

Additional file 1:

Table S1. Limits of quantification (LQ) for the elements determined.

	Al	As	Cu	Fe	Ni	Sb	V	Zn
LQ*	0.5	0.05	0.05	0.20	0.05	0.01	0.10	0.1

* in mg/kg (HM) and Mass-% (N)

Table S2. Statistical values for determinations of heavy metals in reference materials during the measurement period.

	References	Measurements		RR (%)
	MV±SD	MV±SD	RSD (%)	
As	0.04±0.004	0.040±0.001	2.2	101.0
Cu	0.04±0.004	0.041±0.001	2.2	101.3
Ni	0.04±0.004	0.041±0.001	2.4	101.8
Sb	0.04±0.004	0.041±0.001	1.8	101.4
Zn	0.04±0.004	0.041±0.001	2.4	101.4

MV±SD = mean value ± standard deviation (in mg/L, µg/L for Hg); RSD = relative standard deviation; RR = recovery rate, related to the reference

Table S3. Statistical values of the moss reference materials M2 and M3.

	M2	M2*		RR (%)	M3	M3*		RR (%)
	Reference	MV±SD	RSD (%)		Reference	MV±SD	RSD (%)	
Al	178±15	181±3	1.5	101.8	169±10	169±4	2.1	99.8
As	0.98±0.07	0.94±0.05	5.1	96.3	0.105±0.007	0.106±0.01	6.8	101.3
Cu	68.7±2.5	66.65±3.69	5.5	97.0	3.76±0.23	3.58±0.13	3.5	95.3
Fe	262±35	259±7	2.6	98.9	138±12	146±3	2.3	105.6
Ni	16.3±0.9	15.78±0.23	1.4	96.8	0.95±0.08	0.90±0.05	5.2	95.0
Sb	0.21±0.016	0.21±0.01	4.9	98.3	0.052±0.007	0.051±0.00	6.0	101.3
V	1.43±0.17	1.32±0.09	6.7	92.2	1.19±0.15	1.09±0.05	5.0	91.2
Zn	36.1±1.2	34.3±1.6	4.5	95.1	25.4±1.1	24.7±1.5	6.0	97.4

* = Measurements; MV±SD = mean value ± standard deviation (in mg/L, µg/L for Hg); RSD = relative standard deviation; RR = recovery rate, related to the reference

Table S4a. Geostatistical analysis 2015: Methods and parameters of the model

Element	n	Vk	Sk	Method	Transformati on	lamda	Model	Lag Size	No of Lags	Sector type	Max. neighbours	Range
Al	400	98.63	4.83	Ordinary	Box-Cox	-0.1	Exponential	15000	12	4 (45° Offset)	10	103
As	398	94.69	5.49	Ordinary	Box-Cox	-0.3	Exponential	15000	18	4 (45° Offset)	10	104
Cd	398	75.93	6.28	Ordinary	Box-Cox*	-0.5	Spherical	30000	12	4 (45° Offset)	10	223
Cr	399	68.07	3.17	Ordinary	Box-Cox	-0.1	Exponential	15000	12	4 (45° Offset)	10	104
Cu	400	39.07	1.24	Universal 2. Ord.	Box-Cox	-0.7	Spherical	15000	12	4 (45° Offset)	10	117
Fe	390*	76.60	5.60	Ordinary	Box-Cox	-0.5	Exponential	10000	8	4 (45° Offset)	10	58
Hg	397	46.01	3.33	Ordinary	Box-Cox	0.1	Spherical	10000	12	4 (45° Offset)	10	67
Ni	400	28.89	1.28	Ordinary	Box-Cox*	-0.9	Exponential	15000	12	4 (45° Offset)	10	180
Pb	400	77.96	4.42	Ordinary	Box-Cox	-0.2	Exponential	15000	12	4 (45° Offset)	10	202
Sb	400	82.00	3.56	Ordinary	Box-Cox*	-0.1	Spherical	15000	12	4 (45° Offset)	10	139
V	397	54.03	1.99	Ordinary	Box-Cox	-0.1	Exponential	15000	18	4 (45° Offset)	10	106
Zn	400	70.09	4.01	Universal 2. Order	Box-Cox	-0.1	Exponential	15000	12	4 (45° Offset)	10	166
N	400	37.22	1.20	Universal 2. Order	Log*	---	Spherical	15000	12	4 (45° Offset)	10	117

Table S4b. Geostatistical analysis 2015: Quality parameters of the model

Element	Nugget	Partial Sill	N/S	Moran's I	p-value	ME	MSE	RMSSE	MPE	SR	MPEc	r _p
Al	0.0829	0.0309	0.73	0.05	0	-12.23	-0.07	1.27	24.84	0.97	24.02	0.32
As	0.8570	0.2817	0.75	0.08	0	0	-0.04	1.07	20.00	1.03	20.67	0.32
Cd	1.1245	0.5851	0.66	0.02	0	0	-0.07	1.20	12.13	1.03	12.55	0.22
Cr	0.2355	0.1142	0.67	0.06	0	0	-0.01	0.96	21.43	0.95	20.38	0.35
Cu	0.0092	0.0041	0.69	0.19	0	0.04	-0.04	1.08	2.82	1.28	3.61	0.54
Fe	0.0007	0.0002	0.81	0.05	0.02	-3.69	-0.06	0.96	18.15	1.06	19.24	0.14
Hg	0.0538	0.0317	0.63	0.05	0	0	-0.02	1.37	5.37	0.97	5.21	0.33
Ni	0.2372	0.2372	0.50	0.19	0	0	0	1.08	15.24	1.1	16.71	0.38
Pb	0.1758	0.0738	0.70	0.09	0	-0.04	-0.04	1.24	23.27	1.03	23.90	0.28
Sb	0.2911	0.1925	0.60	0.01	0.01	0	-0.02	1.06	10.28	1.07	10.97	0.26
V	0.2534	0.0793	0.76	0.04	0	-0.01	-0.03	1.03	21.43	0.92	19.75	0.28
Zn	0.0362	0.0183	0.66	0.03	0	-0.28	-0.07	1.19	4.83	1.13	5.48	0.46
N	0.0376	0.0185	0.67	0.10	0	0	-0.03	0.97	2.75	1.07	2.96	0.57

n Sample size; *= outliers eliminated (SN1 NI110_104 NI118_125
NI01 NI63 BY86 BW805 BW241 BW803 BW240)

Vr Coefficient of variation [%]

Sk Skewness

Methode Kriging method

Transformation Transformation method (* = significant)

lambda Lambda value for the Box-Cox Transformation

Model Kriging variogram model

Lag Size Lag size in empirical semivariogram

No of Lags Number of lags

Range Rrange of spatial autocorrelation [km]

Nugget Nugget in the semivariogram

Partial Sill Partial Sill (Sill – Nugget)

N/S Nugget/Sill Ratio

Moran's I Moran's Autocorrelation Coefficient

p-value Probability of error

Sector type Sector type for the division of the search window

Max. neighbors Maximum number of samples in the search window

ME Mean Error) [µg/g. %]

MSE Mean Standardized Error

RMSSE Root-Mean-Square Standardized Error

MPE Median Percentage Error [%]

SR Range-Ratio: Ratio between range of cross validation error and range of the measurements

MPEc MPE * SR [%]

r_p Correlation coefficient (Pearson)

Figure S1. Spatial distribution of sample point-specific measured and geostatistically estimated concentrations of Al and As in moss

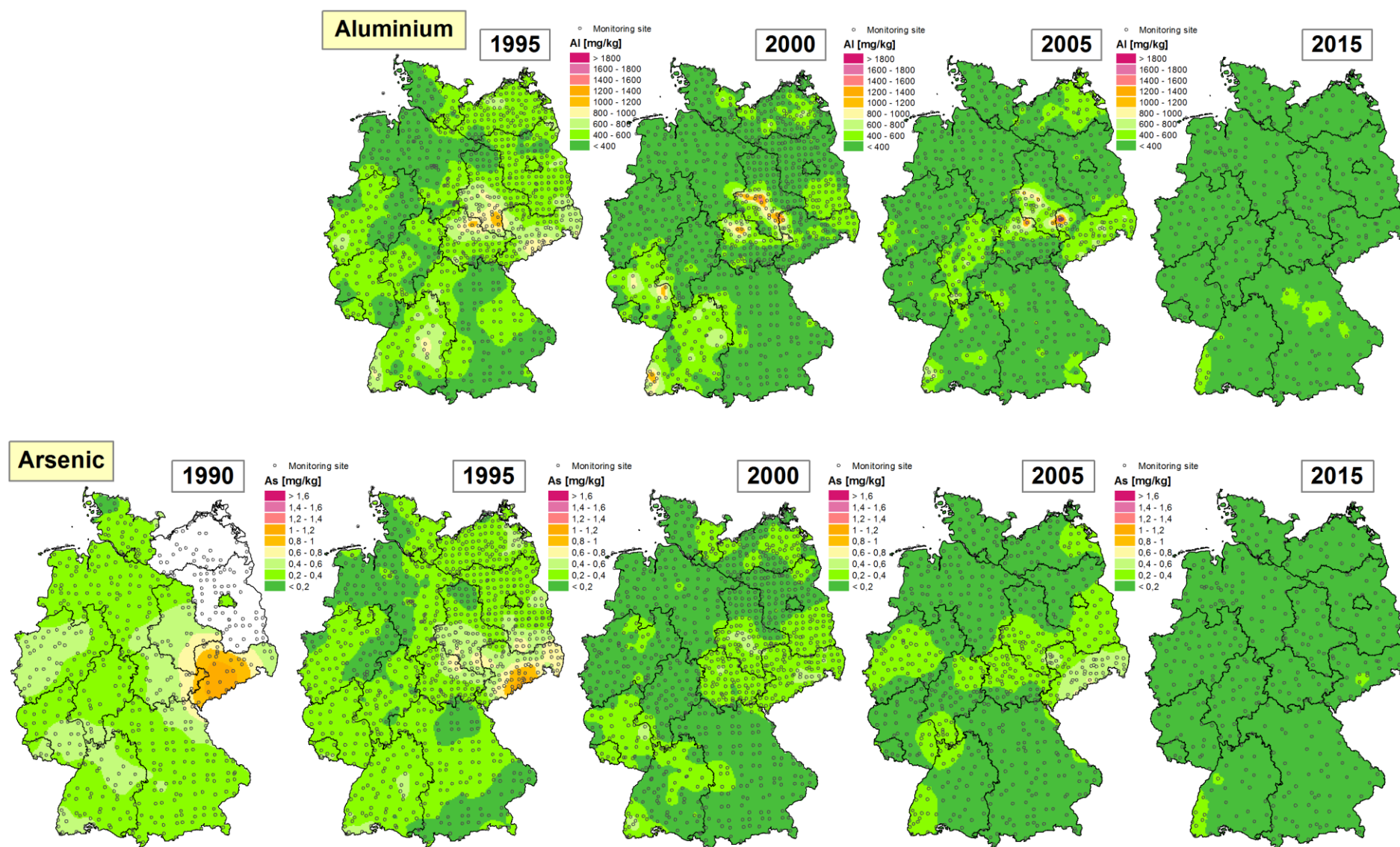


Figure S2. Spatial distribution of sample point-specific measured and geostatistically estimated concentrations of Cu and Fe in moss

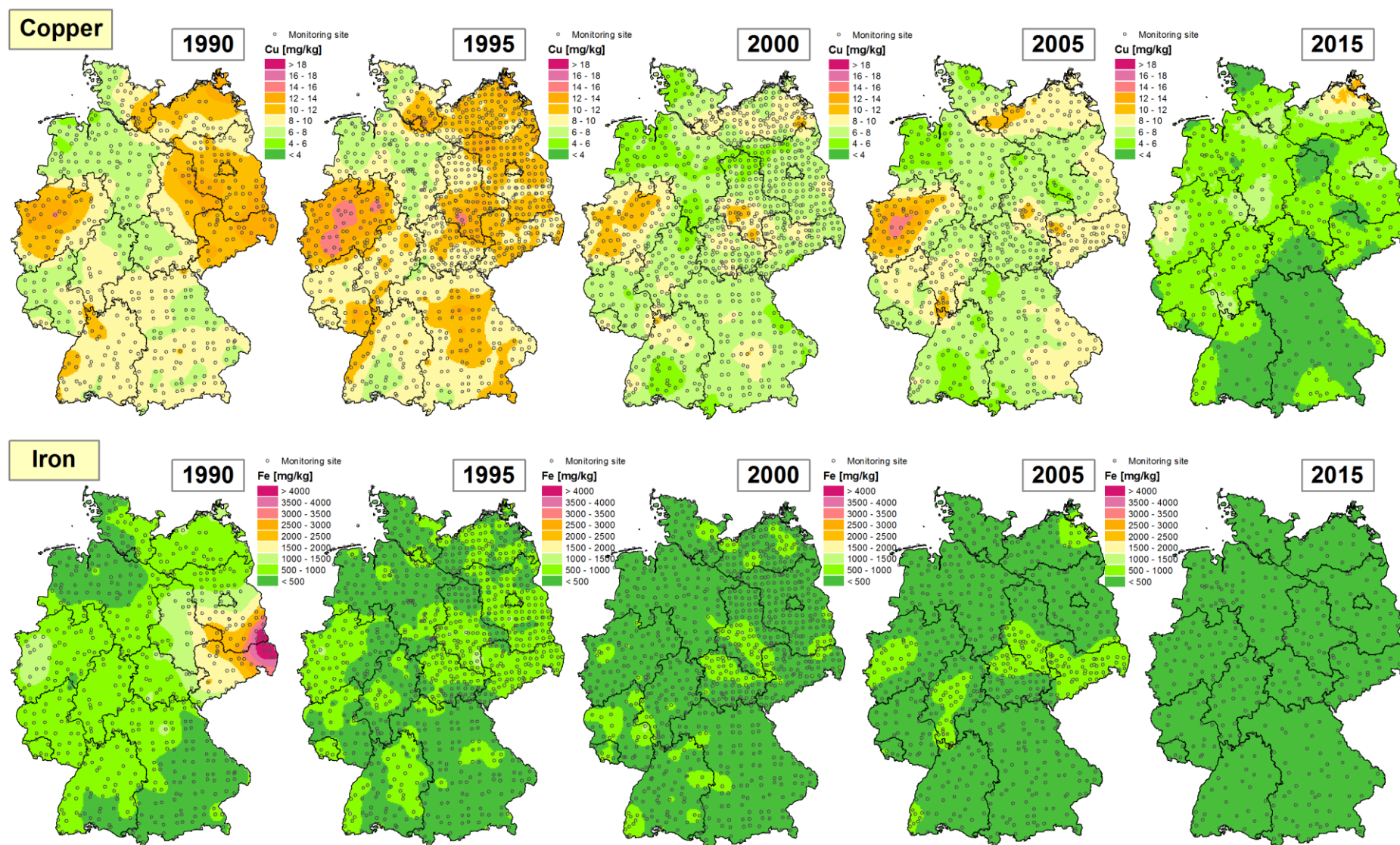


Figure S3. Spatial distribution of sample point-specific measured and geostatistically estimated concentrations of Ni and Sb in moss

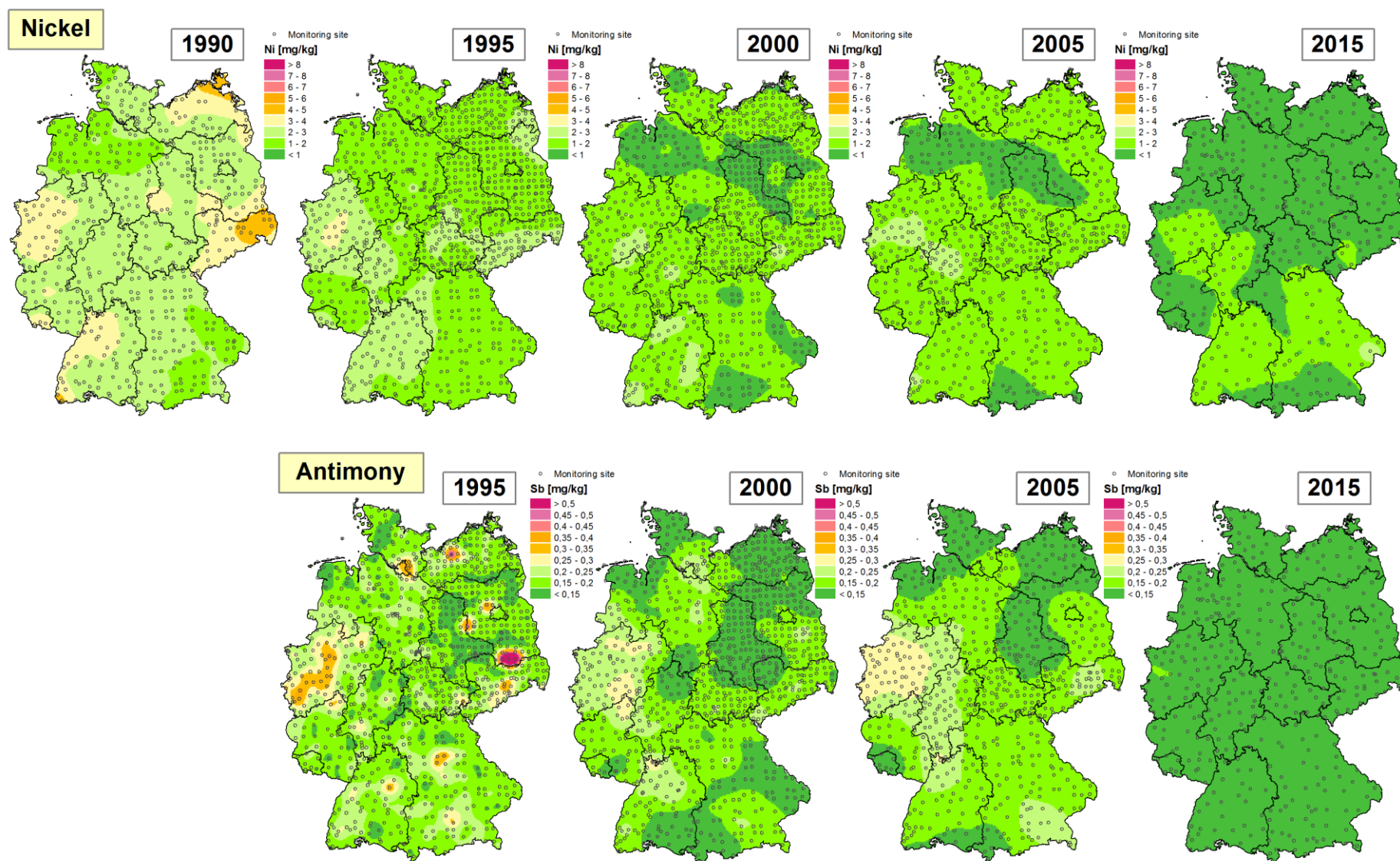


Figure S4. Spatial distribution of sample point-specific measured and geostatistically estimated concentrations of V and Zn in moss

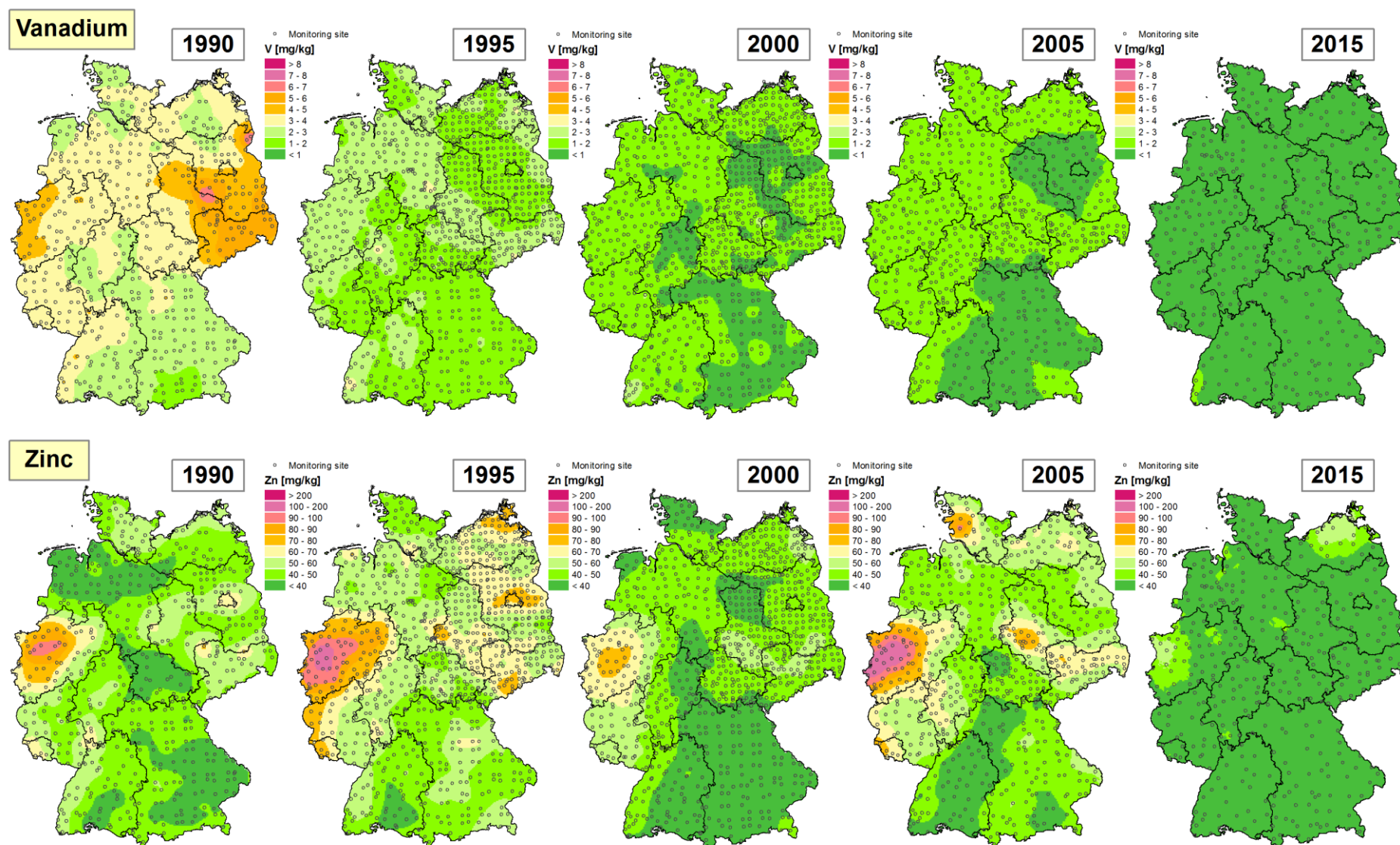


Table S5. Spatial distribution of sample point-specific measured and geostatistically estimated V and Zn concentrations in moss

Element	n	Min [µg/g]	Max [µg/g]	SD [µg/g]	RSD [%]	P20 [µg/g]	P50 [µg/g]	P90 [µg/g]	P98 [µg/g]
Al	39744	103.8	638.3	66.2	28.3	179.5	226.2	327.0	409.1
As	39744	0.0580	0.2970	0.0280	24.2100	0.0940	0.1080	0.1550	0.1890
Cd	39744	0.0786	0.2366	0.0292	21.0785	0.1140	0.1329	0.1760	0.2210
Cr	39744	0.249	1.575	0.155	24.188	0.519	0.610	0.844	1.032
Cu	39744	2.699	13.673	1.303	27.365	3.672	4.497	6.271	8.667
Fe	39744	134.8	454.4	38.2	17.6	185.1	213.3	269.4	311.5
Hg	39744	0.0167	0.0682	0.0069	19.9413	0.0281	0.0343	0.0436	0.0487
Ni	39744	0.382	2.790	0.303	35.235	0.602	0.795	1.234	1.636
Pb	39744	1.115	4.017	0.471	24.161	1.545	1.842	2.615	3.291
Sb	39744	0.0247	0.1523	0.0219	24.6875	0.0732	0.0881	0.1155	0.1342
V	39744	0.228	1.312	0.103	18.286	0.490	0.557	0.691	0.797
Zn	39744	14.2	59.64	7.12	23.16	24.7	30.42	38.72	50.4
	n	Min [%]	Max [%]	SD [%]	RSD [%]	P20 [%]	P50 [%]	P90 [%]	P98 [%]
N	39744	0.604	3.905	0.303	20.258	1.279	1.429	1.845	2.439

n = sample size; RSD = relative standard deviation; P = percentiles