Water Resources and Industry 17 (2017) 43–52

Contents lists available at ScienceDirect

Water Resources and Industry

journal homepage: www.elsevier.com/locate/wri

Carbon and energy footprint analysis of tannery wastewater treatment: A Global overview

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ARTICLE INFO

Keywords:
Carbon footprint
Power demand
Energy
Tannery wastewater

ABSTRACT

In this study the carbon footprint and power demand of tannery wastewater treatment processes for the largest bovine leather producing regions were quantified and analysed. Moreover, we present a case in which we benchmarked the carbon footprint and energy demand analysis of tannery wastewater treatment to municipal wastewater treatment. We quantified the greenhouse gas direct and indirect emissions from tannery wastewater treatment facilities. Our results show that the total CO_2-equivalent emission for tannery wastewater treatment is 1.49 10^3 $t_{CO2,eq}$ d^{-1}. Moreover, the energy intensity of tannery wastewater treatment processes are evaluated at 3.9 kWh $kg^{-1}bCOD_{,removed}$, compared to 1.4 kWh $kg^{-1}bCOD_{,removed}$ of municipal wastewater treatment processes. Based on this work in the field of tannery wastewater treatment, an effort to innovate suitable treatment trains and technologies has the strong potential to reduce the carbon footprint.

1. Introduction

The leather industry is a worldwide segment of the economy, and although not amongst the largest industrial segments worldwide, it can be a dominant regional player in certain areas. Leather tanning is known to be one of the most important industries in Mediterranean countries [1]. The leather manufacturing process consists of several steps, with one of the most important activities being the tanning of the raw hides. The tanning process can be represented in three main phases: acquisition and pre-treatment of raw animal hides; treatment of the hides with a tanning agent; and drying and shining the hides before sending them to product manufacturers. The two main types of tanning are chrome tanning and vegetable tanning, with chrome tanning still constituting the vast majority of the industry.

The production of leather requires high amounts of water for livestock, as well as for all the steps in hide-to-leather processing. The processing water may be associated with high organic load in terms of COD, organic nitrogen, sulphur, chemicals, high levels of suspended solids, and heavy metals. The high complexity of the tannery wastewater matrix originates from a wide range of components such as: raw materials (hide) residues, excess dosage of reagents including a high concentration of proteins, lipids, and salts (sulphide, sulphate, and chloride), tanning agents such as natural and synthetic tannins, and also dyes and surfactants [2]. Like all wastewater treatment, tannery wastewater is associated with direct and indirect emissions of greenhouse gases [3].

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http://dx.doi.org/10.1016/j.wri.2017.03.001
Received 8 February 2017; Accepted 16 March 2017

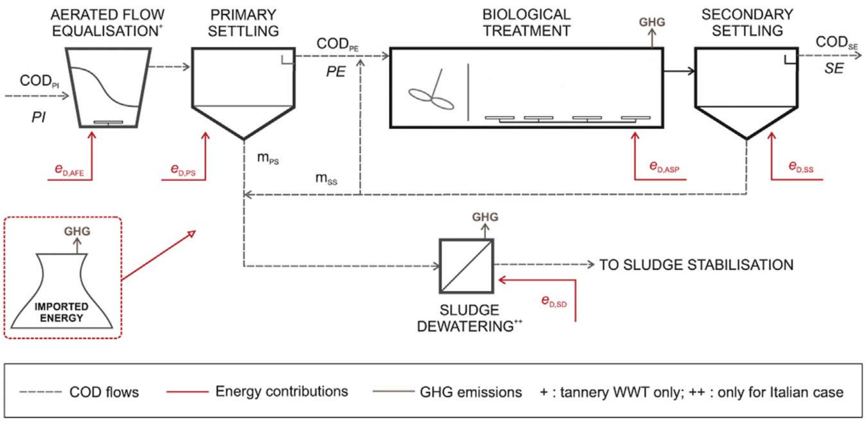

Fig. 1. Typical wastewater treatment process employed for tannery wastewater treatment.

According to the Food and Agriculture Organization (FAO) of the United Nations [4], the demand for leather for footwear, automobile upholstery, and other applications is expected to continue to expand in the medium to longer term, particularly in the faster-growing developing regions of Asia and Latin America. An expansion of the leather industry will cause more water consumption; therefore, more process water will need treatment.

The aim of this study is the evaluation of the carbon footprint (CFP) and the power demand of the most common tannery wastewater treatment process for the five largest leather producing regions (Brazil, China, India, Italy, and Russia). For the purpose of this paper, we only quantified carbon and energy fluxes concerning the main treatment operations (Fig. 1).

The process schematic shown in Fig. 1 is a common activated sludge process with the addition of the aerated equalization tank. The equalization tank must be aerated for two main purposes: mixing of the flow rich of suspended solids and the oxidation of the sulphide.

The goal of this research is the global estimation of the carbon and energy footprint for the tannery wastewater treatment. To put the carbon footprint of tannery wastewater treatment in perspective, a comparison between tannery and municipal wastewater treatment processes is presented for the Italian case study.

The domain of this research is confined to the direct emissions from the process (also referred to as Scope I) and indirect emissions from imported energy (also referred to as Scope II). Hence, all of the indirect emissions associated with chemicals, third party services, etc. (also referred to as Scope III) are not included so as to curb uncertainty. However, a sensitivity analysis of the indirect emission for caustic soda production in Europe was performed.

Therefore, we do not aim at performing a Life Cycle Assessment, nor do we aim at substituting our carbon footprint model to it, but we do intend to provide a quantitative tool with minimum uncertainty to compare process scenarios and their directly attributable emissions.

2. Methods

Datasets on leather production from the Un-Fao [4] were processed and wastewater flow rates were calculated with assumptions based on previous studies [1,5,6].

The main carbon and energy fluxes in the treatment process included in the calculation are reported, summarized, and defined in Fig. 1 and Table 1.

As shown in Fig. 1 and Table 1, the five components of the process energy demand considered were: aeration for equalization and biological oxidation; primary and secondary settling; sludge dewatering. The equalization tank is generally an aerated reactor for this specific industrial treatment with the main aims including the homogenization of the effluent and sulphide elimination (mostly catalytic oxidation) [5]. For that reason, the total energy requirement is characterized by the energy demand of the two aeration systems: aeration for equalization and aeration for biological oxidation.

The energy demand for the equalization tank was evaluated by the simplified approximation reported by Buljan and Kral [5]: 2 kg

Table 1
Summary of energy- and carbon- fluxes included in our calculations.

Flux	Definition
Energy Demand	Aeration For Equalization^{+} ($ED_{,AFE}$) + Primary Settling ($ED_{,PS}$) + Activated Sludge Process ($ED_{,ASP}$) + Secondary Settling ($ED_{,SS}$) + Sludge Dewatering^{++} ($ED_{,SD}$)
Total carbon emitted	Aerobic Respiration (on site) + Energy Generation (off site) + NaOH production$^{, ++}$ (off site)

$^{+}$ is evaluated only for industrial tannery treatment. $^{++}$ is evaluated only for the Italian case study.

Table 2
Sensitivity analysis of the indirect emission for caustic soda production in Europe.

Country	$kg_{CO2,eq}$ tonnes^{-1} NaOH	Country	$kg_{CO2,eq}$ tonnes^{-1} NaOH	Country	$kg_{CO2,eq}$ tonnes^{-1} NaOH	Country	$kg_{CO2,eq}$ tonnes^{-1} NaOH
Austria	3196	Denmark	4136	Italy	3966	Romania	5371
Belgium	5075	France	2536	Latvia	3704	Spain	3881
Bulgaria	4991	Germany	4328	Netherlands	6458	Ukraine	5715
Croatia	3401	Greece	4901	Poland	6711	United Kingdom	4575
Czech Republic	4686	Hungary	4260	Portugal	3280		

of O_2 is needed to oxidize 1 kg S^{2-} to sulphate, whereas the oxygen transfer efficiency is about 1.5 kgO_2 kWh^{-1}. The energy consumption for the equalization tank was estimated at 6 kWh per kgS^{2-}. On the contrary, no oxidation phase is required for a municipal wastewater treatment plant.

The CFP has been evaluated using an evolution (as in [7]) of the rational procedure by Monteith et al. [8], including on-site and off-site emission for power generation.

The CO_2-equivalent emitted for power generation was calculated for each country taking into account the carbon emission intensity for power generation (expressed in $kg_{CO2,eq}$ kWh^{-1}) depending on the source of the energy generation reported by the United States Energy Protection Administration [9]. Off-site emissions for the manufacturing of chemicals and other materials, as well as transportation or other contributions (i.e., Scope III emission), were not calculated as they are outside the scope of this research. We made no distinction between the quantified carbon emission and those for reporting, where applicable.

An additional element was calculated for the Italian case study: the CFP and power demand for sludge dewatering was calculated because it is commonly used in the country.

Moreover, the emission due to NaOH production (off-site, also referred to as Scope III) should be evaluated in the Italian tannery wastewater processes, because NaOH is commonly used in this region for chemical scrubbers to treat the gas stream, which is rich in H_2S to reach the standard quality for the gaseous effluent.

However, even though the chemical market for selected chemicals may be dominated by a single supplier, production may be spread over the world. In the case of NaOH, its production occurs in 52 countries and there is no specification on the origin of the product during distribution. Table 2 reports the sensitivity analysis of the emissions due to energy consumption for caustic soda production in Europe, considering an energy consumption for the production of 3.3 10^3 kWh per electrochemical unit ECU [10], being the ECU a combination of 1 t of chlorine, 1.1 t of caustic soda, and 0.03 t of hydrogen.

As shown in Table 2, in Europe the range of indirect emission for caustic soda produced is 2.8 $10^3 - 7.4$ 10^3 $kg_{CO2,eq}$ tonnes^{-1} of NaOH. For this reason it is difficult to evaluate the Scope III emissions because of the high range of $CO_{2,eq}$ emitted for all chemicals employed which adds to the uncertainty of the emissions due to the transportation of the product as a function of the distance.

Data sets on leather production were gathered from Un-Fao [4] repositories and reported in Table 3.

As shown in Table 3, all over the world the bovine leather production is the most important compared to other sheep and goat leather production, 5.62 10^5 t y^{-1} compared to 496 t y^{-1}, respectively. Fig. 2 shows the bovine leather production trend from 1993 to 2011 throughout the world.

The production in Latin America and East Asia experienced an increasing trend, while a mild decrease was recorded in the other countries, with this decrease possibly associated with the global financial crisis of 2008. However, the leather production worldwide shows a positive trend.

In order to estimate the wastewater to be processed for a tannery, we performed a water intensity analysis. The water is related to the finished product because in this paper all steps in the tanning process are considered, from hide to leather stock. According to the

Table 3
Global leather productions, 2011. Un-Fao 2013 [4].

Country	Bovine leather production (t y^{-1})	Sheep and Goat leather production (t y^{-1})
Latin American and Caribbean	110 10^3	16
Africa	5 10^3	49
Near East	22 10^3	98
Far East	285 10^3	225
North America	21 10^3	6
Europe	71 10^3	73
Rest of Europe	0.4 10^3	1
Area Former USSR	38 10^3	22
Oceania Developed	3 10^3	6
Developed Other	6 10^3	0.3
World	562 10^3	496

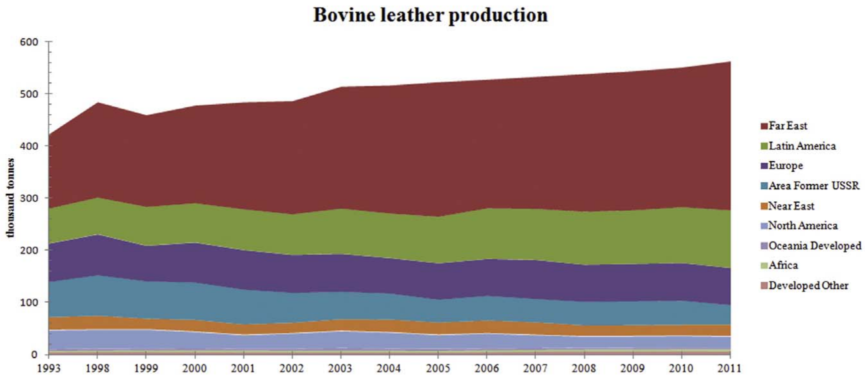

Fig. 2. Bovine leather production trend from hide to leather stock, 1993–2011.

mass balance in leather processing presented in Unido 2012 [11], from 1 kg of raw material was obtained 0.3–0.4 kg of finished product, with water consumption generally reported in a range of 25–45 l of water per kilogram of raw material [5,12,6,1]. Moreover, U.N.I.D.O. 2000 [11] reports a water intensity of 0.13 $m^3\ m^{-2}$ of finished product. Since the specific weight of bovine leather is approximately 1 $kg\ m^{-2}$, the water intensity per finished product is evaluated around 130 $l\ kg^{-1}$.

To compare tannery and municipal water intensity and wastewater production, we used the case region of Italy. The average of water consumption for municipal use in Italy is estimated at 64 m^3 per capita [13].

Due to variations in raw material, process, chemicals, etc., wastewater characterization can be different from district to district. All assumed parameters and wastewater characteristics are summarized in Tables 4, 5.

The wastewater characterization reported in Table 4 and the COD fractions reported in Table 5 were used to calculate the carbon emission and the energy demand of the biological treatment. The choice of COD fractions by Munz et al. [14] was dictated by the application to the Italian region.

3. Results and discussion

Fig. 3 shows the global bovine leather production from rawhide to leather stock in tonnes per annum.

For the 5 largest leather producing regions (alphabetically: Brazil, PR China, India, Italy, and the Russian Federation), the water consumption exceeds 5 $10^7\ m^3\ y^{-1}$. The highest water footprint related to the highest production is due to China with 2.7 $10^7\ m^3\ y^{-1}$.

Fig. 4 shows the bovine leather production per capita for the 10 largest leather producers (kilograms per person).

Fig. 4 confirms the intensity of the Italian leather industry. Moreover, if we consider that the Italian leather industry is located in 4 main industrial poles [1], the real concentration of the production per capita can be considered at 2.12 kg leather per person and 659 kg leather per km^2.

In Italy the tannery industry utilized 6.7 $10^4\ m^3$ of water per year. This country counts a population of approximately 60 million people and the municipal wastewater utilization was around 3.8 $10^9\ m^3$ in 2011. Compared to other important industries, such as the winery industry annually producing more than 10 $10^6\ m^3$ [22] and the textile industry annually producing 6.72 $10^6\ m^3$ [23], the tannery process can be considered one of the least water intensive in the country.

Fig. 5 shows the energy demand of the tannery wastewater treatment processes of the 5 largest producers.

As shown in Fig. 5, the settling process requires less energy compared to the aeration process. Moreover, the activated sludge

Table 4
Typical process characteristics assumed in the model. Industrial wastewater characterization UNIDO, 2011 [5]; Municipal wastewater characterization Metcalf & Eddy [15].

	Parameter	Unit	Industrial wastewater	Municipal wastewater
Water Quality	$[BOD_5]_{PI}$	$mg\ O_2\ l^{-1}$	2000	120
	$[BOD_5]_{SE}$	$mg\ O_2\ l^{-1}$	25	
	$[COD]_{PI}$	$mg\ O_2\ l^{-1}$	4000	300
	$[COD]_{SE}$	$mg\ O_2\ l^{-1}$	125	
	$[\text{Suspended Solids}]_{PI}$	$mg\ l^{-1}$	2000	169
	$[\text{Total nitrogen (TKN)}]_{PI}$	$mg\ N\ l^{-1}$	650	50
	$[S^{2-}]_{PI}$	$mg\ S\ l^{-1}$	160	–
	$[SO_4{}^{2-}]_{PI}$	$mg\ SO_4\ l^{-1}$	1400	–
Process characteristics	Mean cell retention time (MCRT)	d	20	20
	Wastewater temperature	°C	20	20
	Process-water oxygen transfer efficiency per unit depth (αSOTE/Z)	$\%\ m^{-1}$	1.58	3.17

Table 5
COD fraction of Tannery wastewater, all value refers to % of total COD.

	Reference	S_S	S_I	X_S	X_I
Industrial Tannery Wastewater	[16]	44.4	5.8	38.8	11
	[17]	47.5	9.5		11.5
	[18]	59.9	24.2	15.8	
	[19]	27.2	12.7	7.7	52.2
	[2]	42	20	27	11
	[20]	35	8	46	11
Municipal Wastewater	[21]	22	5	50	24

Soluble biodegradable COD (S_S); Soluble non-biodegradable COD (S_I); Particulate biodegradable COD (X_S); Particulate non-biodegradable COD (X_I).

processes aeration tank requires more energy compared to equalization, and in fact it required almost 3 times more energy. The highest total power demand related to the highest producer (PR China) was estimated at 15 MW. The energy intensity of tannery wastewater treatment was evaluated at 3.6 kWh kg^{-1} bCOD removed (4.9 kWh m^{-3}).

Fig. 6 shows the comparison of the power demand between the tannery industry and municipal wastewater, referring to the Italian case study.

In Italy the power demand of the municipal wastewater treatment was evaluated totally at 80 MW compared to 4 MW of the tannery industry; moreover, the energy intensity is 1.4 kWh kg^{-1} $bCOD_{removed}$ and 3.9 kWh kg^{-1} $bCOD_{removed}$ for municipal and tannery wastewater treatment processes, respectively.

In both cases, the energy demand is largely dominated by the aeration process [24,25] at 1.0 and 3.7 kWh kg^{-1} $bCOD_{removed}$ for municipal and tannery wastewater treatment, respectively.

Fig. 7 shows the CFP for tannery wastewater treatment for the 5 largest producers.

The off-site emission due to energy generations dominates the carbon footprint analysis. The CFP of the tannery wastewater treatment amounts to 3.15 $kg_{CO2,eq}$ $kg^{-1}bCOD_{,removed}$ for the on-site emission, while the off-site emission is in the range 9.77–21.4 $kgCO_{2,eq}$ $kg^{-1}bCOD_{,removed}$. The country-specific carbon emission intensity for power generation is responsible for the wide range of off-site emission. As a point of reference, municipal wastewater treatment has a total CFP of approximately 2 $kg_{CO2,eq}$ $kg^{-1}bCOD_{,removed}$ [7]. However, this does not necessarily reflect the high emission of the tannery wastewater process since municipal wastewater is much more diluted, hence its emission per unit volume may be a more level comparison.

Fig. 8 shows the comparison of the carbon footprint between the tannery industry and municipal wastewater, referring to the Italian case study.

The on-site emission of tannery and municipal WWT are almost the same in terms of kg of $bCOD_{removed}$. On the contrary, tannery wastewater treatment requires more energy and for this reason the off-site emission is almost 4 times higher compared to the municipal wastewater treatment. Our results show that in Italy, the carbon footprint of tannery wastewater treatment is 11.17 kg CO_2,eq m^{-3} which is in the same range of the winery industry 9.86–17.1 kg CO_2,eq m^{-3} [22].

The emission due to leather production is evaluated at 73 kg CO_2,eq m^{-2} [26], hence the industrial process emission is 103 10^9 kg CO_2,eq y^{-1}. Moreover, the on-site emission due to tannery wastewater treatment is evaluated globally in this study at 8.15 10^8 kg CO_2,eq y^{-1}. Hence, it is possible to conclude that for global leather production from hide to leather stock, the industrial process and the emission due to wastewater treatment (on-site) is approximately 1.0 10^{11} kg CO_2,eq every year.

An effort was made during these years to reduce the impact of the tannery processes, and according to the report by U.N.I.D.O. (2000) [11], the challenge over the last decade was to reduce the waste of resources as only 53% of corium collagen and 15% of the chemicals purchased are retained in the finished leather.

To reduce emissions for tannery wastewater treatment process, future investigations must be focused on the application of innovative technologies for the wastewater treatment, such as: biological scrubbers to control sulphur emissions; anaerobic sludge digestion to reduce biosolids disposal and reuse energy from biogas; anaerobic digestion of wastewater coupled with autotrophic denitrification to reduce energy consumption while maintaining nitrogen removal. Moreover, future investigations must consider the application of combined solid and liquid waste treatment from the same industrial process, such as the co-digestion of waste sludge and solid waste from tanneries [27–30].

4. Summary and conclusions

The CFP and the power demand of a common tannery wastewater treatment process were calculated for the 5 largest leather-producing regions (Brazil, China, India, Italy, and Russia). Results of the analysis were compared to the impact of the municipal wastewater treatment in Italy, in terms of CFP and power demand.

The carbon emission of the tannery wastewater treatment processes is evaluated at 5.75 10^4 $t_{CO2,eq}$ y^{-1} and the energy demand at approximately 3.5 10^7 kW y^{-1}. In Italy, the leather tanning industry generates 5.8 10^4 $t_{CO2,eq}$ y^{-1} compared to 2.0 10^6 $t_{CO2,eq}$ y^{-1} of the municipal wastewater treatment.

Due to the high concentration of COD, nitrogen, and recalcitrant compounds to be removed from tannery wastewater, the research must be focused on processes with high COD removal and low emissions, such as anaerobic digestion and sulphate reduction in combination with anammox and/or denitrification with sulphide as electron donor; the implementation of biological processes for

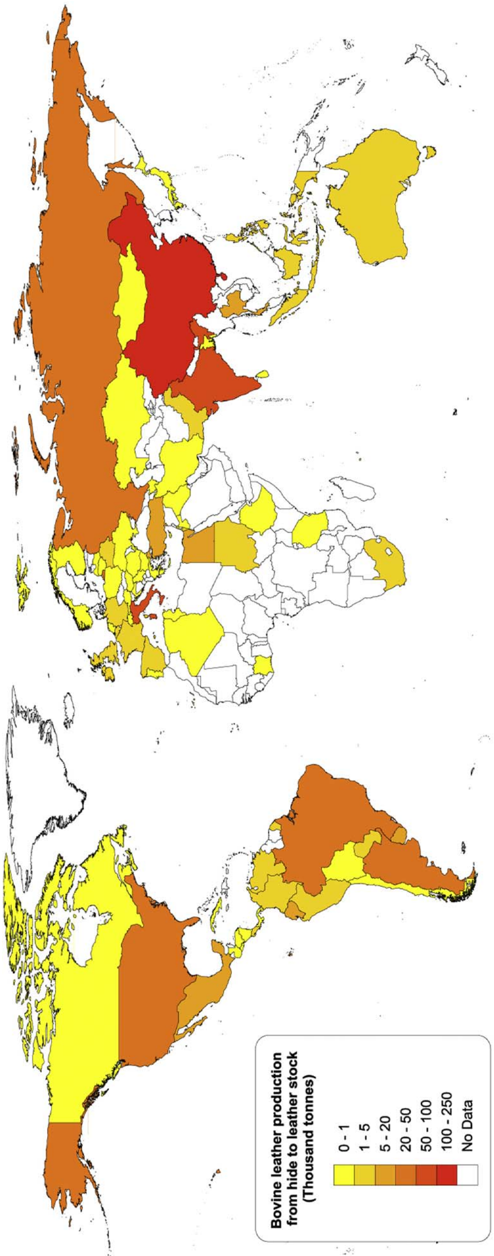

Fig. 3. Global bovine leather production (from hide to leather stock), thousand tonnes per year.

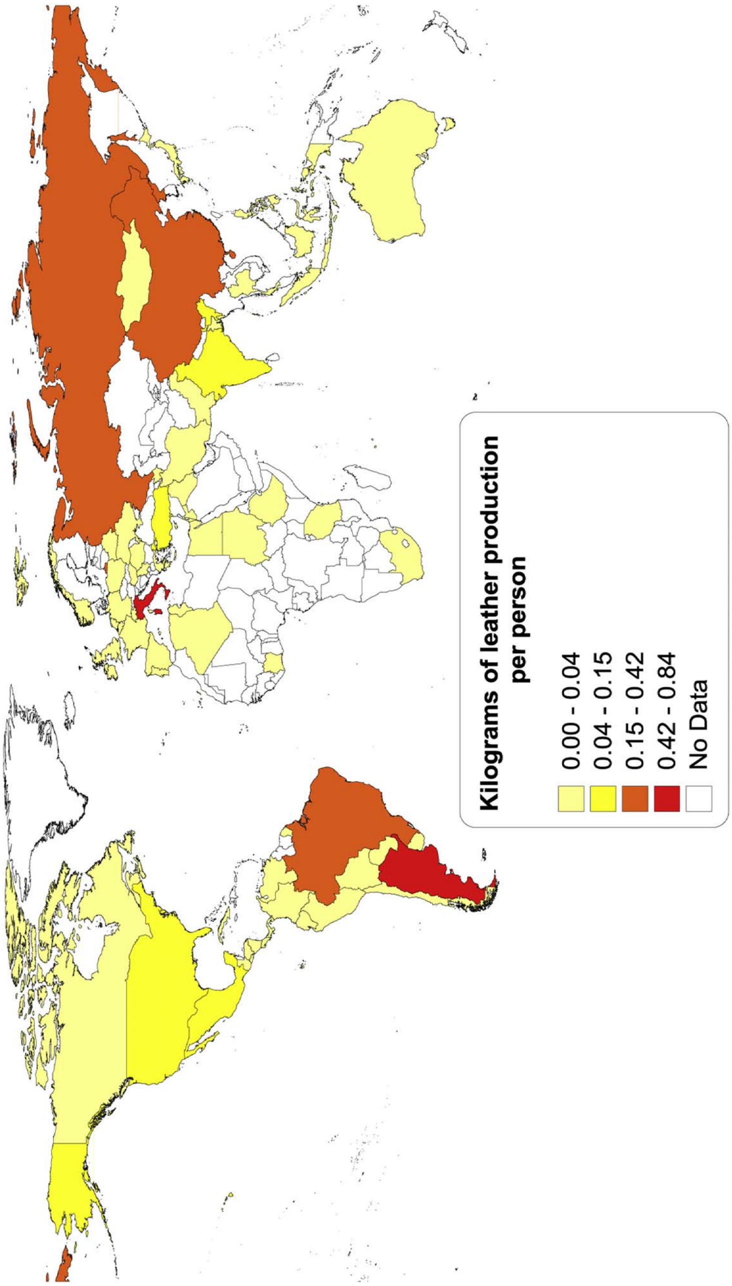

Fig. 4. Bovine leather production per capita for the 10 largest leather producers.

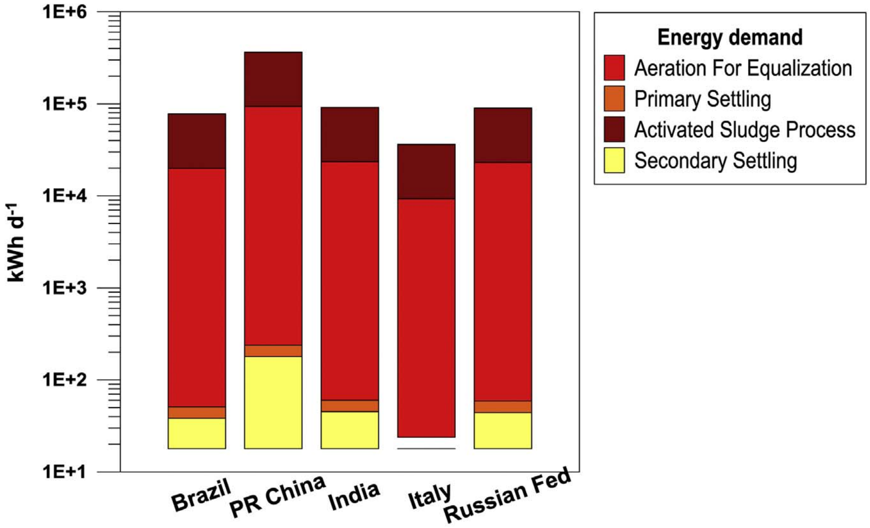

Fig. 5. Power demand of the tannery wastewater treatment of the most producers.

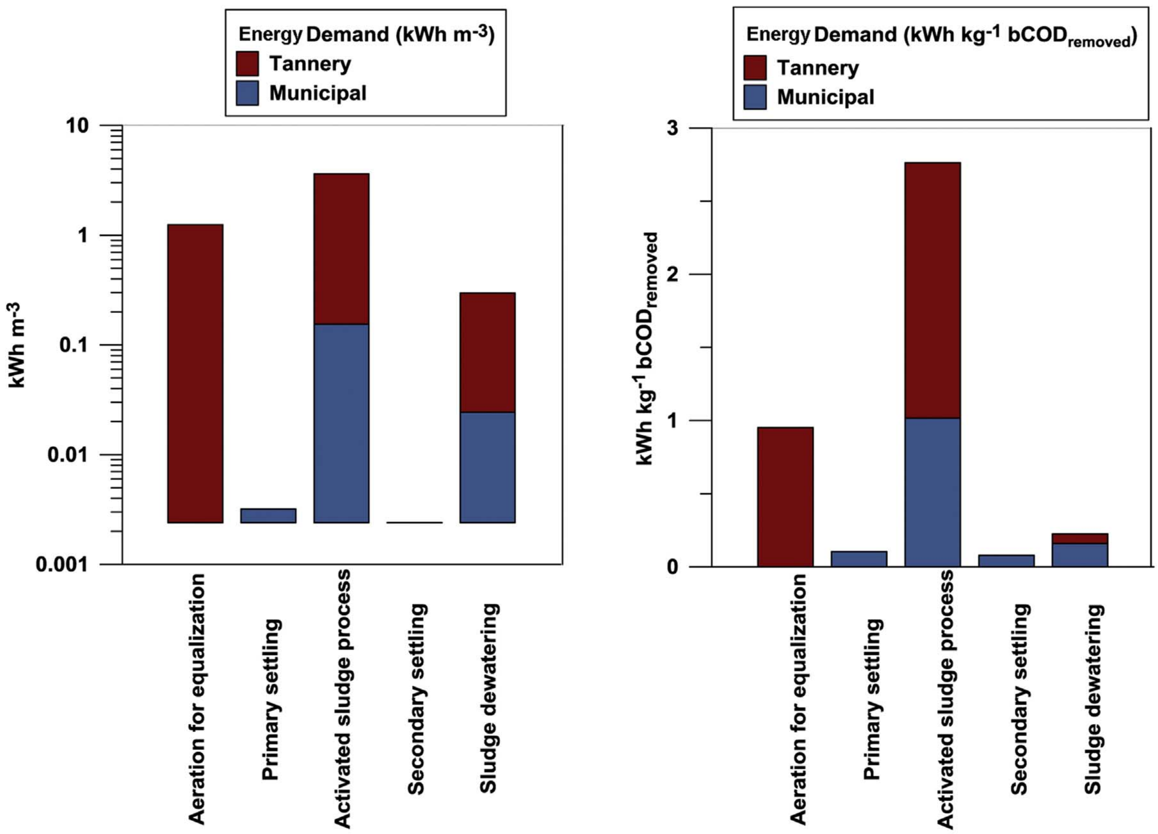

Fig. 6. Comparison of the energy demand of the tannery and municipal wastewater treatment in Italy.

recalcitrant compounds with a low consumption of chemicals such as fungal biomass is also a solution that merits further investigation. Even though the application of these processes would require a complex treatment train, the applicability of this option is favoured by the high temperature of the wastewater (usually above 20 °C) and by the warm weather of most of the countries where the industry is growing; the trend of concentrating tannery industries and wastewater treatment into specialized districts will also favour the application of the above mentioned processes.

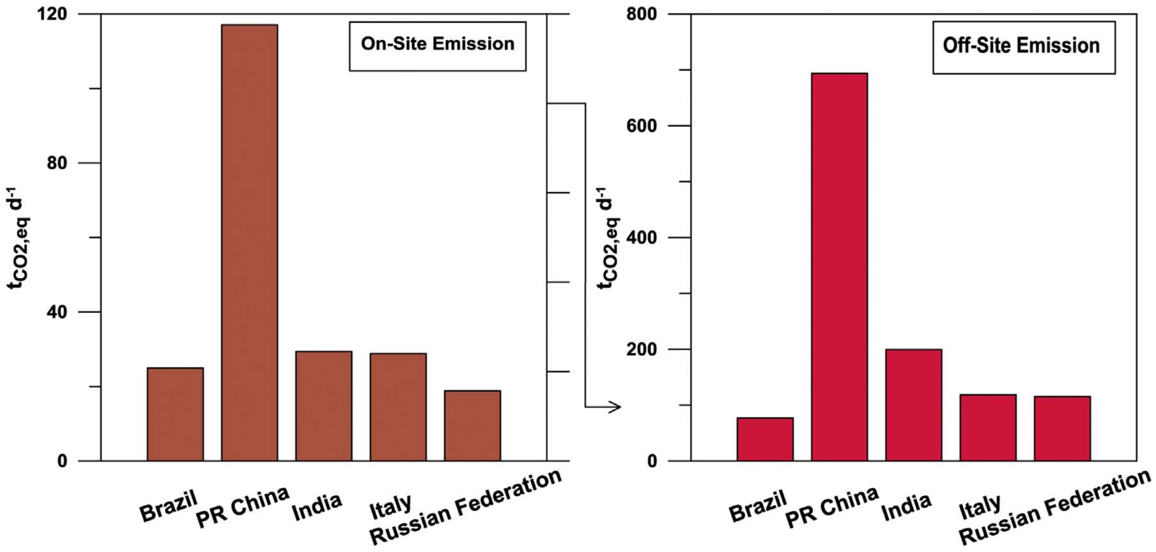

Fig. 7. CFP of the tannery wastewater treatment of the most producers.

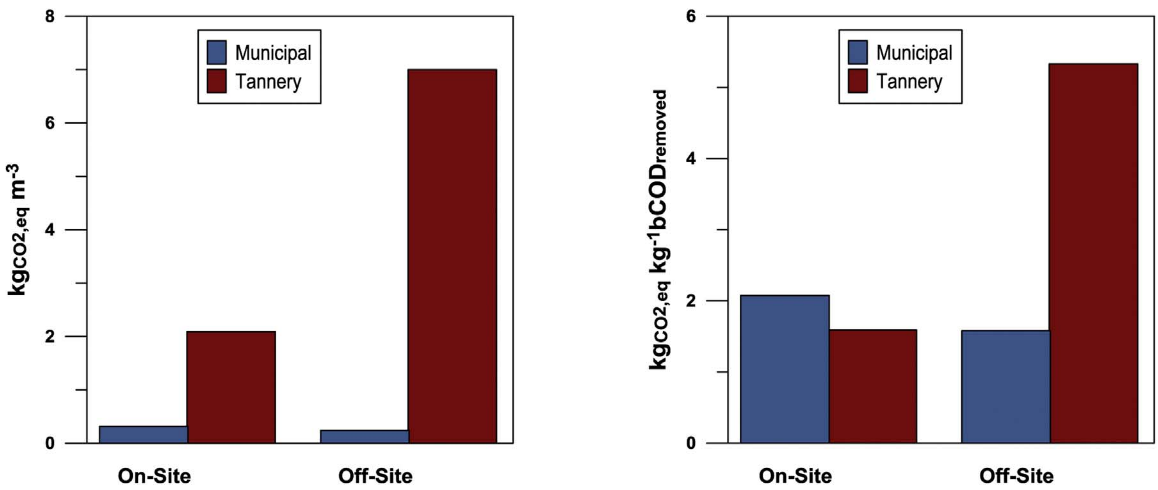

Fig. 8. Comparison of the CFP of the tannery and municipal wastewater treatment in Italy.

Acknowledgements

The authors acknowledge the European Union for supporting this work through the CARBALA Marie Curie Irses program (CARBALA project-295176).

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