Heavy Metal Content in Sewage Sludge: A Management Strategy for an Ocean Island

Contenido de metales pesados en lodos de depuradora: estrategia de gestión para una isla oceánica

Teor de metais pesados nas lamas de tratamento de águas residuais: estratégia de gestão para uma ilha oceânica

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Cita: Hernández Sánchez C, Gutiérrez A, Galindo JM, González-Weller D, Rubio C, Revert C, Burgos A, Hardisson A. Heavy Metal Content in Sewage Sludge: A Management Strategy for an Ocean Island. Rev. salud ambient. 2017; 17(1):3-9.

Recibido: 30 de julio de 2016. **Aceptado:** 9 de marzo de 2017. **Publicado:** 15 de junio de 2017.

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Financiación: Este grupo no ha contado con ningún tipo de financiación para el desarrollo de su trabajo.

Declaración de conflicto de intereses: Los autores declaran que no existen conflictos de intereses que hayan influido en la realización y la preparación de este trabajo.

Declaraciones de autoría: Todos los autores contribuyeron al diseño del estudio y la redacción del artículo. Asimismo, todos los autores aprobaron la versión final.

Abstract

In recent years, the generation of sewage sludge has increased worldwide. Correct processing and management of this waste concerns all countries. This work presents a study of metal contents, i.e. of Cd, Pb, Zn, Cr, Cu, and Ni, in sewage sludge from a wastewater treatment plant in the northeastern region of the island of Tenerife. The study aimed at examining the sludge for potential suitability as a farmland fertilizer. Detected metal levels for Pb, Zn, Cr, Cu, and Ni were extremely low (26.44, 544.01, 24.10, 37.05, and 8.04 mg/kg dw [dry weight], respectively). Cadmium levels were under quantification limit. Season-dependent, statistically significant differences ($p < 0.05$) in metal concentrations in sewage sludge were observed for Cu, Ni, Cr, and Pb. Consequently, the application of sewage sludge to fertilize nutrient-deficient agricultural soils and soils degraded by human activity represents a fast and straightforward solution to the lack of such resources, particularly in an oceanic island.

Keywords: heavy metals; oceanic island; sewage-sludge; waste-management.

Resumen

Durante los últimos años, la generación de lodos de depuradora ha aumentado en todo el mundo. El tratamiento y la gestión correctos de estos residuos son una cuestión que afecta a todos los países. Este trabajo presenta un estudio del contenido de metales, i.e. Cd, Pb, Zn, Cr, Cu y Ni, de los lodos de depuradora de una planta de tratamiento de aguas residuales situada en la zona noreste de la isla de Tenerife. El estudio tuvo por objeto estudiar los lodos para ver su posible idoneidad como fertilizante agrícola. Los niveles de metal detectados para Pb, Zn, Cr, Cu y Ni fueron extremadamente bajos (26,44, 544,01, 24,10, 37,05 y 8,04 mg/kg [peso seco], respectivamente). Los niveles de cadmio quedaron por debajo del límite de determinación. Se observaron diferencias dependientes de las estaciones que son estadísticamente significativas ($p < 0{,}05$) en las concentraciones de Cu, Ni, Cr y Pb. Por consiguiente, el uso de lodos de depuradora para fertilizar suelos agrícolas que son pobres en nutrientes o que han sido degradados por la actividad humana es una solución rápida y sencilla a la escasez de tales recursos, particularmente en islas oceánicas.

Palabras clave: metales pesados; isla oceánica; lodos de depuradora; gestión de residuos.

Resumo

Nos últimos anos, a produção de lamas de águas residuais aumentou em todo o mundo. O tratamento e a gestão corretos deste resíduo são uma preocupação de todos os países. Este trabalho apresenta um estudo sobre os teores de metais, ou seja, Cd, Pb, Zn, Cr, Cu e Ni, em lamas duma estação de tratamento de águas residuais no nordeste da ilha de Tenerife. O estudo teve como objetivo analisar as lamas quanto à sua potencial adequação como fertilizante de terras agrícolas. Os níveis de metais detetados para Pb, Zn, Cr, Cu e Ni foram extremamente baixos (26,44, 544,01, 24,10, 37,05 e 8,04 mg/kg dw [peso seco], respetivamente). Os níveis de cádmio estavam abaixo do limite de quantificação. Foram observadas diferenças, estatisticamente significativas ($p < 0{,}05$), dependentes da estação do ano nas concentrações de Cu, Ni, Cr e Pb. Consequentemente, a aplicação de lamas para fertilizar agriculturas, solos deficientes em nutrientes e solos degradados pela atividade humana representa uma solução rápida e direta para a insuficiência de tais recursos, particularmente numa ilha oceânica.

Palavras-chave: metais pesados; ilha oceânica; lamas de águas residuais; gestão de resíduos.

INTRODUCTION

Wastewater Treatment Plants (WWTPs) generate millions of tons of sewage sludge globally every year[1-3]. After the enactment of Directive 97/271/EEC, which regulates the treatment of urban wastewater, the production of sewage sludge as a by-product of water treatment has increased[4-8].

To date, there are three alternatives for handling wastewater sludge within the European Union: incineration, land burial, and application as a farmland fertilizer. Each European member state employs these options in a different way. Thus, in Spain, the law prohibits burying sludge so as to not affect the groundwater[2]. Incineration of sewage sludge creates an important problem, as the ash contains toxic metals such as lead, cadmium, copper, and zinc, as well as dioxins and furans, which could pass into the atmosphere[9]. In the European Union, 30 % of sewage sludge is reused as fertilizer in agriculture[10]. Other countries like the US, New Zealand, and Australia not only apply wastewater sludge in agriculture but also legislate its reuse in forest, mountain and other soils[11].

Wastewater-derived sludge is high in nutrients and organic matter content, especially nitrogen and phosphorus, which could make it an attractive fertilizer[12,13]. Multiple studies have shown that sewage sludge, applied as an organic supplement on farmland, produces a substantial increase in soil fertility and hence crop yield[14]. On the whole, water treatments concentrate pollutants from wastewater in the sewage sludge[15]. Therefore, this sludge must be reused in environmentally safe conditions[9].

Since 1990, Spanish law permits the reuse of sludge in agriculture, stipulating concentrations of heavy metals below the provisions of Royal Decree (RD) 1310/1990[16] (which implements European Directive 86/278/EEC[17]). Both the EU Directive and the Spanish RD set limits on sewage sludge application on soil for 7 metals: Cd, Cr, Cu, Pb, Hg, Ni, and Zn. The US Environmental Protection Agency (USEPA) regulates 10 metals (Cd, Cr, Cu, Pb, Hg, Ni, Zn, As, Mo, and Se). Several European countries impose additional limits for pollutants other than metals[8].

EU and USEPA legislation[18] differ in the annual pollutant loading rate limits (kg/ha/year) set for the different heavy metals, the European Directive being much more restrictive and thus limiting.

Studies of metal concentrations in sewage sludge from around the worldshow much variation[1,2,7,8,10,19,20-27], largely due to the characteristics of the wastewater. Therefore, each WWTP requires a study to determine its suitability to generate sewage sludge for fertilizing, as the pollution load varies with the location. Obviously, completion of appropriate sludge management entails substantial efforts and financial costs[5,28-30].

The Canary Islands are considered an outermost region for lying in the most southern point of Europe, at a distance of more than 1,000 km from the Iberian Peninsula. High transport costs, due to their isolated location, impact and increase the cost of all import products. The Island of Tenerife, with a land area of 2,058 square kilometers and 852,945 inhabitants, is the largest of the seven Canary Islands[31] and has an economy which is primarily based on tourism and agriculture. Therefore, agricultural production is costlier due to high transport costs. Agriculture should not be the only sector to take advantage of this high-quality sludge for fertilization. It could also be applied to other types of nutrient-deficient soils or soils degraded by human activity.

The *Plan Territorial Especial de Ordenación de Residuos de Tenerife* (the specific territorial regulation for waste management in Tenerife), passed by the Canary Islands' Government[32], proposes composting all sewage sludge generated in the island, but only a small portion is used for gardening purposes. In summary, the use and proper management of sludge from sewage treatment plants would turn waste from being a landfill problem into a high quality resource. Moreover, preventing disposal of sewage sludge in landfills could help avoiding added problems of space, which are always a critical issue in the limited territory of islands.

The aim of this study was to evaluate the annual distribution pattern of metals (Cd, Pb, Zn, Cr, Cu, and Ni) in sewage sludge from the WWTP in the northeastern region of Tenerife.

MATERIALS AND METHODS

The regional wastewater system in the northeastern region of Tenerife has a WWTP for secondary treatment of wastewater from the area. The slurry in this WWTP goes through a primary decanter, a clarifier, and a thickener, and finally passes through a band filter. 60 samples of sewage sludge were taken from this WWTP between August 2013 and July 2014, divided into one sample per day.

Homogenized samples were weighed into 1 g aliquots and oven-dried at 80 °C for 20-24 h. Next, samples were incinerated in a muffle furnace by increasing the temperature every 2 hours in 30 °C steps until reaching 450 °C ± 25 °C for a total of 30 h. After a cool-down period, the resulting white ash was dissolved in 1.5 % nitric acid and completed to a volume of 50 ml. Processed samples were stored in antiseptic polyethylene bottles until analysis.

Metal contents were analyzed by atomic absorption spectrophotometry (AAS), a reference technique for metal determination that is characterized by its high sensitivity and reproducibility. A 2100 Flame Atomic Absorbtion Spectrophotometer from Perkin Elmer (MA, USA) was used.

The detection and quantification limits were estimated based on the instrumental response of the equipment. Specificity was confirmed by analyzing 15 white/blank samples in reproducible conditions.[33] The instrumental detection and quantification limits were 47.54 µg/L and 158.47 µg/L respectively for Cd: 37.61 µg/L and 125.36 µg/L respectively for Pb: 19.03 µg/L and 63.43 µg/L respectively for Zn: 40.86 µg/L and 136.21 µg/L respectively for Cr; 26.76 µg/L and 89.18 µg/L respectively for Cu; and 33.68 µg/L and 112.28 µg/L respectively for Ni.

Quality control was carried out using Standard Reference Material 2782 (Industrial Sludge; USA). The recovery rates obtained with the reference materials were all above 95 % (recovery rates: 101.39 % for Pb; 92.66 % for Zn; 87.89 % for Cr; 92.98 % for Cu and 91.50 % for Ni). During all the analytical processes, each batch of 20 samples was analyzed together with at least a blank and a reference sample. Calibration was performed using the calibration curve method.

For statistical analysis, data was processed using SPSS software (V19.0 for Windows). The normal distribution of the data was tested against the Kolmogorov-Smirnov model[34], and Levene's test was applied to determine variance homogeneity[35]. For inferential statistics, an ANOVA (post-hoc Tukey's test) was applied as a parametric test, and the Mann-Whitney test and the Kruskal-Wallis test as non-parametric tests[36]. Correlation between variables was determined by means of the Spearman and Pearson tests.

RESULTS

Monthly concentrations (mg/kg dw [dry weight]) of the five evaluated heavy metals (Pb, Zn, Cr, Cu, and Ni) in sewage sludge from the WWTP in the northeastern region of Tenerife are shown in Table 1.

The heavy metal contents of that sludge, listed from lower to higher concentrations (mg/kg), were as follows: Ni (8.04) < Cr (24.10) < Pb (26.44) < Cu (37.05) < Zn (544.01). Cd concentrations were below the quantification limit but above the detection limit.

Statistical analyses showed significant differences ($p < 0.05$) among the concentrations of each of the studied metals and the period of the year in which the sludge was collected. Cr and Ni had the highest concentrations in July and August, and Zn also had its peak in July. However, concentrations of Cu and Pb decreased during the summer months.

Table 1. Monthly Concentration (mg/kg dw) of Metals in the Waste Water treatment Plant of the Study

Metals	Pb	Zn	Cr	Cu	Ni
August 2013	9.71±2.94	357.24±75.30	29.82±7.38	18.44±2.82	12.41±1.69
September 2013	20.08±5.81	365.61±37.13	21.21±4.70	45.88±10.01	6.41±2.15
October 2013	26.28±8.98	608.43±265.21	23.13±11.13	51.78±14.92	9.07±2.85
November 2013	14.02±1.81	566.31±43.24	20.64±1.39	53.24±23.08	9.6±1.25
December 2013	28.16±0.95	672.68±213.48	21.82±1.26	45.33±2.16	7.19±0.63
January 2014	32.03±8.91	543.57±28.40	24.12±1.80	47.13±2.18	8.15±0.54
February 2014	34.62±15.39	582.41±88.53	26.44±0.88	48.83±7.84	6.7±0.70
March 2014	39.37±16.48	518.93±66.08	24.46±1.51	38.19±2.29	7.68±0.15
April 2014	34.78±9.80	462.54±28.31	21.44±2.04	22.65±2.12	6.09±0.77
May 2014	28.40±4.26	534.88±39.37	20.42±2.18	22.79±3.46	5.20±0.75
June 2014	22.85±3.48	569.88±52.65	23.08±1.70	26.24±1.53	10.76±3.05
July 2014	27.02±3.29	745.66±133.97	32.61±6.92	24.11±2.12	7.19±0.26
Average concentration	26.44	544.01	24.10	37.05	8.04

Table 2. Maximum Concentrations of Metals Permitted in Sludge by the EPA (1996) and Royal Decree (1990)

Metals (mg/kg dw)		Pb	Zn	Cr	Cu	Ni
Average concentration in this study		26.44	544.01	24.10	37.05	8.04
Maximum concentrations of metals permitted by Royal Decree (1990)	Soils with pH < 7	750	2500	1000[b]	1000	300
	Soils with pH > 7	1200	4000	1500[b]	1750	400
Percentage of the maximum concentrations of metals permitted by Royal Decree (1990) in the analyzed sludge	Soils with pH < 7	3.53 %	21.76 %	2.41 %	3.70 %	2.68 %
	Soils with pH > 7	2.20 %	13.60 %	1.61 %	2.12 %	2.01 %
Maximum concentrations of metals permitted by the EPA (1996)		300	2800	1200[c]	1500	420
Percentage of the maximum concentrations of metals permitted by the EPA (1996) in the studied sludge		8.81 %	19.43 %	2.01 %	2.47 %	1.91 %

[a] The European Directive does not set this value. The value set by Spanish RD 1310/1990 has been used
[b]EPA is re-examining these limits

Table 2 shows the metal contents of the samples from the WWTP of the study in Tenerife compared to the limits set by European Directives 86/278/EEC[37] and 98/15/EEC[38] and the USEPA[18]. In addition, data from the sludge of the studied WWTP are presented as percentages from the European Directives as well as the USEPA's limits.

Concentrations of the heavy metals Cd, Pb, Zn, Cr, Cu, and Ni in sewage sludge from the WWTP in the northeastern region of Tenerife are well below the limits set by European Directive 86/278/EEC and the USEPA (1996). In order to know which of these five metals would be the metal that limits the use of this sludge on soils, we proceeded to calculate the percentages of each metal. Percentages of mean metal contents with respect to the limits set by European Directive 86/278/EEC, listed in increasing order, were: Cr (2.41 %) < Ni (2.68 %) < Pb (3.53 %) < Cu (3.70 %) < Zn (21.76 %) for soils with a pH < 7, and Cr (1.61 %) < Ni (2.01 %) < Cu (2.12 %) < Pb (2.20 %) < Zn (13.60 %) for soils with a pH > 7. The increasing order of percentages of the studied metals referring to USEPA limits is: Ni (1.91 %) < Cr (2.01 %) < Cu (2.47 %) <Pb (8.81 %)

< Zn (19.43 %). Zn had the highest percentages with respect to both limits.

It is of great importance to know the soils' pH when applying any sewage sludge because an acid soil allows greater bioavailability of metals to biota, which may lead to a public health problem.

Table 3 shows a comparison of our results with other Spanish studies. The metal concentrations found in our study were lower than those published for other WWTPs except for Cr, the concentration of which was even lower in sludges from the WWTP of Lugo[27]. Table 4 shows a comparison of our results with sewage sludge studies conducted in other countries. Pb concentrations in the present study turned out to be among the lowest. As for Cu and Ni, values from the study object were only higher than the values found in Italy and Malaysia. Finally, the Zn concentrations observed in the Tenerife wastewater were below the values from other wastewater from around the world except Jalisco (Mexico), Tunisia, Coimbra (Portugal), Italy, China, and Malaysia (Tables 3 and 4).

Table 3: Comparison with Other Spanish WWTP Studies (mg/kg dw)

WWTP	Cd	Pb	Zn	Cr	Cu	Ni	Reference
Nordeste, Tenerife	< QL	26.44	544.01	24.10	37.05	8.04	Thisstudy
Albacete	1.54	110	1455	117	370	77.54	Heras *et ál.*, 2005[2]
Lezama (Biskaia)	5.7	155	8488	803	605	167	Egiarte *et ál.*, 2008[7]
Lugo	1.5	184	1320	74	241	49	Mosquera-Lozada *et ál.*, 2009[25]
Lugo	<0.01	104	753.1	3.9	121.2	95.3	Rigueiro-Rodríguez *et ál.* 2010[29]
Madrid	-	385	1635	468	460	79.6	Cuevas and Walter, 2004[1]
Madrid	4	403	1490	438	438	77.8	Beltrán *et ál.*, 2005[21]
Madrid	2.55	50	1100	210	230	53	Roca-Pérez *et ál.* 2009[26]

QL: Quantification Limit

Table 4. Comparison with Other International WWTP Studies (mg/kg dw)

WWTP	Cd	Pb	Zn	Cr	Cu	Ni	Reference
Nordeste, Tenerife	<QL	26.44	544.01	24.10	37.05	8.04	Thisstudy
Jalisco (Mexico)	1.08	117.22	539.9	22.16	166	9.69	Salcedo-Pérez *et al.*, 2007[39]
Venezuela	6.8	304.29	1474.79	72.81	226.01	76.46	García *et ál.*, 2006[40]
Tunisia	3.3	325	410	52	278	44	Acosta *et ál.* 2003[19]
Greece	1.2	191	728	134	599	99	Manios *et ál.*, 2003[41]
Coimbra (Portugal)	ND	48	500	29	124	15	Moreira *et ál.*, 2008[8]
Gothenburg (Sweden)	1.64	43.79	748.7	23.42	501.9	24.7	Ahlberg *et ál.*, 2006[23]
Italy	<2.0	13.4	152.8	22.7	20.1	11.0	Goi*et ál.*, 2006[42]
Austria	0.82	38.3	683	30.6	166	25.6	Sager, 2007[43]
Poland	1.90	59.5	1385	85.0	201	21.7	Oleszczuk, 2008[44]
Changsha (China)	7.2	152	424.8	-	110	-	Chen *et ál.*, 2008[24]
Xiangtan (China)	15.7	71.8	444.6	-	159.6	-	Chen *et ál.*, 2008[24]
Zhuzhou(China)	7.9	98.4	361.0	-	67.0	-	Chen *et ál.*, 2008[24]
Shenyang (China)	5	255	290	-	170	-	Wang *et ál.*, 2008[10]
India	16	340.5	2164	-	1434.5	168	Kandpal *et ál.*, 2004[20]
Malaysia	8	10	200	500	80	-	Haroun *et ál.*, 2009[45]

ND: Not Detected
QL: Quantification Limit

DISCUSSION

The island of Tenerife and the Canary Islands as a whole have little industry and, therefore, only modest pollution. As expected, sewage sludges produced in this little industrialized area are poorer in toxic heavy metals than sludges from WWTPs located in larger industrialized areas.

We, therefore, propose using this high quality sewage sludge as fertilizer matter for agricultural soils in the island, which would lead to a cost reduction by eliminating the need for imported fertilizer and its transport costs.

Moreover, preventing disposal of sewage sludge in landfills could help to avoid added space problems, which are always a critical issue in the limited territory of islands. Therefore, reuse of this waste would create a resource and an economic advantage for farming on the island.

ACKNOWLEDGMENTS

The authors are deeply indebted to the *Consejo Insular de Aguas de Tenerife* and specifically the Área *de Infraestructura Hidráulica* for facilitating sampling in the WWTP in the northeastern reation of Tenerife.

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