Heavy metal uptake by natural zeolite and metals partitioning in sewage sludge compost

A.A. Zorpas *, T. Constantinides, A.G. Vlyssides, I. Haralambous, M. Loizidou

Laboratory of General Chemistry, Department of Chemical Engineering, National Technical University of Athens, 9 Heroon Polytechniou St., Zographou 157 00, Athens, Greece

Abstract

A major limitation of land application of sewage sludge compost is the potential high heavy metal content due to the metal content of the original sludge. Zeolites may be useful as metal scavengers in metal-rich sludges. The natural zeolite, clinoptilolite has the ability to take up heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn). The aim of the reported work was to determine the metal forms most readily taken up by a natural zeolite when used as a bulking material during the composting process. Using a sequential extraction procedure in the raw sludge and in the final products after 150 days of maturity, the heavy metal content was associated with five fractions; the exchangeable, the carbonate, the reducible, the organic and the residual. It was found that a significant percentage of the metals not taken up by the zeolite was associated with the residual fraction, which is considered an inert form. © 1999 Published by Elsevier Science Ltd. All rights reserved.

Keywords: Sewage sludge composting; Metal uptake; Natural zeolite; Clinoptilolite; Metals partitioning

1. Introduction

In the greater region of Athens, with almost 4 500 000 citizens, the main wastewater treatment plant operating is in the rock-island of Psittalia. At Psittalia, approximately 750 000 m³/day of mainly municipal wastewater along with industrial wastes are subjected to primary treatment. Approximately 250 t/day of primary sludge is produced. Until recently, landfilling has been the main disposal route for sewage sludge, generating potential environmental hazards, including the production of odour and methane gas, as well as the contamination of groundwater by the leachate produced (Papadopoulos et al., 1997). Also decreasing land availability and increasing land cost is a major concern (Drougas et al., 1992).

A major limitation of land application of sewage sludge compost is the potential for metal contamination related to the metal content of the original sludge. Natural zeolite, clinoptilolite, has the ability to take up heavy metals. Zeolite utilization has become popular in the last decade, either for their cation exchange or molecular sieving properties. Currently, natural zeolites are used in soil benefication, and in water and wastewater treatment (Constantopoulou et al., 1994; Kapetanios and Loizidou, 1992; Loizidou, 1989). The aim of the reported study was to examine the content and the chemical forms of the heavy metals Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn in the raw sludge and in the final compost products when natural zeolite was used during the composting process.

A wide variety of sequential chemical extraction (SCE) procedures have been developed for the determination of heavy metal forms in sewage sludge (Lindau and Hossner, 1982; Pierce et al., 1982). According to the procedure of Tessier et al. (1979), heavy metals are associated with five fractions: the exchangeable fraction, which is likely to be affected by changes in water ionic composition as well as sorption–desorption processes; the carbonate fraction, that is susceptible to changes in pH; the reducible fraction, that consists of iron and manganese oxides which are thermodynamically unstable under anoxic conditions; the organic fraction, that can be degraded (oxidizing) leading to a release of soluble metals under oxidizing conditions; the residual fraction, that contains mainly primary and secondary minerals, which may hold metals within their crystal line structure. These metals are not expected to be released in solution over a reasonable time span under the condi-

* Corresponding author.
E-mail address: antoniszorpas@iname.com (A.A. Zorpas)

Table 1
Chemical analysis of clinoptilolite

Component	Mean value of 20 samples			
	mol/100 g		% (w/w)	
SiO₂	1.151	±0.025	69.396	±0.660
Al₂O₃	0.112	±0.001	11.551	±0.045
Fe₂O₃	0.002	±0.0002	0.384	±0.016
Na₂O	0.080	±0.001	4.980	±0.027
H₂O	0.495	±0.005	8.955	±0.039
K₂O	0.028	±0.002	2.753	±0.086
CaO	0.035	±0.002	1.981	±0.049
Total			100	

tions normally encountered in nature. The above procedure, with some modifications (Savvides et al., 1995), was chosen for this study.

2. Methods

2.1. Sewage sludge collection

Dewatered anaerobically stabilized primary sewage sludge (DASPSS) was collected from the Psittalia wastewater treatment plant in Athens. The samples were air-dried at room temperature and prepared in order to determine the following parameters: pH, electrical conductivity, total organic carbon (TOC), organic matter (OM), total phosphorous (TP), total Kjeldahl nitrogen (TKN), ammonia nitrite, nitrate and heavy metals contents (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn). The natural zeolite, clinoptilolite was collected from Evros (region in northern Greece) and analyzed for chemical composition (Table 1) (Maliou, 1994). The above analysis gives an ion-exchange capacity of 1.70 meq/g.

In order to observe the effect of the zeolite and how the metal content of the final products varied, a range of 0–30% clinoptilolite was applied to the DASPSS (Table 2).

2.2. Composting process

The composting process was carried out in the laboratory using an In-Vessel reactor of 10 l volume (Finstein

Table 2
Prepared samples for composting

Sample	Clinoptilolite (%)	DASPSS^a (%)
A0	0	100
A1	5	95
A2	10	90
A3	15	85
A4	20	80
A5	25	75
A6	30	70

^{a} DASPSS: dewatered anaerobically stabilized primary sewage sludge.

et al., 1992). The thermophilic phase in the reactor lasted for 15 days. The temperature was set to approximately 60–65°C in the center of the reactor and the moisture was in the range of 40–50%. After the thermophilic period in which the organic material was biodegraded, the compost was piled into an enclosed package where it remained for about four months to mature. For all the prepared samples, the following parameters were determined: pH value, electrical conductivity, TOC, OM, TP, TKN and ammonia content, and heavy metal Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn content.

2.3. Methods of analysis

For the determination of conductivity, TOC, TKN and ammonia content, nitrite, nitrate, standard methods of analysis were used (APHA, 1985; Adams, 1990). pH values were determined according to previously described procedures (Methods of Sampling and Analysis of Solid Wastes, 1970). TP was determined by stannous chloride method (APHA, 1985) while the OM according to Soil Reference Handbook (Soil Reference Handbook, 1992). For the total metal concentration, a known quantity (1 g) of sample was digested with 10 ml of conc. HNO_3 as described by Zorpas et al. (1998). After the completion of the digestion, the samples were vacuum filtered and the filtrate was used for the determination of heavy metal concentration by atomic absorption spectroscopy, using a Perkin-Elmer 2380 spectrophotometer (Norwalk, Connecticut, USA).

The chemical forms of heavy metals were determined by the use of the sequential extraction procedure of Tessier et al. (1979), with some modifications (Savvides et al., 1995). A known quantity (1 g) of each sample was treated with 1 M CH_3COONa for 1 h at room temperature and (pH 8.2) to extract the exchangeable phase and 1 M CH_3COONa for 5 h at room temperature (pH 5.0) to extract the carbonate phase. Using 0.04 M $NH_2OH \cdot HCl$ in 25% (v/v) CH_3COOH for 6 h at 96°C the fraction bound to Fe–Mn oxides was extracted. To extract the organic fraction 0.02 M HNO_3 and 30% (w/v)

H_2O_2 was used for 2 h at 85°C and pH 2.0, followed by the addition of 3.2 M CH_3COONH_4 in 20% (w/v) HNO_3 with 30% (w/v) H_2O_2 for 3 h at 85°C. The final residue was digested with conc. HF and conc. HNO_3 for 1 h at room temperature to extract the residual fraction. After each extraction step samples were centrifuged (5000 rpm for 30 min) and filtered. The filtrate was used for the determination of heavy metal concentration by atomic absorption spectroscopy, using a Perkin-Elmer 2380 spectrophotometer. For the characterization of all compost samples, the zeolites were removed from the final products. For the statistical analysis, Taylor's (1990) statistical techniques were used for data analysis and specifically the confidence interval (CI) for a mean.

3. Results and discussion

The water content, pH value, conductivity, total phosphorus (P–PO₄), organic matter (OM), total organic carbon (TOC), total Kjeldahl nitrogen (TKN) and ammonia content, C/N and C/P ratio of the DASPSS and of the compost samples are presented in Table 3.

The amount of zeolite added did not affect the total phosphorous and TKN of the final compost. Total phosphorous and TKN, were in the range between 2.50% and 1.50% for all the compost samples. Nitrogen is usually affected by the action of the proteolytic bacteria in the first step of composting (Kapetanios et al., 1993). At high temperature the nitrogen is lost to the atmosphere. Usually nitrogen appeared to increase during the first days of the composting process. This is caused by the decrease of the substrate carbon resulting from CO_2 loss (Kapetanios et al., 1993). It also appears that the ammonia content in the final product decreased when the amount of zeolite used was increased (0.821 mg/g for the A1 sample and 0.395 mg/g for the A6 sample). The compost quality, with respect to agricultural use, depends on its inorganic nitrogen content (Kapetanios et al., 1993). Inorganic N levels, should not exceed 10% of the total nitrogen, and ammonia content should be less than 0.04% of the dry matter. The final total concentration did not exceed the above limit, an indication of an optimal soil conditioner.

Organic matter and total organic carbon was lower in samples A5 and A6 compared to the other samples (Table 3). Zeolite as a bulking material has the ability to increase the porosity of the substrate and as a result, to improve the composting process and the biodegradability of the organic matter. In addition, carbon was lost to the atmosphere as CO_2. It was also observed that the moisture content retained in samples containing clinoptilolite was higher than that of the sample in which no clinoptilolite was added (A0).

The metal content in the DASPSS and final compost products is presented in Table 4. When comparing the metal content of the A0 sample, sewage sludge compost with no zeolite and DASPSS the concentration of Cr, Ni, Mn, Pb and Zn increased while that of Cu and Fe decreased. Composting can concentrate or dilute heavy metals present in sewage sludge. This change in metal concentration depends on the metal loss through leaching and on the overall concentration of metals due to organic matter destruction (Wagner et al., 1990).

Clinoptilolite has the ability to exchange sodium and potassium (Table 4). With increasing amounts of zeolite in the compost, the concentration of all heavy metals in the samples decreased while the concentration of sodium and potassium increased. In samples A5 and A6 containing 25–30% clinoptilolite, maximum metal ($p < 0.05$) uptake was 100% for Cd, 28–45% for Cu, 10–15% for Cr, 41–47% for Fe, 9–24% for Mn, 50–55% for Ni and Pb, and 40–46% for Zn.

The sequential extraction procedure was applied to the raw sludge (Fig. 1(a)), to sewage sludge compost (Fig. 1(b)), after 150 days with no zeolite and to the final products A5 and A6 where maximum metal was taken up at 150 days (Fig. 2). The metal partitioning in the DASPSS sample as determined by sequential chemical extraction is shown in Fig. 1(a). A significant ($p < 0.05$) amount of Cr and Cu was bound to the organic and residual fractions with less than 2% in the exchangeable and the carbonate fractions. Almost 60% of Cd and Mn was bound to the reducible fraction. Iron and Fe were found to be bound in the reducible and residual fractions (about 95% for the Fe and 83% for the Pb). Only Ni and Zn were found in substantial proportions in all phases, (very little in exchangeable phase).

Comparing the results with studies carried out previously (Angelidis and Gibbs, 1988; Rudd et al., 1988; Brennan, 1991; Garcia-Delgado et al., 1994), Cr and Cu were found in similar proportions in all fractions while Fe was mainly associated with the residual and reducible ones. Ni was observed to yield substantial proportions in all phases, whereas Zn was mostly found in the organic and reducible fractions. As far as Pb was concerned, the reducible, organic and residual fractions were over 90%.

Comparing the results of Figs. 1(a) and (b), Cd, Fe and Zn were not affected during the composting process. Additionally, Cr seems to be transformed from the reducible and the organic to the residual fraction. The organic fraction of Cu changed to other fractions, especially residual and exchangeable. The residual fraction increased from 27.53% to 45.37% and exchangeable from 1.55% to 10.22%. Approximately 72% of Pb was bound in the residual fraction. A substantial percentage of Mn was removed from the reducible, carbonate and organic fractions and transferred to the residual fraction. The changes observed in metal partitioning of the

Table 3
DASPSS and cured compost characteristics

Parameter[a]	DASPSS	Cured compost[b]						
		A0	A1	A2	A3	A4	A5	A6
Moisture (%)	70.00±2.00	25.20±1.50	30.10±1.00	32.25±1.00	33.42±1.50	31.05±1.50	30.85±1.00	30.29±1.00
pH	7.02±0.05	7.01±0.05	7.31±0.05	7.21±0.05	7.29±0.05	7.25±0.05	7.25±0.05	7.25±0.05
Conductivity (mS/cm)	1.001±0.005	1.101±0.005	0.987±0.003	0.895±0.003	0.877±0.003	0.987±0.005	0.875±0.005	0.857±0.002
OM (%)	45.00±2.50	32.81±1.00	32.50±1.50	32.93±1.50	29.60±0.80	29.02±0.80	28.36±1.00	27.67±0.95
TOC (%)	26.51±1.50	19.03±1.00	18.81±1.50	19.01±1.50	17.82±0.80	17.08±0.80	16.03±1.00	15.83±0.95
TKN (%)	1.62±0.12	1.65±0.15	1.58±0.12	1.45±0.15	1.52±0.10	1.44±0.11	1.43±0.10	1.41±0.13
TP (P–PO$_4$) (%)	2.51±0.05	2.48±0.05	2.48±0.05	2.50±0.05	2.52±0.05	2.45±0.05	2.50±0.05	2.47±0.05
N–NH$_4$[c]	0.956±0.025	0.739±0.035	0.821±0.020	0.602±0.020	0.531±0.028	0.502±0.015	0.429±0.010	0.395±0.005
N–NO$_3$	0.253±0.015	0.262±0.015	0.253±0.020	0.269±0.020	0.275±0.015	0.260±0.015	0.259±0.015	0.284±0.010
N–NO$_2$ E-6[c]	258±35	260±25	245±20	269±20	270±25	271±18	265±15	266±10
C/N	16.31±1.50	11.53±1.00	11.90±1.50	13.11±1.50	11.72±1.80	11.86±1.40	11.20±1.30	11.22±1.25
C/P	10.56±0.50	7.67±1.05	7.58±1.50	7.61±1.50	7.07±0.80	6.97±0.80	6.42±1.00	6.41±0.90

[a] All values were determined in dried matter, except moisture.
[b] Mean value of 20 samples ± standard deviation.
[c] mg/g dry sludge.

Table 4
Metals content in DASPSS and compost

Metal[a]	DASPSS	Cured compost[b]						
		A0	A1	A2	A3	A4	A5	A6
Cd	0.002 ± 0.0005	0.002 ± 0.0001	n.d.[c]	n.d.	n.d.	n.d.	n.d.	n.d.
Cr	0.552 ± 0.025	0.578 ± 0.023	0.552 ± 0.023	0.550 ± 0.023	0.542 ± 0.020	0.501 ± 0.024	0.488 ± 0.025	0.478 ± 0.025
Cu	0.258 ± 0.005	0.205 ± 0.005	0.265 ± 0.006	0.184 ± 0.006	0.181 ± 0.004	0.172 ± 0.004	0.163 ± 0.006	0.140 ± 0.007
Fe	5.098 ± 0.310	4.118 ± 0.225	3.963 ± 0.170	3.838 ± 0.125	3.217 ± 0.125	3.191 ± 0.150	2.999 ± 0.150	2.673 ± 0.100
Ni	0.041 ± 0.003	0.045 ± 0.003	0.040 ± 0.002	0.038 ± 0.001	0.038 ± 0.001	0.034 ± 0.001	0.019 ± 0.002	0.018 ± 0.001
Mn	0.150 ± 0.003	0.168 ± 0.005	0.150 ± 0.003	0.149 ± 0.003	0.139 ± 0.002	0.136 ± 0.003	0.136 ± 0.002	0.114 ± 0.001
Pb	0.326 ± 0.004	0.335 ± 0.004	0.199 ± 0.004	0.187 ± 0.003	0.178 ± 0.002	0.177 ± 0.003	0.157 ± 0.004	0.145 ± 0.005
Zn	1.739 ± 0.028	1.801 ± 0.030	1.400 ± 0.045	1.216 ± 0.037	1.117 ± 0.040	1.083 ± 0.035	1.027 ± 0.025	0.938 ± 0.027
K	0.723 ± 0.020	0.772 ± 0.012	0.772 ± 0.018	0.902 ± 0.013	0.965 ± 0.015	1.121 ± 0.025	1.215 ± 0.033	1.235 ± 0.045
Mg	1.132 ± 0.010	0.796 ± 0.012	0.786 ± 0.014	0738 ± 0.017	0.695 ± 0.009	0.620 ± 0.009	0.589 ± 0.010	0.538 ± 0.009
Na	0.724 ± 0.006	0.732 ± 0.006	0.783 ± 0.007	0.796 ± 0.007	0.865 ± 0.009	0.901 ± 0.012	1.011 ± 0.011	1.121 ± 0.010
Ca	13.827 ± 1.055	14.001 ± 1.185	13.159 ± 1.150	12.414 ± 1.005	11.825 ± 1.015	11.613 ± 1.005	10.320 ± 0.958	10.002 ± 0.950

[a] mg/g dry sludge; zeolite has been removed from the final products.
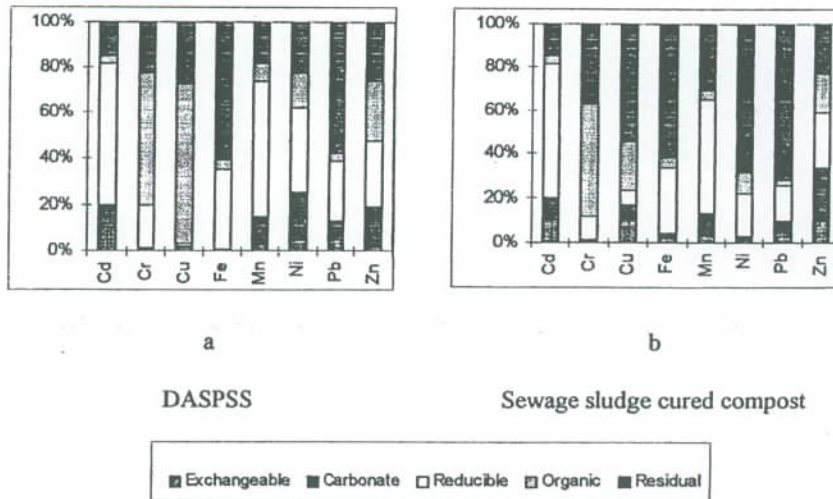[b] Mean value of 20 samples ± standard deviation.
[c] n.d.: not detected.

a   b

DASPSS   Sewage sludge cured compost

☑ Exchangeable ■ Carbonate ☐ Reducible ▨ Organic ■ Residual

Fig. 1. Metals partitioning in DASPSS and in sewage sludge cured compost.

A5   A6

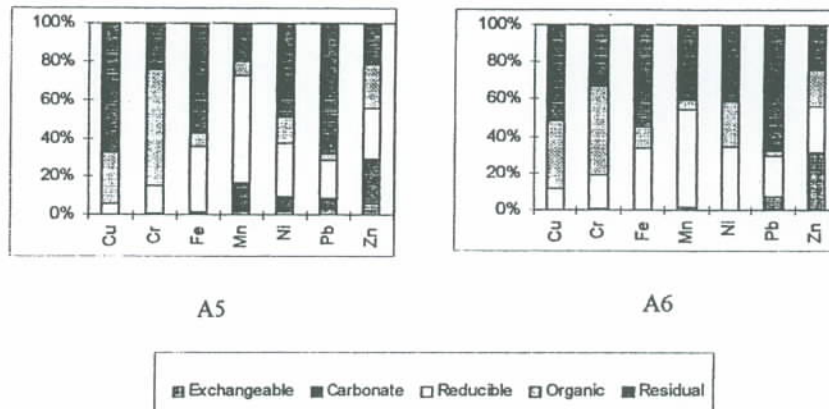☑ Exchangeable ■ Carbonate ☐ Reducible ▨ Organic ■ Residual

Fig. 2. Metals partitioning in cured compost.

sewage sludge compost are the results of several factors. The thermophilic phase of composting, which is the first step of composting, affects the exchangeable fraction. During the first step of composting there are changes in pH value (acetic acid formation) and in ammonia content (affected by the action of the proteolytic bacterial and by the temperature) (Epstein, 1997; Kapetanios et al., 1993,1990). The pH changes, which occur in the beginning of the composting process, affect the exchangeable and carbonate fraction. The pH changes may be due to acid formation during the decomposition of organic matter contained in the sludge (Wong et al., 1995). The oxic and anoxic conditions (produced by acetic acid and ammonia) at the first step of composting (Kapetanios et al., 1993,1990), affect the reducible and organic fractions.

Copper in the compost (sample A5 and A6) was bound in the residual fraction 47–67% (Fig. 2). Chromium in the residual fractions was approximately 20–

32%. A substantial amount (47–63%) of Cr was in the organic fraction. Iron was primarily bound in the residual fraction (67%). The other three fractions contained less than 8%, most of which was bound in the organic fraction. Seventy-five percent of the Ni was bound in the residual and in the organic fraction, while Mn was found in the reducible fraction at 48% and in the residual at 35%. In the case of Zn all fractions for samples A5 and A6 had substantial proportions of metal, while lead was found to be bound in the residual and reducible fractions by almost 90%. It was observed that clinoptilolite has the ability to readily take up almost all metals that are bound to the exchangeable and the carbonate fractions. Clinoptilolite took up all metals bound in exchangeable and carbonate fractions according to the following selectivity series: Cu > Cr > Fe > Ni > Mn > Pb > Zn.

Some researchers (Manios and Stentiford, 1997) observed that 50% of Cd, Cu and Zn was associated with

the residual fraction in compost produced from sewage sludge and the organic fraction of municipal solid waste. Nickel was found in 35% and almost 50% in the residual and exchangeable fractions, respectively, while Pb was 80% associated with the residual fraction. Also Sims and Kline (1991) determined that the concentrations of Cd, Cr, Cu, Ni, Pb and Zn were associated with the more resistant fractions in co-composted sewage sludge.

4. Conclusion

While the amount of added natural zeolite (clinoptilolite) increased in compost pile, the concentration of heavy metals in the final compost decreased. By using 25–30% of clinoptilolite during the composting process 100% of Cd, 28–45% of Cu, 10–15% of Cr, 41–47% of Fe, 9–24% of Mn, 50–55% of Ni and Pb, and 40–46% of Zn was taken up by the zeolite. Using a sequential extraction procedure in the raw sludge (DASPSS) and final products (compost after 150 days) it was found that a significant ($p < 0.05$) amount of sludge metals was bound in the residual fraction, characterized as an inert form. Clinoptilolite most readily took up the metal content bound in the exchangeable and carbonate fractions.

References

APHA, AWWA-WPCF, 1985. Standard methods for the examination of water and wastewater. American Public Health Association, Washington, USA, pp. 479–480.

Adams Dean, V., 1990. Water and wastewater examination manual. Lewis Publishers, Chelsea, MI. pp. 45–47, 121–139, 186–188.

Angelidis, M., Gibbs, R.J., 1988. Chemistry of metals in anaerobically treated sludge's. Water Res. 23, 29–33.

Brennan, B., 1991. Chemical partitioning and remobilization of heavy metals from sewage sludge dumped in Dublin Bay. Water Res. 25, 1193–1198.

Constantopoulou, C., Loizidou, M., Loizou, Z., Spyrellis, N., 1994. Thorium equilibria with the sodium form of clinoptololite and mordenite. J. Radioanal. Nucl. Chem. 178 (1), 143–151.

Drougas, A., Fatsea, H., Kambouri, E., Kapetanios, E., 1992. The problem of sewage sludge management of Psittalia island waste water treatment plant in Attica. ACTA Hort. Int. Soc. Hort. Sci. 302, 105–112.

Epstein, E., 1997. The Science of Composting. Technomic Publishing, Pennsylvania, USA.

Finstein, M.S., Miller, F.C., MacGregor, S.T., Psarianos, K.M., 1992. The Rutgers strategy for composting: process design and control. ACTA Hort. Int. Soc. Horticultural Sci. 302, 75–86.

Garcia-Delgado, R.A., Garcia-Herruzo, F., Gomez-Lahoz, C., Rodriguez-Maroto, J.M., 1994. Heavy metals and disposal alterna-

tives for an anaerobic sewage sludge. J. Environ. Sci. Health A 29 (7), 1335–1347.

Handbook of Reference Methods for Soil Analysis, 1992. Georgia University Station, Athens, USA, pp. 172–174.

Kapetanios, E.G., Loizidou, M., Valkanas, G., 1993. Compost production from Greek domestic refuse. Bioresource Technol. 43, 13–16.

Kapetanios, E.G., 1990. Production and use of compost from municipal waste. Heavy metal uptake using natural clinoptilolite. Ph.D. Thesis. National Technical University of Athens, Athens, Greece.

Kapetanios, E., Loizidou, M., 1992. Heavy metals removal using zeolites in tomato cultivation in compost. ACTA Hort. Int. Soc. Hort. Sci. 302, 63–74.

Lindau, C., Hossner, L., 1982. Sediment fractionation of copper, nickel, zinc, chromium, manganese and iron in one experimental and three natural marshes. J. Environ. Qual. 11, 540–545.

Loizidou, M., 1989. Heavy metals in the effluents of the metal plating industries and their removal by zeolites. Seventh International Conference on Heavy Metals in the Environment. Geneva.

Maliou, D., 1994. Heavy metal removal from waste waters solution using natural zeolite. Ph.D. Thesis. National Technical University of Athens, Athens, Greece.

Manios, T., Stentiford, E.I., 1997. Heavy metals fractionation before, during and after composting of urban organic residues. International Conference on Organic Recovery and Biological Treatment into the next millenium. Harrogate, UK, pp. 227–234.

Methods of Sampling and Analysis of Solid Wastes, 1970. EAWAG, Swiss Federal Institute for Water Supply, Sewage Purification and Water Pollution Control. Dubendorf, Switzerland, p. 55.

Papadopoulos, A., Fatta, D., Loizidou, M., 1997. Stabilised Landfill Leachate and its Treatability. In: Proceeding of the Fifth Conference on Environmental Science and Technology. Molyvos Lesvos, Greece, pp. 337–346.

Pierce, F.J., Dowdy, R., Grigal, D., 1982. Concentrations of six trace metals in some major Minnesota soil series. J. Environ. Qual. 11, 416–422.

Rudd, T., Campbell, J.A., Lester, J.N., 1988. The use of model compounds to elucidate metal forms in sewage sludge. Environ. Pollut. 50, 225–242.

Sims, J.T., Kline, J.S., 1991. Chemical fractionation and plant uptake of heavy metals in soils amended with co-composted sewage sludge. J. Environ. Qual. 20, 387–395.

Savvides, C., Papadopoulos, A., Haralambous, K.J., Loizidou, M., 1995. Sea sediments contaminated with heavy metals: metal speciation and removal. Water Sci. Technol. 32, 65–73.

Taylor, K.J., 1990. Statistical Techniques for Data Analysis. Lewis Publishers, INC, Florida, USA, pp. 42–45, 66–68.

Tessier, A., Campbell, P.G.C., Bisson, M., 1979. Sequential extraction procedure for the speciation of particulate trace metals. Anal. Chem. 51, 844–851.

Wagner, D.J., Bacon, G.D., Knocke, W.R., Switzenbaum, M.S., 1990. Changes and variability in concentration of heavy metals in sewage sludge during composting. Environ. Technol. 11, 949–960.

Wong, J.W.C., Li, S.W.Y., Wong, M.H., 1995. Coal fly ash as a composting material for sewage sludge: effects on microbial activities. Environ. Technol. 16, 527–537.

Zorpas, A.A, Vlyssides, A.G., Loizidou, M., 1998. Physical and chemical characterization of anaerobically stabilized primary sewage sludge. Fres. Environ. Bul. 8, 502–508.