

Simulation of a real plant for the combined treatment of wastewaters and liquid wastes

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ABSTRACT

This paper presents the upgrading of a full-scale domestic wastewater treatment plant, operating in Southern Italy, modified so as to process domestic wastewaters together with liquid wastes, such as leachate and digestate in an integrated manner. The two streams – domestic wastewaters and liquid wastes – are separately pre-treated and then sent into a conventional denitrification/nitrification section. The preliminary treatments consist in grinding, sand decantation, flocculation and sedimentation for wastewaters and in grinding, flocculation, biological pre-treatment and advanced oxidation (Fenton) for liquid wastes. Several scenarios were analyzed using a process simulator (SuperPro Designer[®]): the concentrations of the main parameters (Biochemical oxygen demand (BOD₅); Chemical oxygen demand (COD); ammonium (NH₄⁺), nitrate (NO₃⁻), metals and total suspended solids (SST)) of the effluents were estimated as a function of BOD₅/COD ratio, initial leachate volume and nitrate percentage in the recycle stream. In each case study, the parameter values were compared with the regulatory limits provided show the intervals of variation of the main process parameters that in some cases do not permit to achieve the established limits.

Keywords: Leachate; Digestate; Domestic wastewaters; Process analysis

1. Introduction

The opportunity of treating residual liquid effluents, from solid waste treatment processes, together with domestic wastewater in a combined process is a step forward in the direction of an integrated system for sustainable water and waste management in urban areas. Examples of liquid wastes that can be treated together with domestic wastewaters are landfill leachates and digestates (liquid fraction) produced by the anaerobic digestion of organic solid waste and biomasses.

Municipal landfill leachates have a major environmental impact, because of their high ammonium, organic matter and salt concentration [1]. Leachates resulting from waste degradation in landfill sites [2] have to be processed by means of both biological and physical/chemical treatments to meet current stringent regulations. This is a difficult challenge, as leachate

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is a mixture of organic and inorganic pollutants, often recalcitrant, which include humic acids, ammonia, heavy metals, xenobiotics and inorganic salts. Moreover, its chemical and biological compositions depend on its age: young, medium and old. Young leachates, with high BOD₅/COD value (about 0.6), are characterized by a low total Kejeldahl nitrogen (TKN) concentration; medium leachates are affected by BOD_c/COD value in the range 0.06-0.6; old leachates, with a BOD₅/COD about 0.06, show a high TKN concentration with considerable recalcitrant compound content, that are very difficult to treat [3]. Therefore, medium and old leachates require combination of chemical/physical/biological treatments before discharging in sewage systems or superficial water bodies, following the regulatory limits of the Italian Regulation [4]. As a matter of fact, due to low BOD₅/COD values characterizing mediumold leachate, the biological treatment usually results in low treatment efficiencies.

Anaerobic digestion, also referred to as biogasification, is a versatile technology platform that can serve many purposes in the industry. However, besides the biogas phase - that is, the noble part of the process - a liquid stream effluent (digestate) is produced, which has to be treated before discharging in sewage system. The digestate flow is characterized by a very variable composition, depending on the biological matrix used as feed stream in the anaerobic digester. For example, the liquid digestate, resulting from the organic fraction of solid waste digestion, is a mixture of nitrogen, phosphorus, organic matter and heavy metals (e.g., iron, aluminium, zinc, nickel and chromium), that usually requires great reduction by means of physical and chemical treatments. Differently, the digestate resulting from the anaerobic digestion of other substrates, such as pig manure, contains high NH₄-N concentration and could be used as fertilizer. However, considering the transport charges, the gas emission during the storage and the constraints of the nitrate directive regulations, further treatment to remove nitrogen and organic matter from digestate is necessary before discharging into water bodies or spreading on land [5]. The review by Monlau et al. [6] describes the environmental problems caused by digestate and suggests its possible alternative valorizations. More in details, the liquid phase of digestate can be used as nutrients for algae cultivation, while the solid fraction can be used for the production of energy through biological and thermal processes and for the production of added-value products such as activated carbons through a pyrolysis process [6].

Recently, Collivignarelli et al. [7] reported the results related to the treatment of the liquid fraction of the digestate coming from the anaerobic digestion of two solid waste fractions, bovine manure and corn silage. The process was performed on a pilot scale using a thermophilic aerobic membrane bioreactor. The results after 4 months of treatments showed a removal of COD and total phosphorus greater than 94% and 87%, respectively. As a matter of fact, the digestate characteristics are not so far from those of a medium leachate, with a higher value of total solid contents and a lower TKN concentration, depending on the substrate of source.

Thus, leachate is the more difficult stream to treat. The coagulation process is often used as a pre-treatment step [8] for old and stabilized landfill leachate [9]. It is a physical–chemical technique successfully employed prior to biological or other techniques. Different coagulants are used such as aluminium sulphate $(Al_2(SO_4)_3)$, ferric sulphate

 (Fe_2SO_4) , ferric chloride $(FeCl_2)$ and polyaluminium chloride (PAC) [10,11]. This technique is very effective in suspended solids and colloids removal. Furthermore, it is an essential tool in the presence of heavy metals that may precipitate with the addition of a reactant agent, in order to meet provisions [12–15]. If lime (0–10 g/L) is used alone, COD value reduces by about 10%, thus becoming insufficient. The addition of another reactant agent such as FeCl₃ (20-100 mg/L) is to be preferred [16], together with a coagulant that can facilitate aggregation in clusters, whose dimensions favour the liquid-solid separation process. However, the coagulation process alone can result in only moderate removals of COD contents: so further treatment steps are necessary [17,18]. Particularly, advanced oxidation processes (AOPs) [19-21] appear to be a suitable treatment for such critical wastes in addition to classical biological treatments. Among the AOPs, the Fenton treatment is often used to treat industrial wastewaters or landfill leachate, but the efficiency of this process is influenced by the characteristics of the solutions, therefore, the application of Fenton has to be verified by both experimental tests and modelling before it can be implemented on a full-scale.

The choice of the effective sequence to adopt in order to achieve the best removal of all contaminants is crucial. Deng and Englehardt [22] examined different process schemes for leachate treatment. According to them the most suitable one seems to be the scheme which provides for the following sequence of treatments: biological, Fenton, biological. The first biological treatment consists in an oxidation step, which permits to increase the BOD_z/COD ratio up to 0.5 - where biological treatments are admitted - and to remove 60% of initial COD [23-25]. Besides, Gotvajn et al. [26] demonstrated that Fenton is effective in removing nitrogen (about 6%), too, if used as a pre-treatment before coagulation. On the other hand, Oller et al. [27] showed that if leachate is characterized by high biodegradable substance concentration, Fenton treatment alone can cause a high chemical consumption, such as acids (for the decreasing of pH values for wastewaters with high alkalinity), hydrogen peroxide and ferrous ions. Fenton is also characterized by a considerable production of chemical sludge. For this reason, the Fenton process has significant operating costs that may adversely affect the economic feasibility of the wastewater treatment [27,28]. Yoon et al. [29] studied the precipitation of iron from the Fenton process in the treatment of real industrial wastewaters together with landfill leachate. They investigated the effect of ferrous ions and hydrogen peroxide on the degradation of organic substances. The experimental results showed that, in a short time, a concentration of ferrous ions higher than 1 mM produced large quantities of OH radicals from hydrogen peroxide degradation, but in this case the precipitation of ferric ion occurred. In order to reduce precipitation, a concentration of ferrous ions lower than 1 mM should be considered.

In a recent paper, Prisciandaro et al. [30] explored the possibility of integrating the treatment of domestic wastewater with landfill leachate by modifying a typical domestic wastewater treatment scheme. The process analysis, performed with mass balances and considering efficiencies taken from the literature, showed that concentrations of pollutants of the effluent from the upgraded scheme were within the regulatory limits of the Italian regulation [4] for discharge in surface water bodies.

This paper deals with a treatment plant for domestic wastewater, located in Southern Italy (Calabritto, AV) was integrated with a dedicated line for the treatment of both landfill leachate and digestate stream. The authors investigated an upgraded scheme with a first biological step, where biodegradable species are removed, then a Fenton treatment, which converts non-biodegradables into biodegradables with lower chemicals consumption, followed by a final biological polishing step. The additional line includes a flocculation tank, a biological step (aeration pre-treatment [APT]) and a Fenton treatment (AOP). In this way, the two liquid wastes can be simultaneously purified together with the domestic wastewater, in order to comply with the Italian Legislative Decree 152/2006 [4] for discharge in surface water bodies. This integrated process is recommended in case of leachates or, in general, when wastewaters are characterized by a high biodegradable COD content, with low heavy metal concentrations.

2. Results and discussion

2.1. WWTP description

The wastewater treatment process of an existing plant in Calabritto (Avellino, Italy) was simulated using SuperPro Designer[®] [31], with the main goal to investigate its possible upgrade.

The plant was designed for 19,000 equivalent inhabitants (treatment capacity of 94 m³/h) however it was oversized. For this reason, it was decided to study the possibility of integrating the treatment of domestic wastewaters with leachate and digestate wastewaters.

In its current configuration (Fig. 1), the plant treats a domestic wastewater flow rate of 1.45 m³/h, in a process line composed by the following processes: chemical–physical pre-treatment, followed by flocculation with PAC, primary sedimentation, pre-denitrification, oxidation/nitrification, secondary sedimentation and disinfection. PAC was proposed in order to remove large amounts of microsuspended organic particles in water, such as cellulose, sugar and fat proteins [32].

The proposed upgrade will allow the treatment of liquid fractions of leachate (European Waste Catalogue 19.07.03), classified as not dangerous, together with digestate liquid waste, resulting from anaerobic digestion of solid wastes (EWC 19.06.03-04-05). Fig. 2 shows the Calabritto plant rearranged with additional processes (upgrading): leachate and digestate grinding, coagulation treatment with FeCl₃ and biological oxidation. The existing denitrification basin is adequate to act as an equalization basin, which collects the streams from primary sedimentation (domestic wastewater line) and Fenton's output streams (leachate and digestate line). Finally, oxidation/nitrification, secondary sedimentation (together with recycle streams from nitrification and secondary sedimentation) and disinfection, already present in the initial configuration, are carried out; moreover, a filtration unit is added to remove residual suspended impurities [33].

2.2. Process analysis

In this section, the results of the process analysis, performed using SuperPro Designer[®], are reported. Several simulations have been carried out and can be divided into: basic simulation and sensitivity analysis. The details of the results are shown in the following sections.

2.2.1. Basic simulation

Fig. 3 sketches the process scheme in SuperPro Designer[®] layout, which corresponds to the upgrading of the Calabritto plant illustrated in Fig. 2.

A simplified block scheme of the process, where three different streams were simulated – domestic wastewater (stream #1), digestate (stream #2) and leachate (stream #3) is shown in Fig. 4. Table 1 reports their composition: for domestic wastewater, flow rate and pollutant concentrations (average composition) were the real values of the Calabritto WWTP; for digestate and leachate wastewaters, were hypothesized, according to plant capacity, while pollutant concentrations (average compositions) were assumed by literature. From Table 1, it is possible to see that domestic wastewater is characterized by high nitrogen, sulphur and phosphorus contents; while digestate and leachate wastes are characterized by BOD_{5'} COD greatly higher than those of domestic wastewater.

Simulations were performed by considering the removal efficiencies reported in Table 2, taken from specialized literature [34].



Fig. 1. Current configuration of wastewater treatment plant in Calabritto.



Fig. 2. Proposed upgrading of Calabritto plant.



Fig. 3. Process scheme simulated by SuperPro Designer.

Results of simulation runs in terms of oxygen demand, expressed as $BOD_{5'}$ COD and their ratio, for the various sections of the WWT plant are reported in Fig. 5. As shown, concentrations of BOD_5 and COD are effectively reduced during the pre-treatments (sections IV, V and VI) and the biodegradability of the liquid wastes increases, thus improving their treatability with the subsequent nitro/denitro/oxidation processes.

Results of simulation runs in terms of nitrogen and SST concentrations for the various sections of the WWT plant

are reported in Fig. 6: as for SST, it can be observed that the high values of suspended solids contained in the digestate stream, are conveyed in the mixed stream coming from the equalization/denitrification tank (section 8), resulting in SST concentration increase.

This confirms the necessity to include a final filtration step, such as micellar-enhanced ultrafiltration for reduction of concentrations of residual solids and reagents used for the disinfection process, to respect the discharge limits [35–39]. As it concerns nitrogen in its different forms, the digestate



Fig. 4. Block scheme of the simulated process.

Table 1 Composition of domestic wastewater, digestate and leachate

	Units	Domestic WW (stream #1)	Digestate (stream #2)	Leachate (stream #3)
		(otream #1)	(otream #2)	(orieun #0)
Flow	m³/d	30	35	100
BOD ₅	mg/L	550	12,150	3,000
COD	mg/L	1,250	21,991	10,000
BOD ₅ /COD	mg/L	0.44	0.55	0.3
TKN	mg/L	126	1,350	1,000
NH_4^+	mg/L	101	1,200	800
NO ₃ ⁻	mg/L	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Ni	mg/L	<dl< td=""><td><dl< td=""><td>2</td></dl<></td></dl<>	<dl< td=""><td>2</td></dl<>	2
Fe	mg/L	30	50	40
Pb	mg/L	1	<dl< td=""><td>1</td></dl<>	1
Cr	mg/L	2	2	1
SST	mg/L	825	10,000	3,000

DL, Detection limit.

Table 2 Removal yields used in simulation runs

Equipment/operation Removal yield (%) BOD COD TKN NH, NO. Metals SST 7 5 Grinding 10 _ _ _ Sand decantation 10 15 15 15 70 70 Flocculation/I sedimentation 50 20 90 80 Flocculation 70 70 50 20 90 80 **APT**^a 70 60 10 10 AOP-Fenton^b 10 60 10 10 Denitrification 90 90 90 Oxidation-nitrification 90 80 30 95 **II** Sedimentation 30 25 50 10 20 85 Disinfection 20 20 _ Filtration 75 50 90 90 60 25 10

^aAPT, Aeration pre-treatment.

^bAOP-Fenton, Advanced oxidation process-Fenton.

and leachate are obviously critical streams. Fig. 6 highlights high nitrate content after the pre-treatment of the liquid waste streams, due to the nitrification naturally occurring in the biological APT (section 5, APT outlet). However, acting on nitrate recycle ratio, it is possible to respect the emission limits.

Table 3 compares the results obtained from simulation runs with discharge limits: the upgraded plant was able to produce an effluent whose concentrations were within the limits of the law dictated by the Italian Legislative Decree 152/2006 [4] for surface water bodies.

The final concentration of N–NO₃ for the basic simulation was around 17 mg/L, below the legal limit (20 mg/L). The denitrification process is an important step of the wastewater treatment and NO₃ concentration is highly dependent of bacterial activities. In the performed simulation, it was not considered all complex microorganism activity, but only the average yields of nitrogen removal taken from literature for the main biological reactions. Nitrate concentration, together



Fig. 5. BOD₅₇ COD and their ratio along the treatment line.



Fig. 6. Nitrogen and SST concentrations along the treatment line.

Table 3

Upgraded plant effluent concentrations compared with emission limits (surface water bodies)

Parameter	Unit	Treated water (stream #18)	Italia regulationª
		(0000000000)	
BOD ₅	mg/L	0.16	≤40
COD	mg/L	2.98	≤160
NH_4^+	mg/L	1.70	≤15
N-NO ₃ -	mg/L	17	≤20
Ni	mg/L	0.01	≤2
Fe	mg/L	0.35	≤2
Pb	mg/L	0.01	≤0.2
Cr	mg/L	0.02	≤0.2
SST	mg/L	38.32	≤80

^aLegislative Decree 152/2006 - Part III, Annex 5, Table 3 in [4].

with other pollutants, is very relevant from an industrial point of view and specific experiments in the laboratory and pilot scale should be necessary in order to check the concentrations found with simulation. After that industrial tests should be necessary before to start up the full-scale industrial plant with the new improvements.

The disinfection step is used to remove microorganisms, other parasites and some organic compounds that can be dragged during treatment operations. In the simulation case, these compounds were destroyed by the oxidizing action of sodium hypochlorite. As a consequence there was a further reduction of COD and BOD₅ by 20%. Thus, COD and BOD₅ concentrations were about 3 and 0.16 mg/L, respectively, lower than regulatory limits.

Removal of metals during pre-treatment steps and final filtration was mainly carried out.

2.2.2. Sensitivity analysis

In the second phase of this study, a sensitivity analysis was carried out varying the following key parameters:

- (a) BOD₅/COD ratio of leachate: 0.03, 0.1, 0.3 and 0.5.
- (b) Initial volume of leachate: 4, 6, 8 and 10 m³.
- (c) Percentage of nitrate recycle stream: 70%, 80% and 90%.

In the simulation (a), the flow rates were equal to the basic run: 30, 35 and 100 m³/d for the lines #1, #2 and #3, respectively. In the simulation (b), the volume of leachate was varied while other parameters of the input ($BOD_{s'}$ COD...) for the three lines were kept constant and equal to those of the basic run (Table 1). In the simulation (c), it only changed the recycle percentage of the current from nitrification/oxidation with respect to the basic configuration.

Results of simulation (a) in Fig. 7 are sketched. Simulations show that if BOD_5/COD varied between 0.03 and 0.5, treated water specifics always met Italia regulation limits [4]. In terms of BOD_5/COD of the treated current, an increase of BOD_5/COD at Fenton output was found, highlighting an increase of biodegradability of the mixed flow, while it reduced with denitrification/nitrification steps, sedimentation and disinfection.

If initial volume of leachate increased from 4 until 6 m³, regulatory limits were followed by the treated water. However, some problems arose when 8 m³, or 10 m³ were reached: in these cases nitrate concentration in the treated water reached 20 mg/L, higher than Legislative Decree 152/2006 [4] limits (<20 mg/L; Fig. 8).

Finally, the percentage of nitrate recycle stream was varied from 70% to 90%. It was observed that only in the case of 70% value, nitrate concentration (24 mg/L) exceeded the legal limit (Fig. 9): this behaviour confirms the strong dependence between percentage nitrate recycled and chemical reactions, which influenced final nitrate concentration in treated water stream (effluent). So, to meet legal limits, recycle percentage should be greater than 70%.

3. Conclusions

In this paper, the upgrading of a full-scale WWT plant, located in Calabritto, in Southern Italy, is proposed. The basic configuration includes the typical line of domestic wastewater treatment with pre-treatments, flocculation, primary sedimentation, pre-denitrification, oxidation/nitrification, secondary sedimentation and disinfection. The conventional line with two additional lines for treatment of leachate and digestate wastewaters was integrated. The upgrading scheme consists of liquid grinding to remove the solid particles and flocculation with FeCl, and Fenton process to remove the most of BOD_5 and COD. The output streams are mixed with the flow resulting from primary sedimentation and sent to denitrification, where nitrates are removed as elemental nitrogen during APT, Fenton and recycling of nitrate processes. Afterward liquid stream are sent to nitrification/oxidation, secondary sedimentation, disinfection and filtration, prior to discharge the treated water.

A sensitive analysis based on variation of BOD_5/COD ratio, initial volume of leachate and recycle percentage of nitrates was carried out. The effects of these variations were studied on $BOD_{5'}$ COD, SST, NO_3 , $NH_{4'}$ TKN and metal concentrations in the effluent water.

It was found that if BOD₅/COD of leachate varied between 0.03 and 0.5 the effluent water met Legislative



Fig. 7. Results of the simulation (a): (I) $BOD_{5}/COD = 0.03$; (II) $BOD_{5}/COD = 0.1$; (III) $BOD_{5}/COD = 0.3$; and (IV) $BOD_{5}/COD = 0.5$.



Fig. 8. N–NO₂ concentration as a function of leachate volume.

Decree 152/2006 limits (emission limits for surface water bodies) [4]. This behaviour was also confirmed, when initial volume of leachate increased from 4 until 6 m³. However,



Fig. 9. N–NO $_3$ concentration as a function of percentage nitrate recycle.

some problems arose when 8 m³, or 10 m³ were reached: in these cases, nitrate concentration in the treated water reached 20 mg/L, higher than Legislative Decree 152/2006 limits (<20mg/L) [4]. Moreover, when the percentage of nitrate recycle stream was 70%, nitrate concentration (24 mg/L) exceeded the legal limit.

In all other cases, the proposed upgrading of the plant allows to simultaneously treat domestic wastewaters, leachate and digestate, producing an effluent that complies with Legislative Decree 152/2006 [4].

The process analysis performed by software could be a starting point for other research activities in which the efficiency of the processes and the results should be checked by experimental laboratory, pilot and finally industrial scale tests, where systematic monitoring of the environmental parameters should be performed.

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