

Original Research

Soil and Groundwater Contamination as a Result of Sewage Sludge Land Application

A. Łuczkiwicz*

Department of Water and Wastewater Technology, Faculty of Civil and Environmental Engineering,
Gdańsk University of Technology, G. Narutowicza 11/12, 80-952 Gdańsk, Poland

Received: August 17, 2005

Accepted: June 2, 2006

Abstract

The leaching of chemical compounds (N_{NO_3} , N_{NH_4} , N_{org} , P_{PO_4} and P_{tot}) and trace elements (Cd, Cu, Cr, Ni, Zn, and Pb) from sewage sludge and their migration through the soil profile is the subject of this study. The column experiments were carried out under atmospheric conditions corresponding to naturally occurring storm events – sewage sludge amended soil was irrigated with high rates of water. The column filter media used in the laboratory tests were similar to the soils present in degraded areas: coal fly ash (FA), coarse grained sand (CGS) and medium grained sand (MGS). Sewage sludge was applied on the top of each column in the quantity corresponding to the best land reclamation practice. The data obtained in the experiments showed the dynamics of chemical compounds' leaching from the sewage sludge and their further transport through the column beds to the effluents. It was discovered that nitrogen compounds, such as nitrate (N_{NO_3}) and ammonium (N_{NH_4}), as well as some heavy metals (Ni and Cd) – originating from the sewage sludge – can reach deeper than 0.8 m and cause the contamination of potential shallow aquifers.

Keywords: sewage sludge, nutrient compounds, heavy metals, soil and groundwater contamination, column experiments

Introduction

Wastewater treatment plants generate effluents and sludge that must be disposed of safely and economically. It is well known that sewage sludge represents a potentially valuable source of nutrients and organic matter for arable farming, for the reconditioning of sandy and degraded soils [1- 3], as well as for erosion control and slope stabilization [1, 4]. Although sewage sludge improves soil properties and its use in the rehabilitation of degraded natural and anthropogenic soils seems to be the most convenient, it is controversial because of possible health and environmental risks involved [5-7].

This paper studies the effects of sludge land applications in column experiments. The ability of leaching sludge-borne nutrients and heavy metals as well as their interactions with column beds were analyzed. As column beds coal fly ash (FA), coarse grained sand (CGS) and medium grained sand (MGS) were used. In coarse and medium grained sands low water-holding capacity, no organic component and lack of available macronutrients limit plant colonization and consequently promote water and wind degradation of these soils. Similar problems occur in fly ash, where additionally the content of toxic substances and alkalinity inhibited root growth. One way to overcome this problems is to apply sewage sludge as organic matter and nutrient supplements. This technique is both: a method for treating waste and improving soil conditions.

*e-mail: aneta.luczkiwicz@pg.gda.pl

Materials and Methods

The column experiments were conducted in four series: A, B, C, and D. Cylindrical plexiglass columns were used in all the series. The total length of each column was 100 cm with the internal diameter of 5.4 cm. As column beds, coarse grained sand (CGS), medium grained sand (MGS) and coal fly ash (FA) were used. The basic features of the column filter media are summarized in Table 1.

Air-dried CGS, MGS and FA were sieved (<1mm) and to eliminate the impact of their autochthonous microflora on the obtained results, autoclaved prior to every experiment. The columns were filled to a height of 0.8 m and to achieve high packing density, dry-packed columns were completely saturated from the bottom up with the filter-sterilized ($\phi = 0.45\mu\text{m}$) deionized water

(pH 6.5 – 6.6). Next the columns were drained under the force of gravity.

The stabilized sewage sludge, collected from the local wastewater plant “Wschód” in Gdańsk was a mixture of primary and secondary sludge, which has previously been stored in piles for three months. The dosage of the sludge in each experiment was equal to $100 \text{ Mg ha}^{-1} (\text{dm})$. This value, with regard to the column internal diameter (5.4 cm), was equivalent to 23 g (dm) of sewage sludge per column. To simulate conditions of land reclamation, the sewage sludge was first mixed with coarse grained sand (CGS) in ratio 1:3 (m/m) and then placed as a layer (thickness about 5 cm) at the top of each column bed. The sewage sludge/sand mixture is called as initial deposition (ID) in the paper. The mass of the initial deposition applied to each column was equal to $81.05 \pm 0.50 \text{ g (dm)}$. Basic properties of the ID are shown in Table 2.

Table 1. Basic features of the filter media used in the column experiments.

Constituents		Coarse grained sand (CGS)	Medium grained sand (MGS)	Coal fly ash (FA)
Av. grain size d_{50}	mm	0.70	0.25	0.10
Silt fraction	%	0.00	1.33	25.61
Clay fraction	%	0.00	0.00	1.21
Organic matter	%	0.20 ± 0.01	0.39 ± 0.02	1.31 ± 0.11
pH (H_2O)	%	6.5 ± 0.5	5.9 ± 0.4	8.4 ± 0.7
N_{NO_3}	$\text{g} \cdot \text{kg}^{-1}$ (dm)	nd ^a	nd	0.020 ± 0.005
N_{NH_4}		nd	0.005 ± 0.001	0.030 ± 0.001
N_{org}		nd	0.007 ± 0.002	0.005 ± 0.003
P_{tot}		nd	0.007 ± 0.002	0.020 ± 0.002
Cd	$\text{mg} \cdot \text{kg}^{-1}$ (dm)	1.0 ± 0.3	1.8 ± 0.2	2.4 ± 0.5
Cr		5.1 ± 0.5	8.3 ± 0.7	89.0 ± 0.9
Cu		4.5 ± 0.3	9.3 ± 0.2	80.6 ± 4.8
Ni		5.5 ± 0.2	8.0 ± 1.0	23.7 ± 0.7
Pb		13.0 ± 2.0	15.0 ± 2.1	39.0 ± 1.7
Zn		14.6 ± 2.5	23.3 ± 2.9	120.2 ± 5.9

^a) nd – not detectable; detection limit [$\text{g} \cdot \text{kg}^{-1}$] was less than 0.001 for N_{NO_3} and P_{tot} , and less than 0.005 for N_{NH_4} and N_{org} .

Table 2. Basic features of the initial deposition (ID).

pH	N_{NO_3}	N_{NH_4}	N_{org}	P_{tot}
$\text{g} \cdot \text{kg}^{-1} (\text{dm})$				
6.8 ± 0.8	0.08 ± 0.01	3.50 ± 0.10	1.27 ± 0.09	1.31 ± 0.25
Cd	Cr	Cu	Ni	Pb
$\text{mg} \cdot \text{kg}^{-1} (\text{dm})$				
2.9 ± 0.1	38.0 ± 3.6	99.8 ± 6.8	14.8 ± 0.8	81.3 ± 6.5
				Zn
				242.7 ± 14.9

The column experiments were conducted under atmospheric conditions corresponding to heavy rainfall – 100 mm per day. The volume of water applied to each column was equal to 0.230 dm³ per day, supplied every day in the period of 12 h followed by 12 h intervals. Each day column effluents were collected and subjected to physical and chemical analyses.

In pilot-run series A, the sorption abilities of the selected filter media (CGS, MGS, FA) were determined. For 30 days three parallel experiments were carried out, each in a separate column, packed with a different filter medium: coarse grained sand was in the first column (A1), medium grained sand in the second (A2), and coal fly ash in the third (A3). The data obtained in the pilot series A showed that the concentration of the analyzed, sludge-borne heavy metals

and nutrients in the column effluents reached the highest level during the first 7 days of the experiments, regardless of the filter medium type (see Fig 1). Thus, that period of time was established as sufficiently long for series B, C, and D. The reduction of series duration also helped avoid the effect of the microbiological oxidation of organic nitrogen (N_{org}) and ammonium nitrogen (N_{NH_4}), which was observed in series A. Additionally, to eliminate the incidental data, each of the short series (B, C and D) was conducted in four repetitions – four columns were working under the same flow and filter media conditions. Coarse grained sand (CGS) was used in series B, medium grained sand (MGS) in series C and coal fly ash (FA) in series D.

Solid samples of the filter media (CGS, MGS, FA) and the initial deposition (ID) were subjected to investigation

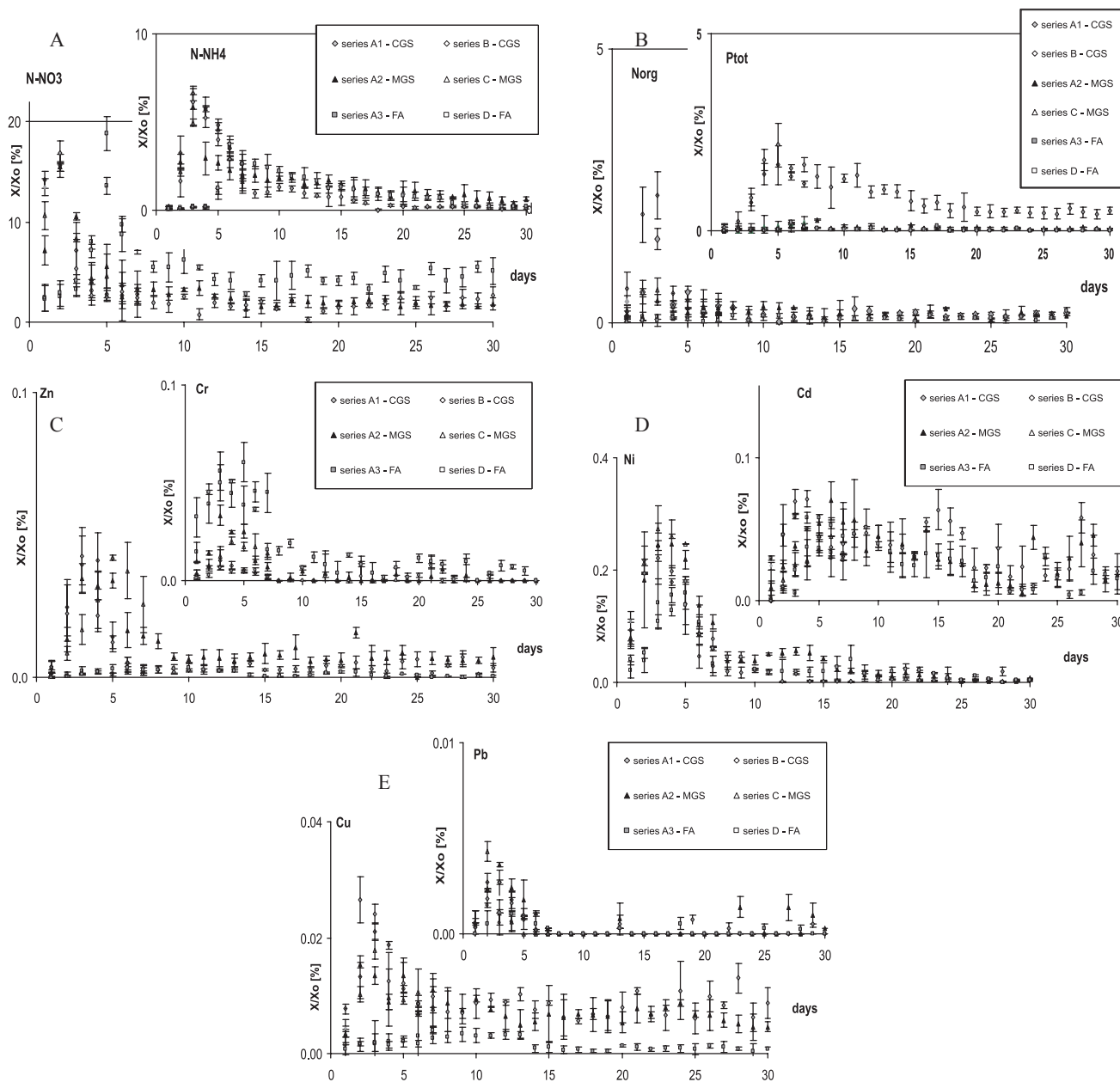


Fig. 1. Relative BTCs, i.e. ratio of analyzed compounds mass present in daily effluents (X) to their initial mass (X_0) introduced to the column in a function of time (days).

of physical and chemical properties. The grain-size sieve analysis were conducted prior to the experiments. Additionally pH, the total content of trace elements (Cr, Zn, Cd, Cu, Ni, Pb), the total phosphorus (P_{tot}), the total nitrogen of Kjeldhal ($N_{\text{tot,Kj}}$) as well as the content of nitrate (N_{NO_3}) and ammonium (N_{NH_4}) were determined.

Column effluents were collected in bottles during the experiments and analyzed once a day for pH, selected heavy metals, nitrate compounds and P_{tot} . Dissolved orthophosphates (P_{PO_4}) were determined every two days.

The measurements were made according to the Polish Standards [8, 9] and to the guidelines given by Ostrowska et al. [10].

Solid and liquid samples were homogenized prior to determination. Values of pH were estimated electrometrically, in solid samples in the 1:5 (volume fraction) suspension of soil in water. Total phosphorus was determined colorimetrically using the ammonium-molibdate (in the presence of glycine and SnCl_2) method, following the wet acidic digestion (H_2SO_4 and HNO_3) of the samples. Dissolved orthophosphates (P_{PO_4}) were determined by colorimetric method with stannous chloride as a reductant. The Kjeldhal nitrogen (the sum of organic and ammonia nitrogen) was determined after wet digestion with concentrated sulphuric acid catalyzed with K_2SO_4 and CuSO_4 . The digested samples were distilled after adding concentrated NaOH. Nitrate nitrogen (N_{NO_3}) was analyzed using the colorimetric method with sodium salicylate, while ammonium nitrogen (N_{NH_4}) was analyzed by the direct nesslerization method.

Prior to the determination of the total concentration of dissolved Cr, Zn, Cd, Cu, Ni and Pb from the column effluents, the initial separation of the suspension on nitrocellulose filters ($\phi = 0.45\mu\text{m}$) followed by complete sample evaporation and further dissolution in $0.1 \text{ mol dm}^{-3} \text{ HNO}_3$ was performed. Simultaneously, a blank sample was analogously prepared. The total concentration of heavy metals was determined by means of flame atomic adsorption spectrophotometry (ASA).

In the solid samples (initial deposition and analyzed filter media), the determination of the total content of heavy metals was made according to the following procedure: 1 g of air-dried soil and sludge sub-samples was digested with 5 cm^3 of aqua regia: HCl and HNO_3 (3:1)

at 80°C for 120 min [11, 12]. The solids were removed by centrifuge and the total content of heavy metals was determined in supernatants using the ASA method.

Certified material (Reference Material LGC6139 Laboratory of the Government Chemist UK) was used to state the accuracy of the procedure of heavy metals determination. Comparing the certified and measured values of the analyzed metals' metal recovery indicated the that accuracy was satisfactory (Table 3, second and third column respectively). Precision of measurements can also be highly evaluated. Relative Standard Deviation (RSD), given in the last column of Table 3, varied from 1% (Pb) to 10% (Cd).

In column experiments, nitrate is accepted as a pollution tracer in soil [13, 14]. Thus, in the current study, the migration of contaminants leached from the initial deposition was compared with the migration of NO_3^- , also originating from the ID.

Results and Discussion

Chemical compounds' migration in the soil environment is important in order to predict the quality of shallow aquifers. In the experiments, the influence of surface-applied sewage sludge on amended soils and on underlying groundwater was assessed by using nitrogen compounds (N_{NO_3} , N_{NH_4} , N_{org}), total phosphorus (P_{tot}) and selected heavy metals (Cd, Cu, Cr, Ni, Zn, Pb). Transport abilities of sand (CGS, MGS) and coal fly ash (FA) were examined. On the basis of the granulometric analysis, coarse and medium grained sands were qualified as permeable media, while FA as less permeable. To simulate the practice of land reclamation, sewage sludge was mixed with sand and applied to each column as initial deposition (ID). The nutrient content in initial deposition was, for all series, at a comparable level (Table 2). The column experiments were conducted in the conditions of heavy rainfall. The daily volume of sterile deionised water applied to each column was equal to 0.230 dm^3 . The volume of the total effluent from each column was close to the total amount of input water (water recovery was over 95%).

The transport of the nutrients and trace elements, originating from the ID, in the column filter beds was analyzed

Table 3. Comparison of the recommended and measured content of heavy metals in the certified material.

Metal	Recommended	Measured	Recovery [%]	RSD [%]
Cd	2.3	2.0 ± 0.2	87	10.0
Cr	126.0	91.7 ± 3.9	73	4.3
Cu	96.0	92.5 ± 4.8	96	5.2
Ni	44.0	40.5 ± 0.9	92	2.2
Pb	176.0	154.6 ± 1.5	88	1.0
Zn	530.0	505.3 ± 39.4	95	7.8

in unsaturated flow conditions. Chemical compounds, released and removed from the initial depositions by water percolating through that layer, were transported through the column beds to the effluents. The dynamic of that leaching was determined as a ratio of the analyzed compound mass present in the daily effluents (X) to the mass (X_0) introduced to the column with the initial depositions. Obtained relative breakthrough curves (BTCs) helped to allow observation of changes in the solutes' concentration in the effluents.

Nitrate nitrogen, originating from the initial deposition, was used as a nonreactive pollutant tracer. The average content of that compound in the dry mass of the initial deposition was equal to $0.08 \text{ g N}_{\text{NO}_3} \text{ kg}^{-1}$ ($\text{SD} = 0.01$), so the initial mass (X_0) introduced to the each column was about $6.4 \text{ mg N}_{\text{NO}_3}$ ($\text{SD} = 0.1$).

Mobile nitrate ions were easily released from the ID. The relative breakthrough curves, ($X_{\text{N-NO}_3} X_{0\text{N-NO}_3}^{-1}$) presented in Fig. 1a, started to increase from the first day and reached the highest level on the second day of the experiments for coarse (series A1 and B) and medium grained sand (series A2 and C). For fly ash filter media (series A3 and D) the maximum concentration of N_{NO_3} was observed on the fifth day (Fig. 1a). The longer retardation time, recorded for FA was connected with high moisture content in that filter beds – the capillary water content was about four times higher than the amount of water applied daily to the columns. However, irrespective of the filter bed, the maximum concentration of nitrate nitrogen in the column effluents for each series was comparable and varied from 4.0 to $5.5 \text{ mg N}_{\text{NO}_3} \text{ dm}^{-3}$.

In the first seven days of the experiments sufficient repeatability and no significant difference in the transport

conditions of the chemical tracer (N_{NO_3}) between the corresponding experiments of short series B, C, D and long series A (respectively A1, A2, A3) were confirmed by the consistency of the obtained data. Correlation coefficients were equal to 0.97, 0.95 and 0.99 for CGS (column A1 and series B), MGS (column A2 and series C) and for FA (column A2 and series D) respectively.

The movement of NO_3^- is closely related to water movement [15] and nitrate can be treated as a nonreactive tracer [13, 14]. The obtained data confirmed that mobile NO_3^- ions were easily removed from the ID layer by percolating water, and transported to the outflow with the meaning of the used column beds being unimportant.

In the experiments a significant difference between the relative breakthrough curves noted for NO_3^- ions and for less mobile nitrogen compounds (N_{NH_4} , N_{org}) was observed. For coarse and medium grained sand, however, obtained N_{NH_4} and N_{NO_3} BTCs were similar in shape (Fig. 1a), ammonium retardation time was here about twice as high as for the tracer. In FA series, the maximum concentrations of N_{NH_4} in the column effluents varied from 4.4 to $4.8 \text{ mg N}_{\text{NH}_4} \text{ dm}^{-3}$, and were one and a half times lower than for CGS and MGS media (6.5 to $6.9 \text{ mg N}_{\text{NH}_4} \text{ dm}^{-3}$). The obtained data indicate, as previously discussed in the literature [15, 16], that ammonium was involved in the chemical reactions with the filter media. Fig. 1b represents the time variations of organic nitrogen and total phosphorus content in the column effluents. The obtained relative BTCs indicate that the breakthrough points were distinguishable only for the experiments performed with CGS (series A1 and B) – the highest concentration of N_{org} and P_{tot} were about $9.1 \text{ mg N}_{\text{org}} \text{ dm}^{-3}$ ($\text{SD} = 1.4$) and about $7.7 \text{ mg P}_{\text{tot}} \text{ dm}^{-3}$ ($\text{SD} = 0.6$), respectively. For other filter

Table 4. Ratio [%] of the cumulative amount of nutrient compounds and heavy metals present in the column effluents (X_{tot}) to their initial mass (X_0) introduced to the columns.

Constituents	30 days series			7 days series		
	column A1	column A2	column A	series B	series C	series D
	kind of filter bed					
	CGS	MGS	FA	CGS	MGS	FA
N_{NO_3}	92.1	100.3	128.9	48.5	48.2	47.9
N_{NH_4}	33.0	45.6	32.0	26.1	22.4	7.6
N_{org}	7.6	4.8	2.4	6.2	2.5	0.8
P_{tot}	25.0	1.9	0.9	6.7	0.6	0.2
Cd	1.0	0.9	0.7	0.3	0.2	0.2
Cr	<0.1	0.1	0.4	<0.1	0.1	0.3
Cu	0.3	0.2	<0.1	0.1	0.1	<0.1
Ni	1.3	1.7	1.0	0.9	1.2	0.6
Zn	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Pb	0.2	0.3	<0.1	0.1	0.2	<0.1

beds (MGS and FA), the concentration of the analyzed compounds in the column effluents stabilized at a constant value, less than 1.5 mg dm^{-3} for N_{org} and less than 1.0 mg dm^{-3} for P_{tot} .

In the case of phosphorus, it was found out that phosphates were the main compounds of P_{tot} (95 – 98%) present in the column effluents. Thus, P_{PO_4} was also the main phosphorus compound leached from the initial deposition. It indicates that the concentration of P_{tot} in the column effluents might depend on phosphate chemical affinity to the kind of filter media used in the experiments. Cheung et al. [17] and Geohring et al. [18] among the others, showed that the amount of phosphorus removed in the process of chemical adsorption depends mainly on the presence of calcium, iron and aluminium compounds in the ground. It should be noted that the bonding of phosphate on the surface of the soil particles, resulting from chemisorption, is relatively stable in environmental conditions [19, 20].

Mass balance – the proportion of the cumulative amount of compounds in the column effluents (X_{tot}) to their initial mass (X_0) introduced to the columns – indicated a significant influence of the filter media on the phosphorus compounds and organic nitrogen migration in the beds (Table 4). Compared to the initial mass, about 25% of P_{tot} and 8% of N_{org} were obtained in the effluents from coarse grained sand in series A. Medium grained sand and fly ash could be treated as more effective filters. In that case, ratio $X_{\text{tot}} X_0^{-1}$ was less than 2% for P_{tot} and less than 5% for N_{org} . Regardless of the column bed, even more than 100% of N_{NO_3} initially present in the ID was leached. So “tailing” of BTCs, obtained for N_{NO_3} in series A, suggests that even in a 30-day-time span, organic and ammonium nitrogen present in the initial deposition could be treated as an additional source of nitrate in the column effluent. In the current experiments, aerobic conditions facilitate the conversion of organic and ammonium nitrogen to nitrate nitrogen, and thus increase the possibility for nitrogen release from sewage sludge and movement through the underlying soil [21, 22]. It should also be indicated that NO_3^- , originating from sewage sludge, can be accepted as a tracer but only for short-term column tests.

In these experiments the total content of heavy metals in the initial depositions and effluents was also analyzed. It gave only general information on suspected leachate of the analyzed trace elements from the ID and their further migration in the filter beds.

As was in the case of the nutrient compounds, the highest concentration of heavy metals in the effluents were recorded mostly in the first 7 days of each series (Fig. 1c-e). With time, the concentration of the analyzed metals stabilized on relatively constant, low level – in many cases closed to the detection limit. Hence, the role of the measurement error, in this case, is significant and must be taken into consideration during the analysis of the results.

Data obtained in the experiments indicated that the heavy metals cumulative load (X_{tot}) in the effluents was

low and usually less than 1% of the initial mass (X_0). Only in the case of Ni was that value higher, but still did not exceed 2%. It is considered that the behaviour of heavy metals depends on their specific biochemical properties and the form in which they are introduced to the ground [23, 24]. The obtained data suggested that heavy metals in the initial deposition were rather present in the inorganic or residual fractions. Similar conclusions were presented, for example, by Walter et al. [11], Richards et al. [25] and Amir et al. [26]. However, opinions on this matter vary [24, 27].

Generally, heavy metals' mobility in soils is negatively correlated with pH [16-18]. The data obtained in the experiments with coal fly ash confirmed that correlation. FA is a rich source of heavy metals, but their alkalinity, also in the current experiments (pH of the effluents varied between 7.7 and 8.2), effectively promotes heavy metals retention. The exceptions were two metals: chromium and cadmium.

The chromium content in the effluents from the fly ash beds (series A3 and D) compared to its amount found in the effluents from the coarse and medium grained sand beds (series A1, B and A2, C) was elevated. It may have been caused by the migration-favorable range of FA's pH, together with the amphoteric character of this metal [29]. Additionally, it may be assumed that in that case, Cr in the effluents may have originated not only from the initial deposition but also from FA. Fly ash leaching may even have been a predominant factor. It is confirmed by the fact that after reaching the maximum value ($0.090 \text{ mg Cr dm}^{-3}$, $SD = 0.021$), chromium concentration in the effluents from FA stabilized on a relatively constant level, about $0.013 \text{ mg Cr dm}^{-3}$. For sand media, chromium concentration stabilized at a level close to zero.

Cadmium, occurred in the column effluents in trace quantities – not higher than $0.01 \text{ mg Cd dm}^{-3}$. Moreover, there was no clear maximum observed during any of the individual series. Cadmium exhibits similar extractability from the initial deposition and transport abilities independent of the kind of filter medium. Thus, the influence of pH of the analyzed filter beds (CGS – pH = 6.2; MGS – pH = 5.9; FA – pH = 8.4) on cadmium migration appeared insignificant. This is confirmed, among the others, by Salomons [31], Richards et al. [25] and Kabata-Pendias [29] that cadmium can stay mobile in a wide range of pH. Nevertheless, it should be taken into consideration that, because of the low concentration of cadmium in the column effluents close to the detection limits, the measurement error could have influenced the shape of the obtained curves.

Conclusions

The results of the experiments show that rainfall can leach the most mobile chemical compounds from sewage sludge applied to land. Undoubtedly, this influences the chemical composition of shallow aquifers, especially

when soils are characterized by high water permeability. Hence, together with heavy metals, nutrient compounds should be regarded as limiting factors for sewage sludge land application. This is due to the fact that the maximum concentrations of nickel, cadmium and ammonia nitrogen, recorded in the column effluents, exceeded limit values for groundwater quality levels [33]. In the real risk assessment of chemical groundwater contamination, however, nitrates also should be taken into consideration. Laboratory studies indicate that in 30-day-long series A about 100% of nitrate nitrogen present in the sewage sludge was leached to the effluents, regardless of the kind of filter bed. Moreover, it should be noted that the source of nitrates was 'renewable', because of organic and ammonia nitrogen oxidation. This phenomenon is important due to the fact that nitrates are common contaminants of shallow groundwater.

Appendix

av. – average

ASA – atomic adsorption spectrophotometry

BTC – breakthrough curve

dm – dry mass

CGS – coarse grained sand

FA – fly ash

ID – initial deposition

sewage sludge/CGS mixture prepared in ratio 1:3 (m/m)

MGS – medium grained sand

nd – not detectable

RSD – relative standard deviation

SD – standard deviation

X – mass of the analysed chemical compound or heavy metal present in daily effluents

X_{N-NO_3} – mass of nitrate nitrogen present in daily column effluents

X_0 – mass of the analysed chemical compound or heavy metal introduced to the column with initial depositions

$X_{0,N-NO_3}$ – mass of nitrate nitrogen introduced to the column with ID

X_{tot} – cumulative mass of analysed chemical compound or heavy metal present in the total volume of column effluents.

Acknowledgements

The work was carried out as part of a research project supported by the Committee for Scientific Research 7 T09D 044 21.

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