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The Use of Heavy Metals Detected in the Soil as Air-Pollution Indicators on a Stemming Slope of the Achenkirch Area

By

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S u m m a r y

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To investigate forest soils as sinks for heavy metal input at the northern slope of the Schulterberg, Cu, Zn, Cr, Ni, Pb, and Cd were analysed on six sample plots (altitudinal difference of 800 m) also with a view to their suitability as air pollution indicators. Both the total heavy metal content and the mobile shares were compared with each other. Only in the case of Pb, which is otherwise little soluble, the correlation between the total content and the mobile shares proved to be extraordinarily narrow. The high share of mobile Pb indicates inputs from atmospheric far-distance transport; Pb therefore appears to be suited as air pollution indicator.

I n t r o d u c t i o n

In the course of the Austrian Forest Soil Monitoring System significant increases of the Pb and Cd concentrations in the soil were determined with increasing altitude above sea-level (MUTSCH 1992). Similar correlations were observed also by other authors (HEINRICHS & MAYER 1982, KAZDA & al. 1986, LINDEBNER 1990). However, not only altitude, but also weather-related exposure of sites (higher deposition rates) was essential in that connection (GLAVAC & al. 1987, KATZENSTEINER 1994, ZECHMEISTER 1994, 1995). The air-borne heavy metals which were finally deposited on the forest floor are apparently of anthropogenic origin. Especially on the north- or west-exposed stemming slopes of the northern

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alpine fringe considerable accumulations of those heavy metals should be detectable. The northern slope of the Schulterberg, which has an altitudinal range of approximately 800 m and no protecting foothills towards the northwest-European industrial zone, is therefore very well suited for the investigation of altitude-related heavy metal depositions; ZECHMEISTER 1994 therefore used the same altitudinal profile to investigate the suitability of mosses, and PEINTNER 1995 the same sample plots to investigate the suitability of fungi for the monitoring of atmospheric heavy metal depositions. The present paper aims at investigating the soil as a sink of heavy metal input.

Methods

Selection of sample plots and sampling

On the northern slope of the Schulterberg six sample plots of an altitudinal distance of between 100 and 200 m were sampled. On each sample plot three profile pits were prepared, with samples being collected from three of the faces of every profile pit and separately for the individual depth layers. Consequently, 9 samples were taken from every depth layer of the plots and combined to form mixed samples. The following depth layers of the mineral soil were used for the investigation: 0-5 cm, 5-10 cm, 10-20 cm, and 20-30 cm.

Analytics

For the analysis, air-dry soil (< 2 mm) was used. To determine the total heavy-metal content, a 2 g sample was decomposed with 16 ml of a mixture made up of 5 parts of nitric acid and 1 part of perchloric acid. For the analysis, the acid extract so achieved was filled up to 100 ml with double-deionized water.

To assess the anthropogenic input, not only the total content, or "weatherable" stock, was determined, but also the so-called "mobile" portion of heavy metals. At present, several procedures are being discussed; however, none of the methods that might be used has so far been internationally accepted, as there is no extractant which allows the selective determination of heavy metals deposited by man. For the present study, NH_4 -Acetate-EDTA was chosen as extractant as the latter had proven appropriate in a Swiss pilot study treating a similar question (ZIMMERMANN & BLASER 1993). The extracting agent is 0.5 molar in respect of ammonium acetate and 0.02 molar in respect of EDTA; its pH value was 4.65. LAKANEN & ERVIO 1971 proved that the elements extracted therewith form a mobile pool which is potentially available to the plants. The extracted amounts of elements are defined operationally, i.e. the extract will in no way have a chemically selective effect.

To achieve reproduceable and comparable results, the extraction must be carried out under well-defined conditions. It is important for the interpretation to know the characteristic features of the extract:

- It acts as a cation exchange agent with NH_4 as exchange-effective ion.
- EDTA, which is highly complexing, extracts more strongly fixed heavy metals from various chemical compounds.
- It acts as a weak acid, as it contains acetic acid as a buffering agent.

The latter property is of importance especially in calcareous soils. Because of its acidity, the extract dissolves part of the calcareous rock with the size of that portion depending on extraction time and temperature, which must therefore strictly be adhered to. However, for the extraction of most trace elements that effect can be neglected because the amount of trace elements of calcareous rock is mostly very low.

For the analysis, 5 g fine soil are added to 50 ml extracting solvent and shook for one hour on the horizontal shaker at room temperature. The heavy metal concentrations of the two extracts were determined on a sequential plasma emission spectrometer (Varian Liberty 200).

Results and Discussion

The following heavy metals were analyzed in connection with potential heavy metal inputs: Cu, Zn, Cr, Ni, Pb, and Cd. For the interpretation of the results, one must take into account on the one hand the different ecological classification of the individual heavy metals (Cu and Zn are important trace elements, while only toxic effects are known about Pb and Cd), on the other the different characteristics of those heavy metals in respect of their relevance for pollution have to be taken into account: These are on the one hand the quantities of anthropogenic emissions, on the other the suitability of the emitted heavy metal compounds for far-distance transport in the atmosphere. According to LANTZKY & MCKENZIE 1979 Pb is by far most important in respect of those aspects, followed by Zn, Cd, and Cu. Nearly insignificant are atmospheric transports of Cr and Ni. In the following, special importance will therefore be attached to the role of Pb as pollution indicator.

The total Cu, Zn, Cr, and Ni concentrations are well in accordance with the results from carbonate-influenced soils which MUTSCH 1992 found in the framework of the Austrian Forest Soil Monitoring System: The Cu content shows almost no depth gradient for the profile; for Zn, a significant decrease with soil depth can be seen; Cr concentrations are markedly lower in the surface layer than in the mineral soil. The same holds true for Ni the concentrations of which in the mineral soil increase with increasing soil depth. The concentrations of those four heavy metals did not seem to depend on altitude on the northern slope of the Schulterberg as in the course of the Forest Soil Monitoring System.

The decrease of the total Pb and Cd concentrations with soil depth, which was observed in most of the cases studied in connection with the Forest Soil Monitoring System, was not found on the six sample plots of the Schulterberg. An increase of the total Pb and Cd concentrations with altitude was not observed, either, although MUTSCH 1992 had described it for the Austrian forest floors and ZECHMEISTER 1994 for Pb in mosses even for the same investigation area on the Schulterberg. If we calculate the mean Pb concentration for all depth layers of the mineral soil of the individual sample plots, the result is a distribution independent of altitude (Table 1).

The difference between lowest (83 µg/g) and highest (118 µg/g) total content is not big. A picture similar to the total content of the acid extract is given by the mobile portion extractable with the NH₄-Ac-EDTA extract, although the difference between the lowest and the highest content (1.62 µg/g and 5.90 µg/g, resp.) is relatively more significant. The highest Pb concentrations were found on the lowest-located sample plot, which is contrary to the hypothesis that heavy metal pollution should increase with increasing altitude.

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Table 1. Mean Pb concentrations ($\mu\text{g/g}$) including all depth layers (0-30 cm) of the mineral soil of the individual sample plots located at the northern slope of the Schulterberg.

Altitude a.s.l. of the sample plots	Acid extract	NH ₄ -Ac-EDTA-extract
	$\bar{x}$ Pb (ppm)	$\bar{x}$ Pb (ppm)
N0: 1650 m	101	2.01
N1: 1505 m	101	4.87
N2: 1350 m	83	1.62
N3: 1145 m	116	1.86
N4: 955 m	94	3.11
N5: 840 m	118	5.90

A mobile heavy metal portion was determined in addition to the total content, as far-distance air transport of heavy metals outside their original mineral associations requires small and easily soluble molecular compounds (aerosols of a certain maximum size), which are finally deposited on the ground. There they increase the pool of the portion of easily soluble elements caused by weathering and biogenic processes (ZIMMERMANN & BLASER 1993). The bigger the share of mobile fractions in the total content, the narrower is therefore the proportion total content/mobile portion; the bigger will therefore the portion of pollution-related deposition and accumulation be.

Which part of the mobile fraction is due to pollution-related depositions and which part is due to biogenic accumulation or weathering processes depends on the respective element and the parent rock of the respective site. Some basic features of the discussed heavy metals are useful when assessing such differentiation, among them the binding strength of metal ions (Table 2) and the transfer coefficient from the soil to the plant (Table 3).

Table 2. Relative binding strength of metal ions depending on parts of the soil upon given limit-pH (acc. to DVWK 1988).

The binding strength increases from 1 to 5. Above the limit-pH a heavy immobilization occurs through the formation of oxides or hydroxo complexes.

Element ordered acc. to increasing binding strength	Limit pH	Substrate-related binding strength below limit pH		
		Humus	Clay	Sesquioxides
Cd	6.0	4	2	3
Zn	5.5	2	3	3
Ni	5.5	3.5	2	3
Cu	4.5	5	3	4
Cr (III)	4.5	5	4	5
Pb	4.0	5	4	5

Table 3. Soil-plant transfer coefficient of several heavy metals (acc. to KLOKE & al. 1984). Values > 1 indicate accumulations in the plant.

Element	Transfer coefficient
Cd	1.00 - 10.00
Zn	1.00 - 10.00
Ni	0.10 - 1.00
Cu	0.10 - 1.00
Cr	0.01 - 0.10
Pb	0.01 - 0.10

In Table 4 not only the total heavy metal content of the acid extract and the mobile portions of the $\text{NH}_4\text{-Ac-EDTA}$ extract, but also the proportions of the concentrations in respect of the different methods of extraction are given. Those quotients depend partly on the depth layer; partly they vary greatly, but within limits that are characteristic of the individual elements. In Fig. 1 the quotients are shown separately for the individual elements, with mean values including all six sample plots and calculated for the respective depth layers.

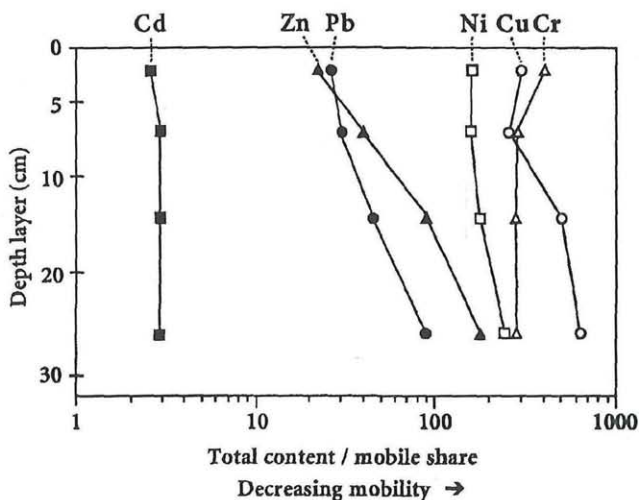


Fig. 1. Quotient total content/mobile share of heavy metal on the northern Schulterberg slope with regard to depth layers.

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Table 4. Heavy Metal Concentrations in the Acid Extract (AE) and in the NH₄ - Acetate-EDTA extract (mob) of the Schulterberg sample plots in µg/g, and proportion of the two methods of extraction (AE/mob).

		Cu			Zn			Cr		
		AE	mob	AE/mob	AE	mob	AE/mob	AE	mob	AE/mob
N0:	1650 m									
	Surface layer	13	0.08	163	121	12.78	9.5	29	0.06	483
	0- 5 cm	15	0.03	500	119	4.72	25.2	42	0.09	467
	5-10 cm	14	0.06	233	115	4.13	27.8	40	0.12	333
	10-20 cm	12	0.03	400	92	1.70	54.1	43	0.12	358
	20-30 cm	12	0.02	600	66	0.60	110.0	42	0.16	262
N1:	1505 m									
	Surface layer	15	0.09	167	134	15.21	8.8	27	0.30	90
	0- 5 cm	11	0.07	157	140	7.29	19.2	36	0.23	157
	5-10 cm	13	0.05	260	138	5.17	26.5	40	0.23	174
	10-20 cm	13	0.03	433	118	1.62	73.8	47	0.31	152
	20-30 cm	11	0.02	550	81	0.51	159.0	47	0.30	157
N2:	1350 m									
	Surface layer	17	0.06	283	139	9.93	14.0	24	0.23	104
	0- 5 cm	18	0.05	360	165	6.19	26.7	37	0.06	617
	5-10 cm	16	0.05	320	176	2.97	59.3	46	0.15	307
	10-20 cm	17	0.05	340	178	1.64	108.0	56	0.21	267
	20-30 cm	17	0.03	567	166	0.82	202.0	54	0.26	208
N3:	1145 m									
	Surface layer	9	0.07	129	114	16.13	7.1	7	0.02	350
	0- 5 cm	16	0.05	320	160	10.02	15.9	39	0.06	650
	5-10 cm	16	0.06	267	109	2.16	50.5	46	0.11	418
	10-20 cm	17	0.06	283	110	1.15	95.7	55	0.13	423
	20-30 cm	18	0.03	600	112	0.32	350.0	62	0.13	477
N4:	955 m									
	Surface layer	13	0.10	130	94	15.86	5.9	20	0.03	667
	0- 5 cm	15	0.05	300	118	5.08	23.2	53	0.14	379
	5-10 cm	15	0.04	375	115	2.09	55.0	60	0.19	316
	10-20 cm	12	0.01	1200	63	0.44	143.0	41	0.23	178
	20-30 cm	11	0.01	1100	38	0.20	190.0	34	0.14	243
N5:	840 m									
	Surface layer	10	0.08	125	90	16.35	5.5	11	0.02	550
	0- 5 cm	10	0.05	200	93	6.08	15.3	36	0.08	450
	5-10 cm	7	0.04	175	86	4.99	17.2	43	0.21	205
	10-20 cm	11	0.03	367	77	0.91	84.6	51	0.23	222
	20-30 cm	9	0.02	450	69	0.76	90.8	53	0.21	252

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		Ni			Pb			Cd		
		AE	mob	AE/mob	AE	mob	AE/mob	AE	mob	AE/mob
N0:	1650 m									
	Surface layer	18	0.17	106	105	7.58	13.9	2.90	0.93	3.1
	0- 5 cm	26	0.26	100	120	3.64	33.9	2.79	0.94	3.0
	5-10 cm	32	0.22	145	105	2.60	40.4	3.01	1.00	3.0
	10-20 cm	37	0.31	119	101	2.00	50.5	2.66	0.96	2.8
	20-30 cm	45	0.19	237	89	0.90	98.9	1.29	0.81	1.6
N1:	1505 m									
	Surface layer	30	0.40	75	127	11.37	11.1	2.51	0.77	3.3
	0- 5 cm	60	0.72	83	118	8.16	14.5	2.17	0.90	2.4
	5-10 cm	65	0.44	148	153	10.11	15.1	2.35	0.87	2.7
	10-20 cm	82	0.60	137	100	4.16	24.0	1.60	0.63	2.5
	20-30 cm	82	0.66	124	68	1.31	51.9	2.20	0.77	2.9
N2:	1350 m									
	Surface layer	25	0.13	192	61	2.43	25.1	2.50	0.71	3.5
	0- 5 cm	40	0.30	133	111	2.73	40.7	3.01	0.91	3.3
	5-10 cm	57	0.42	136	91	2.27	40.1	3.12	0.85	3.7
	10-20 cm	65	0.54	120	85	1.47	57.8	2.76	0.70	3.9
	20-30 cm	71	0.46	154	63	0.90	70.0	2.53	0.61	4.1
N3:	1145 m									
	Surface layer	7	0.13	414	46	5.70	8.1	0.91	0.30	3.0
	0- 5 cm	36	0.23	157	111	2.96	37.5	2.34	0.61	3.8
	5-10 cm	42	0.34	124	111	3.03	36.6	2.64	0.75	3.4
	10-20 cm	53	0.30	177	129	1.42	90.8	2.73	0.70	3.9
	20-30 cm	62	0.29	214	108	1.16	93.1	2.48	0.66	3.8
N4:	955 m									
	Surface layer	6	0.08	75	88	8.34	10.6	1.28	0.53	2.4
	0- 5 cm	35	0.15	233	109	8.01	13.6	1.80	0.85	2.1
	5-10 cm	34	0.23	148	110	5.02	21.9	1.98	0.91	2.2
	10-20 cm	48	0.15	320	90	2.36	38.1	1.04	0.68	1.5
	20-30 cm	49	0.19	258	83	0.44	189.0	0.60	0.38	1.6
N5:	840 m									
	Surface layer	9	0.05	180	71	6.68	10.6	1.03	0.45	2.3
	0- 5 cm	34	0.27	126	116	10.19	11.4	2.68	0.95	2.8
	5-10 cm	40	0.30	133	121	8.72	13.8	2.81	0.98	2.9
	10-20 cm	43	0.23	187	124	4.11	30.2	2.24	0.70	3.2
	20-30 cm	45	0.13	346	110	4.12	26.7	2.20	0.68	3.2

If we follow the increasing binding strength of metal ions at given limit-pH according to Table 2, which describes approximately the mobility of the individual heavy metals, the order of the latter is, apart from one single exception (Pb), in accordance with the representation of Fig. 1: Cd is followed by Zn, Ni, Cu, Cr, and Pb; as regards the quotient, however, Pb takes the second place and therefore shows a much greater mobility (lower binding strength) than was given in Table 2.

Several elements can be taken up through the roots of plants, mobilized biogenically, and finally accumulated in the soil in an easily available form. Again, the order of the elements, according to decreasing transfer coefficient (Table 3),

corresponds exactly to the increasing binding strength of the metal ions. The very low transfer coefficient of Pb does not allow biogenic mobilization or offer an explanation of its narrow proportion of total content and mobile portion.

All the soils of the Schulterberg studied in the present paper have pH values (in CaCl_2 suspension) between 4.5 and 7 (MUTSCH 1995), i.s. ranges which according to Table 2 exclude measurable mobility with certainty for Pb only and which therefore also do not provide an explanation for that element's narrow proportion of total content and mobile portion.

For Cd, the very narrow proportion of total content and mobile portion is on the one hand explicable by the very high limit-pH (Table 2), on the other by the transfer coefficient of Cd, which is high despite the toxicity of Cd (Table 3).

According to Table 2 Zn and Ni are similar in binding strength, but they show a marked difference in respect of the soil/plant transfer coefficient, which contributes to the fact that the proportion of total content and mobile portion of the two elements is narrow only in the subsoil: Because of its biogenic accumulation capacity mobile Zn is accumulated in the topsoil through litter. Cu and Cr have similarly high binding strengths; those of Cr are even a little higher than those of Cu. Nevertheless in the case of Cu the quotient total content/ mobile portion is narrow only in the uppermost 10 cm, which is probably due to the stronger biogenic accumulation capacity of Cu as compared to Cr.

Classified according to its binding strength and transfer coefficient, the proportions of total content and mobile portion of Pb would clearly have to be wider than those of Cu or Cr, proportions which should be between 10 and 20 times wider than the results found. A plausible explanation for the narrow proportion total content/mobile portion of Pb may be the air-borne pollution inputs (far-distance transport in the form of aerosols); Pb is deposited on the ground in the form of micro-grained, mobile compounds and there clearly increases the fraction of mobile Pb. It takes some time until it is transferred into less mobile forms of compounds.

PEINTNER 1995, too, concludes a strong (atmogenic) Pb load: As compared to other investigation areas the Pb concentrations found in the fruiting bodies of fungi on the Schulterberg are markedly increased, despite their low absolute values in comparison to other heavy metals.

The high share of mobile Cd and partly also of mobile Zn might to different degrees also be due to anthropogenic inputs.

C o n c l u s i o n s

First, there is still the question why air pollution could be detected for Pb via the extraordinarily narrow proportion of total content and mobile fraction, while the altitudinal gradient of Pb deposition described in the frame of the Forest Soil Monitoring System was not found at the northern slope of the Schulterberg. That may be due to the following reasons:

As there are no foothills, the entire northern slope of the Schulterberg is relatively regularly affected by weather incidents and thus is regularly affected by air pollution. For the many sample plots of the Forest Soil Monitoring System, however, increasing altitude usually means also (significantly) increasing weather-related exposure and, consequently, increasing deposition stress. Moreover, the particularly steep slope of the Schulterberg with its partly very thick humus layers enables a strong runoff and therefore a shifting of air pollution inputs towards the foot of the slope. In fact, the highest Pb concentrations (both the total concentrations and the concentrations of the mobile fraction) are to be found on the lowest sample plot, which is located in the plain already (Table 1).

The concentrations of the toxic heavy metals Pb and Cd found in the soils of the northern Schulterberg slope are without relevance to the biosphere of forests. What makes the results important is rather that they prove the existing far-distance transport of pollutants. In the course of such transport not only Pb (and Cd) but doubtlessly also many other noxious components, such as acids and organic pollutants from the Central and North-West European areas, are deposited in great amounts at the exposed high-altitude sites of the Northern Tyrolean Limestone Alps. The Alps as a weather divide seem to act as a sink of air pollutants.

From the analytical and methodical points of view the assessment of heavy metal inputs of soils by means of two different methods of extraction ([approximate] total content, mobile fraction), followed by the calculation of the quotient, has been successful. The statements concluded from the results correspond well to other results (ZECHMEISTER 1994, 1995, PEINTNER 1995). The extract according to LAKANEN & ERVIÖ 1971, which was chosen from a great number of possible methods, proved to be a simple, robust method to determine the mobile fraction; it was well suited for the application of modern analytical appliances.

An exact quantification of the pollution input is not possible with the methods described. If samples are collected also from deeper horizons (> 50 cm), maybe even from the parent rock, it is highly probable that at least an approximate determination of the deposited quantities is possible (ZIMMERMANN & BLASER 1993). An exact quantification can be achieved by using the isotopic separation analysis, which in the future might even help to answer the question of the origin of certain pollutants (KLÖTZLI 1995).

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