

SEWAGE SLUDGE MANAGEMENT AT THE CENTRAL WASTEWATER TREATMENT PLANT LJUBLJANA – CHARACTERIZATION OF ITS PROPERTIES, POSSIBILITIES FOR SUSTAINABLE UTILIZATION AND LEGISLATIVE OBSTACLES

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ABSTRACT

The treated sewage sludge under consideration is a hygienized biodegradable waste in the form of pellets. It can be used as a fertilizer, but only for spreading on non-agricultural land. Regarding “waste to energy” philosophy, the specification of pellets as an alternative solid fuel according to EN 15359 resulted in “NCV4; C11; Hg3-4” class. The major problem regarding the final pellets utilization is the lack of facilities for energy and material recovery from this type of waste in Slovenia. According to the newest legislation regarding the waste management, a product status for residues generated in combustion and pyrolysis of pellets on a laboratory and semi-pilot scale was not achieved. The holistic approach to final pellets utilization was studied and regarding the full-scale level of self-sufficient sewage sludge management in Slovenia, some legislative provisions become significant obstacles.

Keywords: *sewage sludge management, sustainable utilization, legislative obstacles*

Abbreviations: AOX - Adsorbable organically bound halogens; ar - as received; BDW - Biodegradable waste, COD - Chemical oxygen demand; CWWTPL - Central wastewater treatment plant Ljubljana; DOC - dissolved organic carbon; DS - Dry solid; EPA - Environment protection Agency (US); n.a. - no analysis, n.e. - not estimated; NCV - Net calorific value; n.r. - no report; n.rl. - not relevant; OMC - organic matter content; OMD - delivered amount of organic matter; PAHs - Polycyclic aromatic hydrocarbons; PE - Population equivalent; PCBs - Polychlorinated biphenyls; POPs - Persistent organic pollutants; PTMs - Potentially toxic metals; SRF - Solid recovered fuel; SRT - Sludge retention time; TIC - total inorganic carbon; TOC - total organic carbon; UWWTP - urban wastewater treatment plant.

INTRODUCTION

EU Water Framework Directive [1] set rules to halt the deterioration of EU water bodies and to achieve “good status” for rivers, lakes and groundwater by 2015. UWWTPs have an important role to play in achieving this goal. In Slovenia, during years 2000 to 2018, there was an intensive upgrade of public sewerage networks and the number of UWWTPs was increased from 117 (by total load of 1,476,275 PE) to 508 (by total load of 2,680,957 PE) [2]. The amount of sewage sludge increased during this period from 8,800 t_{DS} to 38,100 t_{DS} (Figure 1) [3]. So, in 2014, less than 1 wt.% was spread on agricultural land, about 5 wt.% was aerobically or anaerobically treated, about 1.1 wt.% was disposed on landfills, 39.9 wt.% was managed by other procedures and exported to EU members and about 53 wt.% was incinerated (Figure 1). After 2016, despite the ambitious Slovenian waste management program [4], problems with the sewage sludge utilization have begun to accumulate.

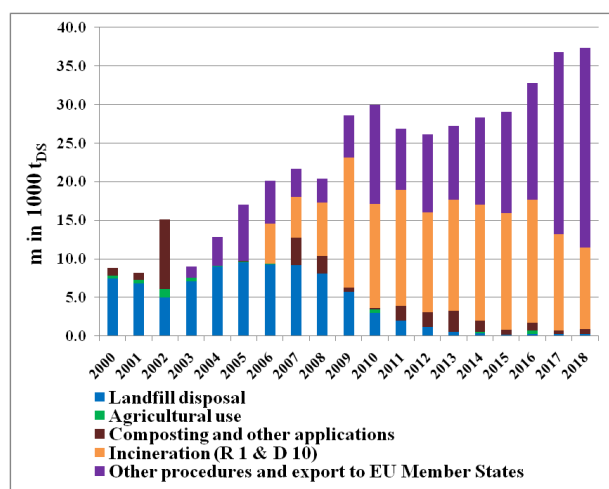


Figure 1. Generation and management of sewage sludge in Slovenia in the period from 2000 to 2018 [3]

In 2017, the interest of the Slovenian cement kiln for the use of dry sludge as an alternative fuel began to decline and the export of sludge for treatment abroad has become inevitable. Given the increase in the number of agglomerations with a load density of ≥ 10 PE/ha and a total load of ≥ 50 PE due to municipal wastewater generation and their

prescribed municipal equipment [5, 6], the amount of sewage sludge will only increase. In accordance with the Slovenian regulation on municipal wastewater management [6], all newly formed agglomerations between 500 PE - 2000 PE must be equipped with a public sewerage network by 31 December 2023 at the latest, and completed with UWWTP. The resulting excess sludge and the contents of septic tanks must be transported to a larger UWWTP, which is equipped for sludge treatment. Since 2014, due to the new method of calculating the costs of removal and treatment of septic sludge in relation to the consumed drinking water, the number of removals and thus the amount of septic tanks have increased significantly. The Regulation [6] stipulates that the public service provider must operate an UWWTP equipped for sludge pre-treatment in order to ensure: i) fulfilment of the requirement for the use of sludge as a fertilizer in agriculture in accordance with the Slovenian Sludge Decree [7], if sludge is finally treated this way and ii) recovery operations or disposal in accordance with the general waste regulations. This means that the final product of pre-treatment of own and taken over foreign surplus sludge and septic sludge must be treated in accordance with the Slovenian Waste Decree [8], namely to ensure the final treatment of sludge within: i) own treatment, ii) delivery to a waste collector or iii) delivery to another recovery or disposal operator. From a holistic point of view, the huge amount of sewage sludge produced should not be a problem because it is a “smart material” of biogenic origin, emerging mainly from domestic wastewater treatment. Due to the composition of the basic functional organs of aquatic microorganisms and their ways of protection against predators, sludge contains a lot of organic carbon and nutrients, such as nitrogen, phosphorus, potassium and minerals, of which the most common elements are Ca, Si and Mg [9]. As a consequence of simultaneous removal of micro-pollutants from raw municipal wastewater and absorption of essential elements for aquatic biomass growth [9, 10], sludge also contains POPs, microplastic, sulphur and PTMs, which may be harmful to the environment and human

health in the event that the sewage sludge is mismanaged.

Waste prevention is the most efficient way to reduce its harmful environmental impact [11], which does not apply to sewage sludge, because a larger amount of treated sludge means more or better treated municipal wastewater and thus better conditions of the environment and consequently human health. In any case, sludge should be seen as a renewable resource [12] that contains valuable raw materials [13]. In order to improve the resource utilization, reduce dependence on certain raw material imports and facilitate the transition to more sustainable materials management and the circular economy model [14], a new paradigm for the valorisation of sewage sludge should be established.

The CWWTPL [15] is an urban one-stage mechanical biological wastewater treatment plant and is intended for conventional secondary treatment of municipal wastewater and small amount of industrial wastewater (approximately 11 % V/V on the yearly level) of Slovenia's capital. Since 2012, CWWTPL's sludge management has been focused on the temporary storage of pellets (pelletized dried sewage sludge) for the purpose of its further utilization with any of the legal recovery operations (procedures from R1 to R11) [8, 11]. From July 2008 to May 2017, the final utilization of pellets was co-incineration in a Slovenian cement kiln. Since June 2017, after concluding a contract for the final treatment of sludge with the waste collector, the pellets have been exported to Hungary, where R3 waste processing was performed. At the end of 2019, the export of sewage sludge to Hungary was banned and thus a huge problem has arisen with the final disposal of sludge, not only for CWWTPL, but for the whole country.

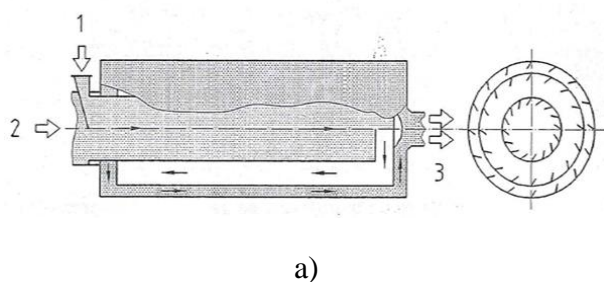
The consideration of legislative provisions and implementation of regular annual assessments of pellets in an accredited laboratory has provided a comprehensive quality control system. Based on the representative's month- and annual samples of pellets, the possibility of their final utilization with appropriate legal

recovery operations (R procedures) [8] in Slovenia was studied: i) as BDW used in soil improvement, as a material for artificial soil or backfilling (R10) and as a biogenic waste for co-composting (R3), ii) as a fertilizer for direct use in agriculture (R10), iii) direct "waste to energy" disposal by incineration and co-incineration (R1), iv) as an alternative solid fuel (SRF) (as a substitute for fossil fuels) in order to reduce greenhouse gases (R1), v) change of the waste classification number from 19 08 05 (sewage sludge) to 19 12 10 (combustible waste), vi) material recovery of the bottom sewage sludge ash (R5), and vii) material recovery of sewage sludge by pyrolysis (R3/R1). The paper will assess all possibilities of final utilization of pellets that are allowed in accordance to the legislation, and compare Slovenian legislative requirements with the legislation of some other EU Member States and EU directives. The purpose of the article is to identify legislative obstacles and their significance, indicate guidelines for optimal energy and material treatment of pellets, and assess the possibility of further material use of generated residues and of the end of their waste status.

EXPERIMENTAL

CWWTPL has a designed capacity of 360,000 PE. The plant has been in full operation since the end of 2005. Because of the mixed sewer system, the amount of removed COD varies between 800 t of O₂ to 1,350 t of O₂ per month and is lower at higher quantity of inflow in the wet weather period. For an effective biological treatment of municipal wastewater, an adequate concentration of activated sludge must be maintained. It is regulated by the appropriate SRT, which means that the excess sludge must be removed on a daily basis to maintain the concentration of activated sludge between 2.5 gL⁻¹ to 3.7 gL⁻¹ in the aeration system according to the seasonal temperature (12 °C to 23 °C) of the input wastewater and desired SRT. The average daily amount of excess sludge is about 18 t_{DS}.

After thickening by gravity and mechanical equipment, along with the addition of polyelectrolyte, the sludge is anaerobically stabilized in a digester, followed by centrifugal dehydration and pelletisation in the drum dryer at 90 °C (Figure 2). The drier was basically designed to generate dried sewage sludge in the form of pellets for agricultural application. Due to the fluctuations of removed COD, the amount of generated pellets and their organic content delivered to the stakeholder varied accordingly (Figure 3).

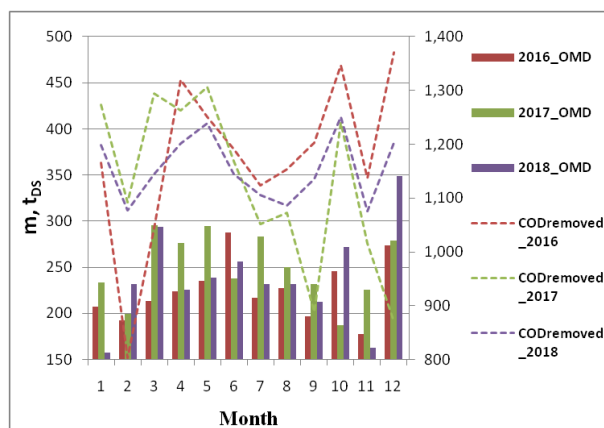


a)

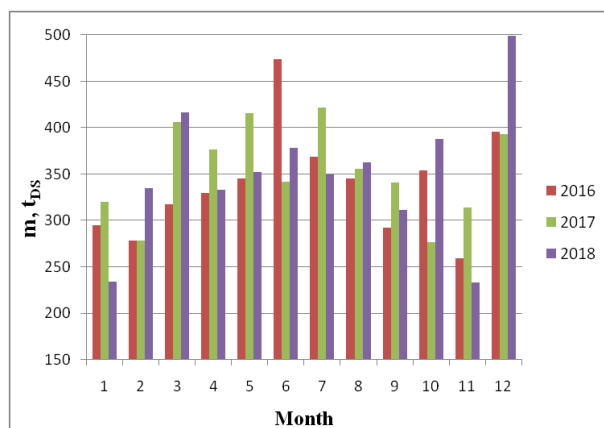


b)

Figure 2. Drum drier at CWWTPL: a) scheme of direct thermal process system (1 - input of wet material, 2 - input of hot off-gases, 3 - output of mixture of pellets, flue gases and evaporated water), b) picture of the dryer



a)



b)

Figure 3. a) Typical quantities of removed COD and OMD of sewage sludge, b) monthly production of pellets in 2016, 2017 and 2018

Sampling techniques

Inflow and outflow on the CWWTPL are measured continuously with a Venturimeter and an in-line Endress+Hauser flow meter. At CWWTPL the quality of raw and treated wastewater is monitored by standardized physicochemical and biological measurements of daily representative 24-hour time-proportional composite water samples. Partial wastewater samples are automatically time-proportionally sampled with Liquistation CSF48, Endress+Hauser. The samples are prepared, cooled and stored in accordance with the standards ISO 5667-10:1992, Water quality, Sampling, Part 10: Guidance on sampling of waste waters, and ISO 5667-3: 2012, Water quality, Sampling, Part 3:

Preservation and handling of water samples. During the dry season, in 2016, in spring (sampling campaign N° 1: May 24; 64,882 m³ per day), and in autumn (sampling campaign N° 2: October 16; 51,720 m³ per day), representative samples were taken at the inlet and outlet in order to determine removal efficiency of the PTMs (Cd, Cr, Cu, Pb, Hg, Ni and Zn), Sb, Co, Mn, V, Tl and some POPs (AOX, PAHs and PCBs) from wastewater. Physicochemical and biological analysis were performed using methods from the groups of standards ISO/TC 14/SC 2 Physical, chemical and biochemical methods. PCBs removal was calculated from the results of samples, collected in similar campaign in 2013, in spring (sampling campaign N° 3: May 14; 79,900 m³ per day) and in autumn (sampling campaign N° 4: October 2; 58,400 m³ per day).

For the pellet sampling procedure, a quality system that includes a manual time-proportionate daily sampling in accordance to the standard ISO 5667-13:2011, Water quality, Sampling, Part 13: Guidance on the sampling of sludge from sewage and water treatment works and EN 15002:2015, Characterization of waste, Preparation of test portions from the laboratory sample, was established, as well as a routine control of pellets as they were shipped to the stakeholder. The pellet sampling plan takes into consideration the relevant Technical Standards: i) EN 14899:2005, Characterization of waste, Sampling of waste materials, Framework for the preparation and application of a Sampling Plan, ii) CEN/TS 15442:2006, Solid recovered fuels, Methods for sampling and iii) CEN/TR 15310-3:2006, Characterization of waste, Sampling of waste materials, Part 3: Guidance on procedures for sub-sampling in the field. From the composite daily sub-samples, monthly composite samples of pellets are made, tested, and stored. Similarly, mass-proportional representative annual composite samples according to monthly delivered amount were prepared out of the refrigerated (5 ± 1 °C) monthly samples. The composite sample was milled to < 1 mm using the Retsch SK1 hammer mill and, when necessary, down to < 0.5 mm in the Retsch ZM 200 mill.

Material and methods

Ten daily samples from December 2019, monthly samples (2016, 2017, 2018) and four annual samples (Pellets 2010, Pellets 2012, Pellets 2016 and Pellets 2018) (Figure 4) were prepared using the described sampling procedure. Standard analytical methods for characterization of BDW and SRF were applied as specified by Technical Committees (TC) CEN/TC 223 “Soil improvers and growing media”, CEN/TC 292 “Characterization of waste” and CEN/TS 343 “Solid recovered fuels”. It should be noted that the pellet digestion to determine the PTMs content of pellets assessed as BDW is done with aqua regia, while the digestion of pellets as SRF is done with a more invasive acid mixture, and results obtained are shown in separate tables.



Figure 4. Hygienized, anaerobically stabilized and dried sewage sludge in the form of pellets

The majority of the methods used for wastewater and pellets characterization are accredited according to the Technical Standard SIST EN ISO/IEC 17025:2017, General requirements for the competence of testing and calibration laboratories and performed by authorized contractors.

RESULTS AND DISCUSSION

Removal rate for metals and some POPs that are relevant for sludge quality assessment for the recovery operation selection is presented in Table 1. Concentrations of pollutants at the inlet to the CWWTPL follow in descending order: Zn > AOX > Cu > Mn > Pb > Cr > Ni >

V > Hg > Tl > Sb > As > Co > PAHs > PCBs, while the removal efficiency follows in the descending order: PAHs > Pb > Cu > Cr > Hg > V > Mn > Ni > Co > Sb > Zn > As > AOX. Given the extent and value of the results for Tl and PCBs, it is not possible to assess their removal rate from wastewater (Table 1). Good wastewater treatment efficiency means a higher content of pollutants in the sludge, so it is necessary to ensure that its life cycle ends with environmentally friendly processing and safety for human health.

Table 1. Wastewater treatment efficiency (TE) estimation (E) for 2016

| PTMs and POPs | Sampling campaign, N° | Concentration, mgL ⁻¹ | | TE | |
|---------------|-----------------------|----------------------------------|------------|------|-----------------------|
| | | Inlet | Outlet | E, % | E, gday ⁻¹ |
| Cd | 1. | [0.0003] | [0.0003] | n.e. | |
| | 2. | 0.00068 | [0.0003] | 77.3 | 26.4 |
| Cr | 1. | 0.026 | [0.005] | 90.4 | 1,532 |
| | 2. | 0.049 | [0.005] | 94.7 | 2,330 |
| Cu | 1. | 0.060 | [0.01] | 91.7 | 3,584 |
| | 2. | 0.18 | [0.01] | 97.1 | 8,776 |
| Hg | 1. | 0.00011 | 0.000016 | 85.5 | 6.1 |
| | 2. | 0.0080 | 0.000042 | 99.5 | 399.4 |
| Ni | 1. | 0.021 | 0.0056 | 73.4 | 1,005 |
| | 2. | 0.033 | 0.0067 | 79.1 | 1,310 |
| Pb | 1. | 0.018 | [0.003] | 91.7 | 1,075 |
| | 2. | 0.060 | [0.003] | 97.4 | 2,934 |
| Zn | 1. | 0.24 | 0.10 | 58.5 | 9,147 |
| | 2. | 0.61 | 0.065 | 89.0 | 27,257 |
| As | 1. | 0.00093 | 0.00046 | 50.7 | 30.7 |
| | 2. | 0.0018 | 0.00043 | 75.4 | 68.1 |
| Sb | 1. | [0.001] | [0.001] | n.e. | |
| | 2. | 0.0021 | [0.001] | 75.5 | 79.5 |
| Co | 1. | 0.0014 | < 0.0005 | 81.0 | 62.7 |
| | 2. | 0.00094 | < 0.0005 | 71.3 | 29.1 |
| Mn | 1. | 0.057 | 0.015 | 73.8 | 2,740 |
| | 2. | 0.11 | 0.011 | 89.7 | 4,952 |
| V | 1. | 0.0030 | 0.00055 | 81.7 | 160 |
| | 2. | 0.0075 | 0.00032 | 95.6 | 360 |
| Tl | 1. | [0.005] | [0.005] | n.e. | |
| | 2. | [0.005] | [0.005] | n.e. | |
| AOX | 1. | 0.13 | 0.061 | 53.3 | 4,510 |
| | 2. | 0.16 | 0.043 | 72.3 | 5,810 |
| PAHs | 1. | 0.0012 | [0.00005] | 97.9 | 76.6 |
| | 2. | [0.00005] | [0.00005] | n.e. | |
| PCBs* | 3. | [0.000003] | [0.000003] | n.e. | |
| | 4. | [0.00000] | [0.00000] | n.e. | |

< below LOQ; [] below LOD; * The sum of six PCB congeners according to Ballschmitter (PCB-28, PCB-52, PCB-101, PCB-138, PCB-153 and PCB-180)

Assessment of pellets utilization as a BDW

Pellets have useful properties for supplying soil with humus, minerals, phosphorus and nitrogen (Tables 2 and 3), but PTMs and POPs must be taken into account (Table 1). Due to the composition and pH value, the pellets are suitable for application on acidic soils (Table 2). Significantly higher content of organic matter in pellets than prescribed by BDW Decree [16] and nutrient content prevent soil depletion and mineralization (Tables 2 and 3). However, when applying BDW on soil, the limit and critical immission values of soil must not be exceeded [17].

Table 2. Basic properties of pellets as a BDW

| Basic properties | | Decree [16] | | Pellets | |
|-------------------------|---------------------------------|-------------------------------|-----------------|---------|-------|
| Parameter | Unit | Quality class 1 | Quality class 2 | 2016 | 2018 |
| Organic matter (450 °C) | wt.% _{DS} | > 15 | > 15 | 65.2 | 67.1 |
| | gkg ⁻¹ _{DS} | No limits; own specification. | | 652 | 671 |
| Dry mater (103 °C) | gkg ⁻¹ | | | 904 | 913 |
| Moisture (103 °C) | gkg ⁻¹ | | | 96 | 87 |
| pH | - | | | 8.0 | 7.3 |
| Electrical conductivity | mSm ⁻¹ | | | 248 | 271 |
| CaO | gkg ⁻¹ _{DS} | | | 51.0 | 89.5 |
| Bulk density | tm ⁻³ | | | 0.646 | 0.628 |

Table 3. Nutrients content specification

| Nutrients | Unit | Pellets | |
|--|----------------------------------|---------|--------|
| | | 2016 | 2018 |
| Total nitrogen, (N _{org} +NH ₄ ⁺ +NO ₃ ⁻ +NO ₂ ⁻), N | mgkg ⁻¹ _{DS} | 57,468 | 60,080 |
| | mgkg ⁻¹ | 51,951 | 54,853 |
| | kgm ⁻³ * | 33.6 | 34.4 |
| Phosphorus, P ₂ O ₅ | % m/m | 3.7 | 3.3 |
| | | 36,978 | 33,324 |
| Potassium, K ₂ O | | 3,400 | 3,800 |
| NH ₄ -N, soluble in water | mgkg ⁻¹ _{DS} | 2,260 | 2,600 |
| NO ₃ -N, soluble in water | | < 2.5 | 16 |

* on the basis of bulk density of pellets (Table 2)

Characterization of pellets as soil improvers (R3)

Preparation of pellets for use as a fertilizer is a legal recovery operation R3 [8, 11]. According to Slovenian legislation on BDW [16], pellets

as a single waste cannot become a product under any circumstances. Characterization of pellets (limit values for digestate with DS > 20 % by wt.) according to relevant Slovenian legislation for the use of BDWs in agriculture [16], is shown in the Tables 2 to 4. The direct use of digestate as a single BDW for nutrients recovery that does not confirm to the Quality class 1 is prohibited. Pellets are specified as a digestate with Quality class 2 due to the high content of PTMs (Table 4) and can be utilized only for spreading on non-agricultural land [16]: i) fertilization of ornamental plants in horticulture and nurseries, ii) soil improvement in parks, lawns or areas for sports, recreation or leisure, iii) reclamation of clay pits, quarries, degraded industrial areas etc., iv) reclamation of landfills in accordance with the regulation governing the disposal of waste in landfills and v) land reclamation of transport infrastructure.

Table 4. Determination of quality indicators according to the content of pollutants

| Pollutants, mgkg ⁻¹ _{DS} | Decree [16] | | Pellets, BDW | |
|--|-----------------|-----------------|--------------|-------------|
| | Quality class 1 | Quality class 2 | 2016 | 2018 |
| Cd | 1.5 | 3 | 0.82 | 0.88 |
| Cr tot. | 100 | 250 | 90 | 110* |
| Cu | 200 | 500 | 230* | 360* |
| Hg | 1 | 3 | 0.83 | 1.4* |
| Ni | 50 | 100 | 49 | 72* |
| Pb | 120 | 200 | 61 | 66 |
| Zn | 400 | 1800 | 720* | 790* |
| (PAH) ₁₆ ** | 6 | 6 | 1.28 | 1.22 |
| (PCB) ₇ *** | 0.2 | 1 | 0.05 | 0.012 |

* The concentration exceeds the limit value for the Quality class 1. ** The sum of sixteen PAHs according to EPA priority list. ***The sum of seven PCB congeners according to Ballschmitter (PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-153 and PCB-180)

Copper, chromium and mercury are the elements with the highest removal rate and copper and zinc have the highest concentration among the pollutants in wastewater (Table 1), so their presence in sewage sludge is a logical consequence. Results show that copper and zinc are the most restricted factors in evaluating the pellets quality class and, depending on fluctuations regarding the average annual concentrations, chromium, mercury and nickel could be the limiting factors as well (Table 4).

The quantities of metals introduced into the soil per unit of area and unit of time are specified and must be monitored after application. Theoretically, the annual applied quantity of dry matter of BDW of Quality class 2 into or onto the soil must not exceed 20 tonnes per hectare (Table 5) for an average of three years, and the introduction of PTMs must not exceed the limit values referred to in Annex 5 of this Regulation (Table 6). Regarding the contamination with PTMs, spreading of > 20 tonnes of pellets per hectare is not allowed (Table 6) [16]. Results show that copper is the most restrictive parameter in spreading the pellets on the non-agricultural land.

Table 5. Land area demand of non-agricultural land (in ha) for annual pellets production quantity (application per one year), treated as a digestate, Decree [16], Article 26 “use of digestate”

| Theoretically permitted spread quantity in the period of three years is 20 t _{DS} ha ⁻¹ | annual pellets production | |
|--|-------------------------------|-----------------------------|
| | 2016: 4,054 t _{DS} | 2018: 4,192 t _{DS} |
| | Land area demand, ha per year | |
| | 608 | 628 |

Table 6. Land area demand of non-agricultural land (in ha) for annual pellets production, treated as a BDW, Decree [16], Annex 5

| Pollutants, PTMs | Permitted annual-spread quantity g _{DS} ha ⁻¹ [10] | 2016, Land area demand, ha per one year | 2018, Land area demand, ha per one year |
|-------------------|--|---|---|
| Cd | 5 | 665 | 738 |
| Cr _{tot} | 300 | 1,216 | 1,537 |
| Cu | 350 | 2,664 | 4,311 |
| Hg | 5 | 673 | 1,174 |
| Ni | 100 | 1,986 | 3,018 |
| Pb | 300 | 824 | 922 |
| Zn | 1,500 | 1,946 | 2,208 |

The potential for applying pellets on non-agricultural land is high, but it is often a problem to get an environmental permit due to high demand regarding the non-agricultural areas, the monitoring of treated soil and used sewage sludge, so interest for this use is uncertain. Another problem is the seasonal use of fertilizers, while the pellets are generated over the whole year with the highest production rate in the summer (Figure 3). The

third problem is unavailability of appropriately large non-agricultural areas (Table 6) near to the CWWTPL, which would lead to high transport costs and air pollution.

Application of pellets for backfilling as a cover material on landfills and degraded land (R10)

Sewage sludge has a status of biogenic waste in which the dry weight ratio of organic to inorganic constituents is approximately 2:1. Since 2009, in the post-landfill-ban period, Slovenia closed 80 % of municipal waste landfills, so, a lot of stabilized and dehydrated sludge was used as a top-cover of the abandoned landfills. The current landfill regulation [18] allows only the use of BDWs where the quality corresponds to grade 1 or 2 (Table 4), assuring that the landfill has an environmental permit for disposal of those wastes. Reclamation of environmentally degraded areas (quarries, gravel pits, clay mines and old industrial zones) also offers pellets utilization. Sewage sludge, however, is not on the list of permitted wastes for the preparation of artificial soil [19]. Such soil may contain BDW if it has been pre-treated and classified in grade 1 or 2 of environmental quality (Table 4), but up to a maximum of 10 % V/V. According to the legislation, artificial soil intended for backfilling construction land and backfilling of degraded areas after extraction of mineral raw materials after excavation up to 2 m, may contain up to 0.4 wt.%_{DS} of total nitrogen, 2 wt.%_{DS} of TOC and 0.1 wt.%_{DS} of total phosphorus, so, the possible addition of pellets is very small. The leaching properties of pellets, which are also a limiting factor, must also be taken into account [19].

Co-composting (R3)

Adding pellets to other BDW for co-composting with industrial, agricultural, and municipal waste is attractive due to their low moisture (Table 2), ability to absorb water and the relatively high nutrients content (Table 3). However, the required proportion of added

pellets to the mixture for composting is 10 - 20 wt.%, which is a small fraction of the whole available amount (Table 5) and contributes little to the sludge disposal problem.

Assessment of the possibility of direct use of pellets as a fertilizer in agriculture

Management of treated sewage sludge (stabilized and hygienized) as a special BDW is additionally regulated with Slovenian Sludge Decree [7], which summarizes the requirements for the European Sewage sludge Directive [20], with the Slovenian requirements for the content of PTMs being stricter. An extensive European study was carried out in 2011 as part of the FATE-SEES campaign [21] on how the quality of sewage sludge and the legislation on agricultural sludge use in the member states differ and whether the limit values in the Sewage sludge Directive [20] need to be recast. The Joint Research Centre, the European Commission's in-house science service, established an independent screening and a survey of the situation regarding the occurrence and levels of problematic compounds in a pan-European dimension. A total of 61 sewage sludge samples were collected in 15 European countries. The participation of UWWTPs from different countries was voluntary and anonymous to the public. The pan-European survey evaluated 22 minor and trace elements and 92 organic compounds including ingredients of personal care products and pharmaceuticals. With the representative annual sample 2010 the CWWTPL (sample code SLF268) also participated in that campaign.

Compared to other campaign results, it was found that the values for Pellets 2010 were close to the average European values (Tables 7 and 8), except for mercury, which was not reported in the separated campaign's report and the values obtained in Slovenia are given for comparison. It can also be seen that all values for PMTs, even the maximum ones, are below the limit values prescribed with the Sewage sludge Directive [20] (Table 7). Main

conclusions of FATE-SEES campaign are: i) national limit values are in some countries significantly lower compared to the ceilings set by the Sewage Sludge Directive, ii) the monitored concentrations do not justify the introduction of new limit values for the considered PTMs, and iii) there is no scientific evidence to introduce a regulation for classical POPs.

Table 7. Comparison of the FATE-SEES campaign results [21] for heavy metals

| Pollutants, PTMs | FATE-SEES | | | | Pellets 2010* | Sewage Sludge Directive [20] |
|---|-----------|-------|---------|--------|---------------|------------------------------|
| | Min | Max | Average | SLF268 | | |
| Total, mgkg ⁻¹ _{DS} | | | | | | |
| As | < 2.63 | 56.1 | n.e. | n.r. | <20 | - |
| Cd | < 0.09 | 5.1 | 0.9 | | 0.92 | 20 to 40 |
| Cr | 10.8 | 1,542 | 79.8 | | 84 | - |
| Cu | 27.3 | 578 | 257 | | 310 | 1000 to 1750 |
| Hg | 0.1 | 1.1 | 0.4 | | 2.6 | 16 to 25 |
| Ni | 8.6 | 310 | 29 | 50.6 | 54 | 300 to 400 |
| Pb | 4 | 430 | 47.6 | 88.72 | 84 | 750 to 1200 |
| Zn | 200 | 1,200 | 700 | 1,090 | 920 | 2500 to 4000 |

* Results of the Slovenian authorized contractor

Table 8. Comparison of the FATE-SEES campaign results [21] for PAH with CWWTPL values

| Pollutants, POPs | EU data | | | Pellets, ngg ⁻¹ _{DS} | | |
|------------------------|---------|------|---------|--|------|------|
| | Min | Max | Average | 2010 | 2016 | 2018 |
| PAH, ngg ⁻¹ | | | | | | |
| Phenanthrene | 29.9 | 5552 | 644 | 50 | 110 | 64 |
| Anthracene | <4.6 | 724 | 95.5 | <50 | 20 | 14 |
| Fluoranthene | 34.5 | 3217 | 814 | 120 | 180 | 130 |
| Pyrene | 47.2 | 2637 | 698 | 160 | 180 | 160 |
| Benzo(a)anthracene | <4 | 1833 | 438 | 50 | 60 | 100 |
| Chrysene | <4.7 | 2021 | 504 | 130 | 130 | 120 |
| Benzo(b)fluoranthene | <7.6 | 1919 | 601 | 130 | 120 | 240 |
| Benzo(k)fluoranthene | 9.9 | 1048 | 260 | <50 | 100 | 120 |
| Benzo(a)pyrene | 17.9 | 1476 | 370 | <50 | 60 | 42 |
| Indeno(1,2,3-cd)pyrene | 24.2 | 1401 | 342 | <50 | 70 | 73 |
| Dibenzo(a,h)anthracene | <4.6 | 548 | 134 | <50 | 10 | 75 |
| Benzo(g,h,i)perilene | 29.7 | 1335 | 356 | <50 | 50 | <34 |

Tables 7, 9 and 10 show a comparison of current European, US (EPA), German (DE), Austrian (AT), Hungarian (HU) and Slovenian (SI) legislations on pollutant limit values in sewage sludge for use as fertilizer in agriculture (R10). It is clear that the Slovenian legislation on PTMs is more restrictive than in the other countries, which is a major obstacle to the use of sludge in agriculture. On the other hand, these countries have further regulated some POPs [22 - 24], but again very

differently. Regarding the use of sewage sludge in agriculture, there is no uniform policy and strategy among member states regarding the updating of regulatory parameters and their limit values. Sewage sludge Directive [20] has one important instruction: where sludge is used on soils of which the pH is below 6, member states shall take into account the increased mobility and availability of heavy metals in the crop and shall, if necessary, reduce the limit values they have laid down in accordance with Annex I A. Table 10 presents the limiting values for concentrations of PTMs in sewage sludge according to relevant Slovenian legislation. CWWTPL has the potential to apply pellets in agriculture, but: i) it does not have an environmental permission for such pellets utilization, ii) there is a problem with the high concentration of copper and iii) there is no experience with an interested stakeholder in Slovenia with sufficient land area for their application regarding the annual pellet quantity.

Table 9. Comparison of limit values for PTMs content in sludge used for agricultural for some EU member states and for the USA

| Pollutants, mgkg ⁻¹ _{DS} | DE [22] | AT [23] | HU [24] | | EPA [25] | SI Decree [10] |
|--|---|---------|-----------------|------------------|----------|----------------|
| | | | SS ¹ | SSC ² | | |
| Total | | | | | | |
| As | 40 | n.rl. | 75 | 25 | 75 | n.rl. |
| Cd | 50 mg (kg P ₂ O ₅) ⁻¹ | 5 | 10 | 5 | 85 | 1.5 |
| Cr | n.rl. | 400 | 1,000 | 350 | n.rl. | 200 |
| Cr ^{VI} | 2 | n.rl. | 1 | 1 | n.rl. | |
| Co | n.rl. | | 50 | 50 | | |
| Cu | 900 | 400 | 1,000 | 750 | 4,300 | 300 |
| Hg | 1.0 | 7 | 10 | 5 | 57 | 1.5 |
| Mo | n.rl. | | 20 | 10 | 75 | n.rl. |
| Ni | 80 | 80 | 200 | 100 | 420 | 75 |
| Pb | 150 | 400 | 750 | 400 | 840 | 250 |
| Se | n.rl. | n.rl. | 100 | 50 | 100 | n.rl. |
| Zn | 4,000 | 1,600 | 2,500 | 2,000 | 7,500 | 1,200 |
| Tl | 1.0 | n.rl. | | | | |

1 - sewage sludge; 2 - sewage sludge compost

A comparison of values for PMTs and PAH for pellets for 2010, 2016 and 2018 shows that the Hg content was reduced by half, while the other values remained at the same level (Tables 7, 8 and 11). Given the stable quality of pellets, nutrient and organic content, appropriate pH and compliance with EU

legislation and some other EU members (Table 9 and 10), pellets could be used as fertilizer on agricultural land intended for animal feed, but the CWWTPL must obtain the environmental permission for this.

Table 10. Evaluation of pellets for direct use in agriculture - comparison of limit values between the Slovenian Decree [7] and the EU Sewage Sludge Directive [20]

| Fertilizer and soil properties | Unit | Directive 86/278/EEC [20] | | SI Decree [7] | | Pellets, BDW | |
|--------------------------------|----------------------------------|--------------------------------|-----------------|---------------|----------------|--------------|------------|
| | | Soil* | Treated sludge* | Soil | Treated sludge | 2016 | 2018 |
| pH | - | 6 to 7 | n.r.l. | 6 to 7 | n.r.l. | 8.0 | 7.3 |
| Cd | mgkg ⁻¹ _{DS} | 1 to 3 | 20 to 40 | 1 | 1.5 | 0.82 | 0.88 |
| Cr | | Limit value is yet not defined | | 100 | 200 | 90 | 110 |
| Cu | | 50 to 140 | 1,000 to 1,750 | 60 | 300 | 230 | 360 |
| Hg | | 1 to 1.5 | 16 to 25 | 0.8 | 1.5 | 0.83 | 1.4 |
| Ni | | 30 to 75 | 300 to 400 | 50 | 75 | 49 | 72 |
| Pb | | 50 to 300 | 750 to 1,200 | 85 | 250 | 61 | 66 |
| Zn | | 150 to 300 | 2,500 to 4,000 | 200 | 1,200 | 720 | 790 |

* Member states may permit the limit values they set to be exceeded in the case of the use of soil and sludge for cultivation commercial food crops exclusively for animal consumption

Assessment of advanced thermal treatment of pellets

Waste incineration (only recovery operation R1) is one of the possible options for energy recovery and reduction of waste, regulated by the Decree on waste incineration and co-incineration plants [26], following the regulations of EU IPPC Directive [27], preventing different approaches to controlling emissions into air, water or soil. Both, hazardous and non-hazardous waste can be used for energy recovery in accordance with the relevant decree. Based on the review of pellets generation technology on the site of origin, the review of input materials, pellet composition and the review of analytical results of the annual representative samples for the years 2016 and 2018, it was concluded that the pellets were not contaminated with substances, classified with one of the hazard statements, and did not contain any of the labels for additional hazard statements related to the hazardous property of HP15 [8, 11].

For efficient thermal utilization, the most important quality parameter is NCV_{ar} , dependent on OMC and water content. In general, the values for $COD_{removed}$, NCV and OMC are lowest in summer, while in the same season the amount of pellets delivered is the highest (Figures 3 and 5). Results in Table 11 show the basic properties of pellets according to fuel characterization and annual values of basic constituents (organic and ash content), delivered to the potential stakeholder. The cement kiln represents a good sink for the use of alternative fuels due to the specific technology used in the production of cement. From 2008 to 2017 pellets were used for energy and material recovery in a Slovenian cement kiln in an amount of 1 to 3 wt.% with regard to the totally utilized fuels (fossil and alternative ones).

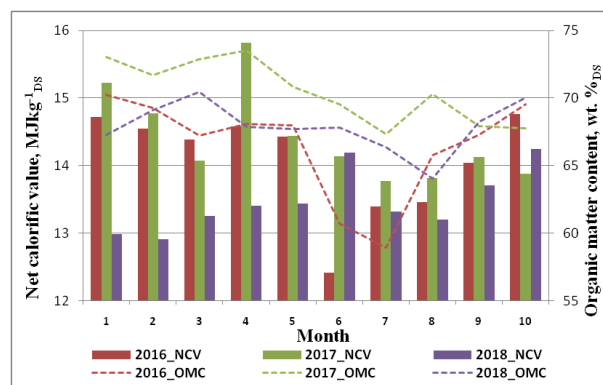


Figure 5. Fluctuations of NCV and OMC content in pellets over the 10-month period

The quality demands for pellets as input material for technological process at cement kiln were: i) max. 10 wt.% of moisture content, ii) min. 10 MJ/kg upper calorific value (as received), iii) pellet diameter d_{50} lower than 3 mm and d_{90} lower than 4 mm, iv) max. 1 wt.% chlorine content, and v) max. 0.5 mg HgMJ⁻¹ (as received). The pellets quality meets the requirements for input control of the cement factory. In addition to the calorific value of the pellets, their mineral composition, that is complementary to the composition of the virgin raw feed, comes to the fore. That is why this type of alternative fuel is not disruptive to the production of cement (Table 11).

Table 11. Proximate and ultimate analysis of pellets as a source of energy

| Basic information about waste to energy utilization, Decree [26], Decree [28] | | Pellets | | |
|---|----------------------------------|---------------------------|-------|---------|
| Basic fuel properties | Unit | 2016 | 2017 | 2018 |
| Moisture (105 °C) | % | 9.2 | 9.7 | 8.5 |
| Net calorific value, NCV* | MJkg ⁻¹ _{ar} | 12.95 | 13.45 | 12.95 |
| Hg* | mgkg ⁻¹ _{ar} | 1.34 | 1.14 | 1.13 |
| Macroelements and mineral components | | 2012 | 2016 | 2018 |
| Si | % m/m _{DS} | 3.3 | 9.4 | 7.6 |
| Ca | | 4.6 | 3.6 | 6.4 |
| Al | | 0.88 | n.a. | |
| Fe | | 0.92 | n.a. | |
| Annual amount delivered to stakeholders | | 2016 | 2017 | 2018 |
| OMC (550 °C) | t _{DS} per year | 2,692 | 2,995 | 2,846 |
| Ash (residue at 550 °C) | | 1,362 | 1,244 | 1,346 |
| TOC | | 1,402.6 | n.a. | 1,496.4 |
| TIC | | 385.1 | | 289.2 |
| N | | 233.1 | | 251.9 |
| S | | 43.0 | | 31.9 |
| Cl (risk of corrosion) | | 4.86 | 4.28 | 3.10 |
| Hg (potential air emissions) | | kg _{DS} per year | 6.89 | 4.84 |

* Average value of 10 representative monthly samples

The study was carried out regarding on co-incineration of coal and Pellets 2012 [29]. Using the laboratory TG method, the comparison of different samples combinations was conducted to predict decomposition temperatures during the heating process in a controlled oxidative atmosphere. It was found that co-incineration led to successful thermochemical conversion of pellets opposed to mono-incineration, and that the co-incineration of coal and pellets was more energy efficient than coal alone because of lower coal decomposition temperature. Most importantly, the addition of pellets to coal shortens the decomposition time [29].

Particularly important is also the information about volatile PTEs and heavy metal content, other than volatile mercury (Table 12), which could cause pollution in the ambient air and consequently in the soil [27]. Additional information about volatile heavy metal content of Pellets 2018, in addition to the data in the Table 12, expressed in mgkg⁻¹_{DS} are: c(Cu) = 350, c(Mn) = 250, c(V) = 20 and c(Tl) = 0.13. But, regarding the non-SRF wastes in Austrian

law [30], it seems only the content of mercury is problematic (Table 12).

Table 12. Assessment of suitability of pellets that would be considered as non-SRF waste under Austrian (AT) law [30]

| Parameter, mgkg ⁻¹ _{DS} | | Sb | As | Pb | Cd | Cr | Co | Ni | Hg | |
|---|-----------------|-----------------|-----|-----|------|------|------|------|------|-------|
| AT demands | 1 | Median | 35 | 10 | 115 | 1.35 | 155 | 7 | 55 | 0.375 |
| | 80th Percentile | 50 | 15 | 205 | 2.7 | 230 | 12.5 | 95 | 0.75 | |
| Pellets, December 2019 | 2 | Median | 2.5 | 3.4 | 56 | 0.79 | 73 | 7.7 | 64 | 1.1 |
| | | 80th Percentile | 2.6 | 3.6 | 57 | 0.83 | 78 | 8.14 | 66 | 1.3 |
| | | Average | 2.4 | 3.4 | 56 | 0.79 | 74 | 7.7 | 63 | 1.1 |
| Pellets 2018 | - | 2.7 | 3.3 | 66 | 0.80 | 100 | 6.8 | 75 | 1.8 | |

1: limit values apply to wastes that are incinerated at the location where it was generated or at other nearby locations; 2: statistical evaluation of 10 daily representative samples for the month of December 2019

Slovenian legislation does not prescribe a limit value for mercury in the waste intended for co-incineration, but only for emissions of substances into the air [26]. For the purpose of efficient flue gas cleaning, it is not enough to control emissions of substances into the air, but also to manage the mass balance of critical pollutants, so, knowing the properties of alternative sources, input and output quantities of pollutants, this is extremely important (Tables 11 and 12).

SRF production [28]

Only non-hazardous waste can be used for SRF production. The classification system for SRF is based on three important parameters, referred to the main SRFs properties: i) heat content (NCV) as an economical/functional parameter, ii) chlorine content - a technical/operational parameter (indicating possible corrosion problem) and iii) mercury content - an environmental parameter/potential air pollution (as a ratio Hg/NCV).

All SRF parameters are chosen to give the stakeholder an immediate but simplified insight into the fuel in question. In Table 13, the statistically evaluated results of the characteristics from ten consecutive monthly composite samples of pellets in the years 2016,

2017 and 2018 are shown. Slovenian legislation also included sulphur and cadmium as additional control parameters, however without specified class limit values (Table 14). Sulphur is one of the main components of sewage sludge, but with regard to the limit value (Table 14) it is a critical parameter and this is an unnecessary obstacle to use pellets as a SRF.

Table 13. Specification of pellets as a SRF: determination of classification classes [28]

| Parameter | Unit | Time period | Average | median | 80th percentile | SRF classification classes |
|------------------|---------------------|-------------|---------|--------|-----------------|----------------------------|
| NCV, as received | MJ/kg _{ar} | 2016 | 12.950 | n.r. | | NCV 4 |
| | | 2017 | 13.451 | | | NCV 4 |
| | | 2018 | 12.952 | | | NCV 4 |
| Cl, dry solid | wt.% DS | 2016 | 0.107 | | | CI 1 |
| | | 2017 | 0.101 | | | CI 1 |
| | | 2018 | 0.075 | | | CI 1 |
| Hg, as received | mg/MJ _{ar} | 2016 | n.r. | 0.089 | 0.120 | Hg 4 |
| | | 2017 | | 0.075 | 0.095 | Hg 3 |
| | | 2018 | | 0.087 | 0.095 | Hg 4 |

Table 14. Additional regulated SRF quality parameters in Slovenia [28]

| Parameter | Unit | Time period* | Limit value | Pellets, SRF |
|-----------|----------------------------------|--------------|-------------|--------------|
| | | | | Average |
| Cd | mgkg ⁻¹ _{DS} | 2016 | ≤ 2 | 1.15 |
| | | 2017** | | 0.87 |
| | | 2018 | | 1.15 |
| S | wt.% DS | 2016 | ≤ 0.2 | 1.06 |
| | | 2017 | | - |
| | | 2018 | | 0.76 |

* January to October; ** January to June

Class codes for the produced SRF are: i) 2016 and 2018: NCV 4; CI 1; Hg 4, and ii) 2017: NCV 4; CI 1; Hg 3. The most challenging issue is changing the classification number of pellets from 19 08 05 (waste sludge) to 19 12 10 (combustible waste, SRF), according to EU legislation. Despite that, this is still a waste according to Slovenian legislation, but pellets are an interesting combustible material. In the case of the CWWTPL, this goal has not yet been achieved. In addition, there is another

obstacle for the recycling of non-hazardous waste into solid fuel because all types of SRFs can only be used in the mixture with at least two SRFs, while the addition of another SRF is at least 3 % V/V. Regarding the binding and renewed Technical Standard EN 15359:2011, Solid recovered fuels, Specifications and classes, there are no obstacles in producing a single SRF from sewage sludge. Indeed, it provides unambiguous and clear classification and specification principles for SRF. The latter can, according to Article 6 of the Waste Framework Directive (2008/98/EC) [14], cease to be waste at community or national level if certain criteria are fulfilled. There are a number of obstacles to the use of SRF in Slovenian legislation. Although in accordance with the requirements of the technical standard regarding SRF [28] the quality of pellets is within the required limit values (class code from 1 to 5), but this does not apply to Slovenian legislation prohibiting the use of SRF if it is classified as: i) 5th class according to NCV, ii) 3rd, 4th or 5th class according to the chlorine content, iii) 2nd, 3rd, 4th or 5th class according to the mercury content. In addition, the use of SRF is only permitted in power plants if the bulk of the SRF is represented as: i) vegetable waste from agriculture and forestry, ii) vegetable waste from the food processing industry, iii) wood waste from industrial wood processing, iv) municipal wood waste and wood waste from construction and demolition, or v) waste cork. The picture is completely different in neighbouring Austria, where the evaluation of SRFs differs in terms of their use and other quality parameters, while the values for NCV are not prescribed and for the content of cadmium and mercury in sewage sludge higher limit values are set, which makes sense when taking into account the properties of sewage sludge [30] (Table 15).

According to Slovenian legislation [28], the rules regarding the use of SRF do not apply in case the waste is used as fuel at the place of its generation. The latter means that the CWWTPL could have its own combustion plant, where the pellets could be used for

energy utilization in the mono-incineration process.

Table 15. Assessment of pellets as SRF under Austrian law [30]

| Parameter, MJkg ⁻¹ _{ar} | Sb | As | Pb | Cd | Cr | Co | Ni | Hg | | | |
|---|------------------------|-----------------|---------------------|------|------|------|---------------------|------|------|-------|------|
| AT demands | 1 | Median | 7 | 2 | 20 | 0.23 | 25 | 1.5 | 10 | 0.075 | |
| | | 80th Percentile | 10 | 3 | 36 | 0.46 | 37 | 2.7 | 18 | 0.15 | |
| | 2 | Median | 7 | 2 | 15 | 0.17 | 19 | 0.9 | 7 | 0.075 | |
| | | 80th Percentile | 10 | 3 | 27 | 0.34 | 28 | 1.6 | 12 | 0.15 | |
| | 3 | Median | 7 | 1 | 15 | 0.17 | 19 | 0.9 | 7 | 0.075 | |
| | | 80th Percentile | 10 | 1.5 | 27 | 0.34 | 28 | 1.6 | 12 | 0.15 | |
| | 4 | Median | Limit values 1 or 2 | | | 0.8 | Limit values 1 or 2 | | | 0.15 | |
| | | 80th Percentile | | | | 0.95 | | | | 0.25 | |
| | Pellets, December 2019 | 5 | Median | 0.17 | 0.23 | 3.77 | 0.05 | 4.91 | 0.52 | 4.36 | 0.08 |
| | | | 80th Percentile | 0.18 | 0.24 | 3.89 | 0.06 | 5.36 | 0.56 | 4.47 | 0.11 |

1: cement kiln; 2: power plant, the proportion of fuel heat output from the incineration of SRF in the total heat output is limited to a maximum of 15 %; 3: conventional co-incineration plants; 4: Cd and Hg limit values for sewage sludge and paper fibre residues; 5: statistical evaluation of 10 daily representative samples for the month of December 2019

Sewage sludge ash (R5, R10)

The ash from special mono-incinerators of sewage sludge has an interesting composition, particularly the content of nutrients (phosphorus and potassium) (Table 3), as well as alkaline earth metals (calcium) (Table 2) and other mineral components (Si, Al and Fe) (Table 11). On the other hand, the content of non-volatile matter and heavy metals in ash during combustion almost triples. A potential option for ash utilization is blending with cement to produce green concretes [31] (the recovery procedure R5). From the environmental and economic point of view, the mentioned material utilization is very interesting, because heavy metals are immobilized in concrete, and at the same time inorganic substances are recovered, either as an aggregate or filler, or as potential pozzolanic substances (Table 11). The ash is an interesting substitute for phosphorus-potassium fertilizer and additionally a good

conditioner for acidic, mineral-depleted soils. But not all useful ash components are bioavailable, so new economically profitable technologies must be developed to free the nutrients from sewage sludge ash. On the laboratory level, the study was carried out regarding the water solubility of nutrients in resulting Pellets 2012 residue after their exposure to oxidative thermal load [32]. It was found that the highest water solubility has the residue generated at 450 °C. At higher temperatures of the oxidizing thermal treatment, the nutrients are chemically transformed to a shape which is not water-soluble anymore, thereby reducing the possibility of nutrients recovery of residues. So, the use of ash as a mineral admixture of artificially prepared soils [19] also has a certain possibility (Table 11).

Sewage sludge pyrolysis (R3/R1)

Pyrolysis is an anaerobic thermochemical decomposition processes for recovery of organic substances and nutrients from waste. The study regarding the water solubility of generated residues after Pellets 2012 thermal treatment [32] revealed that the pyrolysis offers a much greater chance of material utilization of sewage sludge. It was discovered that the water solubility of phosphorus in biochar, produced at 450 °C, is higher when compared to the residues from the oxidizing atmosphere; on the contrary, the water solubility of K and Mg is lower. Another pyrolytic experiments with pellets produced at CWWTPL have been done [33, 34]. The resulting volatile products were fractionated and isolated into separate groups of substances with different chemical properties (recovery procedure R3). Gaseous phase, continuously extracted from the reactor, was burnt concomitantly (procedure R1), while the liquid phase was condensed to a light oily fraction (water/organic condensate) and a heavy fraction (further referred to as bio-oil). By the test on semi-pilot scale 15 wt.% of non-condensable volatile matter (gaseous phase), 15 wt.% of water condensate, 20 wt.% of bio-oil and 50 wt.% of solid residue - mineralized

pyrogenized residue (biochar) were obtained [33]. Most water resulting from the moisture, chemically or crystalline bounded water and water generated during the thermal decomposition of the sludge, is collected in the light oily fraction, and contains a high concentration of condensed water-soluble substances. The yield of the most valuable product, bio-oil, is low because of the low content of macromolecules and their poor quality (oxidized and degraded organic matter). The pyrolysis oil needs to be further refined and water must be completely removed from it to make it useful as a fuel. The solid pyrolytic residue contains 30 wt.% of carbon and has a calorific value of about 10 MJkg⁻¹. It is low value, so it would not have a good price in the secondary fuel market.

According to Guidelines for a sustainable production of biochar [35], it is potentially useful as a fertilizer for non-agricultural land, reclamation of degraded surfaces, and for the preparation of artificial soil. It could be used to supplement depleted soil to increase fertility, porosity and water retention. However, according to the legislation, the content of copper, mercury and nickel is problematic, and their concentration increases due to the volatile and decomposed proportion of pellets, but the concentration of mercury decreases (Table 16). Biochar pH value is very attractive for preventing the soil acidification.

Table 16. Comparison of pH and heavy metal content (in mgkg⁻¹_{DS}) in pellets and biochar

| Fertilizer properties | pH | Cr | Cu | Hg | Ni | Pb |
|-----------------------|-------|-----|------------|------------|------------|-----|
| SI Decree [7] | n.rl. | 200 | 300 | 1.5 | 75 | 250 |
| Biochar [34] | 11 | 29 | 610 | 0.082 | 150 | 160 |
| Pellets 2012 [34] | 7.8 | 120 | 350 | 1.8 | 86 | 93 |

Assessment of thermal treatment residues as a product according to Waste Decree [8]

The list of pollutants to be considered in the residues of pellets thermal treatment to prevent environmental pollution and protect human

health is set out in the IPPC Directive [27], which includes a list of environmental quality standards and special pollutants prescribed by the EU Water Framework Directive [1], and is summarized in the latest version of Slovenian Waste Decree [8]. The relevant regulation determines when a recovered substance or material may lose its waste status.

The condition that the processed substance to be used in the external environment and become a product is the appropriate quality of its leachate (Table 17) and the guaranteed market or completion of the entire chain from waste treatment to customer. The list of parameters for leachate analysis is determined by the authority in the process of obtaining environmental permission. It is clear that the boundary between waste and product is a very thin line and the way waste is valued as a product is more of an obstacle on the path to a circular economy than an incentive.

Table 17. Assessment of residues as a product and comparison of limit values prescribed with other relevant decrees - permissible levels of pollutants in the leachate of the treated waste (W) or material (M) at ratio L/W(M) = 10 L/kg [8]

| Pollutants | Limit values, mgkg ⁻¹ _{DS} | | | Potential product, mgkg ⁻¹ _{DS} | | |
|------------|--|-----------------------------------|--|---|--|-----------------------------------|
| | Waste Decree [8] ¹ | Landfill Decree [18] ² | Artificial soil Decree [19] ³ | Sewage sludge ash, 900 °C [31] ⁴ | Green concrete, 900 °C [31] ⁵ | Biochar, 450 °C [34] ⁶ |
| AOX (Cl) | 0.5 | n.rl. | 0.3 | n.a. | | |
| PAH | 0.01 | | | n.a. | | |
| DOC | n.rl. | 500 | n.rl. | n.a. | | 349 |
| Sb | 0.3 | 0.06 | | 0.0055 | n.a. | n.a. |
| As | 0.1 | 0.5 | 0.3 | 0.0053 | | 0.051 |
| Cd | 0.025 | 0.04 | 0.03 | 0.026 | | < 0.002 |
| Co | 0.03 | n.rl. | | n.a. | | |
| Cr | 0.5 | 0.5 | 0.3 | 5.7 | 0.0031 | 0.013 |
| Cu | 0.5 | 2 | 0.6 | 0.021 | n.a. | 0.025 |
| Hg | 0.005 | 0.01 | 0.01 | n.a. | | |
| Mo | 0.5 | 0.5 | n.r. | 36 | 0.017 | 0.35 |
| Ni | 0.4 | 0.4 | 0.6 | < 0.001 | n.a. | 0.0037 |
| Pb | 0.5 | 0.5 | 0.3 | < 0.002 | | 0.0039 |
| Zn | 2 | 4 | 18 | 0.61 | | 7.3 |

¹ In areas without protection regimes according to water regulations; ² Landfill for inert waste; ³ Backfilling of building land and backfilling of mineral resource areas to fill the soil after excavation, for input up to 2 m; ⁴ Residue of pellets ignition at 900 °C; ⁵ Green concrete produced from sewage sludge ash generated at 900 °C; ⁶ Biochar produced at 450 °C with a TG apparatus

CONCLUSION

The major problem regarding the final pellets utilization is the lack of facilities for energy and material recovery from this type of waste in Slovenia. Certain provisions in Slovenian legislation regarding sewage sludge management are more restrictive than the basic EU legislation and from some Member States as well. This obstacle for successful sewage sludge energy and material utilization must be considered and appropriately eliminated. Treated sewage sludge in the form of pellets can be used as a fertilizer, but only for spreading on non-agricultural land and pellets could be applied on up to 4,300 hectares in one year, depending on the annual production of pellets. Regarding “waste to energy” philosophy, the specification of pellets as an alternative solid fuel according to EN 15359 resulted in “NCV 4; Cl 1; Hg 3-4” class. According to newest legislation regarding the waste management, a product status for residues generated in combustion and pyrolysis of pellets was not achieved. The possibility of using ash obtained from sludge incineration by the recycling of phosphorus, inorganic construction materials and the recovery of inorganic materials in the form of backfilling and conditional R10 (landfill covering) have become relevant. Given the identified possibilities of final pellets utilization and the importance of residues, mono-thermal treatment (in oxidative or inert atmosphere) on the state level is the most optimal solution. It should be carried out within the Slovenian network of facilities and end consumers.

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