



RESEARCH ARTICLE

POLLUTION HAZARDS OF HEAVY METALS IN SEWAGE SLUDGE ASH FROM THE WASTEWATER TREATMENT PLANTS IN CAMBERENE, SENEGAL

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Abstract

Solid wastes and their by-products are gaining interest worldwide given their high environmental impact. Fly ash and Bottom ash from Camberene sludge waste center (Senegal) were characterized to assess the heavy metal contamination (using XRF and the Toxicological Risk) that is very important in type 1 unlike in type 2. The Index of geo-accumulation (I_{geo}), the Pollution load index (PLI), the Enrichment factor (EF) and Contamination factor (CF) have been computed to evaluate the contamination rate. These show that the fly ash has aI_{geo} value of 3.57 for Pb and 3.04 for As which means they are very polluted. For Cu we have an I_{geo} value of 4.23 and for Zn it is 4.67 so these ashes are strongly to extremely polluted by Cu and Zn but unpolluted to moderately polluted by Cr. For the bottom ashes we have I_{geo} values of 3.03 for Cu and 3.02 for Zn, to say they are also strongly polluted. However, they are not polluted by Cr and are only moderately polluted by Pb and As; results confirmed by the EF calculation. Fine and dirty ashes have significantly been enriched by the metal As with an EF of 13.71 while for Pb its EF is 19.10 for the fine ash. As for the bottom ash we have respectively 7.26 and 5.19 for the EF of As and Pb. From the values of PLI these ashes are very highly polluted. Their possible dangerousness depends essentially on their heavy metal contents (criterion H14 of Directive 91 / 689 / EEC). In this Directive the material is toxic if its content in heavy metal is higher to some thresholds (see Annex III, Table 6). Most of their heavy metal contents are below the threshold in Annex III, indicating their harmlessness. This analysis highlighted the principal characteristics to be taken into account before using the SSA properly.

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Introduction:-

Sewage water also called residual water is loaded with pollutants spread or dissolved in the water and served as needed by industrial or agricultural activities [1, 2].

These waters are relatively complex because consisting of organic material, mineral and microorganisms.

Various processes among which pretreatments, primary, secondary, tertiary treatments have been developed in the wastewater treatment plant to minimize the impact of sewage water [3-6].

The treatment of sewage water produces water of acceptable quality but also an important quantity of sludge containing different organic and mineral compound. These compounds contain pathogenic microorganisms, parasites, toxic elements including heavy metals. Produced sludges have thus bad smells because of their ease of putrefaction and occupy a large volume per day [7-14].

The amount of sludge produced differs according to the continents. The production of dry sludge in the United States is 7 millions of tons (Mt) per year whereas in the European Union it was estimated at 10 Mt in 2012 and could reach 13 Mt in 2020 [15-18].

In 2012, Senegal had nine functional wastewater treatment plants. In 2014, the number of " **wastewater treatment plants**" WWTP reached 12 [19]. The most important one is that of Camberene which was built in the framework of the sanitation project of Dakar and its vicinities. The station which has been operating since January 1989, was built by the company DEGRMONT / CSE. It was enlarged in 2007 and had a capacity of 200,000 equivalents/inhabitants with a daily out of rate 19,200 m³/d and a load in biochemical demand in oxygen (DBO5) of 21,696 kg/d. Another extension of the station is in progress. That will allow this station to reach a capacity of nearly 20,000 m³/d to 90,000 m³/d [20].

These sewage sludges are managed between incineration, agricultural and landfill [21, 16]. It is found that the incineration processes reduce the volume of the waste by 90% and its mass by 70%. The incineration process is the most appropriate management option to deal with the volumes produced and the potentially unsafe elements the sewage sludge contains [22].

In 2012, 2.3 Mt of sludges were incinerated, mainly in Germany, France in the Netherlands generating 0.7 Mt of sewage sludge ash. This quantity will probably increase in the future due to a decrease from 50 to 55 % in the disposal option in the agricultural field and the rise of the price of the dumping [23].

The incineration also generates toxic substances like As, Hg, Pb, Zn and the Cd, and an important quantity of ashes (30 to 40% of the dry mass) that must be managed. Hence, some pre-treatment procedures should be adopted to reduce/stabilize the heavy metals contained in sewage sludge ash before their use.

Thoroughly understanding the properties of heavy metals present in sewage sludge ash is important for the control of heavy-metal pollution during their use in civil engineering.

So in this study two types of sewage sludge ash (Fly ash and Bottom ash) from the sludge waste center of Camberene (Senegal) were characterized to assess the heavy metal contamination. XRF, Calculation of the pollution index and the Toxicological Risk were used to characterize samples. The Index of geo-accumulation (I_{geo}), the Pollution load index (PLI), the Enrichment factor (EF) and Contamination factor (CF) have been computed to evaluate the contamination rate of ashes by heavy metals.

Experimental

XRF Chemical Characterization

Samples were weighed and then introduced in oven at 105°C for 24h to remove moisture prior to the preparation of pellets made by mixing them with 10 wt% of a binder called Licowax [24, 25].

The obtained mixture was homogenized in a mortar and the technology press VANEOR FLUXANA considering a force of 10 N on a surface of a disk of a radius of 11 mm was used to form the pellets. After obtaining the pellets,

we used an X-ray portable fluorescence Niton XLT900s (P-XRF) for our X-ray analyzes with a measurement time of 350 s.

XRF was performed with 100% normalization and full fundamental parameter quantification techniques: see Table 1 for specification and operating conditions.

Table 1:- Spectrometer specification and operating conditions.

Resolution	178eV at Mn Ka
Window Thickness	12.7µm Be
Rating	50kV, 40µA maximum power of the tube 2W
Beam diameter	7mm
Filter	Element analysis
Ag excitation source	Sb, Sn, Cd, Pd, Ag, Mo, Nb, Zr, Sr, Rh, Bi, As, Se, Au, Pb, W, Zn, Cu, Re, Ta, Hf, Ni, Co, Fe, Mn, Cr, V, Ti, Th, and U
Sandwich of Al, Ti and Mo	Ba, Sb, Sn, Cd, Pd, Ag
Cu Filter	Cr, V, Ti, Ca, K
No Filter	Al, P, Si, Cl, S, Mg

Calculation of the pollution index

In this study four parameters to evaluate the contamination by metals have been used to check the contamination of fine and dirty sludge ashes from the WWTP of Camberene.

1. Index of geo-accumulation I_{geo}
2. Enrichment factor (EF)
3. Contamination factor (CF)
4. Pollution load index (PLI)

Using Equation (1) of G. Muller in the reference [26] the values of the index of geo-accumulation of the metals studied were calculated:

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5B_n} \right) \quad (1)$$

Where C_n is the measured concentration of an element 'n' into the soil sample, B_n is the geochemical concentration of the background of one element 'n' of the soil [27] and 1.5 is a factor of correction of possible variations due to lithogenic effects. Muller [28] gave, depending on the value of I_{geo} the ranking as below :

Table 2:- Classification of ashes based on the value of I_{geo} .

Class	I_{geo}	Soil Quality
0	$I_{geo} \leq 0$	Unpolluted
1	$0 < I_{geo} < 1$	Unpolluted to moderately polluted
2	$1 < I_{geo} < 2$	Moderately polluted
3	$2 < I_{geo} < 3$	Moderately to strongly polluted
4	$3 < I_{geo} < 4$	strongly polluted
5	$4 < I_{geo} < 5$	strongly to extremely polluted
6	$5 > I_{geo}$	Extremely polluted

We have adopted the same ranking for our ashes:

To establish an enrichment factor, (Simex and Helz, 1981) gave a formula [29].

$$EF = \frac{\left(\frac{C_x}{C_{ref}} \right)_{sample}}{\left(\frac{B_x}{B_{ref}} \right)_{Background}} \quad (2)$$

Were

C_x is the concentration of an x element of the soil sample from the study site, C_{ref} is the concentration of the element of reference, B_x is the concentration of the Background of an x element and B_{ref} is the concentration of the background of the element of reference. In this study, iron which is abundant in the soil was used as the metal of reference as it was distributed independently, compared to other metals.

Five categories of contamination have been recognized on the bases of the enrichment factor [30] :

Table 3:- Classification of ashes based on the value of EF.

$EF < 2$	Deficiency to minimal enrichment
$2 < EF < 5$	Moderate enrichment
$5 < EF < 20$	Significant enrichment
$20 < EF < 40$	Very high enrichment
$EF > 40$	Extremely high enrichment

The measure of the contamination factor is also realized the contamination factor which evaluates the anthropogenic effect of the pollution of the soils by metals. The evaluation is done by using the formula of [30] :

$$CF = \frac{C_x}{B_x} \quad (3)$$

Where C_x is the measured concentration of an element x in the sample and B_x is the concentration of the element x of the geochemical background.

A classification of contaminations according to the values of CF is established by Hakanson 1980 and Loska2004 [31, 32]:

Table 4:- Classification of ashes based on the value of CF.

$CF < 1$	Low contamination factor indicating low contamination
$1 \leq CF < 3$	Moderate contamination factor
$3 \leq CF < 6$	Considerable contamination factor
$6 \leq CF$	Very high contamination factor

The load index of pollution comprises the pollution coming from various heavy metals and reflect directly the level of pollution of all heavy metals on a sampling point [33].

The PLI was established by the equation (4) [34] :

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \quad (4)$$

Where CF is the factor of contamination and n the number of a studied element.

According to the load index of pollution, the pollution is divided into six levels [35]:

Table 5:- Classification of ashes based on the value of PLI.

First	= 0	Background concentration
Second	1~2	Unpolluted
Third	2~3	Middly polluted
Fourth	3~4	Moderately polluted
Fifth	4~5	Highly polluted
Sixth	> 5	Very highly polluted

Results And Discussion:-

X ray Fluorescence Analysis

A portable XRF device Niton XLT900s was used to analyze the chemical composition of the sludge ash in terms of major and minor elements. The minor elements are given in mg/kg. **Table 6**

Table 6:- Results of minor (mg/kg) elements using XRF technique of sewage sludge ash contents.

Elements	Fly ash (type1)	Bottom ash(type 2)
Pb	357	66
Cu	1264	553
Cr	212	67
Zn	3632	1156
As	16	6

Heavy metals are more important in fly ash than in bottom ash, this could be due to their volatility. For example, the concentration of Pb is about five times higher in fly ash than in bottom ash, while that of Zn is 3.5 times greater in fly ash than that of Zn in the bottom ash. Analogous observations are also relayed by Rajamma [36], in their work on the characterization and use of biomass fly ash in cement based-materials. From the point of view of availability, bottom ash is more important than fly ash.

In Table 7 we resume studies from other authors in order to give a comparative asset in term of elemental composition with the samples we have used.

Table 7:- Comparison of minor (mg/kg) element contents measured with other authors working with the same materials.

Element	Sewage sludge ash PW	(Martin Cyr, 2007)	(Nuria Husillos Rodríguez, 2013)	Siew Chin(2016)	Choo
Pb	357	720	40	30	
Cu	1264	2483	320	130	
Cr	212	2636	1270	20	
Zn	3632	7103	1020	810	
As	16	23	5.8	-	

The high variability between values shows that sludge ashes depend on several parameters: sludge composition, treatment conditions such as incineration temperature and additives

Heavy metal contamination

To assess the heavy metal contamination on the ashes, some indices have been calculated: Index of geo-accumulation (I_{geo}) and Pollution load index (PLI). The Enrichment factor (EF) and Contamination factor (CF) have been computed to evaluate the contamination rate of ashes by heavy metals. The obtained results are given in Table 8:

Table 8:- Index of geo-accumulation (I_{geo}), Pollution load index (PLI), Enrichment factor (EF) and Contamination factor (CF) of the ashes.

Heavy metals	I_{geo}		EF		CF		PLI	
	FA	BA	FA	BA	FA	BA	FA	BA
Pb	3.57	1.14	19.10	5.19	17.85	3.30	14.09	4.42
Cu	4.23	3.03	30.06	19.33	28.09	12.29		
Cr	0.65	-1.01	2.52	1.17	2.36	0.74		
Zn	4.67	3.02	40.92	19.14	38.23	12.17		
As	3.04	1.62	13.71	7.26	12.31	4.62		

According to the value of I_{geo} a classification was given: from unpolluted to extremely polluted (Table 2).

The type 1 ashes which we have called by analogy fly ashes have a I_{geo} value of 3.57 for Pb and 3.04 for As which means that they are very polluted by these two metals. For Cu we have an I_{geo} value of 4.23 and for Zn it is 4.67 so these ashes are strongly to extremely polluted by Cu and Zn but unpolluted to moderately polluted by Cr.

The bottom ashes have I_{geo} values of 3.03 for Cu and 3.02 for Zn which means that they are also strongly polluted by these two metals. However, they are not polluted by Cr and they are only moderately polluted by Pb and As.

These results were confirmed by the calculation of the factor of enrichment EF. Fine and dirty ashes have significantly been enriched by the metal As whose EF was estimated to be 13.71 while for Pb its EF was 19.10 for the fine ash. As per the bottom ash we have respectively 7.26 and 5.19 for the factors of enrichment of As and Pb.

The arsenic is a toxic and carcinogenic element. Increased risks of lungs and bladder cancer, as well as skin modification were noticed to exposed persons from arsenic concentrations [37].

The value EF from Cu and Zn in the fine ashes shows that these latter are full of these two elements. The value 2.52 of EF for Cr in the fine ashes shows that these ones are moderately enriched for this metal, whereas the BA have a factor of enrichment of 1.17 for Cr. That may classify the BA among the ashes where the enrichment by the Cr is minimal.

The Cr is a dangerous element. It may cause health problems in humans. In fact the respiratory tracts and causes gastric problems and stomach ulcers[38, 39].

The factor of contamination CF for the whole measured heavy metals for the two ashes except the Cr in the BA was superior to one which can imply the contamination of two ashes.

In addition the values of PLI of two ashes being superior to the unit, that could indicate a pollution for the whole combined heavy metals.

From the values of PLI this ashes are very high polluted.

Globally the fly ash is polluted. This could be due to the volatility of some metals with respect to others.

If we limit ourselves to these indexes, SSA ashes are polluted. However, these indexes are often used for sediments. To be able to really conclude on the pollution of these ashes by the metals, it is necessary to consult the standards. To limit the risks, a leaching operation is required.

Toxicological Risk

The possible dangerousness of the ash from STEP sludge depends essentially on their heavy metal content with regard to the limits of regulated hazardous substances on one hand, and on the other hand of the overall ecotoxicity of the ash (criterion H14 of Directive 91 / 689 / EEC).

In this Directive the material is toxic if its content in heavy metal is higher to some thresholds as defined in Annex III (Table 9).

Table 9:- Comparison of the concentration of minor elements in ashes with thresholds defined in Annex-III.

Element	Concentration (mg/kg) type 1 ash	Concentration (mg/kg) type 2 ash	Threshold concentration (mg/kg)
As	16	6	220
Cr	212	67	150
Cu	1264	553	340
Pb	357	66	840

In some cases, the heavy metal contents of these ashes are below the threshold concentration in Table 9, so the ashes cannot therefore be a priori considered dangerous on the basis of these criteria. The classification of ashes can vary according to the criteria used, depending on the geographic location as well. However some researchers differ on the definition of toxicity of ashes. Some will put them in landfill if there are considered dangerous while others who consider them non dangerous will value them.

Thus a study of the dangerousness of the ash is necessary in order to see their possible compatibility with the recovery in construction.

Conclusion:-

The understanding of the properties of heavy metals present in sewage sludge ash was investigated. Both ashes have been characterized.

Heavy metals are more important in fly ash than in bottom ash, this could be due to their volatility.

The concentration of Pb is about five times higher in fly ash than in bottom ash, while that of Zn is 3.5 times greater in fly ash than that of Zn in ash in the bottom ash

The high variability between values measured with other authors working with the same materials shows that sludge ashes depend on several parameters: sludge composition, treatment conditions such as incineration temperature and additives

The fly ash has an I_{geo} value of 3.57 for Pb and 3.04 for As which means that they are very polluted by these two metals. For Cu we have an I_{geo} value of 4.23 and for Zn it is 4.67 so these ashes are strongly to extremely polluted by Cu and Zn but unpolluted to moderately polluted by Cr.

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From the values of PLI this ashes are very high polluted.

Most of the heavy metal contents of these ashes are below the threshold in Annex III of (criterion H14 of Directive 91 / 689 / EEC) indicating their non-dangerousness. As perspective, other complementary techniques such as the leachable contents of heavy metals by toxicity characteristic leaching procedure (TCLP), the risk assessment code (RAC) to estimate the environment risk of heavy metals by applying a scale to the percentage of metals presented in the acid soluble/exchangeable fraction.

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