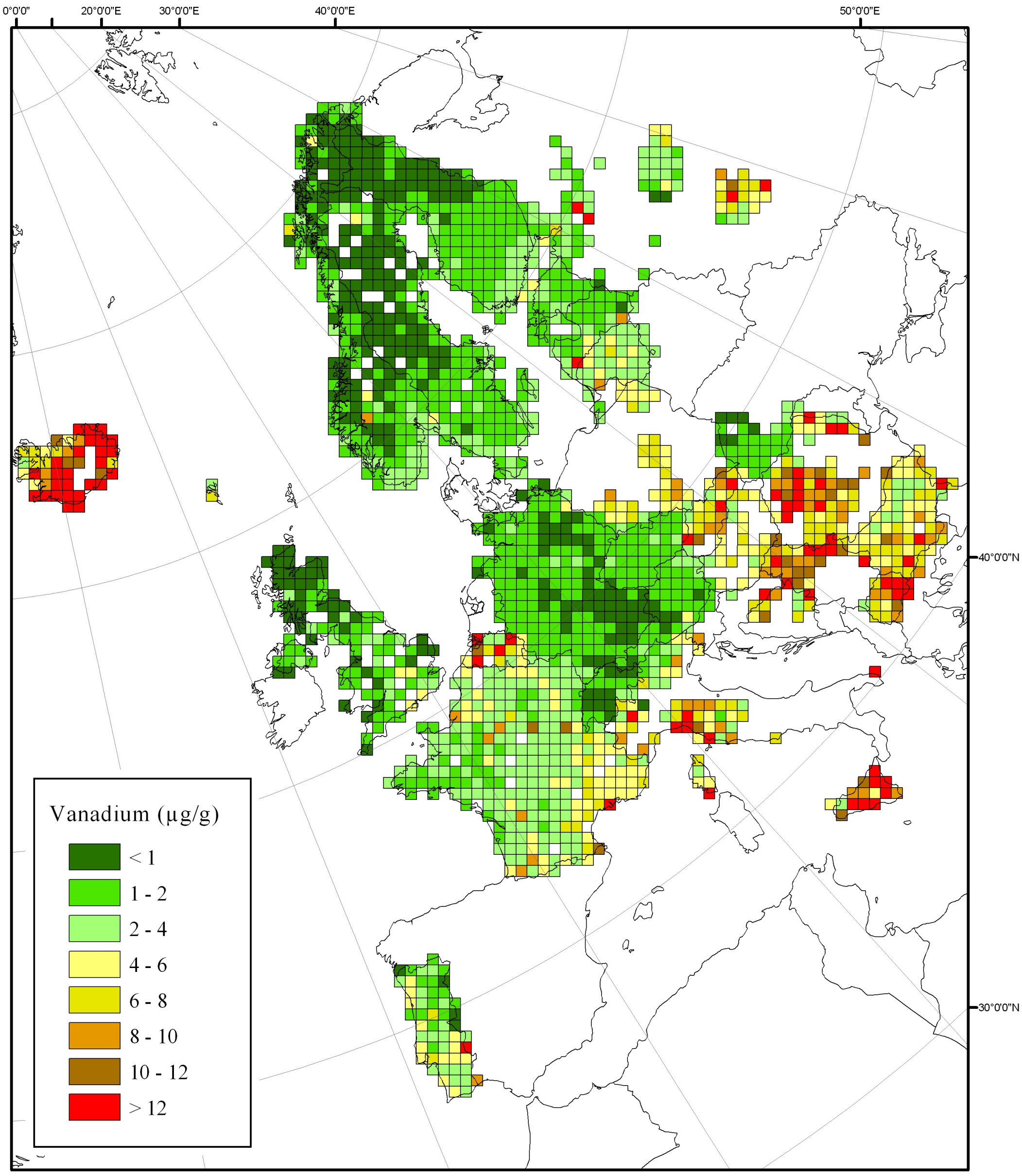
Vanadium (µg/g)
< 1
1 - 2
2 - 4
4 - 6
6 - 8
8 - 10
10 - 12
> 12

2000



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50°0'0"E
40°0'0"N
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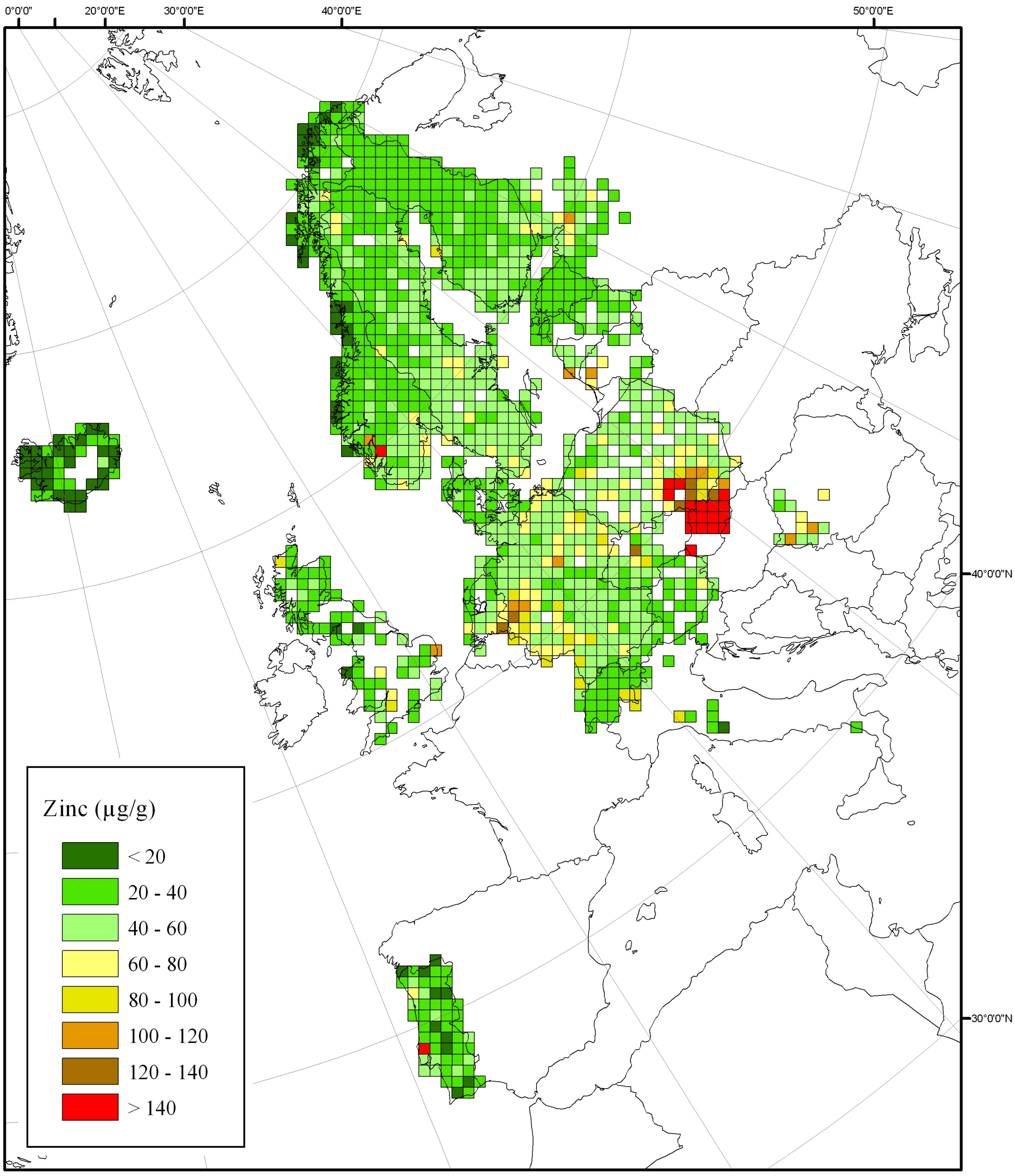
Vanadium (µg/g)
< 1
1 - 2
2 - 4
4 - 6
6 - 8
8 - 10
10 - 12
> 12

1990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40°0'0"N
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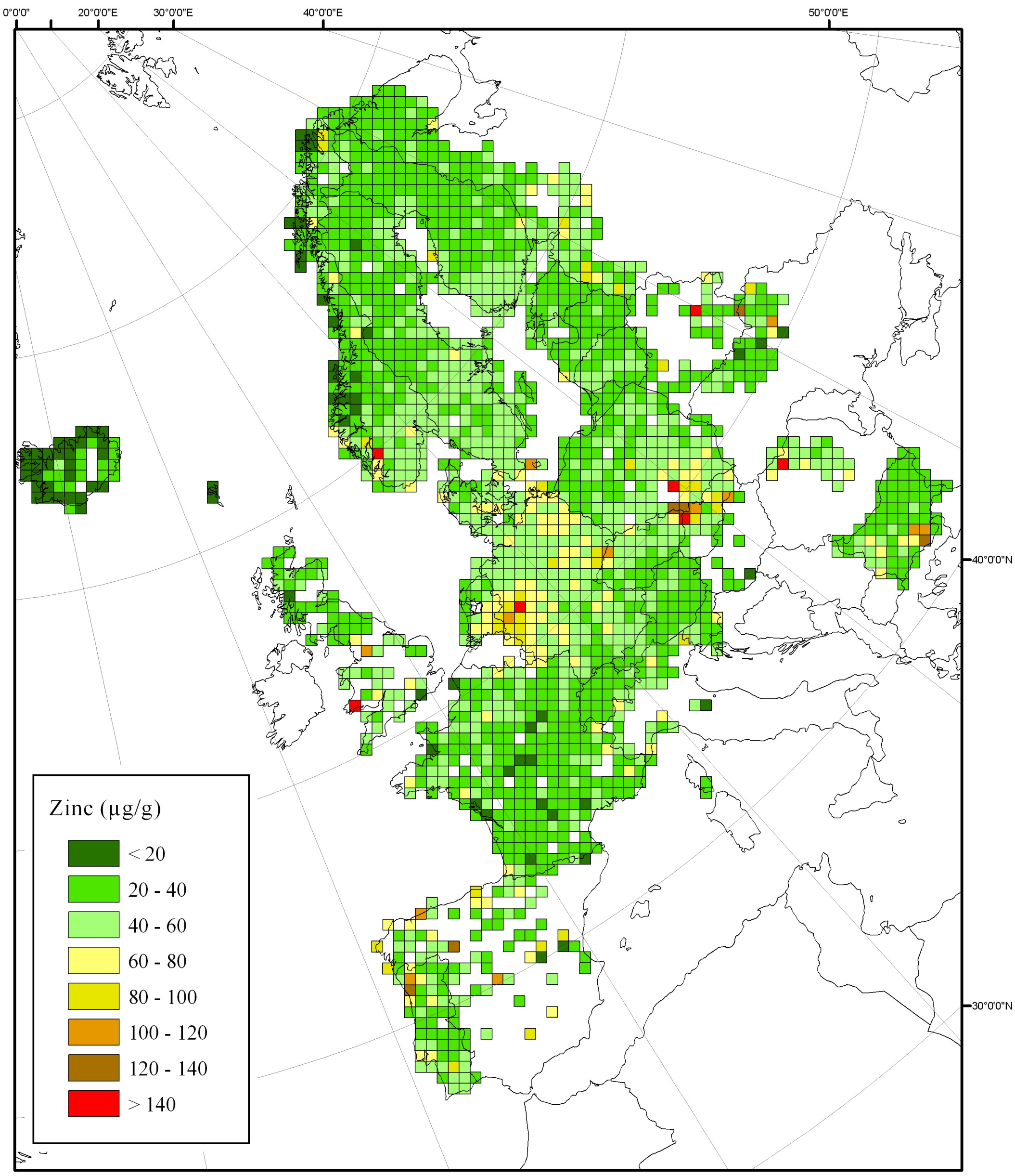
Zinc (µg/g)
< 20
20 - 40
40 - 60
60 - 80
80 - 100
100 - 120
120 - 140
> 140

1995



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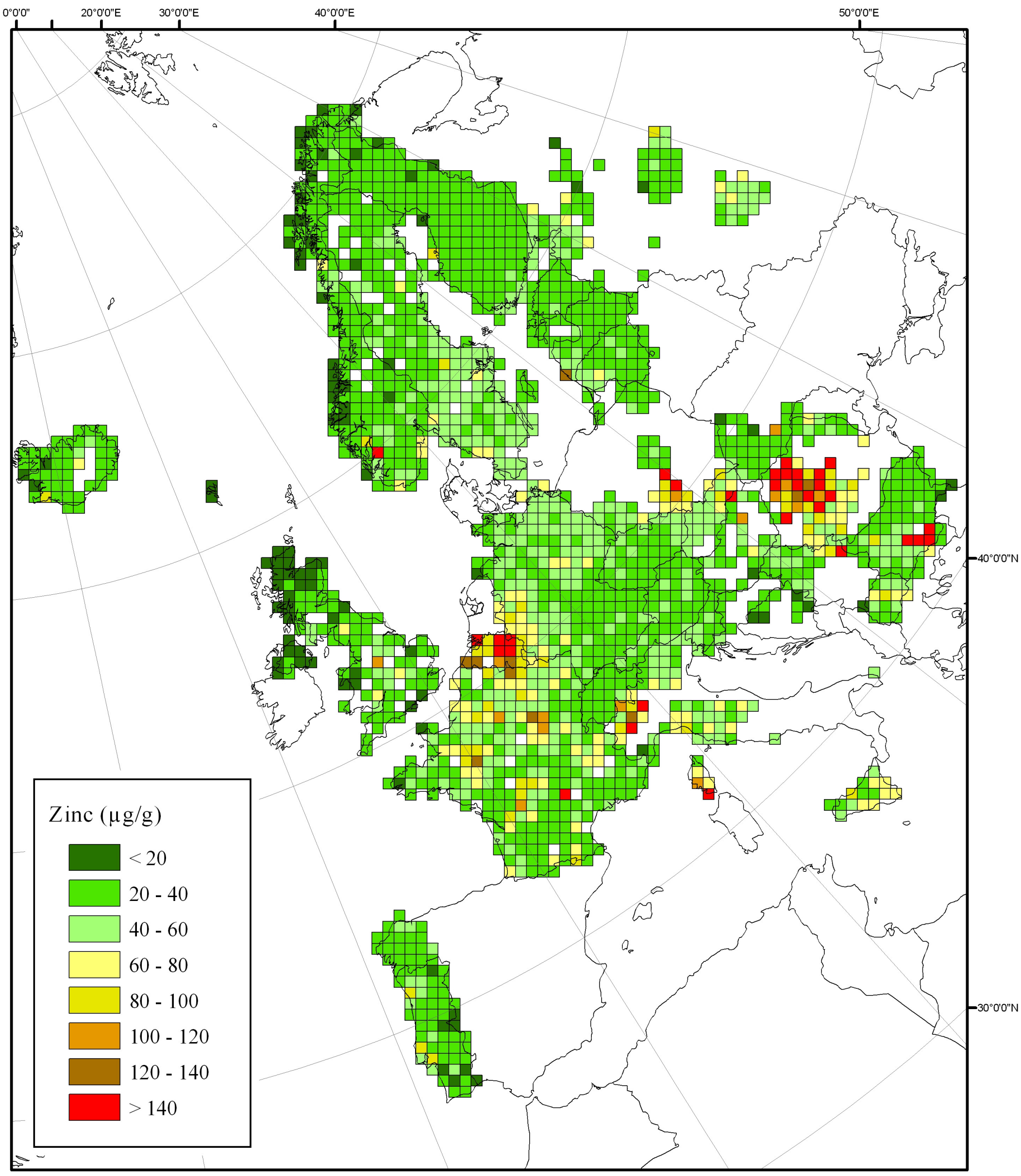
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Zinc (µg/g)
< 20
20 - 40
40 - 60
60 - 80
80 - 100
100 - 120
120 - 140
> 140

2000



0°0'0"
20°0'0"E
30°0'0"E
40°0'0"E
50°0'0"E
40°0'0"N
30°0'0"N
Zinc (µg/g)
< 20
20 - 40
40 - 60
60 - 80
80 - 100
100 - 120
120 - 140
> 140