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Agronomic characterisation of different types of sewage sludge: Policy implications

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ABSTRACT

Spain is one of the main municipal sewage sludge producers of Europe. This paper aims to agronomically characterise different types of sewage sludge stabilised by different methods (anaerobically digested, composted, and pelletised) and deliver policy recommendations from the results of this characterisation. Anaerobic sewage sludge quality is found to be better in plants with a lower volume of water processing. Composted sludge shows the best quality from a heavy metal point of view, but its low available nitrogen content increases the input of heavy metals when spread, as compared to digested or pelletised sludge. Pelletised sludge has higher heavy metal content than anaerobically digested sludge. Despite the good quality of the sludges, future regulations, especially with regard to Cd levels, will limit the use of this waste in agriculture.

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1. Introduction

The preservation of continental water quality is a need for both human beings and for the maintenance of high ecosystem standards. The Community Directive 91/271/EEC on urban waste water treatment (UWWTD) has made it compulsory to provide waste water treatment stations in all agglomerations equivalent to a population of more than 2000 people, in all member states by the 31st of December 2005 (EEA, 2005). During the water depuration process, sludge is produced as the water is cleaned for disposal into a river.

The significant amount of sludge produced by the EU generates an important problem of elimination. Spain is one of the largest sewage-sludge-producing countries in Europe. Article 14 of the UWWTD stipulates that “sludge arising from waste water treatment shall be re-used whenever appropriate”. This is based on a long-term solution of recycling sludge, once its quality can meet environmental protection and public health protection requirements. The criteria for agriculture sewage sludge use are usually based on nitrogen content (EPA, 1994) for two reasons: (a) this is one of the three main macronutrients, together with P and K, needed for increasing crop production, (b) it has greater environmental problems, due to its complex cycling, multiple loss methods (volatilisation and leaching of different compounds), and problems of in-soil retention (mostly associated with organic matter). At the present time, only the heavy metals in sewage sludge are legally regulated for use in agriculture in Europe. The total hea-

vy metal content in soil from sewage sludge is restricted as well as the maximum annual mean quantity of heavy metal application in soils over a period of 10 years, to meet quality requirements for the application of this waste in agricultural soils. Zn, Cu, Cr, Ni, Cd, Pb, and Hg are the heavy metals that are currently regulated, but others are also regulated in different European countries (Smith, 1996).

The use of sewage sludge in agriculture includes several operations that improve its efficiency in crop production, as compared to mineral fertilisers, which are related to the stabilising process before spreading. These are principally aerobic digestion, anaerobic digestion, composting, liming, and pelletisation.

The United States Environmental Protection Agency (EPA, 1994) has pointed out that clear differences exist with regard to mineralisation, depending on the type of stabilisation used for the sludge. For example, for anaerobic sludge, the proportion of total available nitrogen in the first, second, and third year is 20%, 10%, and 15%, respectively. If stabilisation is accomplished by composting, these figures are 10%, 5%, and 2.5%, respectively, although this data should be adjusted for different geographical environments.

In the third draft of the document on bio-wastes (EC, 2000) that intends to mark specific guidelines for complying with the objectives of the Landfill Waste Directive (1999/31/EC), it is established, for the employment of sludge, that “the ground should be protected and that the treated wastes should be beneficial for agriculture, produce ecological improvements, and guarantee that human, plant and animal health is not affected by the use of sludge”. Moreover, European regulations favour the employment of sludges stabilised by composting and anaerobic digestion, due to their lower contamination potential. However, both types of waste contain high proportions of water that could be reduced by drying, with

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the advantage that dried sludge has a lower transport cost and is easier to store and distribute. These processes can alter both the proportions of macronutrients and of micronutrients in the sludge, which can also be affected by the location in which they are produced (industrial or agricultural), the volume of the sewage plant, and the time of the year in which the sludge is produced.

This study aims to evaluate, from an agricultural point of view, the quality of three types of sewage-plant sludge produced in 37 EDARs with different plant volumes, in different regions of Spain.

2. Materials and methods

The experiment was carried out over two years, from 2006 to 2007. Composted, anaerobically digested, and pelleted sewage sludge was taken from 37 plants throughout Spain (Fig. 1). Samples were taken in fresh and sent to the University of Santiago de Compostela laboratories each month.

Once in the laboratory, samples were dried and chemical analyses were performed. Samples were passed through a 2-mm sieve and ground with an agate mortar. Nitrogen and phosphorous levels were determined after a microKjeldahl technique by colourimetry using TRAACS 800⁺, following Castro et al. (1990). Total calcium, potassium, magnesium, sodium, and heavy metal concentrations were analysed with a VARIAN 220FS spectrophotometer using atomic absorption (VARIAN, 1989), after a nitric acid digestion performed in a CEM MDS-2000 microwave (CEM, 1994).

2.1. Statistical analysis

The results were analysed using univariate (ANOVA) and multivariate methods (linear discriminant analysis and cluster analysis). ANOVA and linear discriminant analysis were used to characterise the three different types of sludge, and cluster analysis was used to analyse the plants.

First, ANOVA was performed on a two-factor nested design (hierarchical design), where the levels or categories of the nested factor (sewage plants) were different within each level of the main factor (sludge). The Tukey test ($p < 0.05$) was used for subsequent pair-wise comparisons for the sludge.

This analysis makes use of individual variables and allows us to determine whether there is a linear combination of variables that emphasises the differences among the three types of sludge. When this approach is used, it turns out that it may be possible to determine two linear combinations for separating sludge, referred to as canonical discriminant functions (CDF1 and CDF2). They are chosen in such a way that CDF1 reflects sludge differences as much as possible and CDF2 captures as much as possible of the group differences not displayed by CDF1. If only two functions are needed to account for almost all of the important sludge differences, then a simple graphical representation of the relationship between the three types of sludge can be attained by plotting the values of these functions for the samples.

Thus, to complete the comparison of the three types of sludge, the discriminative power of each variable was tested by discriminant analysis and then the combination of dry matter, N, P, and K was analysed to establish the discrimination between the three types of sewage sludge. For the discriminant analysis, two-thirds of the data was randomly assigned to the building dataset (approximately 60% of the cases) with the remainder being allocated to the validation dataset.

For the multivariate analysis, to reduce missing values in the database when a chemical component was not detected due to the methodology used in the laboratory, the minimum value detected for that component in the other samples was multiplied by 0.5.

In the analysis of the levels of the nested factor (sewage plants), cluster analysis was carried out. Cluster analysis aims to partition a set of objects into groups or clusters in such a way that the profiles of the objects in the same cluster are very similar, whereas the pro-

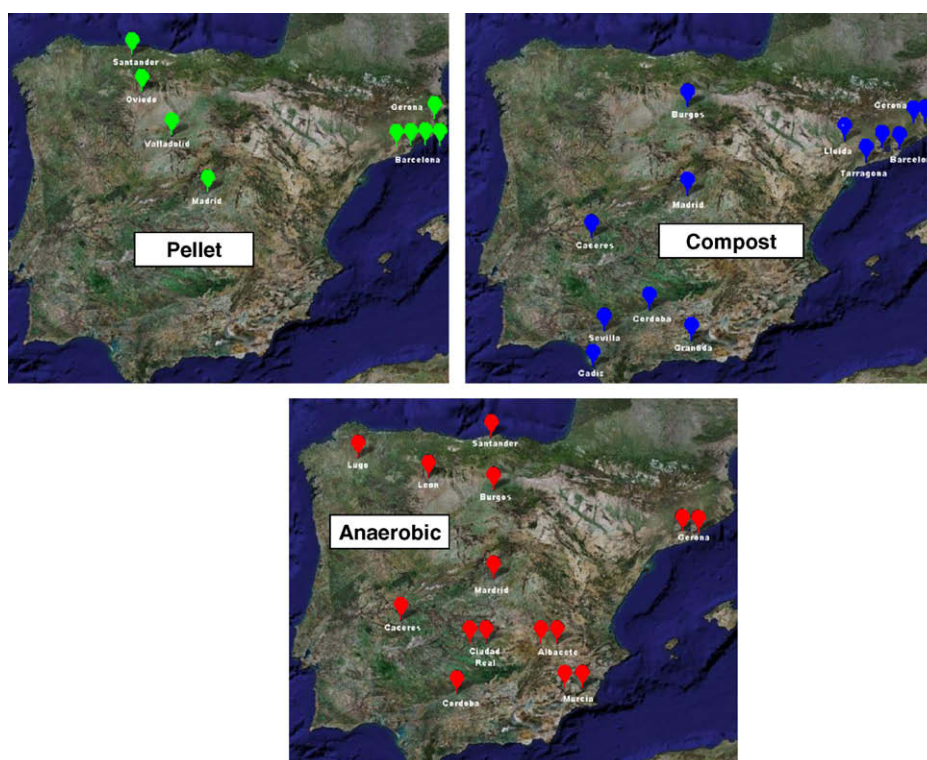


Fig. 1. Geographical distribution of the different plants producing the sludges analysed.

files of objects in different clusters are quite distinct. The most common approach to cluster analysis is the hierarchical method. The method proceeds sequentially, yielding a nested arrangement of objects in groups. Thus, once an object has been assigned to a group, it is never removed from the group later on in the clustering process. The hierarchical method produces a complete sequence of cluster solutions beginning with n clusters (one for each object) and ending with one cluster containing all n objects. The process can be conveniently represented using a tree diagram, called a dendrogram. The partitioning cluster method begins with a given number of clusters as the objective, and then, it partitions the objects to obtain the required clusters. In contrast to the hierarchical method, partitioning techniques permit objects to change group membership throughout the cluster formation process. The partitioning method usually begins with an initial solution, after which reallocation occurs according to some optimality criterion.

In the analysis of the levels of the nested factor (sewage plants), the following procedures were performed in three phases.

In the first phase, a hierarchical cluster analysis was applied. Starting from the standardised variables, we employed hierarchical cluster analysis to determine the optimum number of groups of sewage plants to be included in each type of sludge and to generate some initial centroids of the group. The centroid of a group is the middle point of a multidimensional space determined by the variables that are considered in the analysis.

In the second phase, taking as the starting point the number of previously determined groups, a non-hierarchical analysis of k -averages (partitioning method) was performed to discover the final configuration of the groups of sewage plants within each type of sludge. This procedure considers that the distance between groups is the distance between their centroids and, by starting from these centres of gravity, is less sensible to atypical values. Finally, the groups of sewage plants within each type of sludge were compared using a Student's t -test.

For the multivariate analysis, to reduce missing values in the database when a chemical component was not detected due to

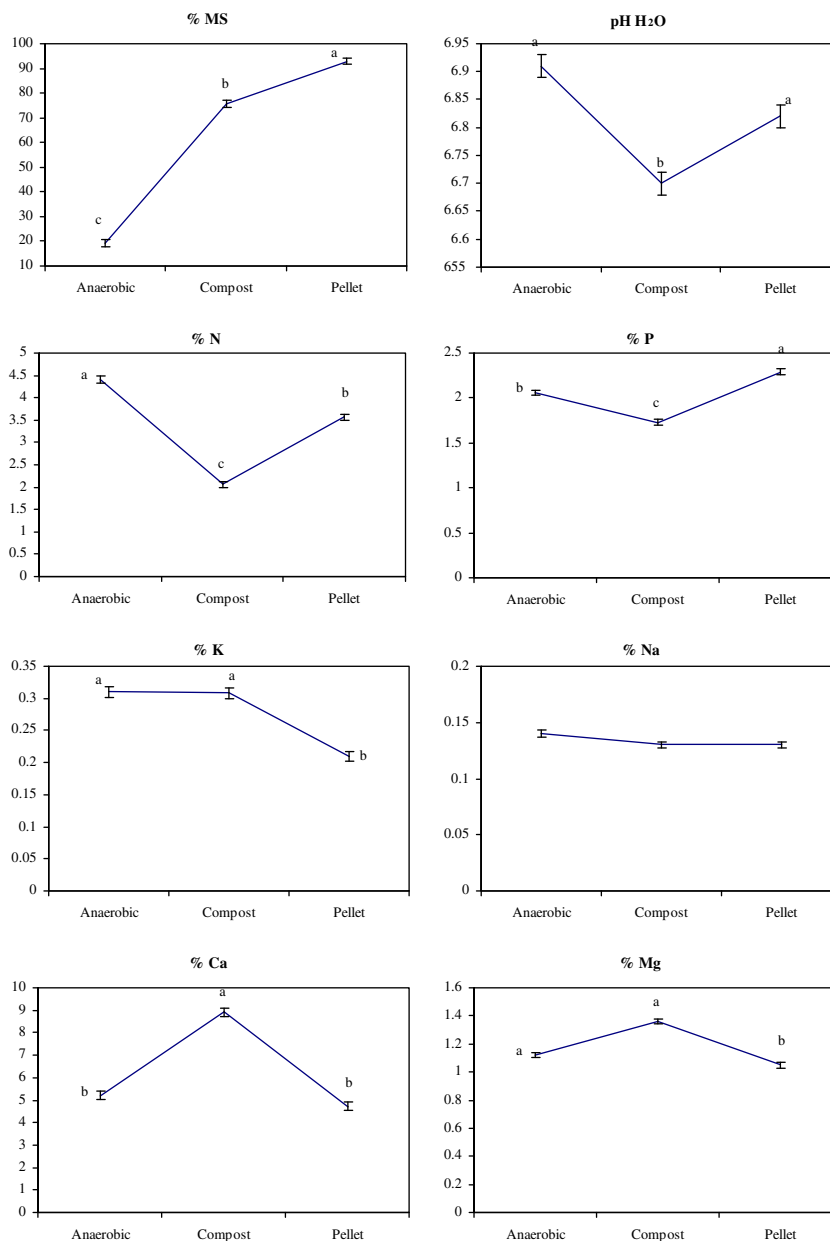


Fig. 2. Dry matter (MS), pH (H₂O), and macroelements of the different types of sludge studied. The bars indicate the typical average error.

the detection limit of the methodology used in the laboratory, the minimum value detected for that component in the other samples was multiplied by 0.5.

The statistical software package SAS (SAS, 2001) and SPSS version 15.0 were used for all analyses.

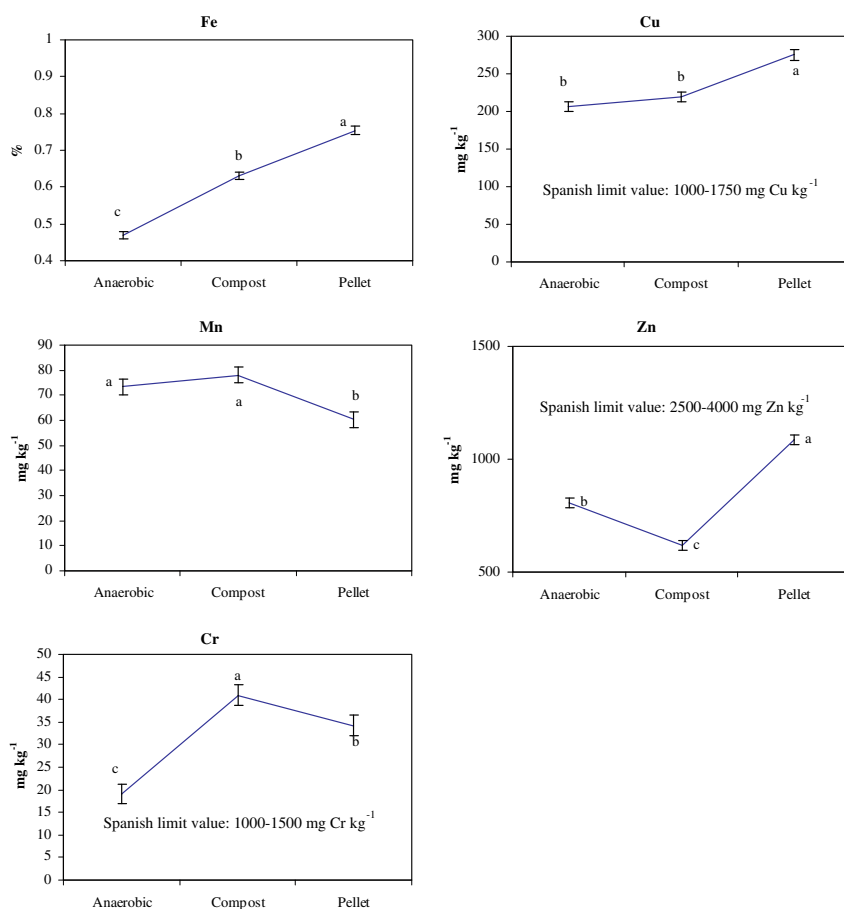


Fig. 3. Contents of Fe, Cu, Mn, Zn, and Cr of the different types of sludge studied. The bars indicate the typical average error. Note that the scale of the ordinate axis varies as a function of the element presented.

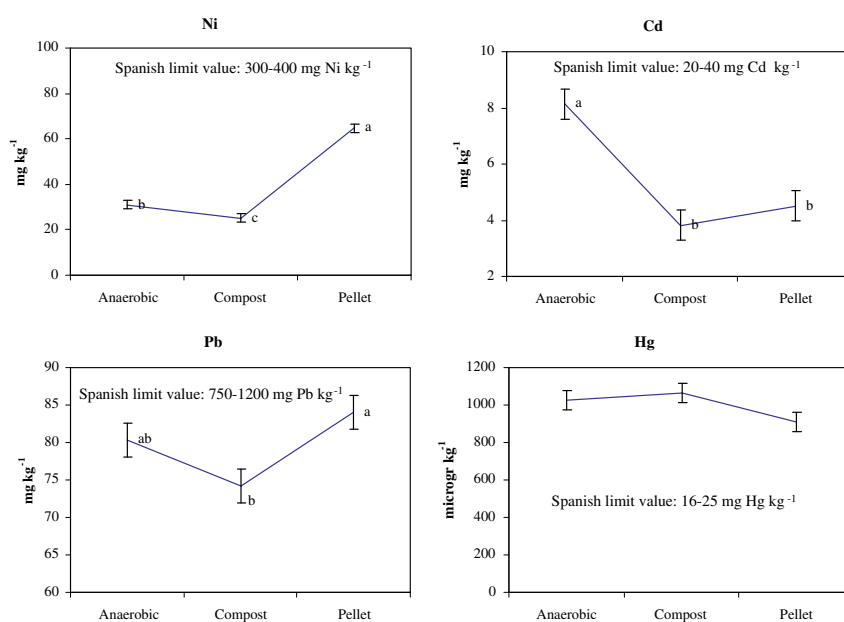


Fig. 4. Contents of Ni, Cd, Pb, and Hg of the different types of sludge studied. The bars indicate the typical average error. Note that the scale of the ordinate axis varies as a function of the element presented.

3. Results

3.1. Concentration of macroelements in the sludge

3.1.1. Macroelements

The principal results with relation to the content of dry matter, pH, and macroelements in the different types of sludge are shown in Fig. 2. The ANOVAs of all of the variables were significant ($p < 0.05$), with the exception of Na, which in turn has the lowest proportion present in the sludge, as frequently occurs in the materials of origin.

As could be expected, the dry-matter content is higher in the sludges that were heat-dried and pelletised when compared to

the other two types: composted sludge and anaerobically digested sludge. With relation to the pH, we observed that this is higher in the case of anaerobic sludge, as at this level of pH (close to 7.0), anaerobic microorganisms that digest the material in the absence of oxygen develop better.

With regard to nitrogen, we observed that it is significantly higher in the anaerobically digested sludge as compared to pelletised sludge, which was higher than the composted sludge. However, the levels of phosphorous are higher in the pelletised sludge, most likely due to a concentrating effect from being dried. The lower levels of phosphorous in the composted sludge can be explained by the fact that the vegetal component with which the sludge is mixed (remains of pallets, cuttings from pruning trees, etc.) contains a

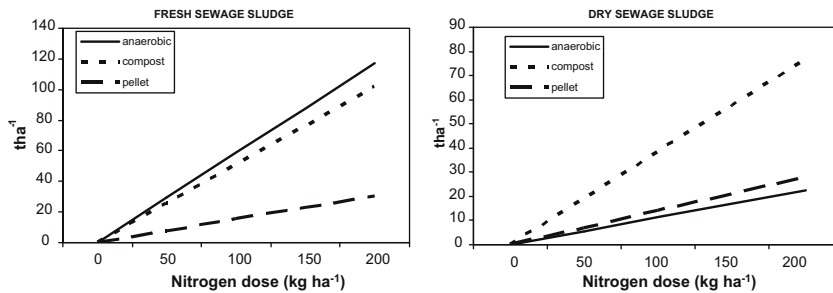


Fig. 5. Doses of sludge to be used ($t\ ha^{-1}$) for the different types of sludge as a function of the nitrogen dose ($kg\ ha^{-1}$).

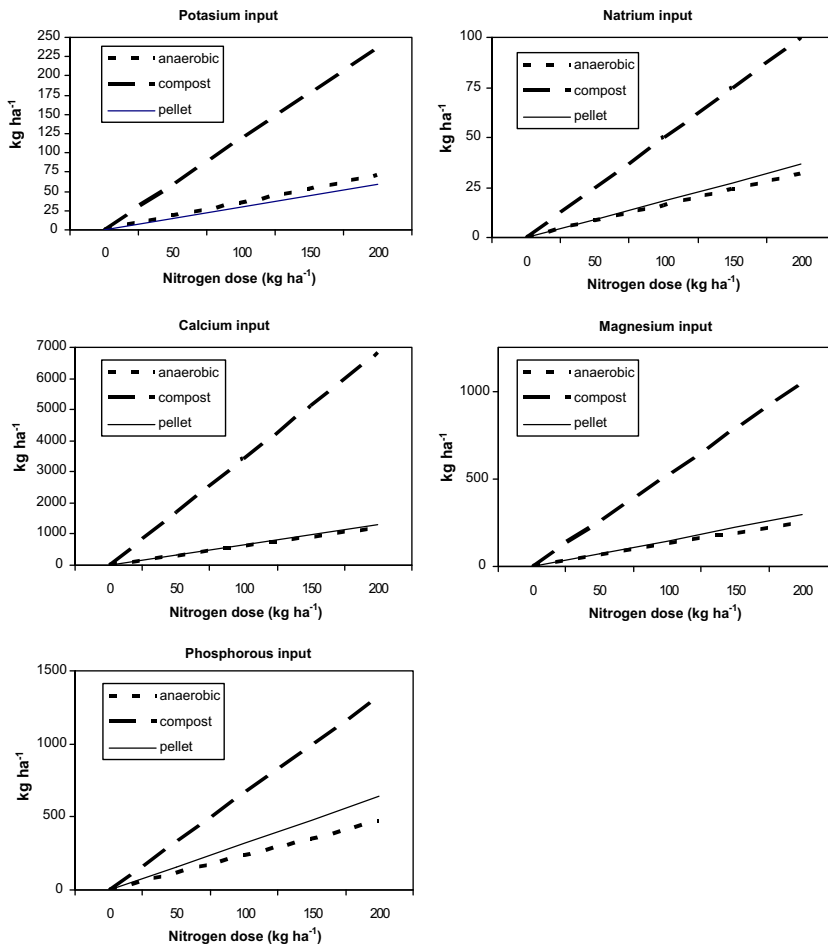


Fig. 6. Contribution of phosphorous, potassium, sodium, calcium, and manganese ($kg\ ha^{-1}$) as a function of the sludge dose and of the nitrogen dose. Note that the scale of the graph of calcium is different from that of the other elements.

low level of phosphorous, which also explains the higher levels of calcium, potassium, and magnesium in the composted sludges. Therefore, from the results obtained, we can see that the element

found in the highest proportion is calcium, followed by magnesium. In this case, the composted sludge has a much higher neutralising potential than the other two types of sludge.

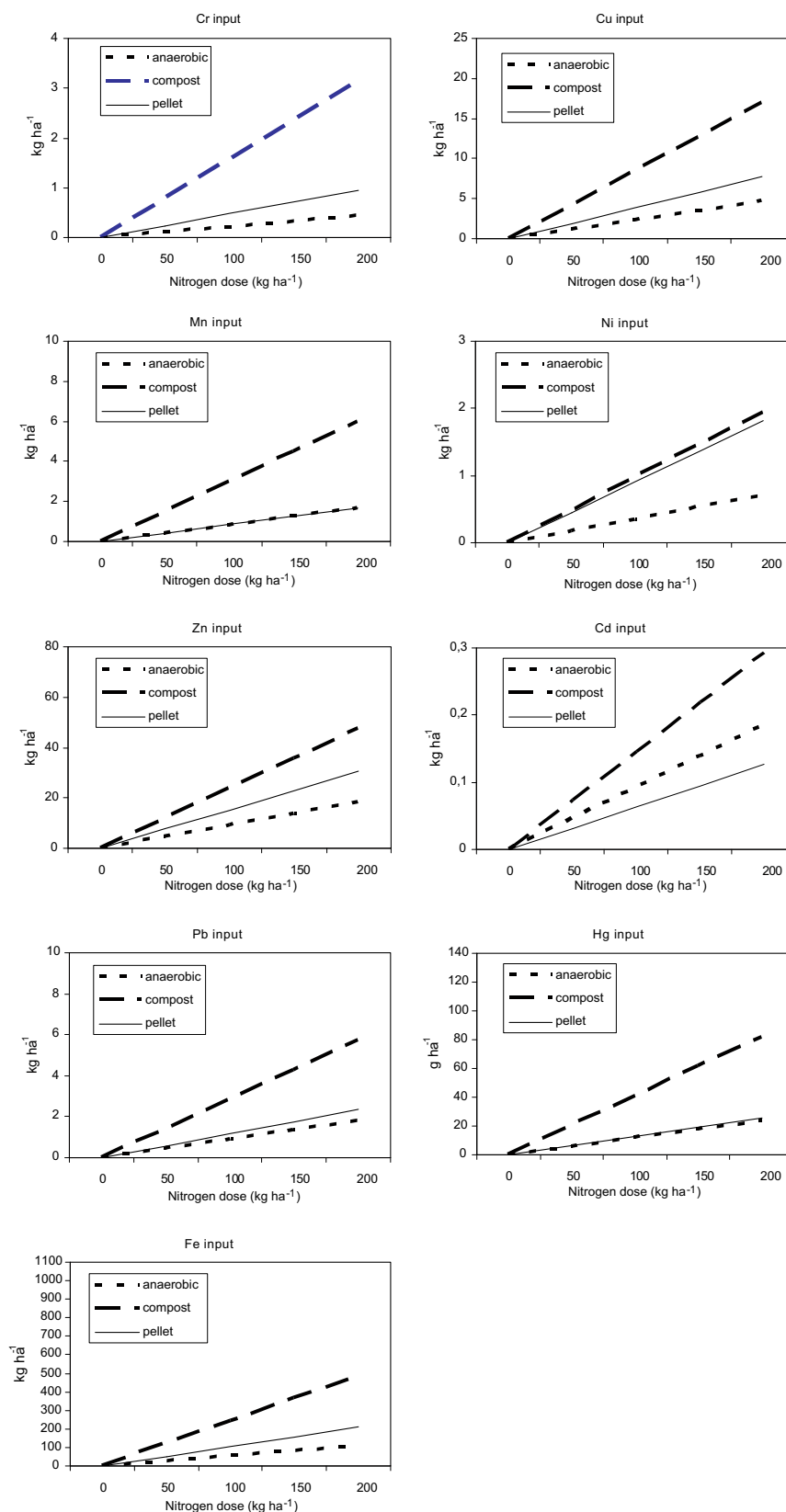


Fig. 7. Contents of Cr, Cu, Mn, Ni, Zn, Cd, Pb, and Hg of the different types of sludge studied. Note that the scale of the ordinate axis varies for the different elements.

3.1.2. Microelements and heavy metals

The levels of the principal microelements (Cu, Fe, Zn, Cr, and Mn) found in the different types of waste can be seen in Fig. 3, whilst those of Ni, Cd, Pb, and Hg can be seen in Fig. 4.

In general, it was found that the levels of the microelements are higher in the pelletised sludge, with the exception of Mn and Cr. This can be explained by the fact that some of the elements can be lost during the drying process, concentrating the remainder. In all cases, the concentration of these elements in the anaerobically digested sludge was similar to that found in the composted sludge, with the exception of manganese.

Concentrating on the trends of the heavy metals (Ni, Cd, Pb, and Hg) in the three types of waste (Fig. 4), it is observed that both Ni and Pb follow the same tendency found in the microelements: a higher concentration in the pelletised sludge. However, the level of Cd appears to be higher in the anaerobic sludge, which could indicate that losses can occur during the process of drying and pelletisation. Finally, no differences are found between the different types of waste with regard to Hg.

3.2. Agronomical implications of the concentrations of elements in the sludge

In the Southwest Atlantic biogeographic region of Europe, the common necessities and recommendations of nitrogen for cultivating pastures (the principal agricultural crop of the region) are between 80 and 120 kg N ha⁻¹, and production does not respond to more than 200 kg of available N per hectare. Therefore, using the necessities of the crops as a reference and taking into account the nitrogen content of the sludge (Fig. 2) and its availability (10% in the case of compost and 20% in the case of pelletised or anaerobic sludge), it is possible to estimate the doses that should be applied. It can be clearly observed (Fig. 5) that, in terms of dry matter, the pelletised and anaerobic sludges are the most beneficial when added to the soil, as they have a higher rate of nitrogen mineralisation and a higher nitrogen content. However, when we observe what occurs in terms of fresh sludge, which is what is usually employed, we can see that pelletised sludge is the most beneficial application. On the other hand, the required doses of fresh composted sludge are very high, as the concentration of nitrogen is much lower in comparison to the other two types of waste.

The main nutrient inputs for each type of sewage sludge (Fig. 6) demonstrate that, using compost, the amount applied is higher and similar to the amounts applied with anaerobic and pelletised sludge, with the exception of phosphorous, which has a higher fertilisation rate with pelletised sewage sludge than with anaerobic sewage sludge. The same response is obtained for the micronutri-

ents and heavy metals (Fig. 7). In this case, it is the composted sludge that exhibits an increased input when compared with pelletised and anaerobic digested sewage sludge. Anaerobic sludge always has a lower input of heavy metals and micronutrients into the soil. Only Ni, Zn, and Fe inputs are notably higher in pelletised sludge, as compared to anaerobic sludge, which is due to their higher concentration in the former.

3.3. Combining dry-matter N, P, and K for discriminating between sludges

Linear discriminant analysis was carried out considering each variable separately; the procedure shows that the percentage of dry matter is the variable that achieves the best discrimination between types of sludge. If a discriminant analysis is applied considering only this variable in the model, the discriminatory function manages to correctly classify 84.5% of the samples in the validation group. The errors in this classification are caused by confusion between samples of composted and pelletised sludge, whilst the samples of anaerobic sludge are all correctly classified (Table 1).

The inclusion of dry-matter N, P, and K as variables in the discriminatory analysis enables the first two discriminating functions (CDF1 and CDF2) resulting from the analysis to correctly classify 94.1% of the samples of the validation group (Table 1), by reducing the confusion between samples of composted and pelletised sludge.

A graphical representation of the scores assigned to the samples for the two discriminatory functions is shown in Fig. 8. Both of the functions have high canonical correlations (0.95 and 0.62), which indicates that both are useful in the differentiation between groups, particularly the first function. Moreover, the significance of the Wilks' lambda statistic ($p < 0.001$) reflects that, using both discriminatory functions, it is possible to establish significant differences between the means of the groups.

Standardised canonical coefficients (Table 2) indicated that CDF1 discriminated sludge based on dry matter. This is the expected discrimination, due to the higher level of dry matter in the pellet, followed by that of the composted sludge. In contrast, the anaerobic sludge has a lower level of dry matter. CDF2 discriminated sludge based on N, P, and K, assigning scores contrary to those of composted and pelletised sludge. Of the two, the pelletised sludge provides the highest values of N and P, together with the lowest values of K, which causes a positive mean scores in this discriminatory function.

In the second discriminatory function, the anaerobic sludge shows as many positive punctuations as negative, which indicates

Table 1
Discriminant accuracy based on the two canonical discriminant functions for 305 samples in the model-building dataset and for 220 samples in the validation dataset. (a) Considering only % of dry matter as the predicting variable.

Actual group	Number of samples in each predicted group (model-building dataset)				Number of samples in each predicted group (validation dataset)			
	Anaerobic	Compost	Pellet	Total	Anaerobic	Compost	Pellet	Total
<i>(a)</i>								
Anaerobic	108(100%)	0	0	108	87(100%)	0	0	87
Compost	0	89(71.2%)	36(28.8%)	125	0	60(72.3%)	23(27.7%)	83
Pellet	0	11(15.3%)	61(84.7%)	72	0	8(16%)	42(84%)	50
<i>(b)</i>								
Anaerobic	108(100%)	0	0	108	87(100%)	0	0	87
Compost	0	111(88.8%)	14(11.2%)	125	0	75(90.4%)	8(9.6%)	83
Pellet	0	5(6.9%)	67(93.1%)	72	0	5(10%)	45(90%)	50

(a) Percentage of samples correctly classified: 84.6% (model-building dataset), 85.9% (validation dataset). As the M of Box test was significant; the co-variance matrices of the separate groups were used to classify them.

(b) Percentage of samples correctly classified: 93.8% (model-building dataset), 94.1% (validation dataset). As the M of Box test was significant; the co-variance matrices of the separate groups were used to classify them.

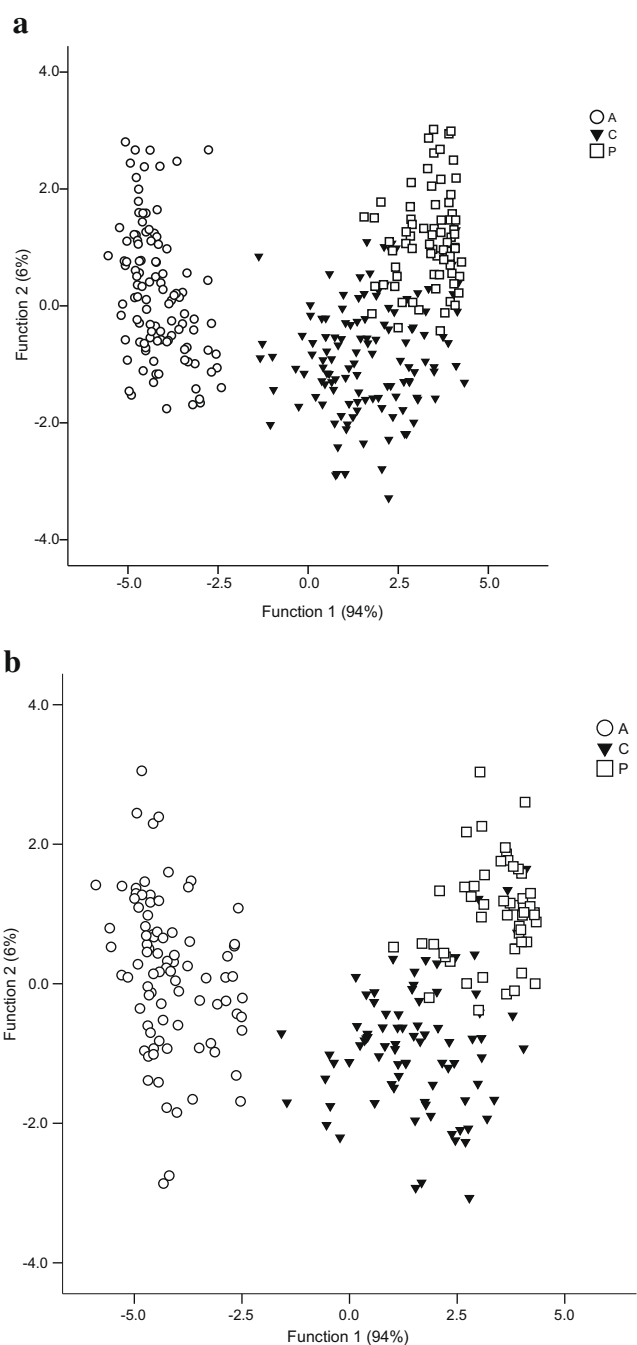


Fig. 8. Separation of the sludge by the two canonical discriminant functions (CDF1 and CDF2) for 305 cases in the model-building dataset (a) and for 220 cases in the validation dataset (b). A: anaerobic; C: compost; P: pellet.

that the combination of N, P, and K has a more variable pattern in anaerobic sludge than in the other two types of sludge.

An analysis of variance of the canonical scores CDF1 and CDF2 indicates that the sludge has significantly different punctuations (Table 2).

3.4. Intra-sludge variation: analysis of the sewage plants

Using the average determinations of macroelements, microelements, and heavy metals in each sewage plant, a cluster analysis was carried out. The dendrograms shown in Fig. 9 illustrate how the agglomerations of the sewage plants take place.

Table 2

(a) The standardised canonical coefficients (SCC) and the correlation coefficients (r) between the canonical discriminant functions (CDF1 and CDF2) and the variables (b). Means of canonical scores of two canonical discriminant functions for the sludges studied. Values labelled with different letters in the same column are significantly different according to the pair-wise Tukey test at the $p < 0.05$ level.

	Function			
	CDF1		CDF2	
	SCC	r	SCC	r
<i>(a)</i>				
Dry matter	.994	.983	.136	.008
N	-.079	-.189	.833	.764
P	.032	-.002	.521	.485
K	-.158	-.052	-.475	-.231
	Mean	SE	Mean	SE
<i>(b)</i>				
Anaerobic	-4.17 ^a	.76	.19 ^b	1.11
Compost	1.63 ^b	1.29	-.87 ^a	.95
Pellet	3.37 ^c	.74	1.09 ^c	.80

3.4.1. Anaerobic sludge

It is possible, within the anaerobic sewage plants, to consider two groups of plants. Beginning with the centroids of these groups and using a non-hierarchical cluster analysis of k -averages, two definitive groups of anaerobic sewage plants are formed. This grouping differs from that established by the analysis of hierarchical conglomerates in the changes in groups No. 8 and No. 1 (Table 3).

A comparison between the two groups using Student's t -test (Table 3) indicates that there are significant differences in all of the elements, except for Na, between the anaerobic sludge sewage plants. Included in the group that agglutinates the highest number of sewage plants are those whose sludges, on average, have the highest contribution of the macroelements N, P, and K and the least contamination. Most of the smaller sewage plants are found in this group. In the sewage plants that generate more contaminating waste, the Zn and Hg levels are doubled, whilst the Cr, Cd, and Mn levels are tripled, and moreover, the standard error indicates that the averages of these sewage plants are more variable in all of the characteristics analysed.

3.4.2. Composted sludge

According to the dendrogram shown in Fig. 9b, two or three groups can be formed from the twelve sewage plants that process composted sludge. Starting from only two groups, the non-hierarchical cluster analysis of k -averages, with respect to the hierarchical grouping, placed sewage plants 1 and 17 in different groups, thus forming two groups with six sewage plants in each one (Table 5).

By comparing the groups of sewage plants, with regard to the average determinations of the different variables (Table 4), it was confirmed that significant differences are found in fewer variables than for the anaerobic sludge. The large difference in Cr is notable between one group of sewage plants and the other, differing by a factor of eight in the group with the most contaminating waste. This is a consequence of the presence, within the group of composted sludge sewage plants, of the two plants that show the highest values of this element (plants 10 and 11). Within the group of the most contaminating sewage plants, Ni and Zn also have higher values, increasing by almost a factor of two. In this case, there is not such a direct relationship as in the anaerobic sewage plants, probably due to the exogenous contribution of material from a different source for composting.

3.4.3. Sludge pellets

Similar to the case of composted sludge, the initial grouping of the sewage plants that process pelletised sludge can be done in

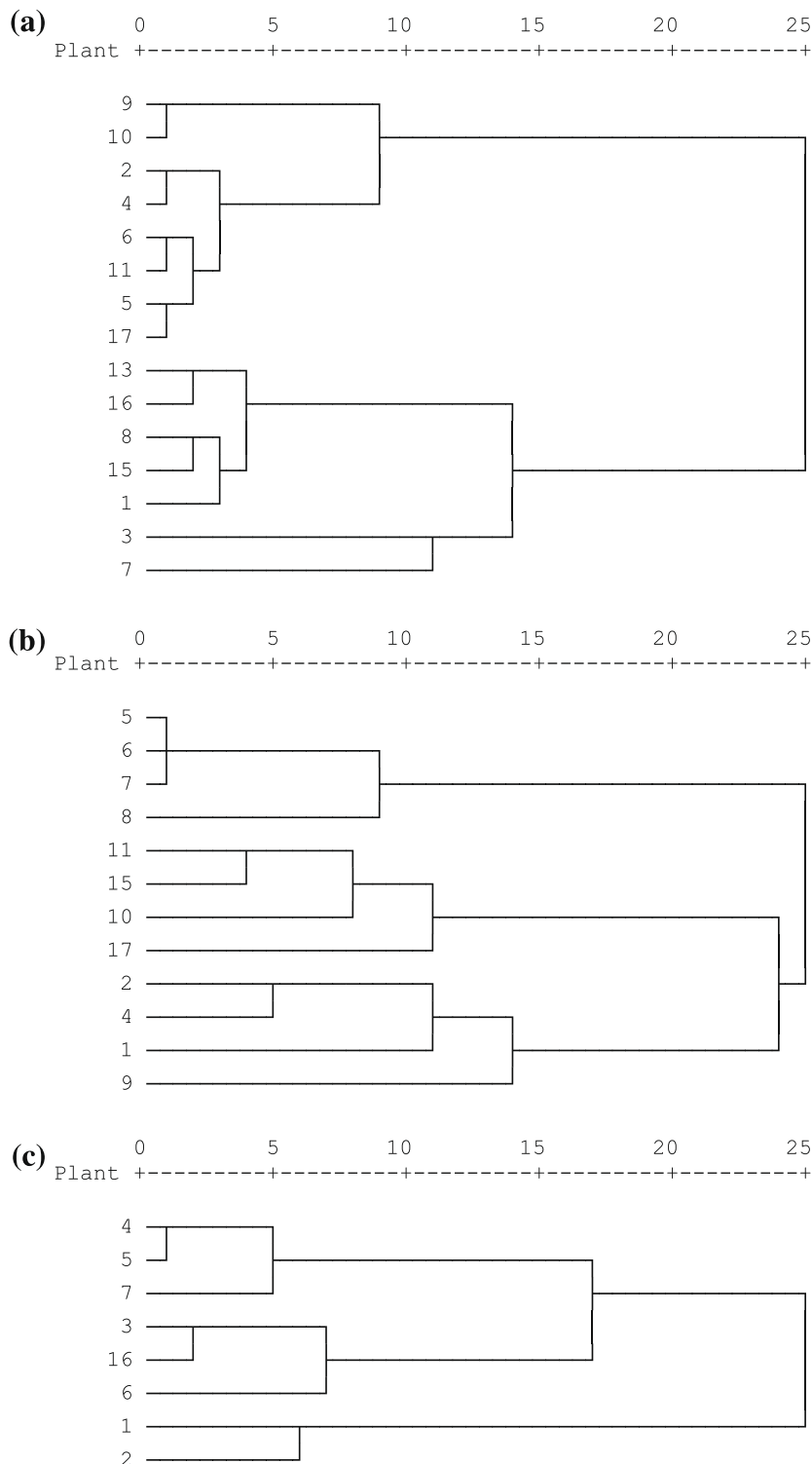


Fig. 9. Dendrograms using the Ward Method as the method of conglomeration and, as the measurement of distance between groups, the squared Euclid distance. Groups of sewage plants that process anaerobic sludge (a). Groups of sewage plants that process composted sludge (b). Groups of sewage plants that process pelletised sludge (c).

two or three groups (Fig. 9c). Considering only two groups, the sewage plants No. 1 and No. 2 are the ones that are separate from the rest. After applying the non-hierarchical cluster analysis of *k*-averages, sewage plant No. 7 joins them. These three sewage plants are separate from the rest because they are the least contaminant (Table 5), with lower values of microelements and heavy metals. They are also the ones that contribute the most nitrogen among the pelletised sludge sewage plants, as also occurs with the anaerobic

sludge. In this case as well, there is no direct relationship between the sewage plant volume and the quality of the sludge.

4. Discussion

The fertilisation potential of the different types of evaluated sludges depends on the stabilising process in such a way that, in general, the composted sludge shows a higher liming potential

Table 3

Sewage plants that process anaerobic sludge. Their groups and distances to the final conglomerating centre. Plant volume of the sewage plants and the determinations with significant differences between groups according to the Student *t*-test.

Conglomerate	1					2									
	3	7	13	15	16	1	2	4	5	6	8	9	10	11	17
Plant	3	7	13	15	16	1	2	4	5	6	8	9	10	11	17
Distance	904.3	540.8	160.4	260.6	304.7	216.8	349.0	138.5	189.3	102.3	353.4	347.0	239.6	289.9	334.7
Plant volume	195,000	1500	28,000	53,500	30,000	285,000	215,000	17,400	7500	7500	2000	12,000	1200	6060	33,000
	<i>n</i>		Mean		ES	<i>n</i>					Mean			ES	
% Dry matter ^{***}	52		24.53		.69	143					17.42			.46	
pH(H ₂ O) ^{***}	52		7.31		.06	143					6.76			.04	
%N ^{***}	52		3.18		.16	143					4.85			.13	
%P ^{***}	52		1.82		.08	143					2.15			.06	
%K [*]	52		.26		.02	143					.33			.02	
%Na	52		.17		.02	143					.14			.01	
%Ca ^{***}	52		7.91		.49	143					4.23			.29	
%Mg ^{***}	52		1.33		.05	143					1.04			.03	
%Fe ^{***}	52		.73		.05	143					.37			.02	
ppm Cr ^{***}	49		37.17		5.12	120					11.76			.96	
ppm Cu ^{***}	52		297.16		34.48	143					172.99			6.92	
ppm Mn ^{**}	52		149.99		24.70	143					45.48			2.36	
ppm Ni ^{***}	49		44.35		3.85	99					24.36			1.70	
ppm Zn ^{***}	52		1373.68		77.41	143					599.37			27.21	
ppm Cd ^{***}	35		15.80		4.42	70					4.30			.48	
ppm Pb ^{***}	52		108.51		12.27	130					69.07			3.93	
ppb Hg ^{***}	17		1551.06		114.45	58					870.40			84.84	

* *p* < 0.05.
 ** *p* < 0.01.
 *** *p* < 0.001.

Table 4

Sewage plants that process composted sludge. Their groups and distances to the final conglomerating centre. Plant volume of the sewage plants and the determinations with significant differences between groups according to the Student *t*-test.

Conglomerate	1					2						
	2	4	9	10	11	15	1	5	6	7	8	17
Sewage plant	2	4	9	10	11	15	1	5	6	7	8	17
Distance	94.4	313.4	231.0	173.6	106.2	141.9	207.2	53.3	49.7	87.6	87.9	147.5
Plant volume	47,500	6060	35,000	135,000	55,000	53,500	285,000	79,400	52,500	24,900	51,250	33,000
	<i>n</i>		Mean		ES	<i>n</i>		Mean		ES		
% Dry matter ^{***}	99		70.99		1.12	109		80.14		1.30		
pH(H ₂ O)	99		6.73		.04	109		6.68		.04		
%N ^{***}	99		2.29		.07	109		1.85		.09		
%P	99		1.76		.09	109		1.71		.06		
%K ^{**}	99		.28		.02	109		.34		.02		
%Na	99		.13		.01	109		.13		.01		
%Ca	99		8.46		.51	109		9.31		.53		
%Mg ^{***}	99		1.21		.04	109		1.50		.06		
%Fe	99		.64		.03	109		.62		.03		
ppm Cr ^{***}	91		71.96		8.77	87		8.53		.47		
ppm Cu ^{***}	99		286.57		18.45	109		158.64		7.10		
ppm Mn	99		76.44		4.08	109		79.48		5.04		
ppm Ni ^{***}	83		30.12		1.87	62		18.67		1.58		
ppm Zn ^{***}	99		812.33		30.62	109		441.54		17.72		
ppm Cd	53		3.89		.37	70		3.76		.28		
ppm Pb	94		73.56		3.93	103		74.71		4.59		
ppb Hg	20		1282.00		153.93	29		915.21		110.94		

** *p* < 0.01.
 *** *p* < 0.001.

(as a supplier of calcium and magnesium cations), but also a lower fertilisation potential (lower concentrations of nitrogen and phosphorous), except for potassium (Walter et al., 2006). This can be justified by the fact that, in many cases, the compost is produced using waste (Deportes et al., 1995) that is characterised by high levels of Ca- and Mg-type cations, with low levels of nitrogen and phosphorous. The pelletised and anaerobic sludges do not combine with these wastes, which increases their fertilisation value and reduces their liming capacity. However, it is notable that the pH of these two sludges is close to neutrality, with an important neutralising potential, particularly in soils with a strong tendency towards acidity. Nevertheless, and given the neutral

character of the sludges, their use can reduce the alkalinity of basic soils, due to their lower pH, and can favour the extraction of calcium and magnesium by the crops, thus generating an increase in production (Mantovi et al., 2005).

On the other hand, the pelletised sludge shows a higher concentration of heavy metals, when compared to anaerobic and composted sludge, which could be due to the heat-drying process itself, which may cause the vapourisation of some nutrients and not others (Anderson et al., 2002). However, Fuentes et al. (2008) obtained similar relative proportions of Ni, Zn, and Cd among sludges subjected to heat-drying and anaerobic digestion studied in the region of Murcia.

Table 5
Sewage plants that process sludge pellets. The groups to which they belong and their distances to the conglomerating centre. Plant volume of the sewage plants and determinations with significant differences between the two groups according to the Student *t*-test.

Conglomerate	1			2			n	Mean	n	Mean
	1	2	7	3	4	5				
Sewage plant	1	2	7	3	4	5			6	16
Distance	196.3	196.3	137.6	438.6	117.2	242.2			224.7	287.1
Plant volume	30,000	215,000	525,000	300,000	410,000	215,000			57,000	50,000
	n	Mean	ES							ES
% Dry matter	35	94.35	1.17	87				92.33		.80
pH(H ₂ O) ^{***}	35	6.23	.09	87				7.07		.05
%N ^{***}	35	4.58	.18	87				3.17		.09
%P	35	2.19	.08	87				2.34		.05
%K [*]	35	.25	.02	87				.20		.01
%Na ^{***}	35	.09	.01	87				.14		.01
%Ca ^{***}	35	2.73	.22	87				5.52		.25
%Mg ^{***}	35	.61	.03	87				1.23		.04
%Fe ^{**}	35	.65	.06	87				.84		.03
ppm Cr ^{***}	34	19.66	2.36	86				39.99		3.76
ppm Cu ^{***}	35	158.83	9.10	87				321.88		15.99
ppm Mn ^{***}	35	37.15	1.96	87				69.76		2.48
ppm Ni ^{***}	26	26.22	3.02	81				77.05		5.93
ppm Zn ^{***}	32	751.02	44.36	87				1210.09		49.05
ppm Cd [*]	28	3.59	.46	65				4.91		.38
ppm Pb ^{**}	35	67.78	6.45	81				91.01		4.68
ppb Hg	13	860.85	156.22	18				947.39		92.41

* $p < 0.05$.

** $p < 0.01$.

*** $p < 0.001$.

It should be noted that in no case did the average levels of concentration of the different metals in the sludge exceed the Spanish, European, or American guidelines (Düring and Gäth, 2002), all of them (except for cadmium) were below the levels indicated in the third draft of the working document on sludge (EC, 2000). If we compare, in the case of composted sludge, the results obtained in a series of bibliographic reviews, we will see that, in most of the cases in this study, the composted sludges are of good quality, probably as a consequence of the recently implemented policies (Deportes et al., 1995; Hargreaves et al., 2008). In fact, they would be within the range of stabilised compost indicated by the EC (2000), but would not belong to Classes 1 and 2 of better quality. In the case of cadmium, none of the evaluated waste complied with the draft guidelines for sludge applications in agriculture in 2025 (2 mg Cd kg⁻¹), and only the composted or pelletised sludge complied with this draft guideline for 2015 (2 mg Cd kg⁻¹). It should be noted that Cd is more mobile than Hg and Pb (McLaughlin et al., 1999), apart from being more available in anaerobic sludge (Fuentes et al., 2008), which may cause an increase in the in-plant concentration, and thus, Cd may reach humans through the trophic chain (Düring and Gäth, 2002). For this reason, the components of these elements should be strongly decreased in the sewage water to be treated, by controlling accidental spillage, to reduce their level in the resulting sludge.

The other metal levels are generally less than half of those marked by the draft guidelines for 2025, with the exception of Cr, which is far from this value, or Zn, which is very close to the guideline level in some types of sludge.

It is important to point out that the correcting, fertilising, and contaminating capacity of the sludges studied also depends on the concentration of the different elements they possess and on the amount each one contributes to the soil, as well as the type of soil on which they are spread.

If we suppose that the soil complies with the current or future guideline for the use of sludge in agriculture (EC, 2000) and that the sludge has a value of heavy metals under the limit established,

it is possible to model the amount to be contributed of each waste, as indicated by the EPA (1994) of the USA, based on nitrogen levels. This would allow the estimation of the quantity interval of the residue to be applied as a function of the nitrogen levels of the different types of sludge and the contribution made by the rest of the elements and heavy metals. In fact, this calculation leads to the conclusion that the composted sludge, despite having reduced levels of heavy metals, has more environmental- and contamination-type problems. The use of composted sewage-plant sludge in agriculture is promoted by the European Union, as well as anaerobic sludge, although the latter has a lower risk of contamination (EC, 2000). This makes necessary either a greater decrease in the level of contaminants or its employment in conditions that require a smaller contribution per surface unit of composted sludge. In this way, among others, its use could be pooled as a substitute for peat in the production of potted trees, as the dose per surface unit is notably reduced (Ostos et al., 2008).

The Spanish guidelines have also established the yearly amount of heavy metals that can be added to the soil through sludge, based on a ten-year mean. If the annual consecutive contribution of different heavy metals over one year is studied, using nitrogen as the criterion, it is observed that all of the sludges comply with the current guidelines in the case of Cr (3 kg ha⁻¹ year⁻¹), Pb (15 kg ha⁻¹ year⁻¹), Ni (3 kg ha⁻¹ year⁻¹), and Hg (100 g ha⁻¹ year⁻¹) and that Cd (0 st 0.33 lb ha⁻¹ year⁻¹), Cu (12 kg ha⁻¹ year⁻¹), and Zn (30 kg ha⁻¹ year⁻¹) exceed the limits in the composted sludge at the highest doses (implying 200 kg N ha⁻¹ of sludge), these being the most restricted heavy metals in sludge in the present legislation, despite the less restrictive levels indicated for Zn and Cu as compared to other metals.

It should be noted that cadmium impedes the contribution of the three types of sludge (0.15 kg ha⁻¹ year⁻¹), which, unless low doses of pelletised and anaerobic sludge are used, limits the number of years sludge can be applied over a period of 10 years. Finally, if these values are taken into account and compared with those established in the draft guidelines for the use of sludge for 2025 (EC, 2000), it can be seen that all of the metals, except for chrome,

exceed the indicated levels. This signifies that, if this guideline is approved, sewage sludge cannot be spread annually unless a serious effort is made to reduce the levels of these heavy metals in the different types of sludge. However, if the average amount of heavy metals that can be contributed in 10 years is multiplied by ten, it can be seen that the addition of the dose of composted sludge that entails the fertilisation of 200 kg ha⁻¹ of readily assimilated nitrogen will never be achieved, as the future guidelines for Zn, Pb, and Hg would be exceeded. However, above all, this could not be done because the criterion for cadmium would be exceeded if doses of more than 50 kg of composted or anaerobic sludge, or doses of more than 100 kg of pelletised sludge, were used. Therefore, it is the level of cadmium that will most limit the use of these types of waste in the future.

The lower proportion of dry matter in pelletised sludge is extremely important from a practical point of view, as there is a direct relationship between the distance between the sewage plant and the point of application and the cost of transport, which is reduced if the dry matter content is high. In fact, and as might be expected, this is the criterion that most differentiates the three types of waste, although nitrogen, phosphorous, and potassium best distinguish the pelletised sludge (with greater concentrations of principal macronutrients) from the composted sludge.

The differences between the types of sludge can be explained by the stabilisation processes, as the heat-drying of the pelletised sludge could cause the losses of determined elements, while concentrating others. The mixing of fibrous vegetal material, which generally has a lower nitrogen content, is probably the cause of the lower proportion of macronutrients found in the composted sludge. In relation to the differences found between the sewage plants within each type of sludge, it should be noted that the level of contaminants is higher in the sewage plants with high volumes of throughput (i.e., large populations), which is a result of the greater contamination per individual produced. However, the separation between the composted-type and the pelletised-type sewage plants is not associated with this parameter as, in the first case, it depends on the type of material added to the sludge in the composting process, which distorts the relation between population and sludge quality, and in the second case, it is associated with much larger towns, which limits differentiation, and depends on the drying process itself, favouring the loss of determined nutrients.

If we take into account the current guidelines, the quality of sewage-plant sludge is adequate, which allows for its use whilst complying with these guidelines. However, if the draft guidelines proposed by the European Union are approved, the use of sludge in agriculture will be severely restricted. It will not be possible to use sludge as a fertiliser in the future, unless the concentrations of Cd, Pb, Zn, and Hg are reduced. The composted sludge shows

an excellent quality as compared to the other two types of sludge, but its use as a fertiliser is very limited, due to its low nitrogen content and the low level of availability of this element. This means that high doses must be used, increasing its contaminating potential.

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