Novel co-valorisation of sewage and marine dredged sludges: preliminary bench scale results

Anna Laura Eusebi*, Matteo Spinelli*, Paolo Battistoni*, Antonio Dell'Anno**, Laura Rocchetti** and Francesca Beolchini**

* Dipartimento SIMAU, Facoltà di Ingegneria, Università Politecnica delle Marche, Via Brecce Bianche, 12, 60100 Ancona, Italy. (E-mail: a.l.eusebi@univpm.it)
** Dipartimento SVA, Facoltà di Scienze, Università Politecnica delle Marche, Via Brecce Bianche, 12, 60100 Ancona, Italy.

Abstract
The co-treatment of urban and the dredged sludges was investigated to produce new fertilizer. Different operative parameters were tested both for washing and ozonation process as previous treatments of dredged sludge. The minimum contact time of 12 hours using effluent from urban wastewater plant was identified to reduce the marine salinity of the dredged sediments. The obtained concentrations of main macro pollutants from the washing unit could permit the discharge of the supernatants liquid flows in the main wastewater plant without overload the traditional urban influent. The following ozonation unit determines the increment of the degradability of the washed materials measured with the SOUR data (up to 4.2 g/kgMLVSS/h). The optimal dose was evaluated equal to 200 gO3/kgTSS also according to the release of the organic substances after the ozonation. The final characterization of the stabilized material was realized comparable with the values for nutrients and organic matter reported in the fertilizer legislation.

Keywords
Sewage sludge, dredged sludge, washing, ozone, fertilizer

INTRODUCTION

Millions of cubic meters of sediments are annually dredged especially in coastal areas (Netzband et al., 2009). These dredged materials become waste and usually for technical and/or economic reasons were dumped into the sea or send to the landfill. Therefore also the non-contaminated or low-contaminated dredged sediments will be finally discharged. This approach determines useless utilization of limited space capacity, high cost and low environmental sustainability and compatibility (Rulkens, 2005). The dredged sediments valorisation is disadvantaged both by the actual mandatory regulation and by the organic and inorganic pollution manly due to the marine salinity and to the compounds from industrial and anthropogenic origin (Fonti et al., 2013). In this framework, development of new economically and environmentally sustainable alternatives for dredged material disposal is desirable. One novel possibility is the co-treatment of marine dredged sediments with the sewage sludge from urban wastewater treatment plants with the main objective of agricultural fertilizer production (Macia et al., 2014). In this sense the integrated treatment of these different wastes (bio-wastes and dredged marine sediments) implements the efficient use of resources and the circular economy (Puchongkawarin et al., 2015). In fact, notwithstanding the dredged sludge is characterized from low level of nutrients contents (N%TS, P%TS, COD%TS), elevated levels of mineral oligoelements (B, Mn, Zn,Cu, Mo, Co, Fe) and salinity are present in this type of matrix (Purves, 2012). To produce suitable material for use in agriculture, the low concentrations of organic substances of the dredged sediments could be provided adding sewage sludge and/or organic fraction of municipal solid waste. In fact, huge amounts (average 30 kg dry matter/inhabitant/year) of sewage sludge are generated all over the world from wastewater treatment plants (WWTPs) increasing the economic and environmental impact (Hospido et al., 2010). The biosolids from WWTPs constitute a complex mixture that, in addition to nitrogen, phosphorus and organic matter, often contains inorganic (metals and trace elements) and organic pollutants (Jones-Lepp et al., 2007). For all these reasons, the optimal processes configuration for the cotreatment has to be finalized to stabilize the amount of matrices used (dredged and sewages sludge and OFMSW), to reduce the main pollutants concentrations and to obtain the final characteristics according to the final destination of fertilization. This paper presents the preliminary results of chemical and physical pre-treatments of dredged sediments to identify the new flow scheme for the final production of fertilizing material from dredged and urban sludges. The defined process configuration was tested to determine the macro characteristics of the final product.

MATERIAL AND METHODS

The proposed flow scheme is composed from washing reactor, ozonation unit, centrifuge, mixing reactor with urban sludge and final composting phase. All the liquid supernatants were supposed to be discharge in the main wastewater treatment plant according with the final obtained concentrations. The washing and ozonation effects were preliminary studied. Two types of dredged sludges (T1 from clay sediments and T2 from sandy sediments) were analytically characterized in terms of main macropollutants concentrations. The sludges were washed using both deionized water and effluent from urban wastewater plant (ratio 1:10 w/w) to define the minimum contact time and the characteristics of the liquid supernatants (10 samples for 48 hours of maximum washing time). The washed matrices with deionized water were ozonized in bench scale at different contact times (0.5, 5, 30 min) changing the oxygen flow from 20 to 40 lO2/h to define the optimal ozone dose. The liquid supernatants were characterized in terms of main macropollutants concentrations and specific oxygen uptake rate (SOUR). In fact, tests of SOUR (at 20°C gO2/kgMLVSS/h) were carried out at same F/M (kgCOD/kgMLVSS) to evaluate the ozone effect as increment of the rapidly biodegradable compounds of the supernatants from washed dredged sludges (T1 and T2). The data were compared with the SOUR values from sodium acetate and urban wastewater influent. The clay sludge T1 was used to realize the complete process configuration at the found optimal operative parameters to determine the macro characteristics of the final product.

RESULTS AND DISCUSSION

The analytical results of the two types of dredged sludges. of urban sludge and of organic fraction from municipal solid wastes were reported in Table 1. The dredged sludges showed basic pH (9.18 for T1 and 7.98 for T2), elevated concentration of TS% higher for the T2 sandy type (66.5%TS for T1 and 84.6 %TS for T2). Moreover, low concentrations of organic compounds (4.70 and 1.40 COD%TS respectively for T1 and T2) and of nutrients (0.20 and 0.14 of N%TS and 0.79 and 0.19 of P%TS respectively for T1 and T2) were determined. Percentages around 0.05 %TS and 0.25 %TS were found respectively as K and Mg in the examined dredged sludges. Finally, the studied dredged sludges presented low levels of metal contamination (Fonti et al., 2013). Differently, the increment of organic matter and of nutrients could be possible during the stabilization phase adding urban sludge and/or OFMSW according to the fertilizer production objective. The chemical and physical characteristics of these two matrices are reported (Table 1).

Table 1. Dredged and urban sludges characterization

		DREDGED SLUDGE		URBAN SLUDGE	OFMSW
		CLAY T1	SANDY T2		
pH		9.18	7.98	7.26	5.93
TS%	%	66.5	84.6	0.79	6.43
TVS/TS	%	10.00	0.98	64.75	55.08
COD%TS	%	4.70	1.40	79.41	98.6
N%TS	%	0.20	0.14	4.78	2.48
P%TS	%	0.79	0.19	0.90	2.57
K%TS	%	0.09	0.03	0.47	n.a.
Mg%TS	%	0.25	0.27	0.34	n.a.

Elevated COD concentrations at lower TS% were determined (0.79 TS% and 79.41 COD%TS for Urban Sludge and 6.43TS% and 98.6 COD%TS for OFMSW). Net increment of structural and organic Nitrogen and Phosphorous is shown up to 4.78 N%TS for urban sludge and 2.57 P%TS for OFMSW.
The first processing problem of the dredged sludges is related with the elevated salinity. Therefore one step of washing phase was scheduled. According with the possible co-treatment with urban sewage the washing unit was tested using both deionized water and effluent from real wastewater treatment plant (Effluent characteristics: pH 7.85, COD 27.2 mg/l, TSS 7 mg/l, NH4-N 5.16 mg/l, NO3-N 7.44 mg/l, PO4-P 2.39 mg/l). In fact, in the potential upscaling of the flow scheme the effluent flow could be greatly sufficient as washing source. Different washing times were studied. The characterization of the liquid supernatants at different contact times were executed for all the main macropollutants. The CODs and Chloride concentrations were reported in Figure 1 after the washing phase with deionized water up to 48 hours. The stationary conditions for the CODs was obtained after 12 hours both for T1 and T2. Final maximum concentrations of 73.4 mgCODs/l (T1) and 40 mgCODs/l (T2) were evaluated in the liquid washing flows. Differently, immediate elevated values was carried out for the Chloride reaching 1440 and 422 mgCl-/l respectively for T1 and T2. Lower salinity was released from sandy sediments for the more elevated dry matter percentage and the different granulometric characteristics. For the washing phase similar results were obtained using the effluent from urban wastewater plant. Therefore, the minimum washing interval of 12 hours was identified as sufficient time for the achievement of the ions equilibrium.

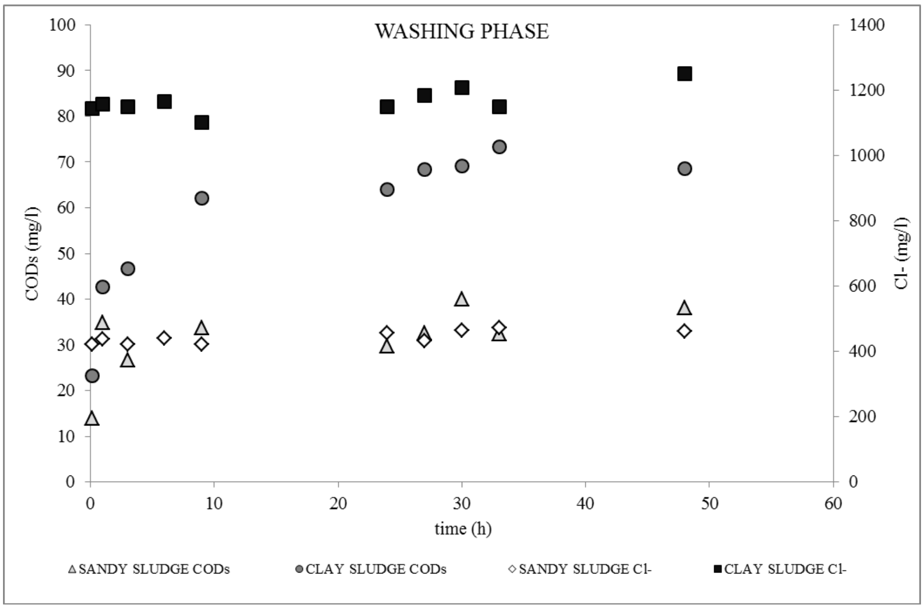


Figure 1. Washing phase of dredged sludges: CODs and Cl concentrations

The characterization of liquid flows at different contact times was realized also to determine some metals concentrations (Zn, Ni, Cu). The data are reported in Figure 2 a) and b) respectively for clay (T1) and sandy (T2) dredged sludge using effluent flow as washing source. Low levels of metals in the final liquid supernatants after the washing unit were found in part related to the contribute of the used effluent flow (for T1: average 0.322±0.186 mgZn/l and 0.177±0.107 mgCu/l; for T2: average 0.442±0.039 mgZn/l and 0.152±0.051 mgCu/l). The amounts of Nickel were almost negligible both for T1 and T2. No net effect of release during the contact times was evaluated for all the metals analysed except for Zinc after 30 h (Figure 2 b). According with the literature (Sabra et al., 2011) simple washing phase did not cause possible release/decontamination from heavy metals of treated sediments. Considering the detected low concentrations the final liquid flows remain conforming with the law discharge limits of metals in surface waters.

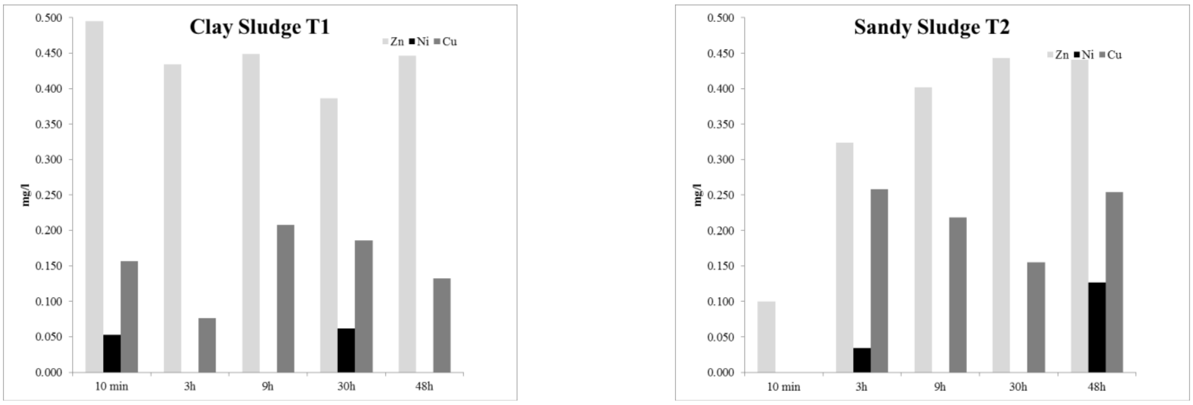


Figure 2 a) Metals in liquid washing flow T1 **b)** Metals in liquid washing flow T2

Following ozonation tests were realized to apply advanced oxidation process at these matrices improving the decontamination effect and the possible solubilisation of the organic compounds. The characterization of the main macropollutants in the ozonized supernatants was realized (Figure 3).
Constant pH values were detected (7.55±0.1 for T1 and 7.55±0.25 for T2). The ozonized and centrifuged liquid phases of the T1 and T2 (Figure 3) defined the increment of CODs up to 220.4 and 80.4 mg/l respectively for T1 and T2 at the maximum ozone dose (about 1000 gO3/kgTSS). Also the calcium concentrations increased after the ozone treatment (up to 133 and 88 mg/l for T1 and T2 at the maximum ozone dose) probably related to the carbonate transformation. No effects for the chloride and sulphate or for other anions (K, Mg, Na, NH4) were found.

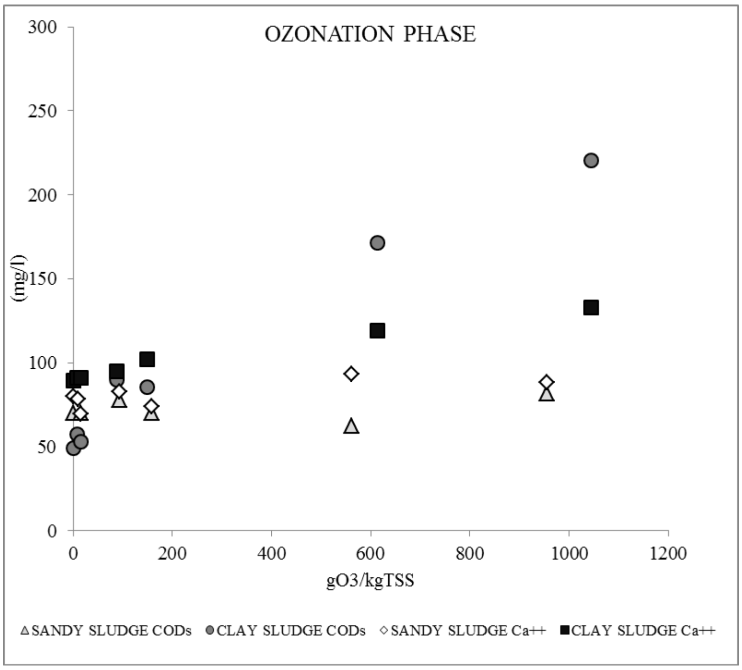
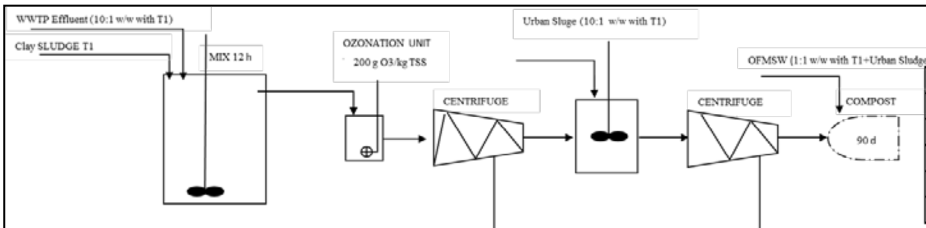


Figure 3. Ozonation phase of dredged sludges: CODs and Calcium concentrations

The ozonized flows was centrifuged and filtered to determine the SOUR values. The clay (T1) and sandy (T2) dredged sludges after washing was characterized from 2.5 and 0.4 gO2/kgMLVSS/h. The values raised up to 4.20 (T1) and 0.72 (T2) gO2/kgMLVSS/h at dose of 200 gO3/kgTSS remaining similar at maximum ozone dose. The SOUR for ozonized T1 was higher than the SOUR from urban influent (2.81 gO2/kgMLVSS/h) and half of the SOUR from the rapidly biodegradable source (7.20 gO2/kgMLVSS/h). Therefore, the ozone amount of 200 gO3/kgTSS was retained the sustainable and optimal dose for the pretreatment of the dredged sludge.

Considering the collected operative data related to the minimum contact time (12 h) and the optimal ozonation dose (200 gO3/kgTSS), the complete flow scheme was realized also to verify the main characteristics of the final compost product. The clay sludge (T1) was washed using effluent from urban wastewater plant (ratio 1:10 w/w Dredged Sludge: Effluent), was ozonized and was coupled with urban sludge source and with OFMSW (10:1 w/w Urban Sludge: Dredged Sludge, 1:1 w/w OFMSW: Urban + Dredged Sludges) (Figure 4). Composting process was realized at ambient temperature (20°C) with natural (daily turning of the compost heap) and mechanical venting.



FINAL MACRO CHARACTERISTICS		
COD%TS	%	59
TS%	%	94
TVS/TS	%	48
N%TS	%	2.34
P%TS	%	2.30

LIQUID SUPERNATANT AFTER WASHING		
pH		8.14
COD	mg/l	71.2
TKN	mg/l	7.13
N-NH4	mg/l	6.46
Na	mg/l	709
K	mg/l	59
Mg	mg/l	84
Ca	mg/l	95
Cl	mg/l	1130
NO2	mg/l	0
N-NO2	mg/l	0
N-NO3	mg/l	11
P-PO4	mg/l	0.0
SO4	mg/l	284
Ptot	mg/l	0.2

LIQUID SUPERNATANT AFTER OZONATION		
pH		7.24
COD	mg/l	134
TKN	mg/l	18
N-NH4	mg/l	6.7
Na	mg/l	727
K	mg/l	66
Mg	mg/l	93
Ca	mg/l	99
Cl	mg/l	1161
NO2	mg/l	0
N-NO2	mg/l	0
N-NO3	mg/l	11
P-PO4	mg/l	0
SO4	mg/l	315
Ptot	mg/l	0.52

Figure 4. Physical and chemical characterization of the main flows

All the liquid supernatants were characterized and the main concentrations of the final product reported in Figure 4. The values are completely conforming with the discharge limits in the wastewater plant. Negligible volumes of liquid leachate were produced during the stabilization process. After 90 days the final macro characteristics of the composted fertilizer was analysed. Dry matter of 91 TS% and mainly composed from COD (59 COD%TS and 48% TVS%TS) was found (Figure 4). The final nutrients amounts in terms of Nitrogen and Phosphorous were detected of about 2.3 as N% and P%TS. The obtained macropollutants contents meet the national and European requirements for the fertilizer production.

CONCLUSIONS

The characterization of the dredged sludges in terms of main macro pollutants was realized. This sediments could be valorised co-treating with urban excess sludge and organic fraction of municipal solid waste to produce fertilizer. The optimal washing time and ozone dose was found for the definition of the operative process parameters. The liquid supernatants were characterized and the values were conforming to the discharge in the wastewater treatment. After the stabilization phase the organic matter and the nutrients contents were in agreement with the values of the fertilizer legislation. Future investigation on the complete destiny of the metals contents have to be realized.

REFERENCES

Fonti, Dell'Anno, Beolchini, 2013, Influence of biogeochemical interactions on metal bioleaching performance in contaminated marine sediment. *Water Research*, **47**, 5139-5152.

Hospido, Carballa, Moreira, Omil, Lema, Feijoo, 2010, Environmental assessment of anaerobically digested sludge reuse in agriculture: Potential impacts of emerging micropollutants. *Water Research*, **44,** 3225-3233

Jones-Lepp, T.L., Stevens, R., 2007, Pharmaceuticals and personal care products in biosolids/sewage sludge: the interface between analytical chemistry and regulation. *Analytical and Bioanalytical Chemistry*, **387 (4)**, 1173–1183.

Macía, Fernández-Costas, Rodríguez, Sieiro, Pazos, Sanromán (2014) Technosols as a novel valorization strategy for an ecologicalmanagement of dredged marine sediments. *Ecological Engineering*, **67**, 182–189.

Netzband, A., Adnitt, C., 2009. Dredging management practices for the environment: a structured selection approach. *Terra Aqua J.*, **114**, 3–7.

Puchongkawarin, Gomez-Mont, Stuckey, Chachuat, 2015, Optimization-based methodology for the development of wastewater facilities for energy and nutrient recovery, *Chemosphere*, **140**, 150–158

Purves D., 2012, Trace-Element Contamination of the Environment. Revised Version. Elsevier Edition.

Rulkens, W., 2005. Introduction to the treatment of polluted sediments. Reviews. *Environmental Science and Biotechnology*, **4 (3)**, 213-221

Sabra, N.Y., Dubourguier, H.C., Benmimouna, A., Duval, M.N., Camuzeaux, S., Hamieh, T., 2011. Lithotrophic bacterial leaching of heavy metals from sediments dredged from the Deuˆ le Canal, France. *Open Environmental Sciences*, **5**, 18-29.