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Research article

Electro-dewatering treatment of sludge: Assessment of the influence on relevant indicators for disposal in agriculture

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ABSTRACT

Waste activated sludge requires effective dewatering, high biological stability and retention of nutrients prior to disposal for agricultural application. The study was conducted to evaluate the impact of pressure-driven electro-dewatering (EDW) on improving sludge characteristics related to disposal in agriculture, including biological stability, pathogen availability, heavy metals concentrations and nutrients content. Thickened conditioned and mechanically dewatered sludge samples were collected from two wastewater treatment plants (WWTPs), characterized by different stabilization processes, and treated by a lab-scale device at 5, 15 and 25 V. EDW increased significantly the dry solid (DS) content, up to 43–45%, starting from 2 to 3% of raw sludge. The endogenous value of specific oxygen uptake rate (SOUR), monitored as indicator of biological stability, increased up to 56% and 39% after EDW tests for sludge from two WWTPs. On the other hand, the exogenous SOUR decreased, indicating a significant drop in the active bacterial population. Likewise, a 1–2 log unit reduction was observed for *E. coli* after EDW tests at 15 and 25 V. However, no remarkable removal of heavy metals, namely chromium, nickel, lead, copper and zinc, was achieved. Finally, the concentration of nutrients for soil, such as carbon, nitrogen, phosphorus and sulfur, was not affected by the EDW process. In conclusion, EDW exerts considerable effects on the biological characteristics of sludge, which should be considered in a proper design of sludge management to ensure safe and sustainable resource recovery.

1. Introduction

In recent years, sludge treatment and management became a bottleneck in wastewater treatment plants (WWTPs) due to the high costs for sludge disposal and the environmental impact resulting from inappropriate dumping (Yang et al., 2015). Therefore, sludge dewatering is required before disposal for ease of handling and minimization of transport costs. Various processes, such as ultrasonication (Pérez-Elvira et al., 2006), Fenton combined with ultrasonication (Ning et al., 2014) or homogenous/heterogeneous Fenton (Tao et al., 2019), alternative Fenton processes (Kałuzna-Czaplińska et al., 2010; Rumky et al., 2018), bioleaching (Pathak et al., 2009) along with others advanced oxidation processes (AOPs) (Liu et al., 2018; Zhen et al., 2012), emerged lately as promising alternatives for enhancing the performance of sludge dewatering with metal and phosphorus recovery. Among these processes, electro-osmosis has gained attention and has been deeply studied as an efficient dewatering process to increase the dry solid (DS) content in the sludge. The application of an electric field combined with pressure can increase the DS content up to 45%, which is much higher than that achieved by mechanical processes only (Xu et al., 2016). During pressure-driven electro-dewatering (EDW) process, the applied pressure reduces the pores volume and squeezes all the free water from sludge, while electro-osmosis enhances the surface adhesion of the sludge and induces the available water to migrate towards the cathode. EDW process may be considered also as an alternative to thermal drying process

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Abbreviations list

AOP	advanced oxidation process
CFU	colony forming units
CST	capillary suction time
DO	dissolved oxygen
DS	dry solid
EDW	electro-dewatering
MD	mechanically dewatered
MRD	maximum recovery diluent
OUR	oxygen uptake rate
OUR_{ENDO}	endogenous oxygen uptake rate
OUR_{EXO}	exogenous oxygen uptake rate
SOUR	specific oxygen uptake rate
$SOUR_{ENDO}$	specific endogenous oxygen uptake rate
$SOUR_{EXO}$	specific exogenous oxygen uptake rate
TC	thickened conditioned
TU	thickened unconditioned
VS	volatile solid
XPS	X-ray photoelectron spectroscopy
WWTP	wastewater treatment plant

due to its lower energy consumption (Mahmoud et al., 2010). In fact, literature reported that EDW process can decrease the energy consumption up to 25% to achieve a sludge dryness up to 60% (Barton et al., 1999; Mahmoud et al., 2011).

Considering sludge disposal routes, while the influence of mechanical dewatering on the sludge characteristics has been widely studied, many related aspects are still unknown for the EDW process. For instance, there is a strong debate regarding the land application of sludge: extensive results about pathogenic contamination have already been reported (Bruce et al., 1990; Gerba and Smith, 2005) but relatively little literature is present in relation to EDW process. Moreover, the effect of EDW on the biological characteristics of treated sludge has not been investigated yet. In the European Union, 30% of sewage sludge is reused as fertilizer in agriculture (Sánchez et al., 2017). Therefore, since sludge disposal in agriculture may determine adverse effects caused by pathogens, with consequent human health risks, and nuisance odour problems, an assessment on the influence of EDW process on the viability of pathogens and biological stability is strongly needed (Huang et al., 2008; Yin et al., 2018).

Besides, the presence of heavy metals in sludge should be considered to minimize health risks when disposed in agriculture. During the EDW process, electro-osmosis and electromigration are regarded as the main phenomena involving heavy metals, since H^+ ions are generated from water electrolysis and move from the anode to the cathode, where water is removed. The migration of protons causes a pH modification in the sludge cake profile, resulting in high acidity next to the anode and high alkalinity next to the cathode (Yoshida et al., 1999), and might allow dissolution of metals at the anodic side, from which they can migrate towards the negatively charged electrode. Some researchers studied the effect of EDW on the concentration of heavy metals in the sludge cake (Feng et al., 2014; Wang et al., 2005) but results were ambiguous and a definitive conclusion was not achieved (Tuan and Sillanpää, 2010; Yuan and Weng, 2003).

Furthermore, from an agronomic point of view, sludge can provide essential nutrients for plants, such as nitrogen, phosphorus, calcium, magnesium and potassium. These elements are present in large quantities whereas micro-elements, such as iron, copper, zinc and manganese are found in variable amounts (Bratina et al., 2016; Frišták et al., 2018). Therefore, the study of the influence of EDW process on the migration and possible separation of nutrients becomes essential when sludge is disposed in agriculture.

By considering all these issues, a robust functional approach was carried out to assess the influence of the EDW process on sludge biological stability, pathogen availability, heavy metals concentration and nutrients content in relation to disposal in agriculture. Lab-scale EDW tests at various fixed electric potentials (5, 15 and 25 V) have been performed on thickened conditioned and mechanically dewatered sludge samples from two WWTPs. To evaluate biological stability, endogenous oxygen uptake rate (OUR_{ENDO}) and exogenous oxygen uptake rate (OUR_{EXO}) have been determined. Moreover, the presence of *Escherichia coli* and heavy metals, namely chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn), has been measured. Finally, nutrient content was assessed by elemental analysis and X-ray photoelectron spectroscopy (XPS).

2. Materials and methods

2.1. Sludge samples and reagents

Thickened unconditioned (TU) sludge samples were collected from two WWTPs located in the Milan metropolitan area, in Italy (Fig. 1). In addition, mechanically dewatered (MD) sludge was sampled after belt filter press in WWTP 1. After collection, sludge samples were stored at 4 °C for less than five days before processing to keep their properties unaltered.

Potassium dihydrogen phosphate (Carlo Erba), magnesium sulfate (Sigma Aldrich), sodium chloride (Merck), iron(III) sulfate hydrate (Carlo Erba), manganese sulfate monohydrate (Baker) and sodium bicarbonate (Carlo Erba) were used to prepare Winogradsky's salt solution for OUR tests. Sodium acetate solution (Sigma Aldrich) was used as nutrient for bacteria during OUR test. Sulfuric acid (Sigma Aldrich) was used to stabilize the sludge at pH = 2 before ICP-MS analyses. For pathogens quantification, peptone (Thomas Scientific) and sodium chloride (Sigma Aldrich) were used to prepare the maximum recovery diluent (MRD) and chromogenic agar (Scharlau Science Group Microinstant Chromogenic Coliforms Agar) was used as selective medium for the detection of *E. coli*.

2.2. Sludge conditioning and characterization

Thickened conditioned (TC) sludge samples were obtained by lab-scale jar tests (Fig. 1), in which the conditioning process at two WWTPs was simulated in order to study its effect on the EDW performance. Polyamidic and high cationic polyelectrolyte (Praestol 645 BC) was used at a dosage of 6 and 10 g/kg_{DS}, respectively.

Initial DS amount, volatile solids to dry solids (VS/DS) ratio and capillary suction time (CST) were measured according to Standard Methods (APHA/AWWA/WEF, 2012). Electrical conductivity was monitored by a conductivity meter (B&C Electronics-C 125.2) and pH by a pH-meter (Metrohm 827 pH Lab). Conductivity and pH on mechanically dewatered sludge samples were measured after a 1:10 dilution with tap water. Sludge samples were filtered under vacuum with a Whatman 42 filter cloth (2.5 μm pores size) and the zeta potential of the filtrate was determined by the instrument Malvern Zetameter ZS90.

2.3. EDW tests

The lab-scale setup for EDW tests was reported in a previous work by Visigalli et al. (2017)a,b. As shown in Fig. 1, the procedure of electro-dewatering tests on TC sludge samples can be summarized in three stages: (1) centrifugation in the laboratory at 4000 rpm for 5 min; (2) compression and filtration of 90 g centrifuged sludge in the device for 10 min, by applying a pressure of 300 kPa; (3) electric potential application at various fixed voltages (5, 15 and 25 V) for 25 min, keeping the pressure constant at 300 kPa. EDW tests on MD sludge were performed with the same procedure, by pouring 55 g of sample directly in the device, without the preliminary centrifugation. At the end of the

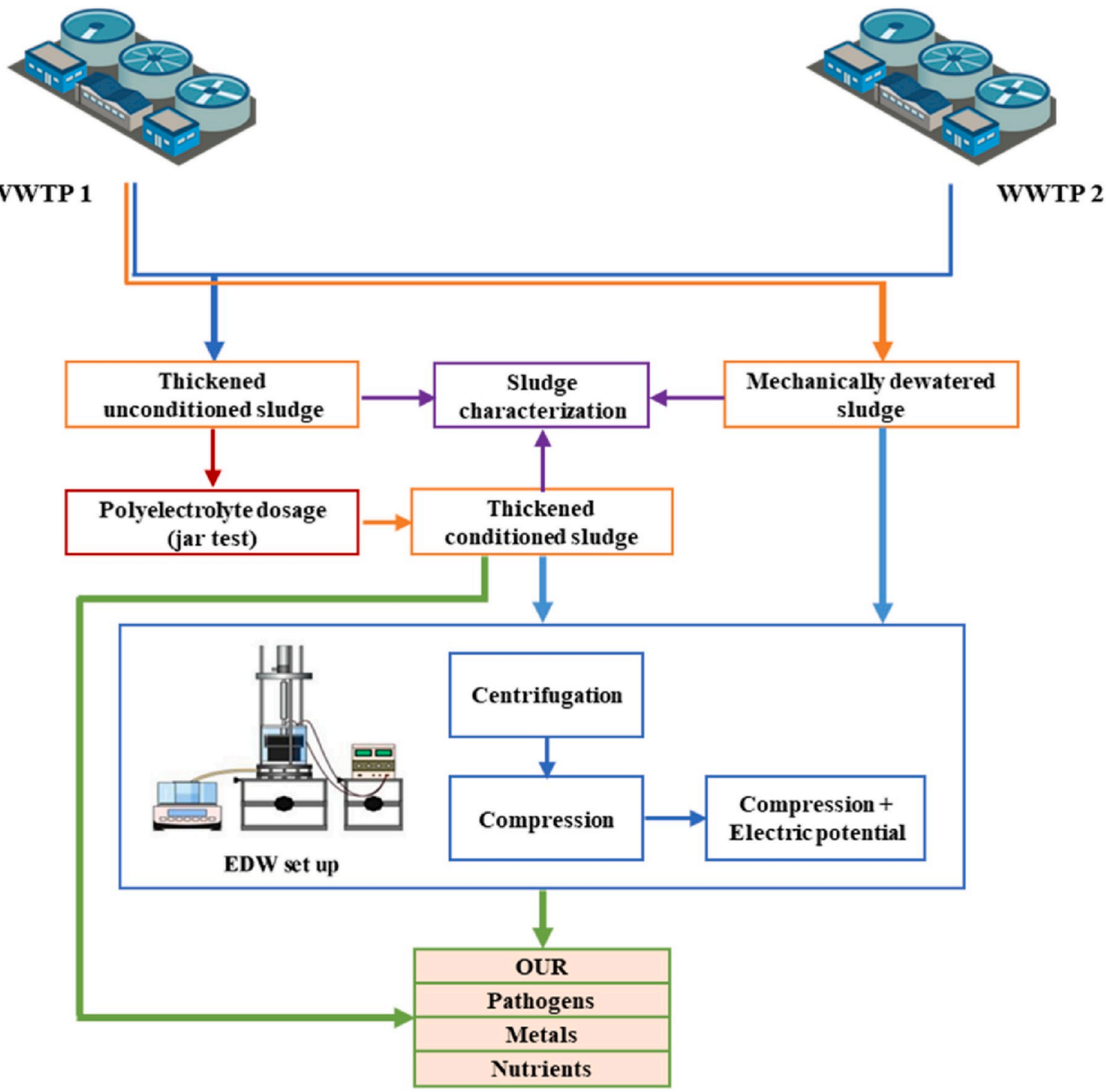

Fig. 1. Flow chart for the structure of the research work.

EDW tests, final DS content and VS/DS ratio were measured by Standard Methods (APHA/AWWA/WEF, 2012). During the EDW tests, the weight of removed water and the electrical current values were recorded every minute. Each test has been carried out in triplicate, as all tests and analyses reported in the following analyses, except for SEM, elemental and XPS.

2.4. Surface morphology of sludge

A scanning electron microscope (SEM, Hitachi S-4800) was used to observe the surface micro-morphology of the thickened conditioned sludge samples before and after the EDW process.

2.5. OUR tests

Sludge samples were diluted and mixed with Winogradsky's salt solution (Sewalem et al., 2011) to get a final concentration of 8 g_{DS}/L. The diluted suspension (500 mL) was poured in a 1 L-beaker, placed on a magnetic stirrer and aerated until oxygen saturation was achieved. During OUR tests, dissolved oxygen (DO) and pH were monitored by a Hach Lange HQ30D portable meter. Then, aeration was stopped and DO values started decreasing (endogenous oxygen uptake); after some minutes, 10 mL of sodium acetate were added to evaluate the exogenous oxygen uptake. OUR tests were stopped when DO reached values around 2–3 mgO_2/L.

The OUR_{ENDO} was computed as the slope of DO values vs. time curve from the point at which aeration was stopped until sodium acetate dosage. On the other hand, the OUR_{EXO} was calculated as the slope of the curve after sodium acetate addition until the end of the OUR test. Specific OUR (SOUR, in mgO_2/gVSS/h) was determined by dividing OUR by the biomass concentration (expressed as volatile suspended solids, VSS).

2.6. Microbial quantification

E. coli enumeration in sludge was carried out by the membrane filtration procedure (Standing Committee of Analysts, 2003). Briefly, peptone (1 g) and sodium chloride (8.5 g) were dosed to prepare the maximum recovery diluent (MRD), which was autoclaved for 20 min at 121 °C. The sludge sample was diluted (1:10) with the MRD and the solution was homogenized by orbital shaking for 1 h at 160 ± 20 rpm. The required volume of diluted sample was filtered on a membrane that was later transferred to the Petri dish containing chromogenic agar. Petri dishes were incubated at 44 °C for 24 h and the blue-violet colonies, corresponding to *E. coli,* were counted to estimate the colony forming units (CFU) per unit of volume.

2.7. Heavy metals

The presence of Cu, Ni, Pb and Zn was measured according to UNI EN ISO 17294–1: 2007 method, while Cr according to APAT IRSA 3150B1:2003 method. Analyses were performed by inductively coupled plasma mass spectroscopy (ICP-MS, Agilent Technologies 7700).

2.8. Elemental and XPS analyses

Sludge elemental composition and carbon, nitrogen and sulfur contents were determined by elemental analyzer (Thermo Fisher Scientific Flash 2000). Phosphorous concentration was measured by XPS (Thermo Fisher Scientific ESCALAB 250Xi). To recognize surface functional groups, XPS was used in an energy range between 0 and 1200 eV.

3. Results and discussion

3.1. Sludge characteristics

Table 1 lists main characteristics of sludge samples collected from two WWTPs. According to Table 1, the DS content of TC samples decreased slightly from the initial value of TU sludge as polyelectrolyte was added for sludge conditioning. Simultaneously, CST decreased and zeta potential increased after conditioning. These results evidence the occurrence of sludge flocculation, resulting in an improvement of sludge filterability.

3.2. EDW tests

TC samples from two WWTPs and mechanically dewatered (MD) sludge from WWTP 1 were tested by EDW. Fig. 2 shows the DS content of sludge samples after EDW.

The DS content of TC samples from WWTP 1 increased from 3.0% to 20.4%, 30.8% and 42.2% at 5, 15 and 25 V, respectively, whereas that of sludge from WWTP 2 increased from 3.4% to 25.9%, 40.3%, and 44.3%. In general, TC sludge from WWTP 2 showed better EDW performance at 5 and 15 V, but the process saturated at 25 V, due to the high electrical resistance of sludge cakes, with a final DS content similar for the two types of sludge. Indeed, as described in Visigalli et al. (2017)a,b, as soon as the sludge cake has formed and a significant amount of free water has been removed, electrical resistance starts rising, currents decrease, and the water removal slows down. The DS content of MD sludge from WWTP 1 reached a final dryness of 31.3% at 15 V and 43.9% at 25 V. These results showed that the electric field application directly coupled with mechanical compression can lead to a considerable reduction of sludge moisture content (Mahmoud et al., 2018).

3.3. Surface morphology of sludge

SEM was used to study surface morphology on sludge samples before and after EDW at 15 and 25 V. In Fig. 3a and d, the morphology of TC samples from WWTP 1 and 2 is shown. Fig. 3b and e shows SEM images after EDW at 15 V. Similarly, Fig. 3c and f shows SEM images after EDW at 25 V: different sizes of pores and sludge particles were observed. It was evidenced that sludge morphology changed after EDW with respect to TC samples, generating smaller pores and a more compact structure, with particles isolated from each other (Feng et al., 2014).

Since MD sludge from WWTP 1 had an initial DS content of about 22%, the sludge structure was compact, with big particles and few pores (Fig. 3g). After EDW, as shown in Fig. 3h and i, particles remained compact but had smaller sizes, due to water separation.

In raw sludge, SEM morphology showed the presence of larger particles with flaky surface. When EDW was applied with external pressure, sludge particles were broken (Yang and Lin, 2017). When subjected to an electric field, a transmembrane potential might have been induced by the presence of opposite charges on either side of the cell membrane of microorganisms. This may have caused a compression of the cell membrane itself due to the attraction between these opposite charges, causing irreversible electroporation and disrupting the cells. Moreover, the increase of temperature due to Joule effect may also have accelerated the rupture of the cells, releasing the cytoplasm (Feng et al., 2014).

Therefore, the electric potential allowed to remove free water and part of the interstitial water from the sludge. The breakdown of particles and the consequent water removal were essential to increase the final DS content in sludge. However, these phenomena may have involved not only the water, but also elements and ions in the sludge cake and dissolved in water.

3.4. OUR tests

Fig. 4a shows the OUR curves for sludge samples from WWTP 1 after conditioning (TC) and after EDW tests. Initial DO was around 7.2–8.0 mg/L and, when aeration was stopped, the DO of TC sample decreased down to 6.8 mg/L. At this point, sodium acetate was added and DO values decreased at higher rate: the rate of exogenous phase was evidently higher than that achieved in the endogenous phase. On the other hand, considering the endogenous phase of samples after EDW, the slope of DO vs. time curves increased with applied electric potential. On the contrary, after the addition of sodium acetate, the slope of curves for samples after EDW at 15 and 25 V did not show any relevant variation with respect to the endogenous phase. The same behavior has been observed for TC samples from WWTP 2 and MD sludge from WWTP 1 (Fig. 4b and c).

Fig. 5 shows average endogenous and exogenous SOUR values of the sludge samples, collected before and after EDW tests. Regarding sludge from WWTP 1, endogenous SOUR increased from 2.3 $mgO_2/gVSS/h$ for TC sample to 3.8 $mgO_2/gVSS/h$ after EDW test at 25 V. On the other hand, exogenous SOUR decreased of 90%, as the value reduced from 10.4 $mgO_2/gVSS/h$ to 1.7 $mgO_2/gVSS/h$. As regards sludge from WWTP 2, endogenous SOUR increased from 1.1 $mgO_2/gVSS/h$ up to 1.9 and 2.5 $mgO_2/gVSS/h$ after EDW at 15 and 25 V, while the exogenous SOUR decreased from 1.6 $mgO_2/gVSS/h$ down to 0.6 $mgO_2/gVSS/h$, meaning a 62.5% reduction with respect to raw sludge. Conversely, considering the MD sample from WWTP 1, endogenous SOUR gradually increased from 1.3 $mgO_2/gVSS/h$ up to 4.0 $mgO_2/gVSS/h$ after EDW at 25 V, while exogenous SOUR decreased of 63.2%, as the initial content dropped from 9.8 $mgO_2/gVSS/h$ to 3.6 $mgO_2/gVSS/h$.

For all sludge samples, endogenous SOUR increased with the electric

Table 1
Main characteristics of sludge samples (mean ± st.dev.) collected from two WWTPs.

WWTP No.	ID	Polymer dosage	DS	VS/DS	CST	Conductivity	pH	Zeta potential
		g/kg_{DS}	wt%	wt%	s	mS/cm	–	mV
1	**TU**	0.0	2.8 ± 1.18	63.2 ± 6.66	19.9 ± 6.52	1.1 ± 0.39	7.0 ± 0.30	−11.1 ± 1.50
	TC	6.0	2.5 ± 0.27	66.9 ± 6.19	10.7 ± 2.36	1.3 ± 0.11	6.8 ± 0.28	−9.3 ± 0.85
	MD	5.3	18.4 ± 1.84	65.3 ± 5.48	–	1.2 ± 0.26	6.6 ± 0.39	–
2	**TU**	0.0	3.5 ± 0.19	60.2 ± 2.11	131.2 ± 45.30	4.7 ± 0.77	7.2 ± 0.20	−11.8 ± 0.98
	TC	10.0	3.3 ± 0.70	59.5 ± 2.29	25.7 ± 18.57	4.6 ± 0.75	7.4 ± 0.40	−8.7 ± 0.76

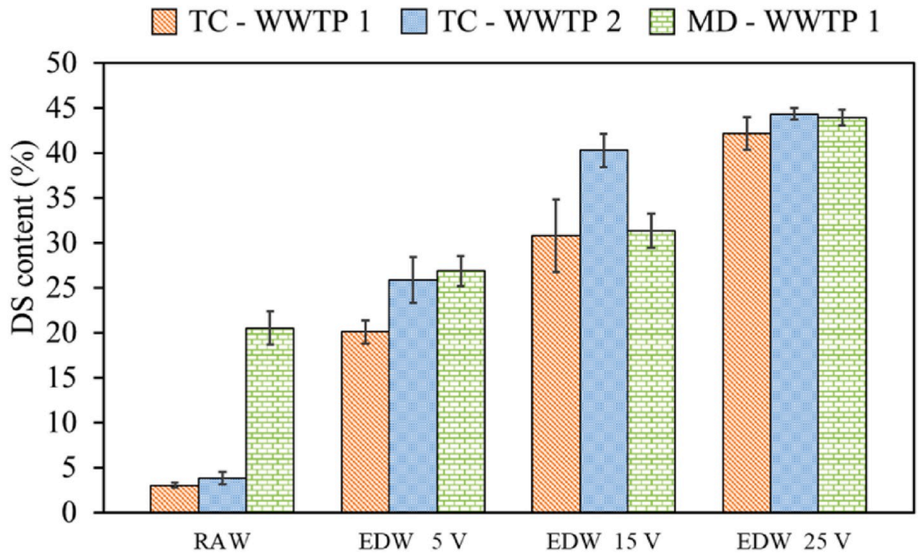

Fig. 2. DS content of TC and MD sludge samples taken from WWTP 1 and WWTP 2 before and after EDW tests at various electric potentials (5, 15 and 25 V).

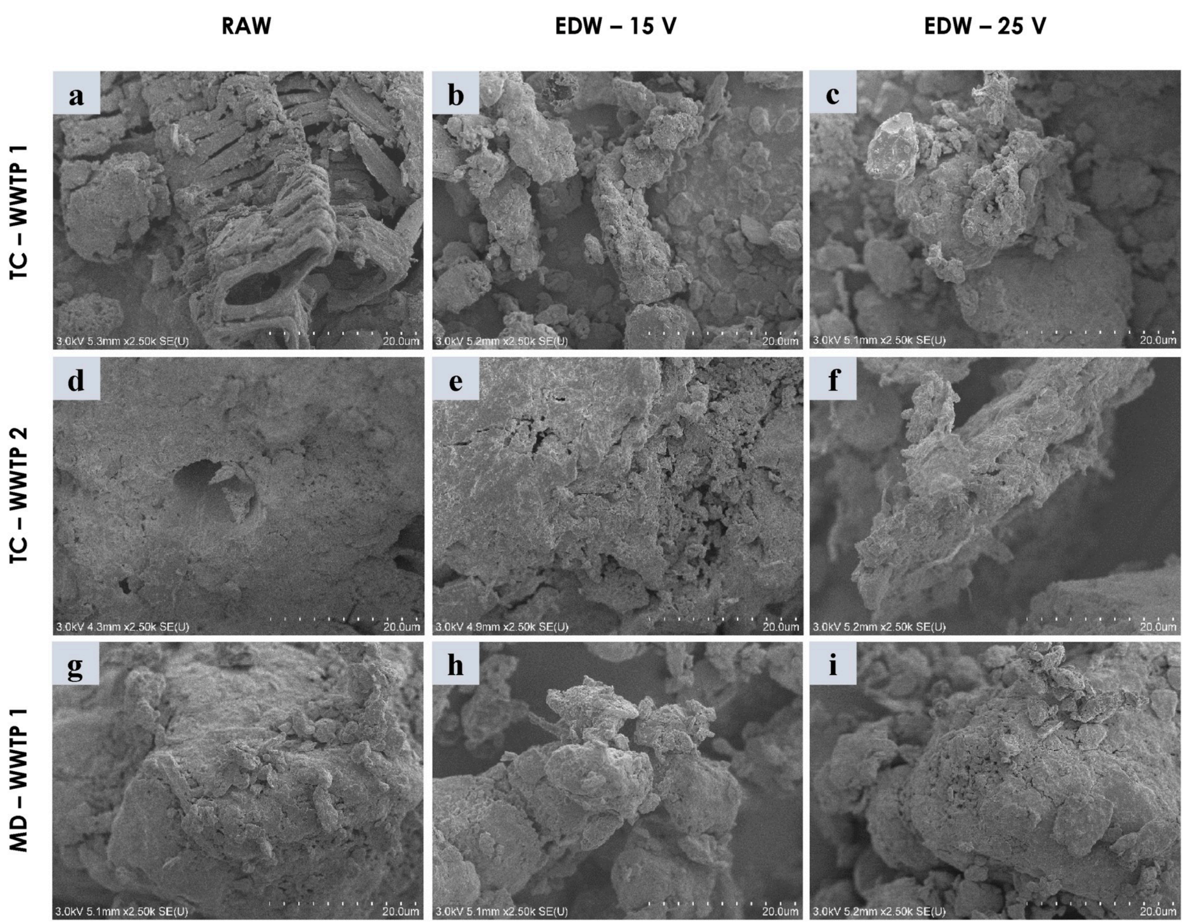

Fig. 3. SEM images (2,500x) of sludge. Samples from WWTP 1: (a) Raw TC, (b) TC after EDW at 15 V and (c) TC after EDW at 25 V, (g) Raw MD, (h) MD after EDW at 15 V and (i) MD after EDW at 25 V. Samples from WWTP 2: (d) Raw TC, (e) TC after EDW at 15 V and (f) TC after EDW at 25 V.

potential applied during EDW tests, while exogenous SOUR tended to decrease. It can be assumed that, by applying an electric potential of 15 and 25 V, the active biomass was reduced due to a significant temperature rise, resulting from ohmic heating (Ibeid et al., 2013), while substrates and enzymes were released in the sludge due to hydrolysis of dead cells. Indeed, higher is the electric potential applied, greater is the increase in the electrical resistance due to a faster removal of free water from the sludge cake. This enhances the ohmic effect, with higher temperatures at higher applied voltages, which in turn increases the extent of bacteria inactivation. Therefore, the increase in the endogenous SOUR after the EDW was caused by the greater availability of biodegradable organic carbon, which became food for the viable

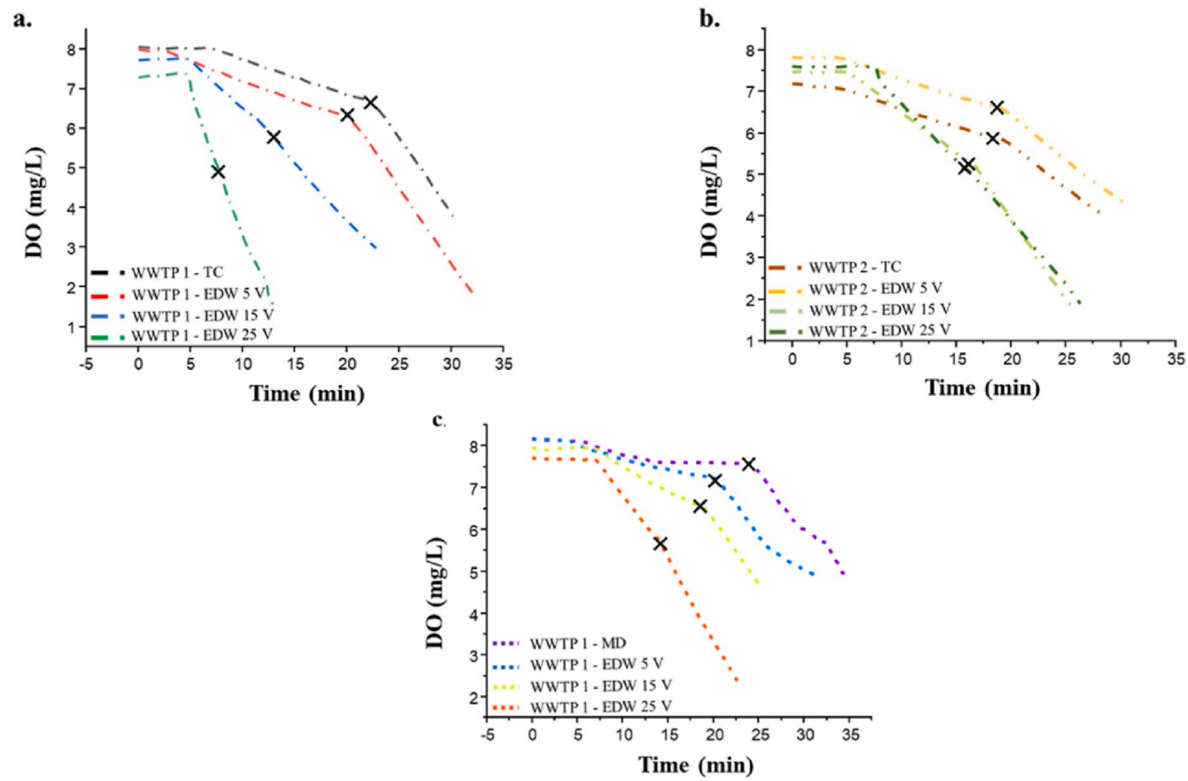

Fig. 4. Results of OUR tests for different types of sludge: (a) TC from WWTP 1, (b) TC from WWTP 2, (c) MD from WWTP 1. *X* shows the time at which sodium acetate was added.

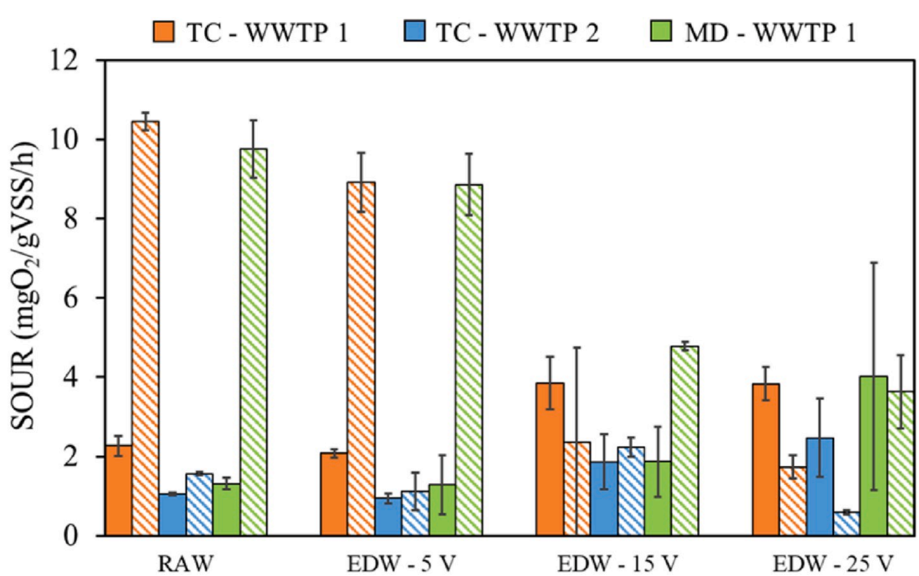

Fig. 5. SOUR for different sludge samples (TC from WWTP 1, TC from WWTP 2, MD from WWTP 1) before and after EDW tests at various electric potentials (5, 15 and 25 V). Endogenous and exogenous SOUR values are reported respectively as solid and patterned bars.

bacteria and increased their respiration rate. However, the overall viable biomass decreased. Exogenous SOUR was significantly reduced after EDW at 15 V and 25 V, meaning that OUR of bacteria was not affected by the addition of external carbon. A possible explanation might be that the remaining viable bacteria were already oversaturated by carbon source released from dead cells. Moreover, Sha et al. (2019) reported that EDW tests at high voltage (e.g. 40–60 V) can decrease SOUR by creating cracking in microorganism structure and releasing organic content. On the other hand, after EDW at 20 V, SOUR increased of 8.7% due to the relatively low current values that can boost up bacterial activity, as in Millanar-Marfa et al. (2018). Therefore, the application of an electric potential at 5 V (Fig. 5) did not significantly affect sludge biological stability.

By comparing TC samples from two WWTPs, the anaerobically stabilized sludge (WWTP 2) had lower initial biological activity with respect to aerobically stabilized one (WWTP 1), indicating a better stabilization during the preliminary treatment phase. Indeed, TC sludge from WWTP 2 had a total SOUR of 2.6 $mgO_2/gVSS/h$, much lower than that of samples from WWTP 1, namely 12.7 $mgO_2/gVSS/h$ for TC and 11.1 $mgO_2/gVSS/h$ for MD sludge. Their endogenous SOUR changed similarly after EDW tests, while the reduction in exogenous SOUR was more evident for WWTP 1, probably indicating a bigger stress caused by EDW to bacteria. TC and MD samples taken from WWTP 1 had similar endogenous and exogenous SOUR, meaning that mechanical stress was not sufficient to modify biological activity of bacteria. This was confirmed also by comparing raw samples and sludge after EDW at 5 V, which had similar SOUR values. Indeed, at 5 V the dewatering efficiency was mainly related to the compression phase due to the low current densities developed during the process and the ohmic effect was negligible.

Besides, sludge can be considered stabilized if the total SOUR is as low as 1 $mgO_2/gDS/h$ (Kazimierczak, 2012), while aerobically stabilized sludge can be categorized as 'class A biosolids' only if the SOUR value is lower than 1.5 $mgO_2/gDS/h$ (US EPA, 1992). In this work, SOUR values after EDW tests ranged from 1.4 to 7.2 $mgO_2/gDS/h$, evidencing that the EDW process was not fully efficient in stabilizing sludge under applied conditions. However, to understand the effect of the electric field on the viability of microorganisms, a deeper insight on the presence of bacteria in the cake after EDW needs to be carried out.

3.5. Presence of pathogens in the sludge before and after EDW

The quantification of microorganisms in sludge samples before and after EDW can be used as a useful indicator to assess the reduction of pathogenic contamination due to EDW (Winfield and Groisman, 2003). As observed from OUR tests, the biological activity on samples taken after EDW at 5 V was similar to that measured on TC sludge. For this reason, the presence of *E. coli* was determined only after EDW at 15 and 25 V, where the effect of EDW process was evident (Fig. 6).

The highest values of *E. coli* were found in TC sludge from WWTP 1, about 1.61×10^3 CFU/gDS, whereas TC sludge from WWTP 2 and MD sludge from WWTP 1 had concentrations of 1.47×10^3 and 1.27×10^3 CFU/gDS, respectively. The presence of *E. coli* in sewage sludge has been reported widely in literature. For example, the presence in an Austrian WWTP was reported as 8.5×10^5 CFU/gDS (Reinthaler et al., 2010). Cooper et al. (2010) reported that anaerobically stabilized sludge, on average, had *E. coli* concentrations around 1.0×10^4 CFU/gDS, much lower than raw sludge samples that had concentrations of 3.2×10^6 CFU/gDS. Moreover, centrifuge dewatering of anaerobically digested sludge evidenced a significant increase in the *E. coli* content with respect to the same sample before dewatering. Therefore, the initial *E. coli* concentrations in the studied sludge samples were one or more orders of magnitude lower than the values commonly found in literature.

After EDW tests at 15 and 25 V, *E. coli* concentration dropped by 1–2 orders of magnitude. *E. coli* in TC sludge from WWTP 1 decreased of 1.0 and 1.7 log units after EDW tests at 15 and 25 V, whereas for WWTP 2 the *E. coli* concentration was reduced of 1.1 and 2.3 log units at 15 and 25 V, respectively. Therefore, the decrease in *E. coli* concentration in the sludge after EDW tests at 15 and 25 V evidenced the effect of EDW process in reducing the pathogens in both aerobically and anaerobically treated sludge samples. As already discussed, the increase of the temperature in sludge cake after EDW at 15 and 25 V might be a reason for the lower residual microbial presence in sludge (Yin et al., 2018). Indeed, Navab-Daneshmand et al. (2012) found that oxidants produced by electrochemical reactions and the extreme pH were secondary factors, while the high temperature arising from Joule heating was the main cause for inactivation of total coliforms and *E. coli* by EDW.

Therefore, the application of an electric field not only increases the DS content of sludge, making it suitable for incineration without preliminary thermal treatments (Visigalli et al., 2017b), but also has the side effect of reducing the concentration of *E. coli* in sludge. The reduction in the viable number of pathogens due to EDW may be helpful in reducing the biological hazard when the sludge is applied to agricultural land. Moreover, also environmental conditions can have a considerable effect on microorganisms when the sludge is used in agriculture. For example, the pathogen concentration may be furtherly reduced when soil is hot and dry, its pH is low or with the influence of ultraviolet irradiation (Andreoli et al., 2007).

3.6. Heavy metals

As already introduced, in the EDW process the highly acidic pH developed in proximity of the anode leads to cations dissolution that may then migrate towards the cathode and be removed together with the filtrate (Feng et al., 2014). Among these cations, heavy metals should be considered in order to evaluate if the EDW process has an effect on their concentration in sludge.

Table S1 in Supplementary material reports the concentrations of heavy metals in sludge before and after EDW tests at 15 and 25 V. Also in this case, only these electric potentials were investigated as a consequence of results obtained for OUR tests, indicating that EDW did not influenced relevantly the sludge at 5 V. No significant results were observed in heavy metals transfer from sludge to the separated liquid phase.

One of the hindering factors for metals removal may be ascribed to the development of a highly alkaline pH at the cathode, which makes the

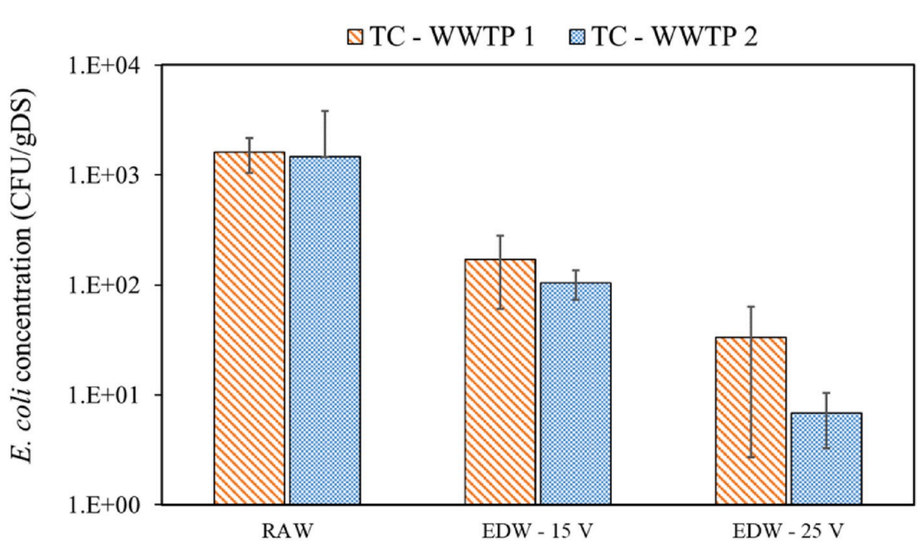

Fig. 6. *E. coli* quantification for different sludge samples (WWTP 1 - TC and WWTP 2 - TC) before and after EDW tests at various electric potentials (15 and 25 V).

cations to precipitate as hydroxides and slows down their electromigration. Virkutyte et al. (2002) reported that if pH is below 4.5 then the metal hydroxide precipitation was minimal. Wang et al. (2005) showed that metal removal could be enhanced by sludge acidification around pH 2 at the cathode: removal efficiencies of 68% for Cr, 95% for Zn, 96% for Cu, for 90% Ni and 19% for Pb were observed.

However, in spite of scarce metal removal efficiency (Table S1), heavy metals in sludge samples were always lower than the maximum threshold allowed for agricultural use of sludge (Council of European Communities, 1986). Therefore, sludge taken after the EDW process was acceptable for use as biosolid in agricultural land, public contact sites and forest.

3.7. Elemental and XPS analyses

Sludge can be used as an organic supplement instead of compost thanks to the significant amount of substrates that allows soil enrichment with nitrogen, phosphorus, calcium, magnesium, potassium and other nutrients (Zaman et al., 2002). It is worth to notice that the bioavailability of elements in soil can be affected by the source of the biological waste, its character, composition and processing (Hudcová et al., 2019). Therefore, it is important to investigate the influence of EDW, which may cause electromigration of nutrients outside the cakes, on the elemental content of sludge. The concentrations of carbon, nitrogen, sulfur and phosphorus in sludge samples before and after EDW are shown in Table 2.

Carbon represents the energy source for composting. Here, the concentration of carbon in sludge samples ranged from 28.2% to 31.0% for sludge samples from WWTP 1 and from 33.7% to 34.5% for sludge samples from WWTP 2, without remarkable influence by the EDW.

The concentration of nitrogen in TC and MD samples from the two WWTPs ranged from 3.5% to 4.6%, with lower values for sludge from WWTP 2. After EDW, the amount of nitrogen was similar to that of raw samples, evidencing that the application of the electric field did not affect its concentrations in sludge. Nitrogen is necessary for the protein synthesis and can be regarded as a top priority nutrient for soil and plants, which generally need concentrations of 3–4% (Andreoli et al., 2007).

In general, a C-range of 22–30% and a N-range of 1–4% are regarded as ideal contents for disposal of sludge in agriculture (Andreoli et al., 2007; Van Oorschot et al., 2000). Therefore, the sludge samples studied in this work, even after the application of the electric field, resulted to be suitable to be used as a fertilizer for plants.

EDW did not result in a noticeable effect on sulfur concentration in TC sludge samples but had a slight influence in the reduction of sulfur amount for MD sludge from WWTP 1. Sulfur belongs to the secondary nutrient category, but it is important in the formation of several proteins and activation of enzymes. However, an excessive concentration can be toxic for the crops, since it can reduce soil pH (Järvensivu, 2015).

Phosphorus, together with nitrogen, is one of the most important nutrients for plant growth. XPS analysis were performed to measure its concentration before and after EDW process (Supplementary Material, Fig. S1 and Fig. S2). The content of phosphorus in sludge samples from WWTP 1 ranged from 1.8% to 2.4%, without any significant correlation to EDW. These results evidenced that no significant reduction in phosphorus content was found after the application of the EDW.

In conclusion, elemental analysis and XPS assessed that EDW does not affect the concentration of nutrients and that the studied sludge samples may be used as fertilizer in agriculture, similarly and with the same advantages of sludge not treated by EDW.

3.8. Future developments

The effect of EDW on pathogen viability, biological stability and concentration of heavy metals and nutrients should be further assessed by considering a larger number of WWTPs. In order to limit the release of organic carbon and enzymes during the EDW process, a further assessment of operating conditions, in terms of voltage and process duration, on sludge biological activity needs to be further studied. For example, the application of an additional stabilization treatment after EDW can be assessed. Moreover, in current experiments, heavy metals removal was probably hindered by their precipitation as hydroxides nearby the cathode. Electromigration of heavy metals and their removal may be encouraged by applying polarity reversal of the electrodes or by acidification of the sludge cake.

4. Conclusion

EDW treatment was studied on different types of sludge to assess the influence on relevant indicators for disposal in agriculture. The overall presence of bacteria in sludge decreased, as evidenced by the reduction in exogenous SOUR, while endogenous SOUR increase indicates that the overall biological activity is higher, although resulting from a smaller bacterial population. Indeed, *E. coli* concentration reduced by 1–2 log units after EDW at 15 and 25 V, respectively. Conversely, the concentration of heavy metals in sludge was scarcely affected by EDW. Moreover, the EDW process did not affect the concentration of nutrients (e.g. carbon, nitrogen, phosphorus and sulfur) in sludge, which can be used as fertilizer for plant growth.

In conclusion, the EDW process not only may lower the disposal costs of sludge, by increasing its DS content, but it also allows the reduction of viable pathogens before spreading sludge on agricultural land. However, as a side effect, the application of EDW may reduce sludge biological stability due to the release of organic carbon and enzymes from dead cells.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Jannatul Rumky: Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft. **Simone Visigalli:** Conceptualization, Methodology, Investigation, Resources, Data curation, Writing - review & editing. **Andrea Turolla:** Conceptualization,

Table 2
Elemental analysis and XPS analysis (% of dry matter) for different sludge samples (TC from WWTP 1, TC from WWTP 2, MD from WWTP 1) before and after EDW tests at two electric potentials (15 and 25 V).

	TC – WWTP 1			TC – WWTP 2			MD – WWTP 1		
	TC	EDW 15 V	EDW 25 V	TC	EDW 15 V	EDW 25 V	MD	EDW 15 V	EDW 25 V
C content (%)	30.98	28.87	29.93	33.84	33.74	34.50	28.30	28.16	28.99
N content (%)	4.65	4.37	4.51	3.70	3.57	3.98	4.47	4.41	4.52
S content (%)	0.25	0.18	0.19	0.43	0.39	0.42	0.18	0.06	0.04
P content (%)	1.76	2.16	2.19	NA	NA	NA	2.39	1.93	2.19

Methodology, Data curation, Writing - review & editing, Visualization. **Enrico Gelmi:** Resources, Investigation. **Chaker Necibi:** Investigation, Writing - review & editing, Supervision. **Paolo Gronchi:** Resources, Supervision. **Mika Sillanpää:** Writing - review & editing, Supervision. **Roberto Canziani:** Conceptualization, Methodology, Resources, Writing - review & editing, Project administration, Supervision, Funding acquisition.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2020.110689.

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