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Sewage Sludge - a Sustainable Material for the Production of Solid Recovered Fuel

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The appropriate quality system of production control of pre-treated sewage sludge and its characterization needs to be established in order to provide confidence in its constant quality as a raw material in the circular economy. Waste to energy procedure with increased energy efficiency in utilization of its residues is gaining an increased recognition. To manage waste to energy procedure the pre-treated (dried) sludge must be continuously assessed in accordance with the CEN/TC 343 technical standards and the European Union (EU) initiative for circular economy. In evaluation of the dried sewage sludge as an alternative solid recovered fuel (SRF), three parameters are important – net calorific value (NCV), chlorine (Cl) and mercury (Hg) content. The statement of the authorized contractor on the quality of sewage sludge as a SRF with the Class code NCV 3 to 4; Cl 1; Hg 3 creates confidence in its stable quality for energy production in cement kilns, power plants and dedicated sludge mono-incinerators. The removal of Hg from municipal wastewater, constant quality control of the dried sewage sludge, and tracking the mass balance of Hg during incineration make it possible to prevent the circulation of this critical pollutant in the environment.

1. Introduction

To ensure sufficient collection and treatment of municipal wastewater, it is expected that the agglomeration for which municipal infrastructure is required will increase, and correspondingly the amount of sewage sludge. It is formed continuously in the process of wastewater treatment and should be re-used whenever appropriate and recovery routes should minimize the adverse effects on the environment (Urban Wastewater Directive Overview, 2021). Waste to energy (WtE) recovery procedure R 1 (Waste Framework Directive (WFD), 2008) with increased energy efficiency is becoming an increasingly recognizable and meaningful way for sludge recovery (Briefing note, 2021). To ensure that the recovery of energy content from waste in the EU supports the objectives of the circular economy (CE) action plan and that WtE is firmly guided by the EU waste hierarchy, EU Commission framed a long-term CE perspective (Communication, 2017). It makes sense that suitable non-hazardous solid wastes with a higher content of organic matter, among them sewage sludge, are processed as a SRF (EN 15359, 2011), thereby reducing the carbon footprint and contributing to reduction of fossil CO_2 emission. According to Article 6 of the WFD (2008) SRF can cease to be waste at the EU Community or national level if certain criteria are fulfilled. WtE technologies may also contribute to heavy metal emissions into the air (IED Directive, 2010), of which Hg is particularly under scrutiny, and as a global pollutant has been selected as an indicator to identify the potential for harmful emissions of substances into the air during SRF combustion. Due to human activity, Hg levels in the atmosphere were 450 % above natural values, which meant a 20 % increase in Hg in the environment between 2010 and 2015 (Chemical, Wastes and Climate Change, 2021). Stationary combustion of fossil fuels and biomass is responsible for about 24 % of global Hg emissions, with coal burning being the primary contributor of 21 %. Under current energy policy scenarios, Hg deposition fluxes will grow in Asia but drop in North America and Europe by 2035 (Chemical, Wastes and Climate Change, 2021). In accordance with European Pollutant Release and Transfer Register (Regulation (EC), 2006) the annual amount of pollutants released into the air, including Hg, should be reported for incineration and co-incineration plants, but not for the efficiency of flue gas treatment. To effectively

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prevent the releases of critical pollutants into the air, it is necessary to control mass balances (input and output) for each pollutant. The basis for achieving efficient flue gases treatment is, in addition to the appropriate technology, a good knowledge of the input materials. This enables the process of specification and classification of SRF, which is primarily the task of its producer, and in the case of sewage sludge, that is municipal wastewater treatment plant (UWWTP) operator. Reporting on the effect of removal heavy metals in flue gases is not yet legally required, but this requirement would certainly contribute to a cleaner environment and human health. Zarei (2020) examined the wastewater resources management approach for energy recovery from the CE perspective. It was found that: i) the processes in the case of energy recovery from wastewater could induce extra costs due to excessive operational and investment cost, ii) geographical and temporal variations between supply and demand, lack of infrastructure, and cost issues are challenging factors for off-site recovered energy, and iii) that several technologies for the recovery of energy, fertilizer, and other products from wastewater have been explored in the academic arena and research institutes. Few of these have been applied on large scale due to technical immaturity and non-technical limitations. The establishment of defined outlet procedures/routes is needed, and a good sludge quality must be guaranteed to properly perform the utilization operations, correctly fulfil the legal requirements, and build stakeholder and public confidence (Spinosa, 2016).

The methods of evaluating the energy and environmental suitability of dried sewage sludge for use as SRF are determined with technical standard EN 15359:2011. According to this standard the classification system for SRF is based on three important parameters, referred to the main SRFs properties: i) an economic functional parameter (NCV), ii) an operational technical parameter, referring to possible equipment corrosion (Cl) content) and iii) an environmental parameter (Hg content). These parameters are chosen to give the stakeholder an immediate but simplified picture of the fuel in question. The standard sets the statistical procedures for generated analytical results of Hg, Cl and NCV. It also sets up specific requirements for sampling procedures to cover the sampling dynamics for preparing partial subsamples and for selection of methods and techniques for analysis, necessary to obtain statistical parameters, as follows: "as received" ($_{ar}$) results i) the 80th percentile and the median for the ratio of Hg (in $mgkg_{ar}^{-1}$) to NCV (in $MJkg_{ar}^{-1}$), Hg/NCV, and ii) the average of the results for NCV (in $MJkg_{ar}^{-1}$) and "as dry matter" ($_{DM}$) results for the average for Cl (in % m/m_{DM}). By statistical evaluation of the results obtained for each analyte separately, in accordance with the limit values set up by the standard, an aggregated quality Class code from 1 to 5 is defined. The Class code 1 means the best quality, and the Class code 5 means the worst quality of designated SRF. At least 10 representative samples of treated waste, sampled evenly over a calendar year, are needed to specify its quality Class code. Thermal behaviour of SRF is also important information for the optimal facility operation. Mislej et al. (2012) examined the thermal behaviour of dried sewage sludge and it was found: i) that carbon can be, in the thermal oxidative process, released from the sludge in the form of CO_2 and C-H, ii) H_2O and CO_2 are released in the temperature range up to 740 °C, iii) in the last part of the thermal decomposition of the sample, in the range from 740 °C to 800 °C, only inorganic CO_2 is produced, iv) in the ash SiO_2, MgO and CaO remained, and v) released inorganic fraction of CO_2 can be seen in an inert and oxidative atmospheres of the thermal treatment. These findings provide a good insight into CO_2 generation from a GHG perspective.

In the case of the Central wastewater treatment plant Ljubljana (WWTPL) (JP VOKA SNAGA, 2019), which is a large municipal wastewater treatment plant, the possibility is presented of a holistic approach to the management of sewage sludge at UWWTPs, which enables the integrated implementation of wastewater treatment, SRF production, and the bridge to WtE in terms of adopted waste hierarchy (Communication, 2017), reducing carbon footprint and pollution of the environment.

2. Materials and methods

The environmental permit for the WWTPL, valid from 2012, sets the technology and procedures for the treatment of municipal and storm wastewaters, pre-treatment of surplus sludge and limit values for the quality of treated wastewater before discharge into the Ljubljanica river (Figure 1, points A, B, C and D). The entire WWTPL quality control system has been set up according to ISO 9001:2015 - Quality management systems, Requirements (Figure 1, point B). The legislative framework for the environmental permit is based on the National Decrees (Figure 1) as follows: i) in the field of industrial pre-treated wastewater, emissions of substances and heat into public sewers (Decree 2012, point A), the management of municipal wastewater, precipitation, and surplus sludge (Decree 2012, Decree 2015, point A), ii) in the field of prevention of emissions of substances into the air (Decree, 2007), and iii) the management of pre-treated dried sewage sludge (pellets) is in general defined by the Decree on waste (Decree 2015) and special national demands on SRF production are prescribed by the Decree 2014 (point C). To achieve the objectives of the latter, the existing quality management system (Figure 1, QMS) must be upgraded with specific demands for the operation of facilities for the production and trade of SRF (EN 15358:2011), covering operations from the point of acceptance of surplus sludge destined for recovery (Figure 1, Waste management) to the point of delivery of the SRF shipment to the final stakeholder in accordance with the contract (Figure 1, point D). This is mainly about resource management, product realization, analysis, measurement and improvements. Physico-chemical analyses of wastewater were performed using standard methods (ISO/TC 14/SC 2). An accredited quality system has also been established for the excess sludge and pellets sampling procedure, which includes manual time-proportional daily sampling in accordance with the standard ISO 5667-13:2011, Water quality, Sampling, Part 13, and EN 15442:2011, from deliveries or from stock (Figure 1, QMS, points A, B and C). Accredited sampling procedure was established in 2009 and continues to a similar extent (Mislej et al., 2012). Mass proportional representative annual composite samples are prepared from refrigerated (5 ± 1 °C) monthly representative subsamples according to the quantity of delivered pellets for each month. For the specification and classification of pellets as a SRF (EN 15359:2011), 10 monthly representative samples (prepared from daily subsamples) were characterized with the accredited standard analytical procedures according to Technical Committees (TC) CEN/TC 292 "Characterization of waste" and CEN/TC 343 "Solid recovered fuels" and are performed by authorized contractors (Figure 1, point D).

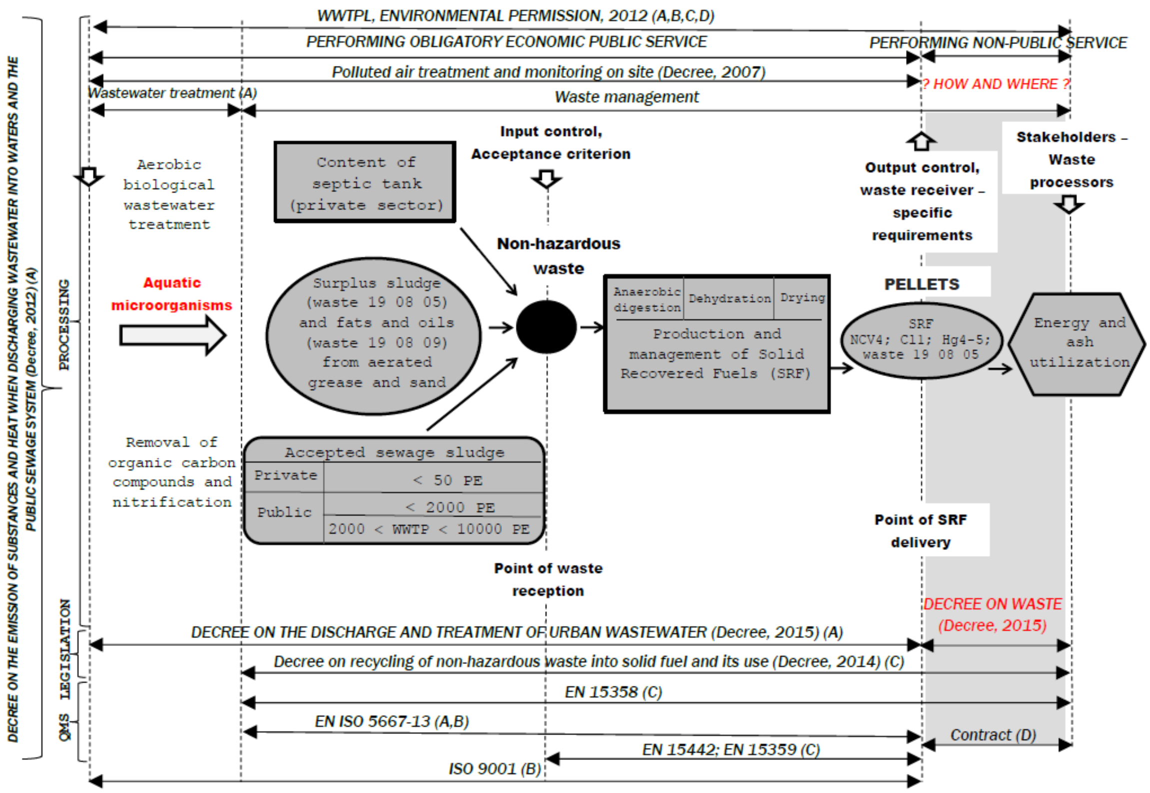

Figure 1: The management system for municipal wastewater treatment and SRF production at WWTPL

3. Results and discussion

The WWTPL is a single-stage mechanical-biological wastewater treatment plant and is designed for conventional secondary treatment of municipal wastewater, precipitation (mixed sewerage system) and 11 % v/v industrial wastewater of the capital city Ljubljana. The efficiency of secondary wastewater treatment is adequate (JP VOKA SNAGA, 2019). Due to seasonal fluctuations and inlet wastewater flow (Figure 2a) the amount of chemical oxygen demand (COD) removed varies between 700 t O_2 to 1,450 t O_2 per month and on average it is lower during summertime. The annual dynamic of nutrients removal from wastewater is stable and fluctuates around 14,000 t COD, from 483 t to 666 t of nitrogen (N) and from 91.0 t to 126.1 t of phosphorus (P) (Figure 2a). Seasonal fluctuation is also characteristic for the efficiency of anaerobic digestion of sludge, and consequently also of the biogas and pellets production. Given the properties of surplus sludge as a substrate (ratio C/N is 6.2/1), 40 % efficiency of anaerobic degradation is expected (Cano et al., 2015). On average, the biogas generation (with approximately 63 % v/v of methane) is 16 to 17 NL per population equivalent (PE) based on BOD_5 analysis, and the pellet's production is 31 $g_{DM}PE^{-1}day^{-1}$. As a plant with a designed capacity of 360,000 PE, the WWTPL has well-equipped facilities for thermal pre-treatment (drying) of dehydrated digestate to pellet shape. On average, particle-size distribution of pellets is d_{10}=2.1 mm, d_{50}=2.6 mm and d_{90}=3.2 mm with bulk density of 647 kgm^{-3} (Table 1). On average, the annual pellets generation is 4,288 t, in volume of 6,630 m^3.

Table 1: Proximate and ultimate analysis of annual samples and average values of 10 consecutive monthly subsamples (*) of pellets.

Parameter	Unit	2012	2013	2014	2015	2016	2017	2018	2019	2020
TOC	% C m/m_{DM}	38.4	37.6	39.5		33.1		35.7		
TIC		2.0	3.6	<0.1	n.a.	9.9	n.a.	6.9	n.a.	
H	% m/m_{DM}	4.40	4.21	5.08		n.a.		n.a.		
Hg*	$mgkg^{-1}_{DM}$	2.05	1.57	1.83	1.69	1.34	1.14	1.13	1.29	1.11
Cd		1.2	1.2	1.1	1.1*	1.2*	0.87*	0.80	0.84*	0.79*
S		1.15	0.93	1.26	n.a.	1.06	n.a.	0.80	0.88	0.90
Cl*		0.081	0.077	0.112	0.106	0.107	0.101	0.075	0.093	0.088
Dry matter at 105 °C	% m/m_{DM}	90.9	90.8	91.5	91.6*	90.4	92.5*	90.5	91.1*	91.0*
Volatile matter at 900 °C		53.9	54.0	53.3	n.a.	53.0		n.a.		
Ash at 550 °C*		32.1	32.9	34.0	32.0	33.1	29.4	31.8	29.7	29.1
Inorganic CO_2*		3.47	3.76	3.63	3.07	4.13	3.20	3.17	2.96	2.85
NCV*	$MJkg_{ar}^{-1}$	13.37	12.99	13.83	12.93	12.95	13.45	12.95	13.37	12.99
Pellet's production	$t_{DM}y^{-1}$	3,839	3,867	3,612	3,831	4,056	4,238	4,192	4,048	3,573
Bulk density	kgm^{-3}	641	676	683	n.a.	646	630*	628*	637*	633*

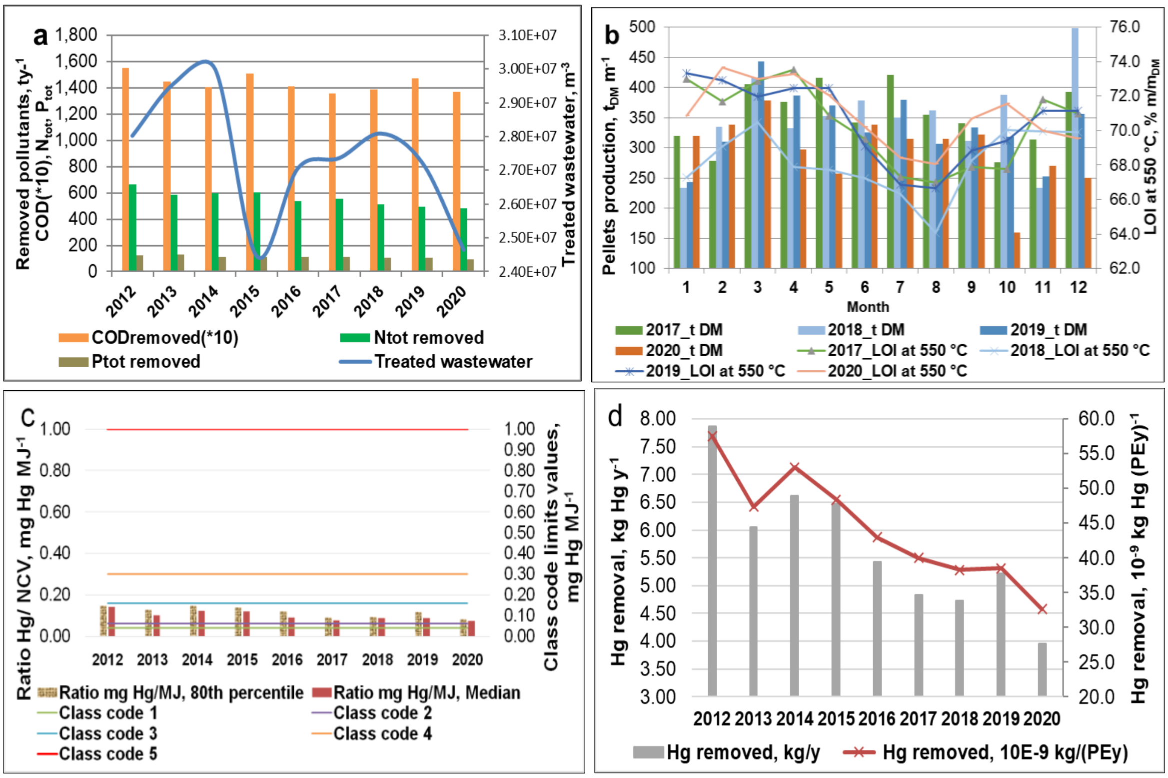

Figure 2: Dynamics of WWTPL: a) outflow, COD, nitrogen and phosphorus removal rate per year, b) pellet's production and LOI at 550 °C, c) SRF classification (ratio Hg/NCV of 10 monthly subsamples), and d) annual removal rate of Hg

Drying of sewage sludge enables low moisture content (<10 %). This reduces the cost of delivery to stakeholder, and CO_2 emissions from transport and increases the calorific value of pellets (Table 1). The biomass composition of aquatic microorganisms, as a tool for biological wastewater treatment and as a substrate for digestate production (Figure 1) and physico-chemical properties of pellets (Table 1) define the

thermal behaviour of pellets and origin of organic and inorganic source of CO_2 (Mislej et al., 2012). For energy production, important aspects are the content of TOC, hydrogen and dry matter, which is reflected in the calorific value (on average 57,071 GJy^{-1}). Due to variations in the removed COD, the amount of pellets produced and their organic content (in terms of loss on ignition (LOI) at 550 °C), delivered to the stakeholder, varied accordingly. The highest pellets production is during spring and summer, while the organic content is the lowest in summer (Figure 2b). At the annual level, there is no significant fluctuation in the amount of pellets and their organic content. Fluctuation in calorific value (from 13.47 $MJkg^{-1}{}_{DM}$ in 2018 to 15.35 $MJkg^{-1}{}_{DM}$ in 2020) and inorganic CO_2 (as a mass difference between LOI at 550 °C and LOI at 900 °C), due to the thermal decomposition of calcium carbonates, is much more significant – Table 1 (Mislej et al., 2012). The major part of pellets composition represents the volatile substances, determined with CEN/TC 343 standard procedure (at 900 °C for 7 minutes in a crucible with a lid) (Table 1). The residue, as the ash determined at 550 °C, contains MgO, CaO and SiO_2 (Mislej et al., 2012; Table 1).
The purpose of producing a SRF is to use the pellets for energy production at the highest possible energy efficiency. Well defined system for classification and specification of pellets as a SRF is therefore of great importance to reach sufficient acceptability on the fuel market and public trust (Figure 1, Point of SRF delivery). To ensure an efficient and economic energy recovery of pellets by a stakeholder site, a permanently control of SRF on both side is required (Figure 1, Contract, point D; Table 1). Figure 2c, shows the statistical characteristics of 10 monthly composite subsamples of pellets from 2012 - 2020, based on the parameters limits for the ratio Hg/NCV prescribed by EN 15359:2011. On average, the removal efficiency of Hg from wastewater at WWTPL is 84.6 ±17.0 %. From 2012 to 2020, a significant decrease of Hg content in the pellets is observed. From 7.87 kg Hg y^{-1} delivered to final stakeholder in 2012 to 4.46 kg Hg y^{-1} delivered in 2020, and the removed Hg from water environment decreased from $57.6*10^{-9}$ kg Hg $PE^{-1}day^{-1}$ in 2012 to $36.7*10^{-9}$ kg Hg $PE^{-1}y^{-1}$ in 2020 (Figure 2d).
For the period from 2012 to 2020, a specification and classification of the pellets has been defined as a SRF with Class code NCV 3 to 4; Cl 1; Hg 3. The values for NCV varies from 12.93 $MJkg^{-1}{}_{ar}$ (limit value for 4th class is 10 $MJkg^{-1}{}_{ar}$) to 13.96 $MJkg^{-1}{}_{ar}$ (limit value for 3rd class is 15 $MJkg^{-1}{}_{ar}$). Cl content in pellets was continuously <0.2 % m/m_{DM}, which is the first (the best) class limit value for SRF (Table 1).

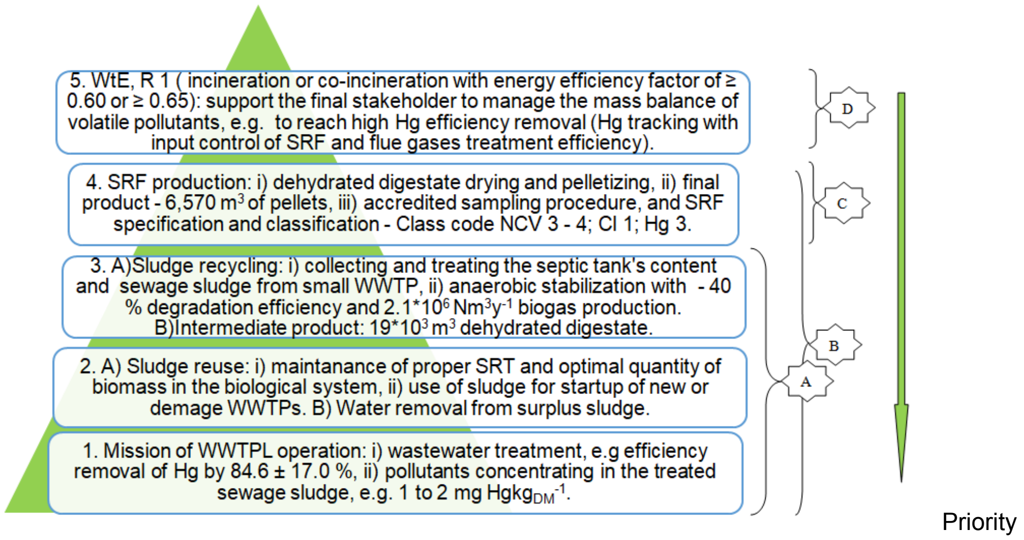

Figure 3: Integrated implementation of wastewater treatment and WtE procedures in terms of waste hierarchy (WFD, 2008), reduction of carbon footprint, and prevention of Hg circulation in the environment

The management system for successfully performing obligatory economic public service and SRF production consists of the following items (Figure 1 and Figure 3): i) legislation on wastewater treatment (point A), ii) quality control system set up according to ISO 9001:2015, and sampling procedures for wastewater and excess sludge (point B), iii) legislation, technical standards for SRF characterization (sampling procedure, specification and classification), and quality control system (point C), and iv) contract with the stakeholder for pellets (point D). By specifying and classifying sewage sludge as a SRF, UWWTPs operators follow the EU WtE strategy. In addition to being a sewage sludge a sustainable material for SRF production, carefully controlled WtE procedures could achieve the decreasing of fossil carbon footprint and the circulation of pollutants in the environment, especially Hg.

4. Conclusions

UWWTPs play an important role in achieving good status for surface and ground waters. Creating a bridge between municipal wastewater treatment, and final sewage sludge recovery procedures without adversely affecting the environment, are the foundations for transition to circular economy in this sector. In the case of UWWTPL, the system of successful processing of excess sludge into SRF is presented. Despite its anaerobic stabilization and the production of biogas, the pellets produced still contain enough organic content to be used as SRF. In order to achieve adequate traceability of pellets, their quality, and classification for use as a SRF, it is necessary to establish an appropriate quality system, covering pellets sampling procedure for preparation of representative daily and monthly subsamples, and further the final representative annual sample for the purpose of extended physico-chemical analysis (Figure 1 and Figure 3, points A, B, C and D). Statistical evaluation of analytical results of 10 monthly representative samples for each calendar year, gives to stakeholder sufficient data on pellets as the input material for WtE. For the period from 2012 to 2020, a classification of the pellets produced at WWTPL, define their conformity as a SRF with Class code NCV 3 to 4; Cl 1; Hg 3. To achieve this result, it is necessary to have a comprehensive quality control system, covering UWWTP basic operation and its business cooperation with authorized waste assessment contractors and stakeholders (Figure 1 and Figure 3, point D). The quality management system for sustainable SRF production provides an integrated approach to the sewage sludge final recovery route, which allows to reduce the sewage sludge quantity, the GHG emission and the circulation of pollutants, especially Hg, in the environment. With increasingly stringent control of emissions of substances into the air, WtE procedures have another great advantage, because efficient flue gas cleaning prevents re-dispersion of pollutants that have been removed from wastewater. The removal of Hg from municipal wastewater and tracking the mass balance of Hg during incineration of sludge make it possible to prevent the circulation of this dangerous pollutant in the environment. The paper also provides the starting point for further research at the level of full-scale facilities for WtE on the efficiency of Hg removal, and not only on the achievement of limit values for Hg emissions into the atmosphere.

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