

## Article

# Sewage Sludge Quality and Management for Circular Economy Opportunities in Lombardy

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## Featured Application: Sewage Sludge management plan.

**Abstract:** From the perspective of a circular economy that prioritizes resource reuse and recovery, sewage sludge could be a source of nutrients for agricultural soils or a source of energy, depending on its characteristics. Lombardy is the region with the highest quantity of sludge production and management in Italy. A methodology was developed to: extract and analyze quantitative data on sewage sludge (EWC 190805) production and management (2017–2018); collect and analyze qualitative data from publicly available documents in tender processes for sewage sludge management (2014–2020). Sludge from Lombardy’s wastewater treatment facilities displayed average qualities that were useful for recovery in agriculture after additional stabilization treatments. Sludge showed generally low heating values and elevated water content and should require additional treatments to be used in mono-combustion. The study discovered that there is still work to be done in sludge recovery in agriculture in Lombardy, taking biosolid quality into account. Sludge, on the other hand, can be converted into energy. The methodology for collecting and analyzing site-specific data presented here can be applied to other areas. The findings can assist and guide decision-makers in developing future regional sewage sludge management strategies.

**Keywords:** biosolids; sewage sludge characteristics; management; recovery; agriculture; combustion; circular economy



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## 1. Introduction

Sewage sludge is a typical by-product of our society. It can be originated from civil or industrial activities. Sewage sludge contains valuable organic matter and can be a source of nutrients to be recovered for soil fertilization and remediation [1]. It also contains pathogens, organic and inorganic pollutants and potentially toxic compounds, such as polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB), dioxins, nonylphenols, and other trace pollutants, such as (phyto)pharmaceuticals, personal care products, and microplastics [2–4], currently so-called “emerging contaminants” [5]. Pollutants can be persistent in the environment, accumulating and biomagnifying [6], and may pose hazards to human health and the ecosystems, such as carcinogenicity, teratogenicity, reproductive toxicity and genotoxicity [3,5]. Moreover, substances can form intermediate products or mixtures resulting in synergistic/additive/antagonistic effects [3]. Consequently, sewage sludge treatment activities focus on improving its quality for recovery opportunities, whilst minimizing the quantities to be treated and disposed of. In line with circular economy principles, the current plan calls for wastewater treatment plants (WWTP) to be turned into water resource recovery facilities [7].

With a view to waste management priorities, which prioritize material recovery, sludge reuse in agriculture potentially represents an optimum solution, due to its valuable

properties: organic carbon content, nitrogen, phosphorus, micronutrients (in particular metals such as nickel and copper) [8–10]. Nitrogen can be easily available from adequately treated biosolids, decreasing the need for chemical fertilizers [11]. Phosphorus has been brought to attention due to the progressive depletion of mines [12] and is included in the list of critical raw materials [13]. Phosphorous and nitrogen could be recovered from the sludge line of WWTPs by precipitation/crystallization; phosphorous is also extracted from sewage sludge and residual ash of incineration by wet chemical recovery technologies [14]. Recovery of micronutrients has become more relevant these days, with fluctuation in micronutrient deficiency levels in soils being a global phenomenon [15]. Treated sludge is indicated to improve soil properties, such as texture and water-holding capacity, favoring root growth and drought resistance [16–18]. Moreover, emerging technologies are aimed at recovering volatile fatty acids and polyhydroxyalkanoates from secondary wastewater sludge [19].

Energy recovery from sludge is becoming more and more interesting due to the need for substitute energy sources and for technology development [20], and a viable alternative pathway for sludge recovery. Biogas from anaerobic digestion can be a supply for heating and electricity generation, and it can be upgraded to biomethane for injection into natural gas networks or as transport fuel [21,22]. Sewage sludge can be dried and converted into energy in cogeneration plants, thermal power plants, waste-to-energy plants, cement kilns, and mono-incineration plants by incineration, pyrolysis, or gasification [23,24].

Besides thermal processes, which significantly reduce sludge, technologies for sludge minimization are employed in the water and sludge lines. Chemical processes include chemical oxidation (ozonation and wet oxidation); among biological processes, there are membrane biological reactors (meso- and thermophilic), and biological predation and Granular Sludge Systems [25]. Electro-osmosis is growing in interest among pressurized electro-dewatering treatments [25].

Currently, the law still in force at the European level is the Council Directive 86/278/EEC [26]: it is aimed at regulating the reuse of sludge in agriculture, increasing its recovery while protecting the environment and health, by ensuring that heavy metals in soil and sludge do not exceed set limits. The Directive is currently obsolete and does not consider the most recent technologies and alternative sludge recovery opportunities. The review of the Directive was started by the European Union (EU) in 1999, but a revision of the law is still absent. Since then, beyond scientific progress and technological developments, the policy framework has changed following the Circular economy action plans, the Bioeconomy Strategy, the new Fertilizing Products Regulation, the Farm to Fork Strategy, and the EU Biodiversity Strategy for 2030 [27]. Besides the regulatory framework, the “ideal” strategy for the reuse/disposal of sludge has not been identified yet, though numerous efforts are ongoing additionally through international projects and research [28].

The produced amount of sewage sludge in Europe (EU27) can be estimated as approximately 10 million tonnes, dry solids [29]. In 2021, approximately 48% was spread on land for agricultural use, and incineration stood at about 27% [30]. In 2016, the EU produced 16.1 Mtoe (Million tonnes of oil equivalent) of biogas, with sewage sludge feedstock contributing with 1.4 Mtoe of biogas (8.7%) [21]. Significant differences between the Member States in sewage sludge management are reported: while the reuse of nutrients in agriculture (land spreading or composting) is the most common practice in Spain, Ireland, Finland, Hungary, and Cyprus, incineration is mainly applied in the Netherlands, Belgium, Germany, and Austria [31]. The main aspects influencing the choice for sludge recovery/disposal are population density and the availability of agricultural lands. Moreover, the low level of acceptance by farmers and the public could represent another significant restriction to biosolid land spreading [32].

In Italy, the production of sewage sludge (European Waste Code EWC 190805) was 3.4 million tonnes in 2020, out of which about 44% was recovered, with reuse in agriculture being one of the possible options [33]. Agricultural reuse is the most prevalent route in the regions of northern Italy, with Lombardy leading this trend [32,33]. In Lombardy, the

production of sewage sludge was around 468,800 t in 2020 (13.8% of Italian production) [33]. In Italy, the European regulation 86/278/EEC has been implemented with the Legislative Decree 99/1992 [34], then integrated in 2018 (Law n. 130, article 41 [35]), which deposes the regions to establish further limits and conditions for the reuse of biosolids in agriculture in relation to the characteristics of the soils, crops, and sludge. At the regional level, Lombardy implemented D.G.R. X/2031/2014 [36], integrated in September 2017 by the D.G.R. X/7076/2017 [37] and by Executive Decree D.D. 6665/2019 [38]. Regional regulations introduced requirements for the acceptability of sludge entering the sludge treatment plants (STPs—authorized for the treatment on behalf of third parties); provided two quality classes for biosolids reused in agriculture (“high-quality” and “suitable for spreading”) with different limit values; and defined the characteristics of agricultural soils receiving the biosolids and spreading methods [32], and set limit values for additional parameters [38].

In 2016, the production of biogas in Italy was about 8259 GWh (3.8% in Lombardy), of which about 129 GWh was produced from sludge [39]. The production of biomethane is recently gaining more attention and the first plants have been realized and tested in the region in the last year, in parallel with the planning and design of full-scale plants.

In short, possibilities of nutrients or energy recovery of sewage sludge are potentially virtuous solutions and promising perspectives for the circular economy, even though regulations and costs have an impact favoring or preventing this process. While there are no ready-made solutions for sewage sludge management, possible options need to be assessed case by case. Sludge quantity, quality, and management are among the necessary information that can support decision-making for practitioners at the local level and at regulatory and research levels. This requires the availability and analysis of raw, disaggregated, site-specific data, which is seldom easily available [40]. This paper focuses on sewage sludge production and management in the Lombardy region in Italy. The paper presents a method to collect publicly available data on sludge quality, and the analysis of their characteristics against regulatory limits and opportunities for nutrients and energy recovery. The goal of this work was to understand sewage sludge management options in Lombardy based on their characteristics to support decision-making processes and help to orient future strategies from the perspective of the circular economy.

## 2. Materials and Methods

### 2.1. Sludge Quantities and Management

For quantitative data, we acquired from the Regional Environmental Protection Agency (ARPA Lombardia) the databases of the single environmental declaration model (MUD) of the years 2017–2018 and data on the amount of sludge produced in Lombardy for the years 2002–2018. The MUD is the single environmental declaration model by which, annually, waste that is produced, collected, disposed of, recovered, transported or brokered must be declared in Italy. MUD databases contain data on waste quantities produced, transported and received by companies and undergoing intermediate or final treatments—recovery (R) and disposal (D) operations (Table S1). In collaboration with ARPA Lombardia, we identified and discussed sources of potential uncertainty in the elaboration of data on sludge (EWC 190805) from MUD databases. MUD databases, while representing an effective punctual control tool, present challenges when used for extracting and processing data to estimate waste flows and mass balances, as double counting of the same waste can occur. As an example, sludge produced by a company, undergoing an intermediate treatment and changing its physical state, could be recorded as a newly produced waste on the MUD form of another company, even if it is, de facto, the same waste.

We generated mass balances for the EWC 190805 for the years 2017 and 2018, presented as Sankey diagrams [41]. A code was written in the R environment [42] to extract data from MUD databases, including the computation of the following waste flows: waste produced in Lombardy; inputs from outside the region; waste undergoing recovery or disposal operations; outputs passing the regional boundaries [43]. We applied the Network

Analysis [44] and established criteria to identify and eliminate data about the intermediate delivery of waste, in order to estimate the total quantities of sludge produced, in input and output, and undergoing final operations. Criteria of clean data, avoiding double-counting, were: eliminate data on sludge produced by companies treating liquid sludge by intermediate D8 and D9 operations, when a new production was reported; eliminate the amount of sludge recorded under intermediate temporary storage operations D15 and R13. After cleaning data, the total production of sludge EWC 190805 in the region (Production) was calculated as:

$$\text{Production} = \text{Outputs} - \text{Inputs}$$

where: Outputs was the total amount of sludge undergoing recovery and disposal final operations in Lombardy and of sludge transported outside the regional boundaries; Inputs was the total amount of sludge received from companies outside Lombardy.

The waste flows by origin and destination were visualized by Sankey diagrams generated using the Sankey software package in R (R Foundation for Statistical Computing, Vienna, Austria) [45].

## 2.2. Sludge Quality

A desk review of publicly available tender processes for the management (collection, transport, and recovery/disposal) of sewage sludge (EWC 190805) treated by wastewater treatment plants (WWTPs) in Lombardy was conducted. Tender processes aim at defining the price for the activities and can include data on sewage sludge quality. First, contracting authorities and water management companies in the region were identified and listed by province. Then, tender documents were collected from the websites of identified contracting authorities and water management companies. The inclusion criteria were: the time frame (years 2014–2020), the EWC code (190805), the location (all provinces in the region of Lombardy) and the presence of sludge analysis certificates.

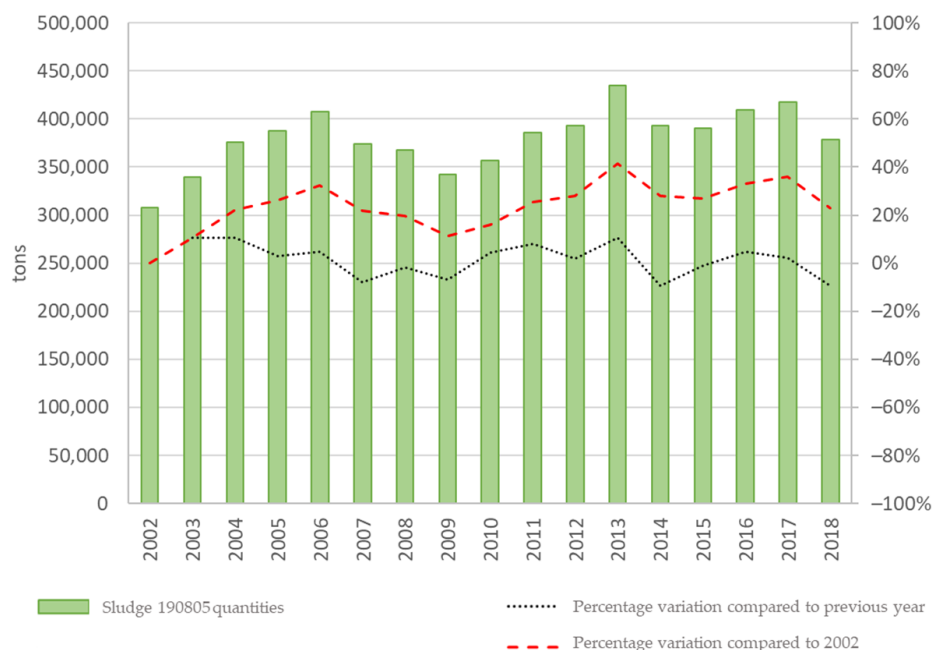
Data collected from certificates were: the concentration of heavy metals and semi-metals (hereafter jointly referred to as “heavy metals”), microbiological parameters, agronomic parameters, organic micropollutants and stabilization degree. Additional information on WWTPs, such as the population served, the capacity, the type of treatment and dewatering, were obtained from the companies’ websites or by internal communication. Data were organized, cleaned and analyzed using Microsoft Excel (Redmond, WA, USA, 2010). Data were elaborated by the calculation of average, median, minimum and maximum, 1st and 3rd quartile for each parameter. Data from analysis certificates were compared with the most recent regulations in Lombardy on sludge reuse in agriculture, D.G.R X/2031/2014 and D.D. 6665/2019, that endorse national and European regulations, whilst posing more restrictive limits. The box plot supported the visualization of the distribution of data.

Energy balance equations were used to study the optimal characteristics of sludge for mono-combustion. Five cases were considered: three cases referred to three samples from the database for which the elemental analysis was available; two cases referred to the literature data for primary and secondary sludge respectively [46]. The Dulong formula was used to calculate the Higher Heating Value (HHV), given the elemental analysis. The temperature that the sludge could reach in the combustion chamber was calculated using the energy balance equations (Section S2 in the Supplementary Material). Then, given the elemental analysis, the calculated HHV, and set temperatures to be reached in the combustion chamber equal to 800 °C and 950 °C, limit values were determined for water content (W), ash (NVS) and volatile solids (VS) by iterative calculation, keeping constant the HHV, the VS/TS ratio and varying the W. For primary and secondary sludge, the iterative calculation was conducted using the average and the minimum and the maximum values of the range indicated for the VS (expressed as % on dry matter DM). Results were reported on a triangular plot.

### 3. Results

#### 3.1. Sludge Quantities and Management

Total sludge (EWC 190805) produced in Lombardy varied from 307,359 t in 2002 to 378,230 t in 2018, with a maximum of 434,677 t in 2013, an average variation compared to the previous year of 6.1% (in terms of absolute value), and an average variation compared to 2002 of 25.2% (Figure 1).



**Figure 1.** Estimated total production of sewage sludge (EWC 190805) in Lombardy, and the percentage variation of quantities compared to the previous year and 2002.

In 2017 in Lombardy, 44.6% of managed sewage sludge was produced in the region, whilst 55.4% was imported (Figure 2). The majority of managed sludge (96%) was recovered, primarily through operations R3 (organic substance recycling/reclamation), R1 (used to generate energy), and R12 (waste exchange for submission to any of the operations numbered R1 to R11), which accounted for 45%, 11%, and 39%, respectively. Only 1.3% of managed sludge was disposed of, whilst 2.8% was transported out of the region.

In 2018 in Lombardy, 47.2% of managed sewage sludge was produced in the region, whilst 52.8% was imported (Figure 3). The majority of managed sludge (93.3%) was recovered, mainly by operations R3, R1, and R12, which accounted for 49%, 14%, and 28%, respectively. Only 2.1% of managed sludge was disposed of, whilst the remaining 4.3% was transported out of the region.

#### 3.2. Sludge Quality

A total of 46 websites were identified and analyzed, and 66 tender processes satisfied the selection criteria. A total of 258 certificates of analysis conducted from 2014 to 2020 on sludge from 102 different WWTPs, across all 12 Lombardy provinces, were identified. The total number of WWTPs in Lombardy in 2019 was 1379, of which 968 (70%) serving < 2000 equivalent inhabitants (EI) and 411 (30%) serving > 2000 EI [47]. Collected data were representative of 7.4% of the total number of WWTPs in Lombardy, and of 23% of plants serving >2000 EI. Most of considered WWTPs (41) have a capacity of 10,000 < EI < 50,000; of the remaining, 24 have a capacity of 2000 < EI < 10,000, 13 have a capacity of 50,000 < EI < 100,000, 12 have a capacity > 100,000 EI, 3 have a capacity < 2000 EI; for 9 plants there was no information.

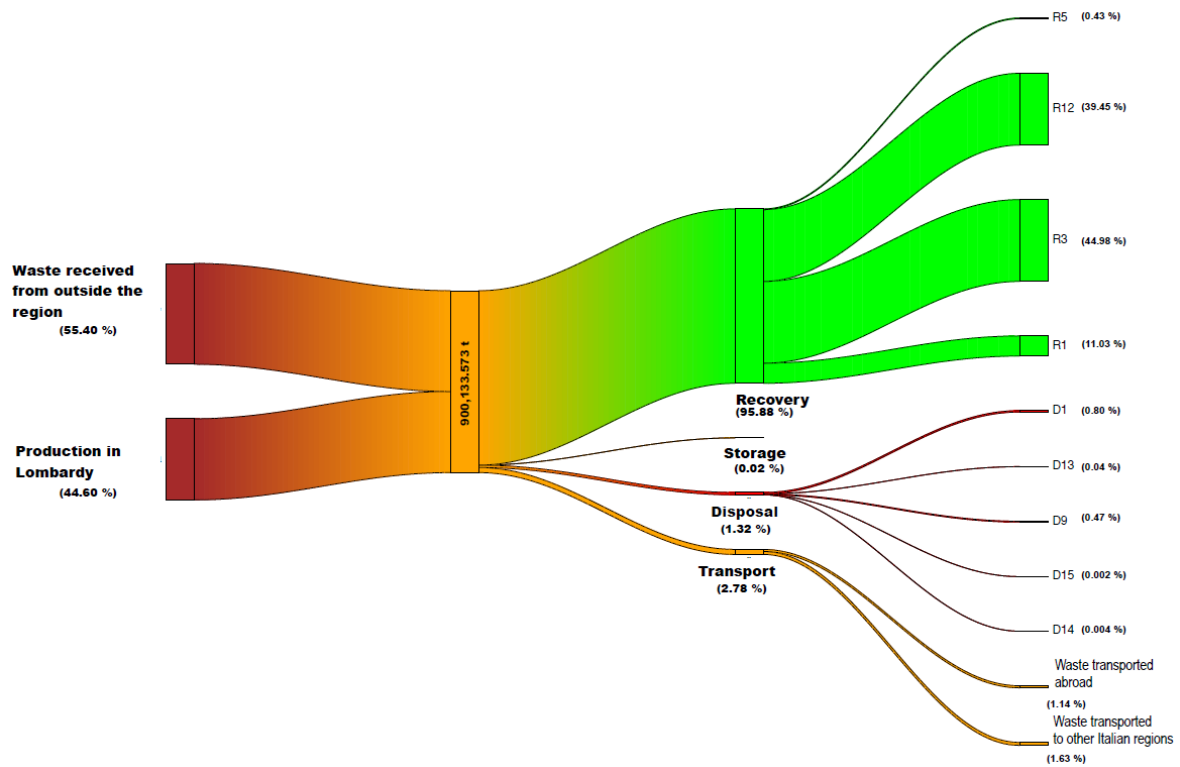


Figure 2. Mass balance of the total production and management of sludge (EWC 190805) in Lombardy for the year 2017, by origin and destination.

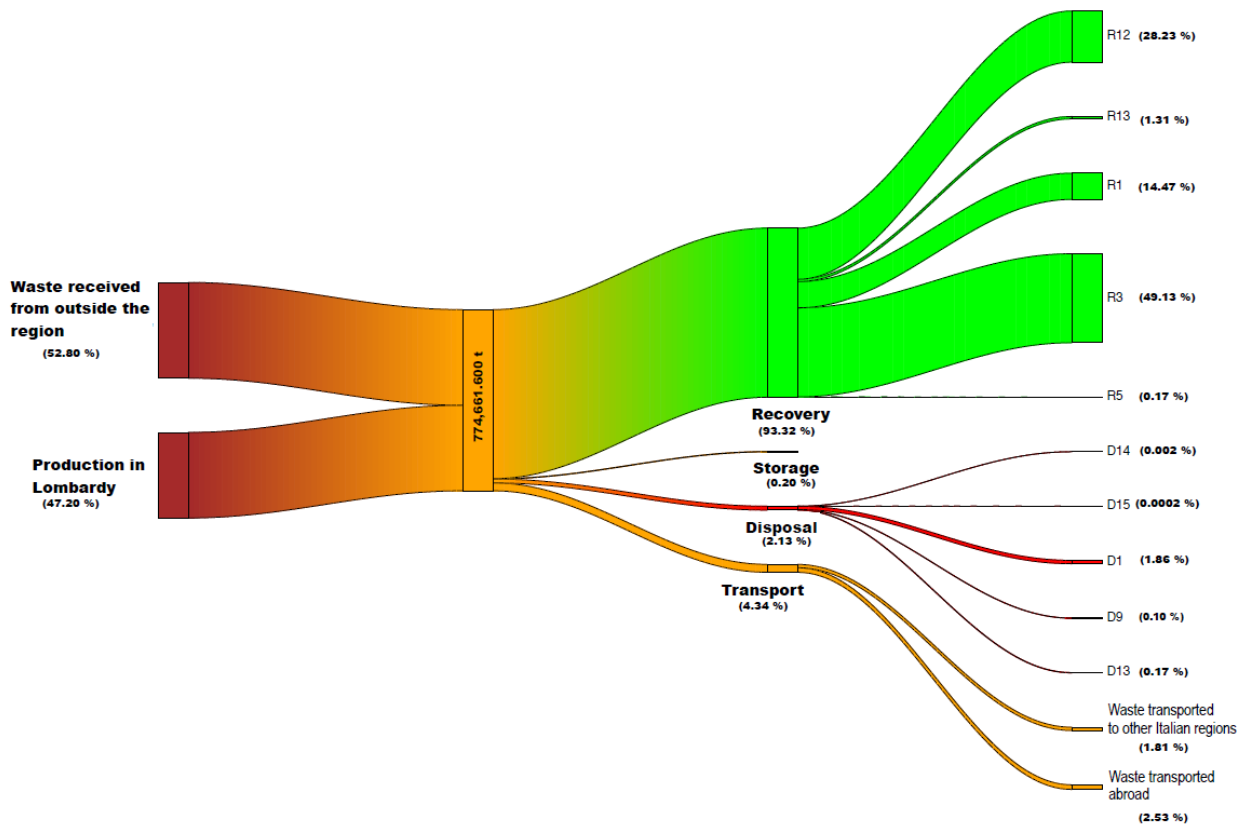


Figure 3. Mass balance of the total production and management of sludge (EWC 190805) in Lombardy for the year 2018, by origin and destination.

Data were from analyses carried out in 2014 (6%), 2015 (8%), 2016 (23%), 2017 (24%), 2018 (15%) and 2019 (21%), and 2020 (3%).

An influence on the degree of stabilization of the waste by the type of stabilization used was not observed ( $n = 78$ ). Plants with aerobic treatment ( $n = 50$ ) produced sludge with an average degree of stabilization of 69% and a median of 71%; plants with anaerobic treatment ( $n = 28$ ) produced sludge with an average degree of stabilization of 70% and a median of 72%. Comparing mechanical dewatering processes ( $n = 152$ ), the percentage of dry matter (DM) achieved by means of filter-presses is the highest (25%,  $n = 16$ ), followed by centrifuges (21%,  $n = 95$ ) and belt presses (17%,  $n = 35$ ). Detailed results are reported in Table S1 (Supplementary Materials).

Table 1 shows the results of elaborations on normed parameters of sludge produced by WWTPs. The comparison with limits set by the most recent regulations for the recovery in agriculture [36,38] helped to delineate sludge quality in relation to the possibilities for direct or indirect recovery in agriculture. The D.D. 6665/2019 defines two sets of limits for some of the parameters for “suitable” and for “high-quality” sludge to be reused in agriculture. Table S2 in the Supplementary Material presents the results of elaborations on non-normed parameters reported in the analyses.

With reference to limits for sludge “suitable” for reuse in agriculture, the average and median values of all regulated heavy metals, trace organic pollutants, agronomic parameters (except the germination index) and of the degree of stabilization complied with the normative limits for recovery in agriculture in the examined analyses and period (Table 1, Figure 4). When considering single values for each parameter, it can be observed that: (a) all analyses satisfied normative limits for lead, cadmium, mercury and chromium VI, total N and total P, PAH, nonylphenols, Bis(2-ethylhexyl)phthalate (DEHP), heavy hydrocarbons and polychlorinated dibenzo-p-dioxins, furans and biphenyls (PCDD/F) + Dioxin-like PCB; (b) non-compliance for one or more heavy metal (total chromium, copper, zinc, nickel, arsenic, beryllium, or selenium) was found in 37 analyses, with the concentrations of total chromium and arsenic being the most frequently over the limits (Figure 4); (c) among agronomic parameters, the percentage of organic carbon ( $C_{org}$ ) respected the limit value in the 96.3% of analyses, with 7 non-compliant values; the limit for the germination index conformed with in 42.7% of analyses; the pH was non-compliant in 2 analyses; (d) only 29.2% of analyses for fecal coliforms and 83% for *Salmonella* sp. complied with the regulatory limits; (e) the limit value for PCB was not respected in 1 analysis (0.5%), and the one for toluene in 4 analyses (4.2%); (f) the degree of stabilization complied with the limit in 29.5% of the analyses (Table 1).

When considering the limits for “high-quality” sludge, the heavy metals which mostly exceeded limits were zinc, arsenic, copper and nickel, while the degree of stabilization for high-quality sludge was respected in 16% of the analyses.

When comparing data with limit values in Table 5.1 of DGR X/2031/2014 [36], which regulates the quality of sludge entering the STPs, it emerged that the same analyses respecting the limit values for sludge “suitable” for recovery in agriculture did respect those for admittance in STPs for copper, zinc, lead, nickel, cadmium, mercury and total N. For total chromium, in 16 cases the value not respecting limits for recovery complies with those for being admitted in STP (97.5%). For  $C_{org}$ , all samples (100%) complied with the limit value for entering STPs (Table 1).

Across all samples for which all normed parameters for recovery in agriculture were analyzed ( $n = 31$ ), only two complied with all limit values.

According to our results, the average quantity of P in the sludge in Lombardy was 2.2% (Table 1), in line with the national average. These data are relevant for considering the possibilities of phosphorus recovery from incineration ashes as well as for recovery in agriculture (through the direct use of sludge or its use for the production of compost, fertilizers or correctives).

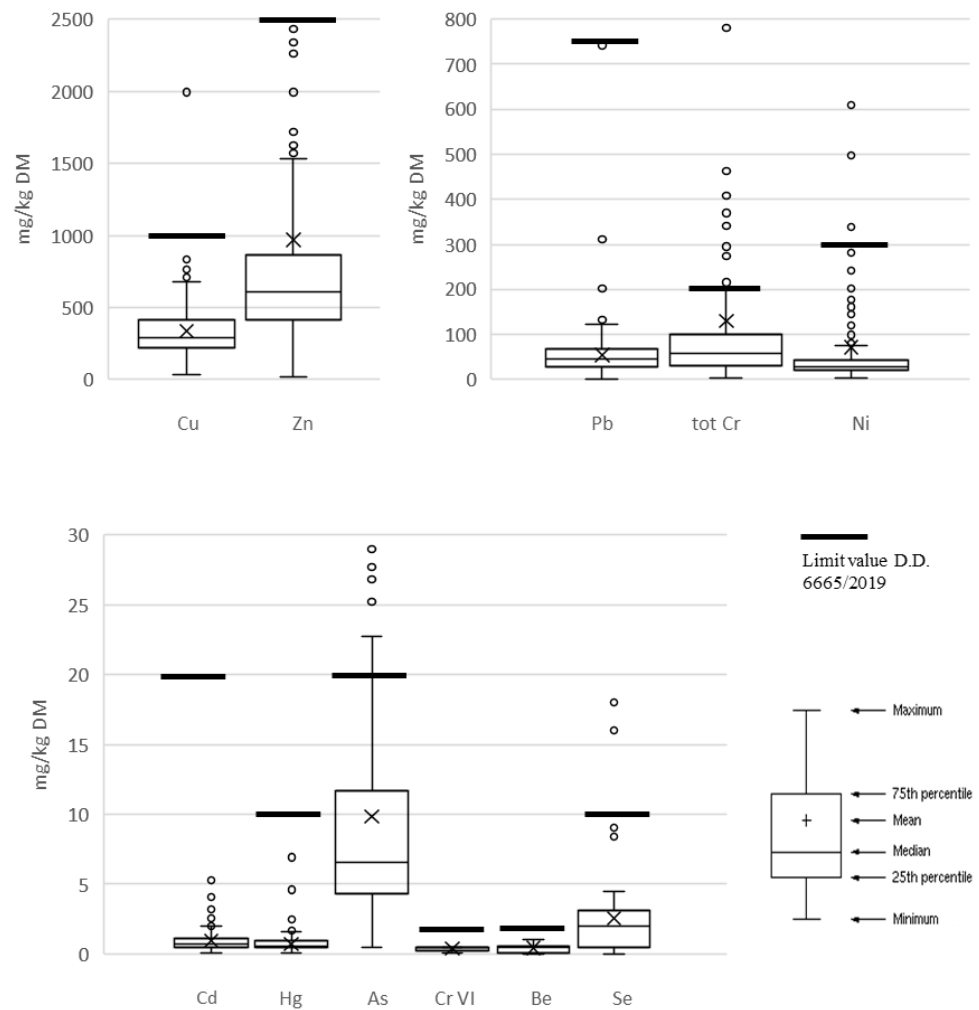
**Table 1.** Results of elaborations of data on sludge produced by WWTPs in Lombardy (2014–2020). Listed parameters are those regulated by D.D. 6665/2019 for agriculture recovery, and those normed by Table 5.1 of D.G.R. X/2031/2014, (in italic), for sludge admitted in STPs.

Parameter	n. of Data	MIN	1st Quartile	Average	Median	3rd Quartile	MAX	D.D. 6665/2019 * and D.G.R X/2031/2014 Limit Values	n. Analysis Complying with Limits	% Analysis Complying with Limits
Heavy metals (mg/kg DM)										
Copper	203	31.2	224.5	335.2	289.0	405.0	1994	≤1000 (≤400) ≤1200	202 (152) 202	99.5 (75) 99.5
Zinc	203	18.0	414.0	967.6	610.0	861.5	32,800	≤2500 (≤600) ≤3000	197 (101) 197	97 (50) 97
Lead	204	1.0	29.0	54.9	46.0	68.0	742	≤750 (≤250) ≤900	204 (202) 204	100 (99) 100
Tot chromium	203	2.5	31.3	129.9	58.9	100.0	2400	<200 (≤150) ≤900	182 (176) 198	90 (87) 97.5
Nickel	203	2.7	20.1	71.4	29.0	42.8	1690	≤300 (≤50) ≤330	197 (159) 197	97 (78) 97
Cadmium	202	0.05	0.5	1.0	0.7	1.1	5.4	≤20 (≤5) ≤22	202 (200) 202	100 (99) 100
Mercury	177	0.05	0.5	0.7	0.5	1.0	6.92	≤10 (≤5) ≤11	177 (176) 177	100 (99) 100
Arsenic	183	0.5	4.4	9.8	6.6	11.6	57.5	≤20 (≤10)	164 (127)	90 (69)
Chromium IV	96	0.1	0.3	0.4	0.5	0.5	0.5	<2	96	100
Beryllium	48	0.1	0.2	0.5	0.5	0.6	1.0	≤2	44	91.7
Selenium	55	0.0	0.6	2.5	2.0	2.8	18.0	≤10	53	96.4
Agronomic parameters (%DM)										
C <sub>org</sub>	190	14.9	29.1	33.3	34.2	38.5	45.0	>20 >10	183 190	96.3 100
Total N	180	1.9	3.9	5.0	4.8	6.0	9.8	>1.5 >1	180 180	100 100
Total P	187	0.5	1.6	2.2	2.1	2.8	4.0	>0.4	187	100
pH	246	5.8	7	7.4	7.34	7.8	12.2	5.5 < pH ≤ 11	244	99.2
Germination index	96	2	19.9	50.0	49.9	80	99.6	>60	41	42.7
Microbiological parameters (MPN/g <sub>DM</sub> )										
Fecal coliforms	154	1.5	8200	11.1 × 10 <sup>6</sup>	0.05 × 10 <sup>6</sup>	0.1 × 10 <sup>6</sup>	640 × 10 <sup>6</sup>	<10,000	45	29.2
<i>Salmonella</i> sp.	176	0	0	114.1	1.5	50	2700	<100	146	83

Table 1. Cont.

Parameter	n. of Data	MIN	1st Quartile	Average	Median	3rd Quartile	MAX	D.D. 6665/2019 * and D.G.R X/2031/2014 Limit Values	n. Analysis Complying with Limits	% Analysis Complying with Limits
Organic micropollutants (mg/kg <sub>DM</sub> )										
PCB	205	0.001	0.005	0.1	0.05	0.1	0.847	$\Sigma < 0.8$	204	99.5
PAH	205	0.005	0.005	0.4	0.05	0.5	4.577	$\Sigma < 6$	205	100
PCDD/F + Dioxin-like PCB (ng TEQ/kg <sub>DM</sub> )	31	0.01	0.6	3.3	1.28	5	22.57	$\Sigma \leq 25$	31	100
Nonylphenols	72	0.0025	2.5	2.9	2.5	2.5	31	$\Sigma < 50$	72	100
DEHP	71	0.05	0.5	5.8	2.65	5	58.7	$\Sigma < 100$	71	100
Heavy hydrocarbons	197	9	410	2309.1	1718	3218	9544	$\Sigma \leq 10,000$	197	100
AOX	86	0.0025	0.5	9.81	3.75	17.5	75.7	$\Sigma \leq 500$	86	100
Toluene	95	0.01	0.12	16.4	2.38	5	385	$\leq 100$	91	95.8
Degree of stabilization (%)										
VS/TS	129	30.1	63.5	68.4	70.9	75	89.1	<65 (<60)	38 (21)	29.5 (16.3)

\* Limit values indicated are those for "suitable" sludge, and in parenthesis limits for "high-quality" sludges.

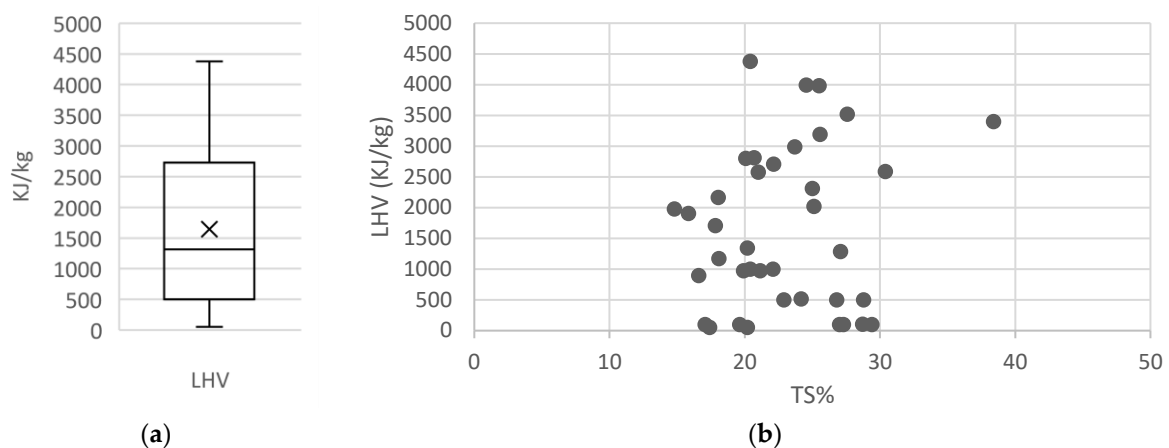


**Figure 4.** Results of elaborations on normed heavy metals for sludge produced by WWTPs in Lombardy (2014–2020) compared with limit values by D.D. 6665/2019. Empty dots indicate outliers. Some very high outliers were not included in the figure for readability reasons.

Another interesting parameter to look at, reasoning on possibilities for sludge recovery, is the lower heating value (LHV) (Table S2, in Supplementary Materials). Considering sludge as a fuel, the higher its LHV, the better the recovery of energy that can be obtained by combustion. The average value for LHV of sludge from available data in Lombardy is 1642 kJ/kg ( $n = 39$ ), the median value is 1315 KJ/kg, with correspondent values of total solids (TS) of 23% on average (min 15%–max 38%) (Figure 5).

The energy balance conducted by using data from the three samples (a, b, and c), for which the certificates of analyses contained the elemental analysis, showed that, in a combustion chamber, they could reach temperatures equal to 253 °C, 617 °C, and 267 °C, respectively (Table 2). The temperature reached would not always be sufficient for the self-combustion of the waste and energy recovery and for ensuring a temperature high enough to prevent chlorinated organic compound formation.

The results of the iterative analysis using energy balance equations conducted on the three samples and average values of primary and secondary sludge are summarized in Table 3 and represented on triangular plots in Figure 6.



**Figure 5.** Sludge outgoing from WWTPs in Lombardy (2014–2020): (a) LHV values ( $n = 39$ ) (b) distribution of LHV in relation to TS.

**Table 2.** Results of the calculation of HHV and of the temperature that samples a, b, and c (for which elemental analysis was available) can reach in a combustion chamber.

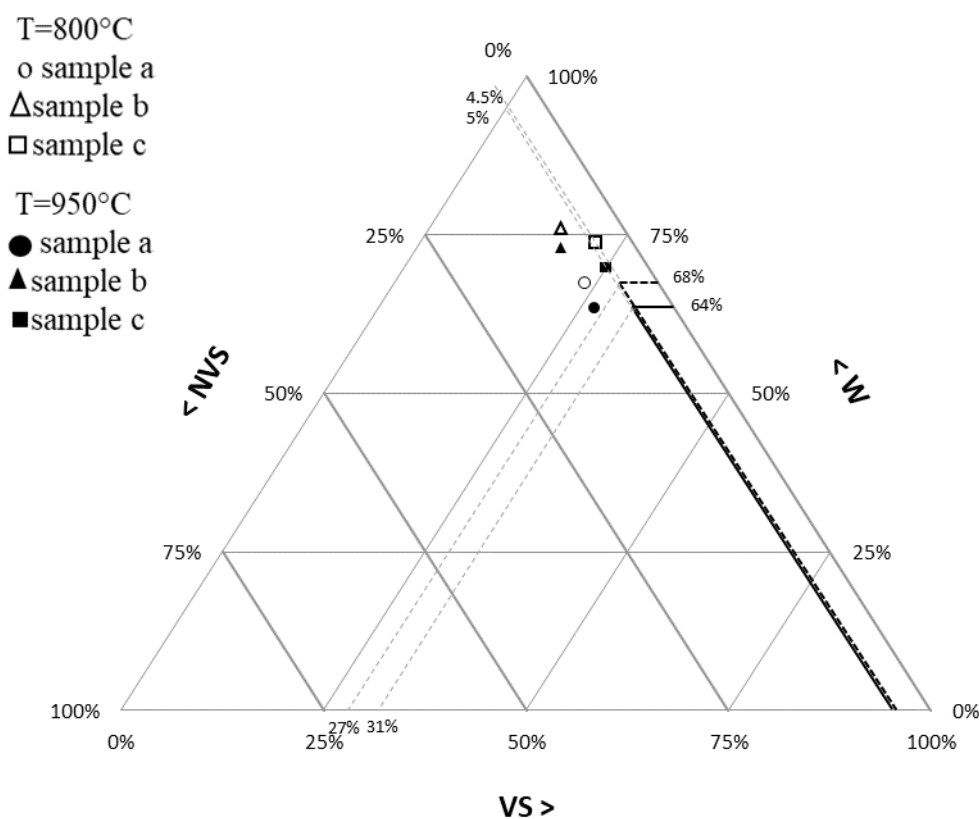
Sludge Sample	Data						Calculated Values		
	C [% <sub>DM</sub> ]	H [% <sub>DM</sub> ]	N [% <sub>DM</sub> ]	S [% <sub>DM</sub> ]	W [%]	NVS [%]	VS [%]	HHV [kJ/kg <sub>DM</sub> ]	T [°C]
a	34.7	5	6	0.1	80.1	5.4	14.5	2811	253
b	39.2	6	7	0.2	79.8	7.0	13.2	3965	617
c	41.7	6	7	0.2	83.4	2.8	13.8	2939	267

**Table 3.** Results of the calculation, for three sludge samples and for primary and secondary sludge, of threshold values for water content (W), VS, NVS and TS to reach the set temperatures of 800 °C and 950 °C. Average, minimum and maximum for primary and secondary sludge refers to results obtained using average, minimum and maximum of values for VS/TS indicated in the literature [46].

Sludge	Results for T = 800 °C				Results for T = 950 °C			
	W [%]	VS [%]	NVS [%]	TS [%]	W [%]	VS [%]	NVS [%]	TS [%]
a	68.1	23.2	8.7	31.9	63.6	26.5	10.0	36.5
b	76.3	15.5	8.2	23.7	73.0	17.7	9.4	27.0
c	73.5	22.0	4.5	26.5	69.6	25.3	5.1	30.4
Primary, average	75.5	17.2	7.4	24.5	72.0	19.6	8.4	28.1
Primary, min-max	72.3–78.1	16.6–17.6	11.1–4.4	27.7–22.0	68.4–74.8	19.0–20.2	12.7–5.0	31.7–25.2
Secondary, average	68.9	19.7	11.4	31.1	64.5	22.5	13.0	35.5
Secondary, min-max	67.1–70.5	19.4–20.1	13.5–9.4	32.9–29.5	62.6–66.3	22.1–22.9	15.4–10.8	37.5–33.7

Figure 6 shows the threshold values for the content of W, VS and NVS (%) needed to obtain a temperature of at least 800 °C and 950 °C, respectively. The threshold values for the water content, both for 800 and 950 °C, were lower than those of the three samples exiting the WWTPs, and the content of VS and NVS was higher. According to the results for the three samples produced by WWTPs in Lombardy, the sludge could reach the temperature of 800 °C in the combustion chamber under the following conditions:  $W < 68.1\%$ ,  $NVS < 4.5\%$ ,  $VS > 27\%$ ,  $TS > 31.9\%$ . The sludge could reach the temperature of 950 °C in the combustion chamber under the following conditions:  $W < 63.6\%$ ,  $NVS < 5.1\%$  and  $VS > 31\%$ ,  $TS > 36.5\%$ . Calculations for an average sample of primary sludge showed that it should contain  $W < 75.5\%$ ,  $NVS < 7.5\%$  and  $VS > 17.2\%$  to reach at least 800 °C in a combustion chamber, and  $W < 72\%$ ,  $NVS < 8.4\%$  and  $VS > 19.6\%$  to reach at least

950 °C (Table 3). Calculations for an average sample of secondary sludge showed that it should contain water < 68.9%, NVS < 11.4% and VS > 19.7% to reach at least 800 °C in a combustion chamber, and water < 64.5%, NVS < 13% and VS > 22.5% to reach at least 950 °C (Table 3). From comparing the obtained values with those calculated for an average sample of primary and secondary sludge, it can be observed that the quality of the three samples of sludge analyzed, from the point of view of energy recovery, is closer to that of primary sludge.



**Figure 6.** Calculated threshold values for sludge produced by WWTPs in Lombardy ( $n = 3$ ) for water content (W), volatile solids (VS) and non-volatile solids (NVS) and identification of the self-combustion areas for  $T = 800$  °C (dotted line) and  $T = 950$  °C (continuous line).

#### 4. Discussion

We collected quantitative and qualitative data on sludge (EWC 190805) produced in Lombardy and analyzed the data against opportunities for recovery.

The quantity of sludge produced by WWTPs in Lombardy has shown an increasing trend in recent years, approximately 10.8% since 2010, in line with estimations of the Regional Waste Management Plan [48]. The quantity of sludge managed in Lombardy in 2017 and 2018 was more than 50% made of sludge that originated outside the region, and the high majority (>93%) underwent recovery operations, the main being recovery of organic substances and energy production. Moreover, the quantity of sludge undergoing R3 is about three times greater than sludge undergoing R1. These results were confirmed by the analysis of qualitative data.

In the last 5 years, sewage sludge from WWTPs in Lombardy showed on average a quality favorable to recovery in agriculture, even if it required, in most cases, additional treatment (chemical, biological, or thermal) in STPs before recovery. The most commonly used chemical treatment is conditioning using hydrated lime or an ammonia solution, or aerobic/anaerobic stabilization [32]. These treatments are oriented towards improving microbiological characteristics, the degree of stabilization, and the percentage of organic carbon, but do not affect other parameters such as heavy metals and trace pollutants, except

for decreasing their concentration by mixing sludge of different quality. Nevertheless, there are limits to heavy metals in sludge admitted to STPs for treatment. With regard to analyzed samples available for this study, in about 91% of cases, the sludge has quality characteristics potentially suitable for recovery in agriculture if undergoing additional treatments to improve stabilization, while the 9% should undergo another type of recovery/disposal. Available data were not sufficient for an appropriate estimation of the quality related to the quantity of produced sludge, but it is interesting to observe that available analyses represent mostly plants with a capacity > 10,000 EI. Other factors influencing the recovery in agriculture, such as odor emissions and anomalous environmental and health risks perception, soil characteristics, and STPs capacity, were not analyzed in this study. It has to be noted that available data corresponds to parameters imposed by the legislation in force, which does not consider many emerging contaminants.

Results on qualitative characteristics of sludge related to its recovery as a fuel for energy generation showed that the water content of sewage sludge in Lombardy is too high for direct recovery by mono-combustion, and it should undergo further treatment to reduce the water content and increase the LHV. This implies higher energy costs to reach the minimum value for TS needed for energy recovery. Moreover, the minimum amount of TS would be even higher if we also consider the need for an excess of air, for process reasons. The excess of air would lower the combustion temperature and, consequently, a higher LVH would be needed to reach the set temperature of combustion (i.e., 800 °C or 950 °C).

Present regulations on the recovery of sludge in agriculture in Europe, and not only, focus on chemical and microbiological indicators on sludge and soil, with some use limitations. The debate on the suitability of these regulations is still open and controversial. A completely different approach is adopted in the US (mainly focused on microbiological contamination) and Europe (mainly focused on chemical pollution). There is a lack of studies and literature on determining sludge reuse based on risk-based approaches and toxicological essays. It is acknowledged that chemical characterization alone cannot account for a mixture of pollutants' antagonistic, subtractive, additive, or synergistic effects [49]. Greater awareness of the real effect of the recovered materials on the surrounding environment may empower their reuse and circular economy with safety. A better understanding of sludge characterization and toxicological properties may also help people understand real risks and, ultimately, increase public acceptance of sewage sludge use in agriculture [50]. Low acceptance of agriculture recovery is one of the main challenges of sewage sludge management, together with the opposition of public opinion to plants with complex technologies such as incinerators and thermal valorization, and the uncertainty of the recovery in cement plants due to the crisis in the construction sector. These factors contribute to uncertainties affecting difficulties in managing the problem, creating difficulties in decision-making and investment. Recovery in agriculture has the advantage from the environmental point of view of resource recovery, making it an elective form of recovery. On the other hand, the need for a certain quality of sludge and the lack of risk-based analysis for their safe reuse are a constraint for adopting this choice. Energy recovery would also need a certain quality of sludge, in particular a lower water content and higher LVH values. Given that the energetic balance does not allow for net energy production, "thermal destruction" (or thermal inertization) appears to be a viable solution. The realization of thermal inertization plants, on the other hand, implies the need for consistent investments. Moreover, thermal drying of sludge could be an interesting hypothesis because it could guarantee certain flexibility, acting on the moisture content of sludge depending on need and consequently on the final destination, leaving open multiple possibilities.

Based on the results of this study and with regard to circular economy principles, the recovery of organic substances and nutrients in agriculture should be prioritized for sludge respecting regulatory limits for agricultural reuse after treatment (e.g., in STPs, co-composting, co-digestion). The alternative route is represented by the mono-combustion of sludge, with phosphorous recovery from ashes. If not viable, other thermal processes

such as combustion with energy recovery in cogeneration plants are recommended. Finally, landfilling of sewage sludge should not be considered as an option.

Anyhow, moving in the direction of retrofitting existing wastewater treatment plants to maximize the recovery of material resources and pushing toward the adoption of innovative technologies does not always represent the best choice in view of the integrated technical, economic and environmental perspective [51]. Thus, it seems that complying with quality standards defined by regulations is not a sufficient requirement to favor, in practice, sludge (and other residues) reuse: the perception of risk of pollution and damage to crops, the environment, and, in turn, to human health is a key factor which affects the whole process. Therefore, the direct application of circular economy principles is seriously threatened unless alternative and more complex routes are promoted, including the development and application of innovative technologies, which are expensive, may lead to a lower extent of material resource recovery and pose other environmental issues (additional environmental footprint).

Finally, it is indispensable, for the examination of possibilities and the selection of solutions appropriate for the territory of interest, to have site-specific information on the variables involved in the problem. This study contributed by studying the quantity and destination, and collecting and assessing the quality, of sludge produced in Lombardy. For qualitative data, a database was created from publicly available data that were jeopardized and often difficult to locate or understand, because they were created for a different objective (the tender process). Even though the methodology developed and applied had some limitations and biases, it had the advantage of providing a significant amount of open-access disaggregated data in real-time. Analyses of sewage sludge characteristics are conducted and available at the WWTPs level, but data are often jeopardized. We recommend the promotion and creation of an open access database of disaggregated data on sludge quality at a regional and at national levels. Moreover, we hope the methodology developed for this study could be useful or inspiring for other researchers and practitioners in the field to investigate sludge quality in other regions or areas.

It has to be noted that this study has some inherent constraints: first, collected tender processes were limited to those available online; second, analyzed parameters and analytical methods were not consistent over time and across different plants; third, not all the parameters were available for all plants for all considered years, resulting in a different amount of data for each plant or year. Anyhow, despite these limitations, we consider the study shows a reliable picture of the quality of sludge from WWTPs in Lombardy and the economics related to its final recovery or disposal. Moreover, by highlighting the point of weaknesses in the availability of data, the study can be a reference for further studies and research in this area.

## 5. Conclusions

Sewage sludge produced by WWTPs in Lombardy was on average about 395 kilotonnes/year (2010–2018), with an increasing trend of 11% per year. More than 50% of managed sludge in Lombardy was imported from outside the region, and overall, more than 93% of managed sludge was sent for recovery. Sludge recovery was mainly oriented toward organic substance recovery and, secondly, energy generation.

Sludge showed properties on average compatible with recovery in agriculture: in many of the considered analyses, sludge could gain a quality suitable for land spreading after additional treatments (e.g., in STPs). The main parameters showing non-compliance with regional regulatory limits were total chromium and arsenic among heavy metals, microbiological parameters, and the degree of stabilization. Sludge produced by WWTPs in Lombardy showed on average an LHV of 1642 kJ/kg and still has an elevated water content, which should be lowered in order to be used for energy recovery by combustion. Whilst thermal processes remain recommended for sludge non-compliance for agricultural reuse, further dewatering and energy recovery imply higher and more complex plants and technologies, involving higher costs and investments. In Lombardy, sludge management is

already oriented to resource recovery in agriculture, and the study shows that there is a potential for improvement in this area considering treated sludge quality. Besides chemical and biological characterization, the study of the toxicological properties of biosolids should also be fostered to ensure their safe reuse. Sludge management remains a big challenge that requires complex and context-specific solutions. The analysis of the quality and management of sludge in the region Lombardy, as a case study, can support and orient decision-makers on a strategy for recovery.

**Supplementary Materials:** The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/app122010391/s1>. Section S1: Recovery and disposal operations; Section S2: Detailed materials and Methods Section; Section S3: Results; Table S1: Results of elaborations on data from WWTPs with a known type of treatment and dewatering used. Comparison of the degree of stabilization of sludge obtained by aerobic or anaerobic treatment. Comparison of the percentage of dry matter obtained by the use of different dewatering processes; Table S2: Results of elaboration of data from analyses of sludge produced by WWTPs in Lombardy (2014–2020) for parameters of relevance for biosolids reuse and recovery (not normed).

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