

Ways of soil development on stony substrate from hard coal mining spoil

Wolfgang Burghardt*¹, Monika Niggemeyer¹, Martin Braunersreuther¹

¹ University Duisburg-Essen, Faculty of Biology, Soil Technology, Universitaetsstr. 5, 45141 Essen, Germany

* Prof. Dr. W. Burghardt, wolfgang.burghardt@uni-due.de

Abstract

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Sulfur content

The effect of stones on soil development is presented with the example from a hard coal mining spoil. The investigation concerns the stone and fine earth content, total bulk density, bulk density of the stone and fine earth fractions, pore volume, total carbon, organic carbon and sulfur content, as well as pH. Samples were taken from two different mine spoil depositions, the Monopol and Achenbach spoils, of the vegetation trial at Waltrop in the Ruhr area, Germany. Sampling occurred at the start of the trial and at two times, 8 and 14 years later, respectively. The samples were taken in big rings which were pushed into the mine spoil soil. The depth intervals of samples were tight with 0–2, 2–5, 5–10 and 10–20 cm below surface. Total organic carbon (TOC) and sulfur content and the pH were measured using standard methods. TOC was determined by destruction with H₂O₂. This proved to be a reasonable method which is relatively easy to perform. Soil formation occurred very fast, within eight years, in substrate with stone content by weight of 60–80 %. However, the way of soil formation differed between the Monopol and Achenbach mining spoils. The results indicate that in Monopol spoil, accumulation of TOC occurred by the intrusion of fine earth into pores between stones. However, in Achenbach spoil, fine earth was formed by the weathering of stones. The TOC content of fine earth achieved 8 to 16 mg g⁻¹. The TOC stock within 20 cm depth and 1 m² of the Monopol and Achenbach spoils was 1.0 and 0.6 kg, respectively. This is a very low value compared to the up to 12 kg in 1 m depth of the farmland soils of the region. The mine spoils contain between 2 and 18 mg g⁻¹ sulfur. Formation of sulfuric acid by the oxidation of sulfides in the spoil decreased the pH from 7.6 at the start of the trial to around pH 4.5 after 8 and 14 years in the Achenbach spoil. In the Monopol spoil, the pH increased with depth from pH 5.5 to 7.2. A negative relationship exists between pH and pore volume for the Monopol spoil. Due to the TOC accumulation of more than 6 mg g⁻¹, the soils can be classified as Regosols from hard coal mining spoil substrate in the German classification system. Within US Soil Taxonomy, the soils would belong to the Entisols. In the WRB soil classification, the soils can be positioned to the Leptosols, with the qualifiers Technic, Skeletic and as an additional qualifier Sulfuric and Intrusic or alternative to the Technosols with the qualifiers Spolic, Skeletic, Sulfuric and Intrusic. The occurrence of stony spoil deposits from deep hard coal mining in the loess belt from Eastern France to Eastern Ukraine changed large areas of originally rich fertile soils to poor soils, but promoted the bio-diversity.

1. Introduction

About the importance and effects of stones in soils exists only few research contributions (Flint and Childs, 1984; Nichols et al., 1984; Hantschel et al., 1992; Schulin et al., 1995; Niggemeyer and Burghardt, 1996; Burghardt, 1997). The effect of stone content on the properties of soils in urban, industrial and mining environments, the SUITMA soils (Soils of urban, industrial, traffic, mining and military areas, Burghardt et al., 2015), was highlighted by Burghardt, 1994. One extreme of stone content in soils, are soils from hard coal deep mining spoils. They are the focus of this publication.

Soils from stone cover are a typical feature of hard coal deep mining areas. Recently hard coal mining areas became of particular interest because of the finding that the release of CO₂ in the atmosphere by coal burning is a main cause of climate change. The consequence will be that coal age will end in the near future, at least in Europe (Alves Dias et al., 2018). Coal for energy generation will be no longer extracted and burnt. What is left besides the CO₂ contamination of atmosphere is a huge amount of stony coal mining spoil which now forms new, human-made mountainous landscapes in mining areas. In the hard coal mining areas of the Ruhr in Germany, approximately 25 km² are covered by coal mining spoil (Schneider, 1989).

Not vegetated, hard coal mining spoil heaps form cool, unfriendly black landscapes, and are sources of dust and of extreme temperature variations. Thus the living quality for humans in former mining areas is affected. To overcome this, the mining spoil is subject to greening, mostly by tree plantation (Pietrzykowski and Krzaklewski, 2018). In respect of greening of the abandoned mining regions, mine soils become of importance (Down, 1975; Jochimsen, 1987, 1989, 1996, 2001; Maiti and Saxena, 1998; Maiti and Ghose, 2005; Sheoran et al., 2010; Kompala-Bąba et al., 2019). Of particular interest is the high contribution of stony soils from hard coal mining spoil to bio-diversity (Zimmermann and Burghardt, 1987; Weiss et al., 2005; Woch et al., 2013; Kompala-Bąba et al., 2019). The question is, what will be the conditions for soil development, and how will the soil formation look like?

The texture of hard coal mining spoil can be diverse which will determine essential the properties and formation of soils from individual mining spoils. The texture differences are due to the processes of sedimentation during the geological era of Carboniferous and the ways of coal mining nowadays. The process of hard coal formation started in the geological era Carboniferous in flat coastal areas. The decrease of land sinking during the phases of the Geosyncline in the Carboniferous era resulted in a reduction of slope between land and sea. Thus the eroded sediments transported into the coastal waters were first sand, followed by silt and finally clay, on which swamps with wooden peat were formed. The recurrence of subsidence covered the peat with mineral, mostly sandy layers. The peat material developed over more than 300 million years into hard coal. That means for the tailings of hard coal mining, they can be from schistose sandstone, silt stone and clay stone. They weather to sand, silt and clay (Kelly and Kelly, 1987). One can observe this at surface outcrops of layers of Carboniferous era. For example, Stagnic-Vertic Cambisols with mighty layers of high clay content and Stagnic properties or Podzols on weathered sandstone occur.

From recent mining operations occur tailings from mine shafts and from tunneling for the access to the coal layers, of mineral layers beside the coal bed and washing tailings from the separation of coal from minerals by washing. Most of the mine spoil is from washing tailings. Some of the earlier washing tailings can still have higher contents of up to 40% fine distributed coal due to the incomplete separation of coal from minerals by the separating technique at that time. Under aeration, this material can burn and will become red-colored and only slightly acid. The individual tailings differ by the size of the stones and the textural composition.

The extracted tailings come mostly from layers in great depth where no aeration and by this oxidation occurs. That means that the sulfur of the original organic matter from peat of the coal was transformed into sulfides. Iron sulfides occur. They are a dominant characteristic of mining spoil which influences essential the soil formation (Kerth, 1988; Burghardt, 1989a; Uzarowicz, 2011). Brought to the earth surface, the sulfide will oxidize and form sulfuric acid. The pH of the hard coal mining spoil will strongly decrease from originally a pH of around 7 or above, down to sometimes a pH of around 3 (Daniels and Zipper, 2018) which is too low for vegetation growth. The water of the swamps of peat formation was partly brackish. That means

it was saline to some extent. Thus some coals, the salt coals, and their tailings are saline.

Another important influence on soil formation on hard coal mining tailings is the way of depositing the material. The techniques used changed with time. Old tailings were comparatively loosely deposited. Thus they were sensitive to spontaneous ignition. Younger tailings were thus compacted, and the surfaces bulldozed and leveled, which means without micro-relief, which would favor natural deposition of seeds, fine earth and water collection, and greening (Burghardt, 1989b).

2. Material and methods

The samples were taken from the greening trial of hard coal mining spoil in the city of Waltrop, Ruhr area, Germany (Fig. 1). The climate is moderately humid, with a mean annual temperature of 9.7°C (from 1980–2010) and a mean annual precipitation of 848 mm (Deutscher Wetterdienst, 2019). Investigated were mining spoils from two different mines of the region: Monopol mine and Achenbach mine. The deposited mine spoil was from recent mining. The spoils of both mines were deposited in a spoil heap which has the form of a long hill with a large plateau, designed as a landscape construction. On the plateau and the slopes, a field trial (Jochimsen, 1989) of 70 parcels with seven trial variants was established for studying soil greening processes. The trial includes different vegetation variants such as air born seeding which was dominated by birch tree seeds, seeding of a seed mixture *Dauco-Melilotion* community (Jochimsen, 1989), weak fertilizing, and three soil variants. The three soil variants concern pure tailings, a mixture of tailings with sandy loam 3:1, and a thin (5 cm) cover with sandy loam (Burghardt, 1989c; Braunersreuther and Burghardt, 2002; Burghardt and Niggemeyer, 2002). In the following, results of texture composition, of bulk density and pore volume, organic carbon content, sulfur content and pH from the soil trial variants, pure Monopol and Achenbach tailings, will be presented.

The investigations were performed on the pure tailing variants of Monopol and Achenbach mine spoil at the beginning and of tow time intervals, 8 years and 14 years after the establishment of the trial. The samples are for each sampling time and mine spoil from 6 parcels of the trial and four depths.

The samples were taken in early spring when the soil was moist from winter precipitation. The sampling was done two days after rainfall. The soil was saturated to field capacity. For gaining volume samples (Fig. 2), a metal ring of 32 cm diameter and 30 cm height was pushed 15 cm into the soil. For each sample 20 times the distance from soil surface to the top of the ring and the soil depth 2 cm, 5 cm and 10 cm was measured. For sampling 10 to 20 cm depth, a ring of 16 cm diameter was used. From the diameter and depth interval, the sample volume was calculated. From the weight of the dried sample, and the sample volume, the soil bulk density (volume weight) was calculated.

The texture fractions from fine earth (< 2 mm) and gravel (2–6, 6–20, 20–60, > 60 mm diameter) were determined by sieving. The fine earth composition of the two soils from Monopol mine and Achenbach mine was described by Braunersreuther



Fig. 1. Hard coal mining spoil greening trial Wal-trop, Ruhr area, Germany, (A) past 3 years (Photo by W. Burghardt), (B) past 12 years (Photo by W. Burghardt)

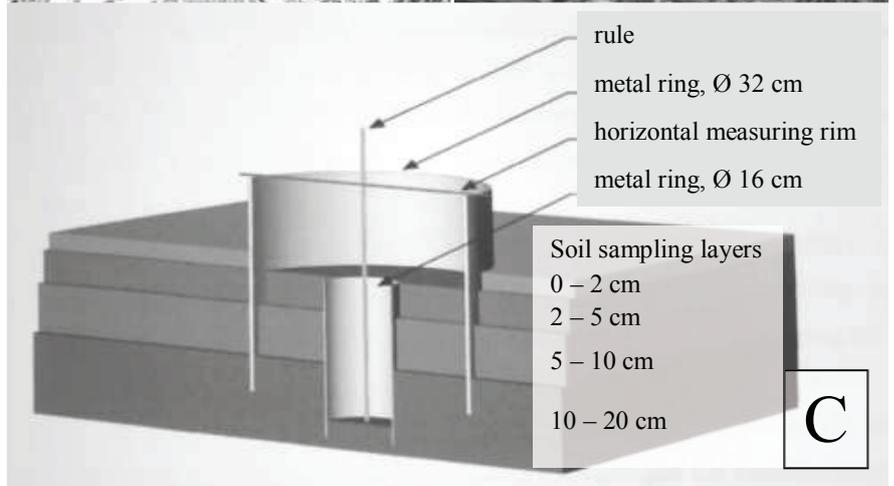


Fig. 2. Scheme of procedure of volume sampling for bulk density determination of soils from hard coal mining spoil, (A) sampling rings Ø 32 cm and 16 cm (Photo by W. Burghardt), (B) measuring depth of soil sample surface and bottom layer (Photo by W. Burghardt), (C) sampling arrangement

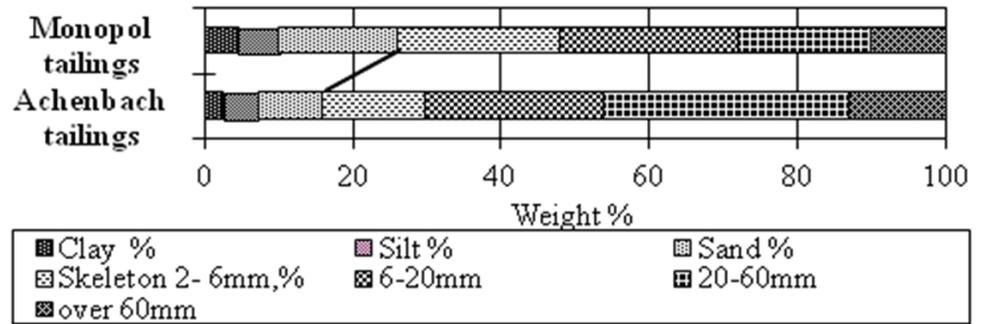


Fig. 3. Grain size distribution of fine earth and skeleton of Monopol and Achenbach mine spoil (Braunersreuther and Burghardt, 2002)

and Burghardt (2002) (Fig. 3). The pH of the fine earth was measured in a 1 : 2.5 soil : 0.01 M CaCl₂ dispersion. The total carbon and sulfur content was obtained with a Carlo Erba C-N-S analytical apparatus by gas chromatography.

The mine spoil contains inorganic carbon from hard coal. Thus the method to determine the organic carbon must be selective enough to measure primarily the organic carbon, and not parts of inorganic carbon. The traditional method to achieve this is the use of H₂O₂ (Jackson, 1958; Savage and Stevenson, 1961; Burghardt, 1989c). The method adds 25 ml of 6% H₂O₂ to 12 g fine earth. After 3 hours, the sample will be boiled down on the laboratory sand bath, and the sample weight will be determined again.

The results were classified as per table 1.

3. Results

3.1. Skeleton content and volume of fine earth, skeleton and pores

The main feature of hard coal mining spoil is the high content of skeleton from rock fragments which dominates the solid matter. It accounts for at least 50% of the solid matter. In the investigated four depth segments of the upper 20 cm of the hard coal mining spoil, the distribution of the content differs between the Monopol and Achenbach spoils (Fig. 4). Apart from in the depth of 0–2 cm, the skeleton content of Monopol spoil was lower than in the Achenbach spoil deposits. With increasing depth, the skeleton content stayed nearly constant in the Monopol spoil deposit. Different from this, the skeleton content of Achenbach spoil deposits increased significantly with depth continuously and was down from 2 cm higher than that of Monopol spoil. The results between the investigations after 8 and 14 years of establishment were about similar.

Table 1
Classification of bulk density and soil organic carbon (STOC) content and stock (Burghardt and Schneider, 2016, improved)

Category	Bulk density ¹⁾ g cm ⁻³	TOC-content ¹⁾ %	TOC-stock ²⁾ kg m ⁻²
Very low	< 1.20	0.6	< 2
Low	1.20–1.40	0.6–1.2	2–4
Moderate	1.40–1.60	1.2–2.4	4–8
High	1.60–1.80	2.4–4.8	8–16
Very high	> 1.80	4.8–8.7	16–24
Extreme high		> 8.7	> 24

¹⁾ Arbeitsgruppe Boden (2005)

²⁾ Calculation of the TOC density from the regression equation: TOC density = -0.59 TOC² + 14.39 TOC + 0.5; R² = 0.84; n = 83;

An insight into the structure of the hard coal mining spoil deposits gives the distribution of the volume of the different size classes of skeleton, of fine earth and of the pores within the spoil (Fig. 5). The fine earth volume was higher in the Monopol spoil as in the Achenbach spoil deposit. With increased depth, there was a slight increase of fine earth volume in the Monopol spoil but a slight decrease in the Achenbach spoil. The coarse skeleton fractions increased with depth. This was more expressed in the Achenbach spoil than in the Monopol spoil. The pore volume of both spoils decreased very strongly with depth from over 60% to less than 50%. This was more expressed in the Achenbach spoil than in the Monopol spoil. The spread of pore volume decrease in the Monopol spoil was about 17 and 20 vol. %, and Achenbach spoil was 30 and 35 vol. %, after 8 and 14 years, respectively, after trial establishment.

3.2. Bulk density of the deposited spoil

The bulk density (volume weight) of the different spoils generated by the fractions fine earth and skeleton varied between very low to low (1200 mg cm⁻³) and very high (>1800 mg cm⁻³) values (Tab. 1). The bulk density increased distinctly with depth from low to very high for the Monopol spoil and from very low to high for the Achenbach spoil (Tab. 1, Fig. 6).

The contribution of skeleton fraction to the bulk density is higher than of the fine earth fraction. The contribution of skeleton fraction increased distinctly with depth in both spoil variants and sample times. The contribution of fine earth fraction to the bulk density of Monopol spoil was overall, in depth and sampling times, about the same and down from 2 cm higher than in the Achenbach spoil. In the Achenbach spoil, the contribution of fine earth fraction decreased strongly with depth.

A close relationship between bulk density of skeleton fraction to the fine earth fraction would indicate a weathering of skeleton

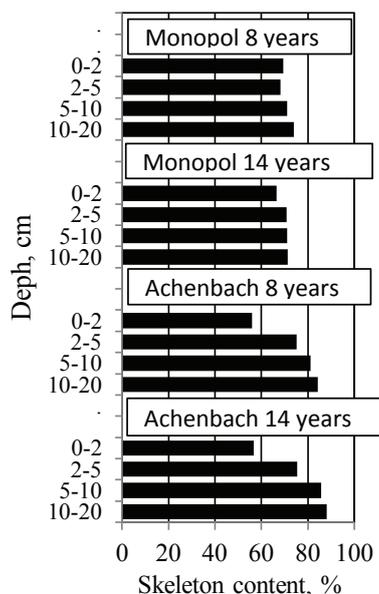


Fig. 4. Distribution with depth of skeleton (stone) contents of the hard coal mining spoil greening variants Monopol and Achenbach spoil of the field trial Waltrop, 8 and 14 years past the trial establishment

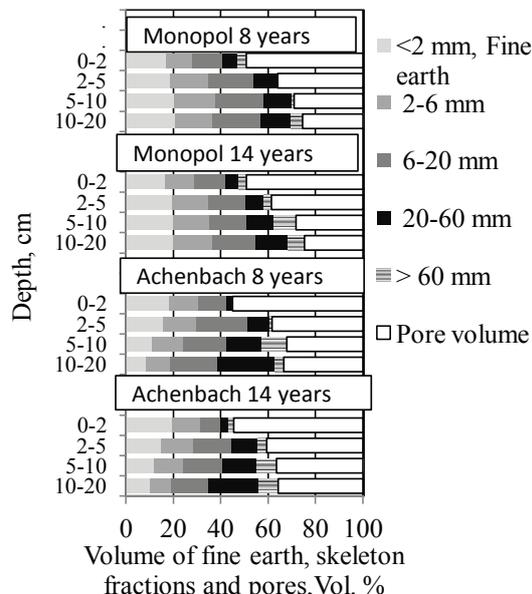


Fig. 5. Distribution with increasing depth of the volume of fine earth (< 2 mm), of skeleton fractions 2–6, 6–20, 20–60, > 60 mm diameter and pore volume of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrop, 8 and 14 years past the trial establishment

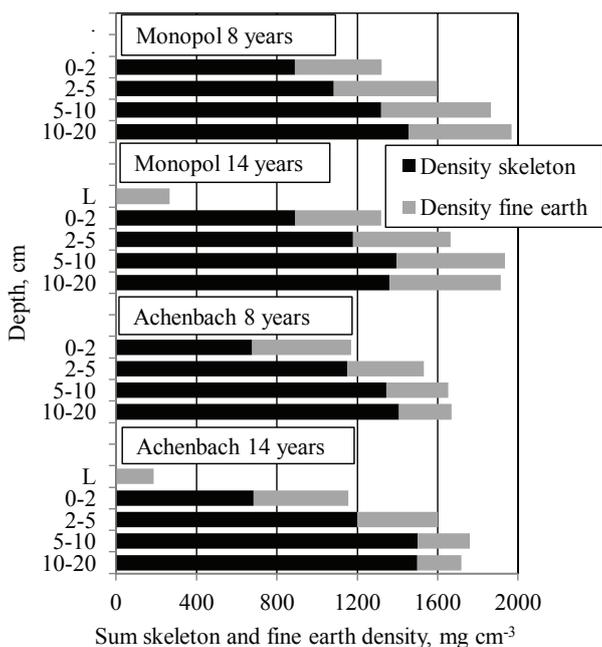


Fig. 6. Distribution with increasing depth of the bulk density (volume weight) and its density of skeleton and fine earth fraction of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrop, 8 and 14 years past the trial establishment

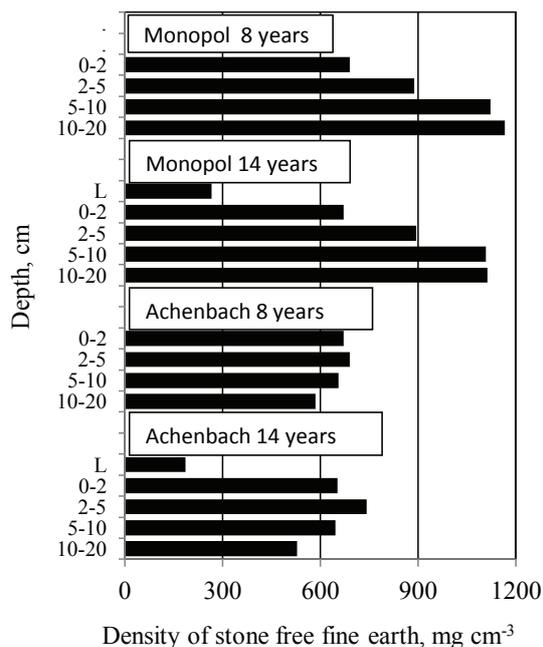


Fig. 7. Distribution with increasing depth of the bulk density of the fine earth in the spaces between the stones of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrop, 8 and 14 years past the trial establishment

from rock fragments. In the Monopol spoil this was not existing, but a close one was determined for the Achenbach spoil (Fig. 8).

After 14 years of the trial, litter (L) did form on the spoil surface. The bulk density of the litter was about 200 mg cm⁻³, and for the Achenbach spoil was slightly lower than for the Monopol spoil (Fig. 6).

The fine earth material will fill the void volume between the gravel and stones which form the skeleton. Thus the density of fine earth filling related to the volume between stones was calculated. The bulk density of fine earth in the voids between stones is very low (between 500 and 1200 mg cm⁻³; Tab. 1). The bulk density of fine earth between stones is lower in the Achen-

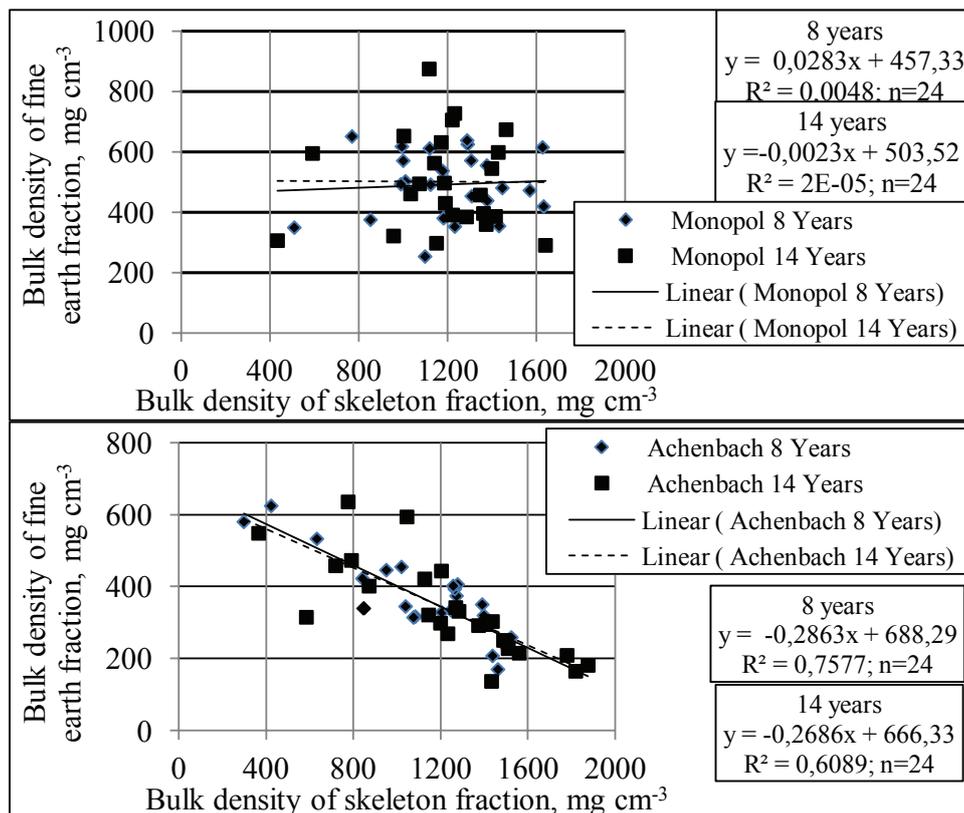


Fig. 8. Correlation of the bulk density of fine earth fractions and the bulk density of skeleton fractions of the hard coal mining spoil Monopol and Achenbach

bach spoil than in the Monopol spoil (Fig. 7). The bulk density of the fine earth in the spaces between the stones increased with depth in the Monopol spoil. In the Achenbach spoil, there was primarily a decreasing trend. For both sampling times of 8 and 14 years after trial establishment, the results of bulk density of fine earth between stones are about similar.

3.3. Inorganic and organic carbon content and stock

The sum of inorganic and organic carbon content from coal and of the organic carbon content of humic substances in the Monopol spoil was between 100 and 150 mg g⁻¹ and in the Achenbach spoil was between 50 and 190 mg g⁻¹. The increase with depth was low in the Monopol spoil, and high in the Achenbach spoil (Fig. 9).

In the hard coal mining spoil of both trial variants, Monopol and Achenbach spoils, within eight years, a marked accumulation of TOC occurred in the fine earth (Fig. 10). This main feature of soil development was present in all trial variants. But it differed significantly between the two mining spoil variants. Eight years after the trial start, in the top 2 cm of Monopol spoil, the TOC content of fine earth was already moderate (Tab. 1). It decreased gradually from 0–2 to 10–20 cm depth and then to low values. After 14 years, the TOC content was also in the 2–5 cm depth moderate.

In contrast to this pattern, the TOC contents of fine earth in the Achenbach spoil was low (Tab. 1) in all investigated depth segments after eight years of soil development. The TOC content was nearly uniform over the total depth of 0–20 cm and thus

did not show the strong decrease of the Monopol mining spoil. After 14 years, the TOC increased in the top 0–2 cm to moderate contents. Also in the depth segment 2–20 cm, an increase of TOC was distinctly determinable, but it stayed nearly uniform low with depth.

In most soils the accumulation of TOC will have an influence on fine earth bulk density. Fig. 14 shows a close relationship between TOC content and bulk density of the Monopol samples, which decreased slightly with time in 14 years. The correlation for the Achenbach spoil after 8 years was weak and increased strongly with time. The results indicate differences for intrusion of organic carbon rich fine earth which occurred in Monopol spoil earlier than in Achenbach spoil.

The stock of TOC which accumulated in 8 and 14 years was in both mining spoils very low (Tab. 1). The TOC stock was 0.98 and 1.0 kg TOC m⁻² in 0–20 cm depth of the Monopol spoil after 8 years and 14 years, respectively (Fig. 11). In the Achenbach spoil, the TOC stock was much lower and achieved 0.56 and 0.65 kg TOC m⁻² in 0–20 cm depth.

3.4. Sulfur content and pH

The sulfur content differed between the two mining spoils, Monopol and Achenbach (Fig. 12). The sulfur content was in the fine earth of Monopol spoil in the top 2 cm 2.0 mg g⁻¹, and the Achenbach spoil 3.7 mg g⁻¹ in the 8 years samples. The sulfur content decreased to 1.4 mg g⁻¹ in the Monopol spoil and 1.9 mg g⁻¹ in the Achenbach spoil in the 14 years samples. With increasing depth, the sulfur content increased in the fine earth of Mo-

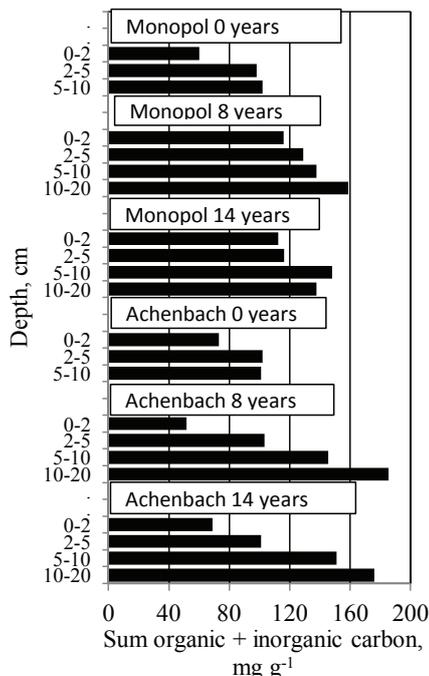


Fig. 9. Distribution with increasing depth of the sum of the inorganic and organic carbon content of the fine earth of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrup, 8 and 14 years past the trial establishment

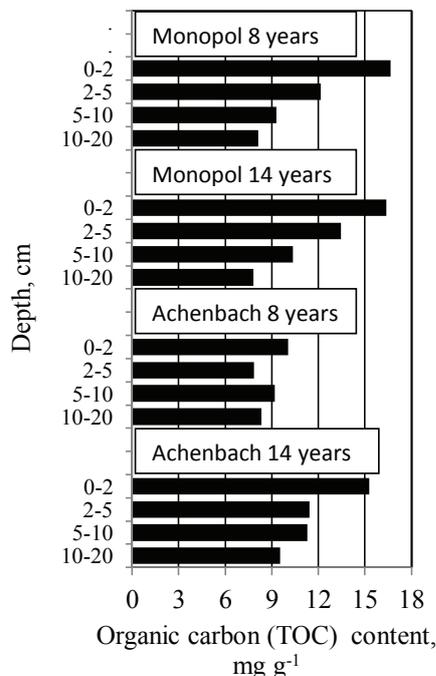
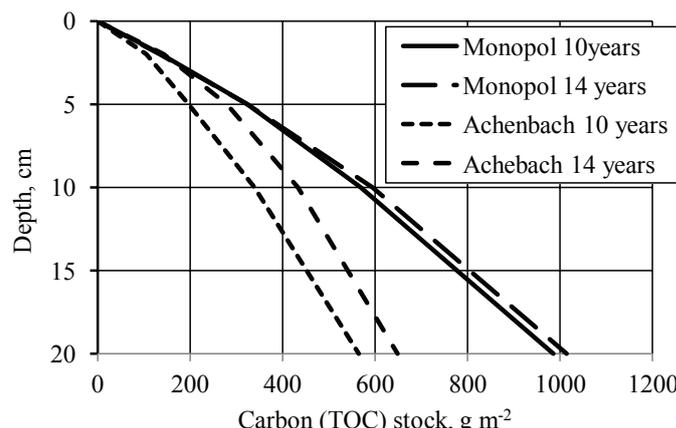


Fig. 10. Distribution with increasing depth of the organic carbon (TOC) content of the fine earth of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrup, 8 and 14 years past the trial establishment

Fig. 11. Cumulative depth curve of the organic carbon (TOC) stock from 0-20 cm depth of the fine earth of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial Waltrup, 8 and 14 years past the trial establishment



nopol spoil only slightly to 3.3 and 2.2 mg g⁻¹ in 10–20 cm depth in the samples taken after 8 and 14 years, respectively. For the Achenbach spoil, the contents strongly increased with depth to 10.9 and 7.7 mg g⁻¹, respectively. The results of both spoils indicate a decrease of sulfur content in the top soil with time.

Past 14 years, at sites of both spoil variants, a litter layer was already formed by the vegetation. The organic material of the litter also contained sulfur (Fig. 12). With 1.3 mg g⁻¹, the sulfur content was in the Monopol spoil variants lower than in the Achenbach variants with 2.1 mg g⁻¹. In both trial variants, the sulfur content in the litter was in the range of the sulfur content of the fine earth in 0–2 cm depth.

The sulfur occurs in the mining spoil as Pyrite and Marcasite. These sulfides will, with the mining spoil, come into contact with air and oxidize to sulfuric acid. This process lowered the pH value which was measured in the fine earth fraction of the

spoils. At the time of establishing the field trial in Waltrup, the pH of the fine earth of both Monopol and Achenbach spoils was pH 7.6 – i.e. very weakly alkaline (Burghardt, 1989c). In the Monopol spoil it decreased after 8 years to a slightly acidic value of pH 5.6 and after 14 years to 5.2 in the top 0–2 cm layer and increased with depth strongly to neutral values of pH 7.2 after 8 and 14 years (Fig. 13).

The results of the Achenbach spoil were totally different from the Monopol spoil. The Achenbach spoil had in its 0–2 cm depth, after 8 years, a pH value of 4.2 which raised to pH 4.6 in the 14 year old spoil. Until the depth of 20 cm, the pH stayed nearly unchanged and achieved values of pH 4.5 and 4.4 after 8 and 14 years trial time, respectively. Thus the fine earth fraction of the Achenbach spoil was strong acid over all investigated depth intervals. But after 14 years the pH showed a slight increase in 0–10 cm depth.

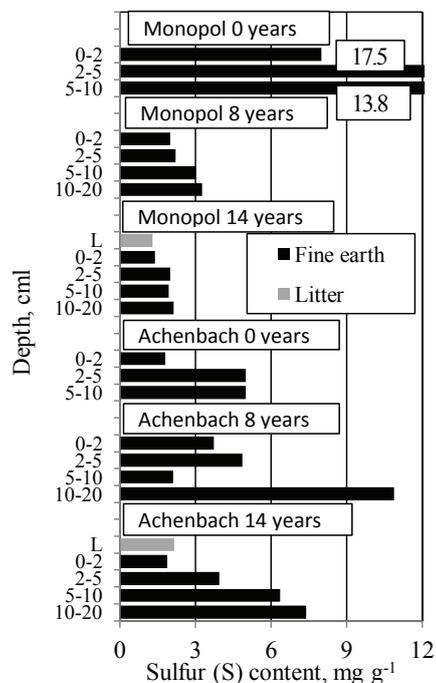


Fig. 12. Depth distribution of sulfur content of the fine earth of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrop, 8 and 14 years past the trial establishment

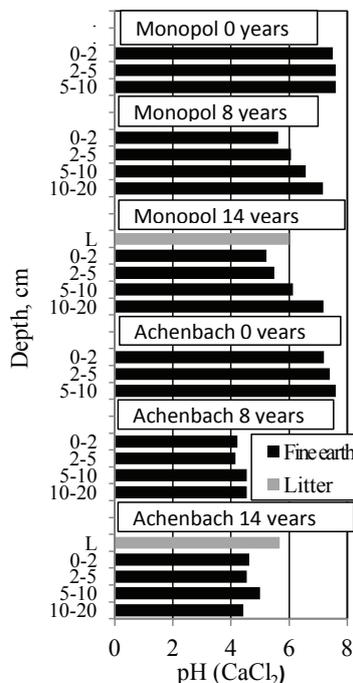


Fig. 13. Depth distribution of the pH(CaCl₂) of the fine earth of the hard coal mining spoil greening variants Monopol and Achenbach spoils of the field trial in Waltrop, 8 and 14 years past the trial establishment

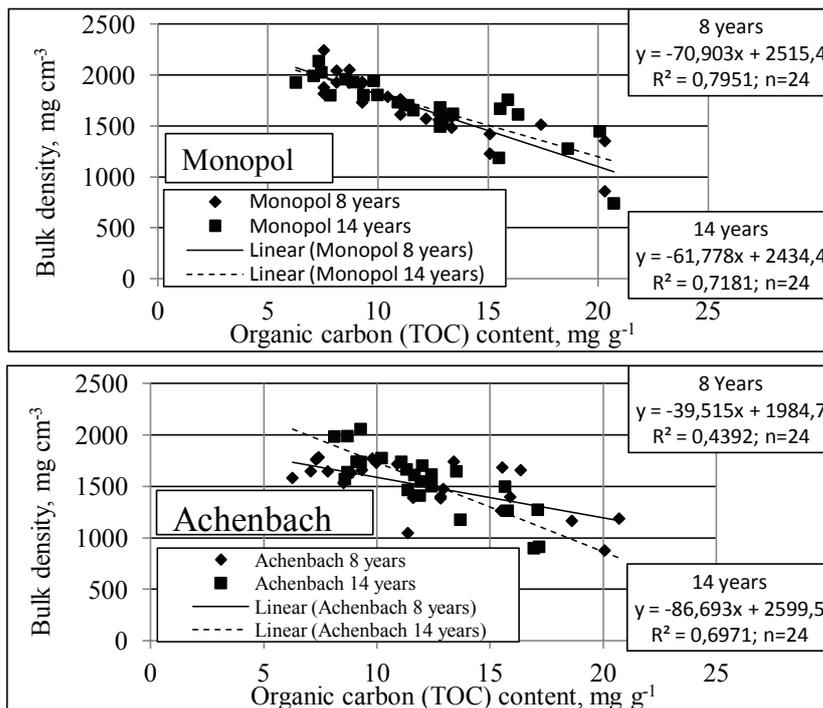


Fig. 14. Correlation of bulk density and of organic carbon (TOC) content of the hard coal mining spoils Monopol and Achenbach

Besides these differences of pH level and depth distribution between the two mine spoils, there were differences in the dependence of pH from the pore volume of the soil samples (Fig. 15). The pH decreased with an increased pore volume of Monopol spoil in both sampling times 8 and 14 years after the trial started. For the Achenbach spoil, this relationship was not ob-

served. The distribution of pore volume with depth was for both spoils and sampling times nearly the same. Different was the increased density of stone-free fine earth, and the higher organic carbon content of the Monopol spoil.

After 14 years of the field trial, the pH in the litter was 6.0 in the Monopol spoil, and 5.7 in the Achenbach spoil (Fig. 13).

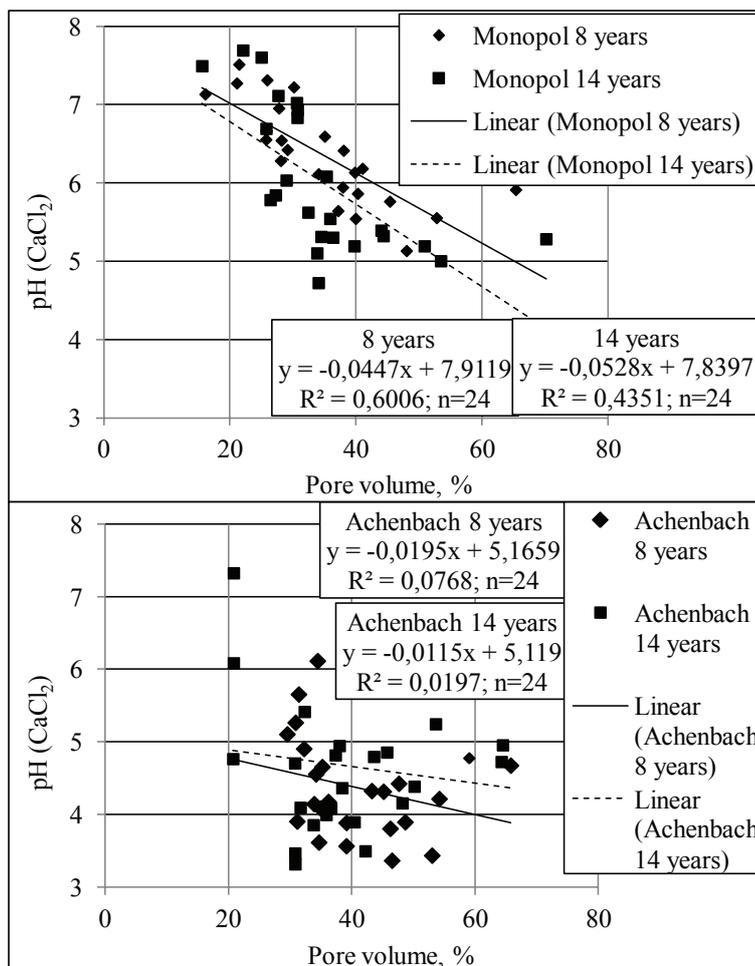


Fig. 15. Correlation of pH to pore volume of the hard coal mining spoil variants Monopol and Achenbach

That means the pH values of the litter were slightly acidic and about 1 step higher than in the following mine spoil layer in 0–2 cm depth.

4. Discussion

The effect of stones in soils on the reduction of soil fertility has been described by Rytter, (2012). Similarly, the change of soil cover of the mining area of Ruhr from silty loam without stones to soils dominated by stones from rock fragments, shifts the soil properties of the region from soil of high fertility farmland (Burghardt et al., 2018) to soil of extremely low fertility. The productive fine earth is now buried under mine spoil. Within the stony mine spoil (Haering et al., 2004; Kompała-Bąba et al., 2019), the content of productive fine earth content is low. Thus the capacity of soils from hard coal mining spoil to store organic carbon, water and plant nutrients is limited.

The landscape from hard coal mining spoil heaps has the character of a mountainous region. The deposited material is from broken and not weathered rocks. As with all surface materials, the fresh mining spoil is subject to soil forming processes by weathering, humus accumulation and water and wind erosion. This way, with time, the new soils of mining spoil will bring

back some low fertility to the area and by this small capacity to fulfill diverse ecological soil functions (Morel et al., 2014).

Stony soils are generated from rocks initially by physical and then by chemical and biological weathering. Stony soils will also develop on fragments of rock fall, and on glacial and fluvial gravel deposits. The hard coal mining spoil bears some resemblance to rock falls according to its edged shape and the fluvial gravel deposits according to its stone size sorting. Besides this, by its geological formation, the hard coal mine spoil has the shape of shale. The shale stones are small when derived from silt and clay sediments, and coarse when derived from sand sediments.

The obtained results show large differences between the two investigated spoils from Monopol and Achenbach mines. The differences concern rock size composition, fine earth content, bulk density, pore volume, TOC and sulfur content, pH, and the distribution of these properties in the investigated soil depth segments, 0–2, 2–5, 5–10 and 10–20 cm. The differences were observed in both sampling times, 8 and 14 years after the field trial was set up.

The skeleton content in the Monopol spoil at all depth segments is nearly equal to 70% per weight at all investigation times. In the Achenbach spoil, the skeleton content increased with depth steadily from 56 to 86 and 88 % from the start of the

field trial to 8 and 14 years past start (Fig. 4). This distribution picture changes distinctly when the results are related to the soil volume (Fig. 5). The bulk density of the skeleton (Fig. 6) of both Monopol and Achenbach spoils increased with depth from about 890 and 680 mg cm⁻³, respectively in 0–2 cm depth, to about 1460 and 1360 mg cm⁻³ past 8 and 14 years in the Monopol spoil, respectively 1410 and 1500 mg cm⁻³ in the Achenbach spoil.

The differences of both spoil variants for the fine earth bulk density are stronger (Fig. 6). They increased with depth from about 430 to 510/560 mg cm⁻³ in the Monopol spoil and decreased from 490/470 to 260/220 mg cm⁻³ in the Achenbach spoil. The differences between Monopol and Achenbach spoils are more considerable when the fine earth content is related to the volume between stones (Fig. 7). It increases in the Monopol spoil from more than 660 to more than 1100 mg cm⁻³, and stayed around 550 to 750 mg cm³ in the Achenbach spoil.

The question is what are the reasons for the differences of fine earth contents and its distribution with increasing depth of the Monopol and Achenbach spoil? Perhaps the answer lies in the organic carbon distribution. The results show a close relationship between TOC content and bulk density of the Monopol samples (Fig. 14). For the Achenbach spoil after 8 years the relationship was weak and increased strongly with time. Thus one can assume that in the early phase of soil development of Monopol spoil, the conditions for intrusion of TOC rich fine earth was given. However, in the Achenbach spoil, the TOC accumulation started later.

Beside the intrusion of organic carbon rich fine earth one can assume a second soil forming factor. The lack of relationship between bulk density of skeleton fraction to the fine earth fraction (Fig. 8) in the Monopol spoil, but a close one for the Achenbach spoil would mean that the mine spoil weathering as a source of fine earth is in the Monopol spoil not a main factor, but it is an important factor for the Achenbach spoil. Late starting TOC accumulation in the Achenbach spoil, let us assume that bioturbation produced the accumulation. In contrast, the intrusion of fine earth plays a dominant role in the soil formation of Monopol soil from the beginning. The two mine spoils seem to represent two different ways of soil formation of stony substrates.

The accumulation of TOC differed between both spoils. Within 20 cm in the Monopol spoil, it was about 1 kg m⁻², and 0.6 kg m⁻² in the Achenbach spoil (Fig. 11). It increased over the period of 8 years to 14 years after the mine spoil deposition by 0.03 and 0.08 kg m⁻². This indicates that the process of soil formation by TOC accumulation within the spoil has nearly finished within 8 years. This means the observed soil development occurred within few years. The formation of an Ah horizon was fast. Similarly, Hearing et al. (2004) reported about Appalachian coal mine soils a development of soils within few years.

How the fast formation of an Ah horizon could occur? The accumulated content of TOC of about 10 mg g⁻¹ (Fig. 10) in fine earth was in the mining spoil about in the same level as in arable land of the region. However, the fine earth volume of mine spoil was only about 20% (Fig. 5) and not 100% as in arable land. That means that a fast accumulation of TOC in the mine spoil oc-

curred due to the low fine earth volume. For the sufficient supply of TOC one can assume that the availability of high amounts of litter from the herbaceous mixture of *Dauco-Mellilotion* community seeding (Jochimsen, 1989, 1996, 2001) was essential.

The TOC accumulated in the total top soil of mine spoil was far below that of the farmland on natural soils of the region which achieves 12 to 14 kg m² within 1 m depth (Burghardt et al., 2018). Thus, hard coal mine spoil landscapes have extremely poor storage capacities for TOC (Fig. 11).

Studies on soil development of hard coal mining spoil are mostly devoted to the content of sulfur compounds Pyrite and Marcasite which developed under reducing conditions in the deep mine environment. These sulfides will oxidize when brought into contact with the atmosphere. The developing sulfuric acid strongly decreases the pH of the soil (Kerth, 1988; Hackelberg et al., 1995; Maiti and Ghose, 2005; Uzarowicz, 2011). Compared to the Achenbach spoil, the sulfur content in the Monopol spoil was high at the start of the field trial but strongly decreased within 8 years in all depths (Fig. 12). In contrast, the sulfur content stayed high in the Achenbach spoil and increased with depth. The pH in the Monopol spoil only lowered to 5.6/5.2 at 0–2 cm depth after 8 and 14 years, respectively and increased with depth to pH 7. The pH values of Achenbach were much lower and stayed between 4.2 and 4.6, and 4.4 and 5.0, respectively after 8 and 14 years of the field trial.

The difference of pH distribution in both spoils (Fig. 13) can be explained by the increased density of stone-free fine earth (Fig. 7), and lowered pore volume (Fig. 5, 15), and the higher TOC of the Monopol spoil compared to Achenbach spoil (Fig. 10). One can assume that the low pore volume and the reduction capacity of organic matter hinder the oxidation of sulfides and thus cause no strong decrease in the pH of Monopol spoil. The effect of organic matter of diminishing the oxidation potential of soils is also known from other soils (Sabien et al., 2010). The higher pH due to higher TOC content will favor plant growth and success of hard coal mining spoil reclamation (Daniels and Zipper, 2018).

Some special methods were used in the investigation of soil properties. To record the soil development, repeated sampling after several years should be undertaken. The sampling 0, 8 and 14 years after the deposition of the hard coal mining spoil showed that already in the first 8 years most of the soil development processes took place. The very differentiated depth segmentation of sampling in 0–2, 2–5, 5–10 and 10–20 cm depth permitted capture detailed the soil forming processes which start at the soil surface.

The content of TOC was determined by the use of H₂O₂ as an agent to destroy humic substances generated during soil formation (Jackson 1958). The organic carbon content strongly influences the bulk density and the exchange capacity of soils. The correlation coefficients gained for the relationship between the bulk density and TOC content (Monopol 8 years: R² = 0.80, Monopol 14 years: R² = 0.72; Achenbach 8 years: R² = 0.44; Achenbach 14 years: R² = 0.70; for each of them: n = 24; old spoil heap > 30 years: R² = 0.85, n = 15), and between the exchange capacity and TOC content of fine earth (R² = 0.75, n = 116) indicate that the method captured the organic carbon.

The achieved TOC content of at least 6 mg g⁻¹ in all inves-

tigated depths to 20 cm below the surface and spoil variants within 8 years characterizes this depth segment as an Ah horizon. The mining spoils had a high stone content. The volume of stones in the spoils was between 40 and 60 Vol. % in 2–20 cm depth, and in 0–2 cm depth 27–28 Vol. %. Thus the soils can be designated as Skeletic. The volume of fine earth is between 8 and 20%. This indicates Leptic properties. The mine spoil substrate is Spolic and thus can be characterized as Technic. As additional important characteristics, the high sulfur content and fine earth intrusion should be considered as Sulfuric and Intrusic.

In the soil classification of Germany (Arbeitsgruppe Boden, 2005) the soils developed on mining spoil are designated as Regosols from hard coal mining spoil substrate. Within US Soil Taxonomy, the soils would belong to the Entisols (Bockheim et al., 2014). The soil classification of the ISSS Working Group WRB (IUSS Working Group WRB, 2015) would position them due to the fine earth content of less than 20% to the Leptosols, with the qualifiers Technic, Skeletic and as additional qualifiers, Sulfuric and for Monopol spoil type Intrusic. Alternative, designations could include classifying the soils as Spolic, Skeletic, Sulfuric, Intrusic (for Monopol spoil type only) Technosols on the basis of WRB classification.

5. Conclusions

Soil formation rapidly takes place in stony substrates of the hard coal mining spoil, in the investigated examples within 8 years. Reasons are the low fine earth content and the accessibility of pore volume between stones. The results show that strong differences exist between mining spoils of different origin. The differences particularly concern the stone content and bulk density of the stone and fine earth fraction, and the depth distribution of these features.

The process of soil formation was not uniform between the two investigated mining spoils. One could distinguish two types of soil formation on stony substrate. In the Monopol spoil, one can assume that the soil formation was dominated by the intrusion of organic matter containing fine earth into the pores between stones. In the Achenbach spoil, weathering of mine stones was the source of fine earth, and organic matter was mixed into the fine earth later perhaps by turbation.

The different soil formation processes result in different amounts of TOC accumulation. The TOC accumulation was within 8 and 14 years with 1 kg m⁻² significant higher in the soils from Monopol mine spoil than with 0.6 kg m⁻² from Achenbach mine spoil. The pH drop with time due to oxidation of sulfides to sulfuric acid was not uniform between both mining spoils. The pH significantly increased with depth in the Monopol mining spoil to neutral values. In the Achenbach spoil, it dropped uniformly to values around pH 4 within 8 and 14 years. In the Monopol mining spoil, an influence of pore volume of the soil occurred. A higher pore volume resulted in a lower pH. Such a correlation did not occur in the Achenbach soil.

Soil formation and development of soil properties can be very diverse in mining spoil. The results show that mining spoil can have very different properties for vegetation growth. Before

starting reclamation and greening projects, the mine spoil properties should be carefully investigated.

The bringing up to the surface of stony spoil from hard coal deep mining causes the landscape to lose its original soil quality. This is particularly the case for the hard coal mining regions from Eastern France and Belgium, via the Ruhr area and southern Poland to Ukraine which natural and farmland areas are from the world most fertile loamy silt soils which developed on loess. The valuable capacities of these soils to store TOC and water, and to produce food and mitigate the climate are lost.

On the other hand soils from hard coal mining spoil can be interesting habitats for vegetation and fauna. The hard coal mining spoil has characteristics of soils of mountain regions. Thus vegetation and fauna of the soils from hard coal mining spoil differ from the original stone free soils from silt loam of the region. The seeding of a seed mixture of Dauco-Melilotion community (Jochimsen, 1989) from mountainous regions supported the establishment of new habitats on the soils of the hard coal mining spoil of the field trial Waltrop. This way the diversity of vegetation and fauna of the region is improved. (Weiss et al., 2005). Particular stony soils from hard coal mining spoil have a high bio-diversity (Woch et al., 2013; Kompała-Bąba et al., 2019). In the present situation, this can be an interesting aspect of the evaluation of mining spoil areas because rural landscapes have become impoverished of plant and animal species. Efforts of investigation of nature and biodiversity of hard coal mining spoil started in the German Federal State North Rhine-Westphalia in the Ruhr area by the project Industriewald – Urban Industrial Woodlands (Weiss et al., 2005). The meaning of the determined differences of the properties of individual mining spoils like Monopol spoil and Achenbach spoil of the field trial Waltrop for the local bio-diversity is until now not known.

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