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V. A. Terekhova, V. V. Demin &
S. Ya. Trofimov**

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Drill cuttings in the environment: possible ways to improve their properties

Ekaterina I. Kovaleva¹ · M. V. Guchok¹ · V. A. Terekhova¹ · V. V. Demin¹ · S. Ya. Trofimov¹

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Abstract

Purpose The disposal of drill cuttings (DCs) in landfills is one of the major impacts of oil extraction on the environment. The DC composition is predetermined by the type of drilling fluid and can include petroleum hydrocarbons (PHCs), salts, and metals. Designing effective approaches to DC recycling is a key problem. We propose the use of certain additives to reduce the toxicity of DC mixtures and improve properties to enable their utilization as artificial soils.

Materials and methods Samples of DCs with different contents of PHCs (from 0 to 50 g kg⁻¹) were taken from landfills and collectors, tested in mixtures with different additives (sand, shungite, diatomite, glauconite, peat, phosphogypsum, and cement). Physical and chemical characteristics of DCs and their mixtures with additives were analyzed. We conducted the leaching experiment by applying amounts of water equal to mean annual precipitation to both pure DCs and their mixtures and analyzed the composition of the leachates. Concentrations of PHCs, chlorides, and metals were measured using gas chromatography, ion liquid chromatography, and inductively coupled plasma mass spectrometry, respectively. We conducted bioassays involving pot experiments with terrestrial plants and aquatic tests on crustaceans and unicellular ciliates to evaluate the toxicity of mixtures.

Results and discussion Sand was the best additive for diluting DCs and improving the structure of mixtures. Phosphogypsum additions caused an intensification of Sr leaching. Cement additions resulted in a dramatic pH increase in mixtures and triggered the formation of soluble Sr and Cu compounds. Additions of 13% peat resulted in the formation of soluble metal-organic complexes, while 5–10% peat additions increased oat and clover biomass. Diatomite was the most efficient additive to absorb PHCs. Mixtures with the PHC concentrations of 1.5–15 g kg⁻¹ caused root and sprout elongation in oat, mustard, and barley. Shungite was efficient in reducing DC contamination levels at pH ≤ 8.5. Paramecium did not show any correlations with contaminants, but the activity of daphnids significantly depended on pH levels and PHC contents.

Conclusions We were able to design soil-like mixtures to be organically integrated into the environment. DCs with PHC contents of ≤ 30 g kg⁻¹ and Cl contents of ≤ 6 g kg⁻¹ could be safely used in mixtures with ≤ 0.7 DC, ≥ 0.2 sand, and ≥ 0.04 diatomite (% v/v) that form soil-like materials. DCs with high HCO₃⁻ and low Cl⁻ contents could be utilized with sand and peat to form fertile horizons of artificial soils.

Keywords Drill cuttings · Artificial soil · Environmental pollution · Plant bioassay · Pot experiments · Toxicity assessment

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✉ Ekaterina I. Kovaleva
ekaterina.kovaleva@soil.msu.ru

¹ Faculty of Soil Science, Lomonosov Moscow State University, Leninskie Gory 1-12, Moscow, Russia 119991

1 Introduction

Increasing demand for energy has led to an unprecedented surge in extracting oil resources worldwide. The drilling industry produces large volumes of oil drill cuttings (DCs) and spent fluids as waste (Kotwica et al. 2015; Huang et al. 2018). The amount of waste from oil well drilling can exceed the drilling cylinder volume by 1.2 times (Calmon et al. 2017). According to estimates of the American Petroleum Institute, the onshore wells in the USA alone produce approximately 150 million barrels of drilling waste per year including 50% solid wastes (Biltayib et al. 2016). This makes the disposal of

generated wastes one of the most pressing current problems (Gutberlet 2015).

Large areas of land resources exploited by the oil industry are owned by indigenous people, from Canada to Peru in the America, from Nigeria to Chad in Africa, and from Northwestern to Far Eastern Russia in Asia (Dana et al. 2008; Yakovleva 2014). Thus, oil and gas projects inevitably influence the well-being of the indigenous peoples (Sawyer and Gomez 2008; Behrends and Schareika 2010) which makes the task of decreasing the environmental impacts and the sustainable development of these areas an important one.

The physical and chemical properties of DCs are highly diverse as they depend on the type of drilling fluid, the local lithology of rock layers, the location of an oil well, and the drilling technology (Gonet 2006). The well drilling wastes can be liquid (drilling fluids or mud) or solid (rock debris) (Onwukw and Nwakaudu 2012), but most frequently they are colloidal suspensions, polymer-mineral micro-composites of diverse chemical and mineral phase compositions (Fijal et al. 2008). Various additives are used to improve the technical performance (Kotwica et al. 2015) in different drilling operations and in the course of drilling itself (Bakke et al. 2013) such as viscosifiers (e.g., polyacrylates and other organic polymers), emulsifiers (e.g., alkyl acrylate sulfonate and polyethylene oxide), pH and shale control agents, and deflocculants.

Drill cuttings are heterogeneous soil-like wastes (EU Commission Decision's 2001) that can be highly hazardous to the ecosystem if disposed of without appropriate treatment, including the removal of contaminants (Ismail et al. 2007; Ghasemi et al. 2017). The drilling industry frequently uses sustainable drilling fluid components. These components include the readily mineralizable organic admixtures for gelification and lubrication based on cellulose, starch, polyacrylamide, glycols, graphitic materials, clay (bentonite), or carbonate (Smagin et al. 2011).

Alleviation of the toxicity and harmful effects of DCs has attracted considerable attention, especially in the light of the sustainable development concept. The treatment methods include the stabilization/solidification process, bioremediation, the thermal desorption method, and vitrification/devitrification techniques (Leonard and Stegemann 2010; Opete et al. 2010; Ghasemi et al. 2017). Currently, drilling wastes are commonly treated with coagulants and flocculants and then buried further on site or introduced into landfills (Maloney and Yoxtheimer 2012). DC treatment at high temperatures (up to 500 °C) helps to remove organic compounds from solid wastes (Geehan et al. 2007). Thermal methods are highly efficient but can be expensive and, therefore, require further development.

DCs consist of stable materials such as shale, quartz, sand, and limestone, which possess good physical and mechanical properties in bulk material (Huang et al. 2018). Composition

of DCs is generally similar to the earth's crust, but certain properties do not allow their sustainable recycling (Kovaleva et al. 2019). They are (specific) structureless finely dispersed systems and can be defined as soil-like bodies (Smagin et al. 2011). According to the world reference base (FAO-ISSS-ISRIC 2015), a DC deposit can be classified as a Technosol, as it is similar to artificial soils formed from wastes (e.g., landfills, sludge, mine spoils). Such Technosol contains toxic chemical compounds such as oil, chlorides, and heavy metals and has a low infiltration capacity, but a high swelling capacity (due to significant contents of swelling clays) upon moistening.

Hence, introducing DCs into terrestrial ecosystems is possible only after appropriate treatment with various additives that should (1) alter their texture and hydrophysical properties and dilute them in order to reduce concentrations of contaminants, (2) improve their toxicant-binding properties such as sorption, and (3) alter their pH in order to reduce the mobility of heavy metals. The main goal of altering DC characteristics is to reduce their toxicity and transform them into a sustainable artificial soil that could be introduced into the ecosystems. This soil-like material should be capable of performing its ecological functions including the subsequent formation of soil horizons.

This artificial soil lacks genetic horizons, and therefore, it cannot be considered as a proper soil according to Dokuchaev's concept (Gerasimova et al. 2003), because genetic horizons can only develop over significant time periods (Targulian and Arnold 2008). However, this artificial soil will immediately be capable of performing its vital ecological functions just like a natural soil, which corresponds to the latest concept of Technosols (FAO-ISSS-ISRIC 2015). The subsequent pedogenic alteration of artificial soil-like materials may result in the formation of a cambic horizon.

The aims of the present work were to improve the physical and chemical characteristics of various types of DCs by mixing them with organic and mineral additives and to propose certain DC mixtures for the most sustainable and environmentally friendly use as artificial soils. This research was focused on assessments of the effects of various additives used in DC mixtures, in particular, the decrease in toxicity levels, which were tested using bioassay endpoints and pot experiments.

2 Materials and methods

2.1 Samples of drill cuttings and additives

Drill cuttings were sampled in oil fields of the Tyumen region, West Siberia taiga zone, Russia. Four types of DC (I, II, III, and IV), which differed by their disposal locations and applied drill fluids, were collected using an Edelman's drill and a

shovel. DC I was taken from DC landfills and DCs II, III, and IV from DC collectors of the drilling rig extension line. DCs I, II, and III were generated with oil-based drilling fluids and DC IV with a water-based fluids and additions of silicate clay (drilling mud). The analyzed DC types differed in pH and contents of PHCs, metals (K, Na, Zn, V, and Sr), and water-soluble salts including chlorides (Table 1).

We tested the ability of certain additives (sand, peat, diatomite, shungite, glauconite, phosphogypsum, and cement) to reduce the toxicity and to improve the properties of the DCs. Sand with a grain size of up to 4 mm was obtained from a quarry within an oil field area, pre-washed, and ignited to avoid the influence of additional contaminants. Peat, which had a pseudo-fibrous condition, pH 5.6, and ash content of 14.3%, was obtained from a quarry in a eutrophic peatland within an oil field area.

Diatomite is a naturally occurring rock consisting of fossilized remains of diatoms and characterized by a porous

structure, a low density, and a large surface area. Shungite is a non-crystalline mineraloid of abiogenic origin that consists mostly of carbon. Glauconite is an iron potassium phyllosilicate (mica group) mineral. The diatomite and glauconite were used in a powder form, while shungite was used in a granular form with grain sizes of ≤ 2 mm. Phosphogypsum is a waste of production of phosphate fertilizers with the free phosphoric acid content of 1% and the Sr content of 15.4 g kg^{-1} .

The samples of DC, phosphogypsum, and peat were air-dried, grinded, and sieved through a 2-mm mesh to reach a homogeneous texture prior to their use in experimental mixtures with DCs (Table 2).

2.2 Experimental design

We made several DC-based mixtures with additions of sand, peat, phosphogypsum, cement, diatomite, shungite, and

Table 1 Chemical properties of drill cuttings. The data is represent mean $\pm$ SD from twenty samples

DCs	I		II		III		IV		Soil, median/Bowen (1979)	Earth's crust/Bowen (1979)
Parameters	Mean	SD	Mean	SD	Mean	SD	Mean	SD		
pH	10.9	0.1	10.3	0.1	8.3	0.1	9.3	0.3	–	
g kg^{-1}										
PHC	9.9	1.2	5.6	0.9	21.8	5.4	1.6	0.5		
Cl^-	62.1	13.2	25.4	9.6	14.9	6.7	0.5	0.1		
HCO_3^-	0.76	0.07	0.02	0.01	0.04	0.02	1.14	0.35		
SO_4^{2-}	2.81	0.94	1.23	0.50	0.41	0.12	0.53	0.35		
Al	41.4	10.8	31.5	8.2	38.4	10.0	36.6	14.7	71	82
Ba	2.99	0.89	2.86	0.86	0.13	0.04	0.29	0.08	0.5	0.5
Ca	73.3	21.9	79.6	23.9	63.7	19.1	22.1	12.6	15	41
Fe	33.9	9.5	33.1	9.3	30.9	8.7	34.4	9.8	40	41
K	37.2	14.9	37.2	14.9	45.7	18.3	7.6	3.3	14	21
Mg	12.6	3.8	9.5	2.8	4.3	1.3	7.2	4.5	5	23
Na	34.6	13.8	3.4	1.4	7.6	3.0	3.4	0.5	5	23
Si	256	52	273	71	269	70	285	99	330	277
mg kg^{-1}										
As	6.5	3.3	7.6	3.8	7.5	3.7	8.4	2.7	6	1.5
Cd	–	–	0.61	0.33	0.22	0.13	0.19	0.02	0.35	0.11
Co	–	–	9.7	3.9	7.6	3.0	12.5	1.9	8	20
Cr	21.9	4.4	46.8	9.5	81.5	16.3	57.5	13.6	70	100
Cu	36.0	7.2	33.9	6.9	36.7	7.4	34.1	9.7	30	50
Mn	509	153	512	154	554	16	735	272	1000	950
Mo	–	–	7.0	2.8	2.3	0.9	1.9	0.5	1.2	1.5
Ni	26.5	9.3	29.5	10.3	36.1	12.6	35.0	7.1	50	80
Pb	24.1	4.6	20.4	5.1	39.1	9.8	55.2	32.5	35	14
Sr	323.2	97.9	16.5	4.9	18.4	5.5	121.3	19.1	250	370
V	235.6	59.2	47.8	12.6	20.8	5.2	85.7	25.8	90	160
Zn	272.7	54.7	94.2	18.9	220.0	44.1	104.2	61.5	90	75

Table 2 Components content in mixtures, % $\%_{\text{v}}$ of DC

Type of DC	DC	Sand	Shungite	Diatomite	Glauconite	Phosphogypsum	Cement	Peat
I	50–70	20–42	3–7	0	0	1–5	0	0
II	60–70	18–33	0	6–10	0	0	1–2	0
III	60–70	21–36	0	2–4	0	0–2	0	0–13
IV	65–70	25–67	0–5	0	0–2	0	0	0–10

glauconite. The rates of additive application were calculated based on the DC characteristics (Table 1), properties of the additives, our previous research (Kovaleva et al.), and also the cost of the additives (because an artificial soil has to be cost-effective).

We used sand in DC-based mixtures to improve the hydrophysical properties of the so-called constructed artificial soils and also to reduce concentrations of certain possibly harmful components by diluting. To reduce the mobility of contaminants in DC mixtures, both carbon sorbents (shungite and peat) and mineral sorbents (glauconite and diatomite) were applied. Phosphogypsum was chosen to neutralize the high alkalinity of DCs, while cement was used as a binding material. Finally, we designed 109 variants of DC-based mixtures to test in experiments (Tables 2 and SA I).

We designed a leaching experiment with DC-based mixtures, which was aimed at assessing the rates of leaching of chemical compounds from the mixtures into filtrates (leachates). A 1.0-kg sample of each mixture prepared from air-dried DCs and additives was put into a nylon ($50 \times 50 \mu\text{m}$ mesh) bag and then placed in a 3.5-L glass container, through which we gradually percolated a water volume of 4.5 L that was equivalent to the mean annual precipitation of 547 mm within the study area (Shah et al. 2015) spread over the 8100 mm² area of the sample.

2.3 Chemical analysis

In the analyzed solid samples (DCs and their mixtures), we measured pH values and the contents of PHCs, Cl^- , HCO_3^{2-} , SO_4^{2-} , HPO_4^{2-} , Al, Ba, Ca, Fe, K, Mg, Na, Si, As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sr, V, and Zn. In the filtrates collected in the leaching experiment, we analyzed pH and contents of PHCs, Cl^- , Na, K, Ba, Sr, Cu, and Ni.

First, PHCs were extracted from solid samples with chloroform, and then the filtered solutions were distilled using the solvent under vacuum. The PHCs were re-extracted with n-hexane and passed through an adsorption column with aluminum oxide and the amount was evaluated by an Agilent 6890N gas chromatograph (USA) with a flame ionization detector, ISO 16703 (ISO 2004). We used a DB-5ms column ($30 \text{ m} \times 0.25 \text{ mm} \times 0.25 \mu\text{m}$), helium, and a gas carrier, with the flow rate of 1 mL min^{-1} and the temperature gradient from

60 to 300 °C. We analyzed the results using the HP-Chem data processing system.

The total contents of metals in solid samples and filtrates were estimated by the use of inductively coupled plasma mass spectrometry (ICP-MS) using an Agilent 7500a gas chromatograph (USA). For a pretreatment, a 0.5-g solid sample was sieved through a 1-mm mesh and dissolved by a microwave acid digestion with 12 mL aqua regia. The digested sample was transferred into a 100-mL flask, and the final volume was adjusted to 100 mL by deionized water.

The Cl^- , HCO_3^- , and SO_4^{2-} contents in filtrates and water extracts from solid samples (with a solid sample to water ratio of 1:5, the samples were filtered through 0.45- μm sieve) were analyzed using an anion liquid chromatograph with an ICS-2000 conductometric detector (Dionex, USA). The pH was determined using an ion meter HI 8314 (Hanna Instruments, USA). DCs and the mixtures were incubated in an oven at 105 °C for 24 h, according to ASTM D2216 (Annual Book of ASTM Standards 1985) to measure the moisture content. The amount of solid residue in filtrates and water samples extracted from solid samples was determined according GOST 26423–85 1985.

2.4 Bioassay procedures

2.4.1 Phytotoxicity of leachates

We assessed the phytotoxicity of leachates, obtained from the leaching experiment, to test the influences of different additives and their proportions on the quality of constructed mixtures.

Higher plant bioassays were performed according to ISO 18763:2016. The seed germination rate and the length of seedling roots and sprouts were determined in experiments with barley (*Hordeum vulgare*), oat (*Avena sativa*), and mustard (*Sinapis alba*). Some of these test cultures are included in the lists of cultures recommended for bioassays published in the guidelines of several regulatory organizations (OECD 2006; US EPA 2012). Each unit of sterilized plastic dishes was filled with the filtrate (leachate) and covered with a filter paper sheet. A total 10 seeds of each of the tested plants were placed on the surface of a paper into individual dishes and incubated for 7 days at 25 °C in a thermostat. After the

incubation, the number of germinated seeds was counted. The root and sprout length were measured manually. Seed germination as a percentage was calculated according to the validity criteria for the higher plant bioassay (OECD 2006). Test validity criterion was fulfilled in 100% of germinated plants in the control test. Sample toxicity was calculated according to the equation $[(A - B)/A] \times 100$, where A was the analyzed parameter in the control sample and B was the analyzed parameter in the mixture sample.

2.4.2 Aquatic bioassay of drill cuttings, mixtures, and leachates

We assessed toxicity levels of the DCs, DC-based mixtures, and their leachates using tests on hydrobionts that could reflect possible impacts of constructed mixtures on aqueous medium, bearing in mind that those mixtures were designed for integration into the environment as artificial soils. The aquatic bioassay involved the use of two test species—crustaceans *Daphnia magna* Straus and infusorians *Paramecium caudatum* Ehr. The crustaceans were not adapted to highly mineralized (i.e., saline) waters of certain leachates, according to ISO 6341:2012. We did not use other test cultures resistant to high mineralization because the mixtures are designed as artificial soils to be used within the zone of peat and podzol soils. We assessed the effects of chemicals on *D. magna* exposed for 96 h (ISO 6341:2012) and *P. caudatum* for 24 h (FER 2006) compared with a control. The water extraction from the samples was carried out at a liquid:solid ratio of 4:1. The toxicity effect of the water extracts was compared with a toxicant-free control sample (the normal medium, without EDTA) to calculate the index of toxicity (IT, %) by the following equation: $IT (\%) = (1 - (N_{\text{sample}}/N_{\text{control}})) \times 100\%$, where N_{sample} is the number of viable specimens in the samples and N_{control} is the number of viable specimens in a toxicant-free control (the normal medium, without EDTA). We interpreted results using the following thresholds: IT of less than 10% corresponded to non-toxic samples, IT between 10 and 50% to samples with a low toxicity, IT between 50 and near 100% to toxic samples, and IT of 100% to highly toxic samples.

2.5 Pot experiments

We carried out the pot experiments using two species of higher plants—oat (*Avéna sativa* L.) and clover (*Trifolium repens* L.)—in order to evaluate their abilities to grow in artificial mixtures constructed from DCs II, III, and IV and the additives of sand, peat, and shungite. A 1.0-kg sample of each mixture was air-dried, sieved through a 2-mm sieve, and placed into a 1.5-L plastic pot. Deionized water was added to experimental pots by drops until its volume reached 60 W% of the moisture capacity of the mixture. Seeds were placed on

the surface of an evenly moistened mixture at an equal distance from each other. Five oat seeds or ten clover seeds per pot were planted taking into account the differences in seed sizes. The rate and timing of watering were determined visually by the state of the soil-like mixtures. We evaluated toxicity of the DC-based mixtures compared with control (sand) by assessing the biomass growth of the oats and clover.

2.6 Data analysis

All experiments were performed using 3 replications per sample. Standard deviations and correlations were calculated using the STATISTICA software package version 8 (StatSoft Inc.).

3 Results

3.1 DC properties

The chemical properties of DCs are essential for considering their possible use as parent materials. It is important to identify the negative effects of their components, which originate from sorbents and ameliorants used in drilling fluids. To interpret the results of our assessments, we compared the elemental composition of DCs with the data on mean concentrations of elements in the Earth's crust and soils (Bowen 1979).

The key characteristics of the studied DCs are listed in Table 1. In DC I, the K concentration exceeded those in the Earth's crust and soils by 1.8 and 2.7 times, respectively, and the Na concentration by 1.5 and 6.9 times, respectively. Concentrations of Ca in DCs I, II, and III were also excessively high (from 63.7 ± 19.1 to 79.62 ± 23.88 g kg⁻¹) compared with those in the Earth's crust (41.00 g kg⁻¹) and soils (15.00 g kg⁻¹). However, the Ca concentration in DC VI (22.05 ± 12.58 g kg⁻¹) was close to its level in the Earth's crust. Concentrations of Mg were 1.9–5.3 times lower than that in the Earth's crust, but 1.4–2.5 times higher than that in soils in all DCs except for DC III, where the Mg concentration was similar to that in soils.

Concentrations of Cd, Cr, Mo, Ni, Pb, Zn, Cu, Sr, Ba, Mn, and V in DC samples did not exceed 0.5%, but they all were significantly higher than the mean concentrations of those elements in the Earth's crust and soils. The most significant deviations from mean concentrations were manifested, e.g., by Zn levels in DCs I, II, and III and Pb, Zn, and As levels in DC IV. The highest Ba concentration was found in DC I, where it was up to 6 times higher than those in the Earth's crust and soils. The DC IV samples were characterized by low levels of Ba and SO₄²⁻. Contents of Mn and Sr in all DC samples, except for DC I, were similar to those in the Earth's crust and soils. Different types of DCs had various contents of V, which reached its highest value in DC I and

exceeded its mean concentration in soils by 2.6 times. The pH values of DC samples were between 8.2 and 11.0. The PHC mass fraction equaled to 1.1–27.0 g kg⁻¹. The composition and features of DC were similar to those reported by Tuncan et al. (2000).

Chloride concentrations in the analyzed DCs varied considerably, with the highest values detected in DCs I, II and III and the lowest in DC IV. The concentrations of SO₄²⁻ varied from 0.41 ± 0.12 (DC III) to 2.81 ± 0.94 g kg⁻¹ (DC I). The HCO₃⁻ ion levels were less significantly increased than the levels of Cl⁻ and SO₄²⁻ in DCs I, II, and III, while DC IV contained a considerable amount of HCO₃⁻ ion (1.14 ± 0.35 g kg⁻¹).

3.2 Effects of additives on chemical properties of leachates from DC mixtures

All leachates (filtrates) from DC I and its mixtures were characterized by nearly neutral pH values (7.1 ± 0.2) and PHC contents from 0.28 ± 0.08 to 18.76 ± 3.28 g L⁻¹ (Table 3). The addition of (35–40%) sand and shungite to the DC I sample, which was characterized by a total salt content of 26 g per liter of its leachate, had resulted in a significant decrease in the concentration of salts including chlorides, i.e., total salt contents in 3, 4, and 5% shungite mixtures were reduced by 1.4, 1.8, and 3 times, respectively (Fig. 1a). The effectiveness of shungite as a PHC absorbent depended on the proportion of shungite in the mixtures (Fig. 1b). For example, PHC concentrations in leachates from 7% shungite mixtures were reduced by 4 times, even in the case where the mixture had a PHC content of 40 g/kg. The total contents of Ba, K, and Sr were significantly influenced by shungite additions to DC I at all experimental rates (from 3 to 7% of the mixture) and concentrations of those elements decreased by 3–7 times in

leachates from the shungite mixtures in comparison with the leachate from pure DC I (Fig. 1c, d).

The addition of 2% cement to the DC II-based mixture containing varied proportions of sand and diatomite caused a rise in leachate pH values that reached 12.4 ± 0.2 (Fig. 2a). The addition of 1% phosphogypsum or 5–13% peat to the DC III-based mixtures without cement shifted the pH of leachates to the near-neutral values (Fig. 2a), with the lowest pH value found in the mixture supplemented with 13% peat.

The solid residue content in filtrates from DC II varied within the range of 15–18 g L⁻¹, one-third of which was represented by Cl content (4.5–6 g L⁻¹) (Fig. 2b). Solid residue contents were relatively lower in filtrates from the sand DC II mixtures. Similar effects of sand additions were observed in DC III-based mixtures.

Cement additions to DC II-based mixtures induced an increase in Cu concentrations in the filtrates by up to 5 times compared to the filtrate from pure DC II (Fig. 2d). It should be taken into account that the mixtures were diluted with sand and diatomite used as sorbents. The Sr concentrations in filtrates from the mixtures were 3 times higher than that in pure DC II filtrate, even in the case of a DC II mixture double-diluted with sand and diatomite (Fig. 2c).

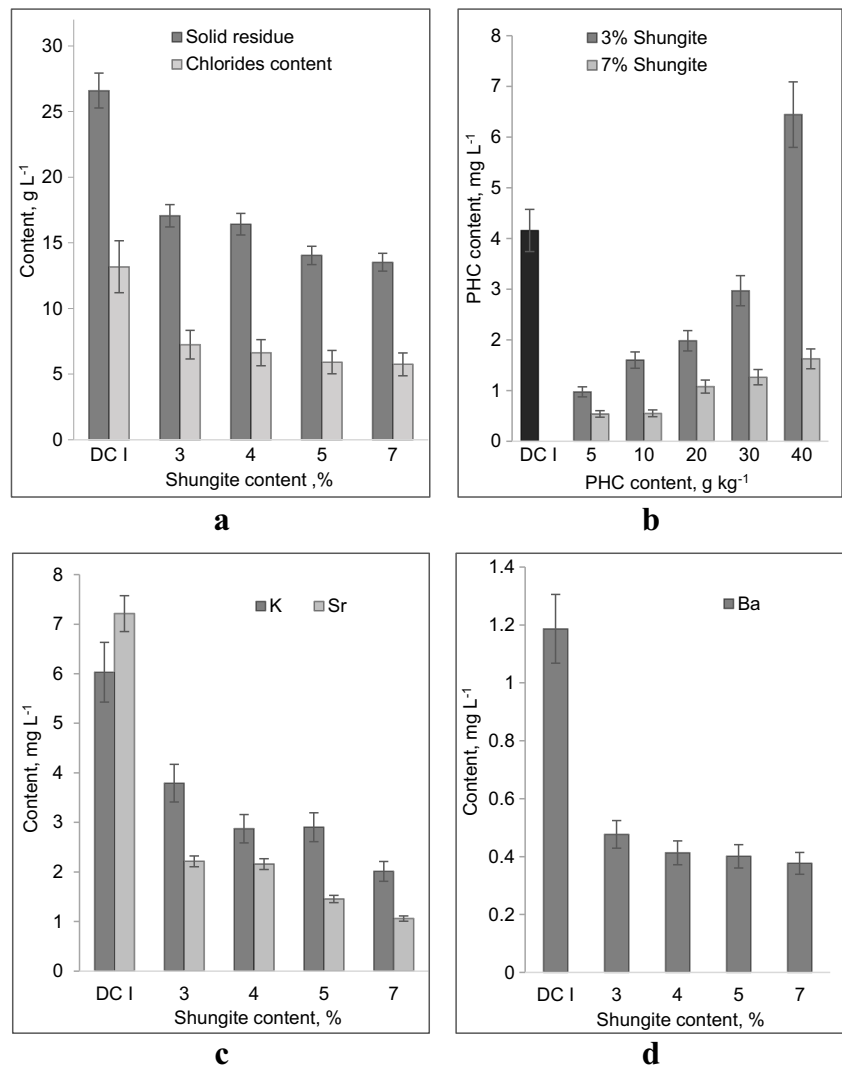
Additions of diatomite to mixtures based on DCs II and III had resulted in reductions of PHC concentrations in their leachates. For example, supplementing a DC III mixture (containing up to 50 g PHC per kg⁻¹ of solid sample) with 2% and 4% diatomite decreased PHC levels in filtrates by 1.5–4 times (Fig. 2e). Adding 13% peat to DC II-based mixtures with the same PHC concentration considerably increased PHC yields in leachates (Fig. 2f).

Supplementing the mixtures containing 65–70% DC IV and 30–35% sand with shungite/glaucinite (Fig. 3a) did not change the pH (8.1 ± 0.3). The filtrate from pure DC IV had the salt content of 2.8 ± 0.5 L⁻¹, which was at least twice as

Table 3 Characteristics of filtrates from the DC samples

Parameters	DC I		DC II		DC III		DC IV	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
pH	7.1	0.2	7.6	0.3	6.9	0.1	8.1	0.3
PHC, mg L ⁻¹	4.16	0.42	3.35	0.87	18.76	3.28	1.45	0.08
chlorides, g L ⁻¹	13.17	2.45	6.85	0.29	6.89	0.45	0.22	0.09
Na, g L ⁻¹	4.58	0.90	0.51	0.09	2.07	0.62	0.67	0.08
K, g L ⁻¹	6.03	0.58	6.76	0.75	3.25	0.25	0.02	0.01
Ba, mg L ⁻¹	1.19	0.14	2.05	0.18	0.79	0.11	0.45	0.09
Sr, mg L ⁻¹	7.21	1.21	0.82	0.12	4.01	0.39	1.19	0.82
Cr, mg L ⁻¹	2.21	0.65	0.07	0.02	3.22	1.43	8.72	1.29
Cu, mg L ⁻¹	18.31	1.24	6.03	0.95	19.78	7.41	16.60	6.81
Ni, mg L ⁻¹	6.73	0.98	15.11	3.46	22.32	7.22	3.12	0.71

Fig. 1 Characteristics of filtrates from DC I with 3, 4, 5, or 7% shungite additive. **a** Solid residue and chlorides content, g L^{-1} . **b** PHC content in filtrates from mixtures with 70% DC I with 3 or 7% shungite; PHC 5–40 g kg^{-1} content. **c**, **d** K, Sr, and Ba content in filtrates from DC I with 3, 4, 5, or 7% shungite



high as those in filtrates from DC IV–based mixtures (Fig. 3b), but significantly lower than those in filtrates from pure DCs I, II, and III. The PHC content increased in filtrates with an increasing share of DC IV containing 1.6 g kg^{-1} PHC (Fig. 3c).

The lowest PHC concentration (0.3 g L^{-1}) was observed in the leachate from a mixture, which consisted of 65% DC IV, 35% sand, and 2–5% shungite. Sr, Zn, Mn, and Cu levels in the leachate were reduced by up to 3, 50, 110, and 6 times, respectively, due to the effects of shungite and glauconite additives.

3.3 Bioassay endpoints and their correlations with contaminant concentrations

3.3.1 Phytotoxicity of leachates

All leachates (filtrates) obtained from DC I and its mixtures demonstrated phytotoxicity, according to the bioassays using

barley, oat, and mustard. The solid residue content in filtrates exceeded 6 g L^{-1} . All species performed identically during the tests and their germination energy of seeds was equal to 0.

Supplementation of DC III with sand and combination of additives (diatomite, phosphogypsum or peat, cement) stimulated the growth of oat sprouts and roots by 3 times and 1.7 times, respectively, and the growth of barley sprouts and roots by 1.5 times and 1.4 times, respectively. The growth of mustard sprouts was enhanced by up to 5 times; however, the root growth was significantly inhibited with the decrease of diatomite share and exclusion of peat in mixture (Fig. 4a, b). The shortest sprouts and roots were found in a pure DC sample. There was a significant correlation between the oat root length, the PHC concentration, and K content ($R=0.97$ and $R=0.98$, respectively, $p<0.05$).

The DC IV–based mixtures containing 0–30 g kg^{-1} PHC were shown to trigger both oat and mustard root elongation, if they were supplemented with sand at a ratio 70:30 or 65:30% by 14 and 28%, respectively, in comparison with DC IV

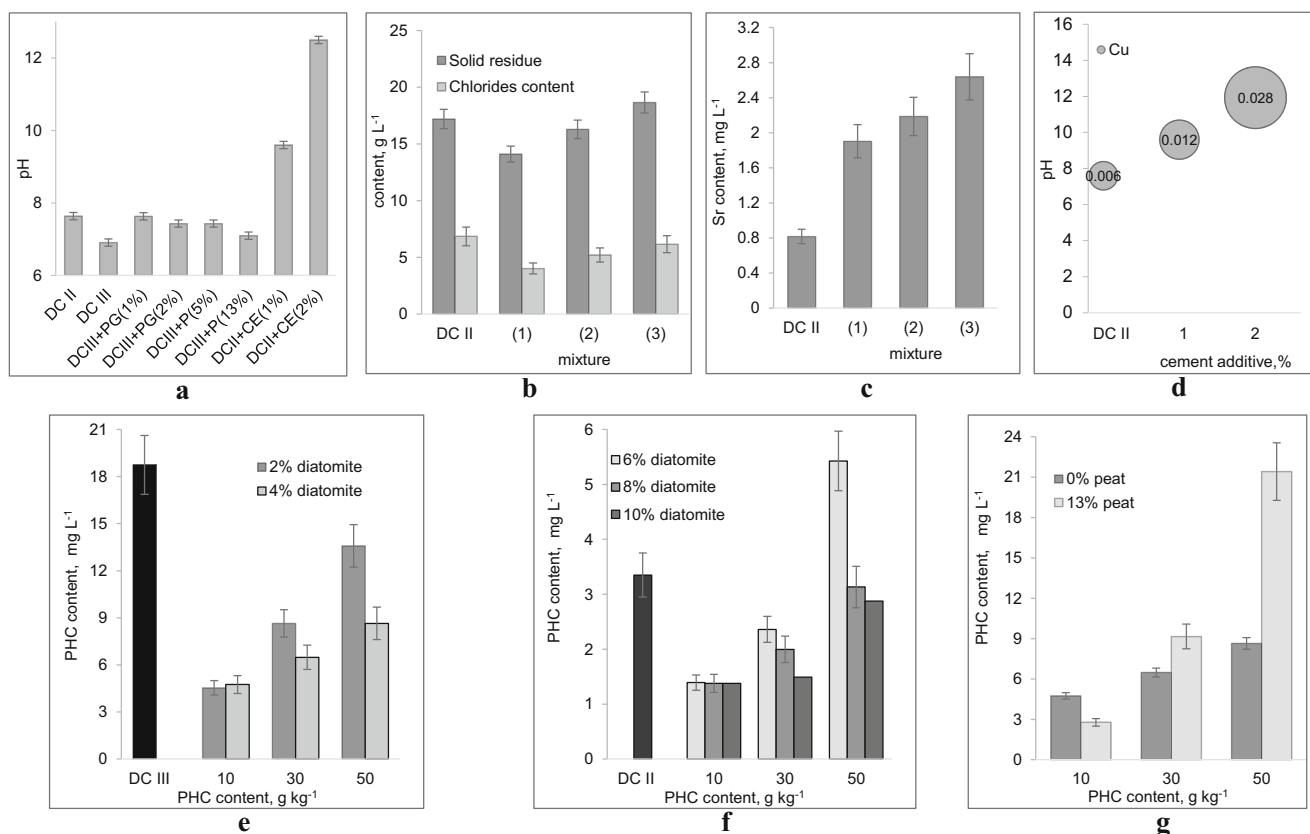


Fig. 2 Effect of additives on filtrate characteristics. **a** pH change in mixtures from DC II and DC III: 60% DC, 27–39% sand (S), phosphogypsum (PG) (1; 2%), peat (P) (5; 13%), and cement (CE) (1; 2%). **b** DC II—solid residue content and Cl content, mg L^{-1} in DC II and mixtures (1, 2, and 3). **c** Sr content in DC II and mixtures (1, 2, and 3), mg L^{-1} . **d** Cu content (mg L^{-1}) in DC II and mixtures with 1–2% cement (CE). **e** PHC content in DC III and mixtures: 60% DC III, 34–36% sand (S), 2–4% diatomite (DE), and 2% phosphogypsum (PG). **f** PHC content

in DCII and mixtures: 60% DC II, 30–34% sand (S), and 6–10% diatomite (DE). **g** PHC content in mixtures: 60% DC III, 21–34% sand (S), 4% diatomite (DE), 0–13% peat (P), and 2% phosphogypsum (PG). Mixtures: (1) 60% DC, 33% sand (S), 6% diatomite (DE), and 1% cement (CE); (2) 60% DC, 31% sand (S), 8% diatomite (DE), and 1% cement (CE); (3) 60% DC, 29% sand (S), 10% diatomite (DE), and 1% cement (CE)

filtrate (Fig. 4e, f). However, we did not find any significant changes in oat and mustard sprout elongation for most filtrates of mixtures in comparison with DC IV filtrate (Fig. 4c, d).

3.3.2 The bioassay for sensitive crustaceans and infusorians

Determining the inhibition of crustaceans *Daphnia magna* and unicellular ciliates *Paramecium caudatum* mobility is an acute toxicity assay. We evaluated the initial concentration of contaminants in aqueous extracts that may immobilize 10% of the *D. magna* compared with the control.

Bioassay of the DC, mixtures and filtrates, obtained in the leaching experiment All filtrates, obtained from the experiment with DC I and its mixtures, as well as solid samples were extremely toxic ($\text{IT} = 100\%$). The toxicity was caused by the transfer of salts (solid residue content $> 6 \text{ g L}^{-1}$) from solid samples into water extracts and filtrates. The toxicity of the filtrates did not depend on PHC concentration ($0\text{--}50 \text{ g kg}^{-1}$ of a solid sample) or pH (equal to ~ 7.0).

Water extracts from DC (III) and its mixtures, treated with diatomite, phosphogypsum, or peat, also exhibited toxicity in *Daphnia magna* as well in *Paramecium caudatum* ($\text{IT} < 10\%$). Only the filtrates of mixtures in proportions of $< 60\%$ DC III, $\geq 35\%$ sand, 2–4% diatomite, $< 0.5\%$ phosphogypsum ($\text{v/v} \%$), and $\leq 6 \text{ g kg}^{-1}$ of solid residue and 50% chlorides were non-toxic to hydrobionts. *P. caudatum* mortality depended on the solid residue content as shown in Fig. 5a. The filtrates, obtained in the experiment with mixtures (prepared from 70% DC IV; 30% sand, shungite, and oil in doses of 0, 0.5, 1.5, 5.0, 15.0, and 30.0 g kg^{-1} ; as well as water extracts from pure DC IV and mixtures), did not exhibit any toxicity for *Daphnia magna* Straus and *Paramecium caudatum* Ehr. The mortality of hydrobionts did not exceed $10.0 \pm 4.0\%$ and $6.0 \pm 4.3\%$, respectively, compared with the control values (Fig. 5b). The maximum effect was determined in filtrate, containing $30 \text{ g PHC per kg}^{-1}$ in a solid phase. Low concentrations of salts composed predominantly (59–72%) of hydrocarbonates were non-toxic for both hydrobionts. The presence of chlorides accounted for the 100% toxicity of

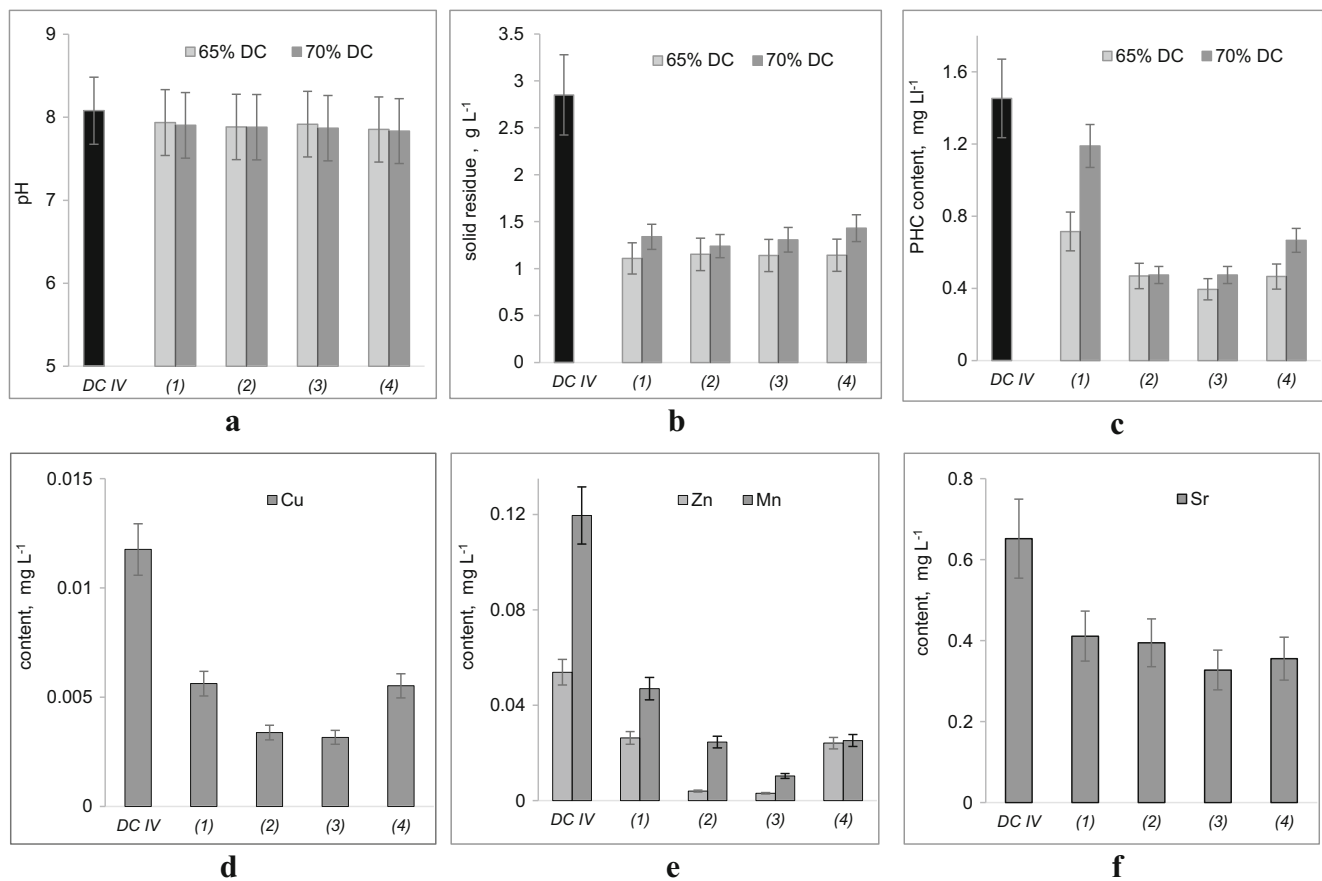


Fig. 3 Effect of shungite and glauconite additives on filtrates' characteristics. **a** pH. **b** Solid residue, g L⁻¹. **c** PHC content, mg L⁻¹. **d** Cu content, mg L⁻¹. **e** Zn and Mn content, mg L⁻¹. **f** Sr content, mg L⁻¹. Mixtures: (1) DC IV 65 or 70% and sand 30 or 35%, respectively; (2) DC

IV 65 or 70%, sand 28 or 33%, respectively, and shungite 2%; (3) DC IV 65 or 70%, sand 25 or 30%, respectively, and shungite 5%; (4) DC IV 65 or 70%, sand 28 or 33%, respectively, and glauconite 2%

leachates from DC I and its mixtures. Toxic effects on ciliates did not correlate with any contaminant concentrations (Table 2), but the mortality of daphnids correlated strongly with PHC contents in leachates ($R = 0.73$, $p < 0.01$).

3.3.3 Pot experiments

The DCs and their mixtures with solid residue contents of more than 3 g kg⁻¹ were 100% toxic to all tested plant cultures. We observed a clear elongation of oat plants, growing in mixtures, containing 30–40% DC IV and 70–40% sand, in comparison with pure DC IV. A significant difference was found for the mixture, containing 50% DC IV:50% sand (Fig. 6a). The largest toxic effect for oat growth was for the mixtures with 60% DC IV share and also for the same mixture but with shungite added. Mixtures supplemented with peat demonstrated the most pronounced positive effect on plant length, especially when it contained 50% DC IV.

The dry biomass indices in oats and clover were more sensitive to the presence of contaminants than the growth parameter. The toxicity did not correlate with the increase of the DC IV share in the mixture. We found that the best mixture

for oat and clover growth had proportions of 5–10% peat, ≤ 50% DC IV, and ≥ 50% sand. The use of shungite as an additive did not influence the plants' growth. The mixture with 30% of DC IV seemed to be toxic for plant growth (Fig. 6b, c).

4 Discussion

This work was focused on the effectiveness of certain ameliorating additives (diatomite, shungite, phosphogypsum, glauconite, peat, cement, and sand) for reducing the contents of contaminants (PHCs, salts, and heavy metals) in the studied mixtures with DCs of different composition and properties.

4.1 Effects of processing techniques on the composition of drill cuttings

Differences between concentrations of most elements in the DCs and the Earth's crust were associated with the waste processing techniques and the constituents of the drilling fluids. The studied DC samples were subdivided into three

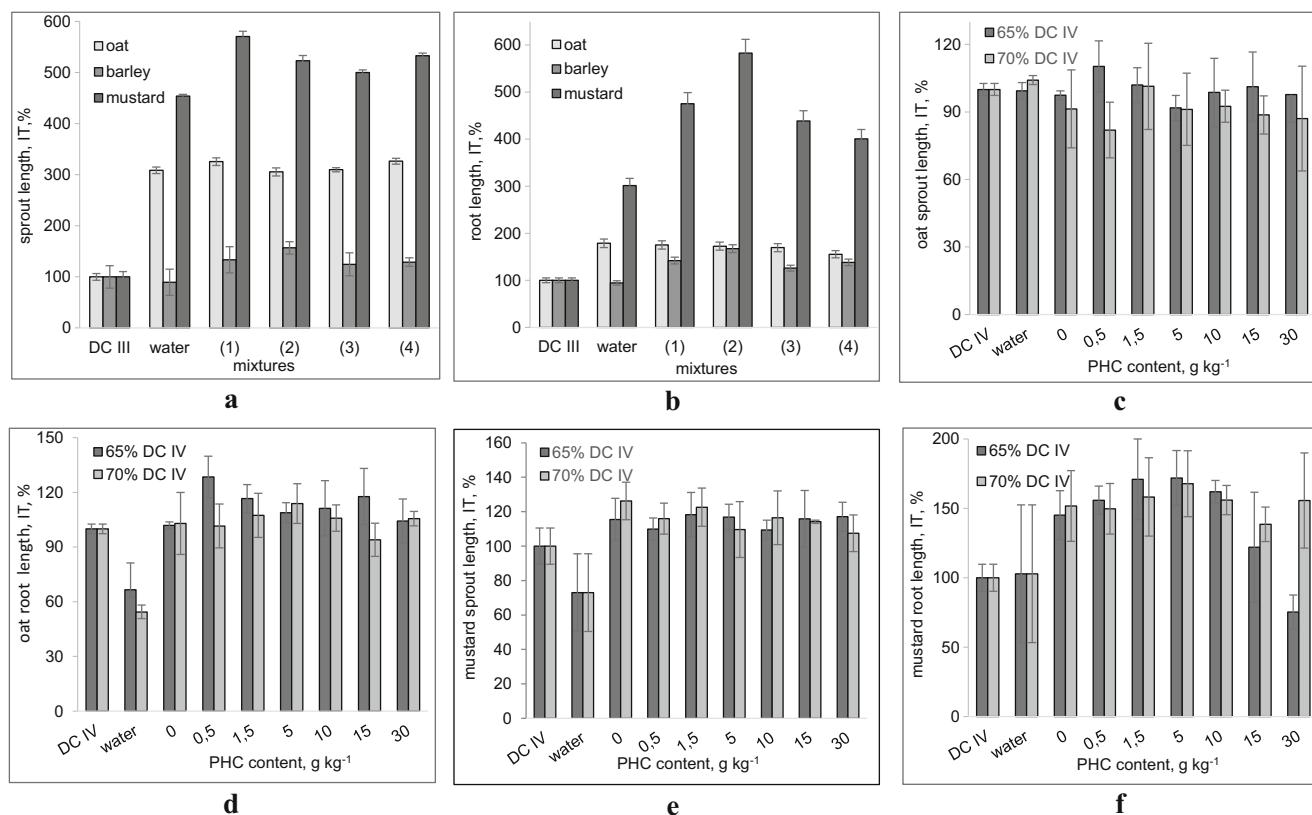


Fig. 4 Additives' effect on the root and sprout length, IT %. **a** Sprout length in DC III and mixtures (1, 2, 3, and 4). **b** Root length in DC III and mixtures (1, 2, 3, and 4). **c** Oat sprout length in DC IV and mixtures: DC IV 65–70%, and sand 30–35%. **d** Mustard sprout length in DC IV and mixtures: DC IV 65–70% and sand 30–35%. **e** Oat root length in DC IV and mixtures: DC IV 65–70% and sand 30–35%. **f** Mustard root length in

DC IV and mixtures: DC IV 65–70% and sand 30–35%. Mixtures: (1) DC III 70%, sand 21%, diatomite 4%, and peat 5% with PHC 5 g kg⁻¹; (2) DC III 70%, sand 21%, diatomite 4%, and peat 5% with PHC 45 g kg⁻¹; (3) DC III 60%, sand 36%, diatomite 2%, and phosphogypsum 2% with PHC 5 g kg⁻¹; (4) DC III 60%, sand 36%, diatomite 2%, and phosphogypsum 2% with PHC 8 g kg⁻¹

types, depending on their composition, as follows: (1) DCs I and III with significant contents of both PHCs and salts, predominantly, chlorides (up to 100% of the total of salts); (2) DC II with a high content of salts including chlorides (up to 50% of the total of salts); and (3) DC IV with low contents of both PHCs and salts represented mostly by hydrocarbonates.

Both DCs I and II were distinguished by high contents of Ba and SO₄²⁻, which resulted mainly from the use of barite (a natural barium sulfate ore) as a density control ingredient (Tuncan et al. 1997; Fink 2015). A high level of sulfates could also be caused by adding calcium lignosulfonate as a lubricant to drilling fluids, which also caused an increase in Ca contents in the DCs.

Fig. 5 Bioassay of filtrates. **a** Dependence of solid residue and paramecium mortality (%) in mixtures' filtrates (DC III (60–70%), sand (21–36%), diatomite (2–4%), and phosphogypsum (0–2%). **b** Paramecium and daphnids mortality (%) at 0–30 g kg⁻¹ PHC doses in mixtures' filtrates (DC IV, sand)

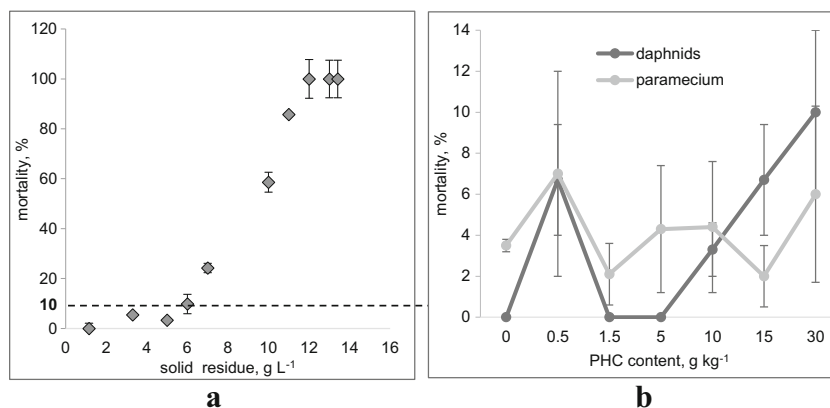
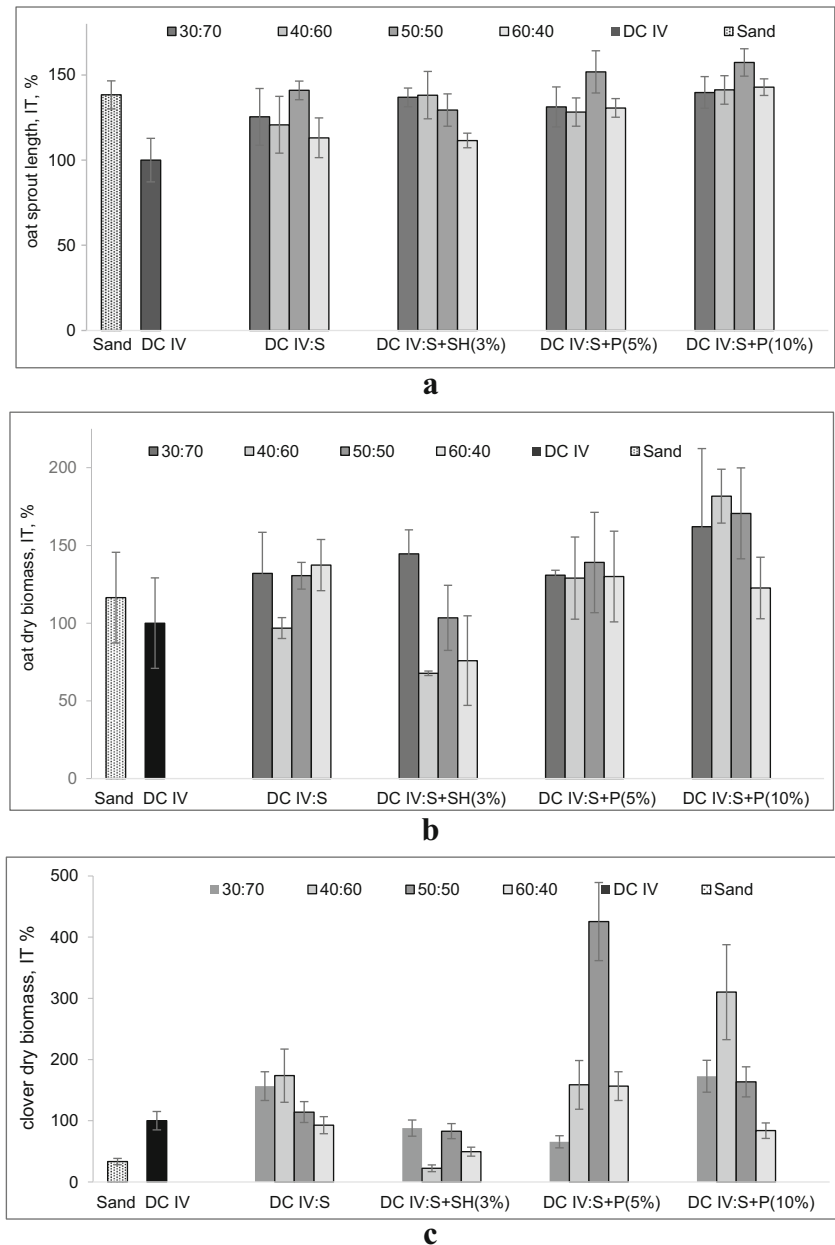


Fig. 6 Plant test parameters in pot experiment with mixtures, based on DC IV and sand (S) in different proportions with adding of shungite (SH) 3% or peat (P) 5 or 10%. **a** Elongation of oat plant, IT (%) **b** Oat dry biomass, IT (%) **c** Clover dry biomass, IT (%)



DCs I, II, and III were characterized by high concentrations of Cl ions, probably due to the use of CaCl_2 in oil-based drilling fluids (Fink 2015; Drilling fluids 2015). DCs I and III had very high Na and K levels, which were also predetermined by the composition of drilling fluids. It has been established that the contents of Na depend on the use of such ingredients as bentonite clay (sodium montmorillonite), caustic soda (sodium hydroxide), and phosphates (sodium acid pyrophosphate and tetrasodium pyrophosphate) as well as the fluid loss control agent (sodium carboxymethyl cellulose) and corrosion and scale control agent (sodium sulfite) (Fink 2015). The presence of K can result from the application of potassium chloride as an extremely efficient shale

stabilizer in cases of drilling through hydro-sensitive clays and shales (Zhong et al. 2013).

It has been reported that applications of zinc chromate (Fink 2015) and zinc chloride (Lyons et al. 2016) to drilling fluids can result in high Zn concentrations in DC samples in comparison with the Earth's crust. The highest Pb content was found in the DC IV sample, obviously due to its parent rock's origin and composition. Vanadium contents in the studied DC types significantly varied, i.e., from comparable with the Earth's crust and soils (e.g., DCs II and III) to being magnified by up to four times (DC IV). The latter could be due to the presence of oil porphyrins, which are known to contain vanadium (Eglinton and Murphy 1969). However, DC IV

generally contained relatively fewer toxic substances, because it was produced using a water-based drilling mud.

4.2 Effects of the ameliorating additives on the quality of leachates from the DC mixtures

The potential abilities of contaminants, including heavy metals, to diffuse from the solid phases of DCs and their mixtures into leachates (filtrates) were analyzed in our leaching experiment. Metals such as Ba, Cd, Cr, Co, Cu, Pb, Hg, Mo, Ni, Zn, and V are toxic, according to the federal or state agencies in the USA (API 1983), and their mobility levels depend on the pH of DC mixtures. Therefore, attaining an optimal pH level is a very important step in designing safe and eco-friendly DC mixtures that can be used as artificial soils. The pH of filtrate is routinely used as an indicator of the behavior of heavy metals (Arya et al. 1990; Conner and Hoeffner 1998).

Despite the fact that the tested DCs and their mixtures had alkaline reactions, their leachates obtained in experimental treatments (leaching with water volumes equal to the mean annual precipitation) were characterized by pH values close to neutral (Table 3). The neutral pH of the leachates resulted, most probably, from their dilution with water. Leachates with much higher pH values were obtained from DC mixtures with cement, e.g., leachates pH values of up to 12 were obtained from DC mixtures with cement contents as low as 1–2%. An alkaline reaction often helps to immobilize heavy metals, for example, it is known that metal hydroxides tend to be least soluble within the pH range from 7.5 to 11 (Conner 1990) and some heavy metals can precipitate as hydroxides within the pH range from 9 to 11 (Lanouette 1977). However, solubilities of other metals, such as Cr, Zn, Ni, and Pb, rapidly increase at a pH above 9 (Conner 1990). The formation of soluble compounds of Cu and other metals under alkaline conditions (pH of 7.5–9.5) can be enhanced in the presence of sulfates and carbonates (Glazovskaya 1988).

In the present study, soluble Cu compounds were detected in all leachates obtained from cement-containing mixtures, e.g., soluble Cu (II) complexes were detected in a strongly alkaline (pH of 12–13) leachate from a mixture of DC II, diatomite, and cement. Negatively charged Cu complexes can inhibit the processes of ion binding due to absorption of clay particles on the surface (Amangusova et al. 2015). Under strongly alkaline conditions, Sr behaves similarly to Cu. Belitsky and Orlova (1968) have shown that the maximal sorption of Sr in soils occurs at the pH of 10–11 and its minimal sorption at pH < 2 and pH > 11. Therefore, in our experiments, the mobility of Sr increased in the alkaline leachates (pH from 11.9 to 12.7) obtained from cement-containing DC mixtures.

DC mixtures with phosphogypsum and peat, as well as their leachates, tended to have a weakly acidic reaction, which

indicated a significant buffer capacity of those additives. Phosphogypsum additions caused dramatic increases in Sr contents in filtrates, probably due to the fact that phosphogypsum is often enriched in this element (Wang et al. 2018). DC mixtures with phosphogypsum and shungite produced leachates that were not enriched in Sr, because shungite is able to adsorb Sr ions and holds them in a solid phase.

The additions of peat to DC III mixtures resulted in increased mobility of heavy metals, probably because peat usually contains fulvic acids that can form soluble complexes with metal ions (Spark et al. 1997; MacCarthy and O'Conneide 1974). Furthermore, additions of 13% peat to the mixtures resulted in a significant increase in PHC leaching, despite the ability of peat to bind hydrocarbons. Deprotonation of acid centers takes place in alkaline medium, and the organic matter of peat works as a poor sorbent of PHC. At the same time, the water-soluble organic matter of peat can solubilize the PHC. In our experiments with DC mixtures, the best results were achieved by adding sand in proportions of 20% or more, together with some diatomite. The sorption properties of diatomite and its ability to firmly hold PHC and metals are probably related to its porous structure and a large surface area (Taylor 1995).

The best quality leachates were obtained from mixtures with DC IV, which was characterized by a high content of HCO_3^- and a low content of Cl^- . Dilution with sand and additions of shungite allowed for significant decreases in PHC and salt contents as well as concentrations of Zn, Mn, Ba, and Sr in DC IV mixtures.

4.3 Effects of the additives on biological parameters of the mixtures

The chemical analyses together with ecotoxicological testing of DCs and their mixtures that are to be disposed of as artificial soils can provide important information on possible environmental risks. A series of aquatic bioassays as well as phytoassays were performed to evaluate the toxic effects of the DC mixtures of different composition and properties. Phytotoxicity tests are based on the ability of plants to respond to environmental changes and serve as highly sensitive indicators for evaluating the toxicity of soils and wastes (Nikolaeva and Terekhova 2017). As plant growth depends on the soil properties, we performed pot experiments with higher plants and assessed their root and shoot lengths following ISO 11269 (ISO 11269-1 2012; ISO 11269-2 2012).

Phytotoxicity tests showed that concentrations of salts, including chlorides, of more than 0.6% reduced the seed germination rates by up to 100%, regardless of DC dilution and adding sorbents. This is in line with the data obtained for soils under salinization conditions (Sun et al. 2019). The PHC concentrations and pH values of DC mixtures were less important factors that influenced the plant germination. The results of

phytotests on mixtures with DCs I and II did not show a clear pattern of the effects of PHC and metals on plant growth due to the high chloride toxicity. Other researchers have reported a greater sensitivity of phytotests compared with other bioassays under normal conditions (Rodríguez-Ruiz et al. 2014). Adding diatomite into the mixtures with solid residue contents of less than 6 g kg^{-1} led to lower toxicity levels in comparison with those of untreated DCs.

Adding sand and combined additives (diatomite, phosphogypsum, or peat) to DC III containing PHC stimulated the plant growth (Fig. 4a, b). This effect could be associated with several factors, e.g., the DC dilution and the sorption of contaminants as well as an additional nutrient intake may positively influence the plant's germination and growth. We found a significant correlation between oat root length and concentrations of PHC and also K, with R values of 0.97 and 0.98, respectively, at $p < 0.05$ (SA II).

All mixtures prepared with DC IV had significantly positive effects on root and sprout lengths in mustard, in comparison with the control and also the pure DC IV. Such effects could be accounted for by the presence of HCO_3^- and Ca^{2+} ions in DC IV and its mixtures, which are responsible for an increase in the buffering capacity of the solid phase. These ions originate from water-soda-clay drilling fluids of DC IV. The presence of Ca^{2+} and HCO_3^- in soils seems to be beneficial for plant growth. There was a significant correlation ($R = 0.9$, $p < 0.05$) between the length of oat shoots and the Mn content in a pot experiment. Manganese can affect the plant's resistance to stress conditions and regulate the supply of other elements. Shungite as an additive had no effect on the growth of the oat and clover plants.

The toxicity assay based on paramecium behavior did not correlate with the presence of any contaminants in water extracts and leachates, but tests with daphnids showed their significant dependence on pH and PHC contents. For example, there was a close correlation ($R = 0.73$, $p < 0.01$) between the mortality of *Daphnia magna* and the PHC concentration (SA II). Chloride-free water extracts from DC IV mixtures displayed no toxicity, which could be explained by the presence of shungite in the composition of those mixtures (Fig. 1d) as well as by the solubility of contaminants. Such a toxic element as barium is mostly associated with sulfates, e.g., barite (BaSO_4) used as a density control agent in drilling fluids. The solubility of BaSO_4 in water is $2.42 \times 10^{-3} \text{ g L}^{-1}$ at 20°C . Our results indicated that the drilling technique and the composition of drilling fluids predetermine the DC composition and, therefore, their proper choice could successfully mitigate the DC toxicity.

The combined disposal of DC solid wastes, drilling fluids, and water can result in unacceptably high concentrations of toxic substances (including salts such as chlorides) in the environment. Based on our findings, we have developed eco-friendly mixtures with $\leq 70\%$ DC and $\geq 20\%$ sand. Small

additions of diatomite were the most efficient at absorbing the PHCs, while shungite most efficiently reduced the heavy metal toxicity. The levels of chlorides should be carefully controlled and should not exceed 6 g kg^{-1} of a mixture used as soil-like parent materials. The DC mixtures with Cl^- concentrations of about 3 g kg^{-1} could be recommended for utilization as artificial soils. The DCs with high contents of HCO_3^- and low contents of Cl^- could serve as optimal ingredients of the artificial soil mixtures.

5 Conclusions

We analyzed the chemical composition and toxicity of pure DCs, which were generated with the use of different drilling fluids, and estimated their possible negative impacts on the environment. We have suggested a combination of additives that can transform such DCs into eco-friendly soil-like mixtures. Sand was shown to be the best major additive in such mixtures, which can dilute and reduce the concentrations of contaminants provided that soluble salts detected in DC leachates were mostly represented by HCO_3^- ions, with low portions of Cl. Our pot experiments confirmed that mixtures containing $\leq 65\%$ DC, $\geq 30\%$ sand and 5–10% peat can be safely used as fertile artificial soils. A high level of Cl^- ($> 3 \text{ g kg}^{-1}$) was the primary cause of toxicity of tested DC mixtures for plants and hydrobionts.

This case study provides solid evidence that adding phosphogypsum to DC mixtures can lead to their excessive contamination, particularly, due to increased solubilities of Cu and Sr. The use of phosphogypsum in DC mixtures requires the continuous monitoring of the contents of contaminants and the pH of leachates from those mixtures. Cement additions (1–2%) caused a dramatic increase in the pH of leachates from the mixtures. Such alkalization could induce an increase in the mobility of some contaminants (e.g., Cu and Sr). An increase in the mobility of metal ions could also be caused by phosphogypsum additions due to the formation of soluble metal-organic complexes. Shungite was shown to be the most effective absorbent of contaminants such as Zn, Cu, Mn, Ba, and Sr, but only at pH levels below 8.0–8.5. We emphasize that the correct choice of the additive is vital, as it can significantly influence the contaminants' behavior under different conditions.

We established that the pot experiments were the best assay to evaluate the ability of plants to grow on DC mixtures. At the same time, bioassays were a useful and handy way to immediately assess the toxicity of mixtures. We underline that the tested plant species responded differently to the various contaminants and toxicity parameters, and so they need to be analyzed together. Hydrobionts were not highly sensitive to PHC contamination.

The investigated parameters were shown to be highly important in determining the correct choice of additives to DC mixtures and in helping to design eco-friendly mixtures to be integrated into the environment as artificial soil materials.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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