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Element budgets of pine stands on lignite and pyrite containing mine soils

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Abstract

To analyze the development of pine ecosystems on lignite and pyrite containing mine soils, four pine stands with ages of 3–35 years were investigated in a chronosequence approach. Bulk precipitation, throughfall and soil solution in depths of 20, 40, 70 and 100 cm were studied over a three-year period to determine element fluxes in these forest ecosystems on extreme acidic and saline soils.

Element budgets are controlled by the processes induced by pyrite oxidation such as intensive weathering of primary minerals, precipitation and leaching of secondary phases.

Despite low water fluxes, element outputs can reach extraordinary high values due to very high concentrations in the soil solution. Although element outputs decrease drastically with stand age, respectively, site age, they exceed those of comparable pine stands on non-mine sites. Nitrogen release from the lignite fraction represents a special characteristic of the examined substrates. Nitrogen losses beneath the rooted zone can be $34 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 100 cm depth. Element input and output in the examined ecosystems are far from balance. Closed cycling of nutrients seems to be recognizable in the case of potassium.

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1. Introduction

In eastern Germany, an area of about 770 km^2 has been subjected to opencast lignite mining. Due to the mean-surface lithologies and the mining technology used, about 60% of the post-lignite mining landscape is characterized by sandy substrates containing various amounts of pyrite and lignite (Katzur, 1995). Oxidation of pyrite in the tertiary substrates releases

large amounts of protons causing enhanced mineral weathering.

Revegetation of these acidic and saline substrates requires substantial efforts to improve the soil reaction and to correct nutrient deficiencies. Besides high amounts of lime, alkaline ashes from lignite-fired power plants were incorporated to the substrates and mineral fertilizer was applied usually (Katzur and Haubold-Rosar, 1996).

Most of the mine sites are afforested with pine species. These pine stands show comparable yield and growth dynamics to those on non-mine sites in the region (Böcker et al., 1998; Katzur et al., 2000).

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Table 1

Characterization of the chronosequence sites

	Weissagker Berg (WB)	Baerenbrueck (BB)	Meuro (MR)	Domsdorf (DD)
Year of dumping	1991	1977	1970	1946
Soil amelioration	1996	1978	1971	1963
	28 t CaO ha ⁻¹	190 t CaO ha ⁻¹	160 t CaO ha ⁻¹	50 t CaO ha ⁻¹
Substrate	depth: 60–80 cm	depth: 40 cm	depth: 60 cm	depth: 30 cm
	Lignite containing, pyritic sand	Lignite containing, pyritic loamy sand	Lignite containing, pyritic sand	Lignite containing, pyritic loamy sand
C (%)	0.5–3.0	4.0–7.0	2.0–5.0	2.0–7.0
S (%)	0.2–0.5	0.2–2.0	0.1–0.4	0.2–1.4
Vegetation	<i>Pinus sylvestris</i>	<i>Pinus nigra</i>	<i>Pinus sylvestris</i>	<i>Pinus sylvestris</i>
Stand density (stems ha ⁻¹)	12800	7375	8944	3409
Stand age in 1999 (years)	3	17	21	35

pyrite containing mine soils are highly influenced by the pedochemical processes induced by pyrite oxidation. High concentrations in the soil solutions, high element fluxes and low water fluxes over several decades point to unfavorable conditions for forest stands.

The long-term development of soils and ecosystems under these conditions is poorly known. Due to the extreme pedochemical situation, analogies to forest ecosystems on non-mine sites do not exist. It is furthermore difficult to predict which equilibrium state these systems are approaching.

By means of a chronosequence study, temporal developments of water and element budgets were investigated. This allowed getting insight into the dynamic of pedogenic processes interacting with forest ecosystems on lignite and pyrite containing mine soils.

2. Materials and methods

Four pine stands with stand ages from 3–35 years and comparable properties concerning substrate and soil amelioration were chosen as investigation sites (Table 1). The substrates were of sandy texture with clay content from 2–10%. pH of the non-ameliorated subsoil was in a range of 2.5–2.9, whereas in the topsoil pH was elevated to 3.5–4.6. The CEC varied in a range of 5–23 cmol_c kg⁻¹ with the higher values in the topsoil and lower values in the subsoil. All sites were ameliorated with alkaline ashes before afforestation. Amounts of given ash depended on the

calculation of the potential acid production that was developed by Illner and Katzur (1964) as a useful instrument for rehabilitation practice in the region. The sites received mineral fertilization at the same time when the alkaline ash was incorporated. At the WB site fertilization was about 120 N, 100 P, 80 K (kg ha⁻¹) and clearly influenced the soil solution, especially in the first months of the study (Wilden, 2000).

Although there had been great efforts in the selection of the investigation sites, the four pine stands do not only differ in stand age, while all other site specifications are the same with view to the initial state of development as theoretically required for chronosequence studies. They vary especially in the amount of the received ash. Still there are unavoidable concessions to the comparability of the site properties; the results give clear tendencies in the development of element budgets of pine stands on lignite and pyrite containing mine sites.

Precipitation and throughfall were collected at each site with 10 bulk samplers (opening area: 104 cm²) from January 1996 to December 1998 fortnightly. At the WB site, the investigation period was May 1996 to May 1998. There, throughfall could not be measured because of the low stand height. Meteorological data (air temperature, humidity, wind speed, precipitation, and global radiation) were recorded by weather stations at cleared areas close to the investigation sites. To determine the temporal distribution of precipitation and throughfall, recording samplers were installed at the cleared areas and within the investigation stands.

Table 2

Used solubility products and constants as found in the PHREEQC1.6 program internal Dataset Wateq4f.dat

Reaction	Log ₁₀ K
CaSO ₄ : 2 H ₂ O = Ca ²⁺ + SO ₄ ²⁻ + 2H ₂ O	-4.58
Al ₂ Si ₂ O ₅ (OH) ₄ + 6 H ⁺ = 2Al ³⁺ + 2H ₄ SiO ₄ + H ₂ O	7.435
KAl ₃ (SO ₄) ₂ (OH) ₆ + 6 H ⁺ = K ⁺ + 3Al ³⁺ + 2SO ₄ ²⁻ + 6H ₂ O	-1.4
Al(OH) ₃ + 3H ⁺ = Al ³⁺ + 3H ₂ O	8.11
FeOOH + 3 H ⁺ = Fe ³⁺ + 2H ₂ O	-1.0
Fe(OH) ₃ + 3H ⁺ = Fe ³⁺ + 3H ₂ O	4.891
KFe ₃ (SO ₄) ₂ (OH) ₆ + 6H ⁺ = K ⁺ + 3Fe ³⁺ + 2SO ₄ ²⁻ + 6H ₂ O	-9.21
AlOH ₄ ⁻ + H ⁺ = Al ³⁺ + SO ₄ ²⁻ + H ₂ O	-3.230
Al ₄ (OH) ₁₀ SO ₄ + 10H ⁺ = 4Al ³⁺ + SO ₄ ²⁻ + 10H ₂ O	22.7

Soil solution was collected using suction cups (Type P 80, UP GmbH) at 20, 40, 70 and 100 cm depths at the sites BB, MR, and DD, and at 20, 60 and 130 cm depths at WB site. Three mixed samples were collected per depth fortnightly.

Electrical conductivity (EC) and pH of precipitation, throughfall and soil solution were determined immediately after sampling. Filtered samples (0.45 µm; nylon) were stored at +4°C until analysis. Samples were not frozen because it would induce precipitation of highly concentrated soil solutions. Main cations were analyzed by AAS (Unicam 701) and ICP-AES (Unicam 939). Anions were measured by IC (Dionex 500 and 120), while dissolved organic carbon (DOC) was analyzed by Shimadzu TOC 5000. Due to the formation of ion pairs and complexes in highly concentrated solutions, ion species were difficult to determine. Therefore, elements in solutions are given as total concentrations if not mentioned otherwise. The median was chosen to show element concentrations because some of the data was not normally distributed.

For the calculation of saturation indices (SIs) of mineral phases and species repartition in soil solution, the program PHREEQC1.6 was used (Parkhurst, 1995). The solubility products and constants (Table 2) were taken from a program internal Dataset (Wateq4f.dat).

The sites were instrumented at 20, 60, 100/130 cm soil depths with pressure transducer tensiometers (5 per depth), TDR-Probes and soil temperature sensors. At the MR and DD site, xylem sap flow for ten average trees was measured using the method by Granier (1985).

To quantify the water fluxes of the chrono-sequence sites the model SOIL (Jansson, 1991) was used. It is a process-based one-dimensional hydrological model that simulates water and heat flow in a layered soil profile. Meteorological data are required as input variables. The model was calibrated by time series of measured soil matrix potential. Details on the simulation of water fluxes are given by Scherzer (2001).

To calculate element fluxes, simulated water fluxes were multiplied with measured soil solution concentrations for each sampling period.

3. Results

3.1. Water budget

The average bulk precipitation at the sites ranged from 562 to 797 mm yr⁻¹ during the period January 1996–December 1998. Deep percolation ranged widely from 51 to 227 mm yr⁻¹ depending on the age and density of the stands. The recently afforested site WB shows highest deep percolation with 31% (227 mm yr⁻¹) of bulk precipitation whereas at BB (17 years) and MR (21 years) deep percolation decreases to 6–13% (51–92 mm yr⁻¹) of bulk precipitation. This decrease relates to the high stand densities causing high interception and evapotranspiration losses. Under the oldest stand DD (35 years) deep percolation is about 21% (120 mm yr⁻¹) of bulk precipitation which can be attributed to the low tree density on this site.

Table 3

Sample volume (*n*) median of pH, EC and element concentrations in soil solutions for the period January 1996–December 1998

Stand (age)	<i>n</i>	pH	EC (mS cm ⁻¹)	Al (mg l ⁻¹)	Fe (mg l ⁻¹)	Ca (mg l ⁻¹)	Mg (mg l ⁻¹)	K (mg l ⁻¹)	Na (mg l ⁻¹)	Si (mg l ⁻¹)	NH ₄ (mg l ⁻¹)	NO ₃ (mg l ⁻¹)	Cl (mg l ⁻¹)	SO ₄ (mg l ⁻¹)
WB (3 yr) ^a														
20 cm	30	2.8	8.1	954	296	471	601	2.2	13.2	61	5.1	5.8	36	9065
60 cm	30	2.5	11.3	1083	1466	469	765	2.3	12.0	87	6.3	7.2	17	12 954
130 cm	34	2.9	12.0	976	1559	497	440	9.1	12.9	52	6.7	2.5	13	14 366
BB (17 yr)														
20 cm	33	4.3	2.1	38	0.3	432	23	5.9	3.3	28	0.3	0.3	11	1348
40 cm	29	3.1	3.4	216	22	466	76	1.6	7.4	45	2.9	1.0	16	2950
70 cm	33	2.5	9.4	1351	440	456	259	1.5	10.7	53	33.0	3.9	21	10 262
100 cm	31	2.3	15.7	2843	1080	439	338	0.5	14.5	55	111.0	0.01	20	18 325
MR (21 yr)														
20 cm	34	5.6	0.6	1.4	0.3	99	4	2.5	2.3	10	0.3	0.1	5	256
40 cm	33	3.6	1.7	23.5	0.9	429	12	1.3	3.3	16	0.2	0.3	7	1208
70 cm	35	2.8	2.6	114.1	15.3	384	28	1.2	3.5	32	3.7	0.5	8	1814
100 cm	36	2.6	3.4	86.1	25.0	511	26	1.1	3.6	42	10.3	0.3	12	2102
DD (35 yr)														
20 cm	33	5.2	0.4	0.3	0.1	77	4	2.2	2.0	11	0.1	42.2	5	103
40 cm	27	4.3	0.9	3.9	0.3	177	7	1.6	2.9	15	0.6	55.3	7	389
70 cm	27	3.5	1.8	15.7	2.1	331	8	1.4	4.4	27	5.8	48.5	9	971
100 cm	32	3.3	2.1	46.9	4.9	376	7	1.1	3.3	33	16.7	16.4	7	1330

^a Period May 1996–May 1998.

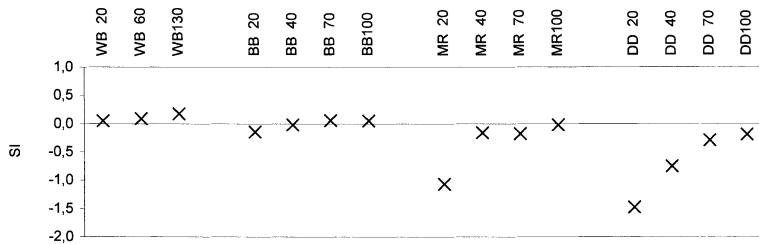


Fig. 1. SIs of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) in soil solutions of different soil depths and investigation sites.

Water balances of the chronosequence sites are discussed in detail in Scherzer (2001).

3.2. Soil solution

Element concentrations of soil solutions especially Al, Fe, Ca, Mg and SO_4 were high and increased with soil depth corresponding with decreasing pH values (Table 3). Highest concentrations are found at the younger sites WB and BB. With stand age concentrations decrease significantly.

The subsoils have low pH with 2.3–3.3, amelioration action raised pH to 4.3–5.5 in the surface soils. An exception is the WB site, where target pH of amelioration (pH 5.5) was not achieved due to insufficient amounts of applied alkaline ash.

Concentrations of Fe and Al were high at deeper soil depth but low in the ameliorated surface soil where Ca dominates the cations composition. Even in the non-ameliorated subsoils Ca remained the main base cation. Independent of depth and stand age SO_4 contributes >90% to anion composition (Schaaf et al., 1998).

K concentrations are in general low and decrease with depth. The higher concentrations in the subsoil of the WB site are caused by mineral fertilization carried out in spring 1996 (Wilden, 2000).

Concentrations of NH_4 are low in the surface soils, but show high values beneath the ameliorated layers. The highest value was found at the BB site. At the WB site, NH_4 concentrations are high in all depths. This might be related to mineral fertilization.

The distribution of NO_3 concentrations does not correspond with those of NH_4 . The sites BB and MR show very low NO_3 concentrations over the whole profile while concentrations at the WB site vary over a wide range. Especially at the beginning of

the study, high NO_3 concentrations were observed at WB due to mineral fertilization (Wilden, 2000). The oldest site DD shows elevated NO_3 concentrations in all depths.

SIs indicate that the Ca concentration of the soil solution is controlled by gypsum on all investigation sites (Fig. 1). A decrease in Ca concentrations and, respectively, a decrease in gypsum saturation are to be observed in the surface soils of the older sites and forthcoming with stand age.

At the BB, MR and DD site, SI indicate the soil solutions to be saturated or oversaturated by the minerals alunite ($\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$), basaluminite ($\text{Al}_4(\text{OH})_{10}\text{SO}_4$), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and gibbsite ($\text{Al}(\text{OH})_3$) in soil depth of 20 cm and at the DD also at 40 cm depth (Fig. 2). The calculated SI of jurbanite (AlOHSO_4) indicates that this mineral is near saturation in all soil solutions.

Soil solutions are oversaturated by goethite (FeOOH) and undersaturated by jarosite ($\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$) in general with exception to 130 cm depth at the WB site, where solution is nearly saturated by jarosite (Fig. 3). SI for amorphous $\text{Fe}(\text{OH})_3$ increase by stand age especially in the surface soil. While the solutions of the younger sites are undersaturated with respect to amorphous $\text{Fe}(\text{OH})_3$, saturation is given at the oldest site DD in 20 cm soil depth.

3.3. Element fluxes

Generally, inputs as bulk precipitation and through-fall deposition are low, and their spatial variation depends mainly on the amount of precipitation (Table 4).

Within the soil Al, Fe, and SO_4 -S fluxes increase significantly with depth. The distribution patterns of

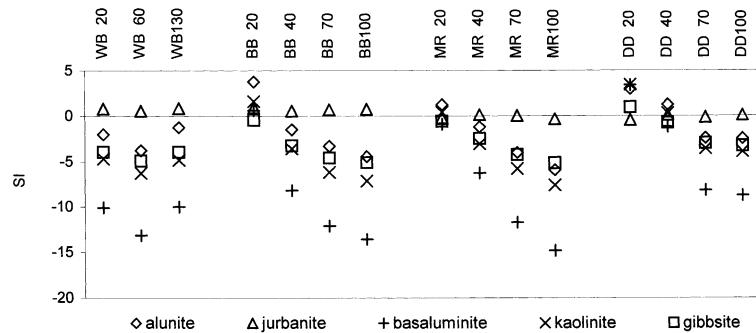


Fig. 2. SIs of the Al-minerals alunite ($\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$), jurbanite (AlOHSO_4), basaluminite ($\text{Al}_4(\text{OH})_{10}\text{SO}_4$), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and gibbsite ($\text{Al}(\text{OH})_3$) in soil solutions of different soil depths and investigation sites.

these fluxes are quite similar to those of the element concentrations. As a combined effect of higher water fluxes and high element concentrations, the youngest site WB shows the highest element fluxes within the soil profile as well as the highest outputs in 130 cm depth. Although element fluxes decrease considerably with site age, values remain high. In the deeper soil layers of the oldest site, DD higher Al and $\text{SO}_4\text{-S}$ fluxes occur compared to the MR site. They are mainly caused by the higher water fluxes. Element outputs of the examined ecosystems are manifold higher than the inputs for Al, Fe, Ca and $\text{SO}_4\text{-S}$ independent from site age.

K input by precipitation is low ($1.7\text{--}3.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$). Together with stand age K leaching from canopy increased to high values (max. throughfall: $13.9 \text{ kg ha}^{-1} \text{ yr}^{-1}$). Within the soil profiles, K flux decreased with soil depth to low values. K output

in 100 cm depth was about $0.3\text{--}1.5 \text{ kg ha}^{-1} \text{ yr}^{-1}$ with exception to the youngest site WB, where output in 130 cm depth was $17.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$.

N input ($\text{NH}_4\text{-N} + \text{NO}_3\text{-N}$) by bulk precipitation was almost similar at all sites ($13.1\text{--}19.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$). $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ each contribute to about 50% to the N input. At the DD site, bulk and throughfall fluxes of N were in same magnitude. Whereas at the BB and MR sites throughfall deposition was significantly lower than bulk precipitation. Although there is a noticeable input with throughfall, $\text{NH}_4\text{-N}$ flux decreased in the surface soils with exception to the WB site, where flux even in the surface soil was high. At the older sites, $\text{NH}_4\text{-N}$ fluxes increased significantly in the subsoils. It was highest in the subsoil of the BB site, where $45.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$ were leached from the rooted zone.

$\text{NO}_3\text{-N}$ fluxes in soils vary in a wide range. The

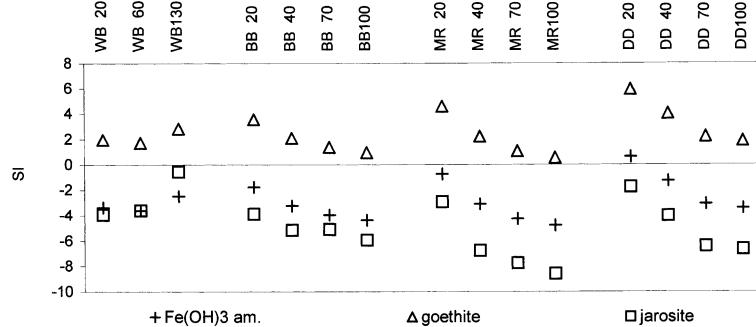


Fig. 3. SIs of the Fe-minerals $\text{Fe}(\text{OH})_3$ (amorphous), goethite (FeOOH) and K-jarosite ($\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$) in soil solutions of different soil depths and investigation sites.

Table 4

Annual element fluxes with bulk precipitation (pre), throughfall (thr) and in different soil depths for the period January 1996–December 1998 (n.d.: not determined)

Stand (age)	H ₂ O (mm)	H (kg ha ⁻¹)	Al (kg ha ⁻¹)	Fe (kg ha ⁻¹)	Ca (kg ha ⁻¹)	Mg (kg ha ⁻¹)	K (kg ha ⁻¹)	Na (kg ha ⁻¹)	Si (kg ha ⁻¹)	Cl (kg ha ⁻¹)	NH ₄ -N (kg ha ⁻¹)	NO ₃ -N (kg ha ⁻¹)	SO ₄ -S (kg ha ⁻¹)
WB (3 yr)^a													
Pre	737	0.25	n.d.	n.d.	21	2	2.4	12	n.d.	13	9.6	9.7	29
20 cm	301	4.00	2504	862	1307	1586	20.6	30	145	114	19.4	18.0	7821
60 cm	246	9.45	2772	3733	1142	1613	8.0	23	233	63	16.6	6.7	10 967
130 cm	227	3.60	2056	5028	1090	1125	17.3	29	192	46	12.6	2.8	11 708
BB (17 yr)													
Pre	797	0.22	0.9	0.9	8	1.9	2.4	7	n.d.	11	9.5	9.1	15
Thr	381	0.15	0.3	0.2	8	1.7	4.8	7	n.d.	8	3.5	4.8	12
20 cm	198	0.12	83.4	0.6	934	41.8	11.0	6	54	16	0.4	0.4	965
40 cm	112	0.71	377.0	31.6	525	80.9	3.0	8	45	17	2.1	0.3	1351
70 cm	70	1.91	885.6	345.5	303	179.9	0.9	7	33	16	17.4	0.7	2659
100 cm	51	2.52	1651.0	702.8	216	174.9	0.3	7	26	10	45.6	0.3	3742
MR (21 yr)													
Pre	704	0.18	0.9	0.6	9	1.2	3.3	8	n.d.	10	7.7	8.2	13
Thr	336	0.09	0.6	0.4	9	1.9	11.4	7	n.d.	8	4.6	5.0	13
20 cm	206	0.06	4.7	0.7	251	6.9	4.8	5	23	10	0.4	0.2	211
40 cm	128	0.40	49.5	1.4	554	18.4	2.1	4	18	12	0.2	0.3	522
70 cm	114	1.74	105.4	15.5	409	26.6	2.2	4	36	8	3.3	0.2	618
100 cm	92	2.29	76.9	23.1	445	23.3	1.1	3	35	10	7.2	0.1	624
DD (35 yr)													
Pre	562	0.15	0.2	0.3	3	0.5	1.7	2	n.d.	5	7.6	5.3	5
Thr	392	0.18	0.5	0.3	10	2.0	13.9	8	n.d.	12	8.0	6.4	16
20 cm	232	0.02	0.9	0.4	163	7.9	5.9	6	24	13	0.2	18.3	76
40 cm	192	0.11	5.6	0.5	337	12.3	3.5	6	26	14	0.6	21.9	244
70 cm	150	0.35	19.8	2.8	398	10.6	1.9	6	37	12	5.0	15.1	379
100 cm	120	0.61	58.1	5.1	419	9.0	1.5	4	39	8	13.7	5.6	488

^a Period May 1996–May 1998.

sites BB and MR showed only small fluxes ($<0.7 \text{ kg NO}_3\text{-N ha}^{-1} \text{yr}^{-1}$) over the whole soil profile in contrast to the WB and DD sites where elevated $\text{NO}_3\text{-N}$ fluxes occurred especially in the upper layers.

4. Discussion

The low pH values and high element concentrations especially of Fe and SO_4 as well as the high EC in the non-ameliorated subsoils are due to pyrite oxidation. High Al and Si concentrations are caused by enhanced weathering of primary minerals as there were detected quartz, feldspars, muscovite, kaolinite and biotite (Heinkele et al., 1999; Neumann, 1999). At the younger sites WB and BB, these processes dominate initially but decline in intensity over time as seen especially at the DD site. This is in agreement to findings by Neumann et al. (1997), and Heinkele et al. (1999) that the contribution of pyrite-S to total S in the subsoil decreases from $>60\%$ at BB to $<5\%$ at the DD site.

Highly concentrated soil solutions allow precipitation of secondary mineral phases. Gypsum crystals have been frequently detected in the investigated soils as well as in precipitations from soil solutions (Heinkele et al., 1999; Neumann, 1999; Schaaf et al., 1999). By the calculation of SI, it is shown that gypsum controls the Ca-solubility from the first year. As a result of advanced gypsum leaching, Ca concentrations decrease in the surface soil solutions of the older sites MR and DD. Formation of gypsum as a typical secondary mineral phase of pyritic spoils is commonly found (Karathanasis et al., 1990; Wisotzky, 1994; Evangelou, 1995), but also in naturally formed acid sulfate soils (van Breemen, 1973, 1982).

Main Ca sources for the soil solutions are the incorporated power plant ashes and the lignite fraction of the substrates. The ash contains about 4.5–14.0% CaO (Neumann, 1999). By amelioration action about $20\text{--}136 \text{ t Ca ha}^{-1}$ were added as ash to the investigated sites. With respect to the current Ca stores in a range of $54\text{--}93 \text{ t ha}^{-1}$ (100 cm depth), Ca budget of the sites has been basically influenced by amelioration. By means of soil microcosm studies Blechschmidt et al. (1999) found that Ca release from the examined lignite and pyrite containing

substrates into percolating soil solution is in the same magnitude whether amelioration ash was added or not. This points to lignite as a considerable source for available Ca because most of the appearing mineral substrates are free of Ca. In lignite samples of the region Schaaf (1997) found $13.0 \pm 0.7 \text{ g Ca kg}^{-1}$. Calculations indicate saturation with respect to gibbsite and kaolinite in the surface soils of the older sites. This is consistent with results of Neumann (1999) who detected these minerals by XRD analysis in the substrates. The observed undersaturation of gibbsite, alunite, basaluminite and kaolinite in all soil depths of the WB site is related to the low pH values. Whereas the saturation with jurbanite in all examined solutions can be explained by its wide ranged stability from $\text{pH} < 0$ up to 5 (Nordstrom, 1982). Although jurbanite was expected in the soils from the calculated SI, it was not observed by XRD analysis.

Jarosite was far under saturation in the solutions with exception to the subsoil of the WB site. But Neumann (1999) detected this secondary precipitate by XRD in every depth at all sites. Obviously, the current pH values are too high and K concentrations are too low for the precipitation of jarosite. Only the solution of the WB site in 130 cm soil depth meets the required conditions for jarosite precipitation. Together with the frequently detected jarosite appearance, this points to precipitation bound to the first years after dumping. In the period before, the amelioration ash is incorporated to the substrates, pH is low in the whole soil profile and K-concentrations are supposed to be high enough for jarosite precipitation.

The reduction of Al and Fe concentrations in the solutions of the ameliorated surface soil layers is most probably caused by the above-discussed precipitations that are mainly controlled by soil reaction (Wisotzky, 1994; Miller, 1995).

High Al concentrations of the soil solutions obviously do not affect tree nutrition deficiencies as seen by element concentrations of the needles (Table 5). This can be explained by simultaneously high Ca concentrations that generate molar Ca/Al ratios far above the critical value of 1 in the ameliorated layers (Ulrich, 1989; Cronan and Grigal, 1995). In the highly concentrated soil solutions of the non-ameliorated layers phytotoxicity of Al is reduced due to formation of ion pairs and sulfate complexes (Cameron et al.,

Table 5

Element concentrations of one- and two-year old pine needles

Site (age in years)	N (mg g^{-1})		K (mg g^{-1})		Ca (mg g^{-1})		Mg (mg g^{-1})	
	One-year	Two-year	One-year	Two-year	One-year	Two-year	One-year	Two-year
WB (3)	13.8	15.0	6.1	5.6	4.3	5.0	2.1	1.6
BB (17)	12.2	9.1	6.0	7.3	4.0	6.1	1.2	1.1
MR (21)	14.5	14.5	5.2	4.9	2.8	6.4	0.9	1.1
DD (35)	22.0	25.7	4.3	3.4	3.0	5.7	1.0	0.8

1986; Kinrade and Parker, 1987; Tanaka et al., 1987; McCray and Sumner, 1990).

As a combined effect of decreasing concentrations and simultaneously reduced water fluxes the element fluxes of Al, Fe, Ca, Mg and $\text{SO}_4\text{-S}$ within the soil decline from the youngest site WB to the oldest site DD. Despite the clear reduction of element outputs over time they remain much higher compared to forest ecosystems at non-mine sites. Outputs of the 35-year old site DD are for Al 1.5-fold, for Fe 17-fold, for Ca

7-fold and for $\text{SO}_4\text{-S}$ 5-fold higher, compared to a 45-year old Scots pine stand on glacial outwash sand (Schaaf et al., 1995).

Plant uptake in the upper layers can explain the decreasing K flux within the soil profile. Nutrition-status of the stands as indicated by K concentrations of the needles show a sufficient K supply (Table 5) although the sandy substrates show only low K contents in general. The small K output fluxes in combination with the enhanced K-leaching from the

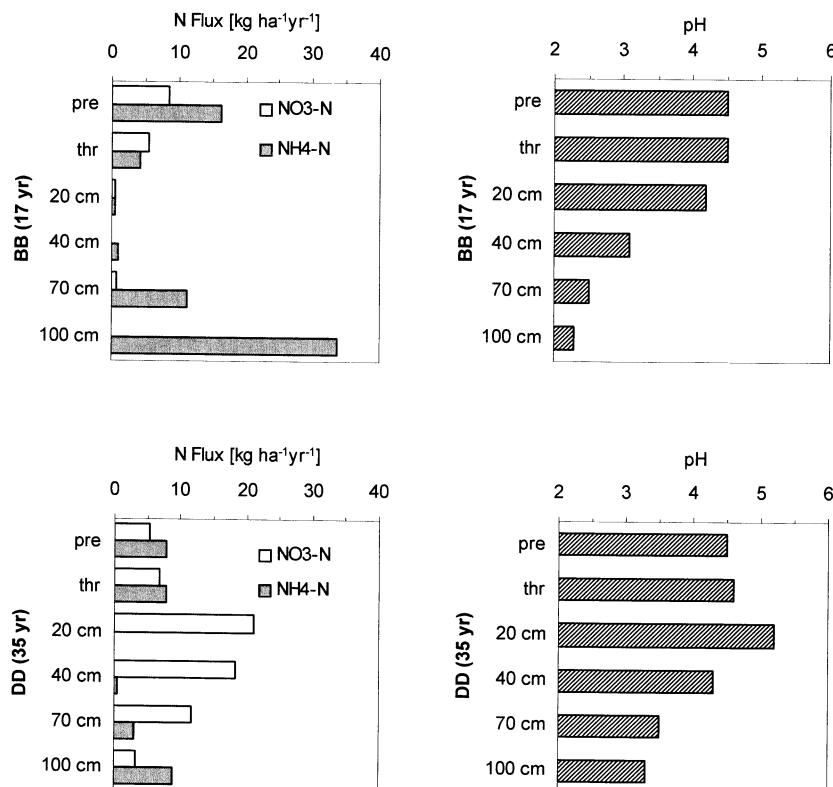


Fig. 4. $\text{NH}_4\text{-N}$, $\text{NO}_3\text{-N}$ flux and pH in bulk precipitation (pre), throughfall (thr) and in different soil depths at the BB and DD sites.

canopy point to considerable cycling within the forest ecosystems only two decades after revegetation. Enhanced K output fluxes on the three years old WB site are related to the mineral fertilization carried out before reforestation at the beginning of the investigations (Knoche et al., 1999).

At the BB and MR sites, direct N uptake by pine needles from atmospheric N-deposition occurs as evident from the difference between bulk and throughfall fluxes. Small N-fluxes within the main rooting zone and N-deficiency in needles (Table 5) indicate that the stands were N-limited. Poor N supply of forest stands on rehabilitated mine sites in the Lusatian region has already been reported by Heinsdorf (1994) who estimated this as a limiting factor for sustainable forest development in general. At the first sight, this is inconsistent with the elevated $\text{NH}_4\text{-N}$ flux in the subsoils especially those at the BB site. The data point to $\text{NH}_4\text{-N}$ release in the subsoil and the lignite fraction of the substrate seems to be the source. N release of examined lignite and pyrite containing substrates into soil solution has already been shown by Blechschmidt et al. (1999) by means of a soil microcosm study. As a result of plant uptake, the N fluxes in the main rooted zone of the BB and MR sites, that means in 20 and 40 cm soil depth are low. At the surface soil of the DD site N flux occurs mainly as $\text{NO}_3\text{-N}$. These $\text{NO}_3\text{-N}$ flux are supposed to be related to the N release by decomposition of the organic surface layers. The DD-site shows highest rates of decomposition compared to the other sites (Dageförde et al., 2000; Keplin et al., 1999). It seems as if the release from the organic surface layer exceeds the plant demand for N at the DD site, whereas at the BB and MR sites it is vice versa. Soil reaction over the whole profile of the DD site is almost one pH unit above the other sites (Fig. 4). This could result in higher nitrification rates from recent organic matter (De Boer et al., 1993). Contribution from decomposition of lignite fractions cannot be excluded. In contrary to the other sites, N flux at the site DD is highest in the rooted topsoil and decreases with depth simultaneous with pH. Needle contents show that this stand is well supplied with N (Table 5). Increase of nitrification in acid forest soils due to improvement of soil reaction is also reported from lysimeter studies (Matzner et al., 1983; Kreutzer et al., 1989; Marschner et al., 1989; Marschner and Wilczynski, 1991) and

from incubation experiments with limed samples (Lang and Beese, 1985; Ibrom and Runge, 1989).

Enhanced N fluxes at the youngest site WB is due to application of mineral fertilizer in context with the amelioration.

5. Conclusions

Outputs from the investigated forest ecosystems decrease strongly with time but show strong quantitative and qualitative differences to non-mine sites. Only for K element outputs are minor compared to the inputs and point to almost closed cycling of this nutrition element in the ecosystem. The N-content of the lignite containing substrates is a special quality of the investigated soils. Although there is a lack in the N-nutrition of the younger stands data point to N release especially from the substrates of the subsoil. This results in elevated output rates of $\text{NH}_4\text{-N}$. At the youngest site element budgets for K and N are still influenced by mineral fertilization carried out with amelioration action before reforestation.

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