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Screening risk assessment of organic pollutants and environmental impacts from sewage sludge management

Study to support policy development on the Sewage Sludge Directive (86/278/EEC)

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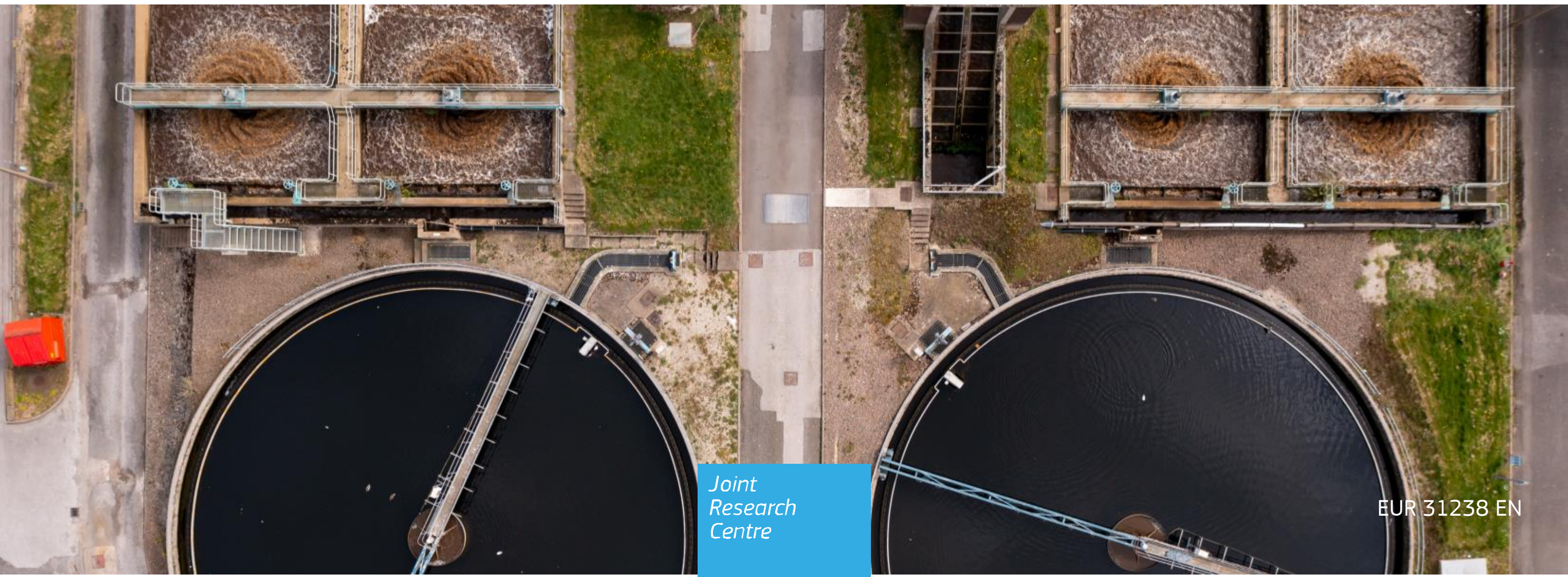

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Contents

Abstract

Sewage sludge is the residue following the treatment of wastewaters. It contains valuable agricultural resources, but also contaminants and pollutants that may pose risks to human health and the environment. Therefore, sewage sludge spreading on agricultural land is a controversial issue, and the occurrence of this practice varies widely amongst EU Member States. This study aims to assess impacts on the environment and human health from main sludge management routes occurring in the EU. For use in agriculture, it is found that a relatively small set of organic pollutants may cause significant risks to both humans and soil organisms following landspreading when present in concentration levels typically documented for sewage sludge. These organic priority contaminants are persistent in soils and have a bioaccumulative and toxic effect on humans and soil organisms. Secondly, it is indicated that sewage sludge management, other than landfilling, has a small impact on the overall global warming potential, and that the enforcement of best sludge management practices will only limitedly offset impacts resulting from wastewater treatment. Thirdly, sound management of sludge could partly contribute to reducing the depletion of the critical raw material rock phosphate. Also options other than landspreading can return a significant share of the phosphorus contained in sludge to agricultural land in a plant-available form. Altogether, it is indicated that a mix of sludge management routes, adapted to the local settings and needs, may be required to maximise benefits and minimise adverse impacts across the different sustainability dimensions affected by sludge management within the EU.

Key messages

- The risk screening assessment shows for a (relatively small) set of organic contaminants that these potentially cause significant risks to both humans and soil organisms, when present in concentrations levels typically documented for sewage sludge.

- A number of data gaps are identified for several contaminants that render their risk characterisation through modelling techniques incomplete. It can therefore not be excluded that risks potentially exist for further substances.

- Humans are found to be a more vulnerable end-point than soil organisms for the priority pollutants as unacceptable risk levels are reached for humans at lower pollutant loads due to bioaccumulation within the food chain.

- Only incineration at high temperatures ensures that all organic priority pollutants are effectively removed. Alternative risk mitigation measures involve setting sludge quality parameters and limiting sludge application rates, or a combination thereof. Sludge applications outside areas used in the food chain could also be further studied.

- Sewage sludge management has a small impact on the overall global warming potential, and the enforcement of best sludge management practices will only limitedly offset impacts resulting from wastewater treatment. The global warming assessment of sewage sludge management routes has a high degree of uncertainty for all pathways. With the exception of landfilling, routes (landspreading, anaerobic digestion, composting, and incineration) are not significant different in terms of global warming potential. Landfilling shows the worst performance, having at all times a net positive contribution to global warming due to methane leakages.

- Supplementary electric energy (estimated at maximum 4.4 TWh per year; or 0.16% of the total net electricity generation in the EU) could be recovered by subjecting sludge to anaerobic digestion and incineration with energy recovery.

- The landspreading of untreated and stabilised sludge returns phosphorus, a critical raw material, and nitrogen to agricultural land. Phosphorus can also be recovered from sewage sludge mono-incineration ashes, a material that can now be used as a secondary raw material in mineral phosphate fertiliser production processes. Nitrogen and phosphorus that could additionally be recovered from sewage using resource efficient sludge management options are reduced compared to other nutrient inputs onto agricultural fields (<2% and <6% for nitrogen and phosphorus in mineral fertilisers, respectively).

- Sewage sludge has potential to contribute to climate change adaptation as higher levels of organic matter help to retain moisture and limit runoff, but organic matter contents in sewage sludge are one to two orders of magnitude lower compared to other sources (manure, bio-waste).

- Guidelines to sewage sludge management have to carefully consider a risk versus resource efficiency trade-off. This suggests that a mix of sludge management options, able to adapt to the local settings and needs, may be required to maximise benefits and minimise adverse impacts across the different sustainability dimensions affected by sludge management within the EU.

- The study identified a number of recommendations for further work, mostly in relation to better quantifying the concentrations and risks of organic contaminants present in sludge as well as to elucidate the importance of sludge relative relative to other soil inputs (e.g. atmospheric deposition) as drivers for soil pollution.

Executive summary

Policy context

Sewage sludge is the residue following the treatment of wastewater. It mainly consists of water (>90% for undewatered sludge) and organic matter, which can contain a range of chemicals. Some of the substances that are found in sewage sludge can be recovered or used as soil amendments (e.g. nutrients) while others may raise environmental and health concerns. In the EU, between 6 and 9 million tonnes of dry sewage sludge is generated annually. Options to manage this sludge and derived materials involve spreading on agricultural land and forests, backfilling (i.e. refill an excavation with sludge), or disposal (e.g. co-incineration and landfilling). Some of the reuse and disposal techniques allow energy to be recovered from sewage sludge.

The objective of this study is to consolidate the technical and scientific knowledge base on the impacts and risks of sewage sludge management on the protection of human health, natural resources and the environment. With respect to pollutants, this study mainly focusses on organic compounds, thus excluding metals. A well-developed scientific evidence base on these aspects is important to support the policy framework on the EU's Sewage Sludge Directive (Council Directive 86/278/EEC)[1].

Screening risk assessment

This assessment presents evidence to identify and prioritise organic pollutants in sludge that may pose risks to soil organisms and human health when applied to agricultural land. The methodology starts with a characterisation of more than 1350 chemicals found in wastewaters and sludge. The long-list of contaminants includes well-known contaminants that have been identified since long in scientific literature and legislation as being of concern (e.g. polyaromatic hydrocarbons (PAHs), perfluorinated alkyl substances (PFAS)) as well as contaminants of more emerging concern (e.g. chlorinated paraffins, pharmaceuticals, personal care products, specialty industrial chemicals).

A risk screening model is developed that calculates the concentrations of numerous contaminants in soil, surface waters, and food following intermittent sewage sludge applications over various time intervals. Subsequently, human exposure from food consumption is estimated using modelled concentrations of contaminants in fish, root and leaf crops, meat, milk, and drinking water. The estimated exposures are then compared to predicted contaminant levels that are considered safe for soil organisms and humans. A number of data gaps are identified for several contaminants that render their risk characterisation through modelling techniques unviable for many compounds. While the conclusions of this work are based on our current understanding and characterisation of contaminants, it cannot be excluded that certain contaminants have been overlooked in this work.

The outcomes of the modelling show that a relatively small set of pollutants may cause significant risks to both humans and soil organisms, when present in concentrations levels typically documented for sewage sludge. Polychlorinated dibenzofuran and dioxins, PAH, long-chain PFAS, short and mid-chain polychlorinated paraffins, and to a lesser extent, alkylphenols, polychlorinated naphthalenes and phthalate acid esters are identified as priority pollutants. These include chemicals that are intentionally manufactured for household and industrial applications, as well as unintentionally released substances, for instance, from the combustion of wastes.

Common features of these pollutants are that they:

(i) persist and accumulate over time in the environment (mostly in soil),
(ii) have a high potential to bioaccumulate along food webs, and
(iii) are toxic to the environment and to humans, even at very low concentration levels.

Most of the priority pollutants are already subject to use restrictions and release reduction provisions under the chemicals legislation, including the POPs[2] and REACH[3] Regulation. In addition to impacts from chemicals,

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[1] Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture

[2] Regulation (EU) 2019/1021 of the European Parliament and the Council of 20 June 2019 on persistent organic pollutants

[3] Regulation (EC) No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency.

the use of untreated and biologically treated sludge on land may cause risks and adverse impacts due to the presence of microplastics and antimicrobial resistant genes in the sludge. Based on the available data, other substances, including most pharmaceuticals and personal care products, are considered of limited concern, even at high application loads of sewage sludge. The study shows that humans are a more sensitive end-point than soil organisms for the priority pollutants as unacceptable risk levels are reached for humans at lower pollutant loads.

Sludge treatment is often proposed as a risk mitigation measure. However, commonly used techniques (e.g. drying, lime stabilisation, composting, anaerobic digestion) do not necessarily ensure the reduction of priority pollutants to safe levels. Some pollutants, such as PFAS, are very resistant to biological and even thermal degradation. Only incineration at high temperatures ensures that all priority pollutants are effectively removed. Because risks are linearly correlated to contaminant loads, risk mitigation measures that involve setting sludge quality parameters and limiting sludge application rates, or a combination, could also be explored. In addition, sludge applications outside areas used in the food chain could be further studied. Such investigations would benefit from recent measurements of priority pollutant concentrations in different types of sludges (e.g. tertiary versus secondary sludge; untreated versus treated sludge; sludge originating from rural versus industrial areas). This may be important to better capture the effects of recent legislative release reduction measures for relevant pollutants.

Environmental and climate change impacts

The aim of this assessment is to assess different sewage sludge management routes in terms of their potential to contribute to climate change mitigation and adaptation, as well as resource efficiency. Selected management routes involved land spreading of untreated, lime stabilised, composted, and digested sludge, as well as landfilling and co-incineration followed by ash use/disposal outside agriculture. In addition, the emerging practice of sludge mono-incineration followed by the re-use of the ashes to manufacture a mineral phosphate fertiliser is studied because of its potential to increase the circularity of phosphorus, a critical raw material. The analysis considers the impacts from sludge processing and from the use- of sludge on land.

The ranking the routes on their climate change mitigation performance in a universal manner across the EU is impossible due to the large uncertainties associated with the technologies applied and emission variations across local geographic contexts. In general, landspreading untreated and treated sludges leads to net negative global warming impacts because sludge can partially displace mineral fertiliser and energy production, in addition to increasing carbon sequestration in soils. These savings are greater than the burdens to global warming resulting from sludge processing and greenhouse gases directly emitted during sludge processing, storage and use-on-land. Disposal routes that include sludge incineration result in estimated neutral net global warming impacts, but net savings or burdens could be observed across technology configurations. This also holds true for incineration ashes that are used for fertiliser production as well as for their use outside agriculture. Finally, landfilling shows the worst performance, having at all times a net positive contribution to global warming due to methane leakages.

In absolute terms, the study estimates that impacts from current sewage sludge management across the EU have a net positive effect on global warming (+1 million tonnes CO_2-eq per year). The application of optimum sewage sludge management routes across the EU may result in a net removal of CO_2 from the atmosphere (-2 million tonnes CO_2-eq per year), that could only to a limited extent offset the estimated upstream emissions from wastewater collection and treatment (25-30 million tonnes CO_2-eq per year).

When applied to agricultural land, sewage sludge also has potential to contribute to climate change adaptation as higher levels of organic matter help to retain moisture and limit runoff. The amounts of organic matter in sewage sludge are, however, one to two orders of magnitude lower compared to other sources, such as manure and bio-waste. Nonetheless, sewage sludge may be a valuable organic asset locally (e.g. in EU regions with a more extensive livestock production system or on soils characterised by a low organic matter content).

Phosphorus is a critical raw material, and its finiteness and criticality for EU agriculture warrants an assessment of how sewage sludge management could contribute to closing the biogeochemical phosphorus cycle and improving resource efficiency. Phosphorus in sewage sludge equals about 12% of the total mineral fertiliser inputs (8% relative to overall inputs including feed), but at present it is estimated that less than half of this amount ends up on agricultural land. Hence, more than 50% of the phosphorus from sewage sludge is irreversibly lost after co-incineration and landfilling. The implementation of innovative techniques, such as phosphorus recovery from mono-incineration ashes, is still limited.

It is indicated that the climate change and resource efficiency assessment generally supports the waste hierarchy that promotes recycling over recovery and disposal. However, the overall environmental impacts of the management routes may vary based on local conditions, including technologies applied and agricultural needs for nutrients and organic matter of the land close to wastewater treatment plants.

Conclusions

Sludge management has to consider different sustainability dimensions, including risks for the environment and human health from organic and inorganic chemicals and pathogens when sludge is further reused, resource use efficiency and impacts on climate change mitigation and adaptation. This study shows a risk versus environmental trade-off across sludge management routes. Limited processing generally has a better environmental performance than sludge incineration and landfilling, but may not be fully effective in reducing pollutant loads, especially when high amounts of sludge are intensively landspread on a limited agricultural land area. Disposal and more intensive sludge processing options typically will not fully contribute to resource efficiency and addressing climate change, but may reduce environmental and health risks from food consumption. Innovative options, such as sludge mono-incineration followed by ash acidulation to retain phosphorus in a plant-available form, can deliver middle-ground impacts. Overall, it is concluded that the application of a mix of established and innovative techniques, as a function of local settings and needs, may help to maximise benefits and minimise adverse impacts on the different sustainability dimensions affected by sludge management within the EU.

1 General objective and structure of the report

The general objective of this report is to support policy development on the Sewage Sludge Directive (86/278/EEC; hereafter SSD)[4] by providing techno-scientific evidence on (i) risks from organic pollutants, contaminants of emerging concern and pathogens in sludge applied to agricultural land, and (ii) environmental impacts arising from sewage sludge management in the EU, including climate change and resource depletion. The report aims to increase the techno-scientific knowledge base on those issues that have been identified in previous works and policy documents as relevant for further policy development.

The report contains six main sections. Section 2 brings forward a brief introduction to sewage sludge management in the EU. The risk screening assessment for chemical substances is developed in section 3 using a model-based calculation approach. This aspect is further complemented by a qualitative review of risks from other contaminants of emerging concern, such as antimicrobial resistance genes and microplastics (section 4). Section 5 assesses the impacts of the use of land of sludge and treated sludge on the soil biogeochemical carbon (C), nitrogen (N) and phosphorus (P) cycles. The result thereof are plugged into a life cycle assessment model that compares the global warming potential of different sludge treatment and management options (section 6). The limitations of our work and further research recommendation to take this work forward are provided in section 7. Finally, section 8 develops the general conclusions of this study. This report is annexed by sections 9-15 that provide supplementary information on the methodology and results of the assessment.

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[4] Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:31986L0278)

2 Background on sewage sludge management in the EU

Sludge is defined in the SSD (86/278/EEC) as:

(i) residual sludge from sewage plants treating domestic or urban wastewaters and from other sewage plants treating wastewaters of a composition similar to domestic and urban wastewaters;
(ii) residual sludge from septic tanks and other similar installations for the treatment of sewage;
(iii) residual sludge from sewage plants other than those referred to in (i) and (ii);

According to Eurostat[5], between 5.2 and 8.6 million tonnes of dry matter of sewage sludge have been managed yearly between 2004 and 2016 in the EU (Wood E&I GmbH, 2021) (Figure 1). The Eurostat terminology 'sludge disposal' involves sludge management options agricultural use, landfill, incineration, compost and other applications, and other.

Figure 1: Eurostat data on the amounts of sewage sludge (dry matter) disposed for the period 2004 – 2016 (reprinted from Wood E&I GmbH, 2021)..

For the year 2016, Member States (MS) reporting data on the implementation of the SSD indicated a total volume of about 6.7 million tonnes of sludge (dry matter). These numbers are similar to Eurostat data of the year 2016, but the latter data are incomplete as no information is available for certain MS, such as Italy, Belgium, and Denmark. After gap filling with data from the period 2010-20115, the updated Eurostat 2016 indicate a tonnage of about 7.5 million of tonnes sludge (dry matter). Sludge management routes for this dataset involve agricultural use (35%), incineration (31%), compost and other applications (12%), landfilling (12%) and other uses (10%). Main uncertainties exist related to the final end use of the categories "compost and other applications" and "other uses" (Anderson et al., 2021). Nonetheless, these data largely align to the conclusions of the report of Wood E&I GmbH (2021) who indicated that the total percentage of sewage sludge used in agriculture falls between around 35% and 50% of the total sewage sludge produced (roughly 2.6 – 3.8 million tonnes (dry matter); based on a total estimated volume of 7.5 million tonnes being disposed). Incineration is also a main sludge management route, with estimated volumes corresponding to 2.3 million tonnes (dry matter).

Large differences in the proportional contribution of sewage sludge disposal routes exist among MS for the fate of sewage sludge (Figure 2). Countries with a high population and livestock density, and strict restrictions on landfilling, incinerate high proportions of their sewage sludge (e.g. Netherlands, Belgium, Germany; 82-98%). Other Member States (e.g. Ireland, Spain; >80%) apply large amounts of the sewage sludge directly on land. Finally, Member States as Greece, Italy, Romania, and Croatia landfill significant amounts of sludge (28-86%) (Figure 2).

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[5] Eurostat, Sewage sludge production and disposal, accessed on 28 September 2021 (http://appsso.eurostat.ec.europa.eu/nui/show.do?lang=en&dataset=env_ww_spd)

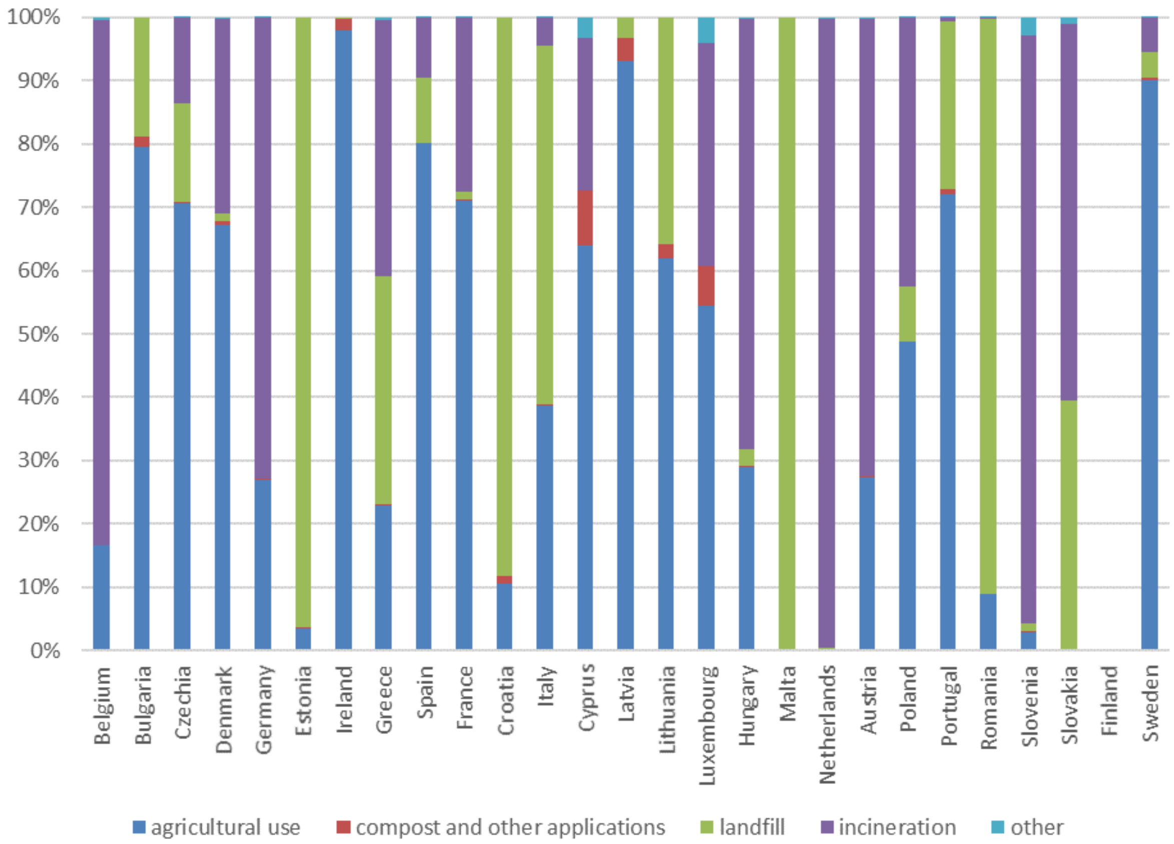

Figure 2: Relative importance of sludge management options within different EU Member States (based on Eurostat 2016 data, with gap filling performed for some Member States based on 2010 – 2015 data).

In addition to primary treatment (e.g. thickening, dewatering), sludge can undergo biological, chemical or heat treatment, long-term storage or any other appropriate process as a secondary treatment so as significantly to reduce its fermentability and the health hazards resulting from its use ('treated sludge' according to the SSD) (Kacprzak et al., 2017). Common techniques that enable further use in agriculture involve lime treatment, solar and thermal drying, anaerobic digestion and composting (Table 1). In addition, a mineral phosphate fertiliser can be manufactured from mono-incinerated sludge after acidulation, with a chemical composition that is similar to a rock phosphate-derived mineral fertiliser (Kabbe, 2017; Huygens et al., 2019; Tonini et al., 2019). Common sludge disposal options co-incineration and landfilling do not return organic matter and/or nutrient to agricultural land. Finally, sludge is used at occasions for other purposes such as backfilling and use in forestry. Hydrothermal carbonisation and pyrolysis are other alternative routes, but minor sludge volumes are currently processed through these pathways. This report will compare and analyse different sludge management options, with a focus on sludge management techniques that remove of biological and chemical pollutants present in sewage sludge.

Table 1: Overview of main secondary treatment (e.g. lime stabilisation, composting, anaerobic digestion, P-fertiliser production from mono-incineration ash) and final end uses of the resulting materials (e.g. use-on-land, landfill, use as construction material of co-incinerated ash)

Sludge treatment/management technique	**Description of technique**	**Main objectives**
Lime stabilisation (Wong and Selvam, 2006; Farzadkia and Bazrafshan, 2014; Teoh and Li, 2020; Anderson et al., 2021)	Lime treatment is a sludge stabilisation technique that involves the addition of CaO or $Ca(OH)_2$ to sludge. The European Lime Association recommends addition of 50% - 90% CaO per unit dry solids for 75 min to treat sludge at >55°C and pH > 12, or the addition of 20%-40% CaO or equivalent Ca(OH)2 per unit dry solids for 3 months.	• Sludge stabilisation (hygienisation and reduction of biological pathogens; odour nuisance reductions) • Increased acid neutralising capacity for receiving agricultural soils; • Decreased mobility of metals.
Composting (Guardabassi et al., 2003; Uçaroğlu and Alkan, 2016; Anderson et al., 2021)	Composting is an aerobic process during which microorganisms convert organic substrates, such as sewage sludge or other organic materials, into stabilized organic matter with production of heat. Different groups of microorganisms develop and become predominant during the different phases of composting (mesophilic phase, thermophilic phase and the maturation phase). A composing process normally takes several weeks, with temperatures that may range between 45°C and 80°C. Several configurations for composting exist (e.g. windrow systems, aerated static pile systems, enclosed (in-vessel) systems.	• Sludge stabilisation (hygienisation and reduction of biological pathogens; odour nuisance reductions) • Volume and moisture reduction; • Improvement of physical properties (e.g. aeration, water retention) of the receiving soil • Partial removal of certain non-persistent organic contaminants.
Anaerobic digestion (Ruffino et al., 2020; Anderson et al., 2021)	Anaerobic digestion is the bacterial breakdown of organic materials in the absence of oxygen resulting in the production of biogas. The anaerobic digester is mostly operated at the mesophilic temperature range (35-40°C), whereas thermophilic digestion involves higher temperatures (up to > 55-70°C).	• Renewable energy production, heat recovery; • Sludge stabilisation (hygienisation and reduction of biological pathogens; odour nuisance reductions) • Volume reduction; • Partial removal of certain non-persistent organic contaminants.
P-fertiliser production from mono-incineration ash (Kabbe et al., 2015; Huygens et al., 2019)	Innovative production processes exist to transform the phosphorus present in sewage sludge into a mineral P-fertiliser. The most common process is a two-stage manufacturing process that starts with the mono-incineration of sewage sludge at temperature of > 850°C - 900°C, typically in a fluidised bed reactor. Afterwards, most processes acidulate (fly) ashes, and the mixture is then potentially purified and/or mixed with other P-rich materials. A high recovery (> 80 - 98%) of the phosphorus present in sewage sludge can be achieved, but nitrogen and organic carbon are destroyed during the incineration process. Alternative manufacturing processes exist based on thermal processing steps instead of acidulation.	• Renewable energy production, heat recovery; • Sourcing of secondary raw materials for P-fertiliser production • Sludge volume reduction; • Sludge decontamination, as biological and chemical contaminants are completely or largely destroyed during the process. • Microplastics removal
Use-on-land	Untreated and treated (e.g. composted, digested, lime stabilised) sludges and P-fertilisers derived from sewage sludge can be used on agricultural land as a nutrient source (fertiliser), and/or to improve the soil environment for crop growth (soil	• Crop growth • Increase resource efficiency and decrease reliance on primary raw materials (e.g. mineral fertilisers, peat).

	improver or liming material)	- Soil restoration and organic matter recycling
Co-incineration (Svoboda et al., 2006)	Co-incineration is a sludge disposal technique where sewage sludge is thermally oxidised together with other materials (mostly municipal solid waste, but possibly also fossil fuels or minerals for cement production). The resulting fly and bottom ashes are either used in construction materials or discarded, resulting in the permanent removal and destruction of nutrients and organic matter from the biogeochemical cycle.	- Renewable energy production, heat recovery; - Sewage sludge disposal.
Landfilling (Stauffer, 2021)	A landfill is an engineered pit, in which layers of waste are filled, compacted and covered for final disposal. Engineered landfills consist of a lined bottom; a leachate collection and treatment system; groundwater monitoring; gas extraction (the gas is flared or used for energy production) and a cap system.	- Renewable energy production, heat recovery; - Sewage sludge disposal.

3 Screening risk assessment for chemical pollutants in sludge

3.1 Objectives and scope

The objective of this section is:

- To develop a methodology that enables the identification of relevant chemical pollutants that are present in sewage sludge and that pose environmental or human health risks when applied on agricultural land. Specifically, the methodology will enable to assess impacts of sewage sludge applications on soil organisms and humans via the intake of food grown on sewage-sludge amended soils;
- To apply the methodology to bring forward evidence to be able to identify relevant organic pollutants that are present in sewage sludge and that pose risks related to the use of sewage sludge as well as a prioritisation of those pollutants that cause most risk.

Although the same method can be applied to assess environmental and health risks from metals, the current study exclusively focuses on identifying relevant organic pollutants because these are currently considered in the SSD. Metals are already considered as relevant pollutants for sludge applied on agricultural land, and covered under the SSD. For relevant organic pollutants, information on baselines of soil conditions relevant to sludge applications will be presented, as well as an estimate of the removal those pollutants may typically undergo during the most common sludge treatment processes. Finally, the availability of screening methods for sampling and analysis of relevant pollutants (e.g. international and national standards, technical standards) in sewage sludge is reviewed.

The scope of this study is limited to soil organisms and human health impacts resulting from indirect exposure via the environment through the consumption of fish, root and leaf crops, meat, milk, drinking and water. The impacts on surface water organisms will not be considered in this study as the contribution of relevant sludge contaminants to waters is for most substances small compared to contaminant loads that are present in wastewater treatment effluent, and soil organism act as the primary species impacted by use-on-land of sludges. Also, risks for sediment organisms and ground water quality were not directly studied in this work.

3.2 Methodology

The present report provides a provisional modelling assessment as to whether selected organic contaminants in sewage sludge could potentially pose a risk to soil ecosystems after soil amendment. It should by no means be considered an exhaustive assessment that extensively covers all substances that could possibly end up in sewage sludge. The selection of substances under assessment is based on a set of criteria, assumptions, availability of ecotoxicity data and the fate of substances in the environment.

3.2.1 Exposure assessment tool

The environmental or health risk of a substance is assumed to be determined by the application rate of the substance on agricultural land, its stability and physico-chemical properties within the atmosphere-soil-plant-water continuum, and its (eco)toxicology that indicates the sensitivity of biota and humans to exposure.

The assessment of the local environmental exposure was carried out based on a tool developed by the European Crop Protection Association (ECPA) that has been further adapted and validated by the JRC (see section 13.1). The Local Environment Tool (LET)[6] is referred to in the REACH R.16 guidance document to fine-tune models developed by the European Union System for the Evaluation of Substances (EUSES) as these do not account for direct intermittent releases to soil in a local assessment. Conceptually, a treated 1 ha agricultural field with an adjacent shallow waterbody is simulated. The LET uses the calculations described in the REACH R.16 (2016) guidance, as well as the “Step 2” calculation approach for surface water devised by the Forum for the Co-ordination of pesticides fate models and their use (FOCUS, 2003). Based on EUSES guidelines (REACH R.16), a sludge application rate to agricultural land is assumed of 5 tonnes dry weight ha^{-1} yr^{-1}. This assumed value a realistic high-end sludge application rate in the EU (Collivignarelli et al., 2019).

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[6] The ECPA-LET tool is freely available at http://www.ecpa.eu/information-page/regulatory-affairs/reach.

Impacts are assessed in the short (after a single year), mid- (after 10 years of continuous applications), and long-term (after 100 years of continuous applications).

To assess the impact on soil organisms, the ECPA-LET model calculates average contaminant concentrations in the top soil layer (20 cm depth), averaged over a 30-day period following sludge applications. The model departs from the contaminant application loads, and takes into consideration contaminant losses via volatilisation, leaching and biodegradation in soil. All calculations are outlined in section 13.1. To assess impacts on soil organisms, standard tests on different trophic levels are available for the soil compartment, including a functional test with soil microorganisms (nitrogen transformation, 28-day; OECD 216) and ecotoxicological tests with soil dwelling invertebrates (Collembola; OECD 232), earthworm/enchytraeid; OECD 222) and plant species (terrestrial plants; OECD 208). In practice, however, data on no-effect concentrations for soil organisms are hardly ever available for most contaminants. However, it can be assumed that contaminant uptake by soil organisms occurs via the soil and its pore water, and that aquatic and soil organisms are equally sensitive. Therefore, a predicted no-effect concentrations for soil organisms is calculated from aquatic organism toxicity data using default methods (equilibrium partitioning).

The daily human intake resulting from indirect exposure via the environment is estimated using modelled concentrations of the contaminant in fish, root and leaf crops, meat, milk, drinking and water, based on a standard diet (Figure 3). To derive human intake of fish, food crops, meat, milk and water, the contaminant concentrations in these media were estimated based on average soil and surface water concentration over a 180-day period after sludge application, thus accounting for biodegradation, volatilisation and leaching in soils as well as for biodegradation and water renewal in surface water.

It is noted that these “human via the environment” calculations show a degree of conservatism as it assesses the impact of exclusive and continuous intake of produce from soils amended with sewage sludge at a yearly application rate of 5 tonnes of sewage sludge per hectare as a worst-case scenario. If one would assume that sewage sludge is homogeneously applied across all agricultural land in the EU, the “average EU produce” consumed by the EU population would have been grown on soils receiving a sewage sludge application rate that is about 2 orders of magnitude lower. Still, it cannot be excluded that some people consume a high share of food products from geographic EU areas receiving higher than average sewage sludge loads; this justifies the use of an additional safety assessment factor. Moreover, many data used in this screening assessment have been collected based on data extrapolation (e.g. equilibrium partitioning methods) and from data repositories; in such case, another additional safety assessment factor to account for data quality is justified. Altogether, the approach is considered valid to identify the most relevant pollutants in sewage sludge.

3.2.2 A stepwise approach

A two-step approach is applied for exposure assessment (Figure 3). As the first step, the exposure to a contaminant is estimated in order to target those compounds that might pose an environmental or health concern based on conservative assumptions. If the estimated exposure exceeds ecotoxicological or toxicological reference values, a more accurate method of dietary exposure assessment is used; if it is below the reference value, no further assessment is conducted.

Step 1 consists out of a broader screening step that models the exposure of soil organisms and humans to sludge-derived contaminants, using data that were retrieved using semi-automated data collected methods (Figure 3). Step 2 then involves the verification and updating of the model input data based on state-of-the-art literature, complemented with most recent data on contaminant concentrations in soils and waters (Figure 3). Such “background concentrations” may originate from sources other than sewage sludge such as atmospheric deposition, municipal and industrial wastewater treatment effluents discharged in surface waters, and other fertilising materials applied to soils.

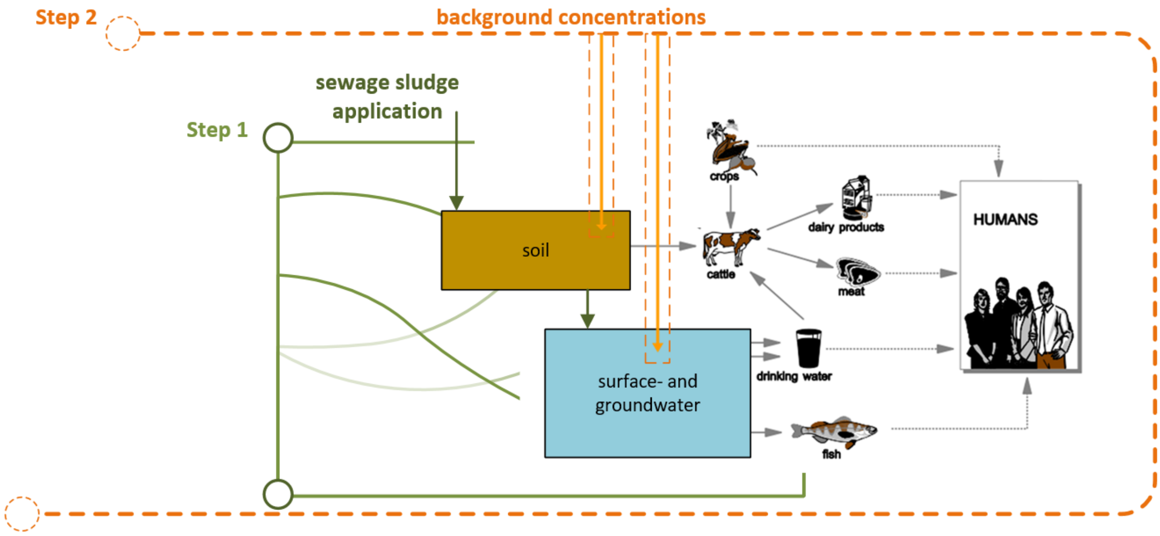

Figure 3: Applied step-wise approach for the exposure assessment. Step 1 involves a local assessment based on the exclusive application of sewage sludge to agricultural soils that is mainly based on data retrieved from data repositories in a semi-automated manner (depectived in green). If the estimated exposure exceeds a risk characterisation ratio above 1, a Step 2 refinement is executed where model input data is further corroborated and background concentrations of the respective contaminant in soils and water bodies is taken into consideration (depicted in orange).

3.2.2.1 Step 1

An extensive list of 1350 potential contaminants in sewage sludge was set up, based on an initial list of 1196 contaminants identified in Urban Wastewater Treatment Plants (UWWTPs). This initial list was based on regulated substances (and relevant for wastewater), monitoring data from UWWTPs[7] and other substances identified as substances of concern by experts or national authorities (e.g. persistent, mobile and toxic). This initial list of contaminants was further complemented with >150 additional entries that were identified from complementary sources, including (national) legislation on sewage sludge land application, as well as techno-scientific publications, reports and public databases that focus on contaminants of emerging concern in sewage sludge (Kinney et al., 2006; Smith, 2009; Clarke and Smith, 2011; Danish Ministry of the Environment, 2012; Mailler et al., 2014; Petrie et al., 2015; Deleebeeck et al., 2021; Rigby et al., 2021).

Available physico-chemical and (eco)toxicological properties required as input parameters were retrieved from data repositories. When available, measured data were prioritised over estimated data using e.g. QSAR software. To that purpose, in addition to the same literature references and datasets used for setting up the initial list, a total of 8 extra databases were used to extract relevant data on properties and toxicity data of the contaminants (when available):

- COMPTOX (US-EPA) database;
- Norman EMPODAT;
- ECOTOC;
- Envirotox;
- IRIS (US-EPA); and
- OpenFOODTOX (EFSA).

Efforts were undertaken to extract the following parameters:

- contaminant concentration in sludge before and after treatment;
- n-octanol/water partition coefficient (Kow);
- organic carbon-water partition coefficient for organic compounds (Koc);

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[7] Based on data from the Helsinki Commission (HELCOM), Dutch monitoring on micro-pollutants in influent and effluent from wastewater treatment plants (WATSON) and Helmholtz Centre for Environmental Research (UFZ)

- solubility in water;
- vapour pressure;
- molar weight;
- biodegradation half-life;
- Predicted No-Effect Concentrations (PNEC) for surface waters; and
- Human toxicity data (Reference Dose, Tolerable Daily Intake, Acceptable Daily Intake, etc.).

The $PNEC_{terrestrial}$ (predicted no-effect concentrations for the terrestrial soil compartment) were estimated from the well-documented $PNEC_{surface\ water}$ values using the equilibrium partitioning method.[8]

Preliminary data analysis indicated that data extraction was generally successful for most parameters, with the exception of safe limit values for human consumption and contaminant concentration values in sludge. To address these issues, safe limit values for human consumption were retrieved on a case-by-case search, with a focus on (i) contaminants that had been identified in the techno-scientific literature and by national competent authorities (in legislation) as relevant, and (ii) contaminants for which total daily intake by humans was highest (approx. above 0.01 mg day^{-1}). The derivation of safe limit values for contaminants based on a detailed review of toxicological data and studies is beyond of the scope of this study. Moreover, for many of the long-listed contaminants, it may simply be impossible to develop safe limit values for human consumption based on the current state of toxicological knowledge and studies available. Missing contaminant concentration values in sludge were estimated from their concentrations in wastewaters and the application of sludge-water partitioning coefficients (cfr. SimpleTreat modelling). In sum, a hybrid approach is applied that enables to focus on the "usual suspects" that have been identified in previous work as being of possible concern, and contaminants of unknown concern within landspreaded sewage sludge that may have been overlooked in previous studies.

Risk characterisation ratios (RCR) were then calculated after a period of 10 years of continuous sludge applications on agricultural land. This mid-term period enables to consider the possible accumulation of persistent compounds in soil.

$RCR_{soil\ 10\ years} = PECsoil_{soil\ 10\ years} / PNEC_{soil}$

With:
$RCR_{soil\ 10\ years}$ = Risk Characterisation Ratio for the soil compartment, estimated after a period of 10 years of continuous sludge applications on agricultural land (-);
$PEC_{soil\ 10\ years}$ = Predicted Environmental Concentrations for the contaminant (mg kg^{-1}) in the soil compartment, estimated after a period of 10 years of continuous sludge applications on agricultural land;
$PNEC_{soil}$ = Predicted No-Effect Concentrations for a contaminant (mg kg^{-1}) in the terrestrial soil compartment.

$RCR_{human\ 10\ years}$ = (Total Daily Intake $_{10\ years}$ / Body Weight) / SLHC

With:
$RCR_{human\ 10\ years}$ = Risk Characterisation Ratio for humans (-) estimated after a period of 10 years of continuous sludge applications on agricultural land (-);
Total Daily Intake$_{10\ years}$ = Daily contaminant intake from food and water (mg day^{-1}), estimated after a period of 10 years of continuous sludge applications on agricultural land (-);
Body Weight = Default value, 70 kg
SLHC = Safe Limit for Human Consumption (mg kg^{-1} body way day^{-1})

Risk characterisation ratios above 1 indicate the presence of a potential concern, after which data for identified contaminants are verified and potentially updated based on a manual literature search in Step 2 (see section 3.2.2.2).

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[8] $PNEC_{terrestrial} = K_{soil-water} / RHO_{soil} \times PNEC_{surface\ water} \times 1000$; with $PNEC_{terrestrial}$ = predicted no-effect concentrations in soil [mg kg^{-1}]; RHOsoil = bulk density of wet soil [1700 kg m^{-3}]; $PNEC_{surface\ water}$ = predicted no effect concentration in water [mg L^{-1}]

The default model (section 13.1) uses Kow to assess the distribution of the contaminant in the environment and the food chain. For fish, root crops, leaf crops, meat and milk, EUSES estimates the concentrations in these food products using methods that rely on log Kow as equivalent measured accumulation factors are not straightforwardly accessible or existent for many substances. For non-ionisable organic contaminants that accumulate primarily in fat tissue, these assumptions are generally accepted as valid. Instead, acids and bases dissolve in water and, because they increase the concentration of either protons or hydroxide ions, they suppress water self-ionization. With the possible exception of bases, these compounds are mostly partitioned to the water phase, and show generally a lower potential to bioaccumulate in fish, soil fauna and plant biomass. For such substances, log Kow is a conservative estimate of the lipid partitioning because the overall partitioning for all of its species (neutral + ionic) may be considerably less when the compound's potential to ionise is taken into account (ECOTOC, 2013). This implies that our step 1 model will overestimate the human intake of ionisable substances from food crops, meat and dairy.

3.2.2.2 Step 2

Starting from Step 1 results, the Step 2 analysis verifies and updates model input data using a targeted literature search for compounds that were considered as posing a potential risk to human health. In addition, data were collected on concentrations in soils and surface water under conditions without sewage sludge applications on land ("background concentrations"), where possible. This enables an initial assessment of the relative contribution of sewage sludge to the contamination issue that is likely the result of pollution from multiple sources (e.g. atmospheric deposition, industrial and municipal wastewater treatment effluents, other fertilising materials). |n case such analysis confirms the importance of the respective contaminant present in sewage sludge as contributing to the risk for human health, a further assessment of the impact of sludge processing as well as the availability of standardised measurement methods is executed.

3.3 Human health assessment

3.3.1 Step 1 results

3.3.1.1 Data comprehensiveness

$RCR_{human\ 10\ years}$ could be estimated for a total of 173 contaminants. For 619 additional contaminants for which sewage sludge concentrations could be retrieved, no RCR_{human} 10 years could be calculated, mostly because of the absence of a Safe Limit for Human Consumption (SLHC). Since targeted efforts were undertaken to list SLHC when daily human intake was above 0.01 mg day^{-1} (see section 3.2.2.1), these substances should, however, be highly toxic to humans (SLHC < 1.4E-04 mg kg^{-1} body weight day^{-1}) to be of human health concern. Therefore, a likelihood exists that these highly toxic substances have already been identified as being of concern, and its SLHC is already listed in the data repositories consulted.

The 173 entry list of organic contaminants assessed for human health risks belong to different substance categories that have been identified in scientific literature as of possible environmental and/or health concern (Kinney et al., 2006; Smith, 2009; Clarke and Smith, 2011; Danish Ministry of the Environment, 2012; Mailler et al., 2014; Petrie et al., 2015; Deleebeeck et al., 2021; Rigby et al., 2021). Specifically, the list covers substances that classify as long-chain perfluoroalkyl susbtances (long-chain PFAS), polyaromatic hydrocarbons (PAH), polychlorinated dibenzo-p-dioxin and furans (PCDD/Fs) and polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), organophosphate flame retardants, alkylphenols, polychlorinated alkanes/paraffins, polychlorinated naphthalenes, organotin compounds, chlorobenzenes, pesticides and biocides, synthetic musks, phthalate acid esters, polydimethylsiloxanes, antibiotics and other drugs, steroids, surfactants, quaternary ammonium compounds, etc.

3.3.1.2 Prioritised substances for step 2 assessments

$RCR_{human\ 10\ years}$ greater than 1 were detected for different compounds belonging to a wide variety of chemical and use classes in the step 1 analysis of the assessment (Table 2).

Table 2: Chemical substances identified in the Step 1 screening risk assessment as having a $RCR_{human\ 10\ years}$ greater than 1, the 10 classes above the horizontal black line will be taken forward for a more detailed analysis.

Substance name	**class**
Benzo[a]pyrene and other polyaromatic hydrocarbons (PAH)	**PAH**
2,3,7,8-TCDD and other polychlorinated dibenzo-p-dioxins (PCDDs) and furans (PCDFs), and dioxin-like polychlorinated biphenyls (dl-PCBs)	**PCDD/F + dl-PCBs**
PFOA, PFOS and other long-chain PFAS	**long-chain PFAS**
Nonylphenol and octylphenol, their ethoxylates.	**Alkylphenols**
Short- and mid-chain parrafins	**Polychlorinated alkanes**
Tributyltin, dibutyltin	**Organotin compounds**
Di(2-ethylhexyl)phthalate (DEHP)	**Phthalate acid esters**
1,2,3,4,5,6,7-HpCN (PCN-73)	**Polychlorinated naphthalenes**
Tris(2-ethylhexyl) phosphate (TEHP) and other organophosphate flame retardants	**Organophosphate compounds**
Decamethylcyclopentasiloxane (D5) and other polydimethylsiloxanes	**Polydimethylsiloxanes**
Aldrin, dieldrin, heptachlor, DDT, and other organochlorine pesticides (e.g.	Organochlorine pesticides
3,3',4,4',5,5'-hexabromobifenyl (PBB-153)	Bromobiphenyls
4,4',4''-(1-Methyl-1-propanyl-3-ylidene)tris 2-(1,1-dimethylethyl)-5-methyl-phenol	Phenolic compounds

With a view to prioritise efforts, it was decided to take 10 classes forward in the assessment for step 2 analysis (indicated in bold in above horizontal line in Table 2). Relevant contaminants for each class have been selected based on occurrence data in sewage sludge, relevance for human health, and availability of toxicological and physico-chemical data to perform the risk modelling.

3.3.1.3 Substances that are not taken forward in the assessment

In spite of their estimated $RCR_{human\ 10\ years} > 1$ during step 1, organochlorine pesticides, bromobiphenyls and 4,4',4''-(1-Methyl-1-propanyl-3-ylidene)tris 2-(1,1-dimethylethyl)-5-methyl-phenol were not taken forward to step 2. The reasons involve relate to a prioritisation of the available resources for this study, as well as technical reasons. Organochlorine pesticides have been phased out since long the EU, and the available dataset on sludge concentrations lacks data of recent measurements. For polybrominated biphenyls (PBBs), the EFSA CONTAM Panel concluded that the risk to the European population from exposure to PBBs through the diet is of no concern (EFSA Panel on Contaminants in the Food Chain, 2010). Since PBBs are no longer produced or used in Europe and taking into account low and declining environmental concentrations, the CONTAM Panel concluded that PBBs are a low priority for further research or monitoring efforts (EFSA Panel on Contaminants in the Food Chain, 2010). For phenolic compounds, such as 4,4',4''-(1-Methyl-1-propanyl-3-ylidene)tris 2-(1,1-dimethylethyl)-5-methyl-phenol, no measured concentration data in sewage sludges could be collected, limiting possibilities for a more in-depth step 2 assessment.

Many substances could not be assessed because no information on safe limit values for human consumption could be retrieved within the limitations and scope of this work (see section 3.3.1.1). Hence, $RCR_{human\ 10\ years}$ could not be calculated for compounds that are associated to high daily intakes by humans, including amongst others tetrabutyl ethylidenebisphenol (AO22E46), 3,5-Bis(1,1-dimethylethyl)-4-hydroxy-, 2,4-bis(1,1-

dimethylethyl)phenyl ester benzoic acid, traseolide, N-methyldodecylamine, tris(4-tert-butylphenyl) phosphate, 4,4'-Thiobis(6-tert-butyl-m-cresol), and many sterols (e.g. cholesterol, campesterol, stigmasterol).

Compounds that could be assessed and were associated to a $RCR_{human\ 10\ years}$ lower than 1 were not taken forward to the step 2 assessment. The results of compounds having an $RCR_{human\ 10\ years}$ in between 0.1 and 1 are presented in Figure 4. These compounds encompass a large number of pharmaceuticals and personal care products (PCPPs), including three different synthetic musks and pharmaceuticals such as miconazole and atorvastatin (Figure 4). It is reiterated that the data used in this assessment originates from data repositories, and has not been verified by JRC on a case-by-case basis, introducing additional uncertainties to the outcomes presented (see section 3.2.2.1 on methodology).

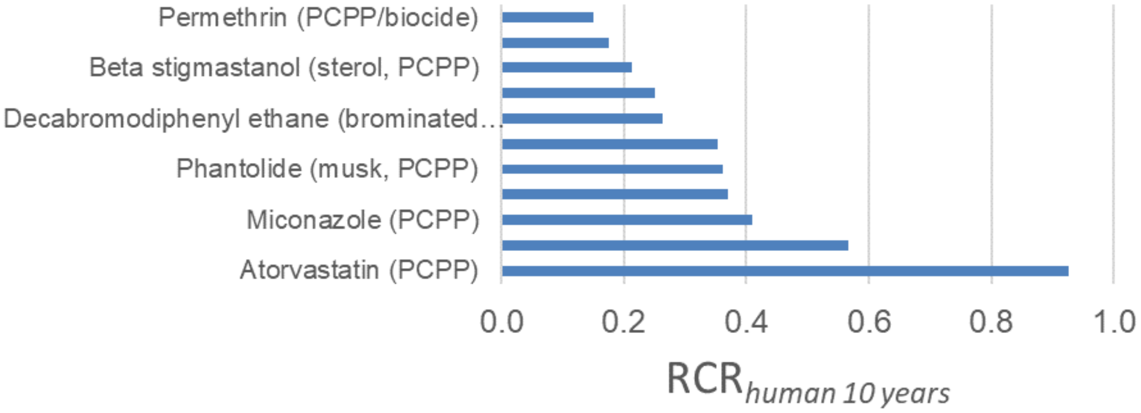

Figure 4: Contaminants present in sewage sludge that showed a $RCR_{human\ 10\ years}$ ratio in between 0.1 and 1 (model input values applied to derive the values presented as Supplementary Information (Table 9)).

In addition, results are presented for compounds selected on an ad-hoc basis that belong to contaminant classes that have been referred in certain scientific publications and by particular stakeholders as being of concern (Figure 5). These involve for instance quaternary ammonium compounds (benzalkonium chloride), pesticides (glyphosate), oestrogens (17β-estradiol), and common personal care products (triclocarban), and pharmaceuticals (e.g. ibuprofen, diclofenacy, acetaminophen) and antimicrobial substances (e.g. ofloxacin) found in sewage sludge (Figure 5). It is reiterated that even though the information of specific contaminants provides a preliminary indication for a wider class of substances, contaminants should be evaluated on a case-by-case basis as even within the same class of substances variations in physico-chemical and toxicological properties may exist.

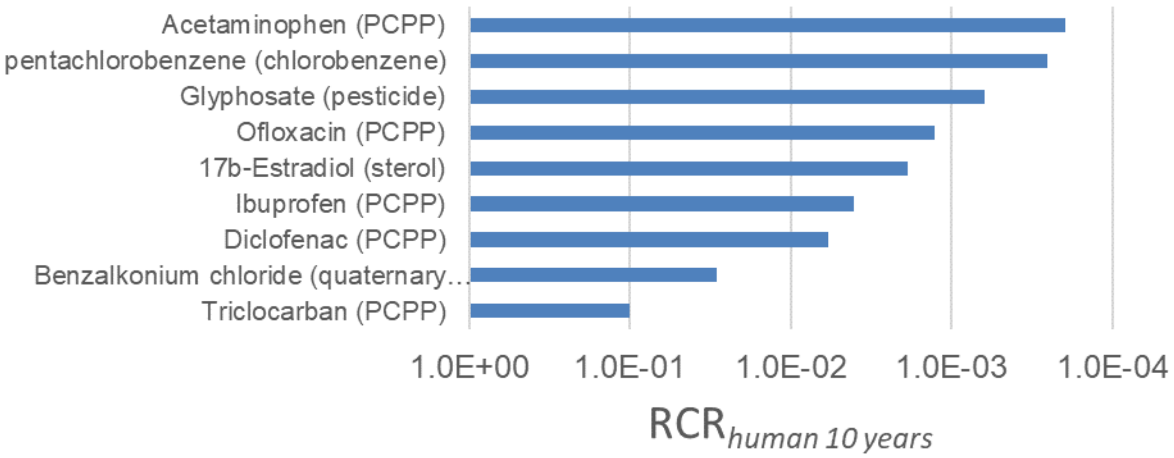

Figure 5: $RCR_{human\ 10\ years}$ values for ad-hoc selected contaminants, identified by some stakeholders as being of potential concern when present in sewage sludge (model input values applied to derive the values presented as Supplementary Information (Table 9) (PCPP: Pharmaceuticals and Compounds and Personal Care.

The main reasons for the low $RCR_{human\ 10\ years}$ for these contaminants is that most of them show either (i) a low concentrations in sewage sludge (usually below 1 mg kg^{-1} dry matter), (ii) a low potential to adsorb to the soil matrix and to bioaccumulate in the trophic chain, and (iii) high degradation rates in the (soil) environment.

3.3.2 Step 2 results

3.3.2.1 Identification and characterisation of pollutants of concern

The results of the Step 2 analysis has been performed for 10 different priority organic contaminant classes, encompassing 29 priority substances in total. Full results for all contaminants, as well as input data applied for the modelling, are presented in section 9.

The refined assessment indicated that relevant substances classified as PAH, PCDD/F+dl-PCB, long-chain PFAS, alkylphenols, polychlorinated alkanes (short- and mid-chain chlorinated paraffins; SCCPs/MCCPs), polychlorinated naphthalenes (PCNs), and phthalate acid esters were associated to an $RCR_{human\ 10\ years}$ above 1 (Figure 6). For the other contaminants that were taken forward, the refined analysis indicated that $RCR_{human\ 10\ years}$ for representative compounds of polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD), organophosphate compounds, organotin compounds, and polydimethylsiloxanes were smaller than 1 (Figure 6).

For many priority pollutants, data on representative[9] present-day concentrations in soils and waters could be retrieved ("background concentrations") that add further up to the RCR_{human}. This holds particularly true for for long-chain PFAS, PCDD/F + dl-PCBs, and PAH, where high background concentrations in soils have been observed (Figure 6).

The pollutants that cause human health risks when present in sewage sludge include both unintentionally produced substances (e.g. resulting from combustion and incineration plants) such as PAH, PCDD/F and PCNs as well as intentionally produced substances that end up in sewage sludge (long-chain PFAS, alkylphenols, phthalate acid esters, and SCCPS/MCCPs). With the exception of MCCPs, the production and use of these pollutants is banned and/or restricted (release reduction provisions) to a variable degree under the POPs[10] and

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[9] It is recognised that background concentrations in EU soils may vary widely depending on a manifold of factors, including the presence of industrial nearby sites. Hence, any outcomes based on these data should be interpreted with the necessary degree of caution. Moreover, for many compounds no representative environmental background data could be collected.

[10] REGULATION (EU) 2019/1021 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 20 June 2019 on persistent organic pollutants (https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019R1021&rid=3)

REACH[11] Regulation. For MCCPs, the procedure for a restriction is ongoing. Nonetheless, pollutants may be restricted to a variable degree for which continued releases to sewage sludge could take place. In addition, some pollutants have been phased out recently, and given their long life span, releases to sewage sludge may therefore continue to occur during the use or end-of-life phase of articles that contain these pollutants.

Common characteristics across the pollutants of most concern are their potential to bioaccumulate in lipids (correlated to their octanol-water partitioning coefficient Kow), their affinity for adsorption to the soil matrix (correlated to their soil-water partition coefficient Koc; with the exception of long-chain PFAS), their low biodegradation potential in soils, and their high toxicity for humans (see section 13.2). Compounds that share these characteristics and end up in sewage sludge are anyway potential hazards for human health.

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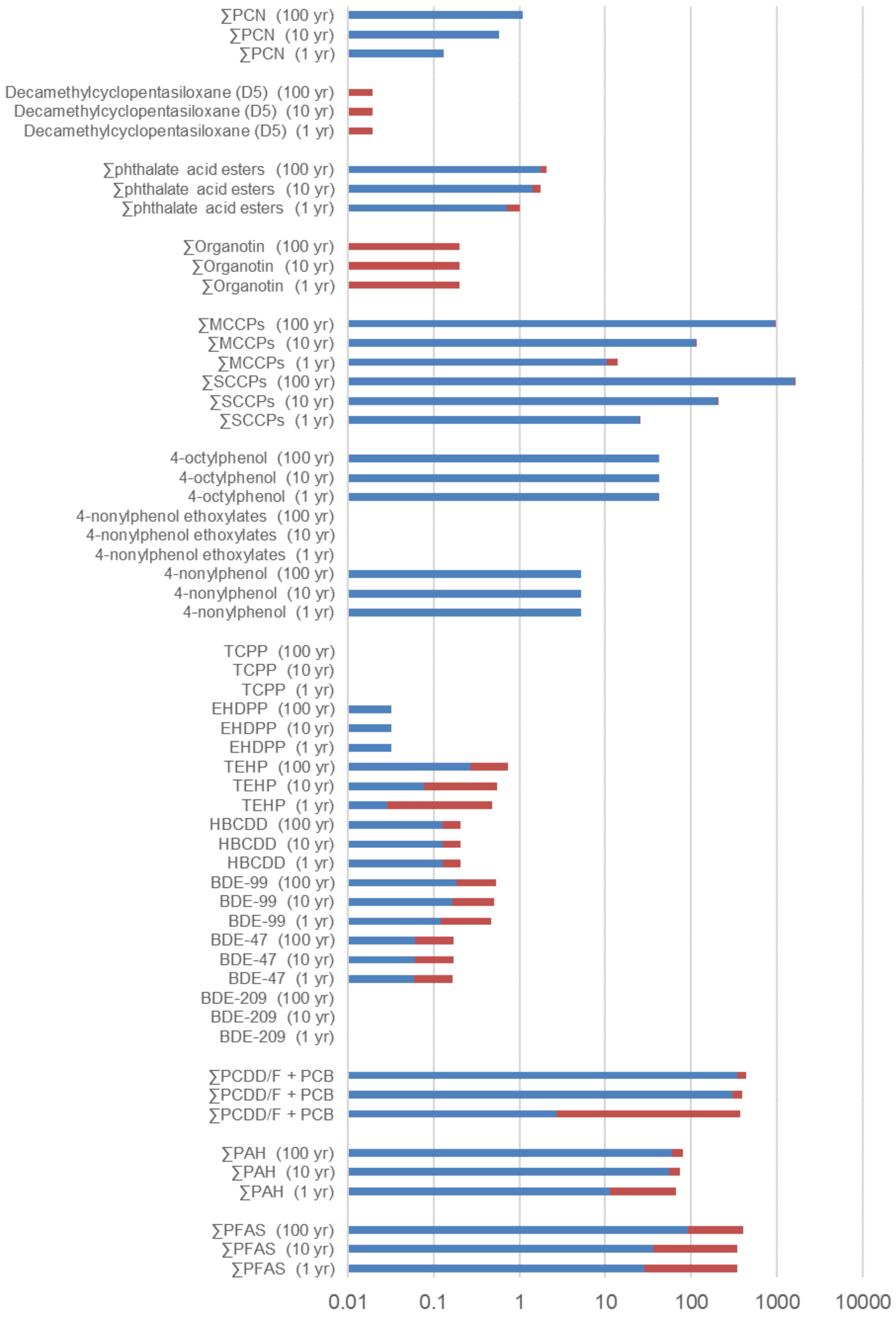
[11] REGULATION (EC) No 1907/2006 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL
of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) (https://eur-lex.europa.eu/legal-content/en/TXT/HTML/?uri=CELEX:02006R1907-20210215)



(see next page for Figure caption)

Figure 6: RCR_{human} values (logarithmic scale) for contaminants that were evaluated in step 2 of the assessment (blue bars indicate RCR_{human} estimated following the exclusive application of sewage sludge as a sole contamination source; the end of the red bar indicates the RCRhuman values when - in addition to sewage sludge applications - background concentrations of the respective contaminants from other sources (e.g. atmospheric deposition) have been taken into consideration; when certain contaminants are associated to a group safe limit value for human consumption, these compound are represented jointly; ∑PCN: sum of naphthalene congeners 66/67 (mixture of 1,2,3,4,6,7-HxCN and 1,2,3,5,6,7-HxCN) and 73 (1,2,3,4,5,6,7-HpCN).; ∑phthalate acid esters: bis(2-ethylhexyl)phthalate (DEHP), and di-isononylphthalate (DINP); ∑organotin: dibutyltins (DBT), tributyltins (TBT), and triphenyltins (TPT), ∑SCCPs: sum of short-chain chlorinated paraffins; ∑MCCPs: sum of mid-chain chlorinated paraffins; BDE-209: decabromodiphenyl ether; BDE-47: 2,2',4,4'-Tetrabromodiphenyl ether; BDE-99: 2,2',4,4',5-Pentabromodiphenyl ether; HBCDD: hexabromocyclododecane); TEHP: tris(2-ethylhexyl) phosphate, EHDPP: 2-ethylhexyl diphenyl phosphate; TCPP: tris(1-chloro-2-propyl) phosphate ∑PCDD/F + dl-PCBS: Polychlorinated dibenzo-p-dioxin and furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs), with values presented for 2,3,7,8-TCDD used as a reference compound); ∑PAH: sum of benzo[a]pyrene, chrysene, benz[a]anthracene and benzo[b]fluoranthene; ∑PFAS: sum of perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), Perfluorohexane sulfonic acid (PFHxS), and perfluorononanoic acid (PFNA); see section 4 and 13.2 for model input data values).

Risk assessment models that estimate the daily human intake of contaminants consider drinking water, fish, crops, meat and dairy as sources. Contaminant concentrations in the water bodies are the driver for fish and drinking water, whereas contaminant concentrations in soils determine the contaminant intake from crops, meat, and dairy. Therefore, assessing the relative contribution of drinking water, fish, and crops/meat/dairy sheds further light on the mechanisms that give rise to human health risks.

3.3.2.2 Contribution of food sources to human health risks

It was indicated that for most compounds (∑PAH, ∑PCDD/F + PCB, 4-octylphenol, ∑MCCPs, ∑phthalate acid esters, ∑PCN), the intake of food grown on agricultural land was the main source (Figure 7). This was, however, this is not the case for long-chain PFAS considered in this study. For these, fish intake was the main contributor (76%) to the observed risk (Figure 7). Hence, for the long-chain PFAS, the model calculations indicate that their concentrations in water bodies appear to be a principal driver to the observed human health risk. This is remarkable since this model only considers runoff, erosion, and drainage from sewage sludge amended soils as inputs to water bodies; other inputs (e.g. wastewater treatment plant effluents) are not considered in these results presented. PFAS ions show a high affinity for the water phase and most PFAS, including the short-chain PFAS, are partitioned to the effluent at wastewater treatment plants (Arvaniti et al., 2014; Brendel et al., 2018). It should be stressed that, while threshold concentrations for PFAS in aquatic ecosystems and drinking water could be identified, we could not retrieve any SLHC for short-chain PFAS (e.g. perfluoroheptanoic acid and perfluorobutanoic acid). Human health concerns related to short-chain PFAS in sludge are smaller than for long-chain PFAS because of their reduced adsorption potential to sludges, reduced bioaccumulation potential and ecotoxicity (Brendel et al., 2018). As a matter of fact, short-chain PFAS will be partitioned preferentially towards the effluent phase at wastewater treatment plants, and the presently observed low concentrations of short-chain PFAS in sludges would only result in limited short-chain PFAS contents in crops, meats, and milk according to our model calculations. Any short-chain PFAS in soil-grown food is likely resulting from the biodegradation of long-chain PFAS that were present in sludges (Brendel et al., 2018).

Therefore, it is likely that PFAS losses to the environment via effluents are the main contributor to the total human health risks. PFAS in sludge may further enhance risks, but a detailed source contribution analysis that considers also wastewater treatment effluents is critical to develop effective mitigation strategies that tackle the root of the contamination issue.

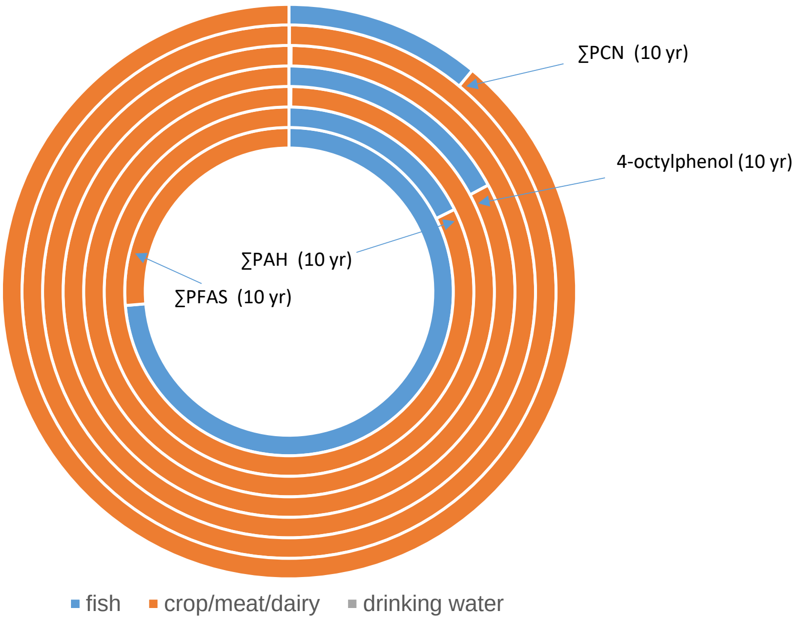


Figure 7: Estimation of the relative contribution of fish (blue), crop/meat/dairy (orange) and drinking water (grey) to the total human intake of selected contaminants resulting from the exclusive assessment of contaminants present in sewage sludge ("no background modelling scenario") (from inner to outer ring: ∑PFAS, ∑PAH, ∑PCDD/F + PCB, 4-octylphenol, ∑MCCPs, ∑phthalate acid esters, ∑PCN; for full abbreviations: see Figure 6).

At present, different EU initiatives are focusing on limiting PFAS pollution to the environment and water bodies. A phased restriction on a broader subset of around 200 linear and branched perfluorocarboxylic acids (PFCAs) substances, a PFAS sub-group, containing 9-14 carbon atoms, their salts and related substances, has been adopted in August 2021 (Regulation (EU) 2021/1297 amending Annex XVII to Regulation (EC) No 1907/2006). The latter Regulation limits C9-C14 PFCAs in substances and mixtures to concentrations below 0.025 mg kg-1. Restrictions will begin to take affect as of February 2023 under Annex XVII of REACH. Regulation (EU) 2019/102 on persistent organic contaminants includes prohibitions for the manufacturing, placing on the market and use of intentionally produced substances. For PFAS, the prohibitions also apply to substances, mixtures and articles that have PFAS as an unintentional trace contaminant. In addition, the 2019 Fitness Check of EU Water Law, which covered the Water Framework Directive together with the Directive on Environmental Quality Standards, the Ground Water Directive and the Floods Directive, confirmed the need to review the lists in the light of scientific developments. The fitness check concluded that, in relation to chemical pollution, the legislation focuses on some less relevant older contaminants while not sufficiently addressing a number of contaminants of emerging concern, including PFAS. The recast of the Drinking Water Directive that entered into force on January 2021 includes a limit of 0.5 µg/l for all PFAS. This is in line with a grouping approach for all PFAS. Finally, the new European Chemicals Strategy has announced the EU's ambitious plans to ban all non-essential uses of PFAS.

3.3.3 Impact of sludge treatments

A detailed overview of the impact of biological (anaerobic digestion and composting) and thermal oxidation (incineration) is given for all relevant pollutants in section 13.3. Other sludge processing techniques such as lime stabilisation, drying and dewatering are not projected to have a main impact on chemical pollutants present in sludge, and were therefore not evaluated. Finally, thermal processing techniques other than incineration (e.g. pyrolysis, gasification, hydrothermal carbonisation) have not been considered due to a lack of a well-developed knowledge and information database related to the removal of relevant pollutants.

In general terms, it is indicated that biological treatment is unable to cause significant removals for some pollutants that cause most risk for human health (e.g. PCDD/F, long-chain PFAS). For others priority pollutants (e.g. PAH, alkylphenols), biological treatment has some potential to decrease pollutant loads, but removal efficiencies are non-consistent in literature and still insufficient to decrease concentrations to below values of concern. Incineration, on the other hand, is an effective treatment options that will remove the overall share of the organic pollutants present in the sewage sludge. However, in order to completely destroy the thermoresistant PFAS substances, good operational conditions and minimum temperatures in the range 1000 - 1100°C are required (Winchell et al., 2021). In addition, incineration has to be executed in line with modern industrial standards in the EU to avoid ashes becoming contaminated with e.g. PAH and PCDD/F generated during the incineration process.

3.3.4 Prioritisation of pollutants

It is projected that PAH, PCDD/F+dl-PCB, long-chain PFAS, alkylphenols, polychlorinated alkanes (SCCPs/MCCPs), polychlorinated naphthalenes (PCNs), and phthalate acid esters are present in sewage sludges at levels that may induce human health risks, and therefore these compounds may cause most risks to human health. Even if a less conservative approach were to be applied that assumes lower pollutant loads and a more regional distribution of the available sewage sludge produced in the EU (see notes in section 3.2.1), PAH, PCDD/F+dl-PCB, long-chain PFAS, and SCCPs/MCCPs will still have risk characterisation ratios above 1. This suggest that the presence of these pollutants in sewage sludge currently causes significant human health risks and should receive absolute priority in view of risk mitigation. Particularly relevant compounds are MCCPs since the use of these intentionally produced substances in the EU is not (yet) restricted. At the same time, it is remarked that sewage sludge is not the sole source of soil contamination and that a more in-depth source contribution analysis is required to develop effective risk mitigation strategies that tackle the core drivers of pollution.

Other persistent pollutants, especially these for which RCR_{human} ratios between 0.1 and 1.0 such as organophosphate and organotin compounds, seem to be of a lower concern, but additional peer-review and verification by other stakeholders/experts in the field of risk assessment may further reinforce these conclusions.

The conclusions of this work align to recent results from Deleebeeck et al. (2021) that assessed risk three different organic compounds present in fertilising materials. It was indicated that PFAS and PCDD/Fs are priority pollutants that may induce human health risks, even when fertilising materials are considered the sole source of environmental contamination. Similar to our results, diclofenac - the model pharmaceutical compound used in Deleebeeck et al. (2021) - was associated to a low risk characterisation ratio in that study.

3.4 Assessment for soil organisms

3.4.1 Results

$RCR_{soil\ 10\ years}$ was calculated for 715 contaminants that cover susbtances across a very broad spectrum of chemicals. Nonetheless, for about 80 additional substances for which sewage sludge concentrations could be retrieved, no $RCR_{soil\ 10\ years}$ could be calculated. In addition, the assessment is incomplete because data on PNEC for soil organisms is only limitedly available and mostly estimated based on extrapolation of results for aquatic organisms. Hence, whereas this assessment provides insights on estimated risks to soil organisms, the results should be interpreted with the necessary degree of caution and it is possible that certain substances that may be of concern to soil organisms have not been identified in this assessment.

$RCR_{soil\ 10\ years}$ were above 1 for about 30 contaminants (Figure 8; Figure 9). It is observed that many of the contaminants that had been identified as being a concern for human health (taken forward for Step 2 analysis), showed $RCR_{soil\ 10\ years} > 1$. Specifically, these involve PAH (e.g. coronene, dibenzo[a,h]perylene), long-chain PFAS (e.g. PFOA, PFOS), PCDD/F (e.g. 2,3,7,8-TCDD), alkylphenols (4-Nonylphenol monoethoxylate), short- and mid-chain paraffins, as well as organochlorine and organophosphate pesticides (dieldrin, heptachlor, p-DDT, ethyl azinphos). Organochlorine and organophosphate pesticides have been phased out since long the EU and the available dataset on sludge concentrations lacks data of recent measurements. The data indicate that RCR_{soil} ratios for these compounds increase as a function of time following continuous applications of sewage sludge on agricultural land (Figure 8). The risks after a few years of sewage sludge application are minimal, but gradually increase over time to significant levels in the mid- and long-term. This observations points towards the urgency to minimise the inputs of these persistent contaminants in the soil so as to ensure soil quality and continued ecosystem functioning in the long-term.

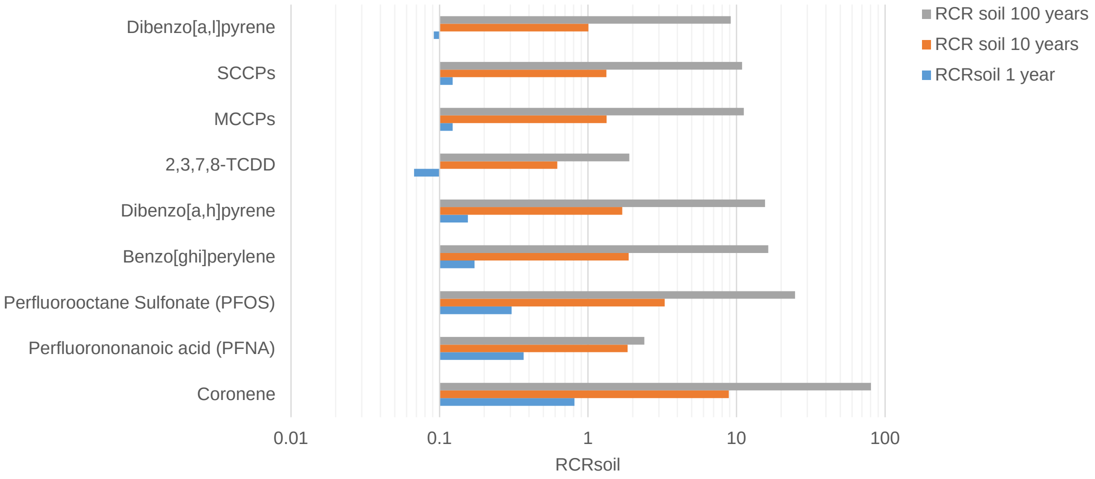


Figure 8: RCR_{soil} estimated after 1, 10, and 100 years of continuous sewage sludge applications for contaminants belonging to classes that had been identified as being of concern to human health (SCCP: short-chain chlorinated paraffins, MCCP: mid-chain chlorinated paraffins; model input values applied to derive the values presented as Supplementary Information (Table 11)).

In addition, the compounds represented in Figure 9 were identified as having a $RCR_{soil\ 10\ years}$ above 1. Most compounds included in the list can be classified as pharmaceuticals and personal care products. In addition, plant sterols, a quaternary ammonium compound and its transformation product, a surfactant and a pesticide degradation period were identified (Figure 9). $RCR_{soil\ 10\ years}$ varied from approximately 1 to about 100 for most compounds, with the exception for dipyridamol with of 339.

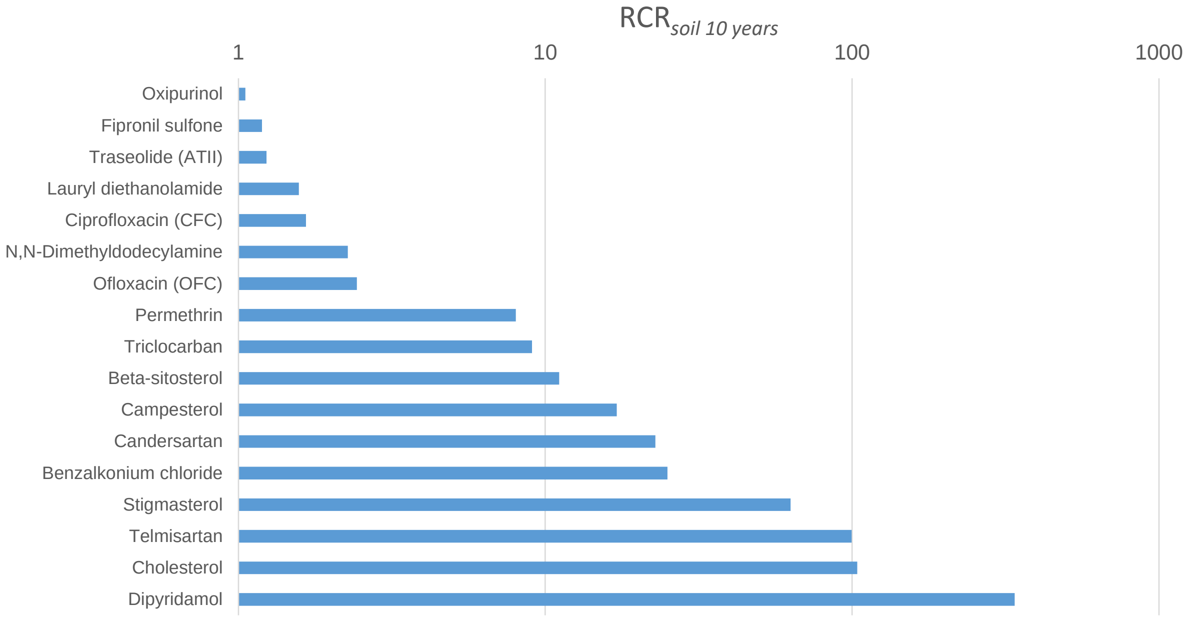


Figure 9: Pollutants present in sewage sludge that showed a $RCR_{soil\ 10\ years}$ ratio above 1 (model input values applied to derive the values presented as Supplementary Information (Table 11)).

The presence of pharmaceuticals in sewage sludge may give rise to their presence in soils shortly after sludge land application. Potential risks for soil organisms have been identified based on the concentration averaged over a 30-day period after the (intermittent) application of sewage sludge. At the same time, it is important to stress that most pharmaceuticals are (readily) biodegradable[12], and therefore no accumulation of these pollutants is expected and observed in agricultural soils (i.e. $RCR_{soil\ 10\ years} = RCR_{soil\ 1\ year}$). This is also the case for dipyridamol (readily biodegradable), telmisartan (readily biodegradable, failing 10-day window), and candersartan (readily biodegradable), the pharmaceuticals characterised by the highest $RCR_{soil\ 10\ years}$ within this class. Biological sludge treatment options, including both anaerobic digestion and composting, are able to remove a share of these pollutants from the sludge (Carballa et al., 2007; Haiba et al., 2016; Thomas et al., 2020). Nonetheless, these compounds might still be present in detectable and removal rates are compound and process-specific, with e.g. high removal rates (>90%) observed for triclosan/triclocarbans but minimal removal for some other substances (Carballa et al., 2007; Haiba et al., 2016; Thomas et al., 2020). Thermal treatment (incineration) will effective remove pharmaceuticals, and no residues are expected to be present in sewage sludge ashes that may be used a source material for P-fertiliser production.

Plant sterols are compounds of eukaryotic cells present in a range of plant foods such as beans, lentils, cereals, vegetable oils, seeds and nuts. Sterols are also food additives added to many foodstuffs to lower cholesterol levels in humans. These are labile compounds that are degraded in soils, presumably due to the action of soil arthropods (Puglisi et al., 2003). No reports have been found that highlight the potential toxicity of plant sterols for soil microorganisms. Therefore, the low $PNEC_{soil}$ values derived fro $PNEC_{water}$ using the equilibrium partition method are likely underestimated. Hence, these compounds are not considered a main concern when present in sewage sludge.

The widespread global use of polycyclic musk, their high log K_{ow} values and persistence has resulted in widespread occurrence in sewage sludge. It is surprising that the assessment identified traseolide as the musk of main concern, whereas no risks for the soil compartment was identified for other synthetic musks such as galaxolide and tonalide. The $PNEC_{aqua}$ and $PNEC_{soil}$ value for traseolide (6.9 E-04 µg L^{-1}) is much lower than for other musks. Musks typically have a half-life in soils around 100-150 days, and show limited potential for accumulation in soils under yearly sludge application scenarios (Balk and Ford, 1999). Therefore, a provisional assessment of the risk of 2 polycyclic musks showed that long term risk to the soil environment as a result of normal sludge application is unlikely (Danish Ministry of the Environment, 2012). In this assessment, the $RCR_{soil\ 10\ years}$ for traseolide is just above 1.

Bactericidal cationic surfactants such as quaternary ammonium compounds (QACs) are widely detected in the environment. Although individual QACs are amenable to biodegradation, it is possible that persistence is increased for mixtures of QACs with varying structure (Khan et al., 2015). Moreover, toxic degradation for soil microorganisms, have been identified (Khan et al., 2015). Hence, QACs, and particularly benzalkonium chloride, may potentially be a risk for soil organisms and soil quality, for which reason a more in-depth assessment of these compounds may be required. For the second surfactant, lauryl diethanolamide, no relevant information related to risks for soil organisms could be retrieved but its low biodegradation potential points towards a potential concern for this substance.

Fipronil is a common insecticide, often used to de-flea household pets such as dogs and cats. Fipronil is not authorised for use in animals for the food chain as it has potential toxicity to humans. The $RCR_{soil\ 10\ years}$ for this substance is only just above 1. Moreover, fipronil sulfone is classified as a readily biodegradable compound, and seems therefore of a reduced concern in view of soil quality.

The detailed results of the step 2 analysis (e.g. RCR_{soil} after 1 and 100 years of application) for compounds relevant for human health (see section 3.3.1) are given in section 13.3. No step 2 analysis were performed for pollutants that are characterised by an $RCR_{soil\ 10\ years}$ above 1, but a $RCR_{human\ 10\ years}$ below 1.

3.4.2 Prioritisation of pollutants

In our view, the pollutants that classify as toxic and persistent in the soil environment could be considered as priority pollutants. The assessment brought forward that particular pollutants identified as being a concern to human health (PAH, long-chain PFAS, PCDD/F, alkylphenols, and short- and mid-chain paraffins) are also of concern for soil quality and biodiversity protection. In our view, these pollutant classes are most relevant in

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[12] Readily biodegradable is defined as the ability of a product to biodegrade quickly and completely within 28 days suing a standard test procedure (≥ 60% by OECD 301A-F/ASTM D7373 testing).

view of soil protection due to the high residence times in soils. Consecutive sewage sludge applications are modelled to result in a further build-up of these pollutants in soils, up to levels that impair soil biodiversity and ecological and socio-economic ecosystem functions of agricultural land.

In addition, a reduced set of pollutants (benzalkonium chloride and its degradation products, and lauryl diethanolamide, traseolide) has been identified in this ecological screening assessment that could be prioritised for a more in-depth ecological assessment.

3.5 Availability of measurement standards

Measurement standards for the priority list of pollutants presented in previous sections have been identified in the European Standard (EN) and International Organization for Standardization (ISO) databases (Table 3).

For most of the priority pollutants, at least one measurement standard (either EN or ISO) was identified as relevant (Table 3). This was the case for polycyclic aromatic hydrocarbons; dioxins, furans and dioxin-like polychlorinated biphenyls; alkylphenols; short- and medium-chain polychlorinated alkanes (no EN standard identified but one ISO standard relevant), and phthalate acid esters.

Nevertheless, for per- and polyfluoroalkyl substances, polychlorinated naphthalenes; benzalkonium chloride and its degradation products (as well as other quaternary ammonium compounds); lauryl diethanolamide (and N-acyl amines or fatty amides), and traseolide (synthetic musks), no relevant measurement standards could be identified, neither in the EN nor in the ISO database (Table 3).

Table 3: Identified measurement CEN and ISO standards relevant for the priority list of pollutants in sludge or soil.

Priority pollutants	Relevant EN standards	EN standard title	Relevant ISO standards	ISO standard title
Polycyclic aromatic hydrocarbons (PAHs)	EN 16181:2018	Soil, treated bio-waste and sludge - Determination of polycyclic aromatic hydrocarbons (PAH) by gas chromatography (GC) and high performance liquid chromatography (HPLC)	ISO 13859:2014	Soil quality — Determination of polycyclic aromatic hydrocarbons (PAH) by gas chromatography (GC) and high performance liquid chromatography (HPLC)
	EN 17322:2020	Environmental Solid Matrices - Determination of polychlorinated biphenyls (PCB) by gas chromatography - mass selective detection (GC-MS) or electron-capture detection (GC-ECD)	ISO 18287:2006	Soil quality — Determination of polycyclic aromatic hydrocarbons (PAH) — Gas chromatographic method with mass spectrometric detection (GC-MS)
	prEN 17503	Soil, sludge, treated bio-waste and waste - Determination of polycyclic aromatic hydrocarbons (PAH) by gas chromatography (GC) and high performance liquid chromatography (HPLC) - Determination of polycyclic aromatic hydrocarbons (PAH) by gas chromatography (GC) and high performance liquid chromatography (HPLC)		
Dioxins, furans and dioxin-like polychlorinated biphenyls (PCDD/Fs,	EN 16190:2018	Sludge, treated bio-waste and soil - Determination of dioxins and furans and dioxin-like polychlorinated biphenyls by	ISO 13876:2013	Soil quality — Determination of polychlorinated biphenyls (PCB) by gas

PCBs)		gas chromatography with high resolution mass selective detection (HR GC-MS)		chromatography with mass selective detection (GC-MS) and gas chromatography with electron-capture detection (GC-ECD)
			ISO 13914:2013	Soil quality — Determination of dioxins and furans and dioxin-like polychlorinated biphenyls by gas chromatography with high-resolution mass selective detection (GC/HRMS)
			ISO/DIS 13914	Soil, treated bio-waste and sludge — Determination of dioxins and furans and dioxin-like polychlorinated biphenyls by gas chromatography with high resolution mass selective detection (HR GC-MS)
Per- and Polyfluoroalkyl substances (PFAS)	-	-	-	-
Alkylphenols (APs/NPs/OPs)	CEN/TS 16182:2012	Sludge treated bio-waste and soil - Determination of nonylphenols (NP) and nonylphenol-mono- and diethoxylates using gas chromatography with mass selective detection (GC-MS)	ISO/TS 13907:2012	Soil quality — Determination of nonylphenols (NP) and nonylphenol-mono- and diethoxylates — Method by gas chromatography with mass selective detection (GC-MS)
Short- and Medium-chain polychlorinated alkanes (S/MCCPs)	-	-	ISO 18635:2016	Water quality — Determination of short-chain polychlorinated alkanes (SCCPs) in sediment, sewage sludge and suspended (particulate) matter — Method using gas chromatography-mass spectrometry (GC-MS) and electron capture negative ionization (ECNI)
Polychlorinated naphthalenes (PCNs)	-	-	-	-
Phthalate acid esters (DEHP/DINP)	CEN/TS 16183:2012	Sludge, treated bio-waste and soil - Determination of selected phthalates using capillary gas chromatography with mass spectrometric detection (GC-MS)	ISO 13913:2014	Soil quality — Determination of selected phthalates using capillary gas chromatography with mass spectrometric detection (GC/MS)
Benzalkonium chloride and its degradation products (Quaternary	-	-	-	-

ammonium compounds)				
Lauryl diethanolamide (N-acyl amines & fatty amides)	-	-	-	-
Traseolide (Synthetic musks)	-	-	-	-

3.6 Impacts of wastewater treatment processes

The assessment in the previous sections is based on observed contaminants concentrations as measured in sewage sludges, mostly originating from wastewater treatment plants across the EU. At present, the implementation of advanced (quaternary) wastewater treatment is limited, for which reason the current database on sludge quality does not capture the effect of such innovative wastewater treatment configurations.

Advanced wastewater treatment typically entails adsorption and oxidation processes, most often through activated carbon and ozonation respectively. Activated carbon can be dosed in powdered form (PAC) directly in the bioreactors of biological treatment plants, or in a separate reactor downstream. Alternatively, it can be provided in the form of a granular bed (GAC) through which the effluents of biological treatment are passed.

In the case of direct dosing in the bioreactor, PAC remains enmeshed in the surplus sludge and, because of its characteristics and potentially high concentration of microcontaminants, it makes the sludge unfit for reuse in agriculture due to the high capacity of the PAC to adsorb and strongly bind a wide spectrum of contaminants. The chemical composition of PAC-enriched sludge and its dynamics in soils with respect to contaminant release are at present unknown. At the same time, the enmeshed PAC enhances the energy content of sludge when it comes to incineration. The dosing of PAC in the bioreactor can be a cheap option for advanced treatment in many cases. The use of PAC is the sole process where there is a transfer of contaminants from the effluents to the sludge, and is therefore applicable only when direct sludge reuse is not contemplated. Its most logical implementation is in cases with sludge incineration, and it could improve the energy balance of incineration.

When PAC is used in a separate reactor, or in the case of GAC, the surplus sludge of biological treatment processes is unaltered. Exhaust PAC or GAC in these cases form a separate waste stream.

Ozonation works on the effluents and does not alter the quality of the sludge. Usually, ozonation does not lead to complete mineralization of micropollutants and yields reaction products that are less stable than the parent molecules, but potentially reactive and harmful. It is therefore recommended to couple ozonation with a further adsorption stage (a sand filter, GAC or PAC). In any case, the waste from these processes is not expected to mix with the surplus sludge of biological treatment, and is therefore irrelevant in terms of sludge quality.

Hence, it is concluded that the possible implementation of more advanced (quaternary) wastewater treatment under the UWWTD with a view to improve effluent quality is not expected to alter the quality of (primary and secondary) sludge brought to agricultural land. However, it may result in an increased proportion of the sludge being incinerated to the detriment of sludge landspreading. Therefore, it is important to minimise contaminant inputs upstream to wastewater treatment plants (e.g. through product design, limiting inflow of certain industrial wastewaters) when sludge is targeted for land application.

3.7 Risk screening assessment conclusions and recommendations

Based on the available data and techno-scientific understanding, this work identified a set of pollutants that are present in sludge at levels that could induce environmental and human health risks. Risk characterisation ratios over 100 were observed, indicating that under a ‘reasonable worst-case’ scenario – significant risks for human health and the environment may be present. The pollutants that were considered to be of most concern for human health are PAH, PCDD/F+dl-PCB, long-chain PFAS, SCCPs/MCCPs, and to a lesser extent alkylphenols, PCNs, and phthalate acid esters. These pollutants are highly toxic to humans, show a high bioaccumulation potential and are persistent in the soil for long periods due to their lack of mobility in the soil matrix and low biodegradation potential. In addition, pollutants such as PFAS may end up in water bodies, where it can further contribute to risks to water quality and fish consumers. The pollutants associated to the greatest risks are already subject to actions and restrictions under the POPs and REACH Regulation.

Most of the pollutant groups identified as being of concern for human health are also associated to risks for soil organisms. Humans are, however, the most sensitive endpoints for these substances for which risks to humans may even occur at lower pollutant concentration levels in sludges. Therefore, risk mitigation strategies that aim to protect human health will also decrease risks to soil organisms and soil quality. However, in addition to the more persistent pollutants identified above, benzalkonium chloride and its degradation products, and lauryl diethanolamide, traseolide were also identified as possibly posing a risk to soil organisms.

Risk mitigation options may involve limiting the concentration of pollutants of concern in sewage sludge. Biological sludge treatment options such as composting and anaerobic digestion, may not be effective to remove the persistent pollutants of concern to below levels of concern. On the other hand, incineration in modern plants has the potential to effectively lower pollutant levels to low or negligible concentrations, after which the ashes could be used as an intermediate in P-fertiliser manufacturing processes. An alternative risk mitigation option is to reduce the volumes of sewage sludge spread on land relative to the modelling scenario of 5 tonnes dry matter ha^{-1} yr^{-1} applied in this assessment. For instance, an application scenario with a maximum load of 2 tonnes of dry matter per ha every 3 years would already reduce pollutant loads by approximately a factor 10. Finally, hybrid options could be envisaged to ensure full environmental and health protection from the effects of organic pollutants.

4 Other contaminants present in sludge

Apart from chemical substances, sewage sludge contains other compounds of concern to humans and the environment. This section focusses on antimicrobial resistance, biological pathogens, and microplastics.

4.1 Antimicrobial resistance

Antimicrobial Resistance (AMR) occurs when bacteria, viruses, fungi and parasites change over time and no longer respond to medicines making infections harder to treat and increasing the risk of disease spread, severe illness and death. As a result of drug resistance, antibiotics and other antimicrobial medicines become ineffective and infections become increasingly difficult or impossible to treat.

Although wastewater goes through numerous treatment and cleaning processes, some antibiotic residues cannot be removed by these methods (Chen et al., 2016). While treatments can remove or reduce the load of AMR bacteria, AMR genes (ARGs) have been detected at all stages of the municipal wastewater treatment process, including in sludge. Both Fouz et al. (2020) and Nguyen et al. (2021) have analysed extensive evidence base to highlight the contribution of wastewater to the emergence, persistence and transmission of AMR under different settings. However they also note inconsistencies in reporting and monitoring data, compounded by a lack of standardised protocol for determining AMR gene removal via wastewater treatments, and the inability to support appropriate risk assessment.

Numerous AMR genes including types and subtypes of almost all common antibiotics have been detected in wastewater influent, effluent and biosolids or sludge. During the treatment process, a large proportion of AMR bacteria and ARGs are removed from the water phase and partitioned into the sludge phase, resulting in high concentrations of ARGs in sludges and biosolids. It is noteworthy that not all ARGs pose the same risk level to human health. The likelihood of AMRG introduction into human pathogens should be assessed based on their hosts (ARB), rather than the ARGs themselves (Vaz-Moreira et al., 2014)

Over the last years, the application of molecular biology techniques has considerably improved the capability to identify AMR bacteria and quantify AMR genes both in wastewater and soil. This offers the possibility to monitor the risk of AMR outbreaks. As part of 2018 LUCAS Soil survey, the JRC selected 1,000 locations to investigate taxonomical and functional diversity in soil by means of metagenomics and metabarcoding (Orgiazzi et al., 2015). At the end of sampling campaign, 885 (out of 1,000) fresh soil samples were collected and analysed for biodiversity (metabarcoding). Bacteria were among the targeted organisms for diversity assessment. The generated dataset may allow the identification of potential ARB. Indeed, most potential ARB belong to *Proteobacteria*, *Firmicutes*, and *Bacteroidetes* phyla, which are dominant bacterial phyla in wastewater and sludge (Su et al., 2017). Their presence and relative abundance in EU agricultural soils can be easily assessed through LUCAS Soil Biodiversity dataset.

Moreover, out of the 885 samples, 630 were also analysed for functional (metagenomics) aspects, including an investigation of the distribution of AMR genes. These 630 samples are mainly from cropland areas, where amenDMents are applied and, thus, the likelihood of an outbreak of antimicrobial resistance events is higher. The JRC is currently carrying out the identification and distribution of AMR genes across EU agricultural soils. The dataset potential is high as, in principle, it allows to identify any AMR genes, with the possibility to focus on genes with higher levels of risk to human health conferring resistance to the most widespread antibiotics (e.g. animal and human health).

Furthermore, each LUCAS point is also well characterized by a large set of ancillary data (e.g. land cover, land use, soil physico-chemical properties, heavy metals, climate and topography). This allows the investigation of how each of these parameters (or a combination thereof) can affect the distribution of AMRG. Furthermore, it might be possible to identify genes that can be considered as indicators for specific agricultural land cover types (e.g. orchards), or specific soil properties (e.g. low/high pH, low/high carbon content). Once developed, these indicators may be considered for inclusion into the progress assessment and review process of EU environmental (e.g. Sewage Sludge Directive and Biodiversity Strategy for 2030), and agricultural (e.g. common agricultural) policies.

4.2 Biological pathogens

Pathogens are any organisms that can produce a disease. Biological pathogens include microorganisms such as bacteria, viruses, protozoa, prions and fungi, and their associated toxins. Pathogens can also be carried by larger organisms. They have the ability to adversely affect human and environmental health in a variety of ways, ranging from relatively mild, allergic reactions and serious medical conditions to death.

Raw sewage may contain a wide variety of pathogenic microorganisms reflecting the health of the community and industrial processes (e.g. hospitals, meat-processing facilities, abattoirs, tanneries). Pathogens of concern (simply an indicative subset) include Astrovirus, Hepatitis A, *Salmonella*, *Cryptosporidium* and *Taenia saginata* (tapeworm). Advances in analytical techniques and changes in consumer habits means that new pathogens are recognized and the impact of others reduce. Microorganisms can mutate to reflect changes in their environment. In addition, many pathogens are viable but non-culturable, which means that actual concentrations in sludge are probably underestimated. Thus, no assessment of the risks associated with the land application of sewage sludge can ever be considered to be complete when dealing with microorganisms. As new agents are discovered and a greater understanding of their ecology is developed. Treatment of sewage sludge by anaerobic or aerobic digestion or dewatering will reduce the number of pathogens, but significant numbers can remain or regrow after storage.

While there are several studies addressing pathogens in sewage sludge (Dumontet et al., 2001), they are often based on a limited number of samples. In addition, many studies are based on laboratory bioreactors with controlled operating conditions (temperature, aeration, and effluent flow), which may not be typical of actual environments. Measures should be applied in order to avoid contamination of crop plants by human pathogens (Major et al., 2020). The emergence of molecular techniques should allow a better characterization of microbial structure and functions directly in the environment (i.e., soil and plant). As for antimicrobial resistance genes and bacteria, LUCAS Soil Biodiversity survey offers several possibilities for detecting the presence and distribution of human pathogens across agricultural soils in the EU. Nonetheless, their eventual source (e.g. sewage sludge application) would require additional investigations.

Knowledge on the effects of sewage sludge on plant pathogens is still poor. Some studies seem to show a no-effect of sludge application on soil suppressiveness (i.e., soil capability to naturally suppress a plant pathogen due to its microbial makeup) to plant pathogens (Ghini et al., 2007). Furthermore, there are evidences that the application of sludge may contribute to the suppression of some plant pathogens (Cotxarrera et al., 2002).

An interesting recent development has been the sampling of untreated wastewater to assess the scale of the SARS-CoV-2 virus in a population (the virus that causes COVID-19). SARS-CoV-2 is shed in feces of infected individuals and can be measured in wastewater. Increased cases of COVID in the community is associated with increased levels of SARS-CoV-2 in wastewater, thus complementing our understanding of the transmission of COVID-19 in the community. The process looks for the non-infectious RNA, not the viable virus. There are no known cases of transmission resulting from exposure to wastewater. The JRC recently published a report[13] that assessed the feasibility of a European Sewage Sentinel System for SARS-CoV-2 based on the experience of several Member States and a laboratory exercise. There is evidence of the presence of SARS-CoV-2 virus in primary sewage sludge, which implies a risk to wastewater treatment workers or those working on sewage systems containing untreated effluent. Conventional wastewater treatment provides only partial removal of SARS-CoVs, thus safe disposal or reuse of sludge will depend on the efficacy of final disinfection (Bogler et al., 2020).

There is also growing concern on plant related diseases. Sewage sludge is also a source for the dissemination of alien plant species if seeds are excreted by humans and retained in the process.

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[13] https://publications.jrc.ec.europa.eu/repository/handle/JRC125065

4.3 Microplastics contamination

While plastic pollution has been extensively documented for aquatic ecosystems, less is known about the state and impacts of plastic in the terrestrial environment, especially in soil. Plastic debris on land is generally the result of three main pathways. These include:

- agricultural practices (e.g. poly-tunnels, mulching substances, baled silage), where large pieces of debris gradually degrade into smaller pieces through wear, weathering and other disintegration processes;
- through waste streams (e.g. landfills, bio-waste, sewage, litter);
- airborne deposition as a result of industrial processes.

The term microplastics are loosely defined as particles of any type of "polymers" with a diameter of less than 5 mm, although they are predominantly composed of polyethylene and polyethylene terephthalate. Microplastics are considered to be primary when relating to any fragment that are already 5 mm or less before entering the environment (e.g. microfibers from clothing, microbeads, and plastic pellets). Secondary microplastics arise from the breakdown of larger pieces that are already in the environment through natural weathering processes (often referred to as macroplastics). For the purposes of this report, the emphasis is on primary microplastics. The new Regulation (EU) 2019/1009 on fertilisers says that macroscopic impurities are above 2 mm, from 16 July 2026, the presence of plastics above 2 mm within shall be no more than 2,5 g/kg dry matter. By 16 July 2029 the limit-value of 2,5 g/kg dry matter for plastics above 2 mm shall be re-assessed in order to take into account the progress made with regards to separate collection of bio-waste.

Recent attention has shown that microplastics are present in sewage sludge. Wastewater Treatment Plants are receptors for microplastics derived from a range of sources (e.g. industry, domestic wastewater, surface drainage) that inevitably are mixed together. Through the settlement processes of wastewater treatment, the majority of microplastic particles becoming entrained in sewage sludge. Differences in water treatment technologies allow for small particles to pass through treatments within solid materials or sludge, which when applied to land, provides a significant pathway to soil.

An interesting aspect to note are studies that show that microplastics in biological wastewater treatment systems reduce the effectiveness of the treatment as they reduce the abundance of: nitrifying bacteria; denitrifying bacteria with nitrogen and phosphorus removal function; heterotrophic nitrification-aerobic denitrifying bacteria; and bacteria capable of degrading phenolic compounds (McCormick et al., 2014).

Nizzeto et al. (2016) estimated that between 125 and 850 tons of microplastics/million inhabitants could be ending up annually on agricultural soils that will receive urban sewage sludge. For example, the study assumes that between 10-90% of microplastics produced from road wear and debris from building coating are collected by WWTP sewers, which means that 360 and 1980 tons microplastics could reach municipal wastewater treatment plants every year.

An unpublished report by the JRC of a recent pilot study to test the feasibility of analysing plastic particles in samples collected during the 2018 LUCAS Survey found on average 58.3 microplastic particles in 1 g of cultivated soil from 50 samples collected across 7 Member States (standard deviation: 146). –Work is ongoing to identify whether the locations sampled were subjected to possible applications of sewage sludge. Rolsky et al. (2020) undertook a literature search on microplastics in sewage sludge, including six EU Member States (Italy, Germany, Finland, Sweden, Ireland, Netherlands). They reported average particle counts of 12.8 ± 5.2 microparticles/g globally (34 microplastics/g for EU MS, but ranging from 0.45 ± 0.2 microplastics g^{-1} in the Netherlands to 113 ± 57 microplastics g^{-1} in Italy). In addition, the paper reported that the most common particle morphology was fiber (present in 100% of the sample), followed by fragment (71%), and sphere (35%).

Van Den Berg et al. (2020) showed that the plastic loads in eastern Spain increased by 280 light density microplastics kg^{-1} and 430 heavy density microplastics kg^{-1} with each successive application of sewage sludge.

These varying outcomes probably reflect differences in soil physical (e.g. soil structure) and chemical (e.g. nutrient status) properties, which in turn can have an impact on the composition of microbial communities and reaction to the plastic material. In addition, most studies focus on single species or groups rather than the complex food webs that exist in reality where changes in a group of soil organisms can have consequences on the abundance, diversity and functioning of other groups.

One factor that hampers the assessment of microparticles in sludge is the lack of recognised standards for the extraction and analytical techniques used to isolate and identify microplastics. This may lead to notable differences in the efficiency to recover plastic particles and reporting of numbers. Density separation is the most common technique to separate plastic particles from sludge while Fourier-Transform InfaRed spectroscopy is widely used for their identification.

Further work is needed to assess the fate and transport of plastic particles in soil, especially as a result of sludge disposal.

Microplastics in soil are an increasing source of concern from both an ecotoxicological and pedological (soil formation and evolution) perspective. Changes in soil characteristics and function due to an accumulation of plastic particles can change particle size distribution and bulk density, while negatively affecting the water holding capacity. Another issue of concern reflects their persistence, leading to a continued accumulation in soil over time.

Most studies generally show that microplastics have a negative impact on soil organisms but with some uncertainties regarding outcomes. Laboratory studies have shown that the addition of polyester fibres can have both positive or negative effects on soil microbial activity (de Souza Machado et al., 2018), while Lin et al. (2020) showed that microplastics negatively affect soil fauna but stimulate microbial activity. On the other hand, other studies have reported harmful effects in some groups of soil fauna following the ingestion of microplastics (Huerta Lwanga et al., 2016) or through disruption to the habitat of soil microarthropods by closing soil pores (Kim and An, 2019). In addition, the degradation of plastics can both release additives that are contained within plastics (e.g. PBDEs, phthalate esters) while also providing a platform for attracting contaminants (e.g. PAH, PCB, metals) due to their large surface area and adsorption characteristics. The latter aspect is of particular concern given the previous discussion on contaminants of concern that are found routinely in sewage sludge. Finally, Li et al (2020) reported the accumulation of microplastics in wheat and lettuce plants in a laboratory setting. Humans are exposed to microplastics through ingestion of food and drink. Risks are currently considered as uncertain, and their understanding will increase significantly over the next decades because it will be further studied (European Chemicals Agency -ECHA, 2019)

5 Soil nitrogen, carbon and phosphorus cycling

5.1 Objective and scope

This section aims to assess the impact of sewage sludge applications on the soil biogeochemical cycles of carbon and nitrogen to characterise the potential of carbon sequestration and related greenhouse gas emissions from soil using a modelling approach. In addition, the possible contribution from sewage sludge management to addressing the EU dependence on phosphate rock imports from abroad will be discussed in a qualitative manner (see section 5.5).

5.2 Modelling approach

Biogeochemical models, such as CENTURY/DAYCENT, simulate the behaviour of C and N within and across the atmosphere, vegetation, and soil, whereas the hydrological compound is able to simulate the spatial dynamics of the N compounds (i.e. the loss through leaching based on soil properties and soil humidity patterns). The models simulate soil and hydrological processes based on daily maximum/minimum air temperature and precipitation, surface soil texture class, and land cover/use data (e.g. vegetation type, cultivation/planting schedules, amount and timing of nutrient amenDMents) at small spatial scale, that are loaded from available spatially explicit databases. In addition to providing data on net primary productivity, biogeochemical models provide information on other aspects brought forward by Member States such as greenhouse gas emissions and soil organic matter balances. The main strength of biogeochemical modelling techniques lies in the possibility to make use of well-calibrated models to simulate the long-term material C and N dynamics and the resulting plant and environmental responses under the full set of EU agroecosystems that vary in plant types, soil types, climate conditions, and fertilisation management practices.

The composition of sewage sludge has been developed from available techno-scientific literature (Fuentes et al., 2004; Walter et al., 2006; Alvarenga et al., 2007; Smith et al., 2008; Tarrasón et al., 2008; Fytili and Zabaniotou, 2008; Tambone et al., 2010; Ferreiro-Domínguez et al., 2012; Kelessidis and Stasinakis, 2012; Koutroubas et al., 2014; Krüger et al., 2014; Samolada and Zabaniotou, 2014; ISPRA, 2015; Rigby et al., 2016; Orzi et al., 2018; Cristina et al., 2019; Di Capua et al., 2020; Campo et al., 2021; Gianico et al., 2021), as well as information sources obtained from experts and organisations (e.g. EurEau). Finally, and in order to align with the closed mass-balance approach for the life cycle assessment (see section 6), information from a process-based understanding of emissions and transformation processes that take place during the composting and anaerobic digestion of sewage sludge were taken into account. The compositions of the dewatered, composted and anaerobically digested sludge are given in Table 4. The uncertainty for dewatered, as observed from the literature (17 peer review papers), indicated a coefficient of variation of 21-30% across the different properties. For the modelling exercises, a coefficient of variation aligned to this spectrum (25%) was assumed for all parameters and sludge types (Table 4).

Table 4: Assumed chemical composition of the selected materials used for biogeochemical modelling (TOC: total organic carbon; TN: total nitrogen, Min N: mineral nitrogen; P: phosphorus, K: potassium; stdev: standard deviation).

	Dewatered sludge		**Digested dewatered sludge**		**Composted sludge**	
	average	stdev	average	stdev	average	stdev
TOC (%, dry matter)	31.2	7.8	26.0	6.6	21.1	5.3
TN (%, dry matter)	3.1	0.78	5.2	1.1	3.0	0.8
TOC:TN (-)	10.1	2.5	5.0	1.5	7.0	1.7
Min N:TN (%)	12	3	32	8	12	3
P (%, dry matter)	1.8	0.45	2.6	0.54	3.2	0.8
K (%, dry matter)	0.4	0.1	0.7	0.1	0.5	0.1
dry matter (%)	23	6	23	6	60	15
methane potential (m3 CH4 /tonne volatile solids)	277	69	-	-	-	-

The approach applied here is developed in such manner that equal crop yields are simulated. Hence, the scenarios where (treated) sewage sludge is applied to agricultural land have a proportionate reduction in mineral fertilisers under the assumption that sewage sludge partially displaces mineral fertilisers. One kg of nitrogen in sewage sludge is assumed to replace about 0.75 kg of mineral N fertiliser. All results are expressed as a net difference of the sewage sludge treatment (sludge treatment) and the corresponding treatment with higher inputs of mineral fertiliser, but absence of sewage sludge in the fertilising mix applied (reference treatment).

The carbon and nitrogen dynamics were modelled for three types of sludges (dewatered, composted and digested sludge), under two different application scenarios (1 tonne dry matter ha^{-1} yr^{-1} and 5 dry matter tonne ha^{-1} yr^{-1}). In the absence of spatially explicit data on sewage sludge applications, it was assumed that sewage sludge applications take place on every spatial point of the LUCAS soil database (22 000 data points in total), and data are presented as such. It is important that point emission data should be interpreted as values for fields amended with sewage sludge, but that data cannot be summed at a wider spatial scale. To obtain aggregated data (e.g. at MS level or EU-wide), the generated sewage sludge volumes at MS level need to be considered (see section 6.3 for such an approach developed at EU scale). The model was run over a simulation period of 32 years.

Three different processes are important to determine the climate change impacts from sludge applications on agricultural land: nitrous oxide (N_2O) emissions, methane (CH_4) emissions, and carbon sequestration in soils that is accounted for as a carbon removal strategy. Results are expressed as emission factors for nitrous oxide (N_2O) emissions (kg N_2O per kg N applied in the (treated) sludge), methane emissions (kg CH_4 per kg of C applied in the (treated) sludge), and carbon sequestration (kg C sequestered per kg of C applied in the (treated) sludge) in agricultural soils. To calculate and present the carbon sequestration factors in soils, the supplementary carbon sequestration from the sludge treatment relative to the reference treatment after 32 years of continuous sludge applications was divided by the number of applications (32).

5.3 Results

EU-wide average values for the absolute greenhouse gas emissions and carbon sequestration in soils after 32 years are presented in Table 5. It is indicated that greenhouse gas emissions factors are higher for the scenarios with the higher sewage sludge applications of 5 tonnes ha^{-1} yr^{-1}, especially for N_2O emissions. This observation is because changes in soil organic C turnover feed back into the N cycle, with soils that are more saturated by soil organic carbon content showing greater N_2O emissions (Lugato et al., 2018). The N_2O emission factors are highest for digested sludge, followed by dewatered and composted sludge (Table 5); this observation may relate to the higher mineral N to total N ratios in digestates that give rise to greater N_2O emissions from nitrification-denitrification pathways (Häfner et al., 2021). N_2O emission factors for the 1 tonne ha^{-1} yr^{-1} scenario are generally well-aligned to the default IPCC emission factor for the storage of dewatered (>20%) organic materials of 0.5%.

In spite of a 5 times higher sewage sludge application rate, carbon sequestration in soils is only marginally higher for the 5 tonnes ha^{-1} yr^{-1} as sequestration under high application scenarios may be limited by the soil organic carbon saturation capacity (Lugato et al., 2014a). Expressed relative to the carbon present in the (treated) sludges applied to land, the carbon sequestration is higher for digested and composted sludge than for dewatered sludge (Table 5). The labile carbon fractions are partly removed during biological sludge treatment, for which higher carbon sequestration can be achieved per unit of carbon applied in composted and digested sludges due to the higher proportion of recalcitrant C added (Torri et al., 2014; Möller, 2015). At the same time, it has to be taken into consideration that biological sludge processing methods significantly reduce the carbon loads of the materials applied to land (with up to more than 50% of the total carbon present in the mixed sludge being removed). Therefore, the potential for carbon sequestration is still larger for untreated sewage sludge than for composted and digested sludge (see section 6).

Table 5: Average CH_4 and N_2O emission factors and C sequestration rates for EU-28.

Parameter	**Type of sludge**	**Scenario 1 tonne ha^{-1} yr^{-1}**	**Scenario 5 tonnes ha^{-1} yr^{-1}**
N_2O emissions	dewatered sludge	0.0055	0.014
(kg N_2O per kg N applied in the (treated) sludge)	composted sludge	0.0049	0.012
	digested sludge	0.0060	0.036
CH_4 emissions	dewatered sludge	0.00100	0.0012
(kg CH_4 per kg of C applied in the (treated) sludge)	composted sludge	0.00093	0.0014
	digested sludge	0.00149	0.0015
Carbon sequestration	dewatered sludge	0.21	0.25
(kg C sequestered per kg of C applied in the (treated) sludge)	composted sludge	0.28	0.36
	digested sludge	0.26	0.30

Due to the high global warming potential of N_2O (265–298 times that of CO_2 for a 100-year timescale), N_2O emissions are the most important contributor the carbon footprint of the use-on-land phase of sewage sludge. The N_2O emission factors for the 1 tonne ha^{-1} yr^{-1} treatment vary across MS, with soils from northern MS characterised by somewhat lower N_2O emissions following continued applications of (treated) sludge land (Figure 11) compared to soils located in other MS such as Italy. Similar to the N_2O emissions, CH_4 emissions vary markedly between northern and southern countries (see section 14.2; not depicted in the main text given their more limited impacts on the total carbon footprint).

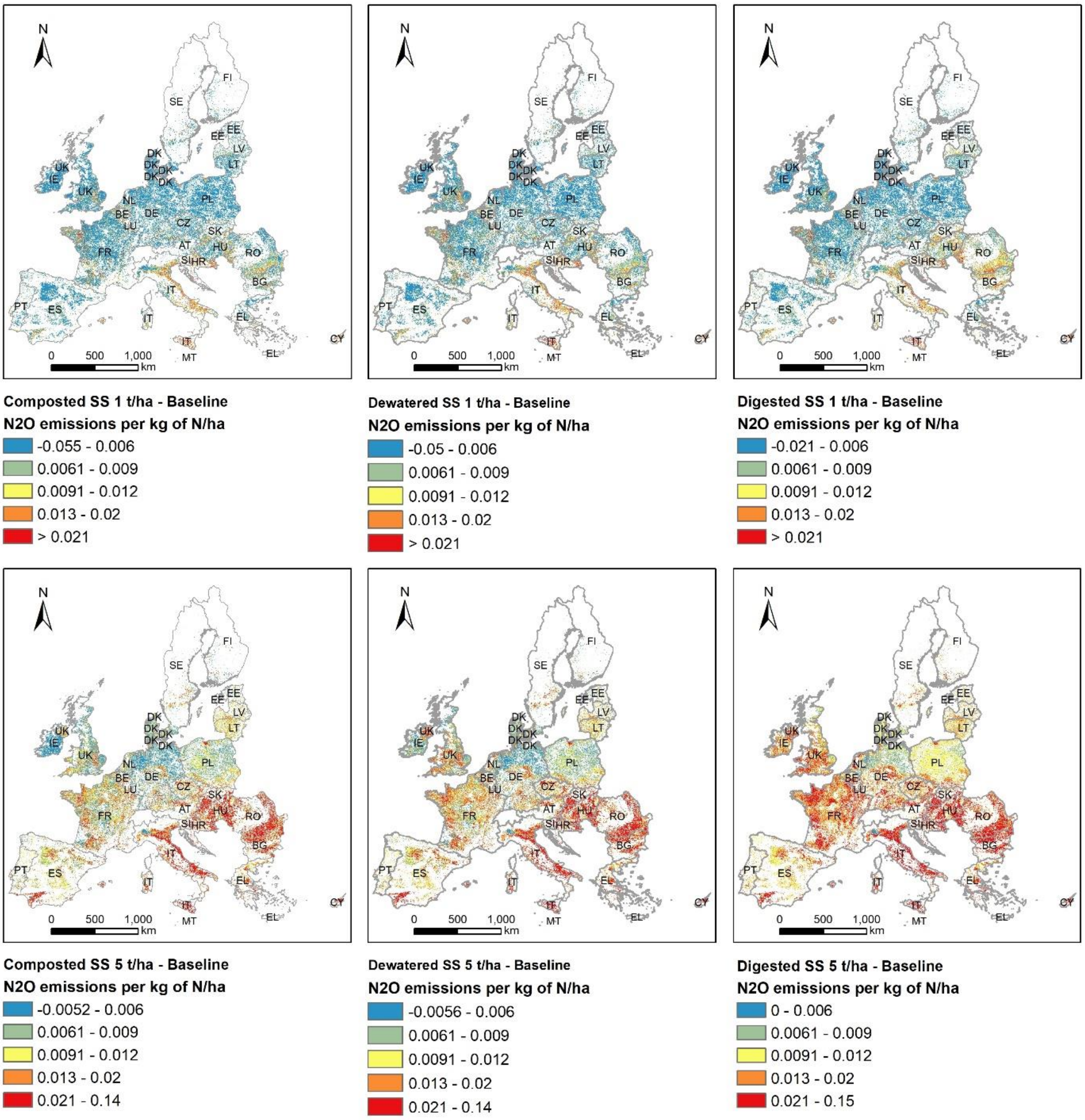
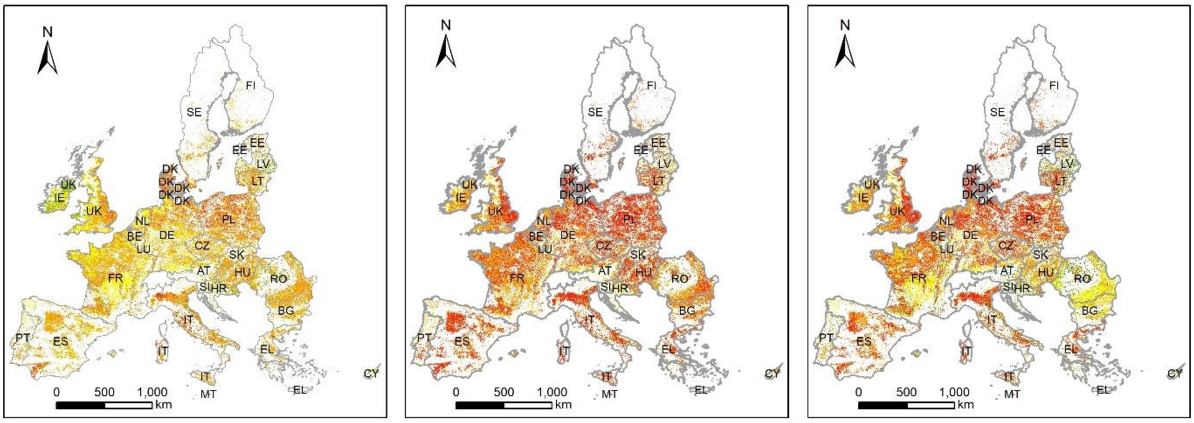


Figure 10: Estimated N_2O emissions across EU Member States for assumed sewage sludge application scenarios of 1 tonne ha-1 yr-1 (top row) and 5 tonnes ha-1 yr-1 (bottom row).

A North-South gradient is observed from the biogeochemical modelling results that estimate carbon sequestration from sewage sludge applications in soils (Figure 12). Across the different types of sludges, soils from northern countries (e.g. PL, DK) show the lowest potential to sequestrate additional carbon relative to the reference scenario of mineral fertiliser applications. A greater potential was observed for soils from MS located at southern and intermediate latitudes (e.g. PT, southern FR, RO, BG), likely due to their lower present-day organic C contents that are well-below the soil C saturation capacity (Lugato et al., 2014a).



Composted SS 1 t/ha - Baseline after 32 yr

Potential carbon sequestration factor

🟥 -0.04 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 >0.51

Dewatered SS 1 t/ha - Baseline after 32 yr

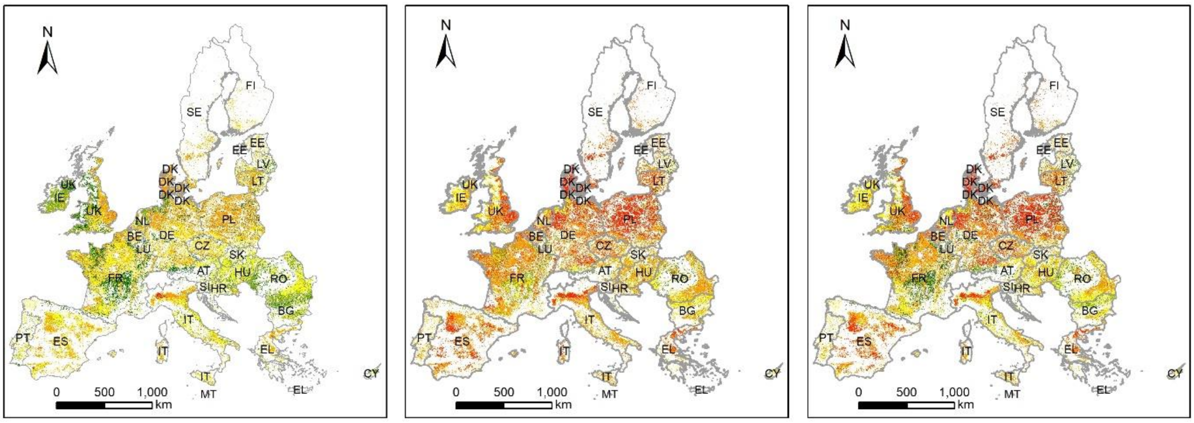
Potential carbon sequestration factor

🟥 0 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 >0.51

Digested SS 1 t/ha - Baseline after 32 yr

Potential carbon sequestration factor

🟥 0 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 >0.51



Composted SS 5 t/ha - Baseline after 32 yr

Potential Carbon factor

🟥 0 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 > 0.5

Dewatered SS 5 t/ha - Baseline after 32 yr

Potential carbon factor

🟥 0 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 > 0.51

Digested SS 5 t/ha - Baseline after 32 yr

Potential carbon factor

🟥 0 - 0.2
🟧 0.21 - 0.3
🟨 0.31 - 0.4
🟩 0.41 - 0.5
🟩 > 0.51

Figure 12: Spatially explicit carbon sequestration factors (kg CO_2 sequestered per kg C in (treated) sludge) as estimated using biogeochemical modelling for application scenarios of 1 tonne ha^{-1} yr^{-1} (top row) and 5 tonnes ha^{-1} yr^{-1} (bottom row) for composted sewage sludge (left hand side), dewatered sewage sludge (middle), and digested sludge (right hand side).

5.4 Climate change adaptation

Soil organic matter improves the soil's water retention capacity enhancing the drought tolerance by crops and improves infiltration-diminishing runoff avoiding that soil particles will be transported with water and wind (Altieri and Nicholls, 2017). Higher levels of soil organic carbon increases biological diversity which in turn creates the conditions that are hospitable to plant roots, allowing the development of strong, healthy and resilient crops (Altieri and Nicholls, 2017). For all these reasons, carbon sequestration in soil is recognised as an effective climate change adaptation strategy.

To assess the potential of sewage sludge to contribute to the climate adaptation potential, it may be relevant to compare organic C inputs from sludge relative to other sources of organic matter for agriculture. The total manure production from pigs, cattle and chicken in the EU amounts approximately 1,381 million tonnes of manure (Foged et al., 2011; Flotats et al., 2013). Solid manure fractions, with an assumed dry matter content of 25%-30%, alone account for about 719 million tonnes, thus equalling about 198 million tonnes expressed on a dry matter basis with an organic carbon content similar to sewage sludge. Food waste is estimated at about 88 million tonnes in the EU, but the (growing) share of municipal waste composted and digested in the EU was only 18% (16 million tonnes) in 2018 (EEA, 2020). Hence, in comparison with other sources, the potential of sewage sludge to deliver organic matter to soils at the EU-wide scale (presently about 2.5 million tonnes out of a total of approximately 7.5 million tonnes produced) is relatively small. Nonetheless, particularly in EU regions with a more extensive livestock production system or for soils characterised by a low organic matter content (e.g. in southern Europe), sewage sludge may be an important source of organic matter and contribute as such to climate change adaptation (Figure 12).

5.5 Phosphorus cycling

The EU is dependent on the input of rock phosphate and finished phosphorus (P) inputs from foreign nations, mostly from Western Sahara and Russia (only <15% of the P originates from mines located in the (north) EU). Moreover, phosphorus is a critical raw material that is finite. This makes the EU particularly sensitive to economic scarcity and price shocks. At present, the annual EU imports about 1.1 million tonnes (Mt) of P in mineral fertilisers, and about 0.5 Mt P yr^{-1} in feed and food for local consumption (net imports, thus substracting e.g. food exports) (Van Dijk et al., 2016). Phosphorus ending up in sewage sludge is estimated at about 0.13 Mt P yr^{-1}, of which less than half ends up on agricultural land. Hence, P in sewage sludge equals about 8% of the primary total P consumed in the EU as fertilisers and feed. Therefore, the recycling of P from sewage sludge is particularly important for the EU in its objective to increase resource efficiency and strategic autonomy.

Sludge management routes that return P to agricultural land involve the landspreading of untreated or (biologically) stabilised sludge, as well mono-incineration followed by P-recovery from the resulting ashes. The agronomic P efficiency of sludge spread on land is typically lower than for P present in mineral fertilisers, even though this is dependent on the degree of complexation with Fe and Al polymers used in wastewater treatment plants (Wilfert et al., 2015). Phosphorus recovery can also take place upstream during wastewater treatment (e.g. precipitation of phosphate salts such as struvite), and this process will affect the P-content of sludge and thus the environmental performance of sludge treatment.

5.6 Conclusions

Greenhouse gas emission and carbon sequestration from the use-on-land phase of sewage sludge may jointly contribute to the carbon footprint of sewage sludge management options. In addition, phosphorus return to agricultural land is a critical point to be considered during the assessment of the environmental performance of sewage sludge management. The biogeochemical modelling results indicate that (i) biological sludge treatment impacts upon the climate change mitigation potential of the use-on-phase of (treated) sewage sludge, and (ii) local climate and soil conditions largely influence the carbon footprint of sludge land application. The biogeochemical modelling data obtained will be further used in section 6 that compares the global warming potential of different sewage sludge management options using a life cycle perspective.

With respect to climate change adaptation, it is indicated that sewage sludge is a small source of organic matter compared to e.g. manure and separately collected bio-waste, Nonetheless, under some regional settings and for some soils, sludge may locally be an important organic matter input that can contribute to the build-up of soil organic C and resilience to climate change. The proper management of P contained in sewage sludge is important of view of reducing the EUs dependence on imported P fertilisers and rock phosphate.

6 Impacts on climate change mitigation

6.1 Objectives and scope

The objective of this section is to assess and compare the global warming impacts from different sludge management options via a life cycle assessment approach (LCA).

This LCA allows comparing the environmental performance from a climate change mitigation perspective of different sludge management options. In addition, it allows bringing forward an absolute estimate of the net global warming potential (kg CO_2-eq per year) associated to sewage sludge management within the EU.

6.2 Methodology

The functional unit of the study is the management of 1 tonne (dry matter) of sewage sludge produced at a wastewater treatment plant. The assessment of the environmental aspects and potential impacts associated with a product was performed following ISO standards for LCA (ISO, 2006a, 2006b). A zero burden global warming potential is assumed for the C present in sewage sludge, a short-lived and wasted biomass of biogenic origin. A consequential approach was applied to capture the environmental consequences of the displacement of conventional market products (e.g. NPK fertilisers and electricity) (Weidema, 2003; Tonini et al., 2019). Hence, system expansion has been performed in order to ensure equivalence of functions across scenarios. The impacts resulting from the transformation of carbon, and nutrients (N, P and K) were considered using a closed mass balance approach in the foreground system of this LCA, focused on Climate Change as the sole impact category (Forster et al., 2007). The temporal scale boundary – especially relevant for long-term processes such as greenhouse gas losses at landfills and carbon sequestration in soils – was set to a period of about 30 years. The assessment was carried out with the LCA tool EASETECH (Clavreul et al., 2014).

The system boundaries start from mixed sludge generated at wastewater treatment plants and expire with the final use of sludge or derived materials on agricultural land or elsewhere. The life cycle stages thus involve primary processing at wastewater treatment plants (e.g. dewatering, thickening), secondary sludge treatment (lime stabilisation, anaerobic digestion, composting, co-incineration, mono-incineration followed by ash acidulation to manufacture a P-fertiliser), transport from wastewater treatment plants to other facilities and agricultural land, sludge storage prior to use on agricultural land, and final use of derived materials (use-on-land, construction material, landfilling). The life cycle approach takes into account both climate burdens (e.g. from energy requirement for sludge processing, chemicals, greenhouse gas emissions during processing and use-on-land) and savings (e.g. substitution of energy, mineral fertilisers, sequestration of sludge-derived carbon in agricultural soils). Internal waste heat recovery was assumed to increase the efficiency of the process and promoting process savings (e.g. for sludge heating and drying). The results from the use-on-land phase of (treated) sewage sludge as developed in section 5 have been plugged into the LCA model (results of 1 tonne ha^{-1} yr^{-1} application scenarios). Greenhouse gas emissions and carbon sequestration from the use-on-land phase of sludge are already expressed as the net impacts relative a scenario with equal crop yields following mineral fertiliser application.

Departing from the assumed sludge composition as outlined in section 5, 7 different scenarios have been selected for this modelling exercise (Figure 13), aiming to represent some main important sludge management options that take place in the EU (see section 2). The options selected are not representing all sludge management options or combination thereof that could take place. Nonetheless, it is believed that assessing these pathways sheds light on the key processes and life cycle stages that contribute to the overall global warming potential. They include options that involve use-on-land of sludges, possibly followed after chemical or biological treatment. Also, the option of the use-on-land of acidulated sewage sludge mono-incineration ashes (in the form of a mineral P-fertiliser) is considered. Finally, two pathways that result in the removal of nutrients and organic carbon from the biogeochemical cycle are considered: co-incineration followed by landfilling and use as a construction material of the ashes, and sludge landfilling (Figure 13). Our entire assessment is based on present-day "scenario analysis modelling", and the results are neither timeless nor exhaustive nor specific to particular manufacturers, industry sectors or situation-specific settings. Rather, this work intends to assess the possible impacts of sludge management options in general, and to provide numerical data that may help to better conceptualise and understand possible impacts from sewage sludge in the broader context. Uncertainties related to assumed parameter values is discussed (section 6.4). Model input data for the different processes applied are given in the Annexes to this report (section 15.1)

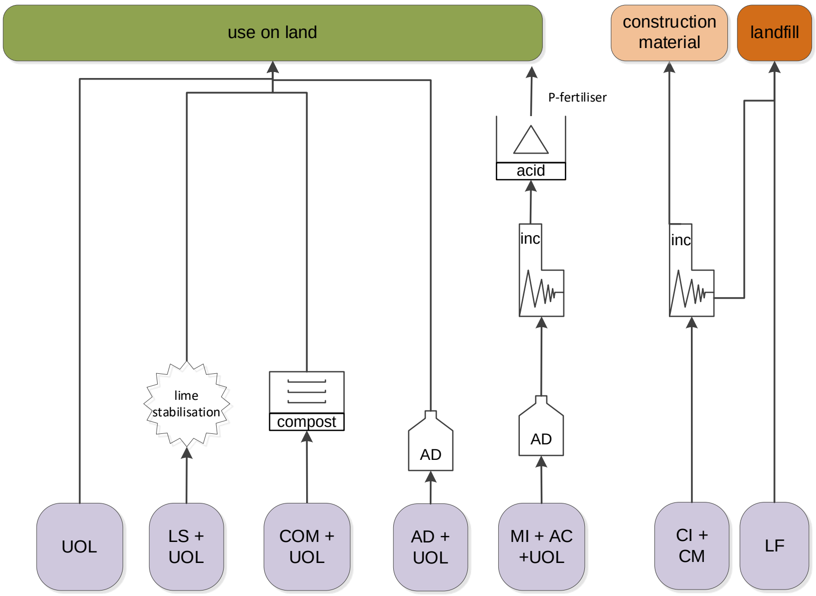


Figure 13: Schematic and simplified representation of the seven modelling scenarios (UOL: use-on-land, LS: lime stabilisation, COM: composting, AD: anaerobic digestion, MI: mono-incineration, AC: acidulation, CI: co-incineration in municipal solid waste incineration plant, CM: use as construction material, LF: landfilling).

6.3 Results (average values)

Net average results sum burdens (positive values) and savings (negative values) vary from -268 to 1650 kg CO_2-eq / tonne sewage sludge DM) (Figure 14).

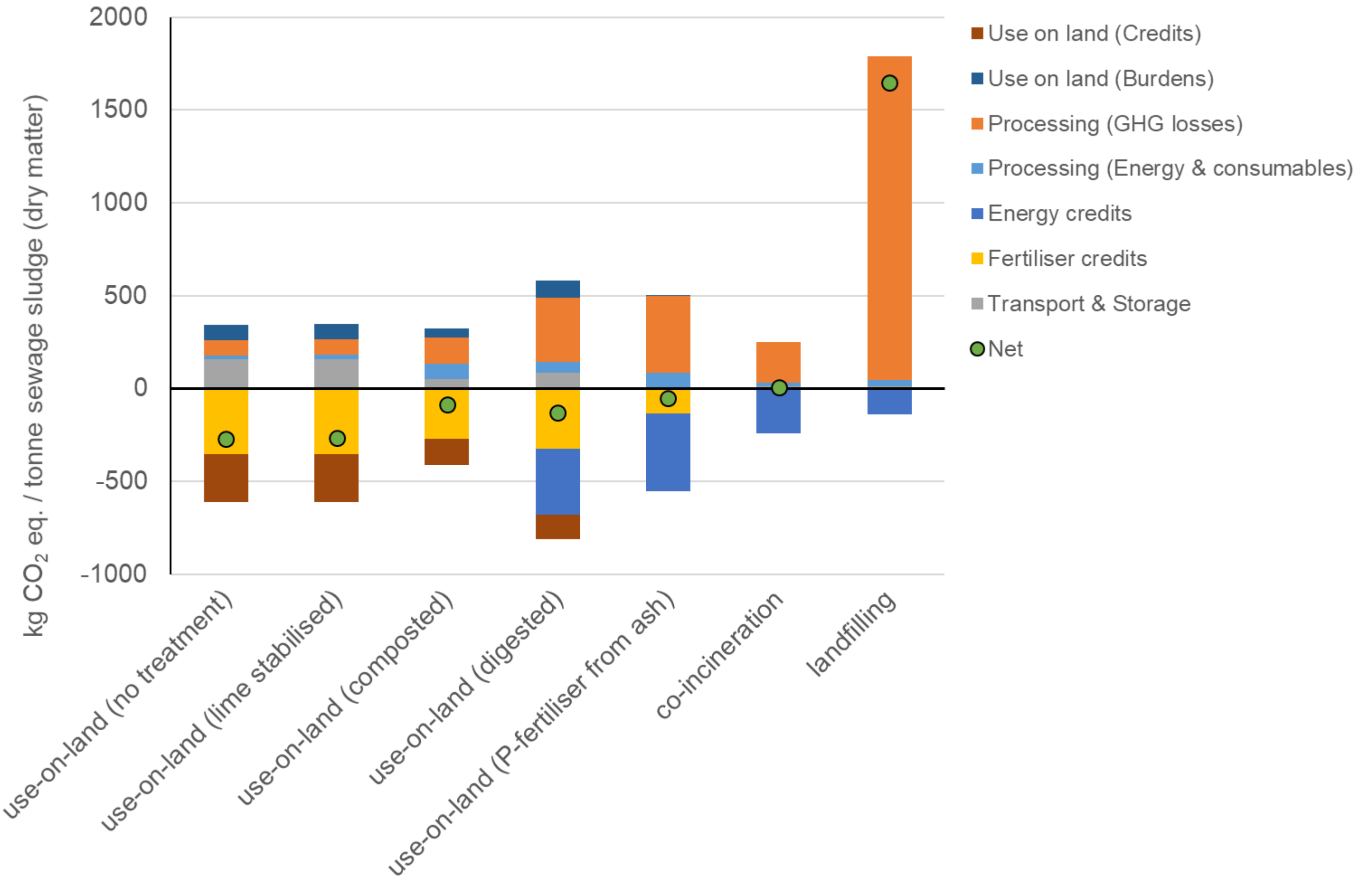


Figure 14: LCA results for the Global Warming Potential for selected pathways (see Figure 13 for pathway description).

The best performing pathways in this analysis are direct use of untreated or lime stabilised sewage sludge on agricultural land. The use-on-land of anaerobically digested and composted sludge, as well as incineration are intermediately ranked options, whereas landfilling is the worst performing pathway associated to a large positive value (Figure 14).

The difference in global warming potential between land application of unprocessed and lime stabilised sludge is minimal given that the production and use of lime to sludge has a minimal carbon footprint, and other emissions were assumed equal in the absence of a well-developed dataset. The processes that contribute to the observed net negative removal of CO_2 through these pathways are the credits from the avoided production, transport and use of mineral fertilisers and carbon sequestration in soils (Figure 14). Emissions occur from the storage and transport as well as to a smaller extent from the initial processing (primary treatment such as dewatering and thickening) of sewage sludge (Figure 14). Nonetheless, since savings are greater than burdens, a net negative carbon footprint is indicated for both pathways (Figure 14).

Biologically stabilised sludges obtained following composting and anaerobic digestion also have a negative global warming potential (Figure 14). Since a significant share of the carbon and nitrogen present in sludge is lost during the composting process, the savings from the use-on-land of composted sludge are smaller compared to use-on-land of untreated sewage sludge. The losses during storage and use-on-land of compost are also smaller compared to that pathway due to the increased stabilisation of the sludge, but the processing stage shows an opposite trend due to the additional energy requirements and greenhouse gases emitted. Anaerobic digestion followed by the use on land of the resulting digestate is the pathway that shows the highest savings in the form of summed fertiliser credits, energy credits and soil carbon sequestration. Fertiliser credits are higher than for compost as N is largely retained during digestion and the slightly higher nitrogen fertiliser displacement value of the mineral nitrogen-rich digestate (Möller, 2015). Nonetheless, the burdens for this pathway are also high due to the methane leakage at anaerobic digesters, as well as higher N_2O losses during the use-on-land phase of the ammonium-rich digestates (see section 5.3).

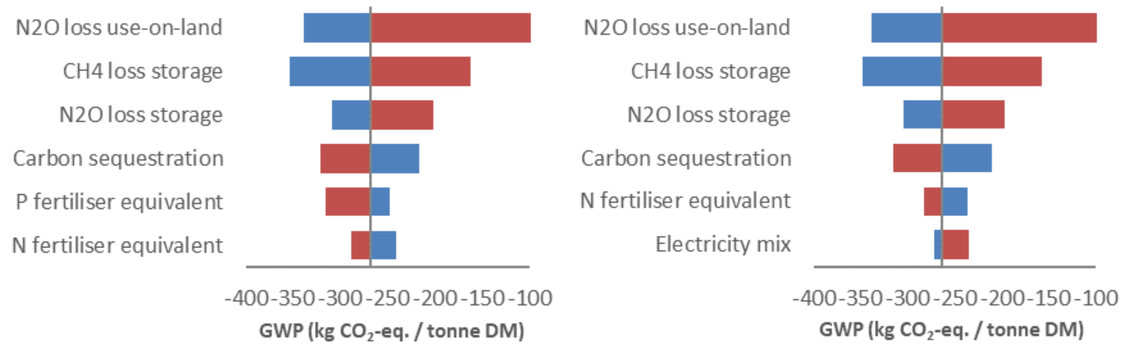
The net impacts of the co-incineration are close to zero as the greenhouse gas emissions (N_2O) (Gutierrez et al., 2005) and energy requirements for the incinerator (e.g. fossil fuels for start-up) are largely compensated by renewable energy production at modern combined heat and power plants (CHP) (Figure 14). Energy credits obtained from energy recovery during incineration are significant, and could be further improved in case sludge is dried using low impact systems (e.g. with waste heat from other industrial neighbouring installations, solar drying).

The emissions from the scenario of P-fertiliser manufacturing from mono-incinerated sewage sludge through acidulation are similar to those from co-incineration. The main process contributing to the burdens are the significant N_2O emissions that have been observed as N_2O emissions concentrations in the off-gasses ranging from approx. 50 to 200 mg N_2O m^{-3} (average value assumed of 125 mg N_2O m^{-3}; slightly higher than for co-incineration (75 mg N_2O m^{-3}) due to the slightly lower temperature and air to fuel ratio for fluidised bed incineration relative to grate incineration) (Svoboda et al., 2006; Gutierrez et al., 2005; Korving et al., 2010). The fertiliser credits are lower compared to the pathways that apply (treated) sludge due to the loss of N during incineration. On the other hand, the P-fertiliser credits are higher compared to other pathways because the increased P availability of the produced (mineral) fertiliser (Oenema et al., 2012; Wilfert et al., 2015).

Landfilling is the worst performing pathway from a global warming perspective due to high methane losses, even in modern facilities (Figure 14).

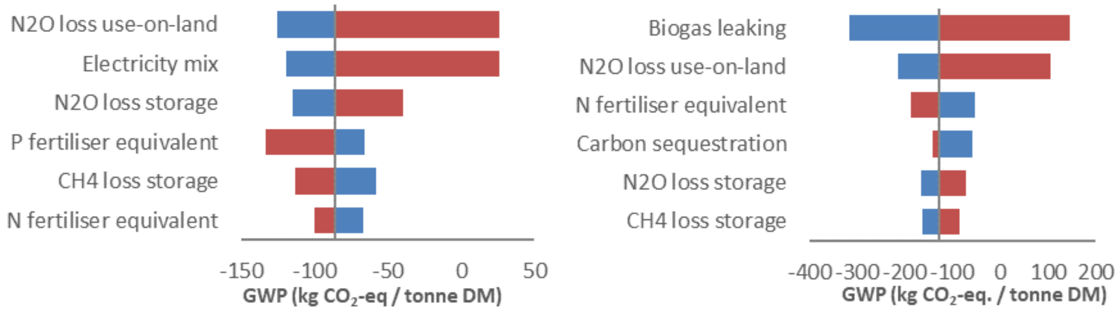
6.4 Uncertainty analysis

Section 6.3 provides results based on assumed average values for process parameters, but it is recognised that process parameters vary across industrial facilities and that greenhouse gas emissions may depend on e.g. storage conditions, electricity mix used, sludge fertiliser equivalency. Assumed parameter values used in this uncertainty assessment are given in section 15.1. The uncertainty analysis indicates that the carbon footprint of a single pathway may fluctuate substantially (Figure 15), and that the performance ranking across pathways can be altered. Figure 15 provides an overview of the parameters that most contribute to the observed uncertainty as well as the expected impacts of their variability.



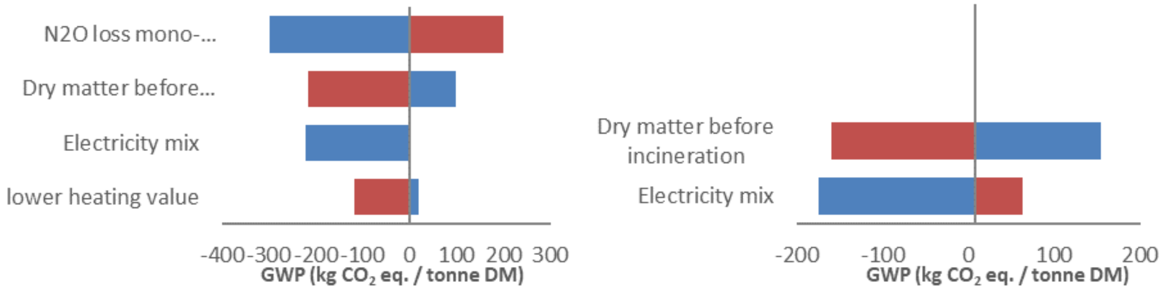
a) use-on-land (no treatment)

b) use-on-land (lime stabilised)



c) use-on-land (composted)

d) use-on-land (digested)



e) use-on-land (P-fertiliser from ash)

f) co-incineration

Figure 15: Sensitivity analysis on the global warming potential for the selected pathways investigated. The red bar represents the global warming potential for the maximum value of each parameter. The blue bar represents the global warming potential for the minimum value of each parameter.

For untreated and lime stabilised sludges used on land, most of the uncertainty originates from greenhouse gas losses during the use-on-land phase and the storage phase. N_2O emissions from the use-on-land phase vary widely across EU regions as a function of soil and climate type (see section 5.3). Storage can be minimised to very low levels when applying best available techniques (e.g. covering, creating a well-aerated environment) and limited storage times (Willén, 2016; Willén et al., 2016; Giner Santonja et al., 2017; Samuelsson et al., 2018), and this may further reduce the global warming potential of this pathway by approximately 200 kg CO_2-eq / tonne sewage sludge DM. The carbon sequestration potential of sludge varies across the EU and may further impact upon the pathway performance, as well as mineral fertiliser equivalence of the sludge (Figure 15 a/b). The phosphorus fertiliser equivalence relative to mineral fertilisers is highly dependent on the Al and Fe content, with sludges characterised by a high (Al+Fe)/P ratio showing a lower plant availability of phosphorus than e.g. sludge produced at biological wastewater treatment plants (Oenema et al., 2012; Wilfert et al., 2015).

The uncertainty related to the global warming potential for the compost pathway is the lowest across all pathways analysed (Figure 15 c). Greenhouse gas emissions from the storage and use-on-land phase vary, but to a smaller extent compared to other pathways given the increased stability of the compost. The main parameter that influences the compost pathway are the N_2O emissions from the use-on-land phase as well as the electricity mix. In case an energy mix with a higher carbon footprint (more fossil fuels) is applied (ventilation, mixing at composting facilities), the carbon footprint of the pathway may show a net positive value (Figure 15.c). Also, emissions from storage are important, but to a smaller extent relative to direct use-on-land of sludge as total losses from storage are lower for the more stabilised compost. Composts characterised by a higher fertilising value show a better performance (Figure 15). Finally, N_2O emissions from compost are important, with much higher greenhouse gas emissions from outdoor windrow composting systems than from controlled aerated systems (Vergara and Silver, 2019).

For anaerobic digestion, the methane leakages from digestion plants are key to the overall impact of this pathway. Methane leakages has been documented to range from 0.1% to 6% (IAEA Bioenergy Task, 2017), and the net impact of best versus worst performing pathways is about 500 kg CO_2-eq tonne^{-1} dry matter. When leakages are minimised, the performance of this pathway is even better than direct use-on-land of untreated sewage sludge, whereas net emissions will turn positive when sludge is being digested in plants characterised by high methane leakages (Figure 15.d). Again, N_2O emissions from the use-on-land are an important contributor to the observed pathway variability.

For mono-incineration plants, the critical factor to the overall emissions for this pathway are the nitrous oxide emissions during the incineration process (Figure 15.e). Concentrations of N_2O in off-gases have been documented to range from 2 to 400 mg N_2O/Nm^3 gas (Korving et al., 2010; Svoboda et al., 2006). It may be important to corroborate if these ranges still apply to modern mono-incineration facilities as emissions may have reduced over time, e.g. due to higher temperature applied in the afterburner chamber (Neuwahl et al., 2019). In case the N_2O emissions from mono-incinerators were minimal, the global warming potential of the pathway that involves the acidulation of mono-incinerated ash to produce a P-fertiliser would be close to zero (Figure 15). Co-incineration technologies typically have much lower N_2O losses than mono-incineration (IGES for the IPPC, 2002). In addition, the moisture content of the sludge has a large impact on the performance of the incineration pathway. In case sludge is dried using low energy demanding techniques (e.g. solar drying, application of waste heat) the performance of the pathway would further increase (Figure 15).

6.5 Impacts of upstream wastewater treatment processes

The Urban Wastewater Treatment Directive (91/271/EEC; UWWTD) requires nitrogen removal between 70 and 80% of influent loads in specifically designed sensitive zones. The current state-of-the-art with biological WWTPs allows a removal of 80%, and a removal of 90% could be achieved with well-targeted and accurate plant operation. Policy options for the UWWTD include an increase of N removal efficiency to more than 80%. When this is the case, the carbon present in the influent needs to be preserved for the stoichiometry of denitrification, for which reason we expect a reduction of carbon in the sludge and, as a consequence, a possible reduction of the potential to produce CH_4 through anaerobic digestion or energy through incineration. While in well-balanced wastewater treatment processes an energy-positive anaerobic digestion is possible also with high denitrification, it should be well understood that it is not possible to maximize at the same time denitrification and downstream renewable energy production from sludge treatment. In terms of global warming potential, the abatement of N_2O emissions due to partial denitrification at the wastewater treatment is expected to have a greater impact than the downstream energy credits from sludge processing on the overall balance. Moreover, enhanced denitrification brings benefits in terms of water quality. All this considered, enhanced denitrification upstream should be regarded as a priority over renewable energy production from downstream sludge management.

6.6 Relative importance of sewage sludge management for climate change

Based on gap-filled Eurostat 2016 data (see section 2), it is assumed that roughly 7.5 million of tonnes sludge (dry matter) are generated and used directly or after digestion in agriculture (35%), incinerated (31%), composted (12%), landfilling (12%) or managed otherwise (10%). Main uncertainties to these data exist (Wood E&I GmbH, 2021), but a further assessment of sludge management pathways falls beyond the scope of this work. To frame the absolute global warming impacts, we will approximate a scenario where 100% of the generated sludge is managed according to one of the pathways evaluated above, as follows:

- Direct use in agriculture: 30%;
- Use-on-land after anaerobic digestion: 11%
- Use-on-land after composting: 12%
- Co-incineration: 25%;
- Mono-incineration: 10%
- Landfilling: 12%

Based on this simplified approach and acknowledging the uncertainties associated to the dataset and limitations of the approach applied, sludge management leads to a net emission of about 0.67 million tonnes CO_2-eq per year. The absolute footprint from sewage sludge management is relatively small relative to operations that take place at upstream (sewage system and wastewater treatment plants), estimated at around 25-30 million tonnes of CO_2-eq per year (unpublished data from JRC policy report on the Urban Wastewater Treatment Directive). The agricultural sector as a whole produced 426 million tonnes of CO_2 equivalent of greenhouse gases in 2015, about 10% of the EU's total greenhouse gas emissions (excluding Land Use, Land Use Change and Forestry net removals) for that year (Eurostat, 2021).

Nonetheless, a worst case alternative scenario with 88% of sludge being incinerated, and 12% of the sludge being landfilled, would increase the global warming potential to about 1.5 million tonnes CO_2-eq per year. In a best possible scenario where all sludge is being applied directly on agricultural land would minimise emission to -2.0 million tonnes per year. Hence, the application of sustainable sludge management has a potential to reduce the global warming footprint with up to 3.5 million tonnes CO_2-eq per year. It is therefore concluded that sustainable sewage sludge management is important to reduce the carbon footprint of the entire wastewater treatment system, including sludge management. Sludge management can be applied as net carbon removal strategy and offset as such a part of the carbon footprint of upstream wastewater collection and collection processes.

6.7 Supplementary potential for energy recovery from sludge from digestion and incineration with energy recovery

Sludge processing techniques such as anaerobic digestion and incineration have the potential to recover part of the energy contained in the sludge. Estimations on the potential are highly dependent on assumptions, including methane potential and lower heating values, technology configurations, and sludge volumes that subject to these treatments. The European Biogas Association estimated that in 2019 about 4642 GWh of gross electricity was produced from the methanation of wastewater treatment plant sludge, with a dominant contribution from plants located in Germany[14]. The main method of taking up biogas energy is producing electricity in CHP or other plants. Assuming an electric efficiency of 35%, this would correspond to about 1625 GWH_{el} per year from sewage sludge. Under the assumption that an additional 30% of the sludge volumes could be subject to anaerobic digestion (2.25 million tonnes dry matter; see above), and an electric energy generation potential of 1500 kWh per tonne dry matter from sludge digestion, about 3375 GWh_{el} of additional renewable gross energy could be produced. Energy production from sludge incineration is subject to high uncertainties because pre-treatments, including sludge drying techniques, have widely differing energy intensities (e.g. solar drying versus rotating drums). Under the assumption of a potential of diverting 1 million tonnes of sludge dry matter from treatments without energetic valorisation (e.g. from landfilling without energy recovery) to incineration with energy recovery, and an electric energy recovery potential of about 1020 KWh per tonne dry matter sludge for incineration, 1020 GWh additional renewable gross energy could be produced. Hence, a reasonable high end potential for additional energy recovery from the anaerobic digestion and incineration of sludge equals about 4.4 TWh_{el}, or 0.16% of the EUs 2020 total net electricity generation.

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[14] See "2020 Biogas Barometer" report available at: https://www.eurobserv-er.org/biogas-barometer-2020/

6.8 Conclusions

Different sewage sludge management options give rise to net emissions of CO_2-eq that range from net negative emissions for direct land application of untreated sludge (-268 kg CO_2-eq / tonne sewage sludge DM), over values closer to zero for biologically treated sewage sludge applied on land and co-incineration, to relatively high net emission values for landfilling (1485 kg CO_2-eq / tonne sewage sludge DM). The results for all the different pathways vary substantially depending on technological implementation (e.g. methane leakage from digesters, N_2O losses from incinerators, coverage at sludge storage facilities) as well as on local agricultural conditions (e.g. N_2O losses from sludge use-on-land, mineral fertiliser equivalence of (treated) sludge). The absolute impacts from sludge management within the EU are small relative to e.g. upstream emissions from wastewater collection and treatment. However, the application of best versus performance techniques for sludge management across the entire EU would result in a net greenhouse gas emissions savings of about 3.5 million tonnes CO_2-eq. Therefore, it is important to consider climate change impacts from sludge management in tandem with other social, environmental and economic impacts when developing sustainable sludge management strategies.

7 Limitations and recommendations for further work

The aim of this work is to increase the techno-scientific knowledge base on (i) risks from pollutants and other contaminants of emerging concern in sludge applied to agricultural land, and (ii) environmental impacts arising from sewage sludge management in the EU. Quantitative and qualitative results on these aspects are presented in the sections 3 - 6 of the report.

The level of analysis is framed within the constraints of the available time, resources and data for this study. This section provides an overview of the limitations that were associated to our work and conclusions, as well as recommendations for complementary work to support policy development on the SSD.

7.1 Issue #1: Compare and discuss risk screening approach with other information sources

Background and observed limitation: Prioritisation of chemical pollutants is needed so actions first address chemicals that drive environmental and human health risks. The outcomes generally align to works of similar nature that have assessed a reduced subset of the pollutants (Norwegian Committee for Food Safety, 2009; Clarke and Smith, 2011; Deleebeeck et al., 2021). However, this is the foremost extensive screening study on risks from chemicals in sludge on the basis of currently available data. Modelling data input is mostly obtained from data repositories and often based on default assumptions and estimations. The data presented in this report have thus been collected in line with existing scientific standards and judgements. However, the authors of this study have mostly a background in environmental engineering, and complementary views from other angles of incidence and scientific domains (e.g. (eco)toxicology) could further enrich this work.

Recommendation: Whereas we are confident on our approach undertaken, additional, independent points of comparison (e.g. studies and expert opinions) and complementary technical discussions with experts in the field of risk assessment would further increase the robustness of this study. Such action could be organised e.g. in the form of a dedicated workshop and/or via the monitoring of newly published techno-scientific literature on this topic.

7.2 Issue #2: Address uncertainty on organic pollutant concentrations present in sewage sludge

Background and observed limitation: This work invested substantial efforts to chemically characterise sewage sludge in an as broad as possible manner. The methodology was developed to spot so-called contaminants of emerging concern for which reason an *a priori* selection of contaminants for assessment was omitted. However, it is noted that a bias is possible as measurements of contaminants in wastewaters and sewage sludges could have overlooked certain contaminants and/or inventorised concentrations that are not representative for all situations within the EU. This may be particularly important as the present database on contaminants in sludge may have been unable to capture (i) the latest trends for certain contaminants, e.g. in response to the latest technological developments ('newly developed chemicals') and/or legislation on chemicals (e.g. REACH restrictions to chemicals in consumer products), and (ii) regional and/or local differences in sludge quality (e.g. distinction between sludge from plants located in rural versus industrialised areas). Finally, for some pollutants in sludge (e.g. polychlorinated paraffins) measurement standards are missing.

Recommendation: Promote further efforts in the monitoring and measurement of organic contaminants in sewage sludge to acquire actual data on sewage sludge quality and variations therein as a function of local conditions within the EU. This could possibly take place in the form of a renewed measurement campaign and effect-based methods on sewage sludge samples collected across the EU (similar to Tavazzi et al., 2012) or through the promotion of such research in EU and national research programmes. In addition, the development of measurement standards for key priority pollutants identified could be envisaged if these are not yet available.

7.3 Issue #3: Improve database on ecotoxicological, toxicological and physico-chemical properties of organic contaminants

Background and observed limitation: The collection of accurate, up-to-date and comparable data is a prerequisite for informed risk assessment and for risk management decisions. A complete database containing data on (i) on the sensitivity of the organisms and/or endpoint under study to the contaminant ('predicted no-effect concentrations'), and (ii) physico-chemical data to model the dynamics and exposure of the contaminant in across different environmental compartments is needed. As indicated in section 3.3.1.1 and 3.4 such data is not available for all contaminants that are present in sewage sludge. For instance, standard tests to assess impacts on different trophic levels of soil organisms are hardly available, and more than 600 contaminants for which sewage sludge concentrations could be retrieved, could not be assessed in the risk assessment due to missing model input data. Hence, in spite of a well-developed database for many recognised contaminants, including those identified in techno-scientific literature as being of concern for sludge, the risk assessment remains incomplete.

Recommendation: Support stakeholder engagement, and calls for data that aim at characterising and inventorying contaminants in terms of (eco)toxicological and physico-chemical properties, as well as data sharing. This recommendation is likely not limited to sewage sludge as a carrier for contaminants to the environment, and should thus be framed within a horizontal initiative to support action programmes on chemicals, e.g. aligned to the zero-pollution action plan. For instance, the European Partnership for the Assessment of Risks from Chemicals (PARC) already seeks to develop next-generation chemical risk assessment, incorporating both human health and the environment in a "One Health" approach.

7.4 Issue #4: Need for an in-depth risk assessment for identified organic priority contaminants and mixtures thereof

Background and observed limitation: The risk assessment is carried out in a stepwise procedure starting from a wider spectrum of potential contaminants to identify pollutants of most concern. The exposure assessment in aims at 'reasonable worst-case' results by applying unfavourable, but not unrealistic, standard exposure scenarios and, as much as possible, mean, median or typical parameter values. If the outcome of the reasonable worst case risk characterisation indicates that the substance is "not of concern", the risk assessment for that substance can be stopped. If, in contrast, the outcome is that the substance is "of concern", the assessment must, if possible, be refined in a subsequent phase. Moreover, the "screening step" has been executed in this study assesses risks to soil organisms and humans following indirect intake of food grown on sewage-sludge amended soils. Other end-points (e.g. aquatic organisms, sediment organisms, birds, other small mammals) were not considered in this assessment. With respect to aquatic organisms and water bodies, it is expected that the impact from sewage sludge applications on agricultural land is small relative to the contaminant load entering water bodies via wastewater treatment effluents. Contaminants that are partitioned to the sludge within the treatment process, rather than to the effluent, show a high affinity for the organic (solid) phase. Therefore, these contaminants are expected to be largely retained within the soil matrix instead of being lost via runoff or leaching. Soil organisms could be a proxy for risks to aquatic organisms and to broader wild life (higher organisms such as birds) for compounds that do not bioaccumulate in the food chain. Humans are considered in this assessment as the top predator that ingest multiple food sources (fish, crops, meat, and drinking water). The results for this end point also could at a maximum be a first proxy for predators that consume fish, plants and soil organisms (e.g. mammals and/or birds), and thus for secondary food poisoning via the aquatic and terrestrial food chain. Finally, the analysis had a limited focus on degradation products of the chemicals that were measured in sewage sludge.

Recommendation: This risk screening assessment points towards priority pollutants that cause most risk when present in sewage sludge. Dependent on the final objective of the risk study, a refined assessment for all or a subset of these priority compounds could be undertaken that further develops and updates this work amongst others with information collected in response to issues #1- #3. Such assessment must in a subsequent step (i) further develop the assessment for additional end points (e.g. aquatic organisms and and top predators (birds, fish of higher trophic levels) for bioaccumulative compounds), (ii) update results by the results of other models (of a higher Tier level) judged to be more suitable for the pollutants under investigation or for certain critical end-points, (iii) perform a detailed analysis of degradation products, (iv) assess the risks from chemical mixtures containing priority compounds, and (v) advance the source contribution analysis by considering inputs from e.g. contaminants present in atmospheric deposition and other fertilising products at the regional scale. Such analysis may ultimately bring forward limit values for a reduced set of pollutants to limit environmental and health risks to acceptable levels.

7.5 Issue #5: Assess environmental and health impacts from metals

Background and observed limitation: Albeit their prevalence in sewage sludge has declined in recent decades, metals are key substances of concern in sewage sludge due to their persistence in soils and toxicity in the environment and food products. The scope of the risk screening assessment excluded metals, and focused exclusively on organic chemicals of concern as the knowledge base on this group of substances is less developed compared to metals.

Recommendation: Assess which metals are of concern in view of the reduced metal contamination in food-chain and based on the revised health recommendations on metals in food products since the entry into force of the Directive.

7.6 Issue #6: Assess innovative sludge management technologies

Background and observed limitation: This assessment of climate change impacts has principally focused on sludge management routes that are commonly applied within the EU, and thus jointly determine the overall climate change impacts from sewage sludge management across the EU. Innovative sludge processing technologies, sludge pre-and post-treatments, and combinations thereof are numerous. Common sludge processing technologies are continuously being refined and optimised with a view to increase process efficiency and minimise adverse environmental outcomes. Rationally, we have been unable to assess each of the individual configurations and facilities that process sewage sludge.

Recommendation: It could be further evaluated if market-competitive technologies exist that show a disruptive potential to lower the carbon footprint of sludge management. Feedback may be requested from stakeholders to corroborate model data input values (averages and ranges reported in this study) in the light of new technological developments.

7.7 Overall considerations and policy recommendations

This study provides an initial evaluation of the environmental and health impacts and risks associated to sewage sludge management. It is essential that the reader and users of the results presented understand the inherent limitations of the estimation methods and, finally, to correctly interpret the results. In our view, the results are useful to complement the evaluation of the effectiveness of the SSD. In addition, they could be used as a starting point to develop and evaluate possible future options and instruments to support policy development on the Sewage Sludge Directive. Aligned to this, it is equally important to further develop the understanding of the actual impacts, risks and contributions of sewage sludge to the wider environmental pressures and food safety issues. Such information may help to better understand the hazards and risks from sewage sludge that are acceptable to stakeholders (e.g. on risks to soil organisms, from microplastics), as well the targeted level of ambition for future policies on sewage sludge.

8 General conclusions

This report further increased the knowledge base on social and environmental impacts from sewage sludge management in the EU.

Firstly, our results on risks from organic pollutants and the environmental performance of sewage sludge management options indicate apparent trade-offs. Thermal (incineration) and to a smaller extent biological (composting and anaerobic digestion) sludge treatment options may completely or partially remove the chemical pollutants as well as microplastics and antimicrobial resistance genes that may cause risks to human health and the environment (e.g. PAH, PCDD/F+dl-PCB, long-chain PFAS, SCCPs/MCCPs.) Nonetheless, seizing greater human and soil health benefits from sludge treatment may – generally speaking – induce opposite impacts on the environmental performace of sewage sludge management. For instance, the direct use-on-land of untreated or lime stabilised sewage sludge has a net negative carbon footprint and returns organic matter phosphorus to agricultural land, and can therefore act as a climate change abatement strategy. In contrast, sludge disposal pathways such as landfilling cause a loss of phosphorus and higher greenhouse gas emissions without causing risks to the food chain. Innovative options, such as mono-incineration followed by P recovery from the ashes, show an intermediate performance when considering all sustainability dimensions.

Secondly, it is shown that climate change impacts as well as risks from the use-on-land phase of sewage sludge may vary spatially across the EU. The use-on-land phase of the sludge contributes significantly to the overall results on global warming potential across different sewage sludge management options, and a great spatial variability is observed for the emissions as a function of soil and climate types. Additionally, environmental and health risks from chemical pollutants in sewage sludge applied to land are linearly correlated to sludge application rates. Hence, cost-benefit ratios may be higher for EU regions characterised by a high ratio of agricultural land to population and livestock equivalents than for EU regions where sewage sludge can be applied at lower application rates, aligned to carbon and nutrient needs of soils close to wastewater treatment plants. This points towards the importance of considering flexibility to implement sewage sludge management options as a function of the local context.

Thirdly, it is indicated that best practices and management techniques may have an impact on the global warming potential, and to a smaller extent on the removal of chemical compounds from sludge. Circular sludge management options, including mono-incineration followed by acidulation to manufacture a sludge-derived P fertiliser and anaerobic digestion, can have a neutral to negative global warming impact when best management techniques (e.g. high incineration temperatures to reduce N_2O emissions from thermal processing, reducing leakages at digestion plants, sludge storage with coverage, minimising transport distances) are implemented. The application of good practices and best available techniques for sludge treatment may also help to further reduce pollutants from sludge (e.g. increased chemical removal when stricter time-temperature profiles are applied during biological and thermal sludge treatment).

Conclusions specific to the different topics in this study are outlined in the respective sections.

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10 List of abbreviations

AMR	Antimicrobial Resistance
AMRG	Antimicrobial Resistance Gene
AP	AlylPhenol
b.w.	body weight
C	Carbon
D5	Decamethylcyclopentasiloxane
DBT	DiButylTins
DEHP	Di(2-ethylhexyl)phthalate
DINP	Di-IsoNonylPhthalate
dl-PCB	dioxin-like PolyChlorinated Biphenyl
DM	Dry matter
ECD	Electron-Capture Detection
ECPA-LET	European Crop Protection Association (ECPA) - Local Environment Tool (LET)
EFSA	European Food Safety Authority
EHDPP	2-EthylHexyl Diphenyl Phosphate
EN	European Standard
EUSES	European Union System for the Evaluation of Substances
GAC	Granular Activitated Carbon
GC	Gas Chromatography
HBCDD	HexaBromoCycloDoSecane
HELCOM	Helsinki Commission
HPLC	High Performance Liquid Chromatography
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
JRC	Joint Research Centre
K_{oc}	Organic carbon-water partition coefficient
K_{ow}	Octanol-water partitioning coefficient
LCA	Life Cycle Assessment
LUCAS Soil	Land Use/Cover Area frame statistical Survey Soil
MCCP	mid-chain chlorinated paraffins
MS	Member Sates
N	Nitrogen
NP	NonylPhenol
OP	OctylPhenol
P	Phosphorus
PAC	Powdered Activitated Carbon
PAH	PolyAromatic Hydrocarbons
PBB	PolyBrominated Biphenyl
PBDE	PolyBrominated Diphenyl Ether
PCB	PolyChlorinated Biphenyl
PCDD	PolyChlorinated Dibenzo-p-Dioxins
PCDF	PolyChlorinated DibenzoFurans
PCN	Polychlorinated Naphthalenes
PCPP	Pharmaceuticals and Personal Care Products
PEC	Predicted Environmental Concentrations
PFAS	PerFluoroAlkyl Susbtances
PFHxS	PerFluoroHexane Sulfonic acid
PFNA	PerFluoronoNanoic Acid
PFOA	PerFluoroOctanoic Acid
PFOS	PerFluoroOctane Sulfonic acid
PNEC	Predicted No-Effect Concentration
POP	Persistent Organic Pollutant
QAC	Quaternary Ammonium Compound
RCR	Risk Characterisation Ratios
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals

SCCP	Short-chain chlorinated paraffins
SLHC	Safe Limit for Human Consumption
SSD	Sewage Sludge Directive (86/278/EEC)
TBT	TriButylTins
TEHP	Tris(2-EthylHexyl) Phosphate
TOC	Total Organic Carbon
TPT	TriPhenylTins
UFZ	Helmholtz Centre for Environmental Research
UWWTD	Urban Wastewater Treatment Directive (91/271/EEC)
UWWTP	Urban Wastewater Treatment Plant
WATSON	Dutch monitoring on micro-pollutants in influent and effluent from wastewater treatment plants

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Annexes

13 Supplementary information – screening risk assessment

13.1 Risk screening model calculations

13.1.1 Soil model

13.1.1.1 Initial concentration in soil after a single application

$$Csoil_{initial} = \frac{Application\ rate * 1000}{DEPTH_{soil} * RHO_{soil}\ x * 10\,000}$$

Equation 1

Explanation of symbols

DEPTHsoil	Mixing depth of soil	[m]	0.20
RHOsoil	Bulk density of wet soil	[kg m_{wwt}^{-3}]	1700
Application rate	Application rate for the pollutant	[g ha^{-1}]	Input
10 000	Area of 1 ha	[m^2]	
$Csoil_{initial}$	Initial concentration in soil	[mg kg_{wwt}^{-1}]	

13.1.1.2 Pollutant losses in soil

$$kbio_{soil} = \frac{\ln 2}{DT_{50}\,soil_{20°C} * exp^{(0.08*(20-12))}}$$

Equation 2

Explanation of symbols

$kbio_{soil}$	First order rate constant for biodegradation in bulk soil	[d^{-1}]
$DT_{50}soil_{20°C}$	Half-life for biodegradation of the pollutant in soil at 20°C	[d]

The inferred degradation at 20°C for surface water and soil is given in Table 6 and Table 7

Table 6: Inferred surface water degradation rates at 20°C reported in the LET on basis of biodegradability screening results and FOCUSsw guidance

Test result	DT_{50} (Half-life, days)
Readily biodegradable	7.91
Readably biodegradable, failing 10 day window	26.36
Inherently biodegradable	79.09
Not biodegradable	10000

Table 7: Inferred soil degradation rates at 20°C reported in the LET on basis of biodegradability screening results and FOCUSsw guidance

Kpsoil (L kg-1)	Readily biodegradable (DT_{50}, days)	Readably biodegradable, failing 10 day window (DT_{50}, days)	Inherently biodegradable (DT_{50}, days)	Not biodegradable (DT_{50}, days)
< 100	15.82	47.46	158.2	1000
> 100, < 1000	158.2	474.6	1000	1000
> 1000, < 10000	1000	1000	1000	1000

Kpsoil = Koc * fraction organic carbon in standard soil (0.02)

$$\frac{1}{k_{volat}} = \left(\frac{1}{kasl_{air} * {K_{air\ water}}/{K_{soil\ water}}} + \frac{1}{kasl_{soil}} \right) * DEPTH_{soil}$$

Equation 3

Explanation of symbols

$kasl_{air}$	Partial mass transfer coefficient at air-side of the air-soil interface	[m d^{-1}]	90.72
$kasl_{soil}$	Partial mass transfer coefficient at soil-side of the air-soil interface	[m d^{-1}]	See ECHA R.16 guidance (2016), Equation R.16-59
Kair-water	Air-water equilibrium distribution constant	[m^3 m^{-3}]	Equation 12
Ksoil-water	Soil-water partitioning coefficient	[m^3 m^{-3}]	Equation 10
DEPTHsoil	Mixing depth of soil	[m]	0.20
k_{volat}	Pseudo first-oder rate constant for volatilisation from soil	[d^{-1}]	

$$k_{leach} = \frac{F\ inf_{soil} * RAIN_{rate}}{K_{soil-water} * DEPTH_{soil}}$$

Equation 4

Explanation of symbols

F inf_{soil}	Fraction of rain water that infiltrates into soil	[-]	0.25
$RAIN_{rate}$	Rate of wet precipitation (700 mm $year^{-1}$)	[m d^{-1}]	1.925E-03
Ksoil-water	Soil-water partitioning coefficient	[m^3 m^{-3}]	Equation 10
DEPTHsoil	Mixing depth of soil	[m]	0.20
k_{leach}	Pseudo first-order rate constant for leaching from soil layer	[d^{-1}]	

$$k = k_{volat} + k_{leach} + kbio_{soil}$$ Equation 5

Explanation of symbols

k_{volat}	Pseudo first-oder rate constant for volatilisation from soil	[d^{-1}]	1/Equation 3
k_{leach}	Pseudo first-oder rate constant for leaching from soil layer	[d^{-1}]	Equation 4
$kbio_{soil}$	First order rate constant for biodegradation in bulk soil	[d^{-1}]	Equation 2
k	First order rate constant for removal from top soil	[d^{-1}]	

13.1.1.3 Maximum concentration in soil after multiple applications

$$Csoil_{max} = Csoil_{initial} * \frac{(1 - exp^{(-k*365)})^{Napp}}{(1 - exp^{(-k*365)})}$$ Equation 6

Explanation of symbols

$Csoil_{initial}$	Initial concentration in soil	[mg kg_{wwt}^{-1}]	Equation 1
k	First order rate constant for removal from top soil	[d^{-1}]	Equation 5
Napp	Number of application events	[-]	1, 10, or 100
$Csoil_{max}$	Maximum concentration in soil	[mg kg_{wwt}-1]	

13.1.1.4 Time-weighted average concentration in soil

$$TWAC_{soil}(t) = Csoil_{max} * \frac{(1 - exp^{(-k*t)})}{(k*t)}$$ Equation 7

Explanation of symbols

$Csoil_{max}$	Maximum concentration in soil	[mg kg_{wwt}^{-1}]	Equation 6
k	First order rate constant for removal from top soil	[d^{-1}]	Equation 5
t	Time period	[d]	Soil organisms: 30; human food intake 180(1)

$TWAC_{soil}(t)$	Time weighted average concentration in soil, over a period t	[mg kg_{wwt}-1]	

(1) In accordance with the EU-TGD (2003) and REACH R.16 guidance (2016), the time weighted average of 30 days has been considered, whereas for the modelling of pollutant transfer from soils to crops for human intake a 180 day period is used.

$$Clocal_{soil} = PEC_{soil} = TWAC_{soil}(30\ days)$$ Equation 8

$$Clocal_{crop\ transfer} = TWAC_{soil}(180\ days)$$ Equation 9

13.1.1.5 *Porewater concentration*

$$K_{soil-water} = Fair_{soil} * K_{air-water} + Fwater_{soil} + Fsolid_{soil} * \frac{Kp_{soil}}{1000} * RHO_{solid}$$ Equation 10

Where:

$$Kp_{soil} = K_{OC} * Foc_{solid}$$ Equation 11

$$K_{air-water} = \frac{HENRY}{R * TEMP}$$ Equation 12

$$HENRY = \frac{VP(TEMP_{env}) * MW}{SOL(TEMP_{env})}$$ Equation 13

Explanation of symbols

$VP(TEMP_{env})$	Vapour Pressure at environmental temperature (12°C)	[Pa]	Input
$SOL(TEMP_{env})$	Solubility in water at environmental temperature (12°C)	[mg L^{-1}]	Input
MW	Molecular weight	[g mol^{-1}]	Input
R	Gas constant	[Pa m^3 mol^{-1} K^{-1}]	8.314
TEMP	Temperature at the air-water interface	[K]	285
Koc	Partition coefficient organic carbon-water	[L kg^{-1}]	Input
Foc_{soil}	Fraction organic carbon in the soil	[-]	0.02
$Fair_{soil}$	Fraction air in soil	[-]	0.2
$Fwater_{soil}$	Fraction water in soil	[-]	0.2
$Fsolid_{soil}$	Fraction solid in soil	[-]	0.2
RHO_{solid}	Bulk density of solids	[kg m^{-3}]	2500
HENRY	Henry's law constant	[Pa m^3 mol^{-1}]	

$K_{air\text{-}water}$	Air-water partitioning coefficient	[-]
Kp_{soil}	Solids-water partition coefficient in soil	[L kg^{-1}]
$K_{soil\text{-}water}$	Soil-water partitioning coefficient	[mg m^{-3}]

The concentration in porewater is then calculated.

$$C_{liq} = \frac{Clocal_{crop\ transfer} * RHO_{soil}}{K_{soil-water} * 1000}$$ Equation 14

Explanation of symbols

$Clocal_{crop\ transfer}$	Time weighted average concentration in soil (180 days)	[mg kg_{wwt}^{-1}]	Equation 9
RHO_{soil}	Bulk density of solids	[kg m^{-3}]	1700
$K_{soil\text{-}water}$	Soil-water partitioning coefficient	[mg m^{-3}]	Equation 10
C_{liq}	Concentration in soil porewater	[mg L-1]	

13.1.2 Water model

13.1.2.1 Input into surface water via runoff/drainage/erosion

As in the FOCUS Step 2 model, the LET runoff/drainage/erosion event is driven by a rainfall event four days after the final application.

$$Eq\ Rate\ Runoff\ Event = Application\ rate * exp^{\left(-4 * \frac{\ln(2)}{DT50soil}\right)}$$ Equation 15

Explanation of symbols

Application rate	Pollutant application rate	[g ha-1]	Input
DT_{50}soil	Half-life of the pollutant in soil	[days]	Input (section 13.1.1.2)
Eq Rate Runoff Event	Equivalent rate for runoff at the time of the runoff event	[g ha-1]	

$$Input\ Runoff = \frac{Eq\ Rate\ Runoff\ Event * Runoff\% * FW\ Ratio}{1000}$$ Equation 16

Explanation of symbols

Eq Rate Runoff Event	Equivalent rate for runoff at the time of the runoff event	[g ha-1]	Equation 15

Runoff %	Runoff percentage	[%]	Default 5%
FW Ratio	Ratio of field to waterbody	[-]	10
Input Runoff	Input via runoff	[mg m^{-2}]	

Equation 16 calculates the amount of co-formulant that will be inputted into the waterbody via runoff/drainage/erosion following a rainfall event. However, the fraction of co-formulant entering the waterbody in the water phase and in the sediment phase will be dependent on the soil adsorption (KOC) of the substance. The fraction entering the waterbody in the water phase via runoff/drainage/erosion is calculated according to the soil adsorption (K_{oc}) of the substance.

$$Fwaterphase_{runoff} = \frac{Water\ Depth}{Water\ Depth + (Eff\ Sed\ Depth * RHOsed * OC * \frac{Koc}{100})}$$ Equation 17

Explanation of symbols

Water Depth	Depth of the surface water	[cm]	30
Eff Sed Depth	Effective sediment depth of the surface water	[cm]	1
RHOsed	Sediment bulk density	[kg L-1]	0.8
OC	Sediment organic carbon content	[%]	5
Koc	Partition coefficient organic carbon - water	[L kg-1]	User input
$Fwaterphase_{runoff}$	Fraction of substance entering in water phase via runoff	[-]	

The total loading to the waterbody entering in the water phase is calculated.

$$Input\ Runoff_{sw} = Input\ Runoff * Fwaterphase_{runoff}$$ Equation 18

Explanation of symbols

$Fwaterphase_{runoff}$	Fraction of substance entering in water phase via runoff	[-]	Equation 17
Input Runoff	Total input via runoff	[mg m-2]	Equation 16
$Input\ Runoff_{sw}$	Runoff input via water phase	[mg m-2]	

Finally, the maximum local concentration in surface water is calculated.

$$Cwater_{initial} = \frac{Input\ Runoff_{sw} * 100}{Water\ Depth}$$ Equation 19

Explanation of symbols

Input Runoff$_{sw}$	Runoff input via water phase	[mg m-2]	Equation 18
Water Depth	Depth of the surface water	[cm]	30
Cwater$_{initial}$	Local concentration in surface water after initial application	[µg L-1]	

To calculate the pollutant by fish in the surface water, it is assumed the diet of the aquatic predator is continuously exposed to the 180-day time weighted average following the runoff/erosion event at day 4 after sludge application. Both surface water residence time and (bio)degradation contribute to removal of the element/substance from surface water.

$$k_{ktotal} = \frac{\ln(2)}{DT50_{biowater}} + kres$$

Equation 20

Explanation of symbols

DT50$_{biowater}$	Half-life for biodegradation in water-sediment phase	[d-1]	Input
kres	First order rate constant for residence time in water body	[d^{-1}]	0.025 [default residence time of 40 days]
k$_{total}$	first order rate constant for removal from water phase	[d-1]	

$$Cwater_{max} = Cwater_{initial} * \frac{(1 - exp^{(-k_{total} * 365)^{Napp}}}{(1 - exp^{(-k_{total} * 365)})}$$

Equation 21

Explanation of symbols

Cwater$_{initial}$	Initial concentration in soil	[µg L-1]	Equation 19
k	First order rate constant for removal from top soil	[d^{-1}]	Equation 20
Napp	Number of application events	[-]	input
Cwater$_{max}$	Maximum concentration in soil	[µg L-1]	

$$TWAC_{water} = Cwater_{max} * \frac{(1 - exp^{(-k_{total} * t)})}{(k_{total} * t)}$$

Equation 22

Explanation of symbols

Cwater$_{max}$	Maximum concentration in soil	[µg L-1]	Equation 19
k$_{total}$	First order rate constant for removal from water phase	[d^{-1}]	Equation 20

t	Time period	[d]	human food intake 180
$TWAC_{water}$	Time weighted average concentration in surface water, over a period of 180 days	[µg L-1]	

13.1.3 Human via food intake

13.1.3.1 Fish concentration

The bioconcentration factor (BCF_{fish}) and the biomagnification factor (BMF) are used to estimate the concentration of a pollutant in the food (fish) of fish-eating predators, that in turn are assumed to be the human food source. When a measured BCF_{fish} is available (e.g. for some pollutants assessed in step 2 analysis), it is used directly. If experimental data are not available, the BCF_{fish} can be predicted from the relationship between Kow and BCF derived by Veith et al. (1979).
For substances with a log Kow in between 2 and 6:

$$\log BCF_{fish} = 0.85 * \log Kow - 0.7$$

Equation 23

For substances with a log Kow higher than 6:

$$\log BCF_{fish} = -0.20 * \log Kow^2 + 2.74 * \log Kow - 4.72$$

Equation 24

Explanation of symbols

Kow	Octanol-water partitioning coefficient	[-]	input
BCF_{fish}	Bioconcentration for fish on wet weight basis	[L $kg_{wet\ fish}$ -1]	

The biomagnification factor in fish (BMF) is determined from the measured BCF (if available) or Kow with default values summarised in

Table 8: Default BMF values for organic susbtances (ECHA R.16 guidance R.16.5.3.5)

Log Kow	BCF_{fish}	BMF
<4.5	<2000	1
4.5-5	2000 – 5000	2
5-8	> 5000	10
8-9	2000 – 5000	3
>9	< 2000	1

It is then assumed that 50% of the predator's diet comes from the local scale and 50% is assumed to come from the regional area.

$$Cfish = 0.5 * \left(TWAC_{water}(180\ days) + PEC_{regional}\right) * BCFfish * BMF \qquad \text{Equation 25}$$

Explanation of symbols			
$TWAC_{water}$	Time weighted average concentration in surface water, over a period of 180 days	[µg L-1]	Equation 22
$PEC_{regional}$	Regional background concentration in surface water	[µg L-1]	input
BCF_{fish}	Bioconcentration for fish on wet weight basis	[L $kg_{wet\ fish}$ -1]	
BMF	Biomagnification factor in fish		Table 8
C_{fish}	Concentration in fish	[µg kg_{wwt}-1]	

13.1.3.2 Biotransfer from soil to plants

The approach applied is described in the EU technical guidance document on risk assessment (ECHA, 2003). Plant products form a major part of the food products for humans and cattle. Contamination of plants will therefore have significant influence on the exposure of humans. When trying to predict concentrations in plant tissues, one will immediately encounter several important conceptual problems:

- there are hundreds of different plant species forming the heterogenous group of food crops.

Furthermore, varietal differences can also account for large differences;

- different tissues from plants are consumed (roots, tubers, fruit, leaves);
- crops differ in pollutant exposure, many crops are for instance grown in greenhouses;
- crops can be exposed through uptake from the soil, but also through gas uptake and aerial deposition.

From the above it may be clear that a modelling approach can only give a rough approximation of the concentrations in plants. To account for the predicted variety in plant products, it is proposed to distinguish between tuberous plants and leaf crops.

Uptake from soil is, in general, a passive process governed by the transpiration stream of the plant (in case of accumulation in leaves) or physical sorption (in case of roots). Uptake into the leaves from the gaseous phase can be viewed as a passive process, in which the leaves components (air, water, lipids) equilibrate with the air concentration. Kow and Kaw (the air-water partitioning coefficient) are used to assess the distribution between the air and the plant. It is proposed to use the modelling approach of Trapp and Matthies (1995) to estimate levels in leaves and roots due to uptake from soil and air.

It should be noted that the applied model is a simplified, generic representation of plant uptake. This model cannot give more than an indication for levels that may occur in plants in the field. Only the concentrations in leaf and root tissue are estimated, fruit is not accounted for. Several plant- specific parameters are required for the model. As there are many different plant species acting as food crops for humans and cattle, an average or typical plant definition is not scientifically feasible. Aerosol deposition is not considered in the model.

The partitioning between water and plant tissue is a key property for the fate of compounds in the soil-plant-air system. This partitioning is assumed to be based on hydrophobic sorption to plant lipids. The Kow is corrected slightly for the differences between plant lipids and octanol. The Kplant-water can than be calculated.

$$Kplant - water = Fwater_{plant} + Flipid_{plant} * Kow^{b} \qquad \text{Equation 26}$$

Explanation of symbols

F_{water}	Volume fraction water in plant tissue	[-]	0.65
F_{lipid}	Volume fraction lipids in plant tissue	[-]	0.01
Kow	Octanol-water partitioning coefficient	[-]	input
b	Correction for differences between plant lipid and octanol	[-]	0.95
$K_{plant\text{-}water}$	partition coefficient between plant tissue and water	[(mg.m plant-3)/(mg.m water-3)]	

When a measured biotransfer factor is available (e.g. for some pollutants assessed in step 2 analysis), it is used directly. Otherwise, concentration in root tissue is mainly governed by physical sorption, and is computed.

Equation 27

$$C_{root} = \frac{\mathrm{K}_{plant-water}}{\mathrm{RHO}_{plant}} * C_{liq} * 1000$$

Explanation of symbols

$K_{plant\text{-}water}$	partition coefficient between plant tissue and water	[(mg.m plant-3)/(mg.m water-3)]	Equation 26
C_{liq}	Concentration in soil porewater	[mg L-1]	Equation 14
RHO_{plant}	Bulk density of plant tissue	[kg m^{-3}]	700
C_{root}	Concentration in root tissue of plant	[µg kg_{wwt}-1]	

When a measured biotransfer factor is available (e.g. for some pollutants assessed in step 2 analysis), it is used directly. Otherwise, the concentration in leaf crops is computed.

Equation 28

$$C_{leaf} = \frac{0.0392 \exp^{-\frac{(\log \mathrm{Kow} - 1.78)^2}{2.44}}}{\dfrac{432}{0.0006 + \dfrac{0.0013 + 0.00002\, Kow^{0.95}}{Kaw}} + 0.035} * C_{liq} * 1000$$

Explanation of symbols

Kow	Octanol-water partitioning coefficient	[-]	input
Kaw	Air-water partitioning coefficient	[-]	Equation 14
C_{liq}	Concentration in soil porewater	[mg L-1]	Equation 14
C_{leaf}	Concentration in leaf tissue of plant	[µg kg_{wwt}-1]	

13.1.3.3 Drinking water

Drinking water can be prepared from surface water or from groundwater. Groundwater can be contaminated through leaching from the soil surface, surface water can be polluted through direct or indirect emission. The drinking water module in the present model assumes a complete removal of suspended particles from surface water.

$$C_{drw} = F_{pur} * TWAC_{water}(180\ days)$$ Equation 29

Explanation of symbols

F_{pur}	Purification factor for surface water	[-]	10 (default value)
$TWAC_{water(}$180 days)	Time weighted average concentration in surface water, over a period of 180 days	[µg L-1]	Equation 22
Cdrw	Concentration in drinking water (wet weight)	[mg L-1]	

13.1.3.4 Biotransfer to meat and milk

Lipophilic substances are known to accumulate in meat, and can be subsequently transferred to milk. Cattle can be exposed to substances in grass (or other feed) with adhering soil, drinking water, and through inhalation of air. When a measured Biotransfer factor is available (e.g. for some pollutants assessed in step 2 analysis), it is used directly. Biotransfer factors can be defined as the steady-state concentration in meat, divided by the daily intake of the substance. Travis and Arms (1988) calculated biotransfer factors for cow's meat and milk by log-linear regression on a number of substances (28 for milk and 36 for beef). Even though the theoretical background is limited, these factors provide a useful tool in risk assessment. It is proposed to use the same exposure estimates for air and crops which have been derived for human exposure for cattle, and the same soil concentration as for plants. It should be noted that no distinction is made between different milk products like cheese or yoghurt. For all dairy products, the concentration in milk is used.

The estimation can be used for substances with a log Kow range of 1.5 - 6.5. Outside this range, the maximum or minimum Kow value should be used.

$$BTF_{meat} = 10^{-7.6+\log Kow}$$ Equation 30

Explanation of symbols

Kow	Octanol-water partitioning coefficient	[-]	input
BTF_{meat}	Biotransfer factor for meat	[(mg kg-1)/(mg d-1)]	

The BTF for milk was derived on data of 28 organic compounds. It should be noted that the uncertainty in this estimation is considerable. The estimation can be used for substances with a log Kow range of 3 - 6.5. Outside this range, the maximum or minimum Kow value should be used.

$$BTF_{meat} = 10^{-8.1+\log Kow}$$ Equation 31

Explanation of symbols

Kow	Octanol-water partitioning coefficient	[-]	input
BTF$_{\text{milk}}$	Biotransfer factor for milk	[(mg kg-1)/(mg d-1)]	

The concentrations in meat and milk can be calculated by applying the biotransfer factors and summing the contributions from soil, grass, and drinking water (air contributions have been assumed to be absent).

$$Cmeat = BTF_{meat}\left(C_{grass} * IC_{grass} + TWAC_{soil}(180 \text{ days}) * IC_{soil} + C_{drw} * IC_{drw}\right)$$ Equation 32

$$Cmilk = BTF_{milk}\left(C_{grass} * IC_{grass} + TWAC_{soil}(180 \text{ days}) * IC_{soil} + C_{drw} * IC_{drw}\right)$$ Equation 33

Explanation of symbols

BTFmeat	Biotransfer factor for meat	[(mg kg-1)/(mg d-1)]	Equation 30
BTF$_{\text{milk}}$	Biotransfer for milk	[(mg kg-1)/(mg d-1)]	Equation 31
Cgrass	Concentration in grass (wet weight)	[mg kg$_{\text{wet weight}}$ -1]	Equation 28
ICgrass	Daily intake of grass	[kg d-1]	67.6
TWAC$_{\text{soil(}}$180 days)	Concentration in soil (wet weight)	[mg kg$_{\text{wet weight}}$ -1]	Equation 9
ICsoil	Daily intake of grass	[kg d-1]	0.46
Cdrw	Concentration in drinking water (wet weight)	[mg L-1]	Equation 29
ICdrw	Daily intake of drinking water	[L d-1]	55
Cmeat	concentration in meat (wet weight)	[mg kg$_{\text{wet weight}}$-1]	
Cmilk	concentration in milk(wet weight)	[mg kg$_{\text{wet weight}}$-1]	

13.1.3.5 Aggregated consumer intake

If concentrations in the intake media are calculated, the total daily intake of humans can be estimated from the daily intake rate of each medium by summing the contribution of each medium.

$$Human\ intake = (C_{fish} * I_{fish} + C_{root} * I_{root} + C_{leaf} * I_{leaf} + C_{meat} * I_{meat} + C_{milk} * I_{milk} + C_{drw} * I_{drw}) * 0.001$$ Equation 34

Explanation of symbols

C$_{\text{fish}}$	Concentration in fish	[µg kg$_{\text{wwt}}$-1]	Equation 25

I_{fish}	Daily human intake of fish	[kg_{wwt} d-1]	0.115
C_{root}	Concentration in root	[µg kg_{wwt}-1]	Equation 27
I_{root}	Daily human intake of root vegetables	[kg_{wwt} d-1]	0.384
C_{leaf}	Concentration in leaf	[µg kg_{wwt}-1]	Equation 28
I_{leaf}	Daily human intake of leaf vegetables	[kg_{wwt} d-1]	1.2
C_{meat}	concentration in meat (wet weight)	[µg kg_{wwt}-1]	Equation 32
I_{meat}	Daily human intake of meat	[kg_{wwt} d-1]	0.301
C_{milk}	Concentration in milk	[µg kg_{wwt}-1]	Equation 33
I_{milk}	Daily human intake of milk	[kg_{wwt} d-1]	0.561
C_{drw}	Concentration in drinking water	[mg L-1]	Equation 29
I_{drw}	Daily human intake of drinking water	[L d-1]	2
bw	Body weight adult	[mg d-1]	70
Human Intake	Total daily human intake	[mg kg-1]	

13.2 Model input data for results reported in main text

The model input data used to calculate the data presented in Figure 4 and Figure 5 (section 3.3.1.3) is presented in Table 9. The data used in this assessment originates from data repositories, and has not been verified by JRC on a case-by-case basis, introducing additional uncertainties to the outcomes presented (see section 1.2.2.1 on methodology).

Table 9: Input data used to calculate the data presented in Figure 4 and Figure 5 (section 3.3.1.3). (Kow = octanol –water partitioning coefficient; Koc = partition coefficient organic carbon – water, DT50 = estimated half-life from laboratory degradation tests; SLHC = Safe limit for human consumption)

CAS number	Substance name	concentration in sludge [mg kg-1 dw]	Water solubility [mg L-1]	Vapour Pressure [Pa]	Log Kow [-]	Koc [L kg1]	DT50 [days]	SLHC [mg kg-1 body weight day-1]
134523-00-5	Atorvastatin (PCPP)	8.5E-02	8.0E+00	4.6E-08	6.6	2.4E+05	8.6E+01	1.0E-03
657-24-9	Metformin (PCPP)	2.9E+00	6.9E+04	8.8E-08	-0.8	9.5E+00	4.8E+00	1.5E-03
22916-47-8	Miconazole (PCPP)	1.0E-01	1.1E-02	1.3E-05	6.1	4.8E+03	3.4E+00	3.0E-02
1222-05-5	Galaxolide (musk, PCPP)	1.1E+01	1.8E+00	6.2E-02	5.3	7.4E+04	2.5E+01	5.0E-02
15323-35-0	Phantolide (musk, PCPP)	1.0E+00	1.3E+00	2.2E-01	5.4	1.3E+04	5.3E+00	2.5E-02
1506-02-1	Tonalid (musk, PCPP)	1.0E+00	1.3E+00	5.6E-03	5.4	6.4E+04	NA	2.5E-02
84852-53-9	Decabromodiphenyl ethane (brom	1.0E-01	2.5E-04	3.2E-06	7.5	3.7E+05	4.4E+02	3.3E-01
139481-59-7	Candersartan (PCPP)	8.5E-01	1.4E-01	5.0E-07	4.5	7.2E+02	7.6E+00	3.3E-03
19466-47-8	Beta stigmastanol (sterol, PCPP)	3.1E+01	2.7E-02	8.8E-06	7.5	5.9E+04	1.4E+01	9.2E+00
5466-77-3	2-Ethylhexyl 4-methoxycinnamate	3.6E+00	0.0E+00	8.9E-04	5.2	9.2E+02	4.9E+00	2.3E+00
52645-53-1	Permethrin (PCPP/biocide)	3.3E-01	5.5E-03	2.1E-06	6.1	6.3E+04	3.4E+00	5.0E-02
101-20-2	Triclocarban (PCPP)	1.1E+01	7.4E-01	3.2E-07	3.2E+00	4.1E+03	5.6E+00	1.8E-02
8001-54-5	Benzalkonium chloride (quaternar	1.0E+02	2.0E+04	3.9E-08	2.71	39810.72	NA	0.1
15307-86-5	Diclofenac (PCPP)	2.0E-01	5.3E+00	1.99E-06	2.84666	5738.4	4.67826	0.0005
15687-27-1	Ibuprofen (PCPP)	5.5E-01	8.0E+01	5.6E-01	1.9E+00	1.4E+02	3.5E+00	2.5E-03
50-28-2	17b-Estradiol (sterol)	6.6E-02	1.3E+01	1.4E-06	2.6E+00	3.5E+05	9.7E+01	5.0E-05
82419-36-1	Ofloxacin (PCPP)	1.2E+00	3.8E+02	9.8E-06	-1.9E-01	4.8E+02	3.4E+00	1.7E-02
1071-83-6	Glyphosate (pesticide)	1.6E-02	1.4E+06	1.3E-05	-3.2E+00	2.1E+03	4.5E+00	1.0E-01
608-93-5	pentachlorobenzene (chlorobenze	5.0E-04	1.3E+00	4.4E-01	3.9E+00	3.2E+03	6.2E+00	8.0E-04
103-90-2	Acetaminophen (PCPP)	5.4E-01	6.0E+03	1.4E-03	2.4E-01	4.8E+01	3.6E+00	5.0E-02

The model input data used to calculate the data presented in Figure 9 (section 3.3.2) is presented in Table 11. The data used in this assessment originates from data repositories, and has not been verified by JRC on a case-by-case basis, introducing additional uncertainties to the outcomes presented (see section 1.2.2.1 on methodology).

Table 10: Input data used to calculate the data presented in Figure 6 (section 3.3.2) (Kow = octanol –water partitioning coefficient; Koc = partition coefficient organic carbon - water, DT50 = estimated half-life from laboratory degradation tests; SLHC = Safe limit for human consumption; BDE-209: decabromodiphenyl ether; BDE-47: 2,2',4,4'-Tetrabromodiphenyl ether; BDE-99: 2,2',4,4',5-Pentabromodiphenyl ether; HBCDD: hexabromocyclododecane); TEHP: tris(2-ethylhexyl) phosphate, EHDPP: 2-ethylhexyl diphenyl phosphate; TCPP: tris(1-chloro-2-propyl) phosphate; PCN 66/67: naphthalene congeners 667/67 (mixture of 1,2,3,4,6,7-HxCN and 1,2,3,5,6,7-HxCN); PCN-73: 73 (1,2,3,4,5,6,7-HpCN).; DEHP: bis(2-ethylhexyl)phthalate DINP: di-isononylphthalate; ∑SCCPs: sum of short-chain chlorinated paraffins; ∑MCCPs: sum of mid-chain chlorinated paraffins)

	concentration in sludge [mg kg-1 dw]	Water solubility [mg L-1]	Vapour Pressure [Pa]	Log Kow [-]	Koc [L kg1]	DT50 in soil [days]	SLHC [mg kg-1 body weight day-1]
Perfluorooctanoic acid (PFOA)	0.00378	9500	4.2	5.3	114.8154	527292.4	6.29E-07
Perfluorohexansulfonic acid (PFHxS)	0.000625	2300	58.9	5.17	3981.072	527292.4	6.29E-07
Perfluorononanoic acid (PFNA)	0.0026	464.08	5.1	2.4	245.4709	527292.4	6.29E-07
Perfluorooctane Sulfonate (PFOS)	0.04646	520	0.00033	6.43	6309.573	527292.4	1.57E-07
benzo(b)fluoranthene	1.919	0.0012	0.000067	6.12	1400000	1241	0.000034
chrysene	2.02	0.002	8.4E-07	5.73	200000	1241	0.000034
Benzo[a]pyrene	1.475	0.0038	7.3E-07	6.5	890000	1241	0.000034
Benzo[a]anthracene	1.832	0.014	0.000028	5.61	790000	1241	0.000034
2,3,7,8-TCDD	0.00001	0.0193	2E-07	6.8	3981072	3650	2.86E-10
BDE-209	0.5	0.062	5.9E-09	9.7	1400000	861	0.68
BDE-47	0.1	0.025	0.000063	6.57	980000	342	6.88E-05
BDE-99	0.1	0.25	0.000015	7.3	980000	342	1.68E-06
HBCDD	0.1	0.0086	3.3E-06	5.47	220000	63	9.79E-05
TEHP	1.2	5.1	0.00019	9.42	620000	4320	0.1
EHDPP	0.5	1	0.00034	5.73	9500	300	0.036
TCPP	1	1600	0.27	2.68	580	47520	0.01
4-nonylphenol	10	5.7	0.3	5.4	19.1	16.5	0.005
4-nonylphenol ethopxylates	1	1.05	0.000018	5.30103	2510	16.5	0.013
4-octylphenol	1	12.6	0.23	4.100371	10000	14	6.7E-08
SCCPs	100	0.33	0.021	6.7	336511.6	527292.4	0.0023
MCCPs	100	0.016	0.00027	7.2	855066.7	527292.4	0.036
Triphenyltin	0.1	5	0.8	3.716667	501187.2	527292.4	0.00025
Dibutyltin	0.3	20	0.16	1.56	41686.94	120	0.00025
Tributyltin	0.1	5.3	48.5	4.063333	31622.78	527292.4	0.00025
DEHP	50	0.003	0.000034	7.5	165000	147	0.05
DINP	30	0.0006	0.00006	8.8	310000	300	0.05
Decamethylcyclopentasiloxane (D5)	10.8	0.017	22.7	8.09	147910.8	12.6	0.25
PCN-65/66	0.00000004	0.00011	0.001	6.7	336511.6	3650	2.86E-10
PCN-73	0.000000068	0.00004	0.00026	6.6	279254.4	3650	2.86E-10

Table 11: Input data used to calculate the data presented in Figure 9 (section 3.3.2). (Kow = octanol –water partitioning coefficient; Koc = partition coefficient organic carbon - water, DT50 = estimated half-life from laboratory degradation tests)

Substance name	concentration in sludge [mg kg-1 dw]	Water solubility [mg L-1]	Vapour Pressure [Pa]	Log Kow [-]	Koc [L kg1]	DT50 in soil [days]	PNECsoil [mg kg-1]
Coronene	2.5E-01	1.0E-10	1.9E-10	7.5	1.0E+05	527292	4.6E-04
Perfluorononanoic acid (PFNA)	2.6E-03	464.08	5.1E+00	2.4	2.5E+02	527292	1.0E-05
Perfluorooctane Sulfonate (PFO	4.6E-02	5.2E+02	3.3E-04	6.4	6.3E+03	527292	2.2E-04
Benzo[ghi]perylene	3.5E-01	5.0E-04	3.7E-08	4.4	4.1E+04	527292	3.0E-03
Dibenzo[a,h]pyrene	2.3E-01	1.0E-10	5.6E-09	7.3	4.3E+05	527292	2.2E-03
2,3,7,8-TCDD	1.0E-05	1.9E-02	2.0E-07	6.8	4.0E+06	3650	2.2E-07
MCCPs	1.0E+02	1.6E-02	2.7E-04	7.2	8.6E+05	527292	1.2E+00
SCCPs	1.0E+02	3.3E-01	2.1E-02	6.7	3.4E+05	527292	1.2E+00
Dibenzo[a,l]pyrene	5.9E-02	1.0E-10	5.4E-09	7.5	1.9E+05	527292	9.5E-04
Oxipurinol	1.6E+02	1.0E-10	7.9E-07	-1.3	1.5E+02	16	1.6E-01
Fipronil sulfone	3.9E-02	5.4E-01	2.9E-07	2.5	1.7E+02	16	3.4E-05
Traseolide (ATII)	3.4E-01	1.0E-10	6.0E-03	5.6	5.4E+03	158	6.9E-04
Lauryl diethanolamide	3.4E+01	1.0E-10	1.9E-05	3.2	1.3E+03	16	2.3E-02
Ciprofloxacin	6.2E+00	7.5E+01	1.0E-07	0.1	3.1E+02	16	4.0E-03
N,N-Dimethyldodecylamine	1.2E+01	2.6E+00	5.6E-01	4.9	2.7E+03	16	5.3E-03
Ofloxacin	1.2E+00	3.8E+02	9.8E-06	-0.2	4.8E+02	16	5.1E-04
Permethrin	3.3E-01	5.5E-03	2.1E-06	6.1	6.3E+04	1582	4.5E-04
Triclocarban	1.1E+01	7.4E-01	3.2E-07	3.2	4.1E+03	16	1.3E-03
Beta-sitosterol	7.1E+01	5.8E-02	3.3E-06	4.6	5.9E+04	527292	1.0E-01
Campesterol	1.0E+02	1.0E-10	7.9E-06	8.9	9.6E+04	527292	9.5E-02
Candersartan	0.853290558	0.14	4.97E-07	4.5	719.679	16	3.96E-05
Benzalkonium chloride	1.0E+02	2.0E+04	3.9E-08	2.7	4.0E+04	527292	6.4E-02
Stigmasterol	3.2E+02	1.0E-10	2.5E-06	8.9	96996.8	527292	0.082168
Telmisartan	8.8E+00	9.0E-02	5.9E-09	4.7	1.3E+05	4746	1.2E-03
Cholesterol	9.0E+02	1.0E-10	9.9E-06	8.3	9.4E+04	527292	1.4E-01
Dipyridamol	5.9E+00	1.0E-10	6.3E-08	2.3	1.9E+02	16	1.8E-05

13.3 Results – Step 2 risk assessment

13.3.1 Perfluoroalkyl substances (PFAS)

13.3.1.1 Background and selection of compounds

Perfluoroalkyl substances (PFAS) are a group of widely used man-made organic chemical substances. PFAS are released into the environment from direct and indirect sources, for example, from professional and industrial facilities using PFAS, during use of consumer products (e.g. cosmetics, clothing or ski waxes) and from food contact materials.

PFAS contain alkyl groups on which all or many of the hydrogen atoms have been replaced with fluorine. Well known PFAS contain fully fluorinated carbon chains of various chain lengths attached to a functional group, like carboxylic or sulfonic acids.

The fluorine-carbon bonds are extremely stabile conferring these substances with very high chemical stability. Even if all releases of PFAS would cease tomorrow, they would continue to be present in the environment, and humans, for generations to come. Certain PFAS are known to accumulate in the bodies of living things and cause toxic effects. Certain PFAS are toxic for reproduction and can harm the development of foetuses. Several PFAS have been demonstrated to cause cancer. Some PFAS are also suspected of interfering with the human endocrine (hormonal) system (ECHA, 2021).

They can be divided into the groups of perfluorinated sulfonic acids (PFSAs), perfluorinated carboxylic acids (PFCAs), fluorotelomer alcohols, high-molecular weight fluoropolymers and low-molecular weight perfluoroalkanamides. Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), often referred to as reference or key substances for the first two groups, have been most intensively studied from a toxicological standpoint.

Since 2009, PFOS and its derivatives have been included in the international Stockholm Convention to eliminate their use. PFOS has already been restricted in the EU for more than 10 years, under the EU's Persistent Organic Pollutants Regulation. In addition, the Stockholm Convention regulates the global elimination of PFOA, its salts and PFOA-related compounds. PFOA has been banned under the POPs Regulation since July 2020. Perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds are being considered for inclusion in the Stockholm Convention and consequent global elimination.

13.3.1.2 Safe limit values for human consumption

Safe limit values for PFAS have recently (2020) been revised and further reinforced by EFSA. Compared to the previous assessment in 2012, tolerable daily intake values for PFAS have been reduced by approximately two to three orders of magnitude. Based on several similar effects in animals, toxicokinetics and observed levels in human blood, the EFSA Panel on Pollutants in the Food Chain (CONTAM Panel) performed a recent risk assessment for the sum of four PFASs: PFOA, perfluorononanoic acid (PFNA), PFHxS and PFOS ((EFSA CONTAM Panel) EFSA Panel on Contaminants in the Food Chain et al., 2020). Longer chain PFCAs (C9-C14) are not known to be intentionally used in the EU, though they can be present as impurities during the manufacture of other PFAS. The above-mentioned four PFAS made up approximately half of the lower bound exposure to those PFASs for which occurrence data were available, the remaining contribution being primarily from perfluorobutyrate and perfluorohexanoic acid, two PFAS with a short half-life. Since physico-chemical and toxicological data are available for these compounds, these 4 PFAS (hereafter $PFAS_4$: PFOA, PFNA, PFHxS and PFOS) have been selected for the screening risk assessment.

A tolerable weekly intake (TWI) of 4.4 ng/kg bw per week was established for the sum of PFOA, PFNA, PFHxS and PFOS ((EFSA CONTAM Panel) EFSA Panel on Contaminants in the Food Chain et al., 2020). Hence, a safe limit of value of 0.63 ng/kg bw per day has been used for $PFAS_4$ in this assessment

13.3.1.3 Concentrations observed in sludge

The median PFOA (3.8 μg kg^{-1} DM), PFNA (2.6 μg kg^{-1} DM) and PFOS (46.5 μg kg^{-1} DM) concentrations reported by Tavazzi et al. (2012) have been retained in the assessment. These values fall within the ranges of studies of other studies (Gómez-Canela et al., 2012; Brambilla et al., 2016; Blytt and Stang, 2018; Fernandes et al., 2019; Kärrman et al., 2019; Rigby et al., 2021). The concentration value for PFHxS (0.62 μg kg^{-1} DM) was taken from Gómez-Canela et al. (2012).

13.3.1.4 Model input values

Table 12: Physico-chemical and toxicological data used for risk modelling purposes for the selected perfluoroalkyl substances.

	PFOA	**PFHxS**	**PFNA**	**PFOS**
Concentration in sludge (mg^{-1} kg^{-1} dry matter)$^{(1)}$	6.7e-3 (Tavazzi et al., 2012)	1.4e-3 (Gómez-Canela et al., 2012)	4.6e-3 (Tavazzi et al., 2012)	66.4e-3 (Tavazzi et al., 2012)
Molecular weight (g mol^{-1})	414.07 (ECHA, 2013)	400.11 (ECHA, 2017a)	464.08 (core database)	500.13 (core database)

Water solubility (mg L^{-1})	9500	2300	0.88	520
	(ECHA, 2013)	(ECHA, 2017a)	(core database)	(ECHA, 2017a)
Vapour pressure (Pa)	4.2	58.9	5.1	3.3e-4
	(ECHA, 2013)	(ECHA, 2017a)	(core database)	(ECHA, 2017a)
Kow (log 10)	5.3	5.17	2.4	6.43
	(ECHA, 2013)	(ECHA, 2017a)	(ECHA, 2017a)	(ECHA, 2017a)
Koc (L kg^{-1})	114.8	3981	245	6310
	(ECHA, 2017a)	(ECHA, 2017a)	(ECHA, 2017a)	(ECHA, 2017a)
PNECaqua (µg L^{-1})	0.1	0.87	0.58	0.002
	(Ankley et al., 2021)	(core database)	(core database)	
PNECsoil (mg kg^{-1} ww)	1.13e-2	1.13e-2	1.13e-2	2.61e-3
	(based on NOEC of 1 mg kg-1 wet soil (Ankley et al., 2021), assessment factor of 100)	(based on NOEC of 1 mg kg-1 wet soil (Ankley et al., 2021), assessment factor of 100)	(based on NOEC of 1 mg kg-1 wet soil (Ankley et al., 2021), assessment factor of 100)	(core database)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	0.63e-6 for the sum of PFOA, PFHxS, PFNA, and PFOS			
	(EFSA Panel on Pollutants in the Food Chain et al., 2020)			
Half-life in soil (days)	not biodegradable	not biodegradable	not biodegradable	not biodegradable
	(ECHA, 2017a)			
Half-life in water (degradation, days)	not biodegradable	not biodegradable	not biodegradable	not biodegradable
	(ECHA, 2017a)			
Regional background in soil (mg kg^{-1} dry matter)$^{(2)}$	0.86e-3	0	0.62e-3	1.2e-3
	(Göckener et al., 2021)			
Regional background in surface water (µg L^{-1})	6.5e-2	5.3e-4	1.45e-3	1.4e-2
	(EMPODAT, 2021)			
Atmospheric dry deposition (mg m^{-2} d^{-1})	1.55	1.66	0.34	7.1
	(Johansson et al., 2018)			
Atmospheric wet deposition (ng L^{-1})	0.57	0.79	0.79	0.59
	(Johansson et al., 2018)			
BCFfish-water (L kg^{-1}ww)	87	257	4570	1862
	(Burkhard, 2021)			

BCFshoot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)$^{(3)}$	2.02E-02	2.74E-01	9.69E-03	2.02E-02
	(Yoo et al., 2011)			
BCFroot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)$^{(3)}$	2.24E-01	2.53E-01	1.79E-01	1.11E-01
	(Wen et al., 2014)			
BTFmeat-grass (day kg-1 dry matter)	0.012	0.041	0.302	0.071
	(Kowalczyk et al., 2013; Vestergren et al., 2013)			
BTF-milk-grass (day kg^{-1} dry matter)	0.01122	0.0007	0.0155	0.0213
	(Vestergren et al., 2013; Takaki et al., 2015)			

(1)The PFOA, PFNA and PFOS concentrations reported by Tavazzi et al. (2012) have been retained in the assessment. These values fall within the ranges of studies of other studies (Gómez-Canela et al., 2012; Brambilla et al., 2016; Blytt and Stang, 2018; Fernandes et al., 2019; Kärrman et al., 2019; Rigby et al., 2021). The concentration value for PFHxS (0.62 µg^{-1} kg^{-1} DM) was taken from Gómez-Canela et al. (2012).

(2)Reported values for regional background concentrations in soils were taken from ten sites across Germany (Göckener et al., 2021), and are thus representative for industrialised countries. These values are, however, one order of magnitude lower compared to global mean values reported by Brusseau et al. (2020).

(3)Values indicated on a dry matter basis have been transformed into values on a wet mass basis (mg kg-1 plant ww)/(mg kg-1 soil ww) assuming a water content in soil of 20vol% (EUSES default, conversion factor for soil concentration wet-dry weight soil of 1.13), and a water content in roots of 65vol%, and a bulk density of plants of 700 kg/m3 (EUSES default),

13.3.1.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of PFAS present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that risk characterisation ratios were above 1 for soils (PFOS, after 10 and 100 years of sludge application) and human health (all time periods) (Table 13). The long-chain PFNA and PFOS contributed most to the total $\Sigma PFAS_4$ uptake, with values that were 2-3 orders of magnitude greater than the contributions of PFOA and PFHxS. The RCR were not exceeded to for surface waters, although the value was only marginally below 1 for PFOS (Table 13).

It was indicated that the fish consumption is the primary driver human health risks, with an average contribution of fish total human intake of 74% after 10 years) (Table 13). In turn, the PFAS concentrations in surface water are the primary driver for PFAS uptake by fish.

Table 13: Environmental and human health risk screening assessment of the exclusive application of PFAS present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		PFOA	**PFHxS**	**PFNA**	**PFOS**	**$\Sigma PFAS_4$**
Soil	PEC$_{year\ 1/10/100}$ (mg kg^{-1} wet soil)	5.5E-06 / 1.5E-05 / 1.9E-05	9.2E-07/ 9.3E-06/ 4.6E-05	3.8E-06/ 1.5E-05/ 1.9E-05	6.8E-05/ 7.3E-04/ 5.5E-03	
	RCR$_{year\ 1/10/100}$ (-)	2.5E-02/ 7.0E-02/ 8.6E-02	1.5E-05/ 1.5E-04/ 7.5E-04	1.4E-03/ 5.8E-03/ 7.3E-03	3.1E-01/ 3.3E+00/ 2.5E+01	
Surface water	PEC$_{year\ 1}$ (µg L^{-1})	1.4E-03	4.1E-05	8.2E-04	2.1E-03	
	RCR $_{year\ 1}$ (-)	1.37E-02	4.7E-05	1.4E-03	1.0E+00	
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	8.6E-06/ 9.7E-06/	9.4E-07/ 6.5E-06/	1.9E-04/ 1.9E-04/	1.1E-04/ 1.9E-04/	3.1E-04/ 4.0E-04/

	1.0E-05	3.1E-05	1.9E-04	7.9E-04	1.0E-03
Relative contribution of fish consumption$_{\text{10 years}}$ (%)	77	4	99	51	74
Relative contribution of crop, meat and dairy consumption$_{\text{10 years}}$ (%)	22	96	1	49	26
Relative contribution of drinking water$_{\text{10 years}}$ (%)	1	0	0	0	0
RCR$_{\text{year 1/10/100}}$					2.8E+01/ 3.6E+01/ 9.3E+01

13.3.1.6 Source contribution analysis

Risks from PFAS are further augmented when regional background concentrations (surface water, soil) and inputs from atmospheric deposition are considered, with a further increase in calculated risk ratios for soils (Table 14). Risk characterisation ratios for PFOS are now also exceeded in surface water bodies. Given the assumed residence time of 40 days in water bodies and single application per year, the PEC in surface water remains largely constant in the long-term. Risks for human health are also further augmented (Table 14) due to increases in PFAS concentrations in crops, dairy products and meat as a result of PFAS soil concentrations.

Sludge-derived PFAS are a relatively small source of contamination relative to existing background levels observed in soils for PFOA and PFNA (maximum 3% after 100 years) (Table 14). For PFOS, the sludge-derived contribution to total soil contamination is more significant, especially in the mid- (10 year, 38%) to long-term (100 year, 82%). For PFHxS, background concentrations in soils have been found to be negligible and sludge is considered to be the sole source of contamination in this study (Table 14).

The PFAS concentrations in water are the sum of local inputs from the sewage sludge amended agricultural field (through runoff, erosion and drainage) and regional background concentrations observed in surface waters (e.g. effluents from wastewater treatment plants, industrial effluents). Sludge-derived PFAS are a rather small source of surface water contamination, with estimated contributions to surface water varying from 3% to 17% for PFOA, PFHxS and PFOS, in the 30-day period following application on agricultural land (Table 14). The contribution of PFNA is somewhat higher, but still not a dominant source (44%) (Table 14). These findings are generally in line with literature observations. Arvaniti et al. (2014) indicated that no more than 48% (PFOS) and 11% (PFOA) in wastewaters end up in sludge faction (mix of primary and secondary sludge, either digested or undigested). These values are expected to be lower for short-chain PFAS that are mainly transferred to the effluent during wastewater treatment (Arvaniti et al., 2014). After land application the overall share (>95%) of the PFAS remain in soils (Costello and Lee, 2020; confirmed by our modelling assessment). These observations corroborate the relatively low contribution of sludge-derived PFAS to surface waters, as well as the conservative nature of our risk assessment methodology (local scale assessment, assessing risks in surface waters surrounding an agricultural fields amended with sewage sludge at a high application rate) at a more regional scale.

In sum, our findings indicates that sludge may be a relevant, but not the principal, source of PFAS contamination for surface waters. Whereas sludge may be a source of PFAS contamination of surface water and induce as such human health risks, other sources including wastewater treatment effluents, need to be tackled simultaneously.

Table 14: Environmental and human health risk screening assessment for PFAS, based on its present background concentrations observed in the environment along with sewage sludge applications to agricultural land (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		PFOA	PFHxS	PFNA	PFOS	$\Sigma PFAS_4$
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	8.6E-04 / 8.7E-04 / 8.8E-04	9.2E-07 / 9.3E-06 / 4.6E-05	6.2E-04 / 6.4E-04 / 6.4E-04	1.3E-03/ 1.9E-03/ 6.7E-03	
	Contribution of sludge to total concentration (%)	1/2/2	100/100/100	1/2/3	5/38/82	
	RCR$_{\text{year 1/10/100}}$ (-)	4.0E+00/ 4.1E+00/ 4.1E+00	1.5E-05/ 1.5E-04/ 7.5E-04	2.4E-01/ 2.4E-01/ 2.5E-01	5.7E+00/ 8.7E+00/ 3.0E+01	

Surface water	PEC$_{year\ 1}$ (µg L^{-1})	3.4E-02	3.1E-04	1.5E-03	9.1E-03	
	Contribution of sludge to total concentration (%)	3	10	44	17	
	RCR $_{year\ 1}$ (-)	3.4E-01	3.5E-04	2.7E-03	4.5E+00	
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	9.7E-04/ 9.8E-04/ 9.8E-04	9.2E-06/ 1.5E-05/ 3.9E-05	1.0E-03/ 1.0E-03/ 1.0E-03	1.8E-03/ 1.9E-03/ 2.5E-03	3.8E-03/ 3.9E-03/ 4.5E-03
	RCR$_{year\ 1/10/100}$					3.4E+02/ 3.5E+02/ 4.1E+02

13.3.1.7 Effect of sludge treatment

PFAS are known as 'forever chemicals' due to their persistence in the environment. They are resistant to biotic degradation processes, including composting and anaerobic digestion (Lindstrom et al., 2011; Lakshminarasimman et al., 2021). Thermal oxidation processes can be applied to incinerate sewage sludge, after which the fly ashes can be used as a rock phosphate substitute to develop a nutrient-dense P fertiliser. In the EU, especially fluidised bed incinerators are commonly used for this purpose. Incineration, mainly fluidised bed incineration characterised by extreme turbulence and long-residence times of the gas phase, at least remove an important share of the PFAS (see recent review of Winchell et al. (2021), and studies cited herein). However, complete destruction of PFAS in thermal processes may require temperatures at 1000°C-1100°C (Winchell et al., 2021). Nonetheless, Khan et al. (2020) calculated a PFOS half-life of 0.2 s at 726°C. MacGregor (2020) reported PFAS is being destroyed through a fluidised-bed sewage sludge mono-incinerator (830°C for 8 s). Wohlin and Karmann (2020) indicated that PFAS concentrations in fly ashes are lower than for bottom-ashes. Hence, uncertainty gaps exist related to the removal of PFAS during incineration (Winchell et al., 2021) that need to be filled. This holds especially true in view of the revised 2020 EFSA guidelines pointing towards human health risks at extremely low levels of PFAS intake. Removal of PFAS (95-99%) during pyrolysis has also recently been observed (Buss, 2021), although the knowledge base and minimum operational conditions for pyrolysis processes remains thin.

13.3.2 Polyaromatic hydrocarbons (PAH)

13.3.2.1 Background and selection of compounds

PAHs are a natural component of coal and oil, which have historically been used in wood preservatives and tar products. They are mainly formed by incomplete combustion of organic material, such as coal, petrol and wood, and are commonly released into the atmosphere as small particulates (Abdel-Shafy and Mansour, 2016). Sources of PAHs into the European environment include the production and processing of metals, vehicle exhausts, coal-fired power generation, domestic heating and forest fires. Atmospheric emissions have been reduced in Europe since the 1980s.

The PAH substance group comprises a large number of substances with different toxicities and environmental fates. Environmental quality standards s have been set for seven of the most toxic PAHs, which act as representatives of the whole group. Three of these are separately listed (anthracene, fluoranthene and naphthalene) while the other five are grouped, with the 'lead substance' being benzo(a)pyrene.

PAHs are extensively metabolised in mammals and but not bioaccumulate. Most PAHs that enter the body leave within a few days, primarily in the feces and urine. Different metabolic pathways can lead to highly reactive intermediates involved in the mutagenic/carcinogenic process of PAHs.

13.3.2.2 Safe limit values for human consumption

Based on the currently available data relating to occurrence and toxicity, the CONTAM Panel concluded that the sum of the high molecular weight PAH benzo[a]pyrene (BaP), chrysene (CHR), benz[a]anthracene (BaA) and benzo[b]fluoranthene (BbFA) (PAH$_4$) is the most suitable indicators of PAHs in food (EFSA, 2008). These PAH, with 4 to 5 aromatic rings in their structure, have been indicated belong to the class of PAH compounds that show the highest persistency in soils, water and sediment (ECHA, 2008a). Together these PAH$_4$ make up 32%

out of total PAH mass (19 PAH compounds) identified and measured in the JRC report on sewage sludge (Tavazzi et al., 2012).

The CONTAM Panel calculated BMDL10 values for PAH_4 using a range of statistical models, and selected the lowest benchmark dose (lower confidence limit with a benchmark response of 10%, $BMDL_{10}$) of 0.34 mg kg^{-1} body weight day^{-1} from the statistical models that adequately fit the data (EFSA, 2008). A minimum Margin of Exposure (MOE) of 10,000 was indicated by the EFSA Scientific Committee to ensure the absence of a potential concern for consumer health and a possible need for risk management action (EFSA, 2008). This MOE was based based on the bench mark dose lower confidence limit for a 10% increase in the number of tumour bearing animals compared to control animals. Therefore, a conservative value of 3.4E-05 mg kg^{-1} body weight day^{-1} is applied in this assessment to flag a potential concern for human health.

13.3.2.3 Concentrations observed in sludge

The PAH concentrations reported in Tavazzi et al. (2012) were used for modelling. Reported PAH concentrations in more recent studies are generally in line or slightly lower compared to these values (Torretta and Katsoyiannis, 2013; Suciu et al., 2015; Poluszyńska et al., 2017; Rorat et al., 2019; Wluka et al., 2021).

13.3.2.4 Model input data

Table 15: Physico-chemical and toxicological data used for risk modelling purposes for the selected polyaromatic hydrocarbons.

	Benzo(b)fluoranthene	**Chrysene**	**Benzo(a)pyrene**	**Benzo(a)anthracene**
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	1.92E+00	2.02E+00	1.48E+00	1.83E+00
	(Tavazzi et al., 2012)			
Molecular weight (g mol-1)	252.32	228.3	252.32	228.3
	(core database)			
Water solubility (mg L^{-1})	0.0012	0.002	0.0038	0.014
	(WHO, 1998a)	(ECHA, 2017b)	(WHO, 1998a)	(WHO, 1998a)
Vapour pressure (Pa)	6.7E-05	8.4E-07	7.3E-07	2.8E-05
	(WHO, 1998a)	(ECHA, 2017b)	(WHO, 1998a)	(WHO, 1998a)
Kow (log 10)	6.12	5.73	6.50	5.61
	(WHO, 1998a)	(ECHA, 2017b)	(WHO, 1998a)	(WHO, 1998a)
Koc (L kg^{-1})	1.4E+06	2.0E+05	8.9E+05	7.9E+05
	(core database)	(core database)	(WHO, 1998a)	(WHO, 1998a)
PNECaqua (µg L^{-1})	3.5E-02	3.0E-02	7.0E-02	1.3E-02
	(core database)			
PNECsoil (mg kg^{-1} ww)	2.56E-01	1.08E-01	1.10E+00	4.59E-02
	(equilibrium partitioning method, based on PNECaqua)(1)			
Safe limit values for human intake (mg kg^{-1}	0.34 e-4 mg kg-1 body weight day-1 for the sum of 4 PAH (benzo(a)pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene)			

body weight day^{-1})	(EFSA, 2008)			
Half-life in soil (days)	1241 (1)			
Half-life in water (degradation, days)	527 292 (2)			
Regional background in soil (mg kg^{-1} dry matter)$^{(2)}$	0.106	0.077	0.089	0.056
	(UK Environment Agency, 2007; average values of documented for the the UK and Norway, assumed to be representative for the EU; Nam et al., 2008)			
Regional background in surface water (µg L^{-1})	Not applied$^{(3)}$			

(1)PAH degrade slowly in soil with half-lives that vary from about 100 days to a couple of years (Smith et al., 1997; WHO, 1998a; Roslund et al., 2018). In this study, the maximum values of the ranges (1.15 – 3.4 years) documented by the UK Environment Agency (UK Environment Agency, 2007) were applied

(2)PAH degrade slowly in surface waters, especially at greater depth where photolysis is absent (Tansel et al., 2011)(ECHA, 2017b). Conservative default half-life values (263 646 days) for non-biodegradable substances (527 292 days) have been used in the assessment. In any event, the main removal pathway from surface would be dilution with inflowing waters (surface water residence time: 40 days)

(3)Background concentrations in surface waters for PAH have sparsely been documented. No data are available in the JRC IPCHEM database. In the NORMAN EMPODAT (EMPODAT, 2021), reported data vary from below detection limit (majority of the samples) to two digit values expressed as µg L^{-1}. These observations indicate that background concentrations are highly variable as a function of location, neighbouring industrial activities, and point-source releases. Therefore, no assessment could be made to assess the impact of regional background concentrations in surface water.

13.3.2.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of PAH present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that risk characterisation ratios below 1 for soils and surface waters. For human health, RCRs above 1 were indicated for the entire time period under study, up to a value of 6.1E+01 indicating significant risks in the long-term (100 years) (Table 16). In the long-term, the contribution of BaP, CHR, and BbFa was on average one order of magnitude greater than the contribution to human risks for BaA (Table 16).

Based on a weighted average of different food sources to total PAH human uptake, it was indicated that crop, meat and dairy were the main sources of PAH uptake (82%), with lower contributions from fish (18%) and drinking water (0.2%). This can be explained by the observed persistence and reduced mobility and strong adsorption of PAH in soils (Smith et al., 1997), causing rather small PAH losses to surface waters.

Table 16: Environmental and human health risk screening assessment of the exclusive application of PAH present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio; BaP: benzo[a]pyrene, CHR: chrysene, BaA: benz[a]anthracene, BbFA: benzo[b]fluoranthene).

		BbFA	**CHR**	**BaP**	**BaA**	**ΣPAH₄**
Soil	PEC$_{year\ 1/10/100}$ (mg kg^{-1} wet soil)	2.8E-03	3.0E-03/	2.2E-03/	2.7E-03/	
		1.9E-02	2.0E-02/	1.5E-02/	1.8E-02/	
		2.8E-02	3.2E-02	2.3E-02	2.9E-02	
	RCR$_{year\ 1/10/100}$ (-)	3.2E-03/	2.8E-02/	2.0E-03/	1.5E-02/	
		2.1E-02/	1.9E-01/	1.3E-02/	1.0E-01/	
		3.3E-02	3.0E-01	2.1E-02	1.6E-01	

Surface water	PEC$_{\text{year 1}}$ (µg L^{-1})	4.3E-04	3.1E-03	5.2E-04	7.2E-04	
	RCR $_{\text{year 1}}$ (-)	1.2E-02	1.0E-01	7.4E-03	5.6E-02	
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	4.1E-03/ 9.0E-03/ 9.8E-03	1.2E-02/ 1.2E-02/ 1.2E-02	8.6E-03/ 1.1E-01/ 1.2E-01	2.1E-03/ 2.2E-03/ 2.3E-03	2.7E-02/ 1.3E-01/ 1.5E-01
	Relative contribution of fish consumption$_{\text{10 years}}$ (%)					18
	Relative contribution of crop, meat and dairy consumption$_{\text{10 years}}$ (%)					82
	Relative contribution of drinking water$_{\text{10 years}}$ (%)					0.2
	RCR$_{\text{year 1/10/100}}$					1.1E+01/ 5.6E+01/ 6.1E+01

13.3.2.6 Source contribution analysis

Risks from PAH are further augmented when regional background concentrations (surface water, soil) and inputs from atmospheric deposition are considered, with a further increase in calculated risk ratios for soils (Table 17). Risk characterisation ratios for chrysene now exceeds the critical ratio of 1 in soils after a period of 100 years. Risks for human health are also further augmented due to increases in PFAS concentrations in crops, dairy products and meat as a result of PFAS soil concentrations. Risk characterisation ratios for surface water remain below estimated limit values that may be of concern to aquatic organisms.

Sludge-derived PAH become a significant contributor to total PAH concentrations in the soil in the mid- to long-term (14% - 34% across all PFAS following 10-100 years of continuous sludge applications) (Table 17). It should be remarked that these simulations are likely overestimated as future inputs from other sources have not been considered in our assessment.

In sum, our findings indicates that sludge may be a relevant source of PAH contamination in soils, that may cause human health risks based on our conservative modelling approaches.

Table 17: Environmental and human health risk screening assessment for PAH, based on its present background concentrations observed in the soil along with sewage sludge applications to agricultural land (PEC: predicted environmental concentration, RCR: risk characterisation ratio; BaP: benzo[a]pyrene, CHR: chrysene, BaA: benz[a]anthracene, BbFA: benzo[b]fluoranthene). Note: due to the absence of EU-representative background PAH values in surface waters, no source contribution analysis could be done for this ecosystem compartment.

		BbFA	**CHR**	**BaP**	**BaA**	**ΣPAH$_4$**
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	1.1E-01/ 1.2E-01/ 1.3E-01	8.0E-02/ 9.7E-02/ 1.1E-01	9.1E-02/ 1.0E-01/ 1.1E-01	5.9E-02/ 7.4E-02/ 8.5E-02	
	Contribution of sludge to total concentration (%)	3/ 15/ 21	4/ 21 /29	2/ 14/ 21	5/ 25/ 34	
	RCR$_{\text{year 1/10/100}}$ (-)	1.3E-01/ 1.4E-01/ 1.6E-01	7.6E-01/ 9.2E-01/ 1.0E+00	8.3E-02/ 9.4E-02/ 1.0E-01	3.2E-01/ 4.1E-01/ 4.7E-01	
Surface water	PEC$_{\text{year 1/10/100}}$ (µg/L)	4.3E-04	3.1E-03	5.2E-04	7.2E-04	
	RCR$_{\text{year 1/10/100}}$ (-)	1.2E-02	1.0E-01	7.4E-03	5.6E-02	
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	1.3E-02/ 1.4E-02/ 1.5E-02	1.3E-02/ 1.3E-02/ 1.3E-02	1.3E-01/ 1.5E-01/ 1.6E-01	2.3E-03/ 2.4E-03/ 2.4E-03	1.6E-01/ 1.8E-01/ 1.9E-01
	RCR$_{\text{year 1/10/100}}$					6.8E+01/

	7.5E+01/ 8.1E+01

13.3.2.7 *Effect of sludge treatment*

Composting has potential to reduce PAH concentrations in sewage sludge (Poluszyńska et al., 2017), with removal rates of 64% to 83% for PAH with 4 to 5 aromatic rings, such as as $PFAS_4$. A recent review article on composting compiled information of about 15 studies for different organic materials, and indicated similar removal rates varying from 47% to 100% removal (Lü et al., 2021). The partial degradation of PAH during mesophilic and thermophilic anaerobic digestion of sludge has also observed (Chang et al., 2003; Trably et al., 2003; Benabdallah El-Hadj et al., 2006; Bernal-Martinez et al., 2009; Zhang et al., 2019). El-Hadj et al. 2006 reported removal efficiencies ranging from 51% to 65% in thermophilic conditions and from 36 to 43% in mesophilic conditions for the sum of 16 PAH. Moreover, according to Trably et al. 2003 the removal rate of the sum of 13 PAH (about 50%) was directly linked to total solids removal rate. It is concluded that, on average, biological treatments of sewage sludge are able to reduce PAH concentrations by a factor 2 to 10, but that the removal efficiency is dependent on process conditions applied and PAH compounds under study.

Incineration of at high temperatures ranging from 870°C to 1200°C effectively destructs PAHs, with removal efficiencies in the 99.0 - 99.9% range (Renoldi et al., 2003; Gan et al., 2009; Van Caneghem and Vandecasteele, 2014). In the boiler and fly ash of industrial facilities, PAHs were not present in concentrations above the detection limit (<0.01 mg kg^{-1} ash) following sewage sludge incineration. This indicates that the ash does not contain PAHs remaining from the waste nor PAHs adsorbed from the gas phase under well-operated conditions that comply with the European Directive on Industrial Emissions (Directive 2010/75/EU) (Van Caneghem and Vandecasteele, 2014). The absence of PAH in incinerator fly ash from waste materials other than sewage sludge was also observed by Wheatley and Sadhra (2004). PAH concentrations in bottom ash above the detection limit (<0.3 mg kg^{-1} ash) have been observed (Van Caneghem and Vandecasteele, 2014), but this fraction is generally not used for the manufacturing of P-fertilisers from ah through acidulation processes. Incineration has the potential to achieve high PAH removal efficiencies, but it is critical that the process is well-controlled so as to avoid contamination of the fly ashes by *de novo* produced PAH during the incineration process (Huygens et al., 2019).

13.3.3 PCDD/Fs and dioxin-like PCBs

13.3.3.1 *Background and selection of compounds*

Polychlorinated dibenzo-p-dioxin and furans (PCDD/Fs) are two groups of tricyclic planar compounds. Dependent on the number of chlorine atoms and their positions at the rings, 75 PCDDs and 135 PCDFs, termed ‘congeners’, can occur (EFSA Panel on Pollutants in the Food Chain et al., 2018). Only 17 of these are relatively persistent in animals and humans and therefore considered relevant. They contain at least four chlorines and at positions 2, 3, 7 and 8. Dioxin-like polychlorinated biphenyls (dl-PCBs) congeners that are non-ortho or mono-ortho chlorine substituted and contain at least four chlorine substituents can easily adopt a coplanar structure and show toxicological properties similar to tetrachlorodibenzo-p-dioxin (TCDD) and other PCDD/Fs (EFSA Panel on Pollutants in the Food Chain et al., 2018).

PCDD/Fs have never been produced on an industrial scale and have no technological use. They are formed unintentionally in a number of industrial and thermal processes. In contrast to PCDD/Fs, PCBs had widespread use in open and closed systems, generally in the form of complex technical mixtures. They were produced with an estimated total world production of 1.2–1.5 million tonnes between 1929 and the end of the 1970s, when their production was abandoned in the majority of countries (EFSA Panel on Pollutants in the Food Chain et al., 2018). Atmospheric PCDD/F emissions have also been reduced significantly over the last decades, amongst others due to the enforcement of stricter EU limits to for industrial facilities (metal industries, waste incineration, etc.) as regulated in the IED.

Due to their lipophilic properties and poor degradation, PCDD/Fs and dl-PCBs accumulate in the food chain. Animal derived food, including fish, contributes most to human exposure, due to the accumulation of these pollutants in fatty tissues and liver (EFSA Panel on Pollutants in the Food Chain et al., 2018).

13.3.3.2 Safe limit values for human consumption

A recent risk assessment by the European Food Safety Authority (EFSA) resulted in a lowering of the Tolerable Weekly Intake (TWI) from 14 to 2 pg TEQ/kg body weight (bw) per week, based on effects on sperm quality observed in humans (EFSA Panel on Pollutants in the Food Chain et al., 2018). This translates into a safe limit value of 2.9E-10 mg kg^{-1} body weight day^{-1} applied in this assessment

Across PCDD/Fs and PCBs, 17 congeners show different toxic potencies, which are expressed as toxic equivalency factors (TEFs) assigned to them based on in vivo and in vitro studies. The toxic potency of a sample is expressed as toxicity equivalents (TEQ) according to the WHO recommended methodology as the sum of the individual congener concentrations multiplied by their respective TEFs(Van den Berg et al., 2006). The use of TEFs provides opportunities to extrapolate the findings for a single compound (2,3,7,8-Tetrachlorodibenzo-p-dioxin, TCDD) and to extrapolate the findings for the broader group of PCDD/Fs and PCBs. The most toxic congener, 2,3,7,8-tetrachlorodibenzop-dioxin (TCDD) is used as the reference compound with an assigned TEF of 1 and good data availability for physico-chemical and toxicological properties.

13.3.3.3 Concentrations observed in sludge

Today, the dioxin contamination of sewage sludge without specific industrial contributions is generally below 10 ng TEQ/kg DM in Europe (Bayerisches Landesamt für Umwelt, 2011; Elskens et al., 2013; Zennegg et al., 2013; Weber et al., 2018), that have decreased significantly relative to previous years and decades (Oleszek-Kudlak et al., 2005; Vacha et al., 2005). Still it cannot be excluded that specific industrial sludges contain high amounts of PCDD/F and PCBs, e.g. following accidental spillage (Fuentes et al., 2007 up to 7000 ng kg-1 sludge; Balasubramani and Rifai, 2015). In this assessment, we assumed a PCDD/F + dl-PCB concentration of 20 ng WHO-TEQ kg^{-1} sludge dry matter, corresponding to a net mass of 10 ng kg^{-1} sludge for 2,3,7,8-TCDD used as reference compound.

13.3.3.4 Model input data

Table 18: Physico-chemical and toxicological data used for risk modelling purposes for the selected PCDD/Fs and dioxin-like PCBs.

	2,3,7,8-tetrachlorodibenzo p-dioxin (TCDD)
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	10E-06 (see main text)
Molecular weight (g mol-1)	321.98 (EQS dossier, 2011a)
Water solubility (mg L^{-1})	1.93E-02 (EQS dossier, 2011a)
Vapour pressure (Pa)	2.0E-07 (EQS dossier, 2011a)
Kow (log 10)	6.80 (EQS dossier, 2011a)
Koc (L kg^{-1})	3.98E+06 (Walters et al., 1989)
PNECaqua (µg L^{-1})	3.1E-09 (EQS dossier, 2011a)

PNECsoil (mg kg^{-1} ww)	1.73E-07 (equilibrium partitioning method)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	2.9E-10 (EFSA Panel on Pollutants in the Food Chain et al., 2018)
Half-life in soil (days)	3650 (EQS dossier, 2011a; Qiu et al., 2015)
Half-life in water (degradation, days)	263 646 (1)
Regional background in soil (mg kg^{-1} dry matter)	2.3E-06(2) (Elskens et al., 2013)
Regional background in surface water (µg L^{-1})	10.5E-09 (Deleebeeck et al., 2021)

(1) Conservative default half-life values (263 646 days) for non-biodegradable substances (527 292 days) have been used in the assessment. In any event, the main removal pathway from surface would be dilution with inflowing waters (surface water residence time: 40 days)

(2) Average levels for PCDD/F in Belgium agricultural soils

13.3.3.5 Estimated environmental and health exposure and risks from sludge application

The risk screening assessment resulting from the application of 2,3,7,8-TCDD present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that RCRs were above 1 for soils after 100 years of continuous application (Table 19). The RCR ratios for human health were above 1 and 100 in the short (1 year) and longer-term (>10 years).

Across the different food sources to total PAH human uptake, it was indicated that crop, meat and dairy were the main sources of 2,3,7,8-TCDD uptake (100%, Table 19, with negligible contributions from fish and drinking water due to the strong 2,3,7,8-TCDD adsorption capacity of the soil.

Table 19: Environmental and human health risk screening assessment of the exclusive application of 2,3,7,8-TCDD present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		2,3,7,8-TCDD
Soil	PEC$_{year\ 1/10/100}$ (mg kg^{-1} wet soil)	1.5E-08 1.4E-07 4.1E-07
	RCR$_{year\ 1/10/100}$ (-)	6.7E-02 6.2E-01 1.9E+00
Surface water	PEC$_{year\ 1}$ (µg L^{-1})	7.8E-10
	RCR $_{year\ 1}$ (-)	2.5E-01
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	5.5E-08/ 6.2E-06/ 7.0E-06

Relative contribution of fish consumption$_{10\ years}$ (%)	0%
Relative contribution of crop, meat and dairy consumption$_{10\ years}$ (%)	100%
Relative contribution of drinking water$_{10\ years}$ (%)	0%
RCR$_{year\ 1/10/100}$	2.7E+00 3.1E+02 3.5E+02

13.3.3.6 *Source contribution analysis*

When regional background concentrations (surface water, soil) and inputs from atmospheric deposition are considered, risk characterisation ratios for soils are exceeded after a single application in year 1 (Table 20), due to the high 2,3,7,8-TCDD background concentrations observed in soils. Risks for human health are also further augmented due to increases in PFAS concentrations in crops, dairy products and meat as a result of PFAS soil concentrations. Risk characterisation ratios for surface water exceed limit values that may be of concern to aquatic organisms.

Sludge-derived PAH become an important contributor to total 2,3,7,8-TCDD concentrations in the soil in the mid- to long-term (6% - 15% across all PFAS following 10-100 years of continuous sludge applications; assuming no further inputs from other sources such as atmospheric deposition) (Table 20).

In sum, our findings indicates that sludge may be a relevant source of 2,3,7,8-TCDD contamination in soils, that may cause human health risks based on our conservative modelling approaches.

Table 20: Environmental and human health risk screening assessment for 2,3,7,8-TCDD, based on its present background concentrations observed in the soil along with sewage sludge applications to agricultural land (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		2,3,7,8-TCDD
Soil	PEC$_{year\ 1/10/100}$ (mg kg^{-1} wet soil)	2.3E-06/ 2.4E-06/ 2.7E-06
	Contribution of sludge to total concentration (%)	1/ 6/ 15
	RCR$_{year\ 1/10/100}$ (-)	1.1E+01/ 1.1E+01/ 1.2E+01
Surface water	PEC$_{year\ 1}$ (µg L^{-1})	6.0E-09
	RCR $_{year\ 1}$ (-)	1.9E+00
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	7.5E-06/ 7.9E-06/ 8.7E-06
	RCR$_{year\ 1/10/100}$	3.7E+02/ 3.9E+02/ 4.4E+02

13.3.3.7 *Impact of sludge treatment*

The impact of biological treatment (composting and anaerobic) on PCDD/F and PCB removal is minimal to absent (Disse et al., 1995; Lü et al., 2021). Because PCDD/Fs do not have high thermal stability, thermal treatment is a viable method for PCDD/Fs and dl-PCB destruction. Similar to PAH, incineration in line with the established conditions for waste incineration in the IED has the potential to remove >99% PCDD/F (Vehlow et al., 2006). Nonetheless, well-designed and state-of-the-art technologies are critical as PCDD/F and PCBs could *de novo* be produced during high-temperature processing during incineration, especially under conditions of incomplete combustion or re-adsorption gas particles on solid material in poorly designed thermal reactors (Huygens et al., 2019).

13.3.4 Flame retardants (polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD), and organophosphate compounds)

13.3.4.1 Background and selection of consumption

Flame retardants are mixtures of man-made chemicals that are added to a wide variety of products, including for industrial use, to make them less flammable. They are used commonly in plastics, textiles and electrical/electronic equipment.

Since the 1970s, the primary flame retardants used were the polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD). However, due to concerns regarding their persistence, toxicity and bioaccumulative potential, these compounds have been added to the Stockholm Convention on Persistent Organic Pollutants, including the most recent addition of deca-BDE (also called BDE-209, referring to the PBDE with 10 bromines) in 2017. However, because of their persistence in the environment there are still concerns about the risks these chemicals pose to public health. In addition, decabromodiphenyl ether (BDE-209) acts as a slow-release reservoir for lower brominated, more toxic PBDEs, via abiotic and biotic transformation processes. As a result, human populations and biota worldwide are exposed to BDE-209 and other PBDEs in combination. Flame retardants, whether in use or waste-derived, can leach into the environment and contaminate the air, soil and water. These pollutants may then enter the food chain where they mainly occur in food of animal origin, such as fish, meat, milk and derived products. In addition, discontinued flame retardants have been substituted by alternative substances such as novel brominated, chlorinated and organophosphate compounds. Some of these substitutes are suspected of being of health and environmental concern, although robust toxicological data are often missing to perform a sound risk assessment (van der Veen and de Boer, 2012; Hendriks and Westerink, 2015).

To select substances belonging to the class of flame retardants, preliminary evidence indicated high daily intake for the legacy flame retardants decabromodiphenyl ether (BDE-209), 2,2',4,4'-Tetrabromodiphenyl ether (BDE-47), 2,2',4,4',5-Pentabromodiphenyl ether (BDE-99) and hexabromocyclododecane (HBCDD) as well as for tris(2-ethylhexyl) phosphate (TEHP) and 2-ethylhexyl diphenyl phosphate (EHDPP). For all these compounds information on sludge concentrations and safe limit values. In addition, also data for tris(1-chloro-2-propyl) phosphate (TCPP) could be retrieved. These compounds were taken forward in the assessment.

Other flame retardants were, such as tetrabromobipshenol A, were not taken forward in a more in-dept assessment because the high safe limit values and associated low risk characterisation ratios observed in the prioritisation step (see section 3.3.1) (EFSA Panel on Pollutants in the Food Chain, 2011)

13.3.4.2 Safe limit values for human consumption

Safe limit values for the selected compound were obtained from a variety of sources (EFSA, ECHA, US-EPA).

Table 21: Safe limit values for human consumption used for risk modelling purposes for the selected flame retardants.

Substance	Safe limit values	Value (mg kg^{-1} body weight day^{-1})	Source
BDE-209	Chronic human intake associated with the body burden calculated at the benchmark dose (lower confidence limit) (BMDL), divided by a minimum Margin of Exposure (MOE) of 2.5	6.80E-01	(EFSA CONTAM Panel, 2011)
BDE-47		6.88E-05	(EFSA CONTAM Panel, 2011)
BDE-99		1.68E-06	(EFSA CONTAM Panel, 2011)
HBCDD	Chronic human intake associated with the body burden calculated at the lowest observed adverse effect level (LOAEL), divided by a minimum Margin of Exposure (MOE) of 24	9.79E-05	(EFSA CONTAM Panel, 2021)
TEHP	Reference dose	1.00E-01	(U.S. EPA, 2017; Gbadamosi et al., 2021)
EHDPP	Derived No Effect Level (oral route), based on repeated dose toxicity as most sensitive endpoint and an assessment factor of 200	3.60E-2	ECHA registration dossier for 2-ethylhexyl diphenyl phosphate

TCPP	Reference dose	1.00E-02	(U.S. EPA, 2017; Gbadamosi et al., 2021)

13.3.4.3 Concentrations observed in sludge

Öberg et al. (2002) indicated that BDE concentrations were in the range not detected to 450 µg kg^{-1} wet weight. Eljarrat (2008) showed PBDE concentrations in sewage sludge from different locations throughout Spain ranging from 197 to 1185 µg kg^{-1} dry matter, being deca-BDE-209 the predominant congener. Kupper et al. (2008) indicated mean concentrations for samples from Switzerland of 310, 149, and 95 µg kg^{-1} dry matter for BDE-209, HBCD, penta-BDE (including BDE-49 and BDE-99 as most abundant congeners), respectively. The ranges from Sweden, Denmark, the Netherlands, reported in the review study of Law et al. (2006) are generally aligned to these results. The PBDE concentrations in sewage sludge samples from 11 German wastewater treatment plants had average concentrations of 48 µg kg^{-1} dry matter (BDE-47), 54 µg kg^{-1} dry matter (BDE-99), and 429 (BDE-209) µg kg^{-1} dry matter (Knoth et al., 2007). Average values of 44 µg and 35 µg kg^{-1} DS for the same congeners BDE-47 and BDE-99 were measured by (Rigby et al., 2021). Gorga et al. (2013) reported PBDE concentrations for Spanish sewage sludges that ranged between 20.7 and 2326 µg kg^{-1} with a median value of 298 µg kg^{-1}. BDE values in German sewage sludge were below 50 µg kg^{-1} for all congeners (Wiechmann et al., 2013). The most abundant PBDE congener in these samples was BDE-209 with concentration levels ranging from not detected to 2303 µg kg^{-1}. For HBCD, Kupper et al. (2008) indicated average values of 149 µg kg^{-1}, whereas average values of 18 µg kg^{-1} (with a maximum of 98 µg kg^{-1}) were reported by Gorga et al. (2013). Recently, Rigby et al. (2021) reported HBCB concentrations between 33 and 45 µg kg^{-1} DS for UK samples. In this assessment, sludge concentration values of 500 µg kg^{-1} dry matter was assumed for BDE-209, whereas for BDE-47, BDE-99 and HBCD a value of 200 µg kg^{-1} dry matter was assumed. For the assessment, sludge concentration values of 0.5 mg kg^{-1} dry matter was assumed for BDE-209, whereas for BDE-47, BDE-99 and HBCD a value of 0.21 mg kg^{-1} dry matter was assumed.

Concentrations values are hardly reported for PBDE replacement chemicals in sludge. Rigby et al. (2021) showed that tris(1-chloro-2-propyl) phosphate (TCPP) was the most abundant organophosphate flame retardant in sewage sludges, with concentrations up to almost 1.0 mg kg^{-1}. These values are very similar to trends among organophosphate flame retardants and maximum values reported for TCPP by Cristale et al. (2016). Maximum concentrations for TEHP and EHDPP reported in the same study were 1.2 mg dry matter kg^{-1}, and 0.5 mg dry matter kg^{-1} matter, respectively. Given the reduced data availability, these maximum values were retained in this assessment.

13.3.4.4 Model input data

The applied model for human intake is for most susbtances based on substance-specific bioconcentration and biotransfer factors as deriving the latter parameters based on Kow values results in an overestimated of daily intake, especially for BDEs characterised by a high Kow value (Kow>7).

Table 22: Physico-chemical and toxicological data used for risk modelling purposes for the selected flame retardants.

	BDE-209	**BDE-47**	**BDE-99**	**HBCDD**	**TEHP**	**EHDPP**	**TCPP**
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	0.5 (see main text)	0.1 (see main text)	0.1 (see main text)	0.1 (see main text)	1.2 (Rigby et al., 2021)	0.5 (Cristale et al., 2016)	1.0 (Cristale et al., 2016)
Molecular weight (g mol-1)	959.17 (core database)	485.8 (core database)	564.69 (core database)	641.7 (core database)	434.65 (core database)	362.4 (Brooke et al., 2009)	327.59 (core database)
Water solubility (mg L^{-1})	6.2E-02 (core database)	2.5E-02 (core database)	2.5E-01 (core database)	8.6E-03 (core database)	5.1E+00 (core database)	1.0E+00 (Brooke et al., 2009)	1.6E+03 (core database)

Vapour pressure (Pa)	5.9E-09	6.3E-05	1.5E-05	3.3E-06	1.9E-04	3.4E-4	2.7E-01
	(core database)	(core database)	(core database)	(core database)	(core database)	(Brooke et al., 2009)	(core database)
Kow (log 10)	9.70	6.57	7.30	5.47	9.42	5.73	2.68
	(deBruyn et al., 2009)	(deBruyn et al., 2009)	(deBruyn et al., 2009)	(EFSA CONTAM Panel, 2021)	(Sühring et al., 2020)	(Brooke et al., 2009)	(Sühring et al., 2020)
Koc (L kg^{-1})	1.4E+06	9.8E+05	9.8E+05	2.2E+05	6.2E+05	9.5E+03	5.8E+02
	(EQS Dossier, 2011)	(EQS Dossier, 2011)	(EQS Dossier, 2011)	(EQS dossier, 2011b)	(core database)	(Brooke et al., 2009)	(core database)
PNECaqua (µg L^{-1})	0.13	0.049	0.049	0.31	6.7E+00	0.18	59.16
	(core database)	(EQS Dossier, 2011)	(EQS Dossier, 2011)	(EQS dossier, 2011b)	(core databse)	(Brooke et al., 2009)	(Xing et al., 2019)
PNECsoil (mg kg^{-1} ww)	98	8.47E-01	8.47E-01	1.20E+00	7.33E+01	3.02E-02	6.12E-01
	ECHA registration dossier	(equilibrium partitioning method)	(equilibrium partitioning method)	(equilibrium partitioning method)	(equilibrium partitioning method)	(equilibrium partitioning method)	(equilibrium partitioning method)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	6.80E-01	6.88E-05	1.68E-06	9.79E-05	1.00E-01	3.60E-02	1.00E-02
	(EFSA CONTAM Panel, 2011)	(EFSA CONTAM Panel, 2011)	(EFSA CONTAM Panel, 2011)	(EFSA CONTAM Panel, 2021)	(U.S. EPA, 2017; Gbadamosi et al., 2021)	ECHA registration dossier for 2-ethylhexyl diphenyl phosphate	(U.S. EPA, 2017; Gbadamosi et al., 2021)
Half-life in soil (days)	861	342	342	63	4320	300	47520
	(Andrade et al., 2017)	(Andrade et al., 2017)	(Andrade et al., 2017)	(Davis et al., 2005)	(Sühring et al., 2020)	(Brooke et al., 2009)	(Sühring et al., 2020)
Half-life in water (degradation, days)	263 646	263 646	263 646	263 646	240	50	3270
	(default value non-biodegradable)	(default value non-biodegradable)	(default value non-biodegradable)	(default value non-biodegradable)	(Sühring et al., 2020)	(Brooke et al., 2009)	(Sühring et al., 2020)
Regional background in soil (mg kg^{-1} dry matter)$^{(2)}$	Not considered	1.0E-03	1.0E-3	Not considered	0.1	Not considered	Not considered
		(Osteras et al., 2014; Kim et al., 2017)	(Osteras et al., 2014; Kim et al., 2017)		(Kurt-Karakus et al., 2018)		
Regional background in	Not considered	2.50E-05	2.50E-05	5.00E-05	Not considere	Not considere	Not considere

surface water ($\mu g\ L^{-1}$)					d	d	d
		(Rauchbüchl, 2015)	(Rauchbüchl, 2015)	(Rauchbüchl, 2015)			
BCFfish-water (L kg^{-1}ww)	50	3.5E+04	1.4E+04	1.8E+04	5.01E+01	1.8E+04	3.6E+00
	(U.S. Department of Health & Human Services, n.d.)	(EQS Dossier, 2011)	(EQS Dossier, 2011)	(EQS dossier, 2011b)	(4)	(default value estimated based on Kow)	(4)
BTFshoot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	4.04E-03	1.21E-02	1.21E-02	2.02E-02	7.18E-03	7.75E-03	5.17E-02
	(1)(EFSA CONTAM Panel, 2011)	(1)	(1)	(default calculation)	(B. Hu et al., 2021)	(B. Hu et al., 2021)	(B. Hu et al., 2021)
BTFroot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	4.04E-03	1.21E-02	1.21E-02	1.11E-01	1.05E-02	8.07E-03	4.60E-01
	(1)	(1)	(1)	(default calculation)	(B. Hu et al., 2021)	(B. Hu et al., 2021)	(B. Hu et al., 2021)
BTFmeat-grass (day kg-1 dry matter)	7.08E-02	1.26E-03	1.26E-03	7.41E-03	7.73E+01	1.35E-02	9.7E-06
	(3)	(Takaki et al., 2015)(2)	(Takaki et al., 2015)(2)	(default value estimated based on Kow)	(McKone, 1994)(4)	(default value estimated based on Kow)	(McKone, 1994) (4)
BTF-milk-grass (day kg^{-1} dry matter)	2.14E-02	1.26E-03	1.26E-03	2.34E-03	2.4E+01	4.27E-03	3.8E-06
	(3)	(Takaki et al., 2015)(2)	(Takaki et al., 2015)(2)	(default value estimated based on Kow)	(McKone, 1994) (4)	(default value estimated based on Kow)	(McKone, 1994) (4)
Biomagnification	1	10	1	10	1	10	1
	(default value based on Kow)	(default value based on Kow)	(UNEP, 2006)	(default value based on Kow)	(default value based on Kow)	(default value based on Kow)	(default value based on Kow)

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(1)Reported $BCF_{plant-soil}$ are invariably below 1 and usually in the range of 0.1-0.01 (EFSA CONTAM Panel, 2011)(Dobslaw et al., 2021), and are inversely related to Kow (She et al., 2013). Based on the Kow values and the values reported by (She et al. (2013), values of BCF values 0.05 (BDE-209), 0.15 (BDE-47 and BDE-99) on a dry matter basis were selected. Values indicated on a dry matter basis have been transformed into values on a wet mass basis (mg kg-1 plant ww)/(mg kg-1 soil ww) assuming a water content in soil of 20vol% (EUSES default, conversion factor for soil concentration wet-dry weight soil of 1.13), and a water content in roots of 65vol%, and a bulk density of plants of 700 kg/m3 (EUSES default), (2)BTF$_{\text{meat-grass}}$ data available for BDE-49, assumed to be equal for BDE-99. Gicen that BDE levels in milk and dairy products are on average below these of meat (EFSA CONTAM Panel, 2011), the same BTF factors were applied to model BDE grass to milk transfer as a conservative estimate.

(3)Default values based on Kow were retained as a conservative estimate for BTF

(4)EPI data obtained from the Risk Assessment Information System, University of Tennessee (https://rais.ornl.gov/cgi-bin/tools/TOX_search?select=chemspef)

13.3.4.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of PAH present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that risk characterisation ratios below 1 for soils, surface waters and human health (Table 23). The risk screening analysis indicated that RCR ratios for soil, surface water and human health were generally below 1. The greatest health concerns were associated to to BDE-99 and to a smaller extent to TEHP (with RCR values of 2.1.E-01, and 2.3E-01 after 100 years, respectively; see Table 23).

For BDE-99, fish (63%) and crop/grass/meat (37%) contributed most to total human intake, in line with observations that fish is the food source for total BDE-99 intake by the population (EFSA CONTAM Panel, 2011). The contribution of food sources to total intake for flame retardants, other than BDE-99, should be interpreted with the necessary caution as (i) no data refinements were performed given the low RCR ratios for human health, (ii) intake values are rather low. Nonetheless, it is clear that the fish bioconcentration factors documented across the range of flame retardants varies markedly in literature (see section 13.3.4.4).

Table 23: Environmental and human health risk screening assessment of the exclusive application of flame retardants present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio; BDE-209: decabromodiphenyl ether, BDE-47: 2,2',4,4'-Tetrabromodiphenyl ether, BDE-99: 2,2',4,4',5-Pentabromodiphenyl ether and HBCDD: hexabromocyclododecane; TEHP: tris(2-ethylhexyl) phosphate; EHDPP: 2-ethylhexyl diphenyl phosphate TCPP: tris(1-chloro-2-propyl) phosphate (TCPP))

		BDE-209	**BDE-47**	**BDE-99**	**HBCDD**	**TEHP**	**EHDPP**	**TCPP**
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	7.3E-04/ 4.2E-03/ 5.7E-03/	1.4E-04/ 4.4E-04/ 5.4E-04	1.4E-04/ 4.4E-04/ 5.5E-04	1.3E-04/ 1.5E-04/ 1.7E-04	1.8E-03/ 1.7E-02/ 5.7E-02/	7.2E-04/ 1.9E-03/ 2.4E-03	1.5E-03/ 1.1E-02/ 1.9E-02
	RCR$_{\text{year 1/10/100}}$ (-)	7.5E-06/ 4.2E-05/ 5.8E-05	1.7E-04/ 5.2E-04/ 6.4E-04	1.7E-04/ 5.2E-04/ 6.4E-04	1.1E-04/ 1.3E-04/ 1.4E-04	2.4E-05/ 2.3E-04/ 7.8E-04	2.4E-02/ 6.5E-02/ 8.0E-02	2.4E-03/ 1.8E-02/ 3.1E-02
Surface water	PEC$_{\text{year 1}}$ (µg L^{-1})	1.1E-04	3.2E-05	3.2E-05	1.9E-04	1.2E-03	3.3E-02	5.6E-01
	RCR $_{\text{year 1}}$ (-)	8.6E-04	6.5E-04	6.5E-04	6.1E-04	1.8E-04	1.8E-01	9.5E-03
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	5.3E-05/ 1.2E-04/ 1.6E-04	2.8E-04 2.9E-04 2.9E-04	1.4E-05 2.0E-05 2.2E-05	8.8E-04 8.8E-04 8.8E-04	2.0E-01 5.5E-01 1.9E+00	8.1E-02/ 8.1E-02/ 8.1E-02	4.4E-04 2.6E-03 4.6E-03
	Relative contribution of fish consumption$_{\text{10 years}}$ (%)	3	98	63	100	0	100	17
	Relative contribution of crop, meat and dairy consumption$_{\text{10 years}}$ (%)	97	2	37	0	100	0	81
	Relative contribution of drinking water$_{\text{10 years}}$ (%)	0	0	0	0	0	0	2
	RCR$_{\text{year 1/10/100}}$	1.1E-06/ 2.5E-06/ 3.5E-06	5.9E-02 6.0E-02 6.1E-02	1.2E-01 1.7E-01 1.8E-01	1.3E-01 1.3E-01 1.3E-01	2.9E-02 7.8E-02 2.7E-01	3.2E-02/ 3.2E-02/ 3.2E-02	6.3E-04 3.8E-03 6.5E-03

13.3.4.6 Source contribution analysis

Given the low risk ratios for most flame retardants, the source contribution was focused on the three compounds that showed the highest RCR ratio, being BDE-99, BDE-47, HBCDD and TEHP (Table 23). Results indicated that the observed RCR values for human health were slightly higher compared to the exclusive sludge modelling results, but even in the long-term all below 1. RCR values after 100 years were 2.1E-01, 6.0E-1, 1.0E-1 and 6.3E-1 for BDE-47, BDE-99, HCBDD, and TEHP, respectively.

13.3.4.7 Impact of sludge processing

Given the recalcitrance of the BDEs and HCBDD under aerobic and anaerobic conditions under lower temperatures, it seems unlikely that microbial degradation during composting and anaerobic digestion may remove significant shares of these compounds. Some microbial degradation has been observed for organophosphorus compounds (Singh and Walker, 2006), but the efficiency of microbial processes to remove organophosphate compounds has not been documented (Lü et al., 2021). Incineration is effective to remove BDEs (Wang et al., 2010), HCBDD (Mark et al., 2015), and organophosphate compounds (Korobeinichev et al., 1997) to low concentrations ranges in ashes, mainly fly ashes.

13.3.5 Alkylphenols

13.3.5.1 Introduction and selection of compounds

Nonylphenol is used in the manufacture of antioxidants, lubricating oil additives and the production of nonylphenol ethoxylates surfactants. Nonylphenol ethoxylates are highly cost effective surfactants with exceptional performance and consequently used widely in industrial, institutional, commercial and household applications such as detergents, emulsifiers, wetting and dispersing agents, antistatic agents, demulsifiers and solubilisers (Soares et al., 2008).

Nonylphenols and nonylphenol ethoxylates are included in the Annex XVII of the REACH Regulation (REACH restricted substances). 4-nonylphenol ethoxylated, branched and linear, and octylphenol are also included in the "Authorisation List" of the Annex XIV of REACH. This annex lists substances for which authorization is necessary in order to be able to place them on the market within the European Union (concerning uses covered by the REACH Regulation). Octylphenols and nonylphenols are on the list of priority substances of the Water Framework Directive. Nonylphenol ethoxylates are now being replaced by other surfactants in Europe, mainly by alcohol ethoxylates.

Nonylphenol persists in aquatic environments and is moderately bioaccumulative. It is not readily biodegradable, and it can take months or longer to degrade in surface waters, soils, and sediments. Ranges for octylphenol are one order of magnitude lower (Bolz et al., 2001).

13.3.5.2 Safe limit values for human consumption

For nonylphenol, a LOAEL for increased incidence of renal tubular degeneration and/or dilation in a three-generation study on rats was 15 mg kg^{-1} body weight (bw) day^{-1} (NTP, 1997), and a total safety factor of 3000, a safe limit value of 5 µg kg^{-1} bw day^{-1} was set, whereas for NPEO a value of 13 µg kg^{-1} bw day^{-1} was derived (Nielsen et al., 2000). Based on the LOAEL for sperm tail abnormalities in rats (20 ng kg^{-1} bw) (Blake et al., 2004) and a safety factor 300, a tolerable intake was assumed 0.067 ng kg^{-1} bw day^{-1} was used for octylphenol.

13.3.5.3 Sludge concentrations

The alkylphenol concentrations reviewed in the 2012 JRC study (Tavazzi et al., 2012) and earlier works (Bolz et al., 2001; Abad et al., 2005; Fountoulakis et al., 2005; Krogh et al., 2007) mostly indicate average nonylphenol and nonylphenol etoxylate concentration values in the range 10 - 100 mg kg^{-1} dry weight. Ranges for octylphenol are one order of magnitude lower (Bolz et al., 2001). Data on actual compositions, following the implementation of new legislation that restricts the use of alkylphenols, are scarce. Nonetheless, preliminary evidence suggests that concentrations of alkylphenols have decreased in last years as a result fo regulatory actions (Veenaas et al., 2018). Lamastra et al. (2018) indicated concentration values for nonylphenols and nonylphenol ethoxylates that vary from 1.32 to 103 mg kg^{-1} DM, with a median of 6.24 mg kg^{-1} DM). Veenas et al. (2018) indicated a maximum concentration observed of 0.73 mg kg^{-1} for octyphenol in sewage sludge from Sweden. Based on these data, a value of 10 mg kg^{-1} DM for nonylphenol/nonylphenol ethoxylate and 1 mg kg^{-1} DM for octylphenol is applied in this study.

13.3.5.4 Model input data

Table 24: Physico-chemical and toxicological data used for risk modelling purposes for the selected alkylphenols.

	4-nonylphenol	**4-nonylphenol ethoxylated (NP2EO)**	**4-tert-Octylphenol**
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	10	10	1
Molecular weight (g mol-1)	220.4	308.5	206.32

		(core databse)	
Water solubility (mg L^{-1})	5.7	1.05	12.6
		(UNEP, 2017a)	
Vapour pressure (Pa)	3.0E-01	1.8E-05	2.3E-1
		(UNEP, 2017a)	
Kow (log 10)	5.4	5.3	4.1
Koc (L kg^{-1})	1.91E+01	2.51E+03	1E+04
		(UNEP, 2017a)	
PNECaqua (µg L^{-1})	0.33	0.33	0.33
	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)
PNECsoil (mg kg^{-1} ww)	1.2	1.2	1.2
	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	5.00E-03 (2)	1.30E-02	6.70E-08
Half-life in soil (days)	16.5	16.5	14
	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)	(Langdon et al., 2011)
Half-life in water (degradation, days)	37	37	50
	(Swedish Chemicals Agency, 2013)	(Swedish Chemicals Agency, 2013)	(UNEP, 2017a)
BCFfish-water (L kg-1ww)	3.14E+03	3.40E+01	4.70E+02
	(UNEP, 2017a)	(UNEP, 2017a)	(UNEP, 2017a)

(1)4-Nonylphenol has a dissociation constant of approximately or or 10, indicating ionic properties. Log Kow is a conservative estimate of the lipid partitioning: when the compound's potential to ionise is taken into account, the overall partitioning for all of its species (neutral + ionic) may be considerable considerably less (ECETOC, 2013). A general rule is to assume that the ionised species will not be available for partitioning to air or organic material, and is thus entirely present in the dissolved phase (ECETOC, 2013).

(2) For nonylphenol, a LOAEL for increased incidence of renal tubular degeneration and/or dilation in a three-generation study on rats was 15 mg kg-1 bw day-1 (NTP, 1997), and a total safety factor of 3000, a safe limit value of 5 µg kg-1 bw day-1 was set, whereas for NPEO a value of 13 µg kg-1 bw day-1 was derived (Nielsen et al., 2000).Based on the LOAEL for sperm tail abnormalities in rats (20 ng kg-1 bw) (Blake et al., 2004) and a safety factor 300, a tolerable intake was assumed 0.067 ng kg-1 bw day-1 was used for octylphenol.

(3)In the absence of a well-developed database on the transfer of alkylphenols from soil to plant, the values reported by Sjöström et al. (2008) for sewage sludge amended soils were used to model plant uptake. The reported value of 0.71 on a dry matter basis was converted towards a wet mass basis (mg kg-1 plant ww)/(mg kg-1 soil ww) assuming a water content in soil of 20vol% (EUSES default, conversion factor for soil concentration wet-dry weight soil of 1.13), and a water content in roots of 65vol%, and a bulk density of plants of 700 kg/m3 (EUSES default),. These values are used for all alkylphenols under study. The concentrations in root were multiplied with a factor 2 to account for the observed enrichment of shoots versus roots (Y. Hu et al., 2021). Further transfer to meat and milk was modelled using the default model based on Kow.

13.3.5.5 *Estimated environmental and health exposure and risks from sludge application*

The results of the assessment that consider the exclusive sludge applications on agricultural land indicates an exceedance of RCR for surface water (4-nonylphenol) and human health in the short- and long-term for 4-nonylphenol and 4-tert-octylphenol. Significant concerns have been observed for the former, whereas moderate concerns are indicated for the latter. Due to relatively fast degradation of alkylphenols in soils (Langdon et al., 2011; Swedish Chemicals Agency, 2013), the RCR ratios do not increase over time.

Table 25: Environmental and human health risk screening assessment of the exclusive application of flame retardants present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		4-nonylphenol	**4-nonylphenol ethoxylated (NP2EO)**	**4-tert-octylphenol**
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	8.9E-03 /	1.1E-03/	1.0E-03/
		8.9E-03/	1.1E-03/	1.0E-03/
		8.9E-03	1.1E-03	1.0E-03
	RCR$_{\text{year 1/10/100}}$ (-)	7.4E-03/	8.9E-04/	8.4E-04/
		7.4E-03/	9.0E-04/	8.4E-04/
		7.4E-03	9.0E-04	8.4E-04
Surface water	PEC$_{\text{year 1}}$ (µg L^{-1})	3.4E+00	8.1E-02	2.4E-02
	RCR $_{\text{year 1}}$ (-)	1.0E+01	2.5E-01	7.2E-02
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	1.8E+00/	2.1E-03/	2.0E-04/
		1.8E+00/	2.1E-03/	2.0E-04/
		1.8E+00	2.1E-03	2.0E-04
	Relative contribution of fish consumption$_{\text{10 years}}$ (%)	17	2	92
	Relative contribution of crop, meat and dairy consumption$_{\text{10 years}}$ (%)	83	98	7
	Relative contribution of drinking water$_{\text{10 years}}$ (%)	0	0	1
	RCR$_{\text{year 1/10/100}}$	5.2E+00/	2.3E-03	4.2E+01/
		5.2E+00/	2.3E-03	4.2E+01/
		5.2E+00	2.3E-03	4.2E+01

13.3.5.6 *Source contribution analysis*

No source contribution analysis has been performed since data on background concentrations in environmental compartments is expected to decrease over time due to regulatory actions, and the relatively low residence times of alkylphenols in soils and waters. Therefore, relevant data could not be acquired.

13.3.5.7 *Impact of sludge processing*

The recent review paper of Lü et al. (2021) investigated the levels and removal of nonylphenol and nonylphenol ethoxylates during sludge composting. During sludge composting, the degradation of nonylphenol ethoxylates could form nonylphenol, leading to slightly higher concentrations of nonylphenol and the sum of (nonylphenol + nonylphenol ethoxylates) in sludge composts than in untreated sludge (González et al., 2010). On the other hand, a considerable removal (64–95%) of nonylphenol and nonylphenol ethoxylates was recorded during sludge composting in other works (Pakou et al., 2009; Priac et al., 2017; Zheng et al., 2018).

Anaerobic digestion has been observed to remove roughly 50% of alkylphenols in sludge (Paterakis et al., 2012). No data on the fate of nonylphenols by incineration have been identified, but most probably the substances would be nearly 100% destroyed by the incineration process, based on similarities with other non-persistent substances (Environment, 2013).

13.3.6 Polychlorinated alkanes

13.3.6.1 Introduction and selection of compounds

Polychlorinated alkanes represent a large class of compounds that are typically classified according to structural characteristics such as carbon chain length and degree of chlorination. Polychlorinated alkanes are also classified according to the variety of feedstocks from which they are manufactured (polychlorinated paraffins and polychlorinated olefins, for example, are two such categories). Especially paraffins are stable compounds, and short-chain chlorinated paraffins (SCCPs) are listed as persistent organic pollutants under the Stockholm Convention (EFSA Panel on Contaminants in the Food Chain et al., 2020). Because of regulations and concerns about SCCPs, the use of mid-chain chlorinated paraffins (MCCPs) has increased and has represented the majority of production over the last decades (Glüge et al., 2018). An estimate derived from the addition of each individual registrant volumes from the ECHA shows that the amount of CPs manufactured and/or imported in the European Economic Area is around 10,000–100,000 tonnes per year (EFSA Panel on Contaminants in the Food Chain et al., 2020).

Polychlorinated alkanes have been produced since the 1930s for a variety of purposes (van Mourik et al., 2015; EFSA Panel on Contaminants in the Food Chain et al., 2020). They are used as additives in lubricants and cutting fluids in the metal industry and are also used as flame retardants in the rubber industry, in PVC plastics and in sealants for use in building, automotive and industrial applications, and the longer chain mixtures are also used as plasticisers in paints and other materials. Of particular relevance for this work is that they are consumed in the textile industry for the production of flame-resistant, water repellent and rot-preventing textile finishes.

13.3.6.2 Safe limit values for human consumption

The recent assessment of EFSA on polychlorinated paraffins selected as reference points a BMDL10 of 2.3 mg/kg bw per day for increased incidence of nephritis in male rats, and of 36 mg/kg bw per day for increased relative kidney weights in male and female rats for SCCPs and MCCPs, respectively (EFSA Panel on Contaminants in the Food Chain et al., 2020). For long-chain chlorinated paraffins, a reference point relevant for humans could not be identified. The Panel also concluded that a Margin of Exposure (MoS) higher than 1000 might indicate that there is no health concern. Therefore, the safe limit values for human consumption were set at 2.3E-3 mg/kg bw per day and 36E-3 mg/kg bw per day for SCCPs and MCCPs, respectively.

13.3.6.3 Concentrations observed in sludge

Recent concentration data for sludges generated in the EU are difficult to collect. Rigby et al. (2021) indicated summed polychlorinated alkane concentrations, including SCCPs and MCCPs and likely other substances, of 6 and 140 mg kg^{-1} dry matter for two sewage sludges from the UK. With specific reference to SCCPs and MCCPs, Thomas et al. (2011) reported average concentration for sewage sludges from Norway of 12 mg kg-1 (SCCPs) and 7 mg kg-1 (MCCPs). Sewage sludge from Swedish WWTPs collected between 2004 and 2010 showed a median concentration for SCCPs of 1.1 mg/kg dry weight (dry matter) and for MCCPs a median concentration of 3.8 mg/kg dry matter (Olofsson et al., 2012). Values in sludge from wastewater treatment plants in the Czech Republic (0.4 and 2.3 mg kg^{-1} dry matter for SCCPs and MCCPs, respectively) and Switzerland (0.5 -1.2 and 0.1 - 80 mg kg^{-1} dry matter for SCCPs and MCCPs, respectively) (Přibylová et al., 2006; Bogdal et al., 2015). Older studies indicated on average higher concentrations (Clarke and Smith, 2011), with for instance SCCPs measured in two German sludge samples, which contained 65 and 47 mg kg^{-1} dry matter of SCCPs (Rieger and Ballschmiter, 1995). Concentrations of MCCPs in samples of digested sludge from the UK were in the range 1.8 to 93 mg kg^{-1} dry matter (Nicholls et al., 2001). In another UK survey, SCCPs and MCCPs concentrations were between 7–200 mg kg^{-1} dry matter and 30–9700 mg kg^{-1} dry matter, respectively (Stevens et al., 2003). Comparison of data from the limited amount of information available is difficult and further complicated by the lack of standardised measurement methods for SCCPs and MCCPs.

For this study, high-end values (aligned to the recent observations from Rigby et al. (2021)) of 100 mg kg^{-1} dry matter were assumed for both SCCPs and MCCPs. Actual and standardised data on polychlorinated alkanes in sewage sludge is required to fine-tune the assessment.

13.3.6.4 Model input data

Table 26: Physico-chemical and toxicological data used for risk modelling purposes for the selected polychlorinated alkanes.

	SCCPs(1)	**MCCPs(1)**
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	100	100
	(see main text)	(see main text)
Molecular weight (g mol-1)	320-500 (410)	300 - 600 (450)
	(ECHA, 1999)	(UK Environment Agency, 2019)
Water solubility (mg L^{-1})	0.15 - 0.47 (0.33)	0.005 - 0.027 (0.016)
	(ECHA, 1999)	(UK Environment Agency, 2019)
Vapour pressure (Pa)	2.1E-2	2.7E-4
	(ECHA, 1999)	(UK Environment Agency, 2019)
Kow (log 10)	4.4 – 8.0 (6.7)	4.7 – 8.3 (7.2)
	(ECHA, 1999)	(UK Environment Agency, 2019)
Koc (L kg^{-1})	3.4E+05	8.5E+05
	(derived from Kow)(2)	(derived from Kow)(2)
PNECaqua (µg L^{-1})	0.5	0.87
	(ECHA, 1999)	(UK Environment Agency, 2019)
PNECsoil (mg kg^{-1} ww)	5.2	10.6
	(equilibrium partitioning method)	(UK Environment Agency, 2019)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	2.3E-03	3.6E-02
	(EFSA Panel on Contaminants in the Food Chain et al., 2020)	(EFSA Panel on Contaminants in the Food Chain et al., 2020)
Half-life in soil (days)	Unknown (default value for not biodegradable, 527292 days)	Unknown (default value for not biodegradable, 527292 days)
Half-life in water (degradation, days)	Not readily or inherently biodegradable (1000)	1-9300 (1000)
	(ECHA, 1999)	(Glüge et al., 2018; UK Environment Agency, 2019)
Regional background in soil (mg kg^{-1} dry	1.0E-03	3.0E-02

matter)(2)	(Halse et al., 2015)	(Glüge et al., 2018)
Regional background in surface water (μg L^{-1})	Not available	0.13 (Glüge et al., 2018)

(1)In contrast to other well-defined substances, SCCPs and MCCPs are complex industrial chemicals with the molecular formula CnH2n+2−xClx are a mixture of different compounds with varying carbon chain length and degree of chlorination that may have different physicochemical and toxicological properties. The values presented here are therefore not representative for one specific compound, but rather aim to provide a general group estimate. Ranges are given for the different parameters, with the values used for modelling indicated in between brackets.

(2)Based on EUSES default for hydrophobic substances: log Koc = 0.81 x log Kow + 0.1

13.3.6.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of SCCPs and MCCPs present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that risk characterisation ratios below 1 for soils after 100 years of continuous sludge application. For human health, RCRs above 1 were indicated for the entire time period under study, up to a value of 1.6E+03 indicating significant risks in the long-term (100 years) (Table 27). Soil grown crops, dairy and meat products were the exclusive sources that contributed to SCCPs and MCCPs intake, whereas contaminant transfer to drinking water and fish were negligible (Table 27).

Table 27: Environmental and human health risk screening assessment of the exclusive application of short- and long-chain polychlorinated paraffins (SCCPs and MCCPs, respectively) present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		SCCPs	**MCCPs**
Soil	$\text{PEC}_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	1.5E-01/ 1.6E+00/ 1.3E+01	1.5E-01/ 1.6E+00/ 1.3E+01
	$\text{RCR}_{\text{year 1/10/100}}$ (-)	1.2E-01 1.3E+00 1.1E+01	1.2E-01 1.3E+00 1.1E+01
Surface water	$\text{PEC}_{\text{year 1}}$ (μg L^{-1})	9.3E-02	3.7E-02
	$\text{RCR}_{\text{year 1}}$ (-)	1.9E-01	4.2E-02
Human	Total daily $\text{intake}_{\text{year 1/10/100}}$ (mg day^{-1})	4.0E+00 3.3E+01 2.6E+02	2.6E+01/ 2.9E+02/ 2.4E+03
	Relative contribution of fish $\text{consumption}_{\text{10 years}}$ (%)	0	0
	Relative contribution of crop, meat and dairy $\text{consumption}_{\text{10 years}}$ (%)	100	100
	Relative contribution of drinking $\text{water}_{\text{10 years}}$ (%)	0	0
	$\text{RCR}_{\text{year 1/10/100}}$	2.5E+01 2.1E+02 1.6E+03	1.0E+01/ 1.1E+02/ 9.4E+02

13.3.6.6 Source contribution analysis

Information on background concentrations in soils and waters for SCCPs and MCCPs is scarce (Halse et al., 2015; Glüge et al., 2018), and measured data may show a high degree of variability due to the lack of standardised measurement methods for these complex industrial mixtures and high total number of possible congeners therein. Given the importance of soil-derived food materials to total human intake, soil background concentration are central whereas background concentrations in water will not influence the results. For SCCPs, the model predicted no increase in $\mathrm{RCR_{soil}}$ or $\mathrm{RCR_{human}}$ compared to model results that only consider sewage sludge applications, whereas for MCCPs, the inclusion of soil background concentrations in the model resulted in marginally higher $\mathrm{RCR_{soil}}$ and $\mathrm{RCR_{human}}$ (Table 28). This is because the observed background concentrations in soils were more than order of magnitude higher for MCCPs than for SCCPs. Acknowledging the uncertainty to the dataset, the source contribution analysis should therefore be interpreted with the necessary degree of caution. Still, it could be observed that for both SCCPs and MCCPs, sludge was a much greater contributor to the total pollutant concentrations in soils than the background (Table 28). This suggests that sludge could be a main source of pollution for SCCPs and MCCPs.

Table 28: Environmental and human health risk screening assessment for short- and long-chain polychlorinated paraffins (SCCPs and MCCPs, respectively), based on its present background concentrations observed in the environment along with sewage sludge applications to agricultural land (PEC: predicted environmental concentration, RCR: risk characterisation ratios)

		SCCPs	**MCCPs**
Soil	$\mathrm{PEC_{year\ 1/10/100}}$ (mg kg^{-1} wet soil)	1.5E-01/ 1.6E+00/ 1.3E+01	1.8E-01/ 1.6E+00/ 1.3E+01
	Contribution of sludge to total concentration (%)	99/ 100/ 100	83/ 98/ 100
	$\mathrm{RCR_{year\ 1/10/100}}$ (-)	1.2E-01 1.3E+00 1.1E+01	1.5E-02/ 1.4E-01/ 1.1E+00
Surface water	$\mathrm{PEC_{year\ 1}}$ (µg L^{-1})		
	RCR $_{\mathrm{year\ 1}}$ (-)		
Human	Total daily intake$_{\mathrm{year\ 1/10/100}}$ (mg day^{-1})	4.0E+00 3.3E+01 2.6E+02	3.5E+01/ 2.9E+02/ 2.4E+03
	$\mathrm{RCR_{year\ 1/10/100}}$	2.5E+01 2.1E+02 1.6E+03	1.4E+01/ 1.2E+02/ 9.5E+02

13.3.6.7 Impact of sludge processing

With few studies that focused on SCCPs and MCCPs in sewage sludge, it is unexpected that little information is available on their removal potential through sludge processing. Microbial degradation processes, involving composting and anaerobic digestion, seem to be vary as a function of the individual susbtances that make up the chloroalkane mixtures, and the relative positions of the chlorine atoms (Heath et al., 2006). Specific aerobic microorganisms have been documented to remove until about 50% of the SCCPs in periods of weeks (Lu, 2013). Mo information could be retrieved on their degradation under anaerobic conditions during sludge digestion processes. Rigby et al. (2021) indicated generally low total polychlorinated alkane concentrations in incinerated waste materials, but it remains unclear if these are the direct result of the incineration process itself or their low occurrence in the feedstock applied. However, nearly complete dehydrochlorination processes in the temperature range 300–600°C have been reported for polychlorinated alkanes (Camino and Costa, 1980) as well as low to negligible concentrations in incinerations char and ash residues above 500°C

(Xin et al., 2018). These observations suggest that thermal oxidation is effective in removing the overall share of the polychlorinated alkanes present in sewage sludge.

13.3.7 Polychlorinated naphthalenes (PCNs)

13.3.7.1 Background and selection of compounds

Polychlorinated naphthalenes (PCNs) comprise of 75 possible congeners in eight homologue groups with one to eight chlorine atoms substituted around the planar aromatic naphthalene molecule. The basic structure of the PCNs has the molecular formula C10H8-nCln, where n=1-8 (UNEP, 2017b). Physical-chemical properties vary considerably due to the degree of chlorine substitution. The physical state ranges from thin liquids to hard waxes. Tri- through octa-CNs are very lipophilic with high log Kow (>5) and their water solubility and vapour pressure decrease with the degree of chlorination (UNEP, 2017b).

Polychlorinated naphthalenes (PCNs) have been commercially produced and used mainly in electrical devices, but also for impregnation of wood, paper, paints, and textiles to attain waterproofness, flame resistance and protection against insects, molds and fungi (Jakobsson and Asplund, 2000). Many of these intentional uses have ceased to take place due to increasing health concerns of PCNs (UNEP, 2017b). Polychlorinated naphthalenes are, however, also present and occurring unintentionally during industrial manufacturing processes (e.g. in technical PCB mixtures, the production of chlorinated solvents, and chlorine via chloralkali electrolysis). In addition, PCNs are formed together with PCDD/PCDFs in thermal processes such as incineration or metal industries (UNEP, 2017b).

13.3.7.2 Safe limit values for human consumption

In the framework of Articles 23 and 33 of Regulation (EC) No 178/2002 EFSA has received from the European Commission a mandate (M-2010-0374), EFSA is currently collecting data on PCNs EFSA's for scientific opinions and reports on pollutants in food and feed.

Li et al. (2020) proposed toxicity equivalence factors (TEFs) relative to 2,3,7,8-TCDD. The TEFs reported for certain hexa- and penta-CNs (0.002 – 0.004) are about 2-3 orders of magnitude higher for penta- and octa-CNs. Therefore, it is proposed (i) to apply the limit value for the related PCDD/F compounds of 2.9E-10 mg kg^{-1} body weight day^{-1}, and (ii) to focus the assessment on selected hexa- and penta-CNs (PCN-66/67 and PCN 73) that are known to mostly accumulate in the human body (Agunbiade et al., 2020).

13.3.7.3 Concentrations observed in sludge

Polychlorinated naphthalene concentrations in sludge have been reported by a limited set of studies (Harrison et al., 2006; Clarke and Smith, 2011; Rigby et al., 2021). The study of Rigby et al. (2021) provides likely the most comprehensive PCN dataset available. The average calculated values for three UK sewage sludge, expressed as toxicity equivalents, of 40 and 68 pg kg^{-1} dry matter, for respectively PCN-66/67 and PCN-73 were retained in this assessment.

13.3.7.4 Model input data

Table 29: Physico-chemical and toxicological data used for risk modelling purposes for the selected polychlorinated naphthalene congeners 667/67 (mixture of 1,2,3,4,6,7-HxCN and 1,2,3,5,6,7-HxCN) and 73 (1,2,3,4,5,6,7-HpCN).

	PCN 66/67	**PCN 73**
Concentration in sludge (mg^{-1} 2,3,7,8-TCDD toxicity equivalents kg^{-1} dry matter)	4.0E-8	6.8E-8

Molecular weight (g mol-1)	335	369.5
Water solubility (mg L^{-1})	0.11E-3	0.04E-3
Vapour pressure (Pa)	0.001	2.6E-4
Kow (log 10)	6.7 (1)	6.6 (1)
	(Puzyn and Falandysz, 2005)(Puzyn and Falandysz, 2007)(Chayawan and Vikas, 2015)	
Koc (L kg^{-1})	3.4E+05	2.8E+05
	(2)	
PNECaqua (µg L^{-1})	1.0E-03	1.0E-3
	(3)	
PNECsoil (mg kg^{-1} ww)	5.94E-03	4.93E-03
	(derived from PNECaqua)	
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	4.0E-09 for the sum of PCNs (WHO, 1998b)	
Half-life in soil (days)	3650	3650
	(4)	
Half-life in water (degradation, days)	263646	263646
	(default value for non-biodegradable substances)	

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(1) Congener-specific estimated Kow values, derived through the extrapolation of measured Kow values for other congeners, have been retained in this assessment.

(2) Based on EUSES default for hydrophobic substances: log Koc = 0.81 x log Kow + 0.1

(3) The ECOSAR-predicted chronic toxicity of hepta-CNs to fish and daphnids is in the low µg/L range (Environment Canada, 2011). An assessment factor of 1000 has been applied to derived the values presented.

(4) Järnberg et al. (1999) did not find any measurable change in the congener composition of tetra- to hexachlorinated naphthalenes in a 28-day aerobic degradation experiment. Half-lifes up to 10 years in sewage sludge amended soils were reported by Meijer et al. (2001).

(5) Meijer et al. (2001) analysed rural soils in the United Kingdom dating back to the 1940s. They found a peak level of 12 µg/kg dry weight in the 1960s, falling to 0.5–1 µg/kg in 1990.

13.3.7.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of PCN 66/67 and PCN-73 present in sewage sludge to agricultural soils (without ‘background’ concentrations of water and soils) indicated that risk characterisation ratios were well below 1 for soils, surface waters and humans after 10 years of sewage sludge application (Table 34. However, when considering a longer time frame of 100 years of application, the ratio was marginally above 1 (1.1E+00, Table 34). The total human daily intake after 10 years of consecutive sludge applications, expressed as PCDD TEQs, for the selected PCNs is about 3 orders of magnitude lower than for 2,3,7,8-TCDD (see section 13.3.3). Previous work measured low PCN concentrations in food (including fish, vegetables, meat and dairy products), at levels that do not not pose toxicological concerns (Fernandes et al., 2010). Hence, the incidence of a human health risks from PCNs in sewage sludge seems to a borderline case, and further detailed analysis will have to shed further light in view of ensuring health protection.

Table 30: Environmental and human health risk screening assessment of the exclusive application of the selected polychlorinated naphthalene (congeners 667/67 (mixture of 1,2,3,4,6,7-HxCN and 1,2,3,5,6,7-HxCN) and 73 (1,2,3,4,5,6,7-HpCN), respectively) present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		PCN 66/67	**PCN 73**	**SUM PCN-2**
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	5.9E-11	1.0E-10	
		4.6E-10	7.9E-10	
		9.0E-10	1.6E-09	
	RCR$_{\text{year 1/10/100}}$ (-)	9.9E-09	2.0E-08	
		7.8E-08	1.6E-07	
		1.5E-07	3.2E-07	
Surface water	PEC$_{\text{year 1}}$ (µg L^{-1})	3.7E-11	7.6E-11	
	RCR $_{\text{year 1}}$ (-)	3.7E-08	7.6E-08	
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	1.0E-09	1.6E-09	2.6E-09
		5.1E-09	6.6E-09	1.2E-08
		9.5E-09	1.2E-08	2.2E-08
	Relative contribution of fish consumption$_{\text{10 years}}$ (%)	8	13	
	Relative contribution of crop, meat and dairy consumption$_{\text{10 years}}$ (%)	92	87	
	Relative contribution of drinking water$_{\text{10 years}}$ (%)	0	0	
	RCR$_{\text{year 1/10/100}}$	5.1E-02	7.9E-02	1.3E-01
		2.6E-01	3.3E-01	5.9E-01
		4.8E-01	6.1E-01	1.1E+00

13.3.7.6 *Source contribution analysis*

No sludge contribution analysis could be performed as no reliable background concentrations in soils and waters could be retrieved. The most harmful PCN congeners; CNs 66, 67, 73, and 52 were mostly found in all their food samples (Fernandes et al., 2010). The dioxin-like toxicity (PCN TEQ) associated with these concentrations is 1–2 orders of magnitude lower than those reported for chlorinated dioxins or PCBs in food (Fernandes et al., 2010).

13.3.7.7 *Impact of sludge processing*

The impact of sludge processing options on PCNs has not been performed. Nonetheless, given the similarities in chemical characteristics with PCBs, a similar behaviour is expected during biological and thermal treatment processes.

13.3.8 Organotin compounds

13.3.8.1 Introduction and selection of compounds

Organotin compounds that are used in the preparation of polyvinylchloride (PVC) plastics. They have also been widely used as general biocides in paints, leather, textiles, the production of paper and as wood preservatives and slimicides. The most important source of exposure of the general population is from food (in particular fish and other seafood). Food contamination is primarily caused by the use of tri-substituted organotin compounds as biocides, components of antifouling paints and as agricultural pesticides. However, tri-substituted organostannic compounds have been banned for use as biocides under Directive 98/8/EC. Water basins, especially those with a low water exchange, may also accumulate OTC due to boating activities and agricultural runoff, with subsequent accumulation in the food chain.

The assessment of environmental and human health risks caused by the organotins is difficult. The large number of compounds with very different properties in the group makes it difficult to make general conclusions. Sludge concentration values have often been reported for dibutyltins (DBT), tributyltins (TBT), and triphenyltins (TPT), where these have been identified as compounds of concern (e.g. Clarke and Smith, 2011). Safe limit values for human consumption of these compounds is available, for which reason these compounds were taken forward.

13.3.8.2 Safe limits for human consumption

An EFSA assessment on organotin compounds focused on the most toxic organotin compounds: DBT, TBT, Dioctyltins (DOT), and TPT. A group TDI of 0.25 µg kg^{-1} body weight day^{-1} for DBT, TBT, DOT and TPT compounds was established by EFSA (EFSA, 2004). This value was used in this assessment, with the caveat that only three compounds were assumed to contribute to the group safe limit value for human consumption.

13.3.8.3 Sludge concentrations

The ranges reported from mono-, di and tributyltin by Voulvoulis et al. (2006) all had similar ranges from 0.3 to values above 8 mg kg^{-1} dry matter. Values for TPT documented in the review paper of Harrison et al. (2006) ranged from 0.3 to 3.4 mg kg^{-1} dry matter, whereas for TBT (up to 297 mg kg^{-1} dry matter) and DBT (up to 8.6 mg kg^{-1} dry matter) higher values were reported based on older studies.

Since 2010, EU restrictions for the use of certain organotin compounds is further extended to cover many consumer products (e.g. including clothes, hygiene products, PVCs) by Decision 2009/524/EC. Therefore, it can reasonably be assumed that organotin concentrations reported in sludge before 2010 may not be representative for current-day situations. This is partially confirmed by studies after that date that generally reported much lower values compared to previous periods. Wiechmann et al. (2013) indicated organotin concentrations below 1 mg kg^{-1} dry matter for DBT (0.22 mg kg^{-1} dry matter), TBT (0.03 mg kg^{-1} dry matter), and DOT (0.06 mg kg^{-1} dry matter). Similarly, Oloffson et al. (2012) indicated values for summed organotin compounds of 0.15 mg kg^{-1} dry matter. Mailler et al. (2014) reported values below 0.05 mg kg-1 dry matter for TPT and TBT, whereas DBT has maximum concentration values of about 0.3 mg kg-1 dry matter.

In this assessment, concentration values of 0.3 mg kg^{-1} dry matter were assumed for DBT, whereas for TBT, and TPT a value of 0.1 mg kg-1 dry matter was assumed. DOT is not considered in this assessment due because the single value reported for sewage sludge was very low. Hence, the assessment will only cover the joint contribution of the DBT, TBT, and TPT for the wider group of organotins.

13.3.8.4 Model input data

Table 31: Physico-chemical and toxicological data used for risk modelling purposes for the selected organotin compounds.

	Triphenyltin (TPT)	**Dibutyltin (DBT)**	**Tributyltin (TBT)**
Concentration in sludge (mg^{-1}	0.1	0.3	0.1

kg^{-1} dry matter)	(see main text)	(see main text)	(see main text)
Molecular weight (g mol-1)	350	249	280.9
		(van Herwijnen, 2012)	
Water solubility (mg L^{-1})	5	20	5.3
		(van Herwijnen, 2012)	
Vapour pressure (Pa)	0.8	0.16	48.5
		(van Herwijnen, 2012)	
Kow (log 10)	3.7	1.6	4.1
		(van Herwijnen, 2012)	
Koc (L kg^{-1})	5.0E+05	4.2E+04	3.2E+04
		(van Herwijnen, 2012)	
PNECaqua (µg L^{-1})	2.6E-02	1.9E-02	2.6E-02
		(Scientific Committee on Health and Environmental Risks, 2006)	
PNECsoil (mg kg^{-1} ww)	1.2E-01	1.9E-02	1.1E-02
		(equilibrium partitioning method)	
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})		0.25 µg kg-1 body weight day-1 for the sum of DBT, TBT, DOT and TPT (EFSA, 2004)	
Half-life in soil (days)	527292	120	527292
	(default value in the absence of biodegradation data)	(van Herwijnen, 2012)	(default value in the absence of biodegradation data)
Half-life in water (degradation, days)	263646	263646	263646
	(default value in the absence of biodegradation data)	(default value in the absence of biodegradation data)	(default value in the absence of biodegradation data)
Regional background in soil (mg kg^{-1} dry matter)(2)	1.0E-02	3.0E-02	1.0E-02
	(Sternbeck et al., 2006)	(Sternbeck et al., 2006)	(Sternbeck et al., 2006)
Regional background in surface water (µg L^{-1})	1.0E-02	5.0E-02	1.0E-01
	(Sternbeck et al., 2006)	(Sternbeck et al., 2006; Cavalheiro et al., 2016)	(Sternbeck et al., 2006; Cavalheiro et al., 2016)

(1)There are no measured data available for the physical and chemical properties for all these substances. Physico-chemical (e.g. Kow) used in this assessment are average values for different compounds that fall under the respective organotin group (averages for bis(tributyltin)oxide, tributyltin chloride, and tributyltin hydride for organotin referred to as tributyltin).

13.3.8.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of organotins present in sewage sludge to agricultural soils (without 'background' concentrations of water and soils) indicated that risk characterisation ratios were well below 1 for soils and humans (maximum up of 6.6E-01 for RCR$_{soil\ year\ 100}$ for TBT, 1E-03 for RCR$_{human\ 100\ year}$) (Table 32. This high differences compared to the step 1 assessment is due to the reduced sludge concentration values that was observed in recent literature (0.1 mg kg-1 versus a value > 100 mg kg-1 applied in the Step 1 assessment).

Table 32: Environmental and human health risk screening assessment of the exclusive application of the organotins Triphenyltin (TPT), Dibutyltin (DBT), Tributyltin (TBT) present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		Triphenyltin (TPT)	Dibutyltin (DBT)	Tributyltin (TBT)	
Soil	PEC$_{year\ 1/10/100}$ (mg kg^{-1} wet soil)	1.5E-04/	4.2E-04/	1.5E-04/	
		1.6E-03/	6.2E-04/	1.5E-03/	
		1.3E-02	7.5E-04	9.5E-03	
	RCR$_{year\ 1/10/100}$ (-)	6.4E-04/	3.0E-02/	1.0E-02/	
		6.9E-03/	4.4E-02/	1.1E-01/	
		5.7E-02	5.4E-02	6.6E-01	
Surface water	PEC$_{year\ 1}$ (µg L^{-1})	6.2E-05	2.2E-03	9.7E-04	
	RCR $_{year\ 1}$ (-)	2.4E-03	1.1E-01	3.7E-02	
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	4.7E-07/	4.5E-07/	1.4E-05/	1.5E-05/
		5.9E-07/	4.6E-07/	1.4E-05/	1.5E-05/
		1.6E-06	4.7E-07	1.6E-05	1.8E-05
	Relative contribution of fish consumption$_{10\ years}$ (%)	76%	50%	97%	
	Relative contribution of crop, meat and dairy consumption$_{10\ years}$ (%)	23%	9%	2%	
	Relative contribution of drinking water$_{10\ years}$ (%)	1%	41%	1%	
	RCR$_{year\ 1/10/100}$				8.5E-04
					8.7E-04
					1.0E-03

13.3.8.6 Source contribution analysis

A source contribution analysis has not been performed due to the low RCR observed from the application of organotins present in sewage sludge to agricultural soils.

13.3.8.7 Impact of sludge processing

No assessment has been undertaken to assess the impact of sludge processing given to the low RCR observed from the application of organotins present in sewage sludge to agricultural soils.

13.3.9 Phthalate acid esters

13.3.9.1 Background

Phthalate esters are colorless sticky liquids and widely used as important additives which impart flexibility in polyvinylchloride resins. Phthalate esters have been widely used in plastic manufacturing since the 1930s, and they can also be found as a common additive in paints, lubricants, adhesives, insecticides, packaging, and cosmetics (Gao and Wen, 2016). Phthalate esters are also gradually released from industrial products during manufacturing, storage, use, and disposal (Gao and Wen, 2016). Degradation rates of these pollutants are very slow under natural conditions

The EU has restricted six phthalates in toys and childcare articles since 1999. EU Regulation (EU) 2018/2005 further expands the restrictions for phthalates that apply as per Annex XVII to Regulation (EC) No 1907/2006 to any products placed on the market, and further tightens the identity of phtalates that should meet the limit value of 0.1%.

13.3.9.2 Safe limit values for human consumption

The EFSA Panel on Food Contact Materials, Enzymes and Processing Aids (CEP) confirmed the tolerable daily intake values of 0.01, 0.5 and 0.05 mg kg^{-1} dry sludge based on reproductive effects for DBP, BBP, and DEHP, respectively (EFSA CEP Panel, 2019). In addition, the EFSA Panel indicated a group TDI for four phtalates di-butylphthalate (DBP), butylbenzylphthalate (BBP), bis(2-ethylhexyl)phthalate (DEHP), and di-isononylphthalate (DINP) of 50 µg kg^{-1} bw day^{-1}, expressed as DEHP equivalents (EFSA CEP Panel, 2019). The relative potency factors (RPFs) are 1 for DEHP, 5 for DBP, 0.1 for BBP and 0.3 for DINP when including the additional assessment factor of 3.3.

13.3.9.3 Sludge concentrations

For DEHP, Lamastra et al. (2018) indicated DEHP concentrations varying from 1.32 to 103 mg kg^{-1} DM (median 6.24 mg kg^{-1} DM) and from 0.602 to 148 mg kg^{-1} DM (median 11.1 mg kg^{-1} DM), respectively. Fromme et al. (2002) reported median DEHP values of 67.3 mg kg^{-1} DM. These values are slightly lower than the values reported by Tavazzi et al. (2012). Lithuanian sewage sludge samples collected in 2019 showed DEHP values of 3.8 – 95 mg kg^{-1} DM (Anne and Paulauskiene, 2021). Based on these data, a value of 50 mg kg^{-1} DM was applied in this assessment. DINP concentrations reported by Rigby et al. (2021) are around 100 mg kg^{-1} DM. Concentrations for DBP and BBP were much lower, with average values of around 1 mg kg^{-1} dry weight (VKI, 1996; Boutrup et al., 1998; Fromme et al., 2002; European Chemicals Bureau, 2007; Dargnat et al., 2009). Therefore, only DEHP and DINP were retained in this assessment, with concentration values of 50 mg kg^{-1} dry matter and 100 mg kg^{-1} dry matter, corresponding to 50 mg kg-1 and 30 mg kg-1 dry matter DEHP Equivalents.

13.3.9.4 Model input data

Table 33: Physico-chemical and toxicological data used for risk modelling purposes for bis(2-ethylhexyl)phthalate (DEHP), and di-isononylphthalate (DINP).

	DEHP	**DINP**
Concentration in sludge (mg^{-1} DEHP equivalents kg^{-1} dry matter)	50	30
Molecular weight (g mol-1)	390.56	420.6
Water solubility (mg L^{-1})	3.0E-03	6.E-04
	(Gao and Wen, 2016)	(Gao and Wen, 2016)
Vapour pressure (Pa)	3.4E-05	6.0E-05
	(ECHA, 2008b)	(INERIS, 2003)
Kow (log 10)	7.5	8.8
	(ECHA, 2008b; Gao and Wen, 2016)	(INERIS, 2003)

Koc (L kg^{-1})	1.65E+05	3.1E+05
	(ECHA, 2008b)	(INERIS, 2003)
PNECaqua (µg L^{-1})	Not available(1)	Not available(1)
	(ECHA, 2008b)	(INERIS, 2003)
PNECsoil (mg kg^{-1} ww)	13	30
	(ECHA, 2008b)	(INERIS, 2003)
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	0.05 mg s DEHP Equivalents kg^{-1} body weight day^{-1}	
	(EFSA CEP Panel, 2019)	
Half-life in soil (days)	147	300
	(Roslev et al., 1998)	(INERIS, 2003; Gao and Wen, 2016)
Half-life in water (degradation, days)	14	50
	(Gao and Wen, 2016)	(INERIS, 2003; Gao and Wen, 2016)
Regional background in soil (mg kg^{-1} dry matter)(2)	0.02	0.02
	(Gawlik and Bidogli, 2006)	(Gawlik and Bidogli, 2006)
Regional background in surface water (µg L^{-1})	Not considered	Not considered
		(4)
BCFfish-water (L kg^{-1}ww)	840	4000
	(ECHA, 2008b)	(ECHA, 2008b)
BCFshoot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	1.00E+00(2)	1.00E+00(3)
	(ECHA, 2008b)	(4)
BTFroot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	1.00E+00(2)	1.00E+00(3)
	(ECHA, 2008b)	(4)
BTFmeat-grass (day kg-1 dry matter)	9.95-01	9.95E-01
	(3)	(4)
BTF-milk-grass (day kg^{-1} dry matter)	3.15E-01	3.15E-01
	(3)	(4)

---

(1) INERIS (2003) concluded that DINP does not have adverse effects towards aquatic or benthic organisms at the limit of water solubility in laboratory tests, no PNECs could be derived.

(2) Based on BCF values of 9.2 (shoot-soil) and 12.4 (root-soil) as reported by EFSA (ECHA, 2008b). Values expressed on a dry basis have been transformed into values on a wet mass basis (mg kg-1 plant ww)/(mg kg-1 soil ww) assuming a water content in soil of 20vol% (EUSES default, conversion factor for soil concentration wet-dry weight soil of 1.13), and a water content in roots of 65vol%, and a bulk density of plants of 700 kg/m3 (EUSES default),

(3)EPI data obtained from the Risk Assessment Information System, University of Tennessee (https://rais.ornl.gov/cgi-bin/tools/TOX_search?select=chemspef).
(4)DINP-specific data could not be retrieved. In the absence of available data for DINP, the DEHP data were used an an approximation.

13.3.9.5 *Estimated environmental and health exposure and risks from sludge application*

The outcome of the risk screening assessment resulting from the application of phthalate esters present in sewage sludge to agricultural soils (without ‘background’ concentrations of water and soils) indicated that risk characterisation ratios were well below 1 for soils, but above 1 for humans (Table 34). Values for RCRhuman after 10 years were for instance estimated at a value of 1.4E+02. In addition, it is noted that risks may be underestimated due to lack of bioconcentration data through plants available for DINP. In this exercise, the data available for DEHP were applied, but bioconcentration factors for DINP may be higher due to increased Kow observed for this compound. Crops, meat and dairy were the dominant food sources contributing to the the total human intake (Table 34).

Table 34: Environmental and human health risk screening assessment of the exclusive application of bis(2-ethylhexyl)phthalate (DEHP), and di-isononylphthalate (DINP) present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		DEHP	DINP	ΣDEHP + DINP
Soil	PEC$_{year\ 1/10/100}$ (mg DEHP equivalents kg^{-1} wet soil)	7.1E-02	4.3E-02	
		1.2E-01	1.1E-01	
		1.4E-01	1.4E-01	
	RCR$_{year\ 1/10/100}$ (-)	5.9E-03	1.4E-03	
		9.8E-03	3.7E-03	
		1.2E-02	4.6E-03	
Surface water	PEC$_{year\ 1}$ (µg L^{-1})	9.3E-02	3.0E-02	
	RCR $_{year\ 1}$ (-)	Not applicable	Not applicable	
Human	Total daily intake$_{year\ 1/10/100}$ (mg day^{-1})	1.5E+00	1.0E+00	2.5E+00
		2.5E+00	2.6E+00	5.0E+00
		3.1E+00	3.2E+00	6.3E+00
	Relative contribution of fish consumption$_{10\ years}$ (%)	0	0	
	Relative contribution of crop, meat and dairy consumption$_{10\ years}$ (%)	0	100	
	Relative contribution of drinking water$_{10\ years}$ (%)	0	0	
	RCR$_{year\ 1/10/100}$			7.2E-01
				1.4E+00
				1.8E+00

13.3.9.6 *Source contribution analysis*

The source contribution analysis indicated that sludge can be a main contributor the observed phthalate ester concentrations in soils, even shortly after application (Table 35). RCR$_{human}$ values further increased with values >1.0E+00 now also being observed after a single sludge application (year 1).

Table 35: Environmental and human health risk screening assessment for bis(2-ethylhexyl)phthalate (DEHP), and di-isononylphthalate (DINP) based on its present background concentrations observed in the environment along with sewage sludge applications to agricultural land (PEC: predicted environmental concentration, RCR: risk characterisation ratios)

		DEHP	DINP	ΣDEHP + DINP
Soil	PEC$_{\text{year 1/10/100}}$ (mg kg^{-1} wet soil)	9.1E-02	6.3E-02	
		1.4E-01	1.3E-01	
		1.6E-01	1.6E-01	
	Contribution of sludge to total concentration (%)	78/ 85/ 88	68/ 85/ 87	
	RCR$_{\text{year 1/10/100}}$ (-)	7.6E-03	2.1E-03	
		1.1E-02	4.4E-03	
		1.4E-02	5.3E-03	
Surface water	PEC$_{\text{year 1}}$ (µg L^{-1})	1.4E-01	8.0E-02	
	RCR $_{\text{year 1}}$ (-)	Not applicable	Not applicable	
Human	Total daily intake$_{\text{year 1/10/100}}$ (mg day^{-1})	2.0E+00	1.6E+00	3.6E+00
		3.0E+00	3.1E+00	6.1E+00
		3.6E+00	3.7E+00	7.3E+00
	RCR$_{\text{year 1/10/100}}$			1.0E+00
				1.7E+00
				2.1E+00

13.3.9.7 Impact of sludge processing

Anaerobic digestion has been shown to cause the partial removal of phthalate acid esters (Gavala et al., 2003; Fountoulakis et al., 2006). Phthalate degradation under aerobic conditions is much higher. Marttinen et al. (2003) observed that 32% of DEHP in sewage was removed during anaerobic digestion of the sludge. Sludge composting could significantly decrease the levels of the pollutants, with removal percentages ranging from 32% to 91% (Lü et al., 2021). For DEHP, removal percentages varied between < 50% to 99% have been observed (Lü et al., 2021). Thermal oxidation results in close to a 100% loss of phthalates at temperatures of around 400-500°C (Aouachira et al., 2014).

13.3.10 Polydimethylsiloxanes

13.3.10.1 Background

Polydimethylsiloxanes are are a group of organosilicon substances that are used in personal care products such as shampoos, cosmetics, and deodorants and in industrial applications such as dry-cleaning solvents and industrial cleaning fluids (Mackay et al., 2015). Following the

13.3.10.2 Safe limit values for human consumption

A NOAEL for increased liver weight of 25 mg kg bw^{-1} day^{-1} were reported for female rats (SCCS, 2010). Therefore, a human toxicological standard was calculated using this lowest NOAEL and an assessment factor of 100, giving a safe limit value for human consumption of 0.25 mg kg^{-1} body weight day^{-1} (Sahlin and Agerstrand, 2018).

13.3.10.3 Concentrations observed in sludge

Few studies have reported on polydimethylsiloxanes in sewage sludge. Tavazzi et al. (2012) indicated that decamethylcyclopentasiloxane (D5) was the dominant siloxane observed in sewage sludge. These results are aligned to 2004 monitoring data from the Swedish national screening programme (Kaj et al., 2005). In the absence of any further data, the D5 concentration value of 10.8 mg kg^{-1} dry matter is retained in this assessment.

The European Union's restriction on the use of octamethylcyclotetrasiloxane (D4) and decamethylcyclopentasiloxane (D5) in rinse-off cosmetic products entered into force on 1 February 2020. Under the REACH Regulation, companies can no longer add the siloxanes in a concentration equal to or greater than 0.1% by weight of the product. It is expected that this measure may result in substantial decreases in D5 concentrations in sewage sludges.

13.3.10.4Model input data

Table 36: Physico-chemical and toxicological data used for risk modelling purposes for the selected polydimethylsiloxanes.

	decamethylcyclopentasiloxane (D5)
Concentration in sludge (mg^{-1} kg^{-1} dry matter)	10.8
Molecular weight (g mol-1)	370.8 (Mackay et al., 2015)
Water solubility (mg L^{-1})	1.7E-02 (Mackay et al., 2015)
Vapour pressure (Pa)	22.7 (Mackay et al., 2015)
Kow (log 10)	8.09 (Mackay et al., 2015)
Koc (L kg^{-1})	5.17 (Mackay et al., 2015)
PNECaqua (µg L^{-1})	0.044 (Sahlin and Agerstrand, 2018)
PNECsoil (mg kg^{-1} ww)	1.15E-01
Safe limit values for human intake (mg kg^{-1} body weight day^{-1})	0.25 (SCCS, 2010)
Half-life in soil (days)	12.6 (Mackay et al., 2015)
Half-life in water (degradation, days)	70.4 (Mackay et al., 2015)

Regional background in soil (mg kg^{-1} dry matter)(2)	3.5E-2 (Sánchez-Brunete et al., 2010)
Regional background in surface water (µg L^{-1})	- (Kaj et al., 2005)
BCFfish-water (L kg^{-1}ww)	4920 (Gobas et al., 2015a)
BCFshoot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	8.07E-02 (1)
BCFroot-soil (mg kg^{-1} plant ww)/(mg kg^{-1} soil ww)(3)	8.07E-02 (1)
BTFmeat-grass (day kg-1 dry matter)	3.09E+00 (2)
BTF-milk-grass (day kg^{-1} dw)	9.77E-01 (2)

(1)Gobas et al. (2015a), Gobas et al. (2015b) and Fairbrother et al. (2015) reviewed in detail the potential for D5 to bioaccumulate or biomagnify in aquatic or terrestrial food webs and concluded that the risk of biomagnification was negligible. Briefly, D5 is a very hydrophobic organic substance with a low affinity for organic carbon relative to its Kow. In terrestrial ecosystems, it is likely that the high rate of loss of D5 via exhalation (as a result of its relatively low octanol-air partition coefficient) and rapid metabolism ensure that D5 depuration rates exceed dietary uptake rates. As such, biomagnification in terrestrial food webs does not occur. Therefore, the values applied for BCFshoot-soil and BCFroot-soil were set to 1 on a dry weight basis. Values expressed on a dry basis have been transformed into values on a wet mass basis (mg kg-1 plant ww)/(mg kg-1 soil ww) assuming a water content in soil of 20vol% (EUSES default, conversion factor for soil concentration wet-dry weight soil of 1.13), and a water content in roots of 65vol%, and a bulk density of plants of 700 kg/m3 (EUSES default).

(2)Default values based on Kow, likely being an overestimation due to the low bioaccumulation potential of D5.

13.3.10.5 Estimated environmental and health exposure and risks from sludge application

The outcome of the risk screening assessment resulting from the application of D5 present in sewage sludge to agricultural soils (without ‘background’ concentrations of water and soils) indicated that risk characterisation ratios were well below 1 for soils, surface waters and humans (Table 34).

Table 37: Environmental and human health risk screening assessment of the exclusive application of decamethylcyclopentasiloxane (D5) present in sewage sludge to agricultural soils (PEC: predicted environmental concentration, RCR: risk characterisation ratio)

		decamethylcyclopentasiloxane (D5)
Soil	PEC$_{year\ 1/10/100}$ (mg DEHP equivalents kg^{-1} wet soil)	7.8E-03/ 7.8E-03/ 7.8E-03
	RCR$_{year\ 1/10/100}$ (-)	6.8E-02/ 6.8E-02/ 6.8E-02

Surface water	$PEC_{year\ 1}$ (µg L^{-1})	1.8E-02
	$RCR_{year\ 1}$ (-)	4.1E-01
Human	Total daily $intake_{year\ 1/10/100}$ (mg day^{-1})	2.3E-02/ 2.3E-02/ 2.3E-02
	Relative contribution of fish $consumption_{10\ years}$ (%)	33
	Relative contribution of crop, meat and dairy $consumption_{10\ years}$ (%)	67
	Relative contribution of drinking $water_{10\ years}$ (%)	0
	$RCR_{year\ 1/10/100}$	1.3E-03/ 1.3E-03/ 1.3E-03

13.3.10.6 Source contribution analysis

A source contribution analysis has not been performed due to the low RCR observed from the application of D5 present in sewage sludge to agricultural soils.

13.3.10.7 Impact of sludge processing

No assessment has been undertaken to assess the impact of sludge processing given to the low RCR observed from the application of organotins present in sewage sludge to agricultural soils.

13.4 Supplementary information - measurement standards for priority pollutants

The available measurement standards (and technical specifications, and technical reports) that may be relevant for the determination of priority pollutants in sewage sludge were searched in both databases: the European Standard (EN) database and the International Organization for Standardization (ISO) database. Standards containing the word '*sludge*' or '*biosolid*' were extracted. In addition, a targeted search, looking for priority pollutant in the standards' title, was also performed. When no relevant standards were found for the determination of a priority pollutant in sludge, the relevant standard for the determination of priority pollutant in *soil* was also considered. Standards for the characterisation of sludge properties or the detection/determination of other pollutants not reported as priority in this report, such as trace elements or metals, nutrients, other pollutants, were not considered for the identification of relevant standards.

The list of priority pollutants used for this exercise was based on the list presented in section 3.

- polycyclic aromatic hydrocarbons (PAHs);
- dioxins, furans and dioxin-like polychlorinated biphenyls (PCDD, PCDFs and dl-PCBs);
- per- and polyfluoroalkyl substances (PFAS);
- alkylphenols, nonylphenols, octylphenols (APs, NPs and Ops); ①
- short- and medium-chain polychlorinated alkanes (SCCPs and MCCPs);
- polychlorinated naphthalenes (PCNs);
- phthalate acid esters (DEHP and DINP);
- benzalkonium chloride and its degradation products (quaternary ammonium compounds);
- lauryl diethanolamide (N-acyl amines and fatty amides); and ②
- traseolide (synthetic musks).

① Identified as priority after human health risk assessment (see section 3.3)

② Identified as priority after soil organisms risk assessment (see Section 3.4)

The results of the identified available measurement standards are presented in Section 3.5.

In addition, other standards covering sampling, sample preparation or the leaching properties may also be relevant, as for example:

- ‘Sludge, treated bio-waste and soil - Guidance for sample pretreatment’ (EN 16179:2012);
- ‘Characterization of sludges - Protocol for preparing synthetic suspensions’ (CEN/TR 16394:2014);
- ‘Characterization of sludges - Laboratory chemical conditioning procedure’ (EN 14742:2015);
- ‘Sludge recovery, recycling, treatment and disposal — Laboratory chemical conditioning procedure’ (ISO/DTR 23594l)
- ‘Water quality - Sampling - Part 1: Guidance on the design of sampling programmes and sampling techniques’ (EN ISO 5667-1:2006);
- ‘Water quality - Sampling - Part 13: Guidance on sampling of sludges’ (EN ISO 5667-13:2011);
- ‘Water quality - Sampling - Part 15: Guidance on the preservation and handling of sludge and sediment samples’ (EN ISO 5667-15:2009);
- ‘Characterisation of waste - Leaching - Compliance test for leaching of granular waste materials and sludges - Part 1: One stage batch test at a liquid to solid ratio of 2 l/kg for materials with high solid content and with particle size below 4 mm (without or with size reduction)’ (EN 12457-1:2002);
- ‘Characterisation of waste - Leaching - Compliance test for leaching of granular waste materials and sludges - Part 2: One stage batch test at a liquid to solid ratio of 10 l/kg for materials with high solid content and with particle size below 4 mm (without or with size reduction)’ (EN 12457-2:2002);
- ‘Characterisation of waste - Leaching - Compliance test for leaching of granular waste materials and sludges - Part 3: Two stage batch test at a liquid to solid ratio of 2 l/kg and 8 l/kg for materials with high solid content and with particle size below 4 mm (without or with size reduction)’ (EN 12457-3:2002); and
- ‘Characterisation of waste - Leaching - Compliance test for leaching of granular waste materials and sludges - Part 4: One stage batch test at a liquid to solid ratio of 10 l/kg for materials with particle size below 10 mm (without or with size reduction)’ (EN 12457-4:2002).

Finally, other standards on the management of sewage sludge and use in agriculture may also be of interest, but these were not considered as measurement standards, as for example:

- ‘Characterization of sludges - Good practice for utilisation in agriculture’ (CR 13097:2001);
- ‘Characterization of sludges - Sludge management in relation to use or disposal’ (CEN/TS 13714:2013);
- ‘Sludge recovery, recycling, treatment and disposal — Beneficial use of biosolids — Land application’ (ISO 19698:2020); and
- ‘Characterisation of sludges - Guide to risk assessment especially in relation to use and disposal of sludges’ (CEN/TR 15584:2007).

14 Supplementary Information – soil biogeochemical modelling

14.1 Model approach

The JRC has developed a state-of-the-art process-based European biogeochemical modelling platform that simulates carbon (C) and nitrogen (N) flows within soil and between soil, the atmosphere and vegetation.

Key sub-models include decomposition of organic input and soil organic matter, mineralisation of nutrients, N gas emissions from nitrification and denitrification, soil water content and temperature by layer, plant production and allocation of net primary production (NPP) and CH4 oxidation in non-saturated soils and CH4 production is flooded soils. Flows of C and N between the different soil organic matter pools are controlled by the size of the pools, C/N ratio and lignin content of material, and abiotic water/temperature factors. Plant production is a function of genetic potential, phenology, nutrient availability, water/temperature stress, and solar radiation. NPP is allocated to plant components (e.g. roots vs. shoots) based on vegetation type, phenology, and water/nutrient stress. Nutrient concentrations of plant components vary within specified limits, depending on vegetation type, and nutrient availability relative to plant demand. Decomposition of litter and soil organic matter and nutrient mineralization are functions of substrate availability, substrate quality (lignin %, C/N ratio), and water/temperature stress. N gas fluxes from nitrification and denitrification are driven by soil NH4 and NO3 concentrations, water content, temperature, texture, and labile C availability (Parton et al., 2001).

In this project, DayCent is run on a 1 km2 grid using the following data (Figure 16):

- soil properties available for ESDAC and derived from spatial interpolation of LUCAS soils (https://esdac.jrc.ec.europa.eu/);
- land cover from the CORINE LAND COVER 1990, 2000, 2006, 2012;
- official statistics (EUROSTAT, FAO, Farm Structure Survey) and spatial datasets, which were used to describe the current management (i.e. crop rotation, mineral and organic N fertilization, tillage, irrigation, cover crop, etc.);
- meteorological data from the E-OBS gridded dataset (http://www.ecad.eu). The dataset provided daily data of maximum and minimum temperature and precipitation on a grid of 0.1° resolution (v22). For the climatic projection, we used the general circulation model CNRM-CM541 run with a RCP4 and downscaled with the RCM CCLM4-8-17, available at the WCR-CORDEX portal.

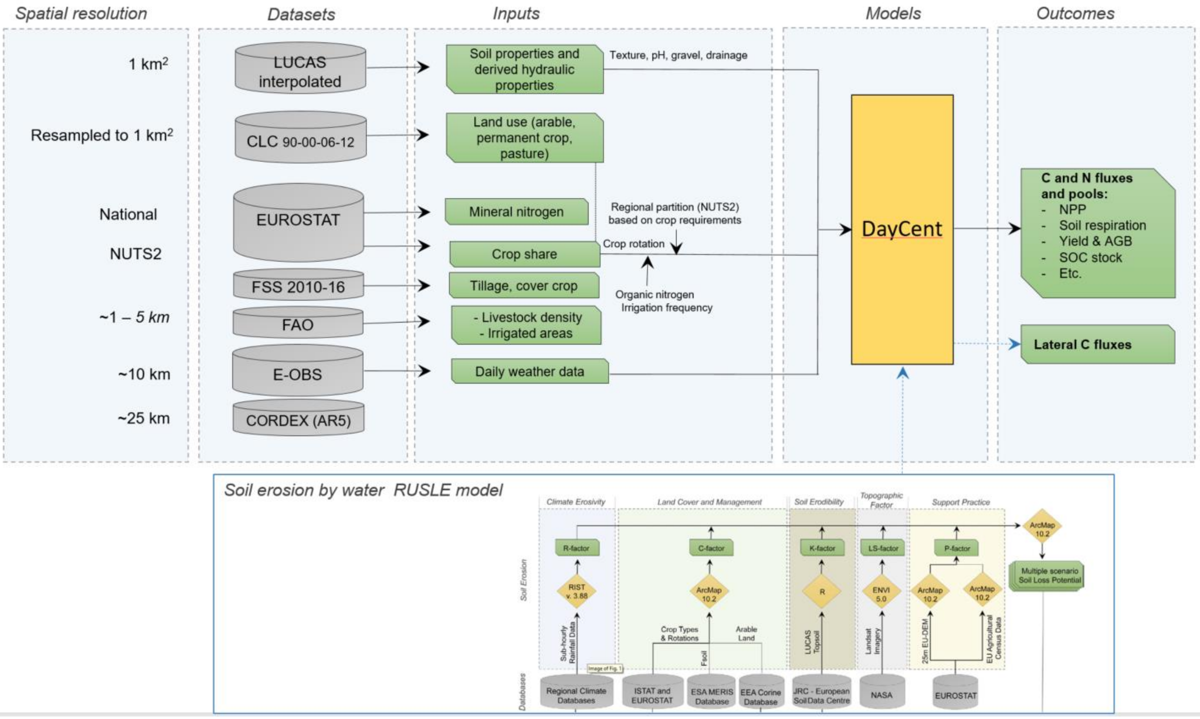

Figure 16: Flow chart showing the datasets utilized and their spatial resolution, the inputs derived and the model integration.

As **inputs**, the amount and timing of nutrient amenDMents is required. The current (baseline) N fertilization was characterised as follow (Figure 17):

- Mineral N fertiliser: it was partitioned in two applications at planting (30%) and standing crops (70%). In each fertilisation the proportion of NH_4 and NO_3 was assumed to be equal to 75 and 25%, respectively;
- Organic: applied generally after harvest or during standing crop in highly demanding crops such as maize. The territorial rates calculated was limited to the maximum rate of 170 kg/ha of N per year.

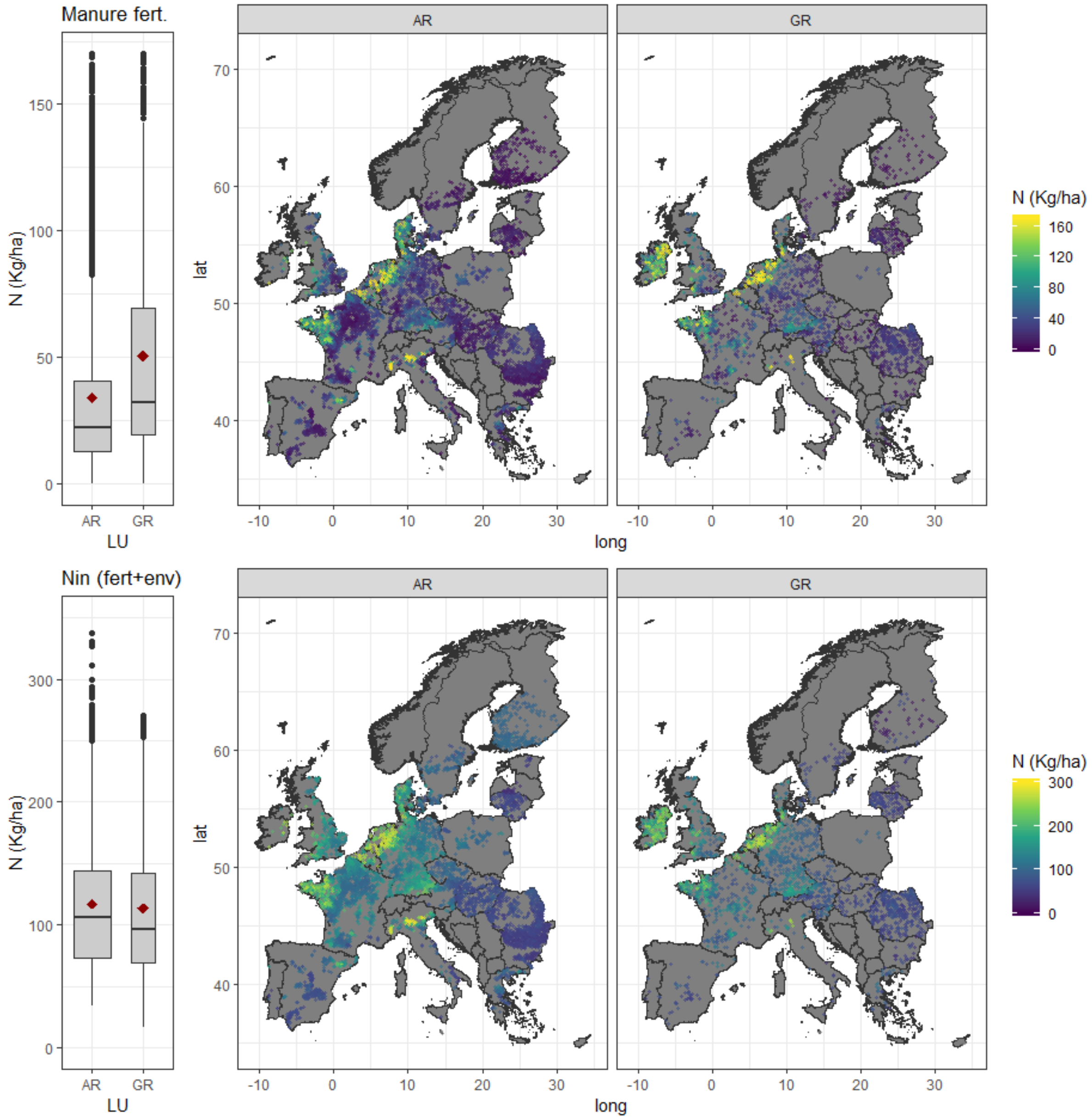

Figure 17: Organic (above) and total N input (below) rates in the baseline. The boxplots represents the values distribution (median and interquartile ranges) of all simulated points with the average in red diamond symbols.

Model **outputs** include daily N fluxes (N_2O, NOx, N_2, NO_3^- leaching), CO_2 flux from heterotrophic soil respiration, soil organic C, NPP (portioned into residues, grains and harvested root crops). The model takes into account land management and cropping practices. As it is driven by a range of climate scenarios, as simulated by Global Climate Models, the model can provide long-term policy perspectives.

The **ability** of DAYCENT to simulate NPP, soil organic carbon, N_2O emissions, and NO_3^- leaching has been tested with data from various native and managed systems (e.g. Del Grosso et al., 2001, 2006). The DAYCENT model is currently being used by the United States Environmental Protection Agency, United States Department of Agriculture and Colorado State University to develop a national inventory of N_2O emissions from U.S. agricultural soils. This inventory will be compared and contrasted with the existing Intergovernmental Panel on Climate Change (IPCC) agricultural N_2O emissions inventory for the USA.

The JRC has developed and continuously improved the modelling framework in the EU in the last decade, initially using CENTURY (the model monthly version) and then DayCent (the model daily version), running both at LUCAS point and gridded 1 km level. This framework was used for many scientific studies and policy scenarios, receiving a scientific recognition. For more information on the general architecture, model performances and different scenarios and agricultural management simulated, we refer to previous publications (Lugato et al., 2014b, 2018; Lugato and Jones, 2015; Borrelli et al., 2016; Scarlat et al., 2019; Quemada et al., 2020).

14.2 Results

14.2.1 Spatially explicit CH emission from the use-on-land of sludge

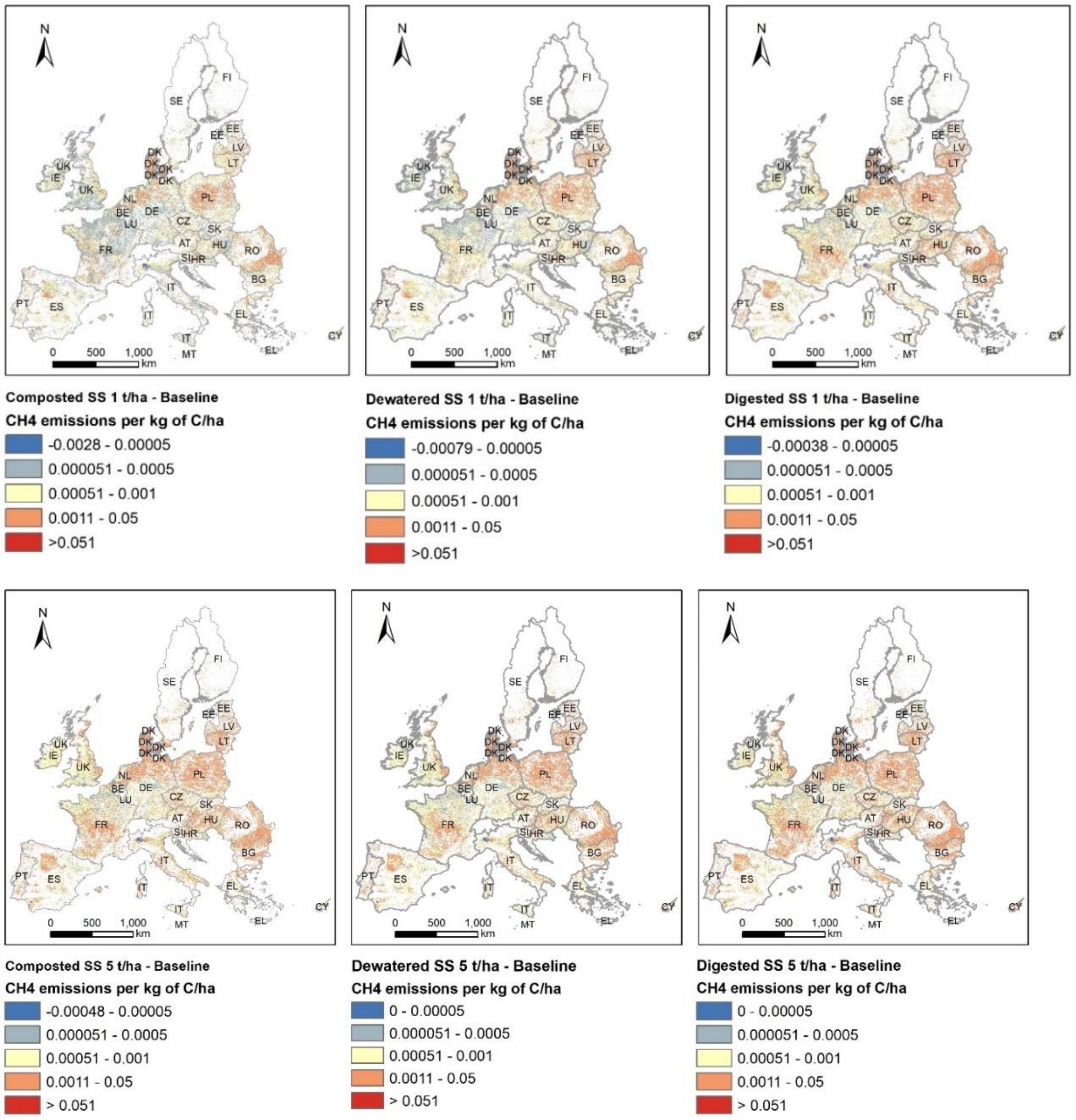

Figure 18: Spatially explicit CH_4 emission factors (kg CH_4 per kg C in (treated) sludge) as estimated using biogeochemical modelling for application scenarios of 1 tonne ha^{-1} yr^{-1} (top row) and 5 tonnes ha^{-1} yr^{-1} (bottom row) for composted sewage sludge (left hand side), dewater sewage sludge (middle), and digested sludge (right hand side).

14.2.2 Carbon sequestration in soils as a function of time for soils in the EU Member States

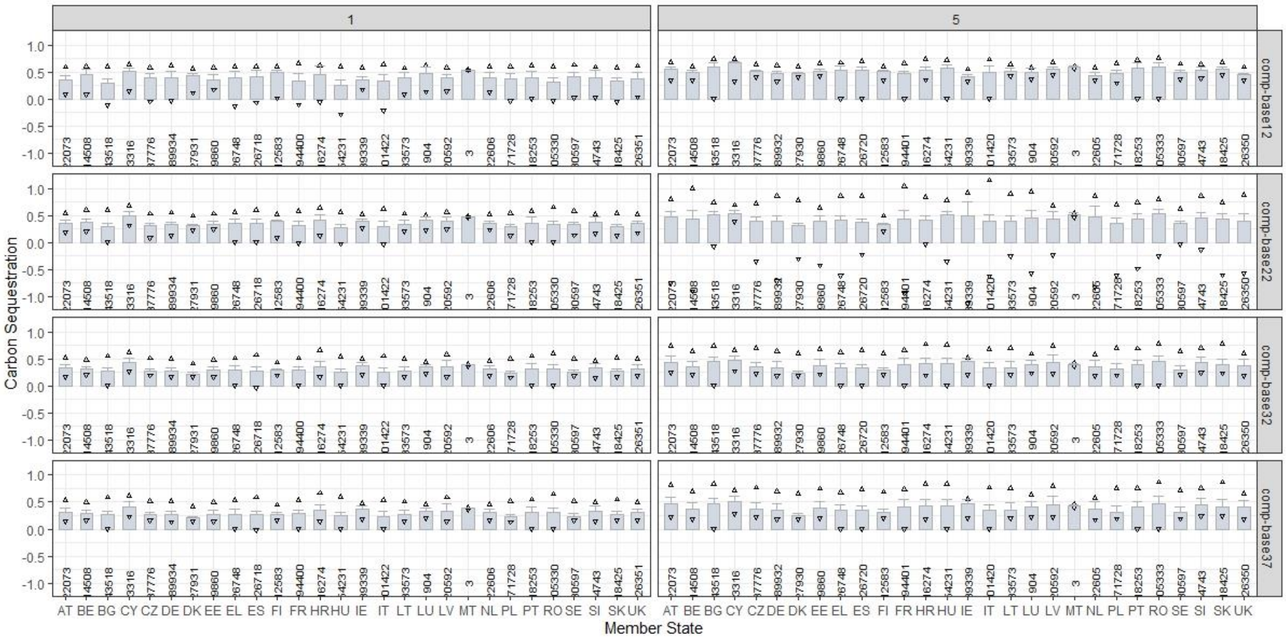

Figure 19: Carbon sequestration (12, 22, 32, 37 years after distribution) in Member States for the scenarios of 1 tonne ha^{-1} y^{r-1} (left panels) and, 1 tonne ha^{-1} y^{r-1} (right panels), Composted SS (comp) – baseline (base) scenario showed a decrease over time of the C sequestration throughout the time.

15 Supplementary information – life cycle assessment

15.1 Model input parameters

Table 38: Default parameter values and ranges applied for the uncertainty analysis of the life cycle assessment presented in section 6.4.

Parameters that are common to all pathways				
Electricity mix	Average EU mix	High share of renewables in the mix: 73%	Low share of renewables in the mix: 15%	
Incineration				
Dry matter content of sludge prior to incineration	24%	75%	18%	expert knowledge and consultations
Lower heating value of de-watered sludge (MJ/kg TS)	22.4	20%	-20%	
N_2O emissions – mono-incineration (mg N_2O /m3)	200	50	400	(IGES for the IPPC, 2002; Korving et al., 2010; Neuwahl et al., 2019)
Anaerobic digestion				
Biogas leak (%)	2.8	0.1	6	(IAEA Bioenergy Task, 2017)
CH_4 in biogas (% vol.)	63%	70%	50%	(Liu et al., 2018)
Electrical efficiency AD	30%	Default x 1.2	Default x 0.8	
Biogas yield (%)	70%	Default x 1.2	Default x 0.8	
Composting and liming				
N_2O emissions during composting (% N transformed)	1.4	Default x 1.3	Default x 0.7	
Storage and transport				
CH_4 emissions storage - compost (kg/kg total C)	0.003	0.001	0.01	(Willén et al., 2016; Samuelsson et al., 2018)
CH_4 emissions storage - sludge and digestate (kg/kg total C)	0.01	0.002	0.02	
N_2O emissions storage (kg/kg total N)	0.005	0	0.013	
Use-on -and				
N2O emissions use-on-land - compost (kg/kg N)	0.0049	0.001	0.015	(own results; see section 5.3)
N2O emissions use-on-land - dewatered sludge (kg/kg N)	0.0055	0.001	0.015	
N2O emissions use-on-land - digestate (kg/kg N)	0.006	0.001	0.015	
Carbon storage in soil - compost (%)	28	Default x 1.2	Default x 0.8	
Carbon storage in soil - dewatered sludge (%)	21	Default x 1.2	Default x 0.8	
Carbon storage in soil - digestate (%)	26	Default x 1.2	Default x 0.8	
K bio-availability (%)	1	1	0.8	
N bio-availability - digestate (%)	0.85	1	0.5	
N bio-availability - compost and dewatered sludge (%)	0.8	0.95	0.5	
P bio-availability (%)	0.55	0.9	0.4	(Oenema et al., 2012; Wilfert et al., 2015)

Table 39: Input-output disaggregated inventory for anaerobic digestion, including utilisation of the biogas in the gas engine. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Tonini et al. (2019)

	Unit	**Anaerobic digestion**
Inputs		
Feedstock-quantity	t (DM)	1
Diesel	L t^{-1}	0.291
Electricity	kWh t^{-1}	6.18
Heat	kWh t^{-1}	0[a]
Outputs		
Digestate	kg (DM) t^{-1}	23.2
Biogas-Electricity	kWh t^{-1}	43.2[b]
Air emissions (fugitive)		
CH_4	kg t^{-1}	0.28
$CO_{2\ \mathrm{biogenic}}$	kg t^{-1}	0.55
Air emissions (gas-engine)[c]		
CH_4	g MJ^{-1}	0.434
N_2O	g MJ^{-1}	0.202
NO_x	g MJ^{-1}	0.0016
SO_x	g MJ^{-1}	0.434
$CO_{2\ \mathrm{biogenic}}$	kg t^{-1}	44

[a] It is assumed that the waste-heat from the gas engine is reused internally to heat the reactor (no external heat source required).

[b] Assuming 30% electrical efficiency

Table 40: Substance mass transfer for dewatering process. TS: total solids; VS: volatile solids; Values are expressed on a wet weight basis, unless otherwise stated (source: Yoshida et al., 2015)

	Dewatered sludge (%)	Centrate (%)
Total mass	10.17%	89.83%
Water	8.70%	91.30%
TS	80.78%	19.22%
VS	80.29%	19.61%
Ash	81.82%	18.38%
C_{bio}	90.72%	9.28%
K	68.99%	31.01%
N	65.00%	35.00%
P	72.07%	27.93%*

*Assuming biological wastewater treatment

Table 41: Input-output disaggregated inventory for dewatering of digestate/sludge. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Yoshida et al., 2015)

	Unit	Dewatering	
Inputs			
Feedstock-type		Digestate	Sludge
Feedstock-quantity	t (DM)	0.45	1
Acrylonitrile	kg t^{-1}	2.31E-03	2.31E-03
Electricity	kWh t^{-1}	2.2	2.2
Outputs			
Centrate	kg (DM) t^{-1}	7.59	9.8
Dewatered	kg (DM) t^{-1}	32.32	40.2

Table 42: Input-output disaggregated inventory for thermal treatment (mono- and co-incineration with municipal solid waste). CO-INC: co-incineration with MSW; DM: dry matter; MO-INC: mono-incineration. Values are expressed on a wet weight basis, unless otherwise stated (source: Tonini et al., 2019)

	Unit	MO-INC	CO-INC
Inputs			
Feedstock-type		Digestate	Sludge
Feedstock-quantity	t (DM)	0.38	0.84
Feedstock-energy	MJ	3358	13520
Activated carbon	kg t^{-1}	1	1
$CaOH_2$	kg t^{-1}	1.04	1.04
$CaCO_3$	kg t^{-1}	5.7	5.7
Electricity	kWh t^{-1}	70	70
HCl	kg t^{-1}	5.60E-03	5.60E-03
NaOH	kg t^{-1}	2.40E-02	2.40E-02
NH_3 25%	kg t^{-1}	1.53	1.53
Polyethylene high density	kg t^{-1}	6.00E-04	6.00E-04
TMT15	kg t^{-1}	0.395	0.395
Water	kg t^{-1}	39.7	39.7
Outputs			
Ashes	kg t^{-1}	94	56
Electricity	kWh t^{-1}	92[a]	197[a]
Air emissions			
As	kg t^{-1}	1.17E-05	6.27E-06
Cd	kg t^{-1}	2.40E-07	1.28E-07
CO	kg t^{-1}	3.31E-02	3.30E-02
CO_2 non-fossil	kg t^{-1}	2.60E+02	4.54E+02
Cr	kg t^{-1}	1.83E-05	9.79E-06
Cu	kg t^{-1}	3.75E-05	2.00E-05
Dioxins	kg t^{-1}	1.81E-11	1.80E-11
HCl	kg t^{-1}	5.32E-03	5.29E-03
HF	kg t^{-1}	3.92E-04	3.90E-04
Hg	kg t^{-1}	3.10E-06	1.65E-06
Ni	kg t^{-1}	1.87E-05	9.96E-06
N_2O	kg t^{-1}	0.79E+00	0.51E+00
NOx	kg t^{-1}	1.15E-01	1.15E-01
Pb	kg t^{-1}	5.58E-05	2.98E-05
PM10	kg t^{-1}	3.01E-02	3.00E-02
SOx	kg t^{-1}	1.22E-02	1.22E-02
Zn	kg t^{-1}	2.68E-04	1.43E-04

[a] Electricity recovery efficiency 15.7%.

Table 43: Input-output disaggregated inventory for acidulation and other post-processing of the ash. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Tonini et al., 2019)

	Unit	Ash acidulation
Inputs		
Feedstock-type		Ash
Feedstock-quantity	t	0.149
Ash-P	kg P t^{-1}	158.93
Diesel	kg t^{-1}	2.1
Electricity	kWh t^{-1}	172
H_2SO_4	kg t^{-1}	710.4
Heat	MJ t^{-1}	29.8
Natural gas	MJ t^{-1}	38
Outputs		
P (granulated)	kg P t^{-1}	158.86

Table 44: Input-output disaggregated inventory for composting. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Tonini et al., 2019; electricity consumption from Boldrin et al., 2009).

	Unit	Composting
Inputs		
Feedstock-type		Sludge
Feedstock-quantity	t (DM)	0.84
Diesel	L t^{-1} (DM)	1
Electricity	kWh t^{-1}	50
Outputs		
Compost	kg t^{-1} (DM)	502.1
Air emissions (biofilters)		
Ammonia	kg t^{-1}	3.72E-02
Carbon dioxide, non-fossil	kg t^{-1}	5.18E-01
Dinitrogen monoxide	kg t^{-1}	6.85E-02
Hydrogen sulfide	kg t^{-1}	2.16E-05
Methane, non-fossil	kg t^{-1}	9.91E-03
Nitrogen	kg t^{-1}	6.23E-03
Terpenes	kg t^{-1}	1.37E-04

Table 45: Input-output disaggregated inventory for lime stabilisation of sludge. DM: dry matter. Values are expressed on a dry weight basis, i.e. per t DM, unless otherwise stated (source: Teoh and Li, 2020)

	Unit	**Lime stabilisation**
Inputs		
Feedstock-type		Sludge
Feedstock-quantity	t	0.732
Lime	kg t^{-1}	300
Outputs		
Stabilised sludge	kg t^{-1}	0.732

Table 46: Input-output disaggregated inventory for landfill construction and operation. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Olesen and Damsgaard, 2014).

	Unit	**Landfill construction**
Inputs		
Feedstock-quantity	t (DM)	0.838
Aluminium	kg t^{-1}	5.80E-11
Clay	kg t^{-1}	8.20E-05
Copper	kg t^{-1}	9.87E-12
Diesel	L t^{-1}	0.24
Electricity	kWh t^{-1}	8
Gravel	kg t^{-1}	1.80E-04
Polyethylene, high density	kg t^{-1}	2.30E-07
Polypropylene	kg t^{-1}	4.00E-11
Polyvinylchloride	kg t^{-1}	1.00E-08
Steel	kg t^{-1}	1.40E-07

Table 47: Input-output disaggregated inventory for landfill gas oxidation in top cover over 20 years. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Olesen and Damsgaard, 2014)..

	Unit	**Landfill gas oxidation**
Emissions to air		
Feedstock-quantity	t (DM)	0.838
1,2-Dichlorobenzene	kg t^{-1}	3.963E-04
Benzene	kg t^{-1}	5.606E-04
Carbon dioxide, non-fossil	kg t^{-1}	3.062E+01
Carbon tetrachloride	kg t^{-1}	7.652E-04
CFC-11	kg t^{-1}	4.430E-05
CFC-113	kg t^{-1}	4.027E-05
CFC-12	kg t^{-1}	3.423E-04
Chloroform	kg t^{-1}	1.667E-05
Cumene	kg t^{-1}	1.450E-04
Ethylbenzene	kg t^{-1}	1.321E-03
HCC-30	kg t^{-1}	9.988E-04
HCFC-21	kg t^{-1}	5.638E-04
HCFC-22	kg t^{-1}	1.933E-04
Hydrogen sulfide	kg t^{-1}	2.900E-03
Mercury	kg t^{-1}	8.055E-08
Methane, non-fossil	kg t^{-1}	1.984E+01
Monochloroethane	kg t^{-1}	5.638E-04
NMVOC	kg t^{-1}	1.692E-03
Phenol	kg t^{-1}	6.283E-05
R-10	kg t^{-1}	2.255E-06
R-40	kg t^{-1}	1.498E-05
Toluene	kg t^{-1}	6.911E-03
Trichloroethylene	kg t^{-1}	2.320E-04
Vinyl chloride	kg t^{-1}	2.030E-04
Xylene	kg t^{-1}	2.642E-03

Table 48: Input-output disaggregated inventory for landfill leachate treatment. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Olesen and Damsgaard, 2014)..

	Unit	Leachate treatment
Inputs		
Feedstock-quantity	t (DM)	0.838
Electricity	kWh t^{-1}	0.44
Water	kg t^{-1}	3.19E-05
Emissions to air		
Ammonium, ion	kg t^{-1}	7.64E-02
Arsenic	kg t^{-1}	6.61E-06
Arsenic, ion	kg t^{-1}	7.45E-06
Barium	kg t^{-1}	1.19E-04
Benzene	kg t^{-1}	8.96E-07
BOD5	kg t^{-1}	1.42E-02
CaDMium	kg t^{-1}	2.94E-06
CaDMium, ion	kg t^{-1}	8.06E-07
Calcium, ion	kg t^{-1}	1.60E-01
Carbon dioxide, non-fossil	kg t^{-1}	-9.74E+01
Carbon tetrachloride	kg t^{-1}	9.86E-07
Chloride	kg t^{-1}	4.03E-01
Chlorine	kg t^{-1}	4.03E-04
Chlorobnzene	kg t^{-1}	5.92E-07
Chloroform	kg t^{-1}	1.55E-08
Chromium VI	kg t^{-1}	4.74E-07
COD	kg t^{-1}	1.15E-01
Copper	kg t^{-1}	2.83E-05
Copper, ion	kg t^{-1}	4.53E-06
Cumene	kg t^{-1}	3.95E-07
Dioctylphthalate	kg t^{-1}	1.10E-06
Ethylbenzene	kg t^{-1}	3.98E-06
HCC-30	kg t^{-1}	8.48E-07
Iron	kg t^{-1}	8.33E-02
Iron, ion	kg t^{-1}	2.19E-02
Lead	kg t^{-1}	1.33E-05
Magnesium	kg t^{-1}	8.20E-02
Mercury	kg t^{-1}	8.49E-08
Monochloroethane	kg t^{-1}	8.42E-07
Nickel	kg t^{-1}	1.42E-05
Nickel, ion	kg t^{-1}	1.87E-05
Nitrogen	kg t^{-1}	3.08E-01
Phenol	kg t^{-1}	7.08E-07
Phosphate	kg t^{-1}	1.53E-04
R-10	kg t^{-1}	3.95E-08

Selenium	kg t^{-1}	2.34E-06
Silver	kg t^{-1}	1.59E-05
Silver, ion	kg t^{-1}	2.11E-06
Sodium, ion	kg t^{-1}	1.25E-01
Sulfur dioxide	kg t^{-1}	1.39E-02
Toluene	kg t^{-1}	7.89E-06
Trichloroethylene	kg t^{-1}	1.15E-06
Xylene	kg t^{-1}	3.98E-06
Zinc	kg t^{-1}	6.24E-04
Zinc, ion	kg t^{-1}	1.24E-04

Table 49: Input-output disaggregated inventory for landfill gas combustion in gas engine. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Olesen and Damsgaard, 2014)..

	Unit	**Landfill gas combustion**
Inputs		
Feedstock-quantity	t (DM)	0.838
Outputs		
Landfill gas-Electricity	kWh t^{-1}	342.3[a]
Air emissions (gas-engine)		
1,2-Dichlorobenzene	kg t^{-1}	5.59E-05
Benzene	kg t^{-1}	7.45E-05
Carbon dioxide, non-fossil	kg t^{-1}	7.01E+01
Carbon monoxide, non-fossil	kg t^{-1}	3.05E-01
CFC-11	kg t^{-1}	4.69E-06
CFC-113	kg t^{-1}	2.35E-06
CFC-12	kg t^{-1}	2.35E-05
Chloroform	kg t^{-1}	1.41E-06
Cumene	kg t^{-1}	1.86E-05
Dioxins	kg t^{-1}	1.30E-10
Ethylbenzene	kg t^{-1}	1.86E-04
HCC-30	kg t^{-1}	9.38E-05
HCFC-21	kg t^{-1}	4.69E-05
Hydrogen chloride	kg t^{-1}	1.44E-03
Hydrogen fluoride	kg t^{-1}	1.44E-03
Hydrogen sulfide	kg t^{-1}	7.51E-05
Mercury	kg t^{-1}	6.70E-08
Methane, non-fossil	kg t^{-1}	2.58E-01
Monochloroethane	kg t^{-1}	6.57E-05
Nitrogen oxides	kg t^{-1}	2.95E-01
NMVOC	kg t^{-1}	5.63E-05
PAH	kg t^{-1}	3.60E-05
Particulates2.5-10	kg t^{-1}	6.48E-03
Phenol	kg t^{-1}	9.32E-06
Polychlorinated biphenyls	kg t^{-1}	3.60E-05
R-10	kg t^{-1}	2.35E-07
R-40	kg t^{-1}	1.41E-06
Sulfur dioxide	kg t^{-1}	1.10E-02
Tetrachloroethylene	kg t^{-1}	4.69E-05
Toluene	kg t^{-1}	1.02E-03
Trichloroethylene	kg t^{-1}	1.88E-05
Xylene	kg t^{-1}	3.73E-04

a Assuming 30% electrical efficiency

Table 50: Substance mass transfer for open storage of biosolids. TS: total solids; VS: volatile solids; Values are expressed on a wet weight basis, unless otherwise stated (Holly et al., 2017 Willén eta l., 2016; source: Delre et al., 2019)

	To air (%)	**To farmland (%)**
C bio	8.68	91.32
N	7.10	92.90
Water	0.00	100.00
VS	15.78	84.22
P	0.00	100.00
K	0.00	100.00
TS	15.78	84.22
Ash	0.00	100.00

Table 51: .Input-output disaggregated inventory for use on farmland of digestate, sludge and compost. DM: dry matter. Values are expressed on a wet weight basis, unless otherwise stated (source: Tonini et al., 2019).

	Unit	**Use on land**		
Inputs				
Feedstock-type		Digestate	Sludge	Compost
Feedstock-quantity	t (DM)	0.341	0.732	0.449
Diesel	L t^{-1} (DM)	6.30E-01	6.30E-01	6.30E-01
Emissions to air				
Carbon dioxide, non-fossil	kg t^{-1}	2.38E+02	3.39E+02	2.98E+02
Carbon dioxide, fossil[a]	kg t^{-1}	-8.38E+01	-9.03E+01	-1.16E+02
Methane, non-fossil[b]	kg t^{-1}	1.75E-01	1.58E-01	1.41E-01
Dinitrogen monoxide[c]	kg t^{-1}	4.48E-01	8.70E-02	1.32E-01
Ammonia	kg t^{-1}	1.44E+00	9.12E-01	1.54E+00

a From data derived by JRC

b From data derived by JRC

c From data derived by JRC

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