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Effects of local soil chemical and hydraulic nonequilibria on element budgets of disturbed sites

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Abstract

Open-cast lignite mining is causing large-scale and long-term disturbance, resulting in a destruction of existing ecosystems and creation of landscapes formed by overburden spoil piles. Overburden sediments can largely differ from parent geological material of previously existing and undisturbed sites. It is expected that new soils and ecosystems will develop from overburden sediments with element budgets differing with respect to rates and directions of development from the undisturbed situation. Amelioration measures further alter the conditions. Typical for such mine soils is a small-distance spatial heterogeneity as a result of mixing of chemically and physically different sediments. Initially, highly different components, such as pyrite/alkaline ash, sand/clay/coal fragments, lignite/soil organic matter, or saline/nonsaline zones, can exist directly next to each other. For such small-distance spatial variability, predictions of long-term development of soils and sites are challenging. Despite disturbed soils show hardly any diagnostic horizons, soil profiles are characterized by distinct depth gradients of soil chemical conditions. Precipitation of secondary minerals, water repellency features of the lignite components, and preferential flow paths may depend on the small-distance spatial heterogeneity. Irregular flow patterns could be observed by dye-tracer studies. Element budgets of disturbed sites are characterized by extremely high soil solution concentrations of most elements. Although solute concentrations decrease with site age, they are still several orders of magnitudes higher compared to undisturbed sites even 50 years after disturbance. Chemical conditions change more rapidly along preferential flow paths than in other soil regions, by enhanced dissolution and transport, thereby affecting plant root growth and soil development. Better understanding of the effects of local non-equilibrium conditions in geochemical and physical properties will help to improve the long-term prediction of the development of disturbed sites.

Keywords: disturbed sites, mine soils, element budgets, spatial variability, local nonequilibrium

Introduction

At “disturbed sites”, the composition and spatial arrangement of soil, plant, and other components may differ from that of the original situation. One of the most striking examples for 'disturbed sites' are resulting from open-cast mining operations. Lignite coal mines, for instance, are mostly large-scale operations which lead to long-term disturbances of landscapes (Hüttl *et al*., 2000). The post-mining landscape is formed by overburden spoil piles. Overburden consists of a technogenic mixture of different geological sediments; e.g., in the Lusatian mining district in eastern Germany, quaternary and tertiary sediment mixtures are forming the basic material of the mostly sandy Regosol soils (Heinkele *et al*., 1999). These sediments can largely differ from parent geological material of previously existing and neighbouring undisturbed sites. Because sediments, which were not existent at the surface prior to mining, are now forming the parent material for soil development, it may be expected that the soils and ecosystems will differ with respect to rates and directions of development from the undisturbed situation or compared to post-glacial development. What makes predictions even more complicated is the fact that due to incomplete mixing of various sediments, the macroscopically homogenized overburden mine soils (i.e., considering a larger scale), typically exhibit a pronounced small-distance spatial variability of chemical and physical properties. Initially, after establishing spoil piles and reclamation, different components, such as pyrite and alkaline ash, or sand, silt, clay, and coal fragments, or lignite and soil organic matter, or saline and nonsaline zones, can exist directly next to each other in an mechanically instable irregularly distributed spatial arrangement (Thum, 1974; Varela *et al*., 1993; Vogler und Vogler 1998; Rumpel *et al*., 1998; Schmidt *et al*., 1999; Wisotzky, 1994).

Existing predictive models for describing water flow, solute transport, geochemistry, or soil mechanics, are generally based on the assumption that the pore-scale or local processes do not affect the volume-averaged processes and properties at the larger macroscopic scale (i.e., local equilibrium). While such models could possibly be used to study the soil forming processes and related effects on element budgets of previously unknown sediments, few models are currently available that are able to consider effects of small-distance spatial variability. Volume-averaged processes and properties appear irrelevant where one component (e.g., coal fragments) has completely different properties than other components (e.g., sand, ash) within the same volume. Small-scale property differences between components are so large that initially present differences as well as dynamically induced gradients in chemical or hydraulic properties, cannot be dissolved in time compared to the time-scale of the macroscopic process (i.e., local nonequilibrium).

The problems may lead to the suggestion of model approaches in which each component needs to be considered separately and for describing the transfer processes between the components. Attempts to consider differently permeable pore water regions are, for instance, two-domain models for improving the description of flow and transport in structured soils, such as the mobile-immobile 2-region transport model (van Genuchten and Wierenga, 1976) and dual-permeability flow and transport models (Gerke and van Genuchten, 1993). However, these models are mostly focussing on hydraulic and diffusion processes and not on more complex small-scale heterogeneity including that of geo-chemical components. Furthermore, physically realistic independent parameters and methods are lacking.

For disturbed sites, any predictions of long-term ecosystem element budgets require the development of a soil element budget model that includes the dynamics of processes which are resulting from small-distance spatial variability of soil physical and chemical properties. The objective to this paper is to discuss effects of nonequilibrium conditions and present examples of first simulation and experimental results for mine spoil heaps and reclaimed afforested mine soils.

Materials and Methods

The studied soils are from afforested and reclaimed mine sites located near the city of Cottbus in Eastern Germany. Dye tracer (Iodide) infiltration experiment were carried at a 1.25 m x 2.5 m ameliorated site that was afforested with 16-year old pine trees. After irrigating about 60 mm of water during 8 hours, the soil was excavated in steps of 10 cm down to 150 cm depth. The blue-colored areas formed by the Iodide-starch reaction were recorded. Water drop penetration time (WDPT) tests were carried on 60 ° C dried samples from the soil block to measure the potential water repellency.

In addition, soil solution chemistry and element cycling was studied in reclaimed mine soils developed on comparable substrates that formed a soil chronosequence of afforested sites varying in age between 5 and 55 years since dumping the overburden sediments (Gast *et al.*, 2001; Schaaf, 2001).

Reactive solute transport in large unsaturated mine spoil heaps was analyzed using a system of numerical models. Two-dimensional (2D) water flow and conservative and reactive solute transport was simulated for characteristic 2D-vertical spoil heap cross section to reflect the typical ripple structure with relatively low hydraulic conductivity for the compacted central areas and alternating low and high permeabilities for the flanks (not shown here). Smaller-scale spatial variability was introduced by randomly changing the proportions of Holocene, Pleistocene, and tertiary components within the mixed sediment stream at the conveyor bridge (Buczko *et al.*, 2001). The 2D-cross sections of hydraulic parameters were used for simulating spatial distributions of flow velocities and non-reactive solute concentrations and outflow and breakthrough curves at the bottom of the cross-section (Buczko *et al.*, 1999). For modeling the transport of multiple reactive solutes considering oxygen diffusion driven shrinkage-core type kinetic of sulphide oxidation and linear geochemical equlibrium processes, using a modified MINTRAN code hydraulical and geochemical parameter distributions in spoil heaps were randomly generated using the SGMS code of GSLIB program library (Gerke *et al.*, 1998, 2001). Figure 1 gives an impression of the generated spatial distributions used for the relative hydraulic conductivity at saturation and for the unoxidized core radius (left) as well as of the steady-state unsaturated water flow velocity field (right) obtained with an finite element code.

Results and Discussion

The reclaimed mine soils show hardly any diagnostic horizons. The soil profiles are dark from coal dust mixed in the sandy matrix. Within the sandy matrix, larger lignitic particles and fragments can be found, generally increasing in both, size and frequency, with soil depth. In the topsoil, remains of amelioration ashes and lime can be present. The lignite content has a strong influence on cation exchange and water holding capacity of the soils (Schaaf *et al.*, 2001). On the microscopic scale, crusts and coatings of iron oxides and gypsum around clods of flue ash, lignite and roots, and polyframboidal pyrite minerals aggregated between sand and coal dust particles can be found. These characteristic features and the spatial distribution of the components differ strongly from naturally developed soils of the region.

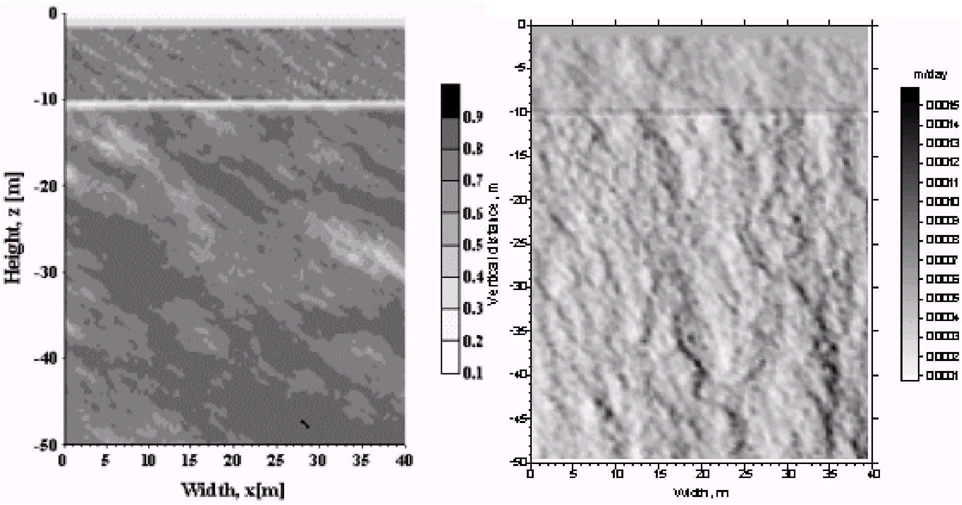

Figure 1 Two-dimensional vertical cross-sections of generated spatial distributions, left of the relative hydraulic conductivity and also of the relative radii of unoxidized pyritic sediment particles, and right of the simulated unsaturated water flow velocity field for imitating heterogeneity in large overburden mine spoil heaps.

The iodide-dye tracer experiment (Figure 2) showed that tracer could only be found at certain spots indicating that tracer movement was limited to preferential flow paths. The largest extension of the flow participating region was in about 10 cm depth (Figure 2, left picture). The dye-covered area was strongly reduced below 50-60 cm depth, however, some flow paths extended down towards 150 cm depth (Figure 2, right).

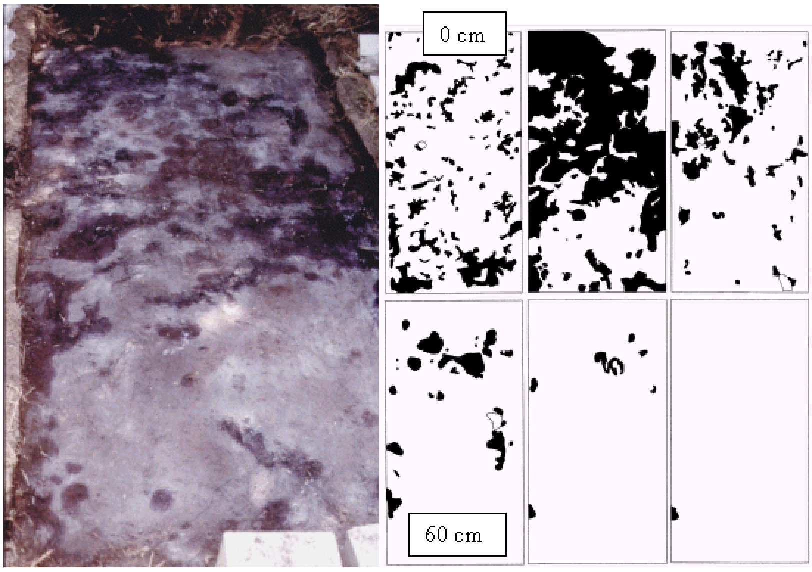

Figure 2 Photo of the mine soil dye tracer infiltration experiment for a 2.5m x 1.25m horizontal plane at 10 cm depth (left), and distribtion of Iodide-starch coloration areas at 0, 10, 20 cm (top), and 60, 110, and 150 cm depths (bottom right). The dark spots show the areas that contained Iodide and the white areas result from the starch dust.

The dye tracer patterns could not simply be explained by the spatial distribution of water repellency. The potential water repellency was found to be bipolar (either large or small) distributed and occurred for the whole block as well as locally (Gerke *et al*., 2001b). The water repellency could probably be correlated with the location of hydrophobic lignite particles or fragments. Actual water repellency could also be affected by the degree of iron coatings around lignite particles. The flow patterns, however, could probably be caused by a combination of different factors including air-water interactions, root channels, and small-distance variability.

Element budgets of disturbed sites are characterized by extremely high soil solution concentrations of most elements. Despite the absence of diagnostic soil horizons, soil solution composition showed distinct depth gradients both due to amelioration measures and soil development processes. Concentrations of major elements vary over several orders of magnitude and show a high spatial heterogeneity (Schaaf *et al*., 1999). In addition, The temporal variability within the soil chronosequence seems to be relatively high. Although solute concentrations decrease with site age, they still are several orders of magnitudes higher compared to undisturbed sites even 55 years after disturbance (e.g., Gast *et al*., 2001; Wilden *et al*., 1999).

In the numerical scenario analyses of flow and transport in mine spoil heaps, effects of soil spatial heterogeneity are described as macroscopic-scale processes. The only local nonequilibrium process considered is kinetic pyrite oxidation. The simulated breakthrough curves (Figure 3) show basic effects of the different types of spatial heterogeneity on solute transport in mine spoil heaps. Multicomponent reactive transport scenarios including kinetically (oxygen-diffusion) controlled weathering of pyrite under otherwise identical conditions are compared assuming 4 different types of hydraulic and chemical parameter distributions (see Figure 3): 1) The case 'het.' indicates that both, the hydraulic (i.e., air and water contents and water flow velocities are derived from variably-saturated flow simulations based on randomly distributed 'Miller-similar' hydraulic parameter functions) and the chemical (i.e., initial pyrite content and degree of 'pre-oxidation', mineral, and solute concentrations) parameters follow a random distribution. 2) The case 'lay, r_a' considers heterogeneous hydraulic and chemical parameter distributions as for 1) except that for the distribution of the radius of the unoxidized core, r_a, of the pyrite-containing idealized sediment particles, indicating the initial stage of pyrite oxidation, mean values obtained from horizontally averaging the heterogeneous case were used. This case implies that the initial distribution of mineral and solute concentration is layered since these initial values are strongly correlated with the initial degree of weathering. 3) The case 'lay, $r_a + f_s$' considers, in addition to parameters of case 2), also the fraction of sulfur, f_s, indicating the initial amount of pyritic sulfur content, to be horizontally layered. 4) Eventually, the case 'lay, $r_a + f_s$ + water + air' considers that also the hydraulic parameters are distributed in horizontal layers (i.e., forming a completely 1D-vertical transport domain), such that horizontal spatial variability is eliminated while the local nonequlibrium process of pyrite oxidation is preserved.

The breakthrough curves of chloride in Figure 3, as a non-reactive or conservative solute, show that a randomly generated heterogeneous spatial distribution of hydraulic parameters generally leads to a spreading of the solute breakthrough curves (i.e., “macrodispersion”).

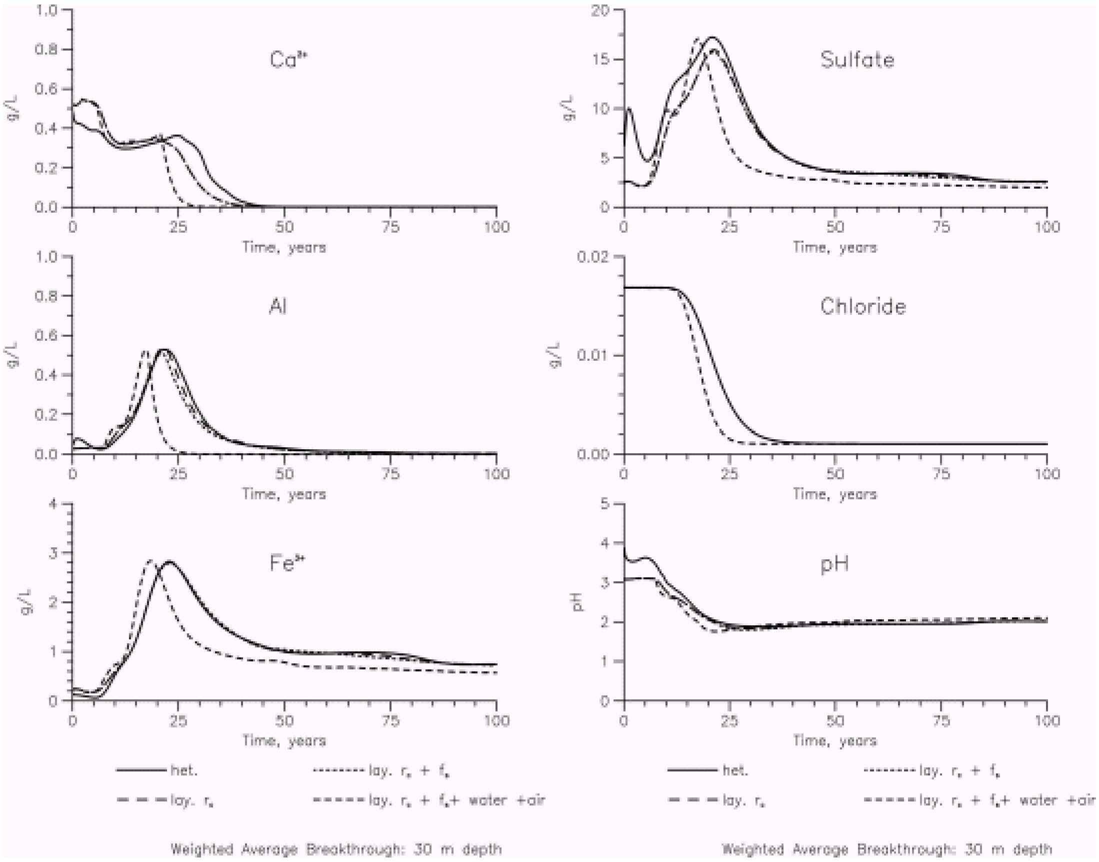

Figure 3 Simulated mass-averaged breakthrough curves at 30 m depth of a generic two-dimensional vertical cross-section imitating a mine spoil heap showing the solute concentrations of cations and anions in the pore water as a function of time calculated with a reactive multicomponent transport model with kinetic pyrite oxidation assuming steady-state water flow (average q = 0.189 m y^{-1}) for vertically layer-averaged and stochastically generated distributions of initial conditions and chemical and hydraulic parameters.

The spatial heterogeneity of chemical properties such as initial pyrite content, solute concentrations, and buffer mineral distributions, generally lead to a smearing and elongation of the leaching period as well as to an increase of the depth of the oxidation front (not shown here) with respect to ions participating in pH-depending equilibrium reactions within the liquid phase and to the pH-depending dissolution and precipitation reactions in contact with the solid mineral phase. The effect of initially pre-oxidized patches within the spoil heap can clearly be reflected, for instance, when looking at the sulfate breakthrough curve (Figure 3) for the 'het.' case where in the initial period, the sulfate concentration peak originates from leaching of sulfate which is already present in the pore water of the spoil heap as a result of burying pre-oxidized sediments deep in the spoil heap, here close above the 30 m depth. The same effect, but to a smaller extent, can be seen in the initial period of the pH- and Al- breakthrough curves. Furthermore, the breakthrough curves of the reactive solutes are controlled by the respective equilibria with the heterogeneous solid mineral phase distributions. Leaching of calcium, for instance, ceases much later in case of the herterogeneous distribution of parameters compared to the layered one.

The nonequilibrium kinetics if compared to an equilibrium approach, obviously would result in a longer duration of leaching period and is affecting all ions participating in the weathering process and in liquid and mineral phase reactions. (Gerke *et al*., 1988).

Conclusions

Chemical conditions change more rapidly along preferential flow paths than in other soil regions, by enhanced dissolution and transport, thereby affecting plant root growth and soil development. Better understanding of the effects of local non-equilibrium conditions in geochemical and physical properties will help to improve long-term prediction of element balances of disturbed sites.

The small-distance spatial variability affects probably also other processes such as for instance the colonization of soil by microorganisms and plants, the establishemnet of preferential flow paths, mineral weathering, mineral precipitation rate and spatial distributions, the leaching of salts and the development of the tree root distribution. Resulting from local nonequilibrium processes, new spatial structures, such as mineral precipitation zones, can develop which in turn may affect the long-term element budgets.

A soil model for disturbed sites that includes local nonequilibrium processes requires flow, transport and chemical submodels that separately consider flow domains for fast and slow velocities, and up to four chemically different pore domains. coupled by diffusive mass transfer terms.

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