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PRODUCTION OF SOIL IMPROVERS FROM WWTP BIOLOGICAL SLUDGE AND INORGANIC BY-PRODUCTS

Dissertation in the ambit of the Integrated Master's in Environmental Engineering, supervised by Professor Margarida Maria João de Quina and Master Andreia Filipa Ferreira dos Santos, presented for Dissertation in the University of Coimbra, Chemical Engineering Department

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Production of soil improvers from WWTP biological sludge and inorganic by-products

A dissertation for graduated study in MSC program for Master of Science in Environmental Engineering.

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A vida natural? O homem pode aguentar-se na vida natural, ou é na vida artificial que está a felicidade? Vestido ou nu? É para a ilusão e a mentira que devem tender os nossos esforços, e a verdade em osso será imagem da inferioridade e da desgraça?

Raul Brandão, in As Ilhas Desconhecidas, 1926.

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At last to all the friends I met during this journey. With them I have been through incredible and joyful moments, that will always mark me.

Abstract

Sewage sludge (SS) production from wastewater treatment plants has been increasing, which raises concerns related to its management. In addition, regarding the circular economy perspective, innovative and sustainable management strategies are required. One of these strategies is the incorporation or production of fertilizers based on SS due to its high organic matter content and nutrients (N and P) that can have a positive impact on soil fertility. However, the requirements for the production of an SS-based fertilizer must be established to ensure that its application to the soil is safe.

This study aims to develop a fertilizer based on SS and other industrial waste, evaluating its interaction with soil and plants. A statistical analysis was conducted about the different agronomic properties that SS can improve after its application to the soil, and it was concluded that SS has a good agricultural potential. To improve the SS potassium content and to ameliorate the heavy metals immobilization, different formulations with coal fly ash and eggshell were developed. The biological stability of the amendments was evaluated measuring the oxygen uptake rate (OUR), which is an important parameter for agronomic applications. Both formulations with dried SS at 100 and 130 ºC were tested, the best results were achieved with the highest temperature. Phytotoxicity essays with several liquid/solid ratios were also conducted, and the germination index was determined. Through the germination index, it was possible to conclude that the samples with SS dried at 130 ºC showed lower phytotoxicity, while the commercial organic mineral fertilizer (OMF) presented the worst results with an extremely toxic effect. Pot experiments with *Lepidium Sativum* L. were carried out, where the effect of the different mixtures and the OMF on the soil and plants was analysed. However, this assessment must be repeated in order for a reliable conclusion to be drawn.

Keywords: sewage sludge; soil improvers; biological stability; phytotoxicity; circular economy.

Resumo

A produção de lamas de ETAR tem vindo a aumentar, o que gera preocupação com a sua gestão. Para além disto, há a necessidade de se aplicar estratégicas de gestão mais inovadores e sustentáveis, tendo em vista a economia circular. A incorporação ou produção de fertilizantes a partir de lamas de ETAR, pode ser uma destas estratégicas, uma vez que as lamas apresentam um elevado teor de matéria orgânica e de nutrientes (N e P), que poderão dar um contributo positivo à fertilidade do solo. No entanto, devem ser estabelecidos os requisitos para a produção de um fertilizante à base de SS, de modo a garantir que a sua aplicação no solo é segura. Neste estudo pretende-se desenvolver um fertilizante a partir de lamas de ETAR e avaliar a sua interação com o solo e com as plantas. Inicialmente, foi desenvolvida uma análise estatística sobre diferentes propriedades agronómicas que podem ser melhoradas após a aplicação das lamas no solo, e concluindo-se que as lamas apresentam um potencial agronómico de interesse. De forma a melhorar o conteúdo de potássio e a imobilização de metais pesados, foram desenvolvidas diferentes formulações com cinzas volantes de carvão e casca de ovo. A taxa de consumo de oxigénio foi determinada para avaliar a estabilidade biológica dos corretivos com o objetivo de decidir a sua aplicabilidade no solo. Foram testados corretivos com lamas secas a 100 e 130 ºC, sendo que os segundos apresentaram os melhores resultados. A toxicidade das amostras foi avaliada com base no índice de germinação para várias razões sólido/líquido. Assim, concluiu-se através do índice de germinação, que os corretivos à base de lamas secas a 130 ºC são os menos fitotóxicos, enquanto que o fertilizante organo-mineral comercial apresentou os piores resultados, com um efeito extremamente tóxico. Por fim, realizaram-se testes em vaso com *Lepidium Sativum* L., nos quais foi avaliado o efeito das diferentes misturas e do fertilizante organo-mineral no solo e nas plantas. No entanto, esta avaliação deve ser repetida para que se possam obter conclusões confiáveis.

Palavras-chave: Lamas de ETAR; corretivos de solos; estabilidade biológica; fitotoxicidade; economia circular

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ACRONYMS

AWSSC – Agricultural Waste and Sewage Sludge Compost
CFA – Coal Fly Ash
DEHP – Di(2-ethylhexyl) phthalates
DS – Dry Solids
ES - Eggshell
FA – Fly Ash
FBBR – Fluidized Bed Bioreactors
GBF – Granular Medium Biofilters
GHG – Greenhouse Gases
GI – Germination Index
LAS – Alkyl-benzene sulphonates
MBBR – Moving Bed Biofilm Reactor
MF – Mineral Fertilizer
OM – Organic Matter
OMF – Organic-mineral Fertilizer
OMW – Olive Mill Solid Wastes
OUR – Oxygen Uptake Rate
PAH – Polycyclic Aromatic Hydrocarbons
PBBR – Packed Bed Bioreactors
PCB – Polychlorinated Biphenyls
PCCD – Dioxins
PTM – Potentially Toxic Metals
RA – Respirometric Activity
RBC – Rotation Biological Contactors
SRI – Static Respiration Index
SS – Sewage Sludge
WPPA - Wet Process Phosphoric Acid

1. INTRODUCTION

1.1. Motivation

Projections estimate that the world population will stand between 9.4 to 10.1 billion in 2050 (Nations, 2019). This factor combined with higher urbanization and industrialization will generate a large volume of wastewater that requires treatment in wastewater treatment plants (WWTP), which forms a considerable quantity of sewage sludge (SS). Indeed, the SS handling may represent 50% of total operating costs in WWTP. This fact highlights the need to develop sustainable strategies for reusing waste materials or taking advantage of their energy content (Spinosa, 2011), considering possible economic and environmental benefits. European Union (EU) has been aware of the issues related to SS management, which lead to the development and implementation of several legislative frameworks regarding or involving this matter (EEC, 1991; European Parliament and Council, 2018; Council of European Communities, 1986).

According to 2nd out of 17 UN Sustainable Development Goals SDG) the "*end hunger, achieve food security, and improved nutrition and sustainable agriculture*" must be promoted. This target may be accomplished using fertilizers in a balanced way, since they are necessary to maintain global crop productivity at current levels (Roberts, 2009). Indeed, nitrogen (N), phosphorous (P), and potassium (P), which are the key elements most rapidly uptaked from the soil by plants, are mainly supplied by rock and/or synthetic fertilizers. However, due to intensive agriculture, the increase in the price of fertilizers, along with tensions on fossil energy and P markets, reliance on synthetic fertilizers can have negative consequences diminishing crop yields (Brunelle et al., 2015). In this scope, SS can have a relevant role in the future of the fertilizers and soil conditioners industry. As many researchers reported, SS can ameliorate the properties of soil, both from the physical and chemical point of view, as a result of the organic matter (OM) and nutrients (e.g., N, P, and K) in its composition (Kirchmann et al., 2017; Kominko et al., 2019; Kelessidis and Stasinakis, 2012).

Agricultural SS application is not a consensual option within the scientific community, since heavy metals, organic contaminants (polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB), dioxins (PCCD), di(2-ethylhexyl) phthalates (DEHP), alkyl-benzene sulphonates (LAS), etc.), and pathogens may be present in its composition. These pollutants in high quantities may have negative impacts on humans and the environment, causing some technical and social issues (Kacprzak et al., 2017; Yang et al., 2018; Bettiol and Ghini, 2011). The solution of environmental problems is complex, and sometimes perverse results can be achieved (Norton, 2012). Generally, these kinds of decisions are associated with long periods, thus affecting unpredictable future contexts, that can be completely different from the one where the decision was made. Furthermore, some factors to be considered are quantitative, like the produced amount of SS, while others are qualitative, with intrinsic subjectivity, such as public acceptance (Bertanza et al., 2016). In fact, there is no management solution that is arguably the best one. For example, the incineration is not aligned with the circular economy regarding cutting of CO_2 emissions and nutrient recycling, mainly organic components and nitrogen (Kominko et al., 2018). Besides, incineration is also an energy-intensive process, involving high-costs (Świerczek et al., 2018). Landfilling is a well-known and consolidated management option but its operation may provoke emissions to the air, soil, and water (European Commission, 2001), public acceptance is low while the costs associated are high in some countries (Mininni et al., 2015). Additionally, the European Waste Framework Directive (Directive 2008/98/EC, 2008) considers that landfill waste disposal should be the last option to be considered in waste management.

Rockström et al. (2009) presented a new and ambitious concept of *Planetary Boundaries*, with the objective of evaluating a safe operating space for humanity concerning the functioning of the Earth System. There are 9 boundaries defined, among which “Interference with the Global Phosphorous and Nitrogen”. Human interference has already provoked the global N cycle’s boundary breach, in particular, due to the growth of fertilizer use in modern agriculture (Rockström et al., 2009; Sayers, 2015). More recently, was also reported that the safe limit to the P cycle has been breached (Steffen et al., 2015). P challenges are many, since it is a nonrenewable resource that is mainly extracted from phosphate mines and, at the ongoing amount of use, the USA are expected to run out their reserves within the next generation, becoming dependents on imports (Jones et al., 2020). Furthermore, P along with

N can lead to eutrophication, caused by the inappropriate use of mineral fertilizers, resulting in the loss of it to the environment (Scholz et al., 2013). Achieving a minimization of the P loss is essential, and its cycle must be closed e (Kominko et al., 2017).

Despite all the obstacles mentioned above, it is important to use SS properly as a resource. When applied to soil and crops, it can improve soil structure and porosity, water retention, increase cation exchange capacity as well as the enzymatic function (Siebielec et al., 2018; Gomes et al., 2019). Considering the positive repercussions expressed above, the valorization of SS in the soil can be a sustainable, cost-effective, and environmentally acceptable method of disposal.

1.2. Aim and objectives

This thesis aims to investigate the requirements for producing a fertilizer based on sewage sludge (SS) formed in wastewater treatment plants and its interaction with the soil and plants. Thus, the studies will involve a statistical analysis conducted to determine the composition of SS regarding the most important parameters that may have impact on its potential as a fertilizer. Despite that, biological parameters, such as stability and phytotoxicity, were assessed for both SS and the produced fertilizers to evaluate its agronomic potential. Finally, pot experiments with garden cress were conducted to predict the plant behavior after the incorporation of this type of soil improvers.

1.3. Structure of thesis

This thesis is organized in six chapters:

- Chapter 1: Introduction, which presents the problematic related to excess SS production and nutrients management.
- Chapter 2: Theoretical background, where a brief description of the current state of wastewater treatments, SS production, characterization and management solutions are indicated. This chapter explores also the fertilizers trends and the possibility of incorporate SS in fertilizers. Moreover, it presents the most relevant legislation to the issue under analysis and the importance of SS biological stabilization.

- Chapter 3: State of the Art, resumes of the main characteristics of the production of fertilizers based on SS. Survey previous studies regarding the conditions applied during respirometric analysis and pot experiments.
- Chapter 4: Materials and Methods, where all the all the materials and experimental procedures are described.
- Chapter 5: Results and Discussion, corresponding to the main part of the thesis. The experimental results are organized and discussed.
- Chapter 6: Conclusions and Future Work, which summarizes the most important conclusions of the experiments and suggestions for future work.

2. THEORETICAL BACKGROUND

2.1. Wastewater treatment plants

Wastewater treatment plants (WWTP) include several processes in which wastewater is depurated aiming at reintroducing water into its cycle, minimizing the damage to the environment. The processes in WWTP are based on a combination of physical, chemical, and biological technologies, which create a synergistic effect between them. Commonly, WWTP can include four steps: preliminary, primary, secondary, and tertiary treatments. The last-mentioned treatment is applied more rarely than the former ones since high costs are standardly required.

Figure 2.1 presents the typical scheme of a WWTP. Preliminary treatments intend the removal of larger suspend solids such as rags, sticks, floatables, grift, and grease. This step aims to prevent downstream operations problems and mechanical equipment damage. Primary treatment is constituted by physical technologies and aims for the removal of suspended solids, which may contain organic matter. Secondary treatment involves biological and chemical processes, and the main objective is the removal of the biodegradable organic load through microorganisms that consume the organic matter present in the mixed liquor, and then the removal of the secondary biological sludge. The removal of organic matter can be accomplished by fungi and bacteria, under aerobic and/or anaerobic conditions. In this stage, the oxidation of most of the biodegradable soluble organic matter occurs, which is mediated by microorganisms subsequently removed by secondary sedimentation (Samer, 2015). Tertiary treatment usually consists of filtration with membranes or disinfection for the removal of residual suspends solids (such as bacteria) or other specific pollutants (Metcalf and Eddy et al., 2014).

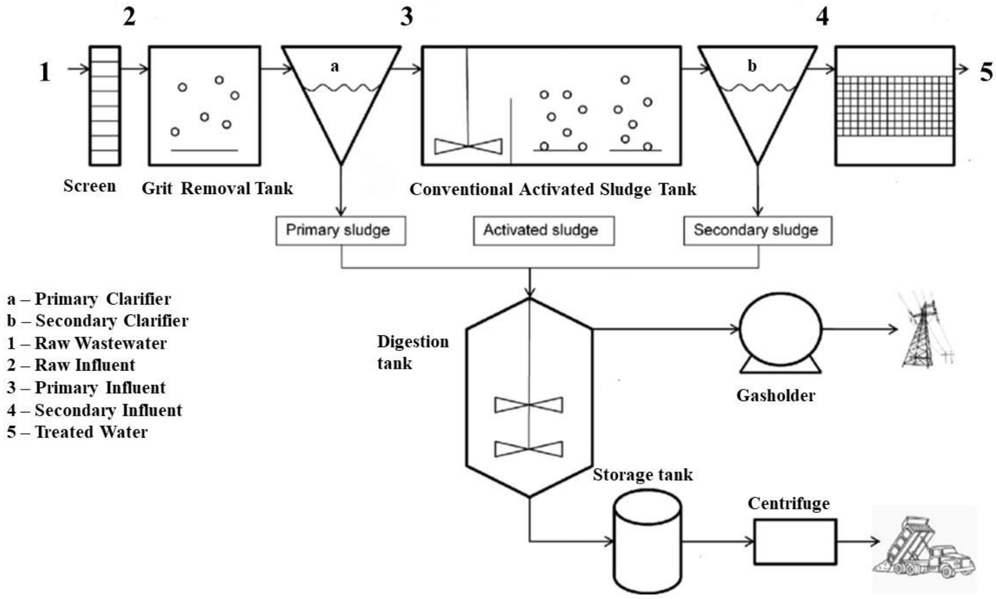


Figure 2.1. Typical wastewater treatment plant processes (adapted from Berthod et al., 2016).

The WWTP in Portugal includes mainly primary followed by secondary treatment, as can be seen in Figure 2.2. Biological processes can be divided into two categories: suspended biomass (in which the microorganisms are kept in constant suspension in the liquid amendment) and attached biomass (in which the microorganisms are fixed into a solid). Many biological treatments can be efficient in removing organic matter through suspended biomass such as aerated lagoons, membrane bioreactors, aerobic digestion, and nitrification processes. However, the activated sludge (AS) process is the most common treatment worldwide (Leyva-Díaz et al., 2013). Some attached biomass processes examples are the moving bed biofilm reactors (MBBR), rotation biological contactors (RBC), fluidized bed bioreactors (FBBR), packed bed bioreactors (PBBR), and granular media biofilters (GBF) (Sonwani et al., 2019).

As already mentioned, the AS is typically used in municipal and industrial wastewater treatment. It consists of aerobic microorganisms that are held in suspension by mixing and aeration systems inside of an aeration reactor. The organic matter or other constituents present in the wastewater are converted into cell tissue and/or gases (Metcalf and Eddy, 2014).

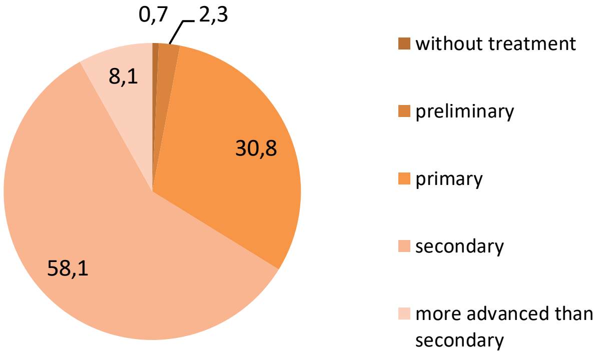


Figure 2.2. Treatment level in mainland Portuguese WWTPs in 2016 (Agência Portuguesa do Ambiente 2019).

2.2. Sewage sludge production

Sewage sludge (SS) is formed during the wastewater treatment all over the world. Regional sludge production depends both on the number of inhabitants and the development of wastewater treatment. Indeed, different regions will have different sludge production rates as well as distinct needs in terms of sludge management (LeBlanc et al., 2006).

The implementation of the Urban Waste Water Treatment Directive 91/271/EEC (EEC, 1991) led the European countries to improve their wastewater collecting and treatment systems. As a consequence, an increase in SS production was reported, from 6.5 million tons of dry solids (DS) in 1992 to 10.9 million tons DS in 2005 (Kelessidis and Stasinakis, 2012). Both Milieu et al. (2010) and Kelessidis and Stasinakis (2012) confirmed that the current specific sewage sludge production at EU-15 is estimated in about 25 kg/(PE·year), which represents 68 g/(PE·day). Mininni et al. (2015) estimated an amount between 66-84 g DS/(PE·day) before sludge processing. Figure 2.3 demonstrates the analysis of yearly per capita sewage sludge production in Europe.

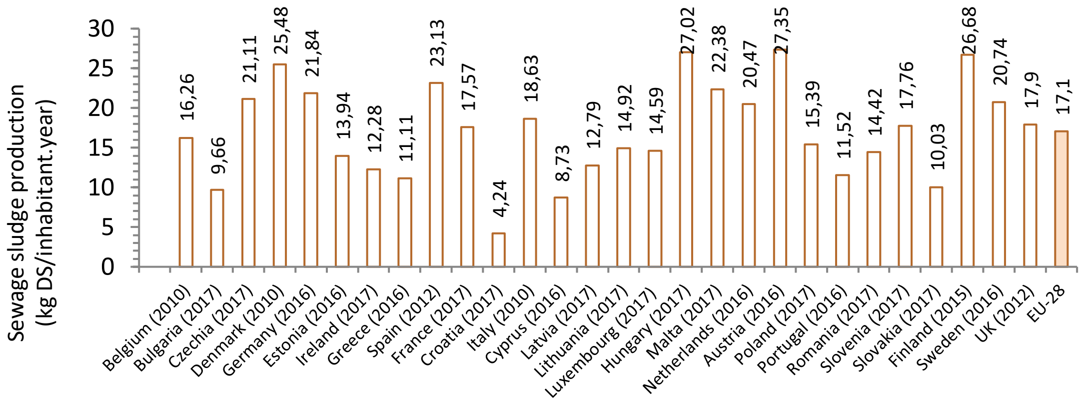


Figure 2.3. Sewage sludge production in Europe (data source: Eurostat).

In Figure 2.3 it is possible to conclude that Croatia has a very low yearly per capita SS production since it was the country joining European Union in 2013, having less time to implement the Urban Wastewater Treatment Directive. The lower SS productions in Croatia lead to a decrease in the EU-28 yearly per capita SS production.

The sustainability is far from being achieved since the EU management system for SS data shows relevant and structural lack of homogeneity and reliability. A clear and standard definition of key-terms, the annual frequency of European and national reports about SS management, and the definition of a standard protocol for data management at a national level are some possible amelioration, among others, to improve the quality of SS information (Pellegrini et al., 2016).

2.3. Sludge composition

The origin of the wastewater, the treatments adopted, and the time and storage conditions are some of the diverse aspects that may affect sludge composition and characteristics (Chen et al., 2012). The primary sludge is composed of settleable solids removed from the treated wastewater in primary settling, and it has a high organic load, which originates from high putrescible conditions. The raw primary sludge is grey and has an extreme fecal odor, conditions that can intensify under prolonged storage beneath anoxic conditions, once putrefaction rapidly initiates (Scholz, 2015).

Secondary sludge includes microorganisms, grown on the biodegradable matter, endogenous residue, and inert solids not removed in the primary settling. Many WWTPs in Portugal, mix primary and secondary sludge, mainly due to economic reasons, generating a type of sludge referred to as mixed sludge (Foladori et al., 2015). The appropriated sludge management may involve Zhou et al. (2014):

- **Thickening:** Developed to separate free bulk water from sludge solids. It aims the reduction of sludge volume to be treated by subsequent processes. Air flotation, biological flotation, centrifugation, flat-sheet membrane filtration, and gravity thickening are some of the options to promote the thickening process;
- **Stabilization:** Targeted to degrade the unstable organics and to inactivate pathogens and odor. Usually achieved through aerobic or anaerobic digestion, or by adding chemicals such as lime;
- **Conditioning:** Boosting the dewaterability of waste activated sludge using physical disruption or the addition of chemicals including flocculants, acid, ferric chloride, and lime;
- **Dewatering:** The main objective is to reduce the water content by mechanical (e.g., press filters and centrifuges) or thermal dehydration (e.g., dryers);
- **Final Disposal.**

As reported by Scholz (2015), digested sludge is the product of either aerobic or anaerobic digestion that is a well-stabilized material. Table 2.1 presents the main properties of the different types of SS.

Table 2.1. Composition of different types of sludge (Andersen, 2002).

Components	Unit	PS (HL)	BS (LL)	BS (L/ML)	MS	DS
Dry Matter (DM)	g/L	12	9	7	10	30
Volatile Matter (VM)	%DM	65	67	77	72	50
pH	-	6	7	7	6.5	7
C	%VM	51.5	52.5	53	51	49
H	%VM	7	6	6.7	7.4	7.7
O	%VM	35.5	33	33	33	35
N	%VM	4.5	7.5	6.3	7.1	6.2
S	%VM	1.5	1	1	1.5	2.1
C/N	-	11.4	7	8.7	7.2	7.9
P	%DM	2	2	2	2	2
Cl	%DM	0.8	0.8	0.8	0.8	0.8
K	%DM	0.3	0.3	0.3	0.3	0.3
Al	%DM	0.2	0.2	0.2	0.2	0.2
Ca	%DM	10	10	10	10	10
Fe	%DM	2	2	2	2	2
Mg	%DM	0.6	0.6	0.6	0.6	0.6
Fat	%DM	18	8	10	14	10
Protein	%DM	24	36	34	30	18
Fibres	%DM	16	7	10	13	10
Calorific Value	kWh/t DM	4200	4100	4800	4600	3000

Note: PS (HL) - primary sludge, primary sludge with physical/chemical treatment or high pollution load; BS (LL) - biological sludge (low load); BS (L/ML) - biological sludge from clarified water (low and middle load); MS - mixed sludge; DS - digested sludge

2.4. Sludge management solutions

Sludge management is an issue of growing importance in developed countries. In the last years, European regulations, legislation, programs, and development strategies have focused on the promotion of sustainable management of sewage sludge. The strategy leads to a replacement of storage management methods that guarantee SS stabilization and safe recycling (Cieślik et al., 2015). Recovering valuable components from SS is possible following several routes, namely through the its application as soil amendment or heat and energy recovery. According to Evans (2016), the main goals of SS management are the reduction of volume (facilitating handling and reducing transport costs), pathogens, and odors (diminishing smell stress in the population). These targets are achieved through intermediate and final technologies. Intermediate technologies were already mentioned in the last chapter, once mechanical dewatering, biological treatment, chemical treatment, and thermal drying are essential to the sludge treatment steps. Agricultural valorization, land recuperation, incineration and landfilling are some of the most common final treatments (Gomes et al., 2019). In EU the preferred method for SS disposal is agricultural use, but some countries such as the Netherlands, Germany, or Belgium adopt incineration (Kelessidis and Stasinakis, 2012).

The regulations of the EU discourage landfilling of wastes by applying strict restrictions to this activity, which turns sludge reuse alternatives as a positive path since the demand for space and undesired impacts of the landfill (gas and leachate) are reduced (European Parliament and Council, 2018; Valderrama et al., 2013). It is possible to accomplish SS agricultural valorization by direct application or through compost. The composting process is based on the aerobic biological conversion of the material, with proper amounts of air and moisture, into a stabilized product often referred as “compost” (Muñoz et al., 2018). The quality of the compost depends on the subtract used, as well as the operating parameters of the composting process. Besides, if SS contains high concentration of heavy metals, organic pollutants, and pathogenic organisms, the utilization of the produced compost may be hampered namely in agriculture (Ozaki et al., 2017). However, the incorporation of other materials in fertilizer production may reduce the concentration of these contaminants, curtailing the possible harmful effects (Kominko et al., 2018).

Incineration is a well-established and reliable technology, in which organic matter is oxidized mainly to CO_2, H_2O and other trace gases. This method reduces significantly the volume, ensures the destruction of pathogens and toxic organic compounds as well as allows energy recovery. Nevertheless, there are some disadvantages, such as the requirement of previous dewatering the SS, increasing the cost of operation. Moreover, after incineration there are ashes that require further management(Raheem et al., 2018). Different authors have studied co-incineration of SS in coal-based power plants and demonstrated that it is possible to reduce the high cost of the incineration technology, minimize greenhouse gases (GHG) emissions, and optimize energy retrieval efficiency along with the public acceptance of the technology (Morais et al., 2011; Hong et al., 2013). However, the emissions of mercury (Hg) should be taken into account (Sun et al., 2020).

In the last years, other types of SS management solutions have emerged, such as pyrolysis, gasification, development of bio-plastic and bio-pesticides, or the incorporation in construction materials, among others (Tyagi and Lo, 2013; Raheem et al., 2018). Once SS management is a global problem, there is a need for different solutions because different world regions have distinctive requirements. Globally, the most important aspect is environmental protection and the safeguarding of human health.

2.5. Fertilizers

2.5.1. Categories and main properties of fertilizers

The main function of fertilizers is to provide plants with nutrients to grow. According to Decreto-Lei n.º 103/2015, there are primary macronutrients (nitrogen, phosphorus, and potassium), secondary macronutrients (calcium, magnesium, sulphur, and in some cultures sodium and silicon), and micronutrients (boron, cobalt, copper, iron, manganese, molybdenum, and zinc). Micronutrients are essential to plant growth in smaller quantities compared to the other two types and can be phytotoxic if applied in excess. Some authors consider carbon, hydrogen, and oxygen as macronutrients. These elements are not considered by the legislation because plants can access them through the air (respiration) and water, while the other elements are assimilated from the soil. Overall, N, P, and K are considered the main nutrients since plants request its presence in larger quantities. Thus, the fertilizer grade expresses the content of each of these elements in percentage, by the order N, P expressed as its oxide (P_2O_5), and K expressed as its oxide (K_2O) (Jones Jr., 2012). The definition of P and K as oxides is associated with the common practice of presenting inorganic elements as oxides.

Even though nitrogen constitutes nearly 79% of the atmosphere, it is in the elemental form (N_2), which is unavailable to most green plants. Nitrogen is available to plants when present in the soil in soluble forms, such as ammonium (NH_4^+), or following the nitrification process, as nitrate (NO_3^-). The last two forms are highly soluble, which can cause eutrophication and decay in water bodies, or even converted to nitrous oxide gas (greenhouse gas). In natural ecosystems, nitrogen can be converted into different forms, some of them associated to complex microbial decay from organic matter (plant and animals within the soil or over its surface) (Hillel, 2008). Figure 2.4 summarizes the nitrogen cycle, where eight flows are indicated. The biochemical function of nitrogen is to combine with carbohydrates, forming amino acids and proteins that will participate in enzymatic processes (Jones Jr., 2012).

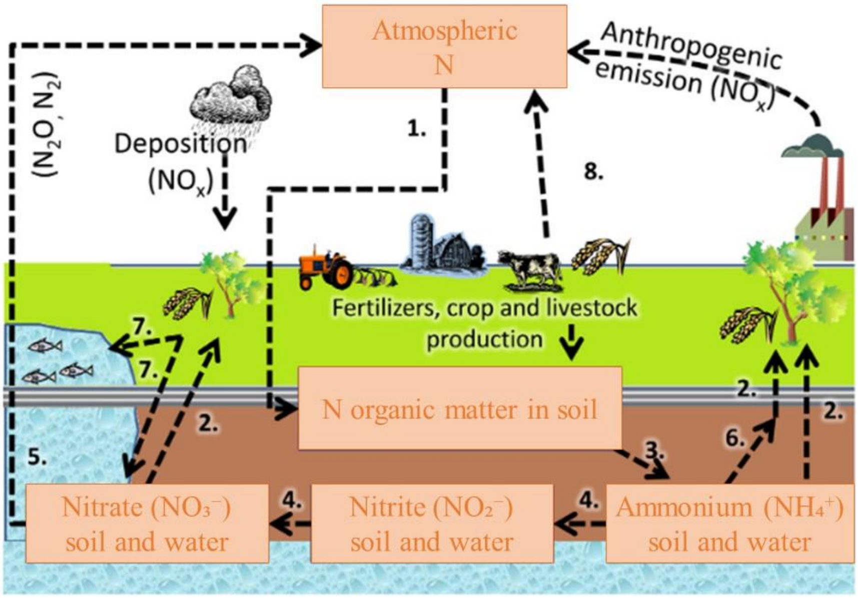


Figure 2.4. The nitrogen cycle: (1) uptake of nitrogen by plants from the atmosphere, (2) uptake of ammonium and nitrate by plants from soil and water, (3) ammonification, (4) nitrification, (5) denitrification, (6) nitrate immobilization by soil sorption, (7) nitrate leaching from the soil, (8) release of ammonia (NH_3), gaseous nitrogen and nitrous oxide to the atmosphere. (Bednarek et al., 2014).

Phosphorous is fundamental for life on earth. For example, it can be found in deoxyribonucleic acid (DNA) and is an important constituent of adenosine triphosphate (ATP), involved in the biological energy transfer reactions. Therefore, a lack of phosphorus reduces plant productivity and slows its maturation. Currently, the major source of phosphorous is the phosphate rock, which is a non-renewable resource (Scholz et al., 2013). Usually, P is present in fertilizers as orthophosphate anion (PO_4^{3-}), dihydrogenphosphate anion ($H_2PO_4^-$), or as hydrogenphosphate anion (HPO_4^{2-}), and its availability for root absorption is a complex process (Jones Jr., 2012). European Commission (2013) states that the major routes for P losses include soil erosion and leaching, as well as an inefficient use of manure, biodegradable waste, and wastewater. These losses should be considered to avoid phosphorous shortage and promoting an efficient way to use this element.

Potassium occurs naturally in the soil when minerals such as feldspars (particularly orthoclase) and micas are present. However, when weathering is intense and prolonged, potassium leaches away, and the soil will require the addition of K through a fertilizer (Hillel, 2008). Potassium is accessible in fertilizers in the form of potassium cation (K^+), and it is involved in osmotic potentials of the plant, balancing anions, controlling membrane permeability, and electro potentials.

Fertilizers can be classified according to its composition or its physical aspect. According to Koli et al. (2019), the physical classification of fertilizers is: solid (e.g., crystals, briquettes, granules, etc.) or liquid, which will determine the storage and the method of application.

The classification of fertilizers based on the Portuguese legislation (Decreto-Lei n.º 103/2015) is:

- Straight/elemental fertilizer: only supply a primary plant nutrient, specifically N, P, or K (nitrogenous, phosphate, and potassic fertilizers);
- Complex fertilizer: contain two to three primary plant nutrients and its obtained through a chemical reaction, by solution, or by granulation in its solid-state. Each granule must contain all the nutrients in its declared composition;
- Mixed fertilizers: mixtures of straight fertilizers elaborated through manual mixing of dry elemental fertilizers, without chemical reaction;
- Compost: the sanitized and stabilized product, resulting from decomposing OM by composting, whose characteristics are beneficial, directly or indirectly, to plant growth;
- Soil amendment: the fertilizer matter, whose main function is to improve physical, chemical and/or biological characteristics, with perspective of a good plant development.

Mineral, organic, and organo-mineral are the terms used to identify the provenance of the materials used in fertilizer processing. The first one declares the nutrients presented in the mineral form. Cyanamide calcium, urea, and the products from the respective condensation and association, as well as fertilizers that contain chelated or complexed micronutrients, can be considered according to this classification. Decreto-Lei n.º 103/2015 states that organic fertilizers are those whose nutrients are totally from vegetal and, or animal sources. On the other hand, organo-mineral are the ones that occur by mechanical mixing of mineral and organic compounds and have at least 1% of organic nitrogen. When in contact with the soil, fertilizer reactivity is determined by both fertilizer and soil factors, Table 2.2.

Table 2.2. Factors influencing the reactivity of fertilizers (Jones Jr. 2012).

Fertilizer Factors	Soil Factors
Physical form	pH
Solubility	Texture
Elemental composition	Organic matter content
Chemical reactivity	Mineral composition
Method of application	
Time of application	

Phosphorus as a critical nutrient

Phosphorus can be seen as a strategic raw material because its world demand is constantly increasing due to the fundamental role in agriculture and food safety. Schröder et al. (2010) pointed out that almost 90% of phosphate (mined P) demand is applied in food production, 82% goes to the development of fertilizers, and a smaller fraction goes to animal feed additions (7%) and food additives (1-2%). The surplus is used in different industries (detergents, oils, pharmaceuticals, and others), and with a growing population, the required quantity of P will expand. Another issue is the geopolitical context, since the phosphate ores are not homogeneously distributed around the globe, with almost three quarters (74% - explored and noun explored) of these deposits located in Morocco. The reserves in Morocco are mainly from sedimentary rocks with a medium level of radioactivity and a medium-high level of hazardous metal contents. Thus, it is unprofitable to exploit the whole of the reserves, which may change with the scarcity and variation in prices. Along with Morocco, Russia, China, and the USA are the principal countries where phosphate is mined, and only about 10% of Europe’s demand has an internal source, Finland (Boer et al., 2019). Figure 2.5 contextualizes the top five countries with explored P reserves. As aforementioned, P is a non-renewable resource, which is the principal reason for being considered a strategic raw material. The scientific community does not have a precise prevision of when the global P resources will run out. Some estimations claim that it will not happen for 200 years (Tan and Lagerkvist, 2011), others claim that it will happen in 100-130 years (Li et al., 2014), and there are even worst evaluations.

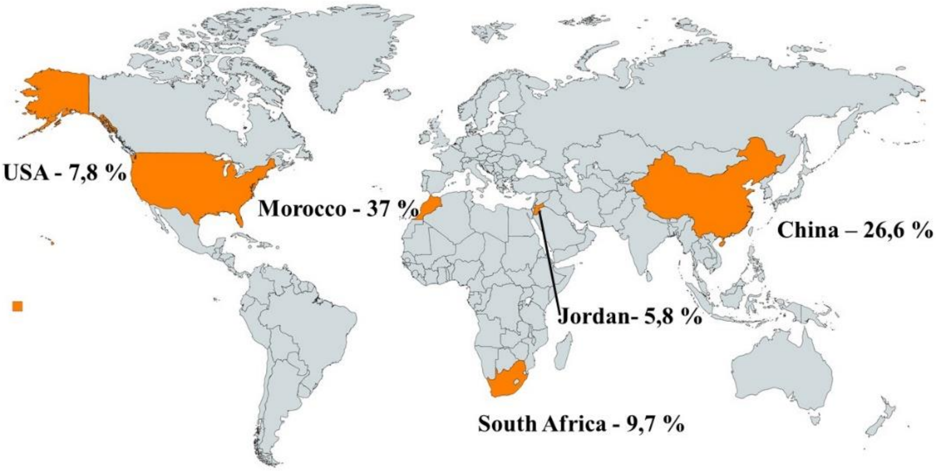


Figure 2.5. Distribution of the top five countries containing phosphorus reserves. (adapted from Rosemarin et al., 2009).

From all the quantity applied of P in agriculture, only a small quantity reaches the consumer mainly because losses during use, which demonstrates the inefficient, non-circular, and dissipative utilization. A significant part of P leaks to ground and surface waters (provoking eutrophication), accumulates in soils, sediments, and on harvested biomass. Beyond all this, there are still big losses from wasted food (Chojnacka et al., 2020). Figure 2.6 frames all the limiting factors considering P availability for productive use in food production.

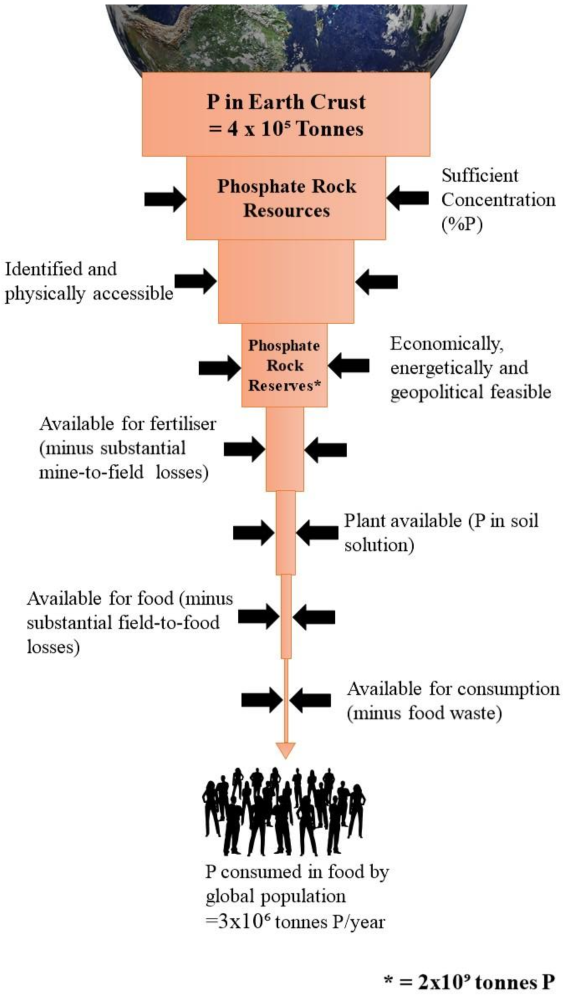


Figure 2.6. Physical, economic, social, and ecological limiting factors for phosphorus availability on the production of food (adapted from Schröder et al., 2010).

The efficiency of P handling can be improved by practicing recycling strategies through the reuse of animals, food, and human wastes. Some good examples of materials with significant amounts of P are slaughter waste (limited by legal constraints), SS or SS ashes (Chojnacka et al., 2020). Bone meal is the waste that contains the highest concentration of P, but is produced in a smaller quantity than SS (Cieślik and Konieczka, 2017).

The easiest way to recover phosphorus from SS is the direct application in soil, without any treatment, but some disadvantages may arise from this management route. The drying process may have a positive influence on SS management since an increases on the bioavailability of P can be observed (Li et al., 2014). Cieślik and Konieczka (2017) highlighted two possible methods for P recovery from SS:

- **direct and from leachates -** the precipitation of phosphoric minerals can occur as struvite, hydroxyapatite, or calcium phosphate. These minerals can contain low heavy metals concentrations and the slow solubilization in soil decreases the possible eutrophication effects. On the other hand, the possibility of transporting pathogens and potential organic pollutants in these materials stands out as the main disadvantages to the environment. Finally, the P obtained can be more expensive compared to regular fertilizers.
- **ashes obtained from thermal treatment** – Ashes incorporate higher amounts of P than SS due to the expressive volume reduction. Typically, SS presents 1 to 5% of P, while ashes contain from 5 to 11%. One of the main drawbacks is that incineration is an expensive technology for SS. Moreover, the bioavailability o P in ashes is limited.

The significant challenge associated with P recovery is to achieve an affordable market price to the consumers. Since each processing step involves additional efforts (labour, energy, chemicals), and it will be reflected in the final price (Kabbe, 2019).

2.5.2. Industrial production of chemical fertilizers and the current demand

Chemical fertilizers can be defined as inorganic materials, with defined composition, that are applied to soil to provide it with nutrients encouraging crops growth (Koli et al., 2019). The first-ever chemical fertilizer, superphosphate, was developed by John Bennet Lawes, an English agricultural scientist that, in 1842, begun the chemical fertilizer industry by patenting a process of treating phosphate rock with sulfuric acid to make the phosphate soluble (Hillel, 2008). At the beginning of the 20th century, just before World War I and aiming the production of explosives, Haber discovers how to synthesize ammonia from air and water, by combining elemental nitrogen with hydrogen under high pressure and temperature, then Bosch refined and industrialized the process (Hillel, 2008). Before the discovery of the Haber-Bosch process, farmers depended on animal manures with or without bedding material, plant residues, leguminous N fixation, and others to guarantee a fertile soil in N (Morari et al., 2011). After World War II, the use of mined mineral phosphorous sources grew exponentially, as it was seen as a low cost and productive phosphorus source, helping to achieve the Green Revolution, which generated twice of crop yield and increased per capita nutritional intake (Ashley et al., 2011).

Although a wide range of fertilizers is available on the market, some products are predominant. A few of these common fertilizers are indicated in Table 2.3.

Table 2.3. Common fertilizers available in the market (Kant and Kafkafi, 2013; Agricultural Extension Service, 2015).

Material	Grade NPK	Physical form	Chemical formulae	Handling precautions
Urea	46-0-0	solid (prills, granules, crystalline)	$CO(NH_2)_2$	Good storage and handling properties
Ammonium Nitrate	34-0-0	solid (prills or granules)	NH_4NO_3	Strong oxidizer; readily absorbs water; can be explosive if mixed with carbonaceous materials
Calcium Nitrate	15-0-0	solid (granules)	$Ca(NO_3)_2{\cdot}4H_2O$	Strong oxidizer; store in tightly closed containers in a cool, well-ventilated area
Diammonium Phosphate (DAP)	18-46-0	solid (granule)	$(NH_4)_2HPO_4$	It should be avoided excess dust*
Monoammonium Phosphate (MAP)	11-48-0	solid (granule)	$NH_4H_2PO_4$	It should be avoided excess dust*
Triple superphosphate (TSP)	0-46-0	solid (granule)	$Ca(H_2PO_4)_2{\cdot}H_2O$	It should be avoided excess dust*
Potassium Nitrate	13-0-44	solid (granule or crystals)	KNO_3	It should be avoided excess dust*
Potassium Chloride	0-0-60	solid (granule or crystals)	KCl	It should be avoided excess dust*

*risk of explosion and potential to harm human health.

The approach to produce fertilizers is well established, offering a reliable and consistent production. Figure 2.7 represents a scheme from traditional fertilizer production., usually it begins with a pre-neutralized or partially ammoniated slurry that is formed by reacting together the feedstock ingredients. Then, in a granulation drum, the slurry is converted into granules, while the reaction completes, and a mechanical action is applied in the material. After this, the material is dried in a rotary dryer and the next step is to classify the particles through a screen. On-sized material is moved on to cooling in a rotary cooler, and over-sized will go to a hammer mill for crushing. The cooling stage is essential once caking is prevented, the final product is much more stable, and the risk of heat damage to downstream material is reduced. The crushed over-size material is combined with under-size material from the screen and then reintroduced in the process.

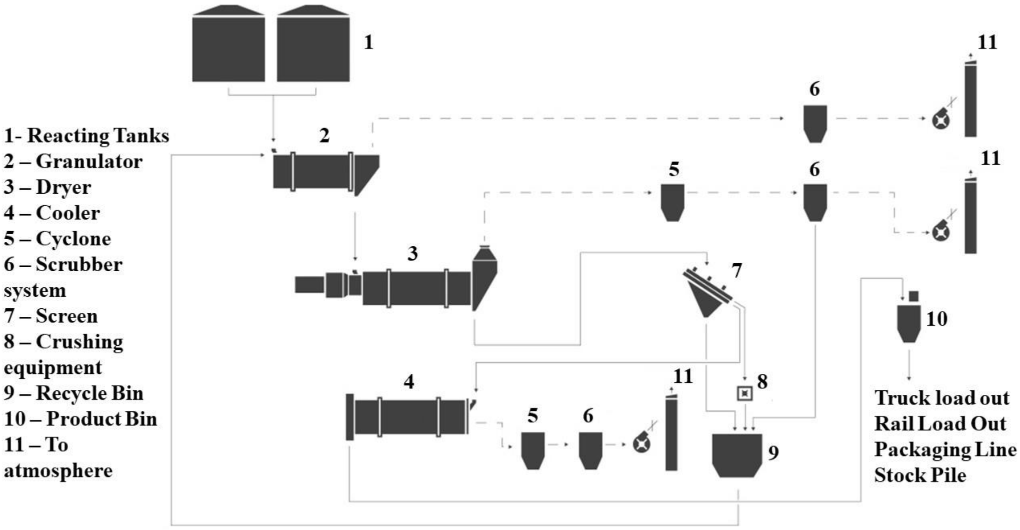


Figure 2.7. Traditional fertilizer process flow diagram (Le Capitaine and Carlson, 2020).

FAO (2019) disclosed that the supply forecast for ammonia, phosphoric acid, and potash, between the years of 2016 and 2022, increased from a total of 244,131 to 269,482 kt. The same happens with the world demand for fertilizer use in the same period, growing from 185,063 to 200,919 kt. Even though the fertilizer industry has a massive and positive impact on the development of modern society and agriculture, it also has some negative environmental impacts, such as TFI (2018) :

- Water – a significant resource in the production of phosphate, potash, and to lesser extent nitrogen.
- Greenhouse gas emissions – carbon dioxide is a by-product of ammonia production and it also occurs in fertilizer transport (rail, truck, waterways, and pipelines), an essential phase connecting the processing facility to the farmer.
- Energy – a lot of energy is required in this type of industry, either in the form of natural gas, fuel, electricity, or steam.
- Land reclamation – a significant physical footprint is associated, especially in the mining sector.

To diminish the negative effects, several measures are taken into account as the use of recycled and reclaimed water, the capture and re-use of CO_2 in the production of urea or the carbonization of soft drinks, energy cogeneration, the use of low-impact energy sources (i.e., solar), among others. Another way to interfere was the launching of the Global 4R Nutrient Stewardship Framework by the fertilizer industry. The framework promotes the application of nutrients using the right source (or product) at the right rate, time, and place to instruct correct management of fertilizer's application to ensure that social, economic, and environmental objectives are achieved (Johnston and Bruulsema, 2014).

2.5.3. Granulation process for fertilizers production

Granulation is a process used to transform fine powders into granular materials with controlled physical properties through a combination of three rate processes: wetting and nucleation; consolidation and growth; and attrition and breakage, Figure 2.8. It is applied in a wide range of industries including mineral processing, agricultural products, detergents, pharmaceuticals, foodstuffs, and specialty chemicals (Reynolds et al., 2005).

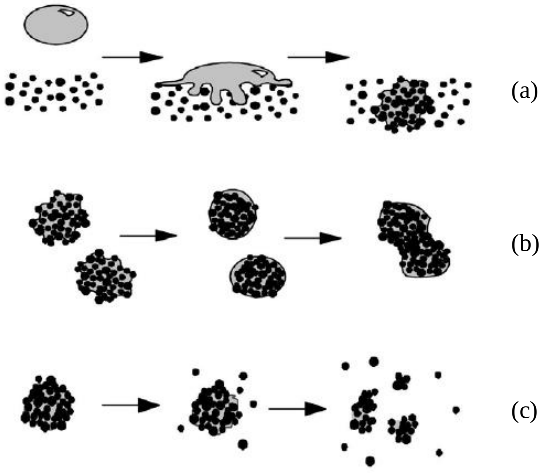


Figure 2.8. Processes in granulation: (a) wetting and nucleation; (b) consolidation and growth, and (c) breakage and attrition (Litster and Ennis, 2004b).

The main objectives of this process concerning fertilizers are to eliminate dust problems during transportation and application, to supply a defined quantity for quantification and distribution, to reduce caking and lump formation, and to eliminate segregating blends of powder ingredients. Granules can be obtained by different techniques and, usually, their size is between approximately 100 µm to 20 mm, while the format can vary from loose aggregates to dense compacts. The preferential techniques applied to fertilizers are tumbling granulators, which included pans/discs and drums, and fluidized bed granulators (Litster and Ennis, 2004b). Figure 2.9 represents a typical disc granulator design as well as the tumbling effect.

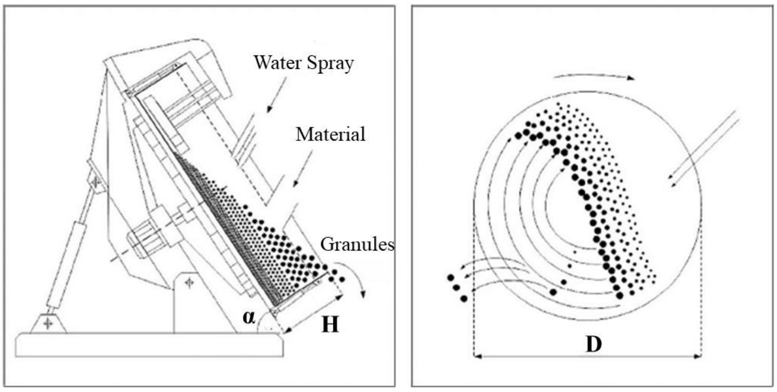


Figure 2.9. Disc granulator with tumbling effect: α is the inclination angle, H is the height of the sidewall and D is the diameter of the plate (adapted from Lieberwirth and Lampke, 2016).

The chemical and physical characteristics of the dust to be granulated, the binder properties, and the granulation process are the principal variables that affect the final product. These variables can be manipulated to obtain different granulation results. The particle size distribution has a huge influence on the characteristics of the formed granules. During granulation, there are different particles that undergo coalescence, forming granules with varying sizes since large and small particles in the raw material come together more easily than two particles of the same size. Nevertheless, wide particle size can difficult the binder infiltration through the layers and do not wet the particles in the lower layers. It lead to the preferential growth of granules containing liquid, resulting in large granules together with primary particles that have not been moistened. Thus, the way that the binder is applied as well as the angle and velocity of the machine (in the case of tumbling granulators) are important factors to a correct granulation (Rodrigues, 2012).

Fries et al. (2011) mentioned that the granulation by fluidized bed granulators is achieved when a liquid suspension or solution is injected into a fluidized bed of particles to attain particle growth by layering or coat the particle surface. The liquid is sprayed homogeneously with the objective of a uniform granule growth. There are five possible configurations, as indicated in Figure 2.10, to fluidized bed granulators:

- Top spray: conventional without draft tube, the nozzle is usually placed at the top outside the moving powder bed and spraying in a descending way;
- Bottom spray: the spray is brought in from a nozzle located at the center and bottom of the bed inside the powder bulk;
- Wurster-coater: is a bottom spray granulator where the bed contains a draft tube to create a circulating flow pattern;
- Rotating fluidized bed: a rotor placed at the bottom of the bed is rotated and the air is fed through a gap between the rotor and the wall, while the liquid is sprayed onto the roping solids flow from a tangential injection nozzle;
- Spouted bed: the fluidization air is introduced from the bottom via adjustable cylinders and the powder enters via a spout at the center, the liquid is injected from a bottom spray nozzle.

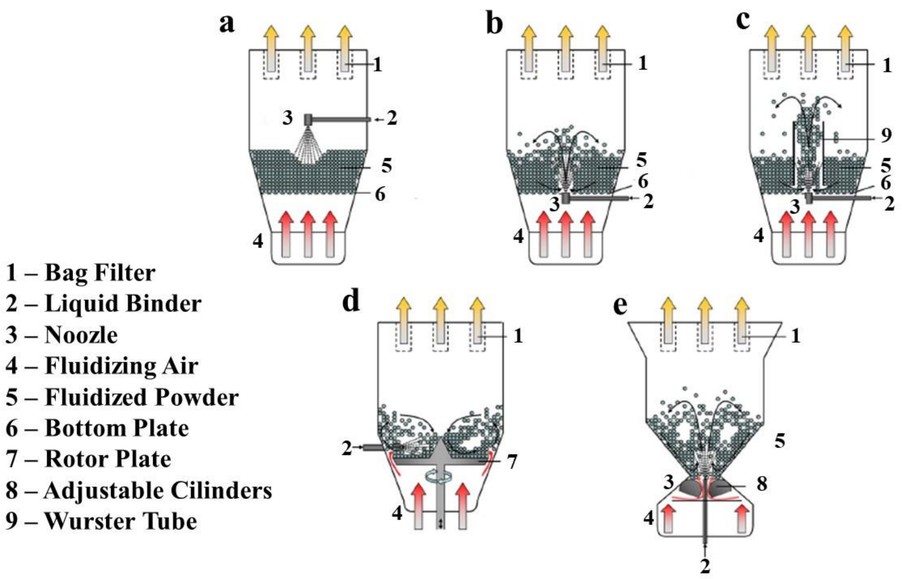


Figure 2.10. Different fluidized bed granulator configurations schematized: (a) top spray granulator, (b) bottom spray granulator, (c) Wurster-coater, (d) rotor granulator, and (e) spouted bed granulator (Fries et al., 2011).

Particles, in tumbling granulators, are impelled by the tumbling action, which leads to segregation and size classification, caused by the balance between gravity and centrifugal forces. These granulators are not suitable for producing very small granules (2 -20 mm) and high porosity agglomerates. However, they are extensively used in mineral fertilizer production because of its large dimensions (up to 4 m in diameter) that enable considerable throughputs. Disc granulators are a tilted rotating disc, with a rim to hold the tumbling granules, that operates between 50 and 75% of critical speed. Critical speed is a key operating parameter defined as the speed at which a granule is just held stationary on the rim by centripetal forces alone, usually with the angle (α) between 45 and 75° (Litster and Ennis, 2004b). The critical speed, N_c (rev/s), is given by equation 2.1, where D is the disc diameter (m) and g the gravity (m/s^2).

$$N_c = \sqrt{\frac{g \cdot \sin \alpha}{2 \cdot \pi^2 \cdot D}} \tag{2.1}$$

The principle in drum granulation is the same as for disc granulators, and there are not many differences in the final products. Drum granulators are the simplest continuous device for the agglomeration of materials. Figure 2.11 shows a tumbling drum granulator comprising a rotating cylinder, with a slight horizontal inclination to facilitate the material transportation over the drum; an inlet dam ring, to minimize back-spill of the inlet material; an exit dam ring, to allow the increase in depth of the bed inside the drum; conveyors, for granulate transportation at the inlet and outlet; and some sprayers to incorporate the binder (Walker, 2007).

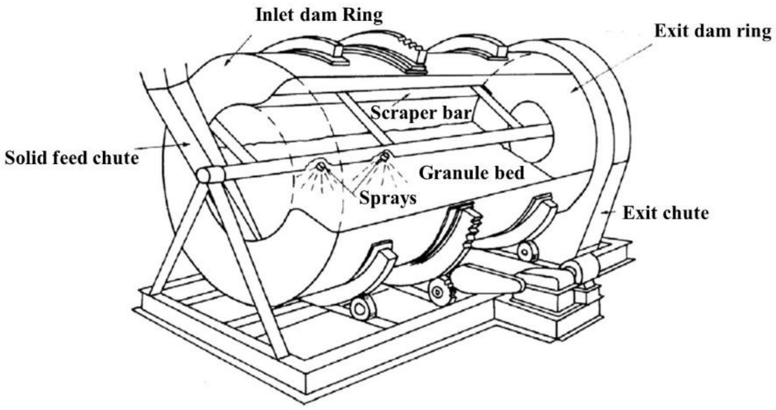


Figure 2.11. Tumbling drum granulator scheme.

2.6. Adjuvants for improving sewage sludge properties

One of the biggest issues of modern society is solid wastes management. The increasing generation of waste potentially represents an environmental burden, but this situation can also represent an opportunity to enhance the circular economy. Indeed, proper waste management can have environmental positive impacts, like a GHG reduction and also in the fertilizers area(Abdel-Shafy and Mansour, 2018). The fertilizer industry always uses ammonia and other by-products, but nowadays this is most urgent with a need to improve the technology by optimizing the used resources, close material loops, and diminish emissions. There are a considerable number of residues that can contain essential nutrients for agriculture applications like post-harvest residues, waste from livestock husbandry, slaughterhouse waste, or food waste (Chojnacka et al., 2019).

Some wastes besides adding nutrients can also have positive effects on the chemical properties of the soil. Ok et al. (2011) reported that eggshells (ES) can be used for heavy metals (Cd and Pb) immobilization, which is beneficial since it reduces their bioavailability. When applied combined with SS, the phytotoxicity of sludge alone decreases and the soil pH can be corrected from acid to neutral. Likewise SS, eggshell is produced in high quantities and can be seen as a raw material for fertilizer production, closing loops (Gomes et al., 2020).

Usually, SS contains low concentration of K, and therefore, there is an interest in combining this material with others to obtain a balanced composition. Different types of materials that can be used to provide K to the soil. Ashes can have different origins (e.g., biomass, coal) and usually have a considerable amount of K in its composition (Aprianti S, 2017; Yunusa et al., 2012). Ashes production in Europe will grow due to directives that point to more sustainable energy production, focusing on renewable sources such as from biomass. Thus, the incorporation into fertilizers can be a way to close the loop of this waste. Besides K, it can be a source of P, Mg, and Ca (Alvarenga et al., 2019). It was proven that coal fly ashes increase the plant uptake of many nutrients and also correct nutrient deficiencies, some of them have high concentrations of alkali and alkaline earth metals (Na, Ca, or Mg) and, as a consequence, high pH. However, not always this affects the neutralization of soil acidity because of the lower buffering capacity in comparison with the soil (Yunusa et al., 2012). Another interesting aspect of ashes is the pozzolanic capacity, which makes them attractive to be used as a binder in the cement production, another possible way to close the loop of this waste (Aprianti, 2017).

2.7. Relevant legislation

The basic EU legislation concerning SS management is the Sewage Sludge Directive (Directive 86/278/EEC), released in 1986, which aims to promote the use of SS in agriculture, along with setting all the requirements and provisions to prevent potential harmful effects on humans, animals, soils, vegetation and surface, and ground-water. Also, it established the limits for potentially toxic metals (PTM), both for the soil and SS (Christodoulou and Stamatelatou, 2016), as Table 2.4 demonstrate.

Table 2.4. Limit values for concentrations of PTM in soil (soil pH 6-7) according to Directive 86/278/EEC and Decreto-Lei n. º 276/2009.

	Directive 86/278/EEC			**Decreto-Lei n. º 276/2009**
Parameters	Soil (mg/kg)[1]	SS (mg/kg)	10-year limit (kg/ha.year)[1]	SS PT (mg/kg)
Cadmium	1 to 3	20 to 40	0.15	20
Copper[2]	50 to 140	1000 to 1750	12	1000
Nickel[2]	30 to 75	300 to 400	3	300
Lead	50 to 300	750 to 1200	15	750
Zinc[2]	150 to 300	2500 to 4000	30	2500
Mercury	1 to 1.5	16 to 25	0.1	16
Chromium[3]	-	-	-	1000

(1) Member States may permit the limit values they fix to be exceeded in the case of the use of sludge on land which at the time of notification of this Directive is dedicated to the disposal of sludge but on which commercial food crops are being grown exclusively for animal consumption. The Member States must inform the Commission of the number and type of sites concerned. They must also seek to ensure that there is no resulting hazard to human health or the environment.

(2) Member States may permit the limit values they fix to be exceeded in respect of these parameters on soil with a pH consistently higher than 7. The maximum authorized concentrations of these heavy metals must in no case exceed those values by more than 50%. The Member States must also seek to ensure that there is no resulting hazard to human health or the environment and to groundwater.

(3) It is not possible at this stage to fix the limit values for chromium.

The Directive 86/278/EEC also has some restrictions for the spreading of SS on grazing and pastureland, and on land in which vegetables and fruits are grown. In Portuguese legislation it is transposed by Decreto-Lei n. º 276/2009, which is stricter since besides the limits already referred, it also restrings the organic compounds and microorganisms presents in sludge and simplifies the national licensing process for the agricultural use of SS. Other important directives related to SS are:

- Directive 91/271/EEC – concerning the collection, treatment, and discharge of urban wastewater and treatment and discharge of wastewater from certain industrial sectors to protect the environment from the mentioned discharges.
- Directive 96/61/EC – proposes to achieve integrated prevention and control of pollution, from different activities, laying down measures designed to prevent or to reduce emissions in the air, water, and land. It is possible to consider agricultural valorisation of sludge in terms of prevention and quantity control.
- Directive 1999/31/EC – aiming the providence of measures, procedures, and guidance to prevent or reduce as far as possible negative effects on the environment from landfills, which should be reduced by 2020. It is transposed into Portuguese Legislation by Decreto-Lei n.º 183/2009.
- Directive 2008/98/EC – that recently was amended by Directive (EU) 2018/851 and specifies measures to protect the environment and human health by preventing or reducing the adverse impacts of the generation and management of waste, reducing the overall impacts of resource use, and improving the efficiency of such use. In this Directive, WWTPs sludge is established as non-hazardous waste and is transposed into national law by Decreto-Lei n.º 73/2011.

It should be highlighted that with the introduction of the waste hierarchy, provided by Directive (EU) 2018/851 (European Commission, 2018), a priority order in waste prevention and management, policy and legislation, shall be applied. This priority order is prevention (e.g., minimization techniques), preparing reuse (e.g., chemical or biological stabilization), recycling, another recovery (e.g., energy), and disposal. The waste hierarchy aligned with the Circular Economy Action Plan (European Commission, 2020a), which includes “Food, water, and nutrients” as one of the seven key targeted value chains, can work in favour of fertilizers produced with SS or its by-products. The focus is to ensure less waste, especially from sectors that use most resources, and where the potential for circularity is high, such as water and nutrients.

The most meaningful legislation concerning fertilizers is Regulation (EU) 2019/1009 that lays down Regulations (EC) No 1069/2009 and (EC) No 1107/2009 while repealing Regulation (EC) No 2003/2003, that regulates especially inorganic fertilizers. In Portuguese legislation, Decreto-Lei n.º 103/2015 ensure compliance, internally, with the provisions of the last-mentioned regulation, it accommodates a broader spectrum of fertilizers, which makes it similar to the most recent legislation.

The implementation of the EU Nitrates Directive 91/676/EEC (Council Directive, 1991), concerning the protection of waters against pollution caused by nitrates from agricultural sources, caused a decrease both in N leaching losses to ground and surface waters, and gaseous emissions to the atmosphere. These phenomena are expected a further decrease since the measures from this directive are likely to become stricter (Velthof et al., 2014). Recently the European Commission announced the preparation of an Integrated Nutrient Management Action Plan (INMAP) that will address nutrient pollution at source and increase the sustainability of the livestock sector, largely because of the excess of nutrients (especially nitrogen and phosphorus) in the environment. The excess of nutrients occurs due to overuse and to the fact that not all nutrients used in agriculture are effectively absorbed by plants, which is an important source of air, soil, and water pollution with different climate impacts. In this way, the Commission will act to reduce nutrient losses by at least 50%, while ensuring that there is no deterioration in soil fertility (European Commission, 2020b).

Finally, it is important to approach that in the document ‘Orientations towards the first Strategic Plan for Horizons Europe’, the largest multinational collaborative research and innovation investment in Europe, is stated that: “A comprehensive EU policy to balance nutrient cycles is not yet well developed. Research and innovation are needed to look at how the EU could move to living within the planetary boundaries, with regard to nutrient flows.” (European Commission, 2019).

2.8. Biological stabilization of sludge for agricultural valorization

To apply SS in soil as a fertilizer it must be ensured that it does not pose a threat to the environment, humans, and animals, namely regarding microbial activity, that can be a source of contamination to crops, water as well as causing odour stress. This can be determined through parameters that describe reactivity/stability of organic matter. Even there are several parameters available, the Respiration Activity (RA), which represents the oxygen consumption, is considered very suitable for the characterization of wastes stability which contain high concentration of organic matter. The *Oxitop*® is one of the possible types of equipment that can be used to determine RA, and it is distinguished from others due to its lower price and compact design (Binner et al., 2012). In this equipment, the type of tests made are static, once that oxygen is not continuously provided, and the oxygen consumption is acquired by the pressure drop, measured by a pressure sensor that belongs to the equipment. Figure 2.12 displays the typical curve for the pressure drop that occurs in a respiration test.

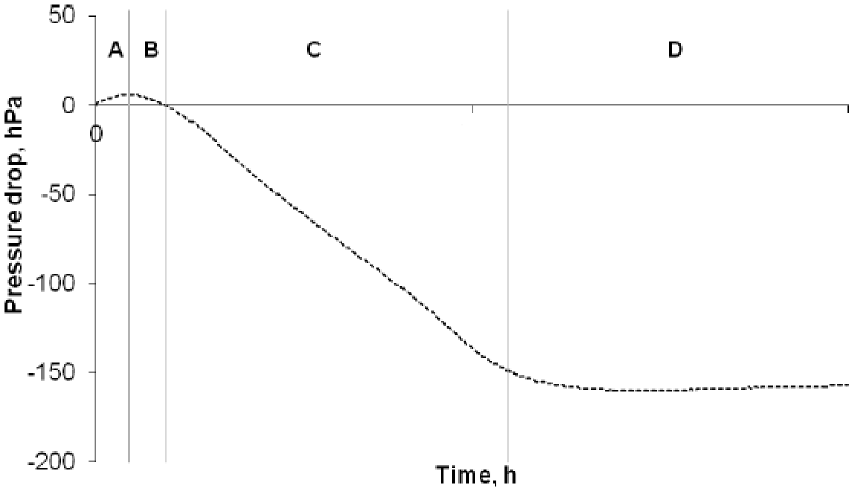


Figure 2.12. The typical pressure drops during a respiration test, with the 4 associated phases (adapted from Malińska, 2016).

From Figure 2.12, it is possible to observe four different pressure phases. The first one (phase A) is due to differences in the temperature of the sample and the incubator, while the second one (phase B) results from a lag period in which microorganisms adapt to the new conditions. After the equilibrium temperature has been reached, and a short adaptation period happens, a gradual pressure drop occurs because of the oxygen consumption (phase C). In the third phase occurs the organic matter biodegradation, where the microorganisms use oxygen and produce carbon dioxide, that is absorbed by NaOH pellets. The fourth and final phase (D) is visible when there is oxygen depletion, limiting aerobic activity from the microorganisms (Malińska, 2016). Equation 2.2 is applied to determine the RA (mg O_2/g OM).

$$RA = \Delta p \cdot \frac{M_{O_2}}{R \cdot T} \cdot \frac{V_v - V_{abs} - V_s}{m_{dm} \cdot OM} \tag{2.2}$$

where, Δp is the differential pressure in (hPa), M_{O_2} is the oxygen molar mass (mg/mol), R is the universal gas constant (mL·hPa/Kmol), T is vase temperature (K), V_v is the vase volume (mL), V_{abs} is the absorbent volume (mL), V_s is the sample volume (mL), m_{ds} is the dry matter mass (g) and OM is the organic matter (%).

As reported by the standard EN 16087-1:2011, the Oxygen Uptake Rate (OUR) should be lower than:

- 50 mmol O_2 /kg OM·h, for digested sludge;
- 25 mmol O_2 /kg OM ·h, for fertilizers;
- 15 mmol O_2 /kg OM ·h, for soil amendments intended to non-professional applications;
- 25 mmol O_2 /kg OM ·h, for soil amendments intended to professional applications,

for considering stable the organic matter in these materials

3. STATE OF THE ART

The first part of this chapter briefly outlines the state of the art of SS based fertilizers. The collected data are summarized in Table 3.1, where the emphasis is given to the utilized materials, measured parameters, and on the main conclusions from all the experiments. The studies from the literature considered in this chapter have the objective of recovery both nutrients and OM (organic matter) from SS, and all of them apply granulation technics.

Kominko et al. (2018) and Kominko et al. (2019) added mineral fertilizers (MF) to improve the nutrient content in the fertilizers produced with SS and different acids (nitric, phosphoric, and sulfuric) to act as a binding agent in the granulation process. (Kominko et al., 2018) concluded that the wet process phosphoric acid (WPPA) introduced considerable amounts of Cr and Cd, exceeding the permissible values established for commercial fertilizers. The organo-mineral fertilizer (OMF) had an NPK ratio of 6% of N, 19.8-25.2% of P_2O_5, and 5.91-6.21% of K_2O. Through a simplified economic analysis, it was shown that the profits from a SS based fertilizer amount to 50.45% compared to conventional fertilizers. The fertilizer from the second study presented an NPK ratio of 7.4-13.2% of N, 3.0-5.6% of P_2O_5, and 9.9-17.6 % of K_2O, and it was concluded that it is possible to adjust fertilizers elemental composition to plant individual needs. Through OMF, it can be recycled annually 82-140 t of P (as P_2O_5) and 42-73 t of N.

Pesonen et al. (2016) applied a granulation method where the utilized binder was water. Granules of fly ash (FA) alone presented a high compressive strength (about 61.3 N), but when the SS was added, in different proportions, this value dropped (>22.4 N). Based on these results, lime was added, but the compressive strength remained low. However, to improve N content, more SS should be used. This fertilizer can be utilized as a forest fertilizer according to the Finish Legislation. Alvarenga et al. (2019) used a similar granulation technique. Pot experiments were carried out, and the results were similar to the granular fertilizers and the fertilizers in bulk form. The used soil was from an acid mine-contaminated site, and the granules were able to correct its acidity and to increase extractable P and K. The granules with a higher portion of sludge caused the best response related to soil enzymatic activities. The selected species for the pot experiments had a low salt tolerance what could justify that a permanent plant cover was not established.

Table 3.1. Literature review in respect to sewage sludge based fertilizers.

Ref.	Term	SS type	Formulations and operation conditions	Measured parameters	Main conclusions
[1]	OMF	Dried municipal sewage sludge	-SS (38 - 42%) + PLA/BA (9 - 12%) + MF (different OMF were produced). -Granulation in disc granulator. Acids used as binder. Granules dried at 70 °C for 3h.	- Compositions; - Nutrient content; - P availability; - Heavy metals content; - Phytotoxicity; - Grain size.	- High Cd content; - Low content of water-soluble compounds; - Longer stem growth compared to control.
[2]	Soil amendment	Dewatered biological sludge from pulp and paper mill	-Dry basis formulations: BFA (70%) + S (30%) BFA (90%) + S (10%). -Manual granulation on a plastic container. Granules, with diameter of 10 to 15 mm, air dried for 2 months.	- Soil physicochemical properties and trace elements; - Soil enzymatic activities; - Soil water-extract properties	- Improvement of soil quality; - The establishment of a plant cover was not permanent.
[3]	OMF	Dried anaerobic digested municipal sewage sludge	-SS + PLA + MF (8 different OMF were produced). -Granulation in a drum granulator. Acids used as binder.	- Phase composition of the obtained product; - Chemical composition; - Total and bioavailable P content; - Particle size distribution; - Bulk density.	- High nutrient content; - High Cr and Cd content; - Uniform grain size (10 mm – 37,4 % and 8 mm – 47.1 %); - Profits can reach 50.45% compared to conventional fertilizers.
[4]	Fertilizer	Hygienized municipal sewage sludge	-Different formulations: FA+ SS + $CA(OH)_2$ (commercial slaked lime with 31%Ca and 3%Mg). -Components and water mixed with a ribbon blade. Granulation in a drum granulator. Granules air dried for 28 days at room temperature (21 °C)	- Crushing strength of single granules; - Elemental concentrations; - Concentrations of soluble nutrients; - Neutralizing values of the granules;	- Ash can be co-granulated with SS and lime; - SS caused the compressive strength of the granules to remain low; - N content remained low, related to initial ash content.
[5]	OMF	Municipal sewage sludge	-SS + dolomite (50% $CACO_3$ and 40% $MgCO_3$) + lime (96% CaO) + microcrystalline cellulose. -Components are mixed and then granulated, in a disc granulator. Granules are dried by a belt dryer at 100 °C.	- Physicochemical parameters; - Flammability and explosivity tests.	- SS granulation is a promising technology to manage it; - Technical, organizational and human aspects may influence safety of the process;
[6]	OMF	Dewatered digested municipal sewage sludge	-SS + Urea (46% N) + ground potash (60% K_2O). -SS cake is dried at 80 °C in a tumble dryer, producing granules. Granules are coated by spraying melted urea and ground potash.	- Composition; - Particle size; - Bulk and particle density; - Porosity of untapped granules; - Static particle strength; - Total surface are per unit mass.	- Lower risk of build-up of soil P; - SS disposal costs could be reduced; - Reduce reliance on mineral fertilizers.

[1] Kominko et al., 2019; [2] Alvarenga et al., 2019; [3] Kominko et al., 2018; [4] Pesonen et al., 2016; [5] Więckol-Ryk et al., 2020; [6] Antille et al., 2013; OMF - Organo-mineral Fertilizer; SS – sewage sludge; PLA - Poultry Litter Ash; BA – Biomass Ash; MF – Mineral Fertilizer; BFA – Biomass Fly Ash; FA – Fly Ash; P – Phosphorous; N – Nitrogen.

The hazards involved in the process of producing fertilizer pellets from SS were evaluated by Więckol-Ryk et al. (2020). To reduce the risk, some measures associated with SS were highlighted: sensors of flammable gases installation (especially in sludge silo), limiting all sources of ignition, monitoring efficiency of earthing the installation and using intrinsically safe devices, and effective disinfection of the contaminated areas.

Subsequently, some studies regarding the respirometric analysis, are presented Table 3. 2. The respirometric tests measure the biological stability of materials (as the degree of stability). The main variables in respect to the stability of a material that must be controlled are the temperature, moisture content of biodegradable organic matter, and the duration of the test. The standard EN 16087-1:2011 lays down these different parameters. Different indexes can be measured to evaluate the respirometric activity (RA), namely the static respiration index (SRI) and the oxygen uptake rate (OUR). The most reliable and adopted parameter to the scientific community is the OUR. Silveira et al. (2019) and Evangelou and Komilis (2020) have performed studies related to biological stability based on the referred standard and emphasized that the ideal temperature to assess biological stability is 30 °C, with an analysis period of 7 days. The standard EN 16087-1:2011 defines that a material (fertilizer or professional application soil amendment) is stable if the maximum limit of 25 mmol O_2/kg OM·h is not exceeded. However, Vanhoof and Tirez (2017) claims that the 25 mmol O_2/kg OM·h limit value is not achievable for the majority of digestate products when the test is performed at 30 °C.

Table 3.2. Studies from the literature related to respirometric analysis.

Ref.	Sample	Operating Conditions	Quantified Indices	Main Conclusions
[1]	Leather	30 °C 7 days	SRI; OUR	The developed method in this study proved to reliable, able to distinguish different levels of biodegradation over 7 days.
[2]	Different Organic Substrates	30 °C 7 days 120 ± 20 rpm	OUR	The tests carried out in liquid matrix do not limit the transfer of O_2 to the solid substrate.
[3]	Compost, Digestates and Organic Biological Waste	30 °C 20 °C	OUR	The incubation temperature has a large effect on the outcome of the stability test

[1] Silveira et al., 2019; [2] Evangelou and Komilis, 2020; [3] Vanhoof and Tirez, 2017; SRI – Static Respiration Index; OUR – Oxygen Uptake Rate.

Pot experiments are a well-established procedure to evaluate fertilizer behaviour. Table 3.3 displays examples from the literature where this method is applied, using organic wastes to improve the soil. Pinho et al. (2017) concluded that cress germination and early shoots growth did not react well to Olive Mill Solid Wastes (OMW). However, Gomes et al. (2020) presents some positive results with eggshell (ES) and SS, related to plant growth and soil pH correction. Alvarenga et al. (2016) achieve better results to plant growth with SS, compared to the produced compost. This can be explained by the lack of readily N presented in agricultural waste and sewage sludge compost (AWSSC), while only 1.1 % of N is present in NH_4^+-N form on AWSSC, about 30 % is present the utilized SS.

Table 3.3. Studies from the literature involving the pot experiment method.

Ref.	Sample	Culture	Conditions	Analysis
[1]	SS1 SS2 AWSSC	2 g of *Sorghum* bicolor x *Sorghum sudanense* var. Rocket	8 d after incorporation of amendments, pots were cultivated; Pots were routinely watered; - Growth period: 60 d period in outdoors.	- Plant biomass; - Plant foliar area; - Plant chlorophyll content.
[2]	SS ES + SS	17 seeds of *Lepidium Sativum* L. (about 2 mm depth)	- Growth in a chamber: photoperiod of 12 h provided by a white LED; 20 °C; and relative humidity of 50%; - Growth period: 4 weeks.	- Root Growth; - Shoot Lengths growth; - Plants Biomass
[3]	OMW	7 seeds of *Lepidium Sativum* L. (about 2 mm depth)	- Pots were in dark for 24 h and then located in a well-lit location; - Photoperiod: 15 h; - Growth period: 2 weeks	- Percentage of relative germination; - Relative shoot growth.

[1] Alvarenga et al., 2016; [2] Gomes et al., 2020; [3] Pinho et al., 2017; SS – Sewage Sludge; AWSSC – Agricultural Waste and Sewage Sludge Compost; ES – Eggshell; OMW – Olive Mill Solid Wastes;

4. MATERIALS AND METHODS

4.1. Sampling

All the samples considered in this work were collected in mainland Portugal. The SS was taken after anaerobic digestion from a WWTP with a capacity of 140,000 population equivalent. The sample was dehydrated in the plant by centrifugation. A weathered coal fly ash (CFA) sample was obtained from a landfill that receives approximately 40 kt/year from a thermoelectric powerplant. This material did not meet the criteria of the cement industry, namely regarding carbon content. ES samples were collected from an egg pasteurization industry that produces 4 kt/year of this waste. The main characteristics of these materials are reported, in Table 4.1. Five different mixtures were prepared mixing SS, CFA and ES, intending to develop an organic fertilizer. For comparison purposes it was also tested an organo-mineral fertilizer (OMF), and its characteristics are also displayed on Table 4.1. Figure 4.1 shows the appearance of these materials.

Table 4.1. Characterisation of the utilized SS, ES and CFA samples and of the OMF.

	SS	CFA	ES	OMF
pH	6.71	8.60	9.10	5.79
EC (mS/cm)	1.7	0.49	0.21	15.3
OM (%)	63.7	-	4.3	33
$N_{kjeldahl}$ (%)	3.9	-	-	8^b
P_2O_5 (%)	3.83	0.20	0.29	3
K_2O (%)	0.22	2.40	0.10	5
MgO (%)	0.38	1.50	0.72	3
CaO (%)	5.5	1.40	86.1^a	-

^{a}: Expressed in $CaCO_3$; ^{b}: Total N, of which 7,5% is organic.

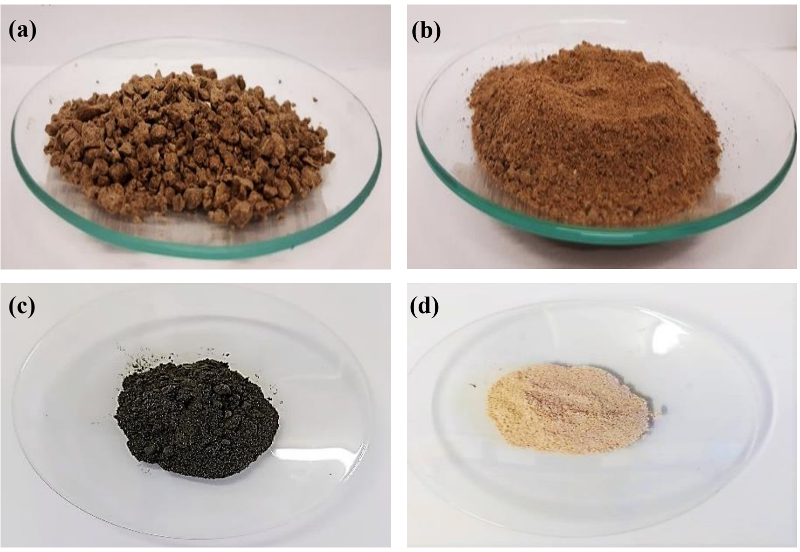


Figure 4.1. Physical aspect of the materials used: (a) OMF before ground; (b) OMF after ground; (c) CFA; (d) ES.

According to Portuguese legislation (Decreto Lei n.º 103/2015), an NPK organic fertilizer must fulfil certain conditions:

- Organic N – 2 (±1,1) %
- Total P_2O_5 – 2 (±1,1) %
- K_2O – 2 (±1,1) %
- N+ P_2O_5+ K_2O – 10 (±1,9) %
- Organic Matter – 50 (±5) %

Based on this information, the five mixtures were prepared to approach the legislation as closely as possible. However, the requirements related to K was not complied, since with the exception of CFA, all the starting materials contain low content in potassium. The formulations of the five different mixtures and the mixtures theoretical compositions are showed in Table 4.2.

Table 4.2. Mixtures prepared and respective theoretical compositions.

	Mixtures				
	M1	M2	M3	M4	M5
SS	0.70	0.70	0.80	0.70	0.70
CFA	0.10	0.20	0.10	0.30	0.00
ES	0.20	0.10	0.10	0.00	0.30
	Theoretical compositions				
OM (%)	45.45	45.02	51.39	44.59	45.88
Organic N (%)	2.73	2.73	3.12	2.73	2.73
Total P_2O_5 (%)	2.76	2.75	3.11	2.74	2.77
K_2O (%)	0.41	0.64	0.43	0.87	0.18
N + P_2O_5 + K_2O (%)	5.90	6.12	6.66	6.35	5.68

A proper mixtures blend was achieved with dried SS at 100 and 130 ºC. The wastes were ground and sieved through a 425 µm screen, and the five mixture were prepared with SS dried at both temperatures. Drying essays were carried out in a Moisture Analyzer, with 1 mg precision (*Precisa XM50*), that uses the gravimetric moisture measurement principle.

4.2. Respirometric Analysis

These essays were developed using *Oxitop*®, an equipment that consists of a hermetically sealed vessel (1 L of capacity). Pressure over time is measured with a sensor on the *Oxitop*® head. The recorded pressure drop is then related to oxygen consumption (defined from the respirometric activity concept) by the application of Equation 2.2, indicated in Chapter 2. The respiration process is characterized by the CO_2 production, which should be retained by NaOH in an aqueous solution or as solid *pellets*.

The respirometric analysis in the aqueous matrix was performed according to EN 16087-1:2011: *Soil improvers and growing media – Determination of the aerobic biological activity – Part 1: Oxygen uptake rate* (OUR). The operating conditions are well established by the standard EN 16087-1:2011: a temperature of 30°C, CO_2 absorbent (NaOH *pellets*), and agitation (120 ± 20 rpm). To guarantee an analysis without nutritional restrictions, with adequate pH, and without interference from nitrification reactions, the reagents to use are also established. The required reagents to perform the analysis are:

- pH buffer: 86 g/L KH_2PO_4, 89 g/L $Na_2HPO_4 \cdot 2H_2O$, with a, pH 7, 1:4 v:v;

- Macronutrients solution: 4.3 g NH_4Cl, 5.4 g $CaCl_2 \cdot 2H_2O$, 4.3 g $MgSO_4 \cdot 7H_2O$, 0.03g $FeCl_3 \cdot 6H_2O$ dissolved in 1000 mL of water;
- Micronutrients solution: 5.0 g EDDHA, 6% iron chelate, 1.4g $MnSO_4$, 1.1 g $ZnSO_4$, 4.2 g $Na_2B_4O_7$, 0.2 g $CuSO_4$, 0.13 g Na_2MoO_4, 1 mL/L HCl (36%) dissolved in 1000 mL of water;
- Composed nutrients solution: 1 mL of micronutrients solution to 1000 mL of macronutrients solution
- Nitrification inhibitor: 4 g/L N-Allylthiourea $C_4H_8N_2S$ (ATU).

Due to limitations associated with reagents, in the laboratory, the micronutrient solution and the nitrification inhibitor were not used.

Based on 2 g of organic matter per liter (EOM), the standard defines the sample quantity calculation to introduce in the vessel:

$$EOM = \frac{2000}{OM \cdot TS} \tag{4.1}$$

where, EOM is on g/L, and OM and TS are the percentage of organic matter and the percentage of total solids, respectively.

$$W = EOM \cdot C_V \tag{4.2}$$

where, W is sample quantity in g and C_V is the vessel capacity in L.

The samples were placed in the *Oxitop*®, and 180 mL of water were added along with 10 mL of macronutrients solution and 10 mL of pH buffer. Then, the equipment was placed in the agitation table (100 rpm) for 4 h before being closed, and the experiment started. The samples analysed according to described procedure were:

- SS dried at 100°C;
- SS dried at 130°C;
- Fresh SS;
- M1;
- M2;
- M3;
- M4;
- M5.

The standard EN 16087-1:2011 defines that to calculate the RA (mmol O_2/kg OM) the Equation 4.3. must be used,

$$RA = \frac{\Delta P \cdot 10}{R \cdot (273{,}15 + T)} \cdot \frac{V_{gas} \cdot 10000}{W \cdot DM \cdot OM} \tag{4.3}$$

where, ΔP is the pressure drop (kPa), R is the is the universal gas constant (mL·hPa/Kmol), T is the analysis temperature (°C), W is the sample mass with the water content (kg), DM is the dry matter content (%), OM is the organic matter content (%), V_{gas} is the gas volume (mL).

Moreover, measures must be considered between the moment that the pressure drop reaches zero and 72 h after. In this period, the pressure drop should not be higher than 100 hPa since a superior value means that the O_2 content diminished from 20 to 10%, and it may represent a limitation on the O_2 supply to aerobic respiration.

To determine the gas volume, Equation 4.4 is applied:

$$V_{gas} = V_{vessel} - \frac{W \cdot DM \cdot 10000}{\rho} - V_{liquid} \tag{4.4}$$

where, V_{vessel} is the *Oxitop*® total volume (mL), V_{liquid} is the total added liquids volume (mL) and ρ represents the measured sample density (kg/m^3), which is determined by:

$$\rho = \frac{1}{\frac{OM \cdot W \cdot DM}{1550} + \frac{(1 - OM) \cdot W \cdot DM}{2650}} \tag{4.5}$$

The oxygen uptake rate (OUR), expressed in mmol O_2/kgOM.h, is obtained with Equation 4.6:

$$OUR = \frac{RA}{\Delta t} \tag{4.6}$$

where, Δt is the time period considered for RA determination, 72 h.

4.3. Germination assays

The phytotoxicity test was performed with *Lepidium sativum* L. (garden cress). For this purpose, liquid extracts were obtained for the different liquid to solid ratios (L/S), based on the standard EN 12457-2:2002, with some adaptions. The solid samples tested were the five mixtures (M1 to M5) described above, SS, ES, CFA, and the commercial fertilizer. In each case, 5 mL of extract was added to a petri dish (90 mm) along with ten seeds placed over the Whatman filter paper. The samples remained in an oven at 25 ± 1°C for 48h, in dark conditions (Gomes et al., 2020). Germination index (GI) was obtained by comparing the percentage of the number of germinated seeds and the percentage of the root lengths of the samples concerning the control sample (Pinho et al., 2017).

$$GI = \frac{RSG \cdot RRG}{100} \tag{4.7}$$

where, RSG is the relative seed germination and RRG is the relative root growth.

$$RSG = \frac{\overline{N}}{\overline{N_b}} \tag{4.8}$$

where, $\overline{N}$ is the mean of root length and $\overline{N_b}$ represents the blank mean root length.

$$RRG = \frac{\overline{L}}{\overline{L_b}} \tag{4.9}$$

where, $\overline{L}$ is the relative root growth and $\overline{L_b}$ stands for the blank relative root growth.

4.4. Pot Experiments

The agronomic potential of the mixtures can be determined with pot experiments. The five mixtures with dried SS (100 °C) were tested, and the selected culture was garden cress, the same used for the germination assays. This culture was selected due to its fast growth and small cost and dimension. The following treatments were applied, considering a rate of application of 24 t/ha:

- P1 – 100% Soil
- P2 – 85.65% Soil + 14.35% M1
- P3 – 85.65% Soil + 14.35% M2
- P4 – 85.65% Soil + 14.35% M3
- P5 – 85.65% Soil + 14.35% M4
- P6 – 85.65% Soil + 14.35% M5
- P7 – 85.65% Soil + 14.35% OMF

The used vases for the mixtures, have a capacity of 300 cm^3, where 10 cm^3 were filled with gravel so that the excess of water could leak. The soil was collected from an agricultural field located in Cesar, Oliveira de Azeméis (40°55’19.3’’N 8°26’13.2’’W), at an approximately 20 cm depth. Its main characteristics are a pH of 6.81, EC of 0.058 mS/cm, OM of 11.8%, and water holding capacity of 292 g H_2O/100 g soil.

Ten seeds of garden cress were placed at a 2 mm depth in the pots, and the water content was constant in all the experience period. The atmospheric conditions were controlled and are presented in Appendix II. The experiment was conducted for 4 weeks. After this period, physical growth parameters were determined, such as shoot, stem, and roots length. By drying all the plant parts at 60 °C until constant weight it was possible to determine the plant biomass quantity (g).

5. RESULTS AND DISCUSSION

In this chapter, the results are presented in 4 different parts. The first one is a statistical analysis of digested SS concerning its characteristics with potential agricultural value. The second one is about the stability of the different produced mixtures, while the third part explores the phytotoxicity essays. Finally, it is evaluated the agronomic potential of the mixture with garden cress.

5.1. Characteristics of digested SS: statistical analysis

This analysis aims to highlight barriers and opportunities of using digested SS to produce fertilizers. Therefore, the parameters with the greatest impact on the fertilizer's quality were considered. Data from the literature were collected and analyzed to provide an overview of the concentrations of these elements in the sludge. It is important to keep in mind that the nutrients must be in a bioavailable form for an immediate uptake or in a form that permits that the nutrients become bioavailable.

5.1.1. Organic Matter

The parameter that stands out the most in the sludge is the OM content. Figure 5.1 shows data gather in the literature (Annex I) regarding OM in 40 samples. Figure 5.1 shows that the range is from 37-83%, while the mean and median are 57.03 and 54.90%, respectively.

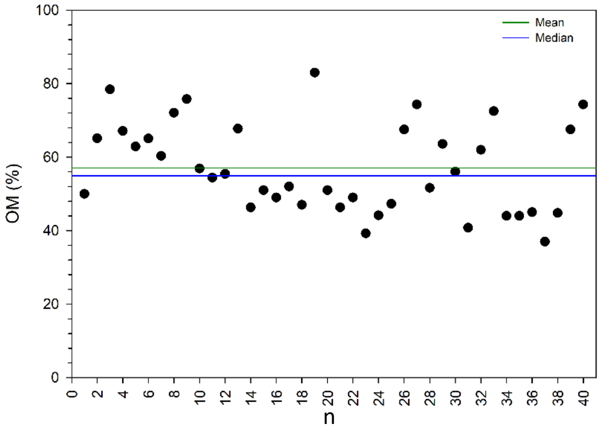


Figure 5.1. Organic matter content in the SS.

The addition of stable organic to the soil matter is important to ensure its good condition. Indeed, the OM content can influence the soil microorganisms (species and population), improving water infiltration and aeration, the water holding capacity, the buffering capacity against rapid changes in pH, among others (Jones Jr., 2012). Considering the values found, it is possible to conclude that SS can be considered a source to increase the OM of poor soils.

5.1.2. NPK nutrients

The essential nutrients for plant growth are N, P, and K, each of them with a specific function. Figure 5.2 (a)-(c) exhibits the content of nitrogen, obtained with the Kjeldahl method or other techniques, phosphorus, in the form of P or P_2O_5, and potassium, as K or K_2O. The absolute values found for N, P, and K are presented at Tables I.1, I.2, and I.3 of Appendix I, while the main values are presented in Table 5.1. The P and K oxides representation is a usual form to exhibit their concentration in the fertilizer industry.

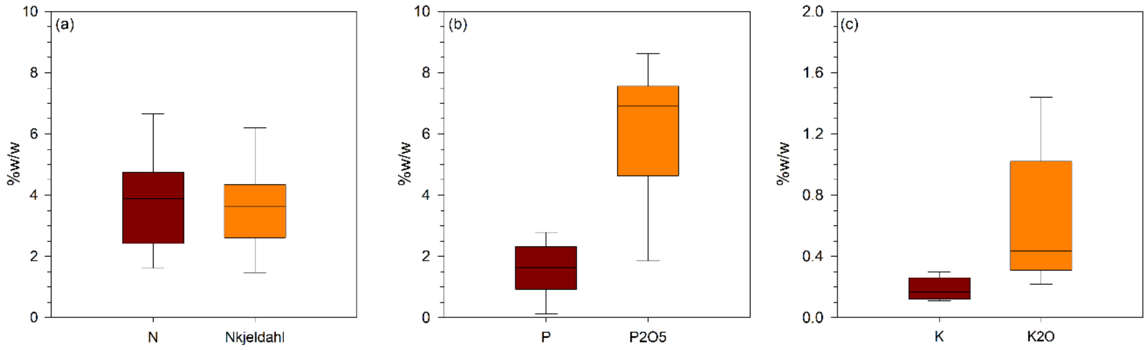


Figure 5.2. Main macronutrients content in the SS (%w/w): (a) N and $N_{kjeldahl}$; (b) K and K_2O; and (c) P and P_2O_5.

Regarding N content, sandy soils usually are associated with high deficit of this element. The shortage of N can be related to heavy rainfall/irrigation, mineral soils with low organic matter content, and a long-history of crop-depleting N supply (due to unbalanced soil nutrition). These types of situations are frequent among the Mediterranean Basin soils (Bancessi et al., 2011). Nevertheless, N is an essential plant nutrient required for an healthy plant growth in concentrations of about 1.4% (Jones Jr., 2012). The obtained values regarding this element vary a lot, from 1.2 to 7.04% N and 0.74 to 6.86% $N_{kjeldahl}$. These may be a consequence of the experimental procedures, but mainly from the initial wastewater and the treatments applied in the WWTP that define all the SS characteristics. However, the results from the Kjeldahl method and other methods are similar, with mean values of 3.63 and 3.93%, respectively (Table 5.1).

P is required in small quantities, approximately 0.2% for healthy plant growth, but it is essential for the plant metabolism, with important participation in various energy transfer reactions. The Portuguese legislation requires 2% P_2O_5 for an Organic NPK Fertilizer and 3% P_2O_5 for an NPK OMF. From the collected data, it was possible to obtain a P_2O_5 mean of 3.64% (Table 5.1.). Thus, it may be possible to develop a commercial fertilizer based on SS regarding the P requirements.

K is fundamental to control the osmotic potential of plants since it is involved, among other things, with the opening and closing of their stomata. About 1% of K is required for healthy plant growth (Jones Jr., 2012). As already referred, K is present in lower concentration. Table 5.1 shows a mean content of 0.19% K and 0.62% K_2O.

Therefore, typical N and P concentrations in plants can easily suppress plants necessity and even empowers SS soil applications in certain parts of the globe, whereas K content is low in SS. The low K concentration in SS can supplement for example with biomass ashes. It is important to note that the N content obtained through this data analysis is close to the one found in the SS sample used in this work (Table 4.1). On the other hand, the P and K values are right above the minimum value found in the data collected.

Table 5.1. Statistical analysis of the data collected in the literature related to N, P, and K content.

Parameter	n*	Mean (%)	Median (%)	Max (%)	Min (%)	SD (%)
N	35	3.93	3.90	7.04	1.2	1.75
$N_{kjeldahl}$	33	3.63	3.64	6.86	0.74	1.63
P	27	1.56	1.64	3.1	0.0018	0.89
P_2O_5	17	6.10	6.90	8.96	1.84	2.18
K	22	0.19	0.17	0.30	0.05	0.07
K_2O	19	0.62	0.44	1.45	0.21	0.41

n-number of values collected in the literature used for the statistical analysis; SD- standard deviation.

5.1.3. Other Macronutrients

Besides the main macronutrients, SS can also provide other macronutrients. Figure 5.3 presents the expected distribution of S, Ca, Mg, and Na in percentage, while Table 5.2

summarizes the essential calculated statistical parameters. The absolute values found for S, Ca, Mg, and Na are presented at Tables I.4 Appendix I.

These elements are relevant once they can have a main role in specific reactions in the plant. For example, Ca serve to detoxify it from the presence of PTM, S can reduce the incidence of some diseases, Mg is part of the chlorophyll molecule, among others. The required concentrations for healthy plant growth are 0.1, 0.5, and 0.2% for S, Ca, and Mg, respectively. According to the data from the literature, it can be concluded that SS can improve these elements in the soil solution, helping to retain a green cover. The secondary macronutrients with the largest proportion are S and Ca, both with a 2.38% mean value (Table 5.2.). Even Mg presents a smaller mean value (0.7%), the content of this element in SS is reasonable to overcome the needs of plants. The OMF used in the pot experiments is an example of it, with 3% MgO (Table 4.1). The standard deviations are also high.

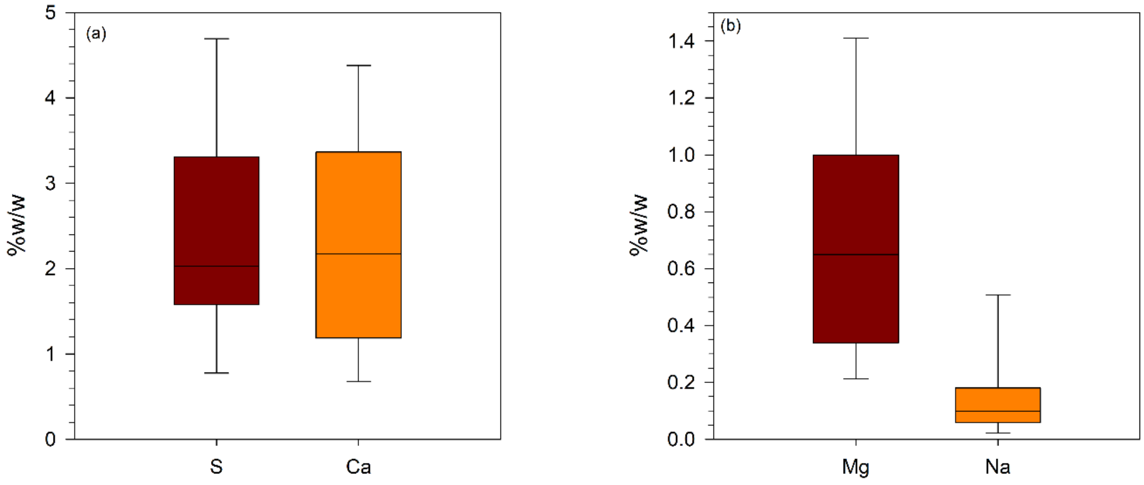


Figure 5.3. Secondary main macronutrients content in the SS (%w/w): (a) S and Ca; and (b) Mg and Na.

Table 5.2. Statistical analysis of the data collected in the literature related to S, Ca, Mg, and Na content.

Parameters	**S**	**Ca**	**Mg**	**Na**
n*	15	51	51	41
Mean (%)	2.38	2.38	0.70	0.17
Median (%)	2.03	2.17	0.65	0.1
Max (%)	4.97	6.12	1.96	0.64
Min (%)	0.53	0.39	0.02	0.001
Standard deviation (%)	1.26	1.47	0.45	0.18
25th Percentile (%)	1.63	1.19	0.34	0.06
75th Percentile (%)	3.09	3.28	1.00	0.18

*number of values collected in the literature used for the statistical analysis.

5.1.4. Micronutrients

SS also contain the essential micronutrients, Figure 5.4 (a)-(b). The absolute values found B, Mn, V, Cl, and Fe are presented at Tables I.5 of Appendix I. From all, Cl and Fe are the most expressive elements in SS. They can achieve up to 1% of SS total solids, while the other elements remain below. Data for Mo is scarce, and it was not possible to achieve meaningful conclusions about this element. Regarding Co, it has achieved 9.21 % mean, a standard deviation of 8.03 % and a 5.65 % median. B and V are found in small quantities (43.79 and 14.8 mg/kg, respectively), while Mn is present with a considerable amount (307.2 mg/kg).

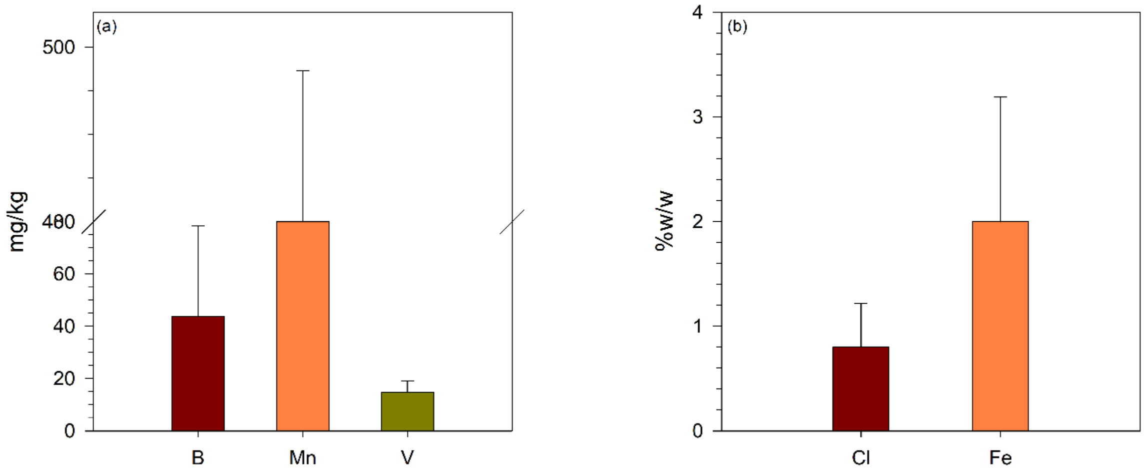


Figure 5.4. Micronutrients content in the SS: (a) B, Mn, and V in mg/kg and (b) Cl and Fe in %w/w.

5.1.5. Potentially Toxic Metals

Even though SS has high agricultural potential, it can also pose a hazard to soil due to several characteristics. For example, the concentration of the several potentially toxic metals (PTM) legislated by European and National legislation (Table 2.4): Pb, Cd, Cr, Cu, Ni, Zn, Hg. Both Zn and Cu can be considered as micronutrients or PTM, depending on their concentrations. As mentioned before, Portuguese legislation (Decreto-Lei n.º 276/2009) establishes legal limits for all the PTM not only in the soil but also in the SS intended to be applied in the soil. Figure 5.5 (a)-(g) presents the data gather from the literature, along with the mean, median, and legal limits, according to Decreto-Lei n.º 276/2009.

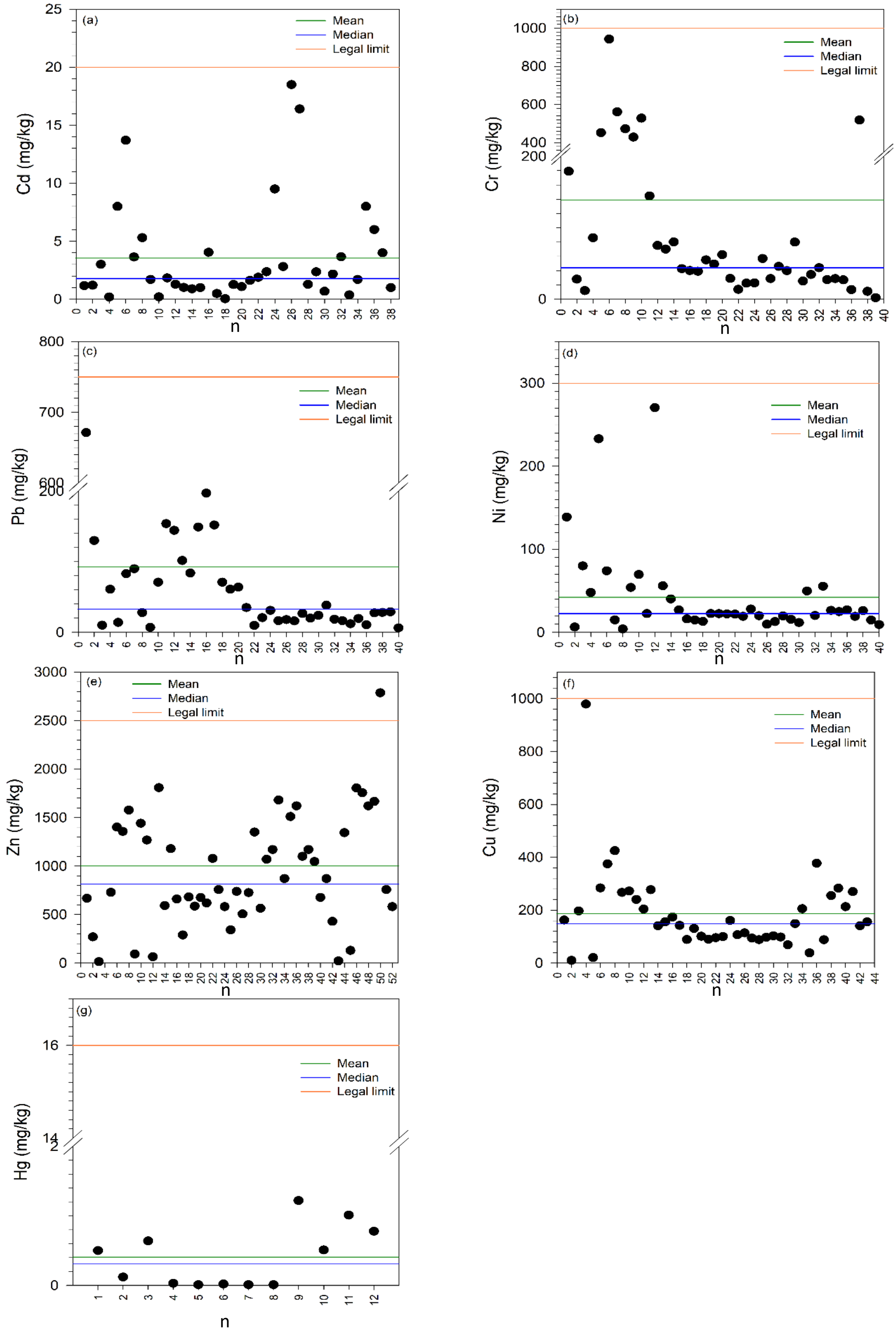


Figure 5.5. PTM concentrations in the SS in mg/kg: (a) Cd; (b) Cr; (c) Pb; (d) Ni; (e) Zn; (f) Cu; and (g) Hg. The sample number is represented by n, according to Table I.6 of Appendix I.

The values found for each PTM are presented at Table I.6 of Appendix I. Considering Figure 5.5, it can be concluded that in general the content of metals in SS complies with legal restrictions. Figure 5.5 (e), reveals that Zn content presents the highest dispersion of data, with mean of 814.1 mg/kg and median of 1001.97 mg/kg. These pollutants are persistent in the environment and cannot be destroyed. Thus, it is crucial to limit PTM concentration in soil to avoid contamination of both cultures and soil, affecting human health and ecosystems downstream.

From all the different samples analysed (n=52), only one sample exceeded limit concentration of Zn set in the Portuguese regulations. Comparing with other restrictive legal limits established by other EU countries, it is possible to conclude that often SS would not be able to be applied in the soil regarding Cd concentration. The Netherlands is the stricter country regarding Cd concentration (1.5 mg/kg), followed by Sweden and Austria with a limit of 2 mg/kg. Germany and Poland are less restrictive than these last three but more than Portugal, with a legal limit of 10 mg/kg. The limits vary from country to country and include different PTM in the list of restrictions. In general, Portugal is one of the less restrictive countries in the EU, adopting limits similar to the ones set by Directive 86/278/EEC (European Commission, 2001).

5.1.6. Microbiological Contamination

Finally, it was studied the presence of microbiological contamination in digested SS and presented in Figure 5.6. From the analysed data, it was possible to determine that *Salmonella* spp. is not present in most of the samples collected (75%). Besides *Salmonella* spp., also *Escherichia coli* is used as a microbiological contamination indicator, and there are even limits in the legislation for these two species. In the Portuguese legislation (Decreto Lei n.º 276/2009), *Salmonella* spp. should be absent in 50 g of original material, while *E. coli* should be less than 1000 CFU/g of fresh matter (3 log CFU/g).

It was not possible to collect enough information related to *E. coli*. Nevertheless, according to a study carried out by Santos et al. (2020), seven out of nine anaerobic digested SS revealed *E. coli* contamination above 4 log CFU/g, while only one fulfils the *E. coli* limit but was contaminated with *Salmonella* spp. The contamination from these types of pollutants poses a risk to human health and to the environment that should be taken into consideration when SS is considered as a soil improver (Santos et al., 2020). In the EU, the current legislation does not consider microorganisms as a pollutant, and thus many EU countries do not regulate this contaminant (European Commission, 2001). Portugal, France, and Italy are some of the exceptions.

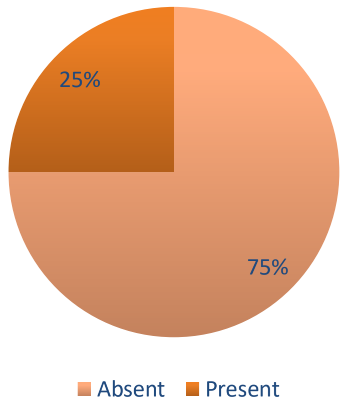


Figure 5.6. Ratio of absence/presence of *Salmonella* spp. in the collect data of anaerobic digested SS.

Besides the PTM and the microbiological contaminants, SS also contain other contaminants of emerging concern (CEC) such as persistent organic compounds, pharmaceutical, cosmetic residues and microplastics. However, the available information about these pollutants are still scarce in the literature, so it was not possible to evaluate it properly. Recently, the contamination of SS with microplastics has been highlighted in the literature (Santos et al., 2020; van den Berg et al., 2020).

5.2. Stability of the different mixtures

Respirometric analyses were performed to study the biological stability of the mixtures. The analysis followed the standard EN 10687-1, which means that aqueous matrices were used. Initially, it was evaluated the RA for the different mixtures with SS dried at 100 ºC, Figure 5.7 (a)-(b). All the samples show a similar pressure drop profile, so the stability will be compared based on the OUR, Figure 5.7 (b). It is possible to observe that the OUR values do not vary much, with a range from 33.41 to 44.29 mmol O_2/kg OM.h. Indeed, the OUR values for each case are statistically similar, according to the Tukey HSD test (p>0.05). The limits defined according to EN 10687-1 for a fertilizer (25 mmol O_2/kg OM.h) or for a professional application soil amendment (15 mmol O_2/kg OM.h) were exceeded in all the mixtures. For this reason, it was studied the effect of SS drying temperature in the OUR.

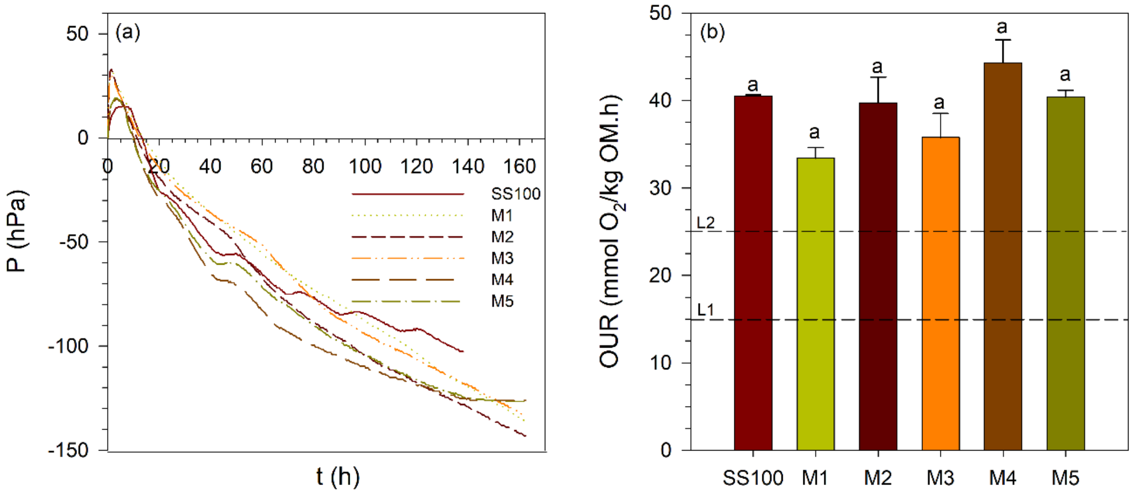


Figure 5.7 (a) Mean pressure drop for different mixtures with SS dried at 100 ºC; (b) OUR value for each mixture and stability limit; [L1 – limit for professional application soil amendment; L2 – limit for fertilizer; Results marked with equal letters are statistically similar (Tukey HSD test with $p < 0.05$)].

For evaluating the drying temperature, three different respirometric tests were conducted: with fresh SS, SS dried at 100 ºC, and SS dried at 130 ºC. The commercial OMF was also analysed for comparison purposes, Figure 5.8 (a)-(b). The OMF was the material which presented the highest OUR (79.11 mmol O_2/kg OM.h), followed by the fresh SS sample (52.08 mmol O_2/kg OM.h). It is important to point out that, even though the OMF is commercialized, it is unable to comply with the limits defined by the standard. As the drying temperature rises, the respirometric activity drops, with the lower OUR (26.25 mmol O_2/kg OM.h) observed for SS dried at 130 ºC. When SS are subjected to high temperatures, the population of microorganisms will decrease, which will directly affect the OUR. The fresh SS was not able to fulfil the EN 10687-1 established limits for fertilizers and soil improvers, and it was also not able to achieve the digested sludge limit (50 mmol O_2/kgOM.h) either. The OUR found for SS dried at 130 ºC is really close to the limit to fertilizers. Since the adjuvants appeared to have a positive influence over the OUR reduction; thus, respirometric tests were applied to the mixtures with SS dried at 130 ºC.

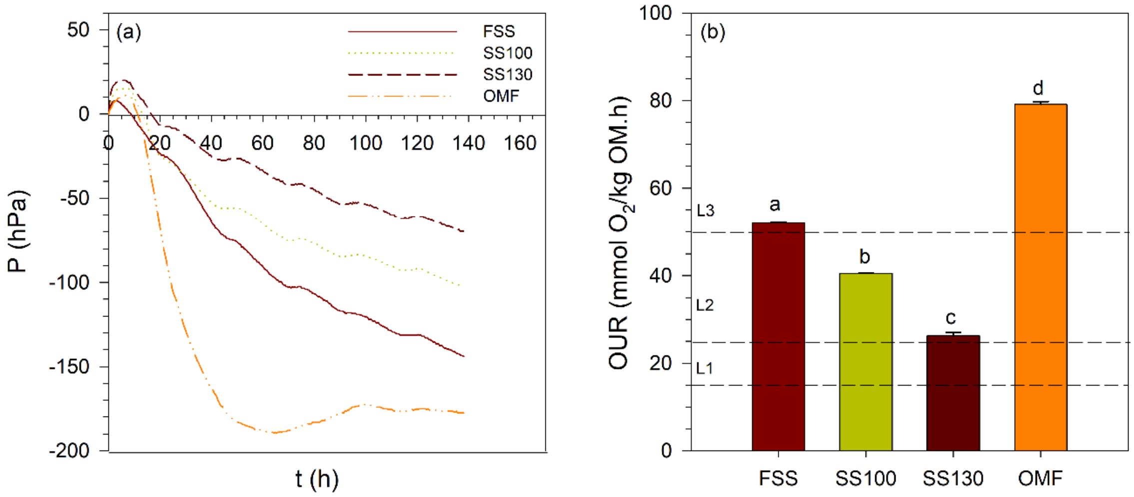


Figure 5.8. (a) Mean pressure drop for fresh SS, dried SS at 100 and 130 ºC, and OMF; (b) OUR value for each case and stability limits [FSS – fresh SS; SS100 – SS dried at 100 ºC; SS130 – SS dried at 130 ºC; L1 – limit for professional application soil amendment; L2 – limit for fertilizer; L3 – limit for digested SS].

The results from the mixtures with SS dried at 130 ºC are presented in Figure 5.9 (a)-(b). The OUR of SS dropped to values in a range of 11.13 to 20.09 mmol O_2/kg OM.h (M1 and M5, respectively). Besides M1, both M2 (11.14 mmol O_2/kg OM.h) and M3 (14.18 mmol O_2/kg OM.h) presented values below the limit value established for soil improvers. M5 and M4 (18.80 mmol O_2/kg OM.h) are above this limit but are below the limit value for fertilizers, which was not possible only with SS dried at 130 ºC.

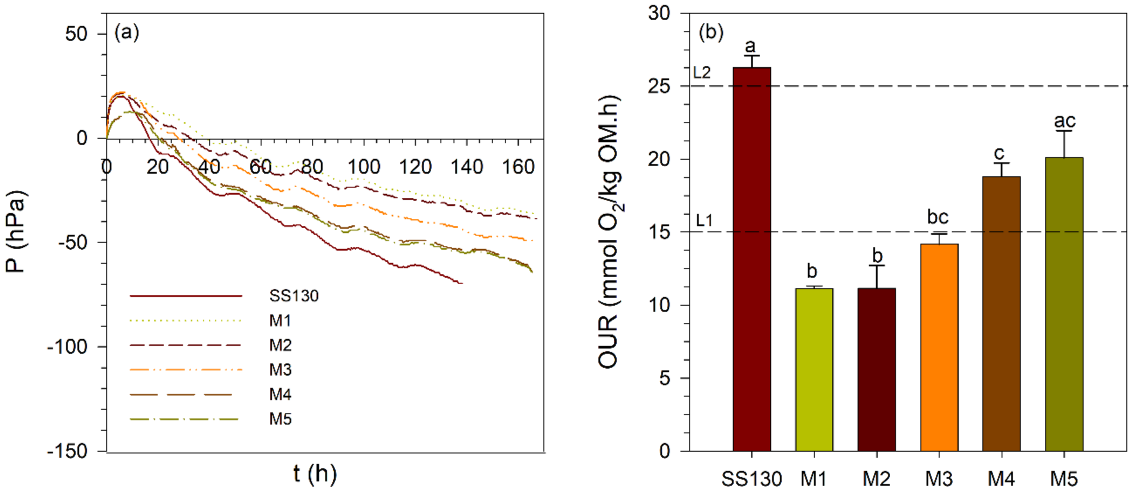
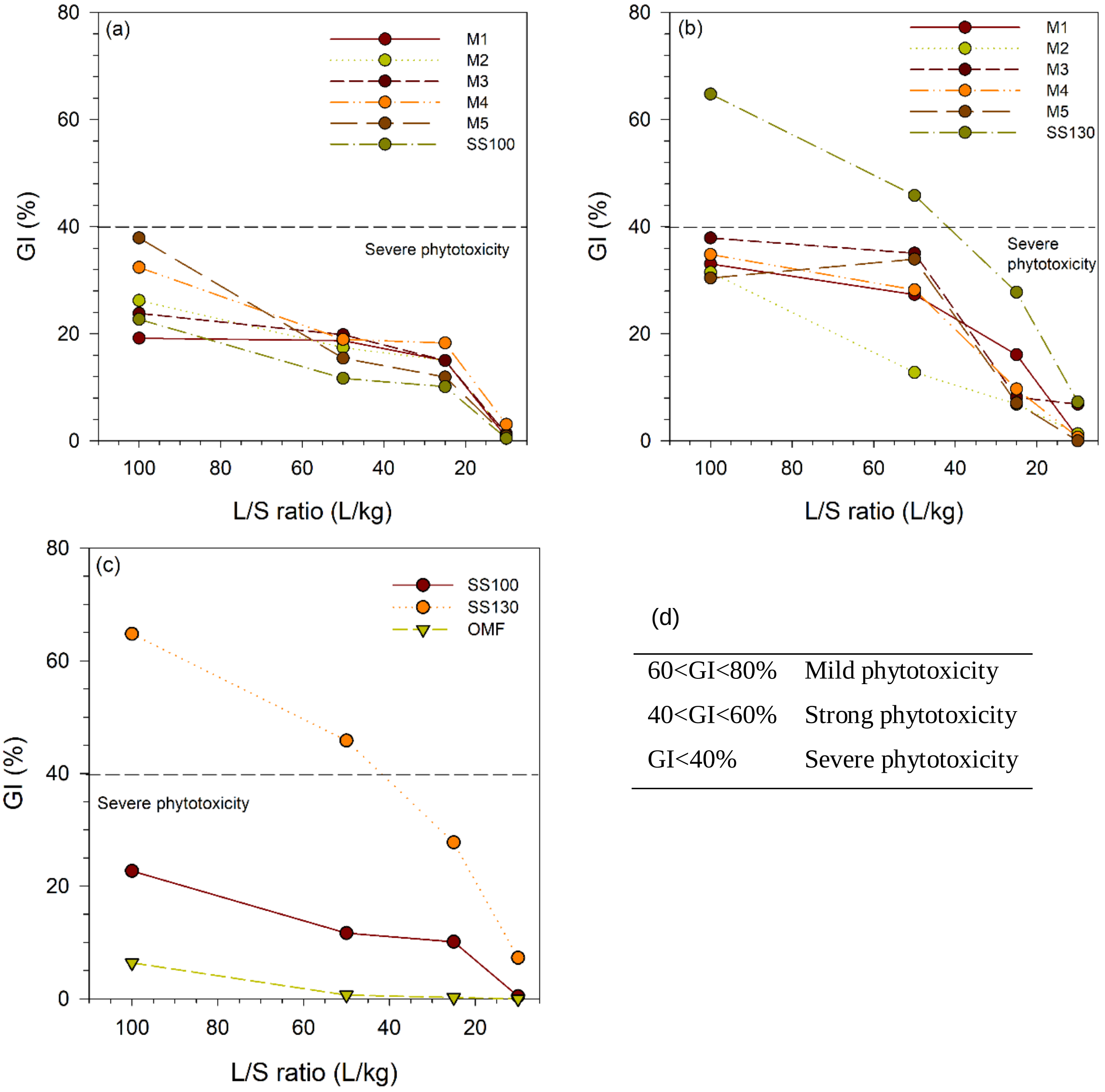


Figure 5.9. (a) Mean pressure drop for different mixtures with SS dried at 130 ºC; (b) OUR value for each mixture and stability limit; [L1 – limit for professional application soil amendment; L2 – limit for fertilizer; Results marked with equal letters are statistically similar (Tukey HSD test with $p < 0.05$)].

5.3. Phytotoxicity essays

The ability of the mixtures prepared with SS, CFA, ES to inhibit plant germination and growth was measured using phytotoxicity assays. The germination index, GI is considered a reliable parameter to characterize the phytotoxicity of a sample: 60<GI<80% - mild phytotoxicity; 40<GI<60% - strong phytotoxicity; and GI<40% - severe phytotoxicity. Figure 5.10 (a)-(c) exhibits the GI measured in liquid extracts obtained at different liquid/solid (L/S) ratios for the mixtures with dried SS at 100 ºC and 130 ºC, and for the commercial OMF used, respectively.



(d)

60<GI<80%	Mild phytotoxicity
40<GI<60%	Strong phytotoxicity
GI<40%	Severe phytotoxicity

Figure 5.10. GI of Lepidium sativum at different L/S ratios of: (a) mixtures with dried SS at 100 ºC, (b) mixtures with dried SS at 130 ºC, (c) OMF, SS100, and SS130, and (d) phytotoxicity classification.

Figure 5.10 (a) shows that the maximum GI for M1 to M5 values range from 19.2 to 37.9% at L/S 100 L/kg, while the minimum GI values stand at the range of 0.66 to 3.08% when using a 10 L/kg. It is possible to conclude that all the samples still exhibit severe phytotoxicity, which indicates that the germination phase is strongly affected by the mixtures. Table 5.3 reports the main data from the GI tests. From those data, it is not possible to indicate a mixture that promoted the best result in all the L/S ratios tested. M5 showed the highest GI in the 100 L/kg ratio, while M3 was better at 50 L/kg, and M4 in both 25 and 10 L/kg. All the samples present GI below 4% at 10 L/kg ratio, and the EC found in the extract is high (> 4 mS/cm). In these conditions, the seeds and radicles are under high osmotic pressure stress, which may impair the germination phase.

Table 5.3. pH, EC and GI values for the different samples tested in the phytotoxicity assays.

Sample	**Parameter**	**L/S (L/kg)**				**Sample**	**Parameter**	**L/S (L/kg)**			
	100 °C	100	50	25	10		**130 °C**	100	50	25	10
SS100	pH	7.01	6.94	6.82	6.79	SS130	pH	7.01	6.95	6.63	6.30
	EC (mS/cm)	0.92	1.57	2.72	5.04		EC (mS/cm)	0.48	0.74	1.42	2.75
	GI (%)	22.7	11.7	10.1	0.44		GI (%)	64.8	45.8	27.7	7.27
M1	pH	6.91	7.04	7.00	6.91	M1	pH	7.25	7.17	6.92	6.74
	EC (mS/cm)	0.77	1.21	2.02	4.13		EC (mS/cm)	0.41	0.61	1.18	2.48
	GI (%)	19.2	18.7	14.9	0.66		GI (%)	33.1	27.3	16.1	0.66
M2	pH	7.08	7.08	6.91	6.91	M2	pH	7.29	7.17	7.17	6.79
	EC (mS/cm)	0.84	1.24	2.14	4.14		EC (mS/cm)	0.46	0.78	1.36	2.60
	GI (%)	26.2	17.4	14.9	1.32		GI (%)	31.5	12.8	6.83	1.32
M3	pH	7.12	7.08	6.87	6.74	M3	pH	7.27	7.22	7.12	6.92
	EC (mS/cm)	0.79	1.34	2.25	4.39		EC (mS/cm)	0.50	0.79	1.39	3.01
	GI (%)	23.8	19.8	14.9	1.32		GI (%)	37.9	35.0	8.15	6.83
M4	pH	7.04	7.00	6.87	6.69	M4	pH	7.30	7.17	7.13	6.96
	EC (mS/cm)	0.78	1.24	1.98	4.01		EC (mS/cm)	0.44	0.74	1.3	2.91
	GI (%)	32.4	18.9	18.3	3.08		GI (%)	34.8	28.2	9.69	0.66
M5	pH9	7.21	7.25	7.17	7.00	M5	pH	7.30	7.39	7.26	7.18
	EC (mS/cm)	0.76	1.21	1.95	4.30		EC (mS/cm)	0.52	0.74	1.30	2.61
	GI (%)	37.9	15.4	11.9	0.88		GI (%)	30.4	33.9	7.05	0
OMF	pH	5.92	5.92	5.83	5.79						
	EC (mS/cm)	3.02	4.87	7.95	15.3						
	GI (%)	6.39	0.66	0.22	0						

Figure 5.10 (b) presents the germination results for mixtures with SS dried at 130 ºC. It is possible to conclude that superior GI values were found at 100 L/S ratio in this case, with a range from 30.4 to 37.9%. At 10 L/kg ratio, the GI is minimum and range from 0 to 1.32%. At 130 ºC, the M3 presents the highest GI values for all liquid/solid ratios, except at 25 L/kg. Indeed, the EC values for the mixtures at 130 ºC are lower than in the case of the mixtures at 100 ºC, which can determine the better results during the germination phase. Figure 5.10 (c) and Table 5.3 shows that the organic-mineral fertilizer is more toxic than the mixtures

prepared with SS, CFA and ES. Indeed, the EC of the OMF is extremely high reaching more than 15 mS/cm at a 10 L/kg ratio. Overall, it is possible to observe that as the EC decreases, the GI increases. Nevertheless, the germination is a sensitive phase, and caution is needed at this stage when the mixtures are applied to the soil.

5.4. Pot experiments

Pot experiments were carried out to evaluate the capability of the mixtures to improve soil quality. However, the results from the growth tests were not as expected since the amendments may have impaired the plants growth. In fact, some plants only germinated after four weeks, when the experiment was finishing. The pots with only soil revealed the highest rate of germinated plants (75%). The soils amended with M1 and M2 showed a rate of germinated plants of 20 and 30%, respectively, while the other amendments presented results inferior to 15%. The tested OMF completely inhibited the germination phase. Indeed, the EC found in the amended soil with OMF reach values of 8.41 mS/cm, while for the remaining amendments, the EC was between 1.71-2.74 mS/cm. These values are high comparing the EC of the soil (0.06 and 0.19 mS/cm before and after the experiment, respectively), which may have negatively influenced germination phase. These results can indicate that the incorporation rate was too high (24 t/ha). In further tests, it is advisable to use lower incorporations rates (e.g., 12 or 6 t/ha).

However, the amendments revealed positive effect in the OM and the pH of the soil after four weeks of growth, Figure 5.11 (a)-(b). Although the soil is already rich in OM (11.78%), after the amendments (with mixtures and OMF), the OM content increased. From Figure 5.11 (a), it is possible to observe that soil amended with M3 presented the highest OM content (23.31%), followed by the amendment with SS (22.90%) and OMF (22.81%). The remaining mixtures have values of OM in the range of 21.88 to 22.67%, except for M5 that presented the second lowest value (18.06%). Also, pots with only soil also exhibited a higher OM content after the experiments. The roots from the plants are very delicate, and when removed, can break and remain in the soil, which could affect the OM content determination. The pH comparison suggests that all the mixtures have a similar effect over the soil, with the pH decreasing. The same happens for both fertilizer and SS. The pH value for the soil after the experiment was almost equal to the original one (6.89 and 6.81, respectively), as expected.

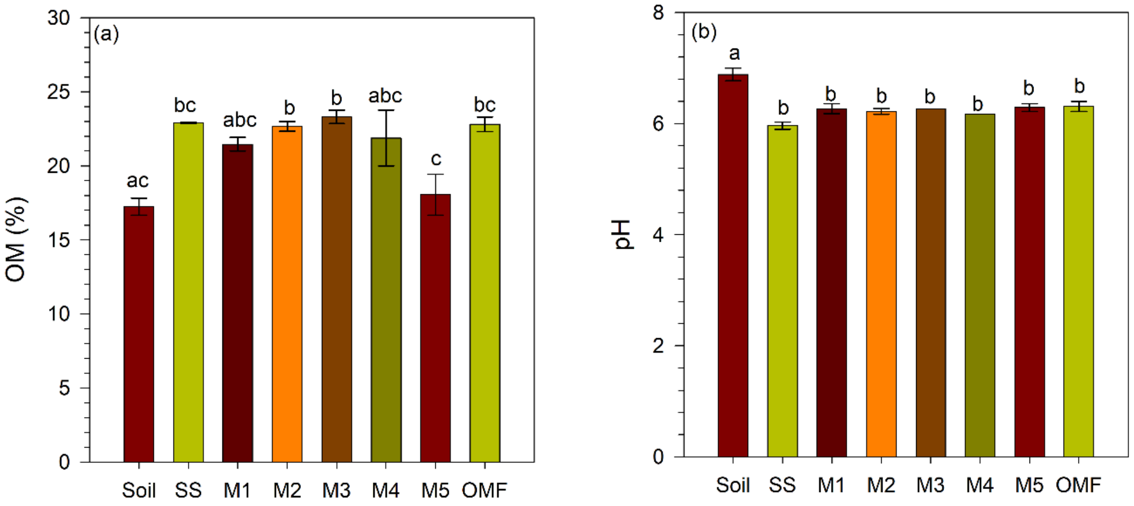


Figure 5.11. (a) OM and (b) pH after four weeks of experiment [Results marked with equal letters are statistically similar (Tukey HSD test with $p < 0.05$)].

Considering that the objective of this section was to obtain reliable information about the effects of the soil amendments in the soil, these assays need to be redesigned and repeated to achieve valid conclusions. The new tests should be conducted with a controlled environment (e.g., humidity and luminosity) and knowing the soil conditions and characteristics.

6. CONCLUSIONS AND FUTURE WORK

The main objective of the thesis was to evaluate the possibility of producing a SS based fertilizer, which is a way of adding value to SS, as well as to promote the circular economy. To accomplish the objective, five different formulations with two adjuvants (eggshell and coal fly ash) were designed. These formulations were tested using phytotoxicity essays, respirometric tests, and pot experiments.

Through a statistical assessment of digested sludges from the literature, it was possible to conclude that these sludges have an interesting content of OM, as well as of N and P, two essential nutrients for plant growth. SS has also the capability to provide other macronutrients and micronutrients. Although some pollutants are part of SS composition, they are usually present within acceptable limits, according to the Portuguese legislation. Nevertheless, a better understanding of potentially toxic pollutants and their harmful effect is needed.

The stability analysis is well established by standard EN 10687-1, which was used as guidance for the determination of the mixtures OUR. According to the results found with SS dried at 100 ºC, it was evident that none of the mixtures was able to accomplish the requirements for fertilizers established in the standard. So, further tests were conducted to investigate the effect of the SS drying temperature in the stabilization process. Thus, it was possible to conclude that using a drying temperature of 130 ºC, the OUR values decreased.

From the phytotoxicity essays, it was observed that the increase of SS drying temperature had a positive impact on the GI and the EC. The commercial organic fertilizer had an extreme toxic impact on the seeds germination, with the highest toxic effect when compared to all the prepared mixtures.

The pot experiments did not lead to the expected results, since the plants germination was reduced for all the tests with soil amendments. Even though the OM content was improved in all the pots and the pH reached acceptable levels, the plants that germinated had low mass production. These experiments will have to be repeated in order to reach reliable conclusions.

6.1. Future Work

The application of SS in agriculture through fertilizer is a prominent management strategy for this material. During the thesis development, many approaches were discarded. Regarding this, some future work is suggested below:

- Investigate the possibility of SS granulation, with a focus on establishing the parameters for obtaining a product with suitable properties as soil corrective;
- Developing a pot experiment in a greenhouse to have controlled conditions (e.g., humidity, temperature, light hours), so that the conclusions are more precise;
- Performing tests with different plants, where the application of the mixtures is evaluated;
- Testing different formulations with other materials, like biomass ashes, slaughterhouse waste ashes, among others, to fulfil the legal requirements established regarding fertilizers;
- Use formulations with dried SS at different temperatures, in soil experiments;
- Assess the potential for soil contamination by microplastics present in SS and how it can affect the crops;
- Elaborate a Life Cycle Assessment to compare the impact caused by SS based fertilizers when compared to the commercialized fertilizers.

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APPENDIX

Appendix I – Statistical analysis of digested SS

Tables I.1-I.7 present the absolute values of the parameters used to perform the statistical analysis described in Chapter 5.1.

Table I.1. Absolute values of N and Nkjeldahl used to proceed with the statistical analysis.

n	N		NKjeldahl	
	Ref n	Value (%w/w)	Ref n	Value (%w/w)
1	1.	6.20	3.	1.50
2	6.	3.87	5.	1.37
3	8.	4.52	7.	1.61
4	9a.	6.80	14.	6.30
5	9b.	4.01	23.	3.72
6	9c.	4.41	24.	1.46
7	9d.	4.75	26a.	6.20
8	9e	6.54	26b.	6.20
9	9f.	6.23	27.	1.20
10	9g.	6.54	28a.	1.69
11	9h.	4.57	28b.	2.43
12	9i.	4.07	28c.	2.91
13	9j.	4.44	29a.	3.78
14	9k.	4.52	29b.	6.86
15	10.	1.63	29c.	4.20
16	11a.	4.88	29d.	2.94
17	11b.	3.59	29e.	4.96
18	12a.	1.70	29f.	2.80
19	12b.	1.60	29g.	3.50
20	12c.	3.70	29h.	4.06
21	12d.	1.80	29i.	3.92
22	12e.	2.30	29j.	4.20
23	12f.	3.90	29k.	3.12
24	13.	7.03	29l.	3.64
25	15.	3.06	29m.	4.48
26	16.	2.42	29n.	3.92
27	17a.	3.21	33.	1.50
28	17b.	3.21	35a.	2.86
29	17c.	4.23	35b.	2.94
30	19.	3.87	35c.	3.06
31	20.	1.50	35d.	4.14
32	21.	1.78	37a.	6.20
33	30.	2.80	37b.	6.20
34	32.	7.04		
35	34.	0.74		

Table I.2. Absolute values of P and P_2O_5 used to proceed with the statistical analysis.

n	P		P_2O_5 (%w/w)	
	Ref n	Value (%w/w)	Ref n	Value (%w/w)
1	1.	20.0	19.	5.67
2	8.	0.28	20a.	3.51
3	12.	0.78	20b.	7.44
4	16.	0.12	20c.	8.53
5	23a.	1.20	20d.	8.96
6	23b.	2.10	20e	8.21
7	23c.	2.20	20f.	7.67
8	23d.	1.50	20g.	5.35
9	23e.	1.00	20h.	7.23
10	23f.	2.40	20i.	6.80
11	24.	1.95	20j.	7.24
12	26.	1.33	20k.	5.67
13	27.	1.64	37b.	6.90
14	28a.	0.93	39a.	1.85
15	28b.	0.52	39b.	1.84
16	28c.	1.65	39c.	3.93
17	29.	0.00181	48b.	6.90
18	34.	2.46		
19	41.	3.10		
20	42.	2.35		
21	43.	1.46		
22	44.	0.12		
23	45.	0.99		
24	46a.	2.76		
25	46b.	2.32		
26	46c.	2.07		
27	46d.	2.90		

Table I.3. Absolute values of K and K_2O used to proceed with the statistical analysis.

n	K		K_2O	
	Ref n	Value (%w/w)	Ref n	Value (%w/w)
1	1.	0.30	19.	0.28
2	8.	0.16	20a.	0.63
3	12.	0.30	20b.	0.24
4	16.	0.15	20c.	0.37
5	23a.	0.05	20d.	0.32
6	23b.	0.12	20e.	0.21
7	23c.	0.14	20f.	1.13
8	23d.	0.16	20g.	1.07
9	23e.	0.28	20h.	1.02
10	23.f	0.12	20i.	0.44
11	24.	0.23	20j.	0.35
12	27.	0.20	20k.	0.22
13	28a.	0.25	39a.	0.45
14	28b.	0.25	39b.	0.31
15	28c.	0.26	39c.	0.42
16	34.	0.30	37a.	0.71
17	42.	0.26	37b.	1.44
18	45.	0.11	48a.	0.71
19	46a.	0.12	48b.	1.45
20	46b.	0.16		
21	46c.	0.11		
22	46d.	0.17		

Table I.4. Absolute values of S, Ca, Mg, Na, and Si used to proceed with the statistical analysis.

n	S		Ca		Mg		Na		Si	
	Ref n	Value (%w/w)	Ref n	Value (%w/w)	Ref n	Value (%w/w)	Ref n	Value (%w/w)	Ref n	Value (%w/w)
1	20a.	1.58	12.	3.03	8.	0.28	8.	0.12	23a.	0.02
2	20b.	4.51	19.	4.37	12.	0.25	23a.	0.07	23b.	0.07
3	20c.	2.03	20a.	2.39	16.	0.25	23b.	0.18	23c.	0.05
4	20d.	2.87	20b.	6.06	19.	0.66	23c.	0.10	23d.	0.07
5	20e.	3.31	20c.	3.64	20a.	0.53	23d.	0.14	23e.	0.08
6	20f.	1.73	20d.	2.17	20b.	0.99	23e.	0.07	23f.	0.05
7	20h.	2.82	20e.	4.38	20c.	1.27	23f.	0.04		
8	20i.	3.32	20f.	1.65	20d.	1.00	26.	0.59		
9	20j.	2.45	20g.	1.69	20e.	0.85	27.	0.001		
10	20k.	4.97	20h.	3.78	20f.	0.88	28a.	0.08		
11	21.	1.23	20i.	5.67	20g.	1.24	28b.	0.02		
12	22.a	1.67	20j.	3.19	20h.	1.50	28c.	0.14		
13	22.b	1.81	20k.	6.12	20i.	1.19	34.	0.18		
14	31.	0.94	23a.	0.67	20j.	0.65	37a.	0.24		
15	43.	0.53	23b.	0.89	20k.	1.10	37b.	0.16		
16			23c.	1.07	23a.	0.06	38.	0.001		
17			23d.	1.22	23b.	0.21	39a.	0.43		
18			23e.	0.51	23c.	0.31	39b.	0.01		
19			23f.	0.77	23d.	0.22	39c.	0.02		
20			24.	1.26	23e.	0.28	40a.	0.11		
21			26.	3.74	23f.	0.12	40b.	0.06		
22			28a.	0.67	26.	0.68	40c.	0.03		
23			28b.	0.69	27.	0.016	40d.	0.05		
24			28c.	0.65	28a.	0.54	40e.	0.10		
25			37a.	1.2	28b.	0.43	40f.	0.04		
26			37b.	2.76	28c.	0.65	40g.	0.08		
27			39a.	3.03	34.	0.65	40h.	0.08		
28			39b.	2.80	37a.	0.46	40i.	0.07		
29			39c.	3.48	37b.	0.79	40j.	0.08		
30			40a.	0.39	39a.	0.54	40k.	0.09		
31			40b.	0.72	39b.	0.36	40l.	0.10		
32			40c.	0.79	39c.	0.51	40m.	0.13		
33			40d.	2.80	40a.	0.34	40n.	0.05		
34			40e.	2.70	40b.	0.49	42.	0.45		
35			40f.	3.36	40c.	1.69	44.	0.06		
36			40g.	2.16	40d.	1.49	46a.	0.64		
37			40h.	1.19	40e.	0.99	46b.	0.46		
38			40i.	1.18	40f.	0.32	46c.	0.59		
39			40j.	1.63	40g.	1.44	46d.	0.52		
40			40k.	2.13	40h.	0.34	48a.	0.24		
41			40l.	1.64	40i.	1.96	48b.	0.16		
42			40m.	2.81	40j.	0.89	Na	0.12		
43			40n.	0.96	40k.	1.02	Ref.	0.07		
44			44.	4.50	40l.	0.49	8.	0.18		
45			45.	3.80	40m.	1.08	23a.	0.10		
46			46a.	3.68	40n.	0.42	23b.	0.14		
47			46b.	2.92	42.	1.16	23c.	0.07		
48			46c.	2.44	43.	0.19	23d.	0.04		
49			46d.	1.92	44.	0.71	23e.	0.59		
50			48a.	1.20	48a.	0.46	23f.	0.001		
51			48b.	2.76	48b.	0.79	26.	0.08		

Table I.5. Absolute values of B, Mn, V, Fe, and Cl used to proceed with the statistical analysis.

n	B		Mn		V		Fe		Cl	
	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (%w/w)	Ref n	Value (%w/w)
1	9a.	21.9	2.	595.7	29a.	16.7	1.	2.00	1.	0.80
2	9b.	79.6	9a.	200	29b.	8.90	8.	2.02	11a.	0.22
3	9c.	92.9	9b.	227	29c.	11.4	9a.	1.28	11b.	0.03
4	9d.	8.64	9c.	154	29d.	12.6	9b.	2.88	35a.	0.89
5	9e.	5.27	9d.	252	29e.	15.6	9c.	2.53	35b.	1.19
6	9f.	9.15	9e.	469	29f.	22.9	9d.	1.70	35c.	1.00
7	9g.	32.5	9f.	559	29g.	13.2	9f.	2.58	35d.	0.71
8	9h.	2.66	9g.	475	29h.	14.3	9g.	1.27		
9	9i.	6.49	9h.	242	29i.	19.2	9h.	1.23		
10	9j.	18.43	9i.	371	29j.	13.1	9i.	1.41		
11	9k.	9.94	9j.	535	29k.	14.8	9k.	2.02		
12	12a.	93	9k.	754	29l.	8.20	12a.	2.15		
13	12b.	70	12.	386	29m.	13.4	12b.	2.46		
14	12c.	79	15.	298.6	29n.	22.9	12c.	2.49		
15	12d.	77	27.	105.1			12d.	2.81		
16	12e.	50	23.	221			12e.	0.91		
17	12f.	88	28a.	168.79			12f.	2.89		
18			28b.	111.34			13.	0.35		
19			28c.	146.24			15.	1.14		
20			29a.	229			16.	1.03		
21			29b.	121			23.	4.64		
22			29c.	191			27.	0.21		
23			29d.	402			28a.	0.46		
24			29e.	245			28b.	0.36		
25			29f.	661			28c.	2.27		
26			29g.	258			31.	2.38		
27			29h.	542			32.	0.07		
28			29i.	465			35b.	4.14		
29			29j.	236			35c.	2.60		
30			29k.	357			35d.	4.47		
31			29l.	399						
32			29m.	228						
33			29n.	661						
34			31.	147.2						
35			32.	28.2						
36			34.	56.9						
37			37a.	238						
38			37b.	231						
39			37c.	139						
40			37d.	182						

Table I.6. Absolute values of the PTM used to proceed with the statistical analysis.

n	**Cd**		**Cr**		**Pb**		**Ni**		**Hg**		**Zn**		**Cu**	
	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)	Ref n	Value (mg/kg)
1	2.	1.17	2.	179.1	2.	671.2	2.	138.7	6.	0.5	2.	667.6	2.	162.6
2	3.	1.20	6.	28	3.	130	7.	6.4	7.	0.12	3.	270	7.	9.8
3	6.	3.00	7.	12	7.	10	8.	80	8.	0.64	7.	14	8.	197
4	7.	0.19	8.	86	8.	61	12a.	48	25a.	0.03	8.	3804	13.	979
5	8.	8.00	12a.	452	12a.	14	12b.	233	25b.	0.01	12a.	729	16.	20.5
6	13.	13.7	12b.	943	12b.	83	12c.	74	25c.	0.02	12b.	1402	19.	284
7	15.	3.64	12c.	561	12c.	90	12d.	15	25d.	0.01	12c.	1357	20.	375
8	20.	5.30	12d.	472	12d.	28	12e.	4	25e.	0.01	12d.	1577	23.	425
9	24.	1.70	12e.	429	12e.	7	12f.	54	28a.	1.22	12e.	93	25a.	267
10	25a.	0.20	12f.	528	12f.	71	15.	69.6	28b.	0.51	12f.	1441	25b.	272
11	25b.	1.83	15	144.6	15.	153.9	16.	22.6	28c.	1.01	13.	1268	25c.	240
12	25c.	1.29	16.	75.3	16.	144.4	19.	270.4	31.	0.77753	15.	64.6	25d.	204
13	25d.	1.01	25a.	70.1	19.	101.7	20.	56			16.	1808.4	25e.	277
14	25e.	0.89	25b.	80.2	20.	84	25a.	40.2			19.	592.7	26a.	140.8
15	26a.	1.00	25c.	42.7	24.	149	25b.	26.9			20.	1179	26b.	155.8
16	27.	4.04	25d.	40.1	25a.	197	25c.	16.2			23.	660	27.	174
17	28b.	0.49	25e.	39	25b.	152	25d.	14.9			24.	288.9	28a.	142.11
18	28c.	0.04	28a.	55.03	25c.	71	25e.	13.2			25a.	681	28b.	89.03
19	29a.	1.27	28b.	49.27	25d.	61	26a.	22.6			25b.	586	28c.	130.55
20	29b.	1.08	28c.	62.29	25e.	64	26b.	22.5			25c.	675	29a.	101
21	29c.	1.62	29a.	29	27.	35	27.	22.2			25d.	619	29b.	89.9
22	29d.	1.89	29b.	13.7	28a.	9.83	28a.	21.88			25e.	1079	29c.	95.8
23	29e.	2.36	29c.	22.4	28b.	20.75	28b.	19.19			26a.	757.2	29d.	100
24	29f.	9.50	29d.	22.7	28c.	31.12	28c.	28.014			26b.	581.1	29e.	161
25	29g.	2.81	29e.	56.9	29a.	16.4	29a.	20			27.	342	29f.	107
26	29h.	18.5	29f.	28.8	29b.	17.9	29b.	9.7			28a.	738.41	29g.	114
27	29i.	16.4	29g.	45.9	29c.	16.2	29c.	12.9			28b.	505.71	29h.	94.1
28	29j.	1.28	29h.	40	29d.	26.7	29d.	19.7			28c.	726.11	29i.	88.2
29	29k.	2.36	29i.	80	29e.	20.1	29e.	15.7			29a.	1350	29j.	97.2
30	29l.	0.68	29j.	25.4	29f.	24.1	29f.	11.8			29b.	564	29k.	102.9
31	29m.	2.16	29k.	34.6	29g.	38.5	29g.	49.7			29c.	1070	29l.	98.4
32	29n.	3.66	29l.	44.1	29h.	18.5	29h.	20.2			29d.	1170	29m.	68.8
33	31.	0.37	29m.	27.3	29i.	16.4	29i.	55.5			29e.	1680	29n.	149
34	33.	1.70	29n.	28.8	29j.	12.1	29j.	26.4			29f.	871	31.	205.3
35	35a.	8.00	31.	27.12	29k.	19.6	29k.	24.9			29g.	1510	32.	38.2
36	35b.	6.00	32.	13.4	29l.	10.6	29l.	27			29h.	1620	33.	377
37	35d.	4.00	33.	518	29m.	27.6	29m.	19.2			29i.	1100	34.	88
38	37a.	1.00	34.	11	29n.	28.2	29n.	26.1			29j.	1170	35a.	255
39			35c.	2	31.	29.03	31.	14.73			29k.	1046	35b.	283
40					32.	6.3	32.	9.3			29m.	676	35c.	213
41					33.	229	33.	63			29n.	871	35d.	270
42					34.	4.9	34.	5.5			31.	429.5	37a.	140.8
43					35a.	376	35a.	52			32.	21.7	37b.	155.8
44					35b.	456	35b.	68			33.	1344		
45					35c.	208	35c.	55			34.	130		
46					35d.	223	35d.	55			35a.	1805		
47							35a.	22.6			35b.	1756		
48							35b.	22.5			35c.	1620		
49											35d.	1667		
50											36	2786		
51											37a.	757.2		
52											37b.	581.1		

Table I.7. Absolute values of *Salmonella* spp. and OM used to proceed with the statistical analysis.

n	***Salmonella* spp.**		**OM**	
	Ref n	Value	Ref n	Value (%w/w)
1	7.	Absent	1.	50.00
2	25a.	Absent	2.	65.10
3	25b.	Absent	9a.	78.42
4	25c.	Absent	9b.	67.09
5	25d.	Absent	9c.	62.86
6	25e.	Absent	9d.	65.04
7	28a.	Absent	9e	60.31
8	28b.	Absent	9f.	72.05
9	28c.	Absent	9g.	75.77
10	37a.	Absent	9h.	56.87
11	37b.	Present	9i.	54.38
12	38.	Present	9j.	55.42
13	38.	Absent	9k.	67.74
14	38.	Absent	10	46.30
15	38.	Present	11a.	51.00
16	38.	Present	11b.	49.00
17	38.	Present	11c.	52.00
18	38.	Absent	12d.	47.00
19	38.	Absent	12e.	83.00
20	38.	Absent	12f.	51.00
21			15	46.30
22			17a.	49.00
23			17b.	39.25
24			17c.	44.12
25			18	47.26
26			26a.	67.50
27			26b.	74.30
28			28a.	51.62
29			28b.	63.55
30			28c.	56.01
31			30	40.77
32			31	61.97
33			32	72.50
34			35a.	44.00
35			35b.	44.00
36			35c.	45.00
37			35d.	37.00
38			36	44.80
39			37a.	67.50
40			37.b	74.30

Table I.8 present the list of references used to perform the statistical analysis showed in Chapter 5.1.

Table I.8. List of the references used to proceed with the statistical analysis.

Ref n	**Ref**
1	(Andersen, 2002)
2	(Eid et al., 2017)
3	(Grobelak and Napora, 2015)
4	(Roig et al., 2012)
5	(Antonkiewicz et al., 2020)
6	(Laura et al., 2020)
7	(Alvarenga et al., 2019)
8	(Kominko et al., 2018)
9 a-k	(Kominko et al., 2019)
10	(Xu et al., 2012)
11 a-b	(Werle and Dudziak, 2014)
12 a-f	(Mtshali et al., 2014)
13	(Wong and Su, 1997)
14	(Yilmaz and Temizgül, 2012)
15	(Delibacak et al., 2009)
16	(Rigueiro-Rodríguez et al., 2010)
17 a-c	López-Díaz et al., 2007
18	(Barros et al., 2006)
19	(Lakhdar et al., 2012)
20	(Paz-Ferreiro et al., 2012)
21	(Oleszczuk and Baran, 2005)
22	(Uysal et al., 2010)
23	(Perez-Espinosa et al., 2000)
24	(A. Grobelak et al., 2017)
25 a-e	(De Abreu et al., 2017)
26 a-b	(Alvarenga et al., 2015)
27	(Zoghlami et al., 2016)
28 a-c	(Romanos et al., 2019)
29 a-n	(Oleszczuk, 2006)
30	(Asses et al., 2018)
31	(Carbonell et al., 2009)
32	(Ashekuzzaman et al., 2019)
33	(Gascó and Lobo, 2007)
34	(Seleiman et al., 2020)
35 a-d	(Samaras et al., 2008)
36	(Leila et al., 2017)
37 a-b	(Alvarenga et al., 2016)
38	(Santos et al., 2020)

Appendix II – Climate conditions during the pot experiments

Table II.1. Weather conditions for the 4-week period of the pot experiments.

Date	H (%)	T (°C)	P (mm)	max. SR (w/m²)
21/set	80	19.72	0.254	946
23/set	83	20.94	1.524	1028
25/set	74	16.89	0	940
27/set	82	18.83	1.524	986
29/set	74	19.22	0	715
01/oct	80	17.56	1.016	937
03/oct	82	14.44	1.016	940
05/oct	86	15.61	0.508	935
07/oct	80	19.56	0	687
09/oct	83	17.94	0	784
11/oct	55	18.72	0	694
13/oct	83	14.50	0	870
15/oct	76	14.11	0	693
17/oct	78	14.22	0	773
19/oct	16	18.00	24.892	580

H – relative humidity; T – temperature; P – precipitation; max. SR – maximum solar radiation