

CHEMICAL COMPOSITION OF DRAINAGE WATER FROM SEWAGE SLUDGES, USED AS PLANT SUBSTRATES

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INTRODUCTION

Sewage sludge as such may be seldom used as a plant substrate, but there are many situations in which it has to be stored for a shorter or longer period. In most cases a spontaneous vegetation will then arise and, unless the sewage sludge is sheltered, there will always be leaching, at least under West-European conditions with a precipitation surplus.

In 1972 we started an experiment, which is still going on, to find out to what extent a light sandy soil and a heavy clay soil, both needing large amounts of organic manure for optimum physical fertility, could be replaced by air-dried sewage sludges without damage to crop yield and/or crop and ground water quality. Crops and drainage water are analysed for macro- and micro-elements.

The soils were replaced by sewage sludges for up to 100%. Results of drainage water analyses for the 100% sewage sludge substrates in comparison with the soils without sewage sludge are presented in this paper. Results for the soil-sewage sludge-mixtures and the yields and chemical composition of the crops had to be left out of consideration here in view of the available time and space.

MATERIALS AND METHODS

In April, 1972, pots having a volume of 150 litres and a height of 75 cm were filled with six air-dried sewage sludges and two soils in layers of 65 cm in depth. The bottoms of the pots had been covered with a 5-cm deep gravel layer, which again was covered with a nylon sheet. Immediately above the bottom there is an outlet pipe, ending in a 2-litre polyethylene bottle, in which the drainage water is collected. Full bottles are stored in a refrigerator at 4 °C prior to analysis.

Characteristics of the sewage sludges and soils are given in table 1. In numerical sequence the sewage sludges were from the towns of Apeldoorn, Assen, Eindhoven, Heerlen, Leeuwarden and Leiden. All sludges were from drying beds, except nos. 1 and 3, which had been stored for a shorter or longer period in the open after mechanical dewatering. The chemical composition of the sludges varies widely and the same is true for the soils. Especially in the micro-element concentrations there is considerable variation. Until now samples of drainage water collected in the following periods have been analysed: April - September 1972 (1); September 1972 - April 1973 (2); April - December 1973 (3); January - June 1975 (4); July - December 1975 (5); January - December 1976 (6); January - June 1977 (7) and July - December 1977 (8). In 1974 there were no analyses because of lack of funds.

Characteristics determined in the drainage water were: COD (chemical oxygen demand as a measure of organic pollution), EC (electrical conductivity as a measure of total inorganic pollution), pH, N, P, K, Ca, Mg, Na, Cl, S, As, B, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb and Zn. Some characteristics were not determined in all periods. Details are given where results of separate characteristics are discussed.

Table 1. Chemical composition of soils and sewage sludges. Concentrations in % or ppm of dry matter

	soils		-----sewage sludges-----					
	sandy	clay	1	2	3	4	5	6
% organic matter	3.35	3.18	23.3	34.3	51.7	21.2	40.6	55.3
nitrogen (N)	0.09	0.19	0.72	2.04	2.20	1.10	1.98	2.83
phosphorus (P)	0.04	0.07	0.64	1.62	1.70	1.20	2.46	2.79
potassium (K)	0.05	0.78	0.05	0.27	0.18	0.18	0.28	0.19
calcium (Ca)	0.05	0.34	0.88	1.00	6.00	0.52	1.43	0.71
magnesium (Mg)	0.02	0.55	0.32	0.25	0.15	0.16	0.40	0.25
sodium (Na)	0.02	0.06	0.02	0.04	0.10	0.03	0.09	0.08
chlorine (Cl)	0.01	0.01	0.11	0.01	0.08	0.01	0.04	0.05
ppm arsenic (As)	2.0	11.6	7.5	5.8	11.6	10.3	45.9	19.5
boron (B)	58	n.d.	n.d.	134	75	148	n.d.	100
cadmium (Cd)	0.3	1.1	4.4	2.5	168	4.4	2.9	135
chromium (Cr)	19	69	515	148	1800	83	193	1693
copper (Cu)	2	77	542	497	3422	197	933	1084
% iron (Fe)	0.23	3.64	1.16	2.97	5.42	1.16	2.65	2.58
ppm mercury (Hg)	0.1	0.1	3.7	5.9	3.1	8.4	27.5	4.9
manganese (Mn)	67	1024	124	327	1190	248	1084	625
nickel (Ni)	8	42	56	22	336	24	46	934
lead (Pb)	17	56	1580	282	1874	195	538	637
zinc (Zn)	14	151	1269	984	5249	931	2391	5533

The analyses were carried out partly at the Central Laboratory of our institute (periods 1, 2, 3, 6 and 7), partly at the Laboratory for Soil and Crop Analysis in Oosterbeek. Both laboratories use the same methods, but there may be some discrepancies in results. A description of the methods used can be sent to those interested at request.

RESULTS

Main results for the drainage water are given in table 2, presenting weighted mean values for the first (period 1 and 2) and the last year (period 7 and 8), and showing the changes which took place in the experimental period. If no changes occurred or if a characteristic was not determined in the first year only one value is given.

For most values of the drainage water from the sewage sludges there was a sharp decrease in the course of time, which is shown in detail for COD in figure 1. For some characteristics and some sludges the picture was different, as is shown for the P content in figure 2. The separate characteristics mentioned in table 2 are briefly discussed below.

Amounts of Drainage Water

In our climate there is a precipitation surplus of about 300 mm = 3000 t/ha/year. The rest (about 400 mm) is evaporated, mainly by the crop.

In our experiment the amounts of drainage water over the whole period varied from about 200 - 500 mm/pot/year, due to differences in amount of precipitation (579 mm in 1976; 814 mm in 1973) and crop yield (cf. De Haan, 1978). Differences in amounts of drainage water between years and treatments

Table 2. Amount and chemical composition of drainage water from soils and sewage sludges. Where two values are given, separated by a diagonal, the first is for the first and the second for the last year of the experimental period considered.

	soils		sewage sludges					
	sandy	'clay	1	2	3	4	5	6
amount mm	280/370	275/385	290/370	325/385	310/410	270/405	275/405	325/475
COD mg/l	90/40	70/30	1120/140	4230/270	15200/250	1510/270	850/190	1430/170
EC μ S/cm	500	500	10600/1850	13300/2760	16900/2490	12000/2270	11800/2430	10400/2680
pH	6.9	7.3	7.4	7.2	7.3	6.4	7.2	6.0
N mg/l	20	20	820/90	1900/240	2250/180	1030/150	1040/230	1100/110
P "	0.15	0.29	2.1/0.4	16.8/1.0	23.8/2.4	43.7/64.9	5.5/5.2	25.9/97.3
K "	2.4	2.0	56/1.3	255/1.3	291/2.1	66/2.1	52/0.8	69/27
Ca "	70	94	2140/680	2310/960	1440/820	2520/760	2050/870	1640/760
Mg "	4.5	8.4	157/5	210/23	150/12	330/18	490/42	320/14
Na "	9	13	48/3	122/7	382/14	210/3	350/4	560/8
Cl "	33/7	28/6	1070/11	290/7	3830/9	230/8	370/10	260/13
S "	38	48	380	400	260	360	420	460
As μ g/l	0	20	10	10	30	250	70	190
B "	49	77	330	420	380	300	400	240
Cd "	<1	<1	10	1	16	5	3	250
Co "	1	1	8	6	14	12	5	120
Cr "	10	20	195/20	265/30	1720/50	250/30	190/20	250/10
Cu "	27	27	450/120	1040/120	5530/460	240/88	340/120	340/160
Fe "	120	180	160/50	2300/50	38300/210	250/24	41/34	170/44
Hg "	<0.1	<0.1	<0.1	0.2	0.2	<0.1	<0.1	<0.1
Mn "	62	44	2640/30	1670/140	4290/135	3360/375	4120/26	10700/5630
Mo "	18	9	200	57	3260	280	30	380
Ni "	10	10	450/100	422/36	3900/630	112/80	67/20	9800/12400
Pb "	<5	<5	<5	26/<5	47/<5	<5	<5	8/<5
Zn "	42	20	1040/240	490/80	1600/300	100/70	150/40	5600/3900

Figure 1

COD-values of drainage water from sewage sludges and soils in the experimental period

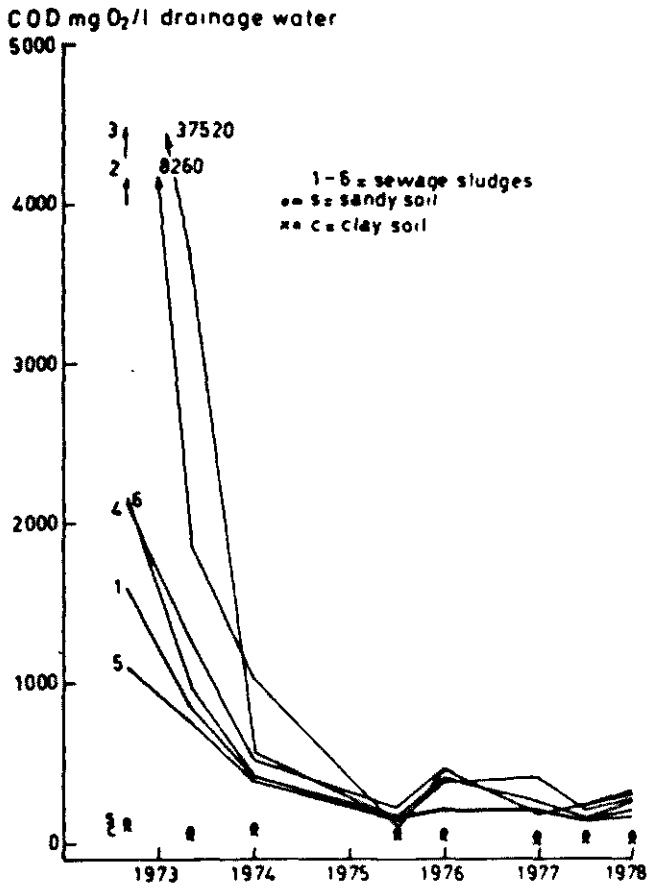
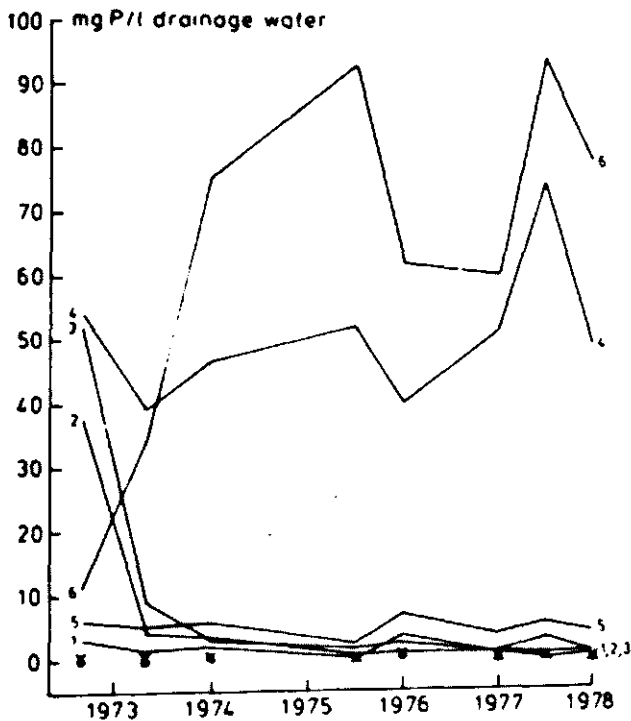


Figure 2
 mg P/l drainage water from sewage sludges (1-6), a sandy soil (•) and a clay soil (x)



were leveled to a certain extent by artificial water supply in times of drought, but may have affected differences in concentrations in the drainage water given in table 2.

Chemical Oxygen Demand

COD-values (mg O₂/litre) for the separate periods are given in figure 1. For the sewage sludges there is a sharp decrease in the values in the first year. In the third year a rather constant and low level was reached for all sludges. Mean values varied from 850 - 15200 in the first year and from 140 - 270 in the last year (table 2).

Corresponding values for the soils were 90/40 for the sandy soil and 70/30 for the clay soil. The rather high values for the soils in the first year may be due to the fact that the soils had been stored in an air-dry state for some years. An increased breakdown of organic matter afterwards can then be expected and may be accompanied by an increased production of water-soluble organic substances.

A COD-value of 40, which may be regarded as normal for drainage water from soils, means an oxygen demand of about 120 kg/ha/year, which is the same amount as is necessary for the oxidation of the organic pollution of 6 population equivalents. Corresponding values for the sewage sludges are 20 - 400 times as high in the first year and 3 - 7 times as high in the last year. Although in the last year the values for the sludges are still higher than those for the soils, per unit of organic matter they are lower for the sludges than for the soils and for that reason a further significant decrease in the values for the sludges seems to be unlikely.

Remark. No study was made as to the nature of the organic matter in the drainage water from the sewage sludges. From results with drainage water from town refuse deposits it seems likely that in the beginning the major part may have consisted of organic acids of low molecular weight and at the end of humic or fulvic acids (Hoeks, 1977; Ehrig, 1978). Contrary to the latter, the former are readily decomposable (Knoch and Stegmann, 1971; Cook and Foree, 1974).

Electrical Conductivity

EC-values (in micro-Siemens) in the drainage water from the sewage sludges varied from 10400 - 16900 in the first year and from 1850 - 2760 in the last year. These values are 20 - 34 and 4 - 6 times as high as those for the soils in the first and last year, respectively. The level of the last year was reached already in the third period.

It can already be stated that Ca and, to a lesser degree, Mg, Na and K as cations, and NO₃, Cl and probably also SO₄ and organic acid radicals as anions are responsible for the high initial EC-values of the drainage water from the sewage sludges.

pH

pH-values of the drainage water were determined not before the fourth period. Then the drainage water from sewage sludges no. 4 and 6 proved to be acid as opposed to that from the other sewage sludges and soils. A conclusive argument for the different behaviour of sludges no. 4 and 6 cannot be given, but almost certainly it is related to their low Ca contents.

Remark. It is unfortunate that pH-values were not determined from the beginning. In another experiment, in which the ripening process of sewage sludges is studied in the absence of crops, pH values were determined from the beginning and proved to be 7 or more. By contrast, leachates from town refuse deposits may have low initial pH-values, due to the presence of organic acids.

Nitrogen

N was determined in the drainage water as mineral N ($\text{NH}_4 + \text{NO}_3$) and as NO_3^- -N. Sometimes also total N, including organic N, was determined, but values for total N for some reason or another proved to be lower than those for mineral N.

Mineral contents of the drainage water from the sewage sludges varied from 820 - 2250 mg/litre in the first year and from 90 - 240 mg/litre in the last year. That is about 40 - 120 and 4 - 12 times as much as in the drainage water from the soils in the first and last year, respectively. All mineral N was in the form of nitrate, except in the first year, when it was partly in the form of ammonia, especially in the winter period, in the drainage water from the sludges 2 and 3 which had the highest COD-values. Therefore lack of oxygen may have inhibited oxidation of ammonia to nitrate here.

Phosphorus

Details of the P content of the drainage water in the different periods are given in figure 2. From this figure it appears that sludges no. 4 and 6 behave anomalously, as the P content of their drainage water remains or becomes high in the course of time. It should be noticed that the pH-values of the drainage water from these sludges were low; there may be a relationship between these phenomena.

P content of the drainage water from the soils was very low and even below the level for rain water in the Netherlands. For the sewage sludges, it was from about 15 - 300 times as high in the first and from 3 - 600 times as high in the last year of the experimental period considered.

Potassium

The K content of the drainage water from the sewage sludges showed the same trend as COD, with rather low initial values for the K content, in accordance with low K contents in the sewage sludges. Already in the fourth period, K contents in the drainage water from soils and sewage sludges were at the same level. Only for sludge no. 6 the final level was somewhat higher, no doubt as a consequence of reduced crop growth due to heavy-metal activity. The somewhat higher K level in the drainage water may have originated in this case from fertilizer K not used by the crop (De Haan, 1978).

Calcium

Ca contents of the drainage water from the sewage sludges were very high at the beginning of the experimental period (up to 3000 mg/l in the first period) and still rather high at the end (800 mg/l). Values for the drainage water from the soils were at a much lower level.

Magnesium

Magnesium contents of the drainage water from the sewage sludges varied in the first year from 150 - 490 mg/l and were at the same level as, or somewhat higher than, for the drainage water from soils in the last year.

Sodium

Sodium (Na) was leached rather rapidly from the sewage sludges. Na contents in the drainage water were not determined after the fifth period, but were then for sewage sludges and soils at the same level.

Chlorine

Cl contents of the drainage water from sewage sludges varied from 230 - 3830 mg/l in the first year, but rapidly fell, and the same level as for the drainage water from soils was reached already in the third period.

Sulphur

S contents of the drainage water were determined only for periods 4, 5 and 8. Weighted mean values are given in table 2. They proved to be rather high for the sewage sludges, but also for the soils, relative to the contents in the substrates, which were not determined, but can be assumed to be about 1% in DM for sewage sludges and 0.01 - 0.02% for soils.

Micro-elements

Arsenic

As contents were determined in the drainage water only in the fourth and fifth period. Average values varied then from 5 (detection limit) to 250 ppb.

Boron

B contents were determined only in the last 5 periods. Average values varied from 280 - 530 for sewage sludges and from 50 - 80 ppb for soils.

Cadmium

Cd contents of the drainage water were very low or became so after the first period. The only exception was sludge no. 6 with a rather value for the whole experimental period considered, probably as a consequence of a high content in the sludge in combination with an acid reaction of the drainage water.

Cobalt

Co contents in the drainage water were determined only in the fourth and fifth period. Average values were very low, except again for sludge no. 6.

Chromium

Cr contents were rather high, but only in the first year. They then fell to the same low level as for the drainage water from soils.

Copper

Cu contents were rather high in the first year, especially for sludges no. 2 and 3 with high COD-values for their drainage water. Though at a much lower level than in the first year, Cu contents remained rather high throughout the rest of the experimental period considered.

Iron

Fe contents of the drainage water were high only in the first year for sludges no. 2 and 3, possibly as a consequence of reducing conditions in these sludges at that time. After the first year, oxygen contents of the soil air were determined and proved to be normal. Fe contents of the drainage water after the first year were very low, both for sewage sludges and soils.

Mercury

Mercury (Hg) contents of the drainage water were determined in the last period only and proved to be below the detection limit, except for sludges no. 2 and 3.

Manganese

Mn contents of the drainage water were rather high for most sewage sludges in the first year and remained so for sludge no. 6 and to a lesser degree also for sludge no. 4, both sludges with acid DW. For the other sludges, Mn contents in the drainage water after the first year dropped to about the same level as for the drainage water from soils.

Molybdenum

Mo contents in the drainage water were determined only for the periods 4, 5 and 8. Average values are given in table 2. A rather high value was found for sludge no 3. Mo contents in the crop from this sludge were also very high.

Nickel

Rather high Ni contents were found in the drainage water from sludges no. 3 and 6, corresponding with rather high contents in the sludges themselves. For sludge no. 6, with acid drainage water, contents remained high throughout the whole experimental period.

Lead

Pb contents were only above the detection limit in the first year in the drainage water from sludges no.2 and 3, with high COD-values for the drainage water.

Zinc

Zn contents in the drainage water were high for sewage sludges no. 1, 3 and 6. For no. 6, with acid drainage water, they remained high throughout the whole experimental period.

REVIEW OF RESULTS

Data presented show that the drainage water from sewage sludges in comparison with soils is heavily polluted, both by organic and inorganic substances, but that the pollution dropped to a much lower level in the course of two to three years, except for elements as P, Cd, Ni and Zn, when the drainage water was acid.

There were large differences, for the same sludge among the elements, and for the same element among the sludges. Many of these differences are still unexplained, but no doubt they depend on differences in concentrations in the sludges, but also on a factor as state of maturity of the sludges, resulting in differences in COD-values of their drainage water. In the first period, concentrations of nearly all elements correlated well with COD-values. For some elements, this may be due to their ability to form soluble complexes with organic compounds, or to become more soluble under reducing conditions (Fe, Mn).

For the same element, there may also be differences among the sludges due to differences in chemical or physical form in which they are present in the sludges, but hardly anything is exactly known about this.

To show once again the differences between soils and sludges and among the sludges for the elements determined throughout the whole experimental period, amounts in the drainage water are expressed in % of the amounts present in the sludges and soils at the beginning of the experiment in table 3. In general, also these values are higher for the sludges than for the soils, and higher for the macro-elements than for the micro-elements, but these rules are not without exception. For instance, the macro-element P behaves as a micro-element, and the micro-elements Mn and Ni behave as macro-elements in the case of sludge no. 6, and Cu seems to be more mobile in the sandy soil than in all sewage sludges.

Without going into details, it may be said that, in general, concentrations in the crop were high when concentrations in the drainage water were high. For the sludges, the amount of an element in the drainage water was mostly much larger than that taken up by the crop in the same period.

DATA FROM LITERATURE

Data from literature relative to the topic of this paper are scarce. Bradford (cit. Page, 1974) determined trace elements in saturation extracts from Californian sewage sludges and soils and found concentrations of the same order of magnitude as in our experiment.

Comparable values for characteristics determined in our experiment have also been found for leachates from town refuse deposits (Ehrig, 1978; Hoeks, 1977), though there are variations due to differences in chemical compositions of the waste material and treatment. Town refuse is usually dumped in deep layers and compressed by its own weight and by heavy equipment (trucks, bulldozers), leading to anaerobic conditions in the system. Ripening processes take much more time under these conditions, and COD-values such as found in the drainage water from sewage sludges in our experiment only in the first year, may be found in leachates from town refuse deposits for a period of at least 10 years (Ehrig, 1978). From this it can be concluded that the results of our experiment are valid only for a thickness of layer of the same order as we used, and for an exploitation as agricultural land without using heavy farm equipment, as in our experiment, but which might be impracticable under farming conditions.

Table 3. Amounts of elements in the drainage water throughout the experimental period in % of the amounts in the soils and sewage sludges at the beginning of the experimental period

	soils		-----sewage sludges-----					
	sandy	clay	1	2	3	4	5	6
N	2.40	2.80	9.50	12.60	13.60	9.80	9.60	10.30
P	0.11	0.18	0.09	0.11	0.31	1.60	0.13	2.40
K	0.95	0.03	7.10	8.40	22.00	2.40	2.20	15.10
Ca	22.30	5.80	29.10	41.00	6.00	60.50	32.60	>100
Mg	2.60	0.30	3.90	13.00	1.80	20.00	19.10	29.30
Na	5.00	2.00	20.70	32.60	49.90	40.70	44.70	51.50
Cl	>100	>100	64.00	>100	>100	>100	97.00	91.00
Cd	0.26	0.09	0.69	0.18	0.08	0.41	0.69	1.65
Cr	0.20	0.09	0.03	0.19	0.11	0.27	0.16	0.04
Cu	3.30	0.09	0.12	0.28	0.24	0.24	0.11	0.17
Fe	0.01	0.001	0.003	0.01	0.08	0.003	0.002	0.004
Mn	0.28	0.02	1.65	1.08	0.57	1.47	0.55	10.90
Ni	0.22	0.09	2.22	2.96	1.95	4.10	1.70	11.80
Pb	0.04	0.01	0.001	0.01	0.003	0.01	0.002	0.02
Zn	0.52	0.03	0.12	0.08	0.05	0.03	0.04	0.77

CONCLUSIONS FOR PRACTICAL USE

The main conclusion for practical use from our experiment seems to be that air-dry sewage sludges under aerobic conditions can be changed in a relatively short period (2 - 3 years) from a material with a high potential for ground water pollution into a material, which could be used as soil or soil amendment with a minimum of risk of ground water pollution. To maintain or re-establish aerobic conditions, an incidental turnover of the material may be desirable under practical conditions and use in crop production, as in our experiment, might then be unpractical in the first years.

As the leachate may be heavily contaminated in the first 2 - 3 years, storage of the sewage sludges on an impermeable subsoil to protect the groundwater, and catchment and treatment of the leachate before discharging into the surface water may be necessary.

If the sludges are to be used as soil or soil amendment for crop production, their heavy metal contents should be low and their Ca content high to avoid mobilizing effects due to acidity.

SUMMARY

Six air-dried sewage sludges and two soils were placed into 150-litre pots to a depth of 65 cm and crops were grown and drainage water collected since 1972. Drainage water was analysed periodically for COD, EC, pH, N, P, K, Ca, Mg, Na, Cl, S, As, B, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb and Zn and results up to 1977 are presented in this paper.

Values for practically all characteristics determined were very high for the drainage water from the sewage sludges in the first period, but dropped in the course of 2 - 3 years to a much lower level, comparable with that for the drainage water from soils, except when the pH of the drainage water was low. Then the P content remained high, and also the contents of Cd, Ni and Zn

if the contents of these metals in the sludge were high.

The most important conclusion for practical use seems to be that sewage sludges, which in view of their chemical composition could be used as soil or soil amendment, after drying should be stored for 2 - 3 years under aerobic conditions in the open, with the possibility to catch and treat the drainage water. Afterwards they can be used with a minimum of risk of ground water pollution.

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