CZECH UNIVERSITY OF LIFE SCIENCES PRAGUE

Faculty of Economics and Management

Diploma Thesis Title:

IS THERE NEED FOR AN UPDATED SLUDGE DIRECTIVE? A RAPID EVIDENCE ASSESSMENT

This Diploma Thesis has been written and defended at the **Cranfield University** in the **United Kingdom** under the Double Degree Agreement between the Czech University of Life Sciences Prague, and the **Cranfield University**. In accordance with the Double Degree Agreement, this Diploma Thesis is fully recognized as part of the MSc programme study at the Czech University of Life Sciences Prague.

Author: Pavel Chyška

Diploma Thesis Supervisor: Jason Weeks and Philip Longhurst

CRANFIELD UNIVERSITY

Pavel CHYSKA

IS THERE NEED FOR AN UPDATED SLUDGE DIRECTIVE? A RAPID EVIDENCE ASSESSMENT

SCHOOL OF ENERGY, ENVIRONMENT AND AGRIFOOD
Environmental Risk Management

MSc THESIS
Academic Year: 2014 - 2015

Supervisor: Jason Weeks and Philip Longhurst
September 2015

CRANFIELD UNIVERSITY

SCHOOL OF ENERGY, ENVIRONMENT AND AGRIFOOD
Environmental Risk Management

MSc THESIS

Academic Year 2014 - 2015

Pavel CHYSKA

Is there need for an updated Sludge Directive?
A rapid evidence assessment

Supervisor: Jason Weeks and Philip Longhurst
September 2015

This thesis is submitted in partial fulfilment of the requirements for the degree of Master of Science

ABSTRACT

Agricultural re-use of sludge, as a way of sludge management, is a common practice in many EU countries. Despite its inherent qualities, sludge may contain potentially toxic elements (PTEs), which are regulated by the European legislation. The current European sludge legislation sets out the limit concentrations for certain heavy metals in sludges and soils for agricultural purposes. There are concerns that the Sludge Directive 86/278/EC may be out-dated since it has been in place for almost 30 years. Over time, advances in legislation regarding drinking water quality legislation have lead to an increase in sludge production. Additionally, the ban on disposal of sludge at sea has contributed to the problem of sludge management. The aim of this work was to analyse pertinent evidence related to the adverse effects of sludge application to agricultural soils and to determine whether there is a need for an updated Sludge Directive or UK update. Code of Practice For Agriculture Use of Sewage Sludge provided the basis to measure appropriateness of UK legislation regarding the sludge application. A systematic rapid evidence assessment approach (REA) was employed to obtain pertinent scientific studies, which were used to pose case scenarios building on a qualitatively structured what-if technique (SWIFT). Due to the complexity of the topic, the work is focused on evidence regarding one essential metal (Cu), one non-essential metal (Cd) and an organic compound (Bisphenol A). No significant evidence of harm arising from direct application of biosolids within the current limits were identified; however, the research highlighted areas that need to be addressed and appropriate measures put in place to reduce future potential risks arising from the application of sludges to agricultural land.

ACKNOWLEDGEMENTS

The author is grateful for all the support and guidance to Jason Weeks, Philip Longhurst and Heather Woodfield. Further thanks are due to Ian Martin and Danielle K. Ashton for their support and interest in this work.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ABBREVIATIONS

AOX	Halogenated organic compounds
BPA	Bisphenol A
DEHP	Di(2-ethylhexyl) phthalate
LAS	Linear alkylbenzene sulphonates
NPE	Nonylphenol and nonylphenol ethoxylates with 1 or 2 ethoxy groups
PAHs	Polycyclic aromatic hydrocarbons
PCBs	Polychlorinated biphenyls
PCDDFs	Polychlorinated dibenzo-p-dioxins/dibenzofurans
PTEs	Potentially toxic elements
REA	Rapid evidence assessment
SWIFT	Structured what-if technique

This thesis has been prepared in the format used for scientific papers appearing in the journal of *Environmental Science & Policy*.

Is there need for an updated Sludge Directive? A rapid evidence assessment

Pavel Chyska, Jason Weeks, Philip Longhurst

School of Energy, Environment and Agrifood, Cranfield University, College Rd, Cranfield, Bedford MK43 0AL, United Kingdom

Abstract

Agricultural re-use of sludge, as a way of sludge management, is a common practice in many EU countries. Despite its inherent qualities, sludge may contain potentially toxic elements (PTEs), which are regulated by the European legislation. The current European sludge legislation sets out the limit concentrations for certain heavy metals in sludges and soils for agricultural purposes. There are concerns that the Sludge Directive 86/278/EC may be out-dated since it has been in place for almost 30 years. Over time, advances in legislation regarding drinking water quality legislation have lead to an increase in sludge production. Additionally, the ban on disposal of sludge at sea has contributed to the problem of sludge management. The aim of this work was to analyse pertinent evidence related to the adverse effects of sludge application to agricultural soils and to determine whether there is a need for an updated Sludge Directive or UK update. Code of Practice For Agriculture Use of Sewage Sludge provided the basis to measure appropriateness of UK legislation regarding the sludge application. A systematic rapid evidence assessment approach (REA) was employed to obtain pertinent scientific studies, which were used to pose case scenarios building on a qualitatively structured what-if technique (SWIFT). Due to the complexity of the topic, the work is focused on evidence regarding one essential metal (Cu), one non-essential metal (Cd) and an organic compound (Bisphenol A). No significant evidence of harm arising from direct application of biosolids within the current limits were identified; however, the research highlighted areas that need to be addressed and appropriate measures put in place to reduce future potential risks arising from the application of sludges to agricultural land.

Keywords: Biosolids, Heavy Metals, Agriculture, Cadmium, Copper, Bisphenol A

1 INTRODUCTION

The European Sewage Sludge Directive 86/278/EC provides mandatory limit values for certain heavy metal concentrations present in biosolids and sludges applied to agricultural soils (CEC, 1986). The main purpose of the Directive is to prevent any negative effects resulting from sludge applications to agricultural soils impacting on soil quality, soil ecology, or livestock, crops, human health and the wider environment. The Directive was introduced almost 30 years ago, and since that time, new contaminants and organic products are increasingly present in the introduced sludge that could potentially cause harm to receptors. The identified receptors that could be negatively affected by sludge re-use in agriculture are shown in Table 1. Given the time that has passed since the ratification of the Sludge Directive and the dynamic nature of the waste that is being generated, there is a probability that the current limits set out by the Directive and British Code of Practice For Agriculture Use of Sewage Sludge (DoE, 1996) may be out-dated and are no longer protective of the environment.

Table 1 – Pathways and receptors for contaminants found in sludges

Pathway	Receptor
Evaporation of organic compounds	Air
Leaching through soil profile	Aquifer
Aquifer	Contained water bodies
Direct application	Crops, soil microorganisms
Inhaling organic compounds, direct contact	Field workers
Ingesting the biosolids and contaminated crops	Grazing animals
Ingesting contaminated crops and animals	Human
Ingesting soil contaminated with biosolids	Playing children

Additional limits for potentially toxic elements (PTEs) that were not covered by the Sludge Directive, as well as more robust rules for sludge application to agricultural grasslands represent a few examples that were introduced by the Code of Practice For Agriculture Use of Sewage Sludge (DoE, 1996) specifically for the UK. The Code serves as guidelines for use of sludges/biosolids in agriculture in the UK, which complements the Sludge Directive. The UK has tightened the limits introduced by the Directive and developed the guidelines to ensure sustainable agricultural practice and to protect the wider UK environment. The UK limits for PTEs introduced by these guidelines have been considered during the synthesis of evidence within this work.

Sludge is a by-product of wastewater treatment, which is increasingly re-used in Europe as a soil improver (UKWIR, 2007). However, it is still regarded as a waste, despite the fact that re-use in agriculture might present lower costs for the wastewater treatment operators as well as a lower environmental burden overall (European Commission, 2002). In contrast to sludge recycling in agriculture, incineration and landfilling are still common ways of disposing of sludge as they account for 51% of sludge disposal in the EU (Fytili and Zabaniotou, 2008). Gendebien (2009) showed that 68% of sludge, produced in the UK in 2006, was re-used in agriculture, whereas countries such as Slovakia, Netherlands, Romania and Greece depend heavily on incineration and landfilling.

Sewage sludge contains phosphates, nitrogen, potassium, magnesium, sulphur and organic matter (MAFF, 1987). These qualities provide benefit, when sludge is re-used in agriculture (Singh and Agrawal, 2008). Phosphate recycling via sludge application in agriculture also helps to slow phosphate rock depletion, which may trigger global scale problems in 100-200 years (Cordell et al., 2009). Sludge application to poor quality land can, through its inherent properties, help in the remediation of such contaminated or brownfield sites (Stuczynski, 2000) and improve eroded soils (UKWIR, 2007). Sludge is also used in the production of biogas through anaerobic digestion (FWR, 2011).

As well as PTEs, sludge can contain various pathogens such as the eggs of tapeworms and potato cyst nematodes. The Sludge Directive does not currently set limits for their concentrations in sludges or soils. However, the Code of Practice For Agriculture Use of Sewage Sludge (DoE, 1996) suggests safety-monitoring measures for sludge that is obtained from any waste source from animal or poultry processing plants, since the sludge may contain elevated levels of pathogens.

PTEs in the form of heavy metals such as Cd, Cu, As, Pb and Hg appear in sludge in various concentrations and may pose a threat to soil, ground water, plants and people resulting in harm (Carrondo et al., 1978). Even though heavy metals may be toxic, not all the metals are toxic at all times. Living organisms need essential metals such as Cu; however, its excessive quantities may introduce carcinogenic effects (Theophanides and Anastassopoulou, 2012). On the other hand, non-essential metals such as Cd form the group of the most toxic metals, which may introduce carcinogenic as well as mutagenic effects (Eisler, 1985). Heavy metals along with other hazardous chemicals are entering the wastewater stream through domestic wastewater, medical facilities, industrial effluent and water run-off from roads, which eventually find their way into sludge during the wastewater treatment process (DEFRA, 2002).

Heavy metals accumulate in soils and tend to be immobilised in the soil environment (Tack, 2010). Plant uptake, or bioavailability, of heavy metals depends on metal interactions with organic matter in soil; however, their bioavailability decreases over time. Factors such as soil pH and metal adsorption on soil particles also affect bioavailability of metals (McGrath, 1987). Experiments shown that microbial biomass decreased by 40%, when sludge containing Zn and Cu at concentrations two and half times higher than the permitted concentration were added to soil (Chander and Brookes, 1991). In terms of identification of a metal with the most toxic effect on soil microbial processes and crops research remains inconclusive (UKWIR, 2007).

Advances in Waste Water legislation (CEC, 1991) contribute to cleaner water that is returned into rivers, but also resulted in increased amounts sludge as well as potential contaminants (DEFRA, 2002). On the contrary, concentrations of Cd and Zn present in the sewage sludge have decreased by 89% and 53% respectively in the UK over the last few decades (IC Consultants Ltd London, 2001). The current UK limits for PTE concentrations in sludge are shown in Table 2.

Table 2 – Maximum permissible concentrations of potentially toxic elements in soil after application of sewage sludge and maximum annual rates of addition

PTE	Maximum permissible concentration of PTE in soil (mg/kg dry solids)				Maximum permissible average annual rate of PTE addition over a 10 year period (kg/ha)
	pH 5.0<5.5	pH 5.5<6.0	pH 6.0-7.0	pH >7.0	
Zinc	200	200	200	300	15
Copper	80	100	135	200	7.5
Nickel	50	60	75	110	3
	For pH 5.0 and above				
Cadmium	3				0.15
Lead	300				15
Mercury	1				0.1
*Chromium	400				15
*Molybdenum	4				0.2
*Selenium	3				0.15
*Arsenic	50				0.7
*Fluoride	500				20

*These parameters are not subject to the provisions of Directive 86/278/EEC (In 1993 the European Commission withdrew its 1988 proposal to set limits for addition of chromium from sewage sludge to agricultural land), (DoE, 1996).

Besides heavy metals and pathogens, sludges are likely to contain a large variety of organic chemicals. A review by Harrison et al. (2006) has identified the presence of 516 organic chemicals in sludge across various study samples. These chemicals included detergents, personal care products, pharmaceuticals, plasticizers etc. Table 3 shows the classes of organic chemicals along with the particular examples of organic chemicals that may be found in sewage sludge. None of these chemicals are regulated within the current Sludge Directive. The past research in the field of organic chemicals has not demonstrated risks to human health when sludge is re-used in agriculture (FWR, 2011). Smith (2009) stated that the use of sludge for agricultural purposes should not be restricted due to current concentrations of organic chemicals present in the sludge.

Table 3 – Organic chemicals found in sludge

Class of organic chemicals	Examples of organic chemicals found in sludge
Aliphatics	Butadiene (hexachloro-1,3-), Ethane (hexachloro), Ethylene (dichloro), Methane (dichloro), Propane
Chlorobenzenes	Benzene (dichloro) isomers, Benzene (hexachloro), Benzene (monochloro), Benzene (pentachloro), Benzene (tetrachloro), Benzene (trichloro) isomers
Flame retardants	Brominated diphenyl ether congeners (BDEs), Cyclododecane (hexabromo) isomers, Tetrabromobisphenol A, Tetrabromobisphenol A (dimethyl)
Monocyclic hydrocarbons and heterocycles	Benzene, Benzene (ethyl), Benzene (mononitro), Benzoic acid, Analine (chloro) (P-), Styrene, Toluene
Nitrosamines	N-nitrosdiphenylamine, N-nitrosodiethylamine, N-nitrosodimethylamine, N-nitrosodi-n-butylamine,
Personal care products	Ibuprofen, Naproxen, Salicylic acid, Antibiotics, Triclosan, Fluorescent whitening agents, Fragrance material
Pesticides	Aldrin, Chlordane, Cyclohexane isomers, DDT and related congeners, Dieldrin, Endrin, Heptachlor epoxides
Phenols	Phenol, Phenol chloro congeners, Phenol methyl congeners, Phenols nitro congeners
Phthalate	Bis(2-chloroethyl) ether, Bis(2-chloroisopropyl) ether, Bis(2-cloroethoxy) methane, Phthalates
Polychlorinated biphenyls, naphthalenes, dioxins and furans	Biphenyl (decachloro), Biphenyls (polybrominated) Dibenzofuran, Dioxins and furans (polychlorinated dibenzo), PCB congeners
Polynuclear aromatic hydrocarbons	Benzo(a)anthracene, Benzofluoranthene congeners, Benzopyrene congeners,, Chrysene, Dibenzoanthracene congeners, Indeno(1,2,3-c,d) pyrene, Naphthalene
Sterols, stanols and estrogens	Campestanol (5a+5b), Campesterol, Cholestanol (5a-), Cholesterol Coprostanol, Estradiol (17b), Estrone
Surfactants	Alcohol ethoxylates, Alkylbenzene sulfonates, Alkylphenolcarboxylates, Alkylphenolethoxylates,
Triaryl/alkyl phosphate.esters	Cresyldiphenyl phosphate, Tricresyl phosphate, Tricresyl phosphate, Tri-n-butylphosphate, Triphenylphosphate

(Adapted from Harrison et al., 2006)

Nevertheless, Schnaak et al. (1997) concluded that surfactant and toluene concentrations in sludge exceeded the terrestrial ecotoxicology values protective of soil health. With regard to dioxins and furans, European legislation (CEC, 1985) helps to protect sludge from these organics, which belong to the most hazardous chemicals. However, Jones and Sewart (1997) stated that the continuous addition of sludges to agricultural land leads to the accumulation of polychlorinated dibenzofurans (PCDDFs) in the soil due to their persistent qualities. Example of hazardous organic chemical of high concern include Bisphenol A (2,2-Bis-(4-hydroxphenyl)propane; BPA). BPA is oestrogen-like chemical found in sludges (Fromme et al., 2002), which was found to be an endocrine disruptor (Delclos et al., 2014; Newbold et al., 2007). This plasticizer, which is used in many products, is commonly found in sludges in concentrations 0.1–3.2 × 10^7 µg kg^{-1} (Harrison et al., 2006; Song et al., 2014).

In order to mitigate the potential risk of harm, arising from the application and reapplication of PTEs contained in the sludges; that may result in harm to soil function or structure, sewage sludge is treated prior to its application and use (DoE, 1996). Treated sludge, sometimes called biosolids, may differ depending on the type of treatment received, since the treatment type determines the amount of residual PTEs (Stylianou et al., 2006), other contaminants and pathogens (Hospido et al., 2005). However, Member States under the Directive may permit use of sludge without treatment if the sludge is injected or otherwise incorporated into soil (CEC, 1986).

The global climate is changing (Parmesan and Yohe, 2003); therefore factors such as humidity and temperature are likely to play a more significant role in the bioavailability of metals and the persistence of pathogens in the future (Hooda and Alloway, 1993). Climate change therefore may be a significant factor driving regulatory changes regarding hazardous elements in sludges in the future.

The main focus of this work was to determine whether there is a need for an update of the Sludge Directive along with the UK Code of Practice For Agriculture Use of Sewage Sludge, given the published evidence on the accumulation of contaminants in soils amended with sewage sludge and potential for harm to be caused in spite of the implementation of the Directive and its UK interpretation (SI, 1989). Many studies focusing on the bioavailability of metals and the effects of other contaminants in soil have been published since the introduction of the Directive. This report summarises a review of a large body of relevant evidence synthesized using a systematic approach using a rapid evidence assessment method (REA) (Government Social Research, 2010). Complementary to the REA approach, a structured what-if technique (SWIFT) was used to determine whether some of the identified contaminants present in the sludge (Card et al., 2012), based on the evidence obtained, might pose a threat to the environment and whether there are drivers for the Sludge Directive update as well as its UK interpretation.

2 METHODS

This assessment sought to identify relevant evidence from the published scientific literature by using a rapid evidence assessment method (REA). The REA process offers a systematic approach for generating the most pertinent evidence quickly, despite the complexity and amount of documents regarding the topic (Government Social Research, 2010). On the down side, this technique may introduce a degree of bias into the review by omitting pertinent information, or information published elsewhere, information in different languages, unpublished information or less developed search strings are several examples of factors that may introduce bias into this approach.

In order for a selected published study to be included within this REA it had to comply and meet with the following inclusion criteria: (1) studies must evaluate effects of sewage sludge in soils, intended as a soil improver in agriculture; (2) studies must consider the effects of heavy metals or organic pollutants on the environment; (3) studies must be peer reviewed; (4) the studies must use empirical data; (5) the location of the study must be within Member States of the EU; (6) studies must be published in English language; (7) studies must be published in the year 1995 and later.

Qualitative analysis of the evidence was conducted via employing risk-based structured what if-technique (SWIFT) on the process of sewage sludge application in agriculture with the focus on particular non-essential metal (Cd), essential metal (Cu) and an organic pollutant (BPA). The SWIFT is a flexible tool for identifying hazards, assessing the risk(s) and proposing actions (Card et al., 2012). This technique is traditionally a team based brainstorming method, where the conceptual understanding of the processes under scrutiny drives the analysis. The participation of a review team and their contribution in this work is substituted by the use of the REA outcomes, where the evidence obtained by REA formed the basis for assessing the hazards arising from sewage sludge applications to land.

Setting out the boundaries of the problem is an important step for the initial analysis using SWIFT. Regulatory response to a multifaceted problem of protection from negative effects of sewage sludge could be dealt with on different levels. Legislation could regulate the waste producers, wastewater treatment facilities and agricultural operators in order to address the problem. This work focused on the problem of application of the sewage sludge in agriculture. A graphical representation of the SWIFT boundaries (red boundary) is shown in Figure 1. Based on the conceptual understanding of the process and likely effects of contaminants introduced by sludges/biosolids, a set of questions were generated, which guided the analysis of the literature.

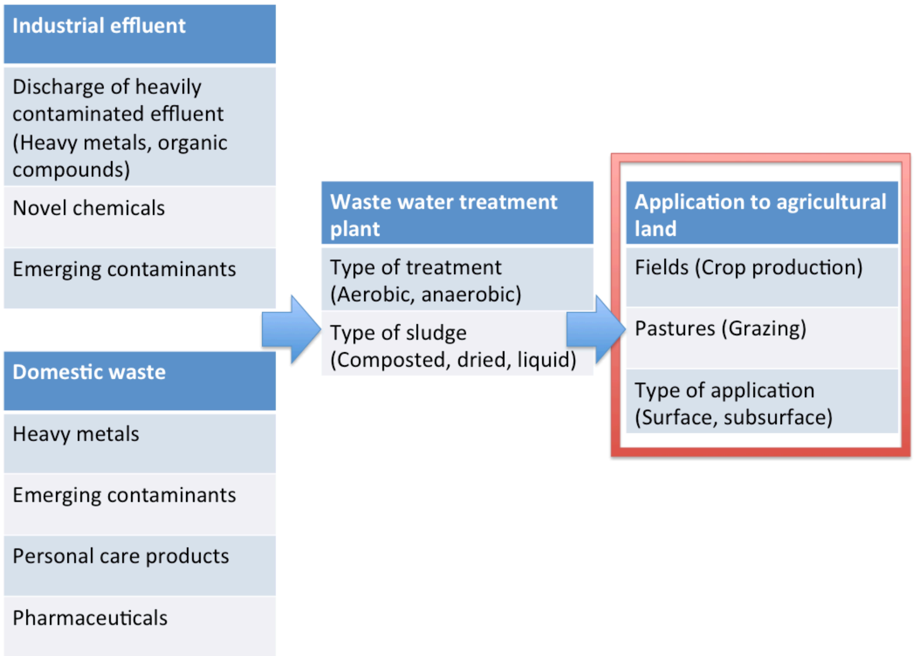

Figure 1 – A portion of sludge management flow and boundaries (red boundary) that served as the basis for the SWIFT

Two online scholarly scientific databases, pertinent to the topic, i.e. Scopus and Environment Complete, were searched using a combination of search strings with Boolean search operators. The following search strings were used to obtain the relevant articles: (1) sludge OR biosolid*; (2) soil* OR agricultur* OR farm* OR arable OR pasture OR grassland; (3) "heavy metal*" OR "toxic element*" OR toxic* OR "trace element*" OR "trace substance*" OR "priority substance*" OR "trace metal*" OR "trace contaminant*" OR pathogen* OR phytotoxicity OR contamin* OR "food chain transfer" OR harm OR damage*. Year 1995 was set as an initial year for relevant articles because it is one year before publishing of the Code of Practice For Agriculture Use of Sewage Sludge. Duplicate articles, studies that were not conducted within the EU, irrelevant articles, studies regarding only tannery sludges or dairy sludges were filtered out. Additionally, studies considering the use of sewage sludges in forestry, bioremediation and long-term effects of landfilling sewage sludges were excluded. Additional filter procedure was imposed to reduce the amount of articles for further study. Articles must have examined at least one of the following contaminants: (1) Cd; (2) Cu; (3) BPA. The resulting data set did not include evidence regarding the effects of BPA in sewage sludge used in agriculture. Therefore, additional search for relevant articles was conducted by employing the above-mentioned online databases. The articles that were identified were obtained in full copy. The resulting data set consisting of 61 final articles can be seen in Table 1 within Appendix A.

3 Results and discussion

Studies under this review provided a diverse set of information about the effects of sewage sludge applications to land, mainly with regard to heavy metal contaminated sludges. The repeating themes within the identified studies were the accumulation of trace metals in soils, mobility of heavy metals, plant uptake of trace metals, fractioning of trace metals and intake of heavy metals by animals. The problem with many studies (Rastetter and Gerhardt, 2015; Escrig and Morell, 1998; Obbard and Jones, 2000; Walter et al., 2002) was that they provided only limited insight into any negative effects of metals contained within sewage sludges due to their laboratory setting; the omitting important soil processes and/or apply high metal concentrations, which are unlikely to occur in the real world environment. The analysis of the literature revealed many variables that affect the application of biosolids to soils that may help to inform decision making, when reviewing the sludge legislation.

A brainstorming procedure, as an important part of structured what-if technique (SWIFT), identified the following hazards arising from sludge applications in agriculture: (1) Biosolids within the permissible concentrations are currently applied to soils – The question regarding the current permissible concentrations and whether there is an evidence of harm; (2) Biosolids are applied over long time periods – The question is trying to understand the long-term effects of sludge application; (3) Biosolids are applied to the soil surface – The question is trying to determine risks arising from such surface applications of sludges; (4) An increase in ambient temperature – The question is trying to find out whether the current legislation is sufficient to understand future risks in terms of likely climate change setting; (5) Biosolids contained BPA – The question is trying to determine the fate of this carcinogenic compound in biosolids. The results of the SWIFT along with the “what-if” questions are displayed in Table 4. The following sections contain results and discussion based on the findings from the rapid evidence assessment (REA) in the context of hazards identified by the SWIFT.

Table 4 – “What-if“ hazard analysis

What if?	Answer	Likelihood	Consequences	Recommendations
Biosolids within the permissible concentrations are applied to soils?	Possible phytotoxicity depending on many factors; promotion of leaching through soil gradients into controlled waters, accumulation of metals.	Possible	Significant	Review the permissible concentrations of heavy metals with regard to different fractions of heavy metals and soil and environment specific conditions.
Biosolids are applied over long time periods?	Possible leaching of contaminants (may be promoted by weathering).	Likely	Significant	Further monitoring of soil, conduct more research on fractioning of heavy metals.
Biosolids are applied to the soil surface?	Possible uptake by plants; possible intake by grazing animals; possible human intake; possible nuisance (smell).	Likely	Highly significant	Extend the “no grazing period”; injection of biosolids into soil may reduce the risk of direct contamination as well as nuisance.
There is an increase of ambient temperature?	Release of available heavy metals after the degradation of organic matter, which provides sorption surface.	Likely	Highly significant	Take into account climate projections and tweak the limit concentrations of heavy metals to reduce risks.
Biosolids contain BPA?	Possible accumulation of BPA; possible leaching into the lower gradients and entering aquifer.	Possible	Significant	Set up the regulatory limits for concentration of BPA in biosolids; promote aerobic digestion as a favourable treatment method.

3.1 Biosolids within the permissible concentrations are applied to soils

The two basic variables, when judging the effects of biosolid application, are dose and concentration of contaminants in the material applied. Studies reporting on plant uptake of trace metals/phytotoxicity stated that the efficiency of absorption of certain metals such as Cd is higher with higher doses of sludge, however all metals studied by Antoniadis et al. (2010) did not increase plant concentrations. Similar results about plant concentrations of metal were reported by Canet et al. (1998), who examined uptake by lettuce in a long-term study. Maize uptake experiment by Delgado et al. (2002), used sewage compost set within the EU limit concentrations for metals, and concluded that there were no toxic effects to plants and thus such materials may be used as a fertiliser on maize.

Studies showed that variables other than dose and concentration may determine the effects of biosolids, which complicates extrapolation, and therefore questions whether the dose and concentration is sufficiently low to protect the environment. Experiments (Moreno et al., 1998; Hooda et al., 1997) with higher concentrations of heavy metals in sludges than those permitted by the Directive revealed that crop type plays an important role in the uptake of heavy metals. The significance of different plant species and their various affinities to heavy metal uptake is also mentioned by Mihalache et al. (2014). Plant mechanisms in certain crops restrict metal transport to above ground parts of plants (Kid et al., 2007; Soriano-Disla et al., 2008).

The application of different sludges or their different mixtures may yield different results in terms of phytotoxicity as well. A study examining three types of sludges (anaerobically digested sludge, heat-dried sludge and composted sludge), in a field experiment, inhibited the germination of cress, even though the research used metal concentrations within the limits adhering to the Directive (Walter et al., 2006). Similar results were observed in wheat-seed germination in a column study (Benítez et al., 2000). Nevertheless, composted biosolids or biosolids ash did not show any phytotoxic effect

(Walter et al., 2006). This suggested that the differences in sludge treatment have an effect on overall phytotoxicity. On the other hand enhanced and conventionally treated biosolids did not show significant leaching of heavy metals (Gove et al., 2010). Moolenar and Beltrami (1998) who studied the effects of sewage sludge and Bordeaux mixture, a commonly used fungicide, concluded that the EU sludge use policy is not efficient in the sustainable management of heavy metals.

The effects of sewage sludge, with metal concentrations well above the UK permissible maximum concentrations, over a long-term experiment on the rhizobium did not show any significant adverse effects on nodulation (Smith, 1997). Moreover, no consistent effects of liquid sludges, containing Zn, Cd and Cu in concentrations close to their respective upper limit concentrations set out by the Sludge Directive at nine sites, were observed on the rhizobium during the four year experiment (Gibbs et al., 2006a). No inhibitory effects to microbial activity were recorded that could be explained by the metal contaminations from sludge application to agricultural land (Gibbs et al., 2006a; 2006b).

The mobility of heavy metals and its bioavailability in sludge-amended soil was tested by many authors who concluded that sorption of certain trace elements such as Cd is determined by a competitive process of adsorption (Antoniadis et al., 2007). This means that it is not the total concentration of metals in soils that determines their bioavailability, but also the competitiveness to adsorb to organic matter and other minerals in a soil. The fractioning of heavy metals plays an important role in determining their mobility, bioavailability and eventually their toxicity (Miller et al., 1986; Tsadilas et al., 1995). The addition of sewage sludges to soil increased non-residual forms of metals, which were more bioavailable (Morera et al., 2001; Sánchéz-Martín et al., 2007, Kalembasa and Pakula, 2009). Gove et al. (2010) noted that metal leaching losses were more associated to soil complexes than to the application of biosolids. Composting of sewage sludge may also alter the forms of available heavy metals into more available fractions (Zn, Ni, Pb); in the case of Cu, different treatment types (composted, dehydrated and liquid) when applied over 6 years did not affect its bioavailability (Rossi et al., 2002). Fractioning studies regarding Cu concluded that it is the least problematic metal due to its inherent ability to readily form complexes in the soil, which are unavailable to plants (Moreno et al., 1997). A bioluminescence study also concluded that soluble Cu in soil amended with high amounts of sludge up to 349 mg kg^{-1} had no effect on bacterial toxicity (Chaudri et al., 1999). In contrast, other research studying the uptake of heavy metals by barley from sludge-amended soil stressed concerns about Cu, Cd and Zn, which posed the highest risk for human intake (Soriano-Disla et al. 2014). Lombi and Gerzabek concluded that extractable concentrations of Cu and Zn in soils increased by the addition of biosolids. Different researchers have suggested that Cu is retained in the root system (Gondek et al., 2010).

The aforementioned evidence suggests that the total permissible amounts of heavy metals added to soils may be an inaccurate endpoint measurement to determine appropriate safeguards to harm. An alternative way to distinguish hazardous concentrations would necessitate measuring the bioavailable metal concentration in soils in preference to total concentrations. Nevertheless, Loveland and Thompson (2000) argue that research on fractioning of metals and their bioavailability has been using methods, which are not able to precisely determine the available metal concentration for all metals of concern. Heavy metal fractions may play a significant role in determining the level of possible hazard and should be taken into account when deriving new regulations or codes for sewage sludge use in agriculture. There is no widely accepted method to determine the bioavailable soil concentration (Loveland and Thompson, 2000).

3.1.1 Key findings

- Cu complexes may or may not be available to plants in sludge-amended soils due to many physical and biotic factors.
- Available soluble Cu had no effect on soil bacteria.
- Composted sewage sludge did not affect the bioavailability of Cu.

- Fractions of available heavy metals are more important than their total amount.
- Phytotoxicity is dependent on type of crop and sludge.
- Leaching of contaminants is dependent upon soil complexes.

3.2 Biosolids are applied over long time periods

Studies on the accumulation of metals in soils reported that long-term repeated application of biosolids increased the amount of bioavailable metals in the soil in initial first years; however, with time the availability reduced to values measured in control plots (Antoniadis et al., 2010). No differences in accumulation of heavy metals compared to the use of manure were observed, though permeability of soil may have played a role (Raffaella et al., 1997). Subsequent application of biosolids at sites with a history of sludge applications may lead to leaching loses of Cu (Kidd et al., 2007). Canet et al. (1998) evaluated metal movement within the soil profile and concluded that metal movement existed, however their research was inconclusive, whether it may be attributed only to leaching or combination of agricultural processes that occurred over time. Weathering may also have an impact on leaching of heavy metals due to their migration through fissures, thus impacting aquifer (Proust et al., 2011). Such a migration of metals such as Cd and Zn may affect plant root systems, which favour fissural environment (Proust et al., 2011).

Testing of agricultural sites prior to the application of biosolids is required by the sludge legislation (SI, 1989). It requires testing of soil pH and metal concentrations in the depth of 25 cm. UK guidance makes differences in depths depending on the site use (DoE, 1996). This means that the representative grassland samples, tested for presence of PTEs, are taken to 7.5 cm depth compared to 15 cm in arable soils. The UK approach seems reasonable, given the evidence that even though concentrations of heavy metals in a 25 cm depth may be within the concentrations of statutory limit, depths of 2.5 cm may contain amounts that are almost twice the permitted concentration (Wilkinson et al., 2001). Acid soils are known to promote the bioavailability of heavy metals (Serrano et al., 2005); however, it is not the only factor affecting the behaviour of contaminants in soils. Factors affecting the bioavailability of metals, according to the reviewed studies, include the type of soil (Gondek et al., 2010; Antoniadis et al., 2010; Morrera et al., 1997), soil structure (Soriano-Disla et al., 2014) and pH (Antoniadis et al., 2010; Hooda et al., 1997; Planquart et al, 1999). Morrera et al. (2002) reported that soil type had a greater effect on uptake of heavy metals by sunflower than the actual dose of sewage sludge. Soil type is a variable that might affect seed germination when biosolids are applied on soil (Oleszcuk et al., 2012). It also appears to be a toxicity modulator to collembolans in agricultural soils (Domene et al., 2010). Factors affecting the solubility of heavy metals in sewage sludge-amended soils are either soil specific or environmental (Ashworth and Alloway, 2008). Another factor related to soil type is soil texture, which showed significant effects on heavy metal bioavailability for barley (Soriano-Disla et al., 2008). Environmental factors including temperature, soil moisture, rainfall, type of land use and time of input of the organic amendment all impact in the bioavailability of heavy metals in sludge-amended soils. These variables have a significant short-term effect especially on organic matter contents. However, long-term field experiments concluded that organic matter is also significant in metal extractability over time (Walter et al., 2002; Petruzzelli et al., 1997).

Soil specific factors tend to be significant in the long-term (Oleszczuk et al., 2012). Metals such as Cu, Ni and Pb are likely to form organo-metal complexes, which may become more hazardous than their original form. In other words, the effects of co-solubilization may result in increased mobility and bioavailability of metals (Ashworth and Alloway, 2008) over time following application in sludge. In the light of this evidence, the total concentrations of heavy metals should not be derived solely to assess the toxicity and availability of PTEs but must give consideration to the total amount of pre-existing (natural background) trace elements and soil pH, but also site specific environmental and soil variables should be considered, when setting up the limits for the application of biosolids.

3.2.1 Key findings

- Cu is likely to form organo-metal complexes that may be more hazardous to key receptors.
- Bioavailability of metals typically decreases over time.
- Weathering may promote leaching of PTEs and other contaminants.
- Legislation should consider the specific environmental and soil variables (soil type, texture, permeability) for each site.

3.3 Surface application of biosolids

Grazing animals represent a possible pathway for the intake of contaminants from sewage sludges by humans (Schowanek et al., 2004). Restrictions on grazing following the application of biosolids or sludges in the UK is given by the Code of Practice For Agriculture Use of Sewage Sludge (DoE, 1996) and states that no grazing is permitted 3 weeks after the application of biosolids. Aitken (1997), who conducted a short-term experiment on the surface-application of biosolids within the prescribed agricultural limits in the UK, revealed that zootoxic levels of Cu and Fe persisted even after the "no grazing period" advocated by the guidelines. Cu appeared toxic in these studies up to 8 days and Fe 21 days, respectively after the 3-week period.

Animals fed with biosolids during their normal diet in the controlled experiment accumulated Cd, Cu, and Pb in their kidneys and livers. Indicating that the current UK limits for concentrations of Cd and Pb in sewage sludge-amended soils do not provide the desired safety margin (Hill et al., 1998). Wilkinson et al. (2001) tested the accumulation of heavy metals in sheep tissues and organs in a farm environment with repeated applications of biosolids at levels close to the UK Statutory Limits and concluded that no margin of safety existed for Cd in sludge-amended soils within current limits. Other long-term experiments conducted in a field setting concluded that application of sludge to pasture does not automatically increase the accumulation of heavy metals in the tissues of animals, but rates of accumulation of heavy metals become altered (Rhind et al., 2005). Nevertheless, experiments in controlled environment do not have to necessarily reflect the real world situation. The research by Rhind et al. (2005) identified many factors that may influence PTE uptake and suggested consideration of each class of animal alongside the spectrum of PTEs separately. In the light of this evidence, a revision of the "no grazing period" seemed to be an appropriate measure to ensure protection of all the receptors. Changes in the sludge legislation that would allow for better protection of grazing animals, from accumulation of heavy metals in their tissues, may be hard to implement due to need for extensive testing of individual animal types and different ways of exposure. On the other hand, the current Cd limit concentrations in sludge-amended soils should be revised in order to decrease the possible negative impacts on grazing animals. With regard to leaching and the type of application of biosolids (surface/subsurface), higher leaching losses were attributed to subsurface application of biosolids (Gove et al., 2010).

3.3.1 Key findings

- Cu may have zootoxic effects even after the "no grazing period".
- Current Cd limits do not provide a sufficient safety margin to protect grazing animals.
- Different classes of animals have shown different effects with regard to heavy metal accumulation in their tissues.

3.4 Increase of ambient temperature

Future changes in ambient air (and hence soil) temperatures will also tend to affect the bioavailability of PTEs such as heavy metals, since the higher temperature allows for rapid degradation of organic matter (Antoniadis et al., 2007; 2010), which provides a binding medium for heavy metals in soils. This may result in long-term higher bioavailabilities of metals. Alternatively, and confoundingly, droughts may not cause problems to plants with regard to heavy metals introduced by sludges (Pascual et al., 2004). As droughts may not promote changes in the metal pools or fractions, and thus not contribute to higher availability of metals. On the other hand, fissures and cracks in soil caused by higher temperatures may contribute to leaching behaviour of heavy metals (Proust et al., 2011).

Based on future UK climate projections, the average temperature in the future is likely to increase (Jenkins et al., 2009). This will have an effect on biosolids applied to agricultural land. In order to avoid possible risks related to effects of heavy metals, future sludge legislation should acknowledge the effects of higher ambient temperature and more sudden changes in weather on the availability of heavy metals and organic and other PTEs in sludges.

3.4.1 Key findings

- Higher temperatures help degrade organic matter that binds heavy metals.
- Fissures and cracks in soil caused by droughts may contribute to leaching of contaminants.

3.5 Biosolids contain Bisphenol A

Numerous pathways have been identified that could relate to the exposure of BPA and other organic compounds from sludge-amended soils (US-EPA, 1993). Soil, plants, animals, groundwater and humans were identified as the possible receptors for organic compounds in sludge. Lysimeter experiments demonstrated concern about disrupting chemicals and their mobility to aquifer and surface waters (Dizer et al., 2002). Despite the possible exposure pathways, direct or chronic effects of BPA, to all the receptors are yet to be determined (Zhang et al., 2015). It may be hard to assess effects of individual organic compounds due to their occurrence in complex mixtures of different organic compounds in sludge. The addition of biosolids to agricultural soils increases concentrations of BPA, which may pose risks to living organisms that are exposed to amended soil. Zhang et al. (2015) examined the effects of BPA and other endocrine disruptors present in sludges, and have observed this effect in the long-term experiment (13 years). In relation to the "no grazing period" the current 21 days limit seems to be an appropriate measure in relation to BPA; however, this regards sites where biosolids have not been extensively applied over long time. Such a concern is due to increased amounts of the recalcitrant fraction of BPA, which may pose currently unknown risks to organisms ingesting amended soils. Other studies focusing on sewage sludge-amended pastures, where sheep ingested mixture of compounds, reported adverse effect on foetuses as well as multiple organs (Rhind, 2009; Lind et al., 2009, 2010; Rhind et al., 2010; Hombach-Klonisch et al., 2013). However, a direct causal link was not established. The types of sludge treatment determine the amount of BPA in biosolids, where aerobic digestion promotes its biodegradation (Tran et al., 2015). The positive effects of aerobic conditions and higher temperatures on the biodegradation of BPA were also confirmed by Press-Kristensen et al. (2008). This work suggested that the aerobic treatment of biosolids might reduce the risk of exposure to BPA introduced by sludge application in agriculture. Despite that, BPA concentrations along with other organic compounds present in sludges should be monitored due to the current inconclusive research about their potential effects to soils from their application in sludges (Zhang et al., 2015).

3.5.1 Key findings

- Direct and chronic effects of BPA are yet to be determined in sludge-amended agricultural soils.
- It is hard to assess the effects of individual organic compounds in sludges, as they are present in complex mixtures.
- BPA quickly degrades, when exposed to atmospheric conditions.
- Aerobic digestion is a favourable treatment method with regard to BPA.
- Organic compounds in sludges should be monitored in greater detail to reduce future risks.

3.6 In the light of previous attempt

In the year 2000, the third draft revision of the EU Sludge Directive took into consideration the advanced treatment and conventional treatment of sludges with respect to different sites (European Commission, 2000). With regard to pastures, which were highlighted in this work in terms of “no grazing period”, conventional treatments required deep injection and no grazing for the following subsequent six weeks. This should be appropriate measure regarding the “no grazing period” based on the reviewed evidence in this study.

The European Commission draft promoted a lowering of the limit concentrations of PTE's and hazardous substances in sludge with respect to pH even below the current UK statutory limits. Such a precaution is compatible with the revelations in this study as it increases the safety margin for all the heavy metals as well as taking into consideration soil pH as modulation factor for all the metals. The prevention of pollution plan proposed by the draft Directive aimed at sequential lowering of heavy metal limit concentrations. The idea of regularly reviewing the limits seems by the evidence from this study as reasonable regarding future climate change and its effects on contaminants in sludges. Various organic compounds (LAS, DEHP, PCB, NPE, AOX, PCDDF) in sludges with their limit concentrations are also acknowledged in the proposed draft. Despite the valid approach introduced in the Sludge Directive draft, no action by Member States has since been taken.

4 Conclusions

Agricultural re-use of sludge provides a sustainable approach to increasing the re-use of a valuable product. The risks connected to its re-use should be regularly reviewed due to the dynamic and evolving nature of the waste composition. Even though there is a lack of clear evidence showing the direct cause and effect of sludge applications applied within the regulatory limits, future regulation should provide safety margin based on the precautionary principle to reduce possible risks to minimum. This work examined a relevant body of literature and analysed risks connected with biosolid additions to agricultural soil, in order to determine whether the current European sludge legislation/UK Code of Practice For Agriculture Use Sewage Sludge is sufficiently protective of human health and the wider environment. The review of pertinent literature helped to reveal factors contributing to understanding of the different behaviours of biosolids in agricultural soils, which tended to increase or decrease the possible risks arising from sludge re-use. The current out-dated legislation governing sludge re-use proved efficient; however, such provided little margin of safety in some areas and did not reflect the changing character of domestic as well as industrial wastes. There was a fairly good understanding of acute effects of heavy metals introduced with sludges into soils. On the other hand, evidence about the chronic effects of elements and compounds added to soils during normal agricultural practice remained sparse. Given this uncertainty, long-term sludge continuous amendments provide the highest risk to receptors. Cd limit concentrations showed no margin of safety with regard to repeated surface applications of biosolids and their effects on grazing animals. The continuous monitoring of sludges as well as sludge-amended soils with regard to chronic effects of sludge application to agricultural soils should be encouraged. Any future sludge legislation should try to reflect all the possible factors affecting the possible risks from sludge re-use. Evidence regarding Cu did not reveal significant risks related to the application of biosolids within the permissible concentrations. The type of sludge and its mode of application largely determine the fate of BPA in soils since it tends to degrade quickly in the environment. Even so the fate of other organics along with BPA should be monitored to mitigate the possible risks.

The logical next step in order to prevent possible risks arising from sludge re-use in agriculture is to initiate a discussion with all the stakeholder groups, on changing the current legislation to reflect the findings of this work. Further research regarding the appropriateness of sludge legislation should consider the effects of emerging contaminants and nanomaterials that are being introduced into wastewater streams and determine whether there is a need to regulate these contaminants.

4.1 Key recommendations

- “No grazing” period of 21 days given by the Code of Practice For Agriculture Use of Sewage Sludge should be extended in order to protect grazing animals.
- The current Cd limit concentrations in sludges used in agriculture should be lower to protect all receptors.
- Different modulating factors, which may influence the risks arising from the application of biosolids and sludges to agricultural soils, should be acknowledged when setting up the limit concentrations.

- The fate of organic compounds in sludges should be monitored in greater depth to prevent potential future risks.

REFERENCES

Aitken, M.N. (1997), "Short-term leaf surface adhesion of heavy metals following application of sewage sludge to grassland", *Grass and Forage Science*, 52(1), pp. 73-85.

Antoniadis, V., Tsadilas, C.D. and Ashworth, D.J. (2007), "Monometal and competitive adsorption of heavy metals by sewage sludge-amended soil", *Chemosphere*, 68(3), pp. 489-494.

Antoniadis, V., Tsadilas, C.D. and Samaras, V. (2010), "Trace element availability in a sewage sludge-amended cotton grown Mediterranean soil", *Chemosphere*, 80(11), pp. 1308-1313.

Ashworth, D.J. and Alloway, B.J. (2008), "Influence of dissolved organic matter on the solubility of heavy metals in sewage-sludge-amended soils", *Communications in Soil Science and Plant Analysis*, 39(3-4), pp. 538-550.

Atanassova, I., Velichkova, N. and Teoharov, M. (2012), "Heavy metal mobility in soils under the application of sewage sludge", *Bulgarian Journal of Agricultural Science*, 18(3), pp. 396-402.

Benítez, E., Romero, E., Gómez, M., Gallardo-Lara, F. and Nogales, R. (2001), "Biosolids and biosolids-ash as sources of heavy metals in a plant-soil system", *Water, Air, and Soil Pollution*, 132, (1-2) pp. 75-87.

Canet, R., Pomares, F., Tarazona, F. and Estela, M. (1998), "Sequential fractionation and plant availability of heavy metals as affected by sewage sludge applications to soil", *Communications in Soil Science and Plant Analysis*, 29(5-6), pp. 697-716.

Card, A.J., Ward, J.R. and Clarkson, P.J. (2012), “Beyond FMEA: The structured what-if technique (SWIFT)”, *Journal of Healthcare Risk Management*, 31(4), pp. 23–29.

Carrondo M. J. T., Lester J.N., Perry R. and Stoyeland S. (1978), *Analysis of Heavy Metals in Sewage Sludge, Sewages and Final Effluent; Final Report, to the Department of the Environment for Contracts DGR/480/66 and DGR/480/240*, Public Health and Water Resource Engineering Section, Civil Engineering Department, Imperial College, London.

CEC (1985), *Council Directive 85/467/EEC of 1 October 1985 amending for the sixth time (PCBs/PCTs) Directive 76/769/EEC on the approximation of the laws, regulations and administrative provisions of the Member States relating to restrictions on the marketing and use of certain dangerous substances and preparations*. Official Journal L 269, 11/10/1985 pp. 0056 – 0058.

CEC (1986), *Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC)*. Official Journal L 181, 04/07/1986 pp. 0006 – 0012.

CEC (1991), *Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment*, Official Journal L 135, 30/05/1991 pp. 0040 – 0052.

Chander, K. and Brookes, P.C. (1991), “Effects of heavy metals from past applications of sewage sludge on microbial biomass and organic matter accumulation in a sandy loam and silty loam UK soil”, *Soil Biology and Biochemistry*, 23, pp. 927-932.

Chaudri, A.M., Knight, B.P., Barbosa-Jefferson, V.L., Preston, S., Paton, G.I., Killham, K., Coad, N., Nicholson, F.A., Chambers, B.J. and McGrath, S.P. (1999), "Determination of acute Zn toxicity in pore water from soils previously treated with sewage sludge using bioluminescence assays", *Environmental Science and Technology*, 33(11), pp. 1880-1885.

Cordell D., Drangert, J.O. and White, S. (2009) “The story of phosphorus: Global food security and food for thought”, *Global Environmental Change*, 19, (2), pp. 292-305.

Cuevas, G. and Walter, I. (1999), "Chemical fractionation of heavy metals in a soil amended with repeated sewage sludge application", *Science of the Total Environment*, 226(2), pp. 113.

DEFRA (2002), *Sewage treatment in the UK: UK Implementation of the EC Urban Waste Water Treatment Directive*. Available at: https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/69582/pb6655-uk-sewage-treatment-020424.pdf (Accessed: 30 August 2015).

Delclos, K.B., Camacho, L., Lewis, S.M., Vanlandingham, M.M., Latendresse, J.R., Olson, G.R., Davis, K.J., Patton, R.E., da Gosta, G.G., Woodling, K.A., Bryant, M.S., Chidambaram, M., Trbojevich, R., Juliar, B.E., Felton, R.P. and Thorn, B.T. (2014), “Toxicity evaluation of Bisphenol A administered by gavage to sprague dawley rats from gestation day 6 through postnatal day 90”, *Toxicological Sciences*, 139(1), pp.174–197.

Delgado Arroyo, M.D.M., Porcel Cots, M. A., Miralles De Imperial Hornedo, R., Beltrán Rodríguez, E.M., Beringola Beringola, L. and Martín Sánchez, J. V. (2002), "Sewage sludge compost fertilizer effect on maize yield and soil heavy metal concentration", *Revista Internacional de Contaminacion Ambiental*, 18(3), pp. 147-150.

Delgado, G., Aranda, V., Pérez-Lomas, A.L., Martín-García, J.M., Calero, J. and Delgado, R. (2012), "Evolution of available heavy metals in soils amended with sewage sludge cocompost", *Compost Science & Utilization*, 20(2), pp. 105-119.

Dizer, H., Fischer, B., Sepulveda, I., Loffredo, E., Senesi, N., Santana, F. and Hansen, P.D. (2002), “Estrogenic effect of leachates and soil extracts from lysimeters spiked with sewage sludge and reference endocrine disrupters”, *Environmental Toxicology*, 17(2), pp. 105–112.

DoE, (1996), *Code of Practice For Agriculture Use of Sewage Sludge*. Available at: http://adlib.everysite.co.uk/resources/000/247/164/sludge-report.pdf (Accessed: 30 August 2015).

Domene, X., Colón, J., Uras, M. V., Izquierdo, R., Àvila, A. and Alcañiz, J. M. (2010), "Role of soil properties in sewage sludge toxicity to soil collembolans", *Soil Biology and Biochemistry*, 42(11), pp. 1982-1990.

Eisler, R. (1985), Cadmium hazards to fish, wildlife, and invertebrates: A synoptic review, *Contaminant Hazard Reviews, Biological Report,* 85(1.2), Report No. 2.

Escrig, I., and Morell, I. (1998), "Soil pollution by some heavy metals from sewage sludge", *Toxicological and Environmental Chemistry*, 66(1-4), pp. 243-252.

European Commission (2002), *Disposal and Recycling Routes for Sewage Sludge. Economic subcomponent report, DG Environment-B/2*, Brussels, 29 January.

European Commission, (2000), *Working document on sludge, 3rd draft, DG Environment E.3.*, Brussels, 27 April.

Fromme, H., Kuchler, T., Otto, T., Pilz, K., Muller, J. and Wenzel, A., (2002), “Occurrence of phthalates and bisphenol A and F in the environment”, *Water Research*, 36, pp.1429–1438.

FWR, (2011), *A Review of Current Knowledge, SEWAGE SLUDGE: Operational and Environmental Issues*. Available at: http://www.fwr.org/sludge.pdf (Accessed: 30 August 2015).

Fytili, D. and Zabaniotou, A. (2008), “Utilisation of sewage sludge in EU application of old and new methods – a review”, *Renewable and Sustainable Energy Reviews*, 12(1), pp. 116–140.

Gendebien, A. (2009), *Environmental, economic and social impacts of the use of sewage sludge on land. Draft Summary Report 1, Assessment of existing knowledge*, Milieu Ltd and WRC for the European Commission.

Georgieva, S.S., McGrath, S. P., Hooper, D.J. and Chambers, B.S. (2002), "Nematode communities under stress: The long-term effects of heavy metals in soil treated with sewage sludge", *Applied Soil Ecology*, 20(1), pp. 27-42.

Gibbs, P.A., Chambers, B.J., Chaudri, A.M., McGrath, S.P., Carlton-Smith, C.H., Bacon, J.R., Campbell, C. D. and Aitken, M. N. (2006a), "Initial results from a long-term, multi-site field study of the effects on soil fertility and microbial activity of sludge cakes containing heavy metals", *Soil Use and Management*, 22(1), pp. 11-21.

Gibbs, P.A., Chambers, B.J., Chaudri, A.M., McGrath, S.P. and Carlton-Smith, C.H. (2006b), "Initial results from long-term field studies at three sites on the effects of heavy metal-amended liquid sludges on soil microbial activity", *Soil Use and Management*, 22(2), pp. 180-187.

Gondek, K., Filipek-Mazur, B. and Koncewicz-Baran, M. (2010), "Content of heavy metals in maize cultivated in soil amended with sewage sludge and its mixtures with peat", *International Agrophysics*, 24(1), pp. 35-42.

Gove, L., Nicholson, F.A., Cook, H. F. and Beck, A.J. (2002), "Comparison of the effect of surface application and subsurface incorporation of enhanced treated biosolids on the leaching of heavy metals and nutrients through sand and sandy loam soils", *Environmental technology*, 23(2), pp. 189-198.

Government Social Research, (2010), *Rapid Evidence Assessment Toolkit Index*. Available at: http://www.civilservice.gov.uk/networks/gsr/resources-and-guidance/rapid-evidence-assessment (Accessed: 30 August 2015).

Harrison, E.Z., Oakes, S.R., Hysell M. and Hay A. (2006), “Organic chemicals in sewage sludges”, *Science of The Total Environment*, 367(2–3), pp. 481-497.

Hill, J., Stark, B.A., Wilkinson, J.M., Curran, M.K., Lean, I.J., Hall, J.E. and Livesey, C.T. (1998), "Accumulation of potentially toxic elements by sheep given diets containing soil and sewage sludge. 1. Effect of type of soil and level of sewage sludge in the diet", *Animal Science*, 67(1), pp. 73-86.

Hombach-Klonisch, S., Danescu, A., Begum, F., Amezaga, M.R., Rhind, S.M., Sharpe, R.M., Evans, N. P., Bellingham, M., Cotinot, C., Mandon-Pepin, B., Fowler, P. A. and Klonisch, T. (2013). “Periconceptional changes in maternal exposure to sewage sludge chemicals disturbs fetal thyroid gland development in sheep”, *Molecular and Cellular Endocrinology*, 367 (1–2), pp. 98–108.

Hooda, P.S., McNulty, D., Alloway, B.J. and Aitken, M.N. (1997), "Plant availability of heavy metal in soils previously amended with heavy applications of sewage sludge", *Journal of the Science of Food and Agriculture*, 73(4), pp. 446-454.

Hooda, P. S. and Alloway, B. J. (1993), “Effects of time and temperature on the bioavailability of Cd and Pb from sludge-amended soils”, *Journal of Soil Science*, 44(1), pages 97–110.

Hospido, A., Moreira, M.T., Martín, M., Rigola, M. and Feijoo, G. (2005), “Environmental Evaluation of Different Treatment Processes for Sludge from Urban Wastewater Treatments: Anaerobic Digestion versus Thermal Processes”, *International Journal of Life Cycle Assessment*, 10(5), pp. 336-345.

IC Consultants Ltd London, (2001), *Pollutants in urban waste water and sewage sludge*. Luxembourg: Office for Official Publicatinos of the European Communities.

Jenkins, G.J., Murphy, J.M., Sexton, D.M.H., Lowe, J.A., Jones, P. and Kilsby, C.G. (2009), *UK Climate Projections: Briefing report*. Exeter: Met Office Hadley Centre.

Jones, K. C. and Sewart A. P. (1997), “Dioxins and furans in sewage sludges: A review of their occurrence and sources in sludge and of their environmental fate, behavior, and significance in sludge amended agricultural systems”, *Critical Reviews in Environmental Science and Technology*, 27(1), pp. 1-86.

Kalembasa, D. and Pakuła, K. (2009), "Heavy metal fractions in soils fertilized with sewage sludge", *Environment Protection Engineering*, 35(2), pp. 157-164.

Kidd, P.S., Domínguez-Rodríguez, M.J., Díez, J. and Monterroso, C. (2007), "Bioavailability and plant accumulation of heavy metals and phosphorus in agricultural soils amended by long-term application of sewage sludge", *Chemosphere*, 66(8), pp. 1458-1467.

Lind, P. M., Gustafsson, M., Hermsen, S. A. B., Larsson, S., Kyle, C. E., Örberg, J. and Rhind, S. M., (2009), “Exposure to pastures fertilised with sewage sludge disrupts bone tissue homeostasis in sheep”, *Science of the Total Environ*ment, 407(7), pp. 2200–2208.

Lind, P. M., Öberg, D., Larsson, S., Kyle, C. E., Örberg and Rhind, S. M., (2010). “Pregnant ewes exposed to multiple endocrine disrupting pollutants through sewage sludge-fertilized pasture show an anti-estrogenic effect in their trabecular bone”, *Science of the Total Environ*ment, 408(11), pp. 2340–2346.

Lombi, E. and Gerzabek, M.H. (1998), "Determination of mobile heavy metal fraction in soil: Results of a pot experiment with sewage sludge", *Communications in Soil Science and Plant Analysis*, 29(17-18), pp. 2545-2556.

Loveland, P.J. and Thompson T.R.E. (2000), *The Draft Revised EC Sewage Sludge Directive (March 2000) in Relation to Concentrations of Copper, Lead, Nickel and Zinc in the Soils of England and Wales*, R&D Project Record P5/049/01, Environment Agency.

Łuczkiewicz, A. (2006), "Soil and groundwater contamination as a result of sewage sludge land application", *Polish Journal of Environmental Studies*, 15(6), pp. 869-876.

MAFF (1987), *The use of sewage sludge on agricultural land*. Ministry of Agriculture Fisheries and Food/ADAS booklet 2409. Revised 1987. London: MAFF Publications.

MAFF (1993), *Review of the Rules for Sewage Sludge Application to Agricultural Land. Soil Fertility Aspects of Potentially Toxic Elements*, Report of the Independent Scientific Committee. London: MAFF Publications.

McGrath, S.P., Knight, B., Killham, K., Preston, S. and Paton, G.I. (1999), "Assessment of the toxicity of metals in soils amended with sewage sludge using a chemical speciation technique and a lux-based biosensor", *Environmental Toxicology and Chemistry*, 18(4), pp. 659-663.

Mihalache, M., Ilie, L. and Madjar, R. (2014), "Translocation of heavy metals from sewage sludge amended soil to plant", *Revue Roumaine de Chimie*, 59(2), pp. 81-89.

Miller W. P., Martins D. C. and Zelazny L. W., (1986), “Effect of sequence in extraction of trace metals from soils”, *Soil Science Society of America Journal,* 50, pp. 598-601.

Moolenaar, S.W. and Beltrami, P. (1998), "Heavy metal balances of an Italian soil as affected by sewage sludge and bordeaux mixture applications", *Journal of Environmental Quality*, 27(4), pp. 828-835.

Moreno, J.L., García, C., Hernández, T. and Pascual, J.A. (1996), "Transference of heavy metals from a calcareous soil amended with sewage-sludge compost to barley plants", *Bioresource Technology*, 55(3), pp. 251-258.

Moreno, J.L., García, C., Hernández, T. and Ayuso, M. (1997), "Application of composted sewage sludges contaminated with heavy metals to an agricultural soil: Effect on lettuce growth", *Soil Science and Plant Nutrition*, 43(3), pp. 565-573.

Moreno, J. L., García, C. and Hernández, T. (1998), "Changes in organic matter and enzymatic activity of an agricultural soil amended with metal-contaminated sewage sludge compost", *Communications in Soil Science and Plant Analysis*, 29(15-16), pp. 2247-2262.

Moreno, J. L., Hernández, T., Garcia, C., (1999), "Effects of a cadmium-contaminated sewage sludge compost on dynamics of organic matter and microbial activity in an arid soil", *Biology and Fertility of Soils*, 28(3), pp. 230-237.

Morera, M.T., Echeverría, J.C. and Garrido, J.J. (2002), "Bioavailability of heavy metals in soils amended with sewage sludge", *Canadian Journal of Soil Science*, 82(4), pp. 433-438.

Morera, M.T., Echeverría, J.C. and Garrido, J.J. (2001), "Mobility of heavy metals in soils amended with sewage sludge", *Canadian Journal of Soil Science*, 81(4), pp. 405-414.

Newbold, R. R., Jefferson, W.N. and Padilla-Banks, E., (2007), “Long-term adverse effects of neonatal exposure to Bisphenol A on the murine female reproductive tract” *Reproducive Toxicol*ogy, 24(2), pp. 253–258.

Obbard, J. P. and Jones, K. C., (2000), "Measurement of symbiotic nitrogen-fixation in leguminous host-plants grown in heavy metal-contaminated soils amended with sewage sludge", *Environmental Pollution*, 111(2), pp. 311-320.

Obrador, A., Rico, M. I., Alvarez, J.M. and Mingot, J. (1998), "Mobility and extractability of heavy metals in contaminated sewage sludge-soil incubated mixtures", *Environmental Technology*, 19(3), pp. 307-314.

Oleszczuk, P., Malara, A., Josko, I. and Lesiuk, A., (2012), "The phytotoxicity changes of sewage sludge-amended soils", *Water, Air, and Soil Pollution*, 223(8), pp. 4937-4948.

Parmesan, C. and Yohe, G., (2003), “A globally coherent fingerprint of climate change impacts across natural systems”, *Nature*, 421(6918), pp. 37-42.

Pascual, I., Antolín, M. C., García, C., Polo, A. and Sánchez-Díaz, M., (2004), "Plant availability of heavy metals in a soil amended with a high dose of sewage sludge under drought conditions", *Biology and Fertility of Soils*, 40(5), pp. 291-299.

Petruzzelli, G., Petronio, B. M., Gennaro, M. C., Vanni, A., Liberatori, A., Barbafieri, M. and Pezzarossa, B., (1997), "Residual effect of application of sewage sludge on heavy metals sorption by soil", *Annali di Chimica*, 87(11-12), pp. 733-742.

Pisarek, I. and Moliszewska, E. B., (2004), "Effect of the addition of sewage sludge on the composition of humic substances, content of heavy metals and the presence of Trichoderma spp. in the soil", *Polish Journal of Soil Science*, 37(2), pp. 171-179.

Planquart, P., Bonin, G., Prone, A. and Massiani, C. (1999), "Distribution, movement and plant availability of trace metals in soils amended with sewage sludge composts: application to low metal loadings", *Science of the Total Environment*, 241(1-3), pp. 161-179.

Press-Kristensen, K., Lindblom, E., Schmidt, J.E. and Henze, M. (2008), “Examining the biodegradation of endocrine disrupting bisphenol A and nonylphenol in WWTPs”, *Water Science and Technology,* 57, pp. 1253–1258.

Proust, D., Caillaud, J., Fontaine, C., Fialin, M., Courbe, C. and Dauger, N. (2011), "Fissure and mineral weathering impacts on heavy metal distribution in sludge-amended soil", *Plant and Soil*, 346(1), pp. 29-44.

Raffaella, B., Ettore, C., Elisabetta, G. and Vittoria, L., (1997), "Soil accumulation of heavy metals after repeated application of sludges", *Fresenius Environmental Bulletin*, 6(11-12), pp. 642-647.

Rastetter, N. and Gerhardt, A. (2014), "Toxic potential of different types of sewage sludge as fertiliser in agriculture: ecotoxicological effects on aquatic and soil indicator species", *Journal of Soils and Sediments*, 15(3), pp. 565-577.

Rhind, S.M., Kyle, C.E. and Owen, J. (2005), "Accumulation of potentially toxic metals in the liver tissue of sheep grazed on sewage sludge-treated pastures", *Animal Science*, 81(1), pp. 107-113.

Rhind, S.M. (2009), “Anthropogenic pollutants: a threat to ecosystem sustainability”, *Philosophical Transactions of the Royal Society B,* 364, pp. 3391–3401.

Rhind, S.M., Kyle, C.E., Mackie, C., McDonald, L., Zhang, Z.L., Duff, E.I., Bellingham, M., Amezaga, M.R., Mandon-Pepin, B., Loup, B., Continot, C., Evans, N.P., Sharpe, R.M. and Fowler, P.A. (2010). “Maternal and fetal tissue accumulation of selected endocrine disrupting compounds (EDC) following exposure to sewage sludge-treated pastures before or after conception”. *Journal of Environmental Monitor*ing, 12, pp. 1582–1593.

Rossi, G., Pennelli, B., Socciarelli, S. and Figliolia, A. (2002), “Effects of medium-term amendment with sewage sludges on heavy metal distribution in soil”, in: A. Violante, P. M. Huang, J.-M. Bollag and L. Gianfreda, (Ed), *Developments in Soil Science*, 28, Part A, pp. 99-107.

Sánchez-Martín, M.J., García-Delgado, M., Lorenzo, L.F., Rodríguez-Cruz, M.S. and Arienzo, M. (2007), "Heavy metals in sewage sludge amended soils determined by sequential extractions as a function of incubation time of soils", *Geoderma*, 142(3-4), pp. 262-273.

Samaras, V. and Kallianou, C. (2000), "Effect of sewage sludge application on cotton yield and contamination of soils and plant leaves", *Communications in Soil Science and Plant Analysis*, 31(3-4), pp. 331-343.

Schnaak, W., Küchler, T., Kujawa, M., Henschel, K.P., Süßenbach, D. and Donau, R. (1997), “Organic contaminants in sewage sludge and their ecotoxicological significance in the agricultural utilization of sewage sludge”, *Chemosphere*, 35(1–2), pp. 5-11.

Schowanek D., Carr R., David H., Douben P., Hall J., Kirchmann H., Patria L., Sequi P., Smith S. and Webb S., (2004), “A risk-based methodology for deriving quality standards for organic contaminants in sewage sludge for use in agriculture—Conceptual Framework”, *Regulatory Toxicology and Pharmacology*, 40(3), pp. 227-251.

Serrano, S., Garrido, F., Campbell, C.G. and García-González, M.T. (2005), “Competitive sorption of cadmium and lead in acid soils of Central Spain”, *Geoderma*, 124(1-2), pp. 91-104.

Sieciechowicz, A., Sadecka, Z., Myszograj, S., Włodarczyk-Makuła, M., Wiśniowska, E. and Turek, A. (2014), "Occurrence of heavy metals and PAHs in soil and plants after application of sewage sludge to soil", *Desalination and Water Treatment*, 52(19-21), pp. 4014-4026.

Singh, R.P. and Agrawal, M. (2008), “Potential benefits and risks of land application of sewage sludge”, *Waste Management*, 28(2), pp. 347-358.

Smith, S.R. (1997), "Rhizobium in soils contaminated with copper and zinc following the long-term application of sewage sludge and other organic wastes", *Soil Biology and Biochemistry*, 29(9-10), pp. 1475-1489.

Soriano-Disla, J.M., Gómez, I., Guerrero, C., Jordan, M.M. and Navarro-Pedreño, J. (2008), "Soil factors related to heavy metal bioavailability after sewage sludge application", *Fresenius Environmental Bulletin*, 17(11) A, pp. 1839-1845.

Soriano-Disla, J.M., Gómez, I., Navarro-Pedreño, J. and Jordán, M.M. (2014), "The transfer of heavy metals to barley plants from soils amended with sewage sludge with different heavy metal burdens", *Journal of Soils and Sediments*, 14(4), pp. 687-696.

Stöven, K. and Schnug, E. (2009), "Long term effects of heavy metal enriched sewage sludge disposal in agriculture on soil biota", *Landbauforschung Volkenrode*, 59(2), pp. 131-138.

Smith, S.R., (2009), "Organic contaminants in sewage sludge (biosolids) and their significance for agricultural recycling", *Philosophical Transactions of the Royal Society* A, 367, pp. 4005–4041.

Song, S., Song, M., Zeng, L., Wang, T., Liu, R., Ruan, T. and Jiang, G. (2014), "Occurrence and profiles of Bisphenol analogues in municipal sewage sludge in China", *Environmental Pollut*ion, 186, pp. 14–19.

SI (1989) United Kingdom Statutory Instrument No. 1263, *The Sludge (Use in Agriculture) Regulations 1989*, HMSO: London.

Stuczynski, T., Pistelok, F., Siebielec, G., Kukla, H., Daniels, W., Chaney, R. and Pantuck, K. (2000), "Biological aspects of metal waste reclamation with sewage sludge in Poland", *In Proceedings of the Symposium on Mining, Forest and Land Restoration: The Successful Use of Residuals/Biosolids/Organic Matter for Reclamation Activities*, Denver, CO.

Stylianou M. A., Kollia D., Haralambous K. J., Inglezakis V. J., Moustakas K. G. and Loizidou M. D., (2007), "Effect of acid treatment on the removal of heavy metals from sewage sludge", *Desalination*, 215(1–3), pp. 73-81.

Tack, F.M.G. (2010), "Trace elements: general soil chemistry, principles and processes", in: Hooda, P. (Ed), *Trace Elements in Soils*, Wiley-Blackwell, pp. 9-32.

Theophanides T. ans Anastassopoulou J. (2012), "Copper and carcinogenesis", *Critical Reviews in Oncology/Hematology*, 42 pp. 57–64.

Tran, B.C., Teil, M.J., Blanchard, M., Alliot, F. and Chevreuil, M. (2015), "Fate of phthalates and BPA in agricultural and non-agricultural soils of the Paris area (France)", *Environmental Science Pollution Research International*, 22(14) pp. 11118-26.

Tsadilas, C. D , Matsi T., Barbayiannis N. and Dimoyiannis D., (1995), "Influence of sewage sludge application on soil properties and on the distribution and availability of heavy metal fractions", *Communications inSoil Science and Plant Analysis,* 26(15-16) pp. 2603-2619.

UKWIR (2007), *Effects of sewage sludge applications to agricultural soils on soil microbial activity and the implication for agricultural productivity and long-term soil fertility? Phase III*. Available at: http://randd.defra.gov.uk/Document.aspx?Document=SP0130_6505_FRP.pdf (Accessed: 30 August 2015).

US-EPA (1993), “Standards for the use and disposal of sewage sludges”, *Federal Register,* 58(32), pp. 9248–9415.

Walter, I., Martínez, F., Alonso, L. and Gabriela Cuevas, J.D.G. (2002), "Extractable soil heavy metals following the cessation of biosolids application to agricultural soil", *Environmental Pollution,* 117(2), pp. 315-321.

Walter, I., Martínez, F. and Cala, V. (2006), "Heavy metal speciation and phytotoxic effects of three representative sewage sludges for agricultural uses", *Environmental Pollution*, 139(3), pp. 507-514.

Weightman, R. M. (2006), "Heavy metal and microbial contamination of Valerian (Valeriana officinalis L.) roots grown in soil treated with sewage sludge", *Journal of Herbs, Spices and Medicinal Plants*, 12(3), pp. 77-88.

Weissenhorn, I., Mench, M. and Leyval, C. (1995), "Bioavailability of heavy metals and arbuscular mycorrhiza in a sewage-sludge-amended sandy soil", *Soil Biology and Biochemistry*, 27(3), pp. 287-296.

Wilkinson, J. M., Hill, J. and Livesey, C. T. (2001), "Accumulation of potentially toxic elements in the body tissues of sheep grazed on grassland given repeated applications of sewage sludge", *Animal Science*, 72(1), pp. 179-190.

Zhang, Z., Le Velly, M., Rhind, S.M., Kyle, C.E., Hough, R.L., Duff, E.I. and McKenzie, C., (2015), “A study on temporal trends and estimates of fate of Bisphenol A in agricultural soils after sewage sludge amendment”, *Science of the Total Environment*, 515–516(15), pp. 1-11.

APPENDIX

Appendix A – Reviewed studies

No.	Author(s)	Study location	Focus
1	Aitken (1997)	UK	Surface adhesion of biosolids
2	Antoniadis (2007)	Greece	Adsorption
3	Antoniadis (2010)	Greece	Bioavalability
4	Ashworth and Alloway (2008)	UK	Partitioning and sorption
5	Atanassova et al. (2012)	Bulgaria	Mobility of heavy metals
6	Benítez et al. (2000)	Spain	Mobility of heavy metals
7	Canet et al. (1998)	Spain	Extractibility and bioavailability
8	Chaudri et al. (1999)	UK	Ecotoxicity of metals
9	Delgado et al. (2002)	Spain	Yield and chemical properties of soil
10	Delgado et al. (2012)	Spain	Physiochemical soil parameters
11	Dizer et al. (2002)	Germany	Leaching of EDCs
12	Domene et al. (2010)	Spain	Toxicity for collembolans
13	Escrig and Morell (1998)	Spain	Pollutant effect of sludges and toxicity
14	Frommme et al. (2002)	Germany	Occurrence of BPA in the environment
15	Georgieva et al. (2002)	UK	Ecotoxicity of metals
16	Gibbs et al. (2006a)	UK	Effects of individual metals
17	Gibbs et al. (2006b)	UK	Effects of individual metals
18	Gondek et al. (2010)	Poland	Metal transfer to plants
19	Gove et al. (2001)	UK	Mobility of metals and soil attenuation capacity
20	Hill et al. (1998)	UK	Accumulation of metals in animals
21	Hooda et al. (1997)	UK	Metal transfer to plants
22	Kalembasa and Pakula (2009)	Poland	Fractioning of metals
23	Kidd et al. (2007)	Spain	Bioavalability
24	Lombi and Gerzabek (1998)	Italy/Austria	Mobility of metals
25	Luckiewicz (2006)	Poland	Leachability
26	McGrath et al. (1999)	Germany	Bioavalability and ecotoxicity
27	Mihalache et al. (2014)	Romania	Metal transfer to plants
28	Moolenaar and Beltrami (1998)	Italy	Bioaccumulation and adsorption
29	Moreno et al. (1996)	Spain	Metal transfer to plants
30	Moreno et al. (1997)	Spain	Metal transfer to plants

31	Moreno et al. (1998)	Spain	Plant uptake and enzymatic activities
32	Moreno et al. (1999)	Spain	Microbial activity and mineralization
33	Morera et al. (2001)	Spain	Mobility of metals
34	Morera et al. (2002)	Spain	Bioavalability
35	Obbard and Jones (2000)	UK	Symbiotic nitrogen fixation
36	Obrador et al. (1998)	Spain	Mobility and extractability of metals
37	Oleszczuk et al. (2012)	Poland	Phytotoxicity
38	Pascual et al. (2004)	Spain	Bioavalability
39	Petruzzelli et al. (1997)	Italy	Sorption
40	Pisarek and Moliszewska (2004)	Poland	Soil properties
41	Planquart et al. (1999)	France	Metal transfer to plants
42	Proust et al. (2011)	France	Migration of metals in soil
43	Raffaella et al. (1997)	Italy	Accumulation of metals
44	Rastetter and Gerhardt (2015)	Germany	Ecotoxicity of metals and organic pollutants
45	Rhind et al. (2005)	UK	Accumulation of metals in animals
46	Rossi et al. (2002)	Italy	Accumulation of metals
47	Samaras and Kallianou (2000)	Greece	Yield and contamination of soil
48	Sánchez-Martin et al. (2007)	Spain	Fractioning of metals
49	Sieciechowicz et al. (2014)	Poland	Concentration of heavy metals and PAHs
50	Smith (1997)	UK	Soil properties
51	Soriano-Disla et al. (2008)	Spain	Bioavalability
52	Soriano-Disla et al. (2014)	Spain	Metal transfer to plants
53	Stöven and Schnug (2009)	Germany	Accumulation of metals
54	Tran et al. (2015)	France	Fate of BPA in soils
55	Walter and Cuevas (1999)	Spain	Fractioning of metals
56	Walter et al. (2002)	Spain	Extractibility of metals
57	Walter et al. (2006)	Spain	Phytotoxicity
58	Weightman (2006)	UK	Metal transfer to plants
59	Weissenhorn et al. (1995)	France	Bioavalability
60	Wilkinson et al. (2001)	UK	Accumulation of metals in animals
61	Zhang et al. 2005	UK	Fate of BPA in agricultural soils

Table 1 - Reviewed studies