

Investigation of the Potential Transfer and Uptake of Contaminants into Food Arising from the use of Biosolids and other Recycled Wastes in Agriculture

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Abstract

A research programme has commenced to determine the potential for transfer of organic contaminants to the human diet from biosolids and other recycled wastes applied to agricultural land as fertilisers and soil improvers. This includes an assessment of the potential transfer of selected organic contaminants to grazing dairy cattle, and milk, via ingestion of contaminated soil and foliage, and to agricultural crops via plant uptake.

In general, various organic contaminants were detected at low concentrations in two samples of biosolids from different urban areas. The concentrations of polychlorinated dioxins/furans, polychlorinated biphenyls and polycyclic aromatic hydrocarbons were between 5-50 times lower than limit values, proposed by the EC in 2000 for biosolids applied to agricultural land. A detailed discussion of the full suite of organic contaminants analysed in the range of wastes is presented in the paper. The research is expected to provide evidence that will improve confidence in the use of biosolids and waste-derived materials in agriculture and establish or update guidelines to protect the food chain where necessary.

Key words

agriculture; biosolids; land application; organic contaminants; persistent organic pollutants, sewage sludge

Introduction

Biosolids have been applied extensively to agricultural land for decades and the impacts on human health are well researched, but the significance of emerging contaminants needs to be considered. This paper presents the initial findings of a UK Food Standards Agency (FSA) funded research programme to investigate the potential transfer of organic contaminants into food arising from the use of biosolids and other recycled wastes in agriculture. The programme includes: i) crop transfer studies to investigate the potential uptake of organic contaminants from biosolids and other waste materials spread to agricultural land by plant tissue, including controlled growth chamber studies with barley and carrots and a field experiment with winter wheat; and ii) dairy cattle trials to investigate the potential transfer of organic contaminants to milk from wastes spread to agricultural land via ingestion of contaminated soil and foliage. A number of contaminant types have been selected that are recognised as the most abundant or important chemicals in their type class, taking into account the hazard assessment for emerging organic contaminants in biosolids conducted by Clarke and Smith (2011), which prioritised chemicals of concern for agricultural recycling. The programme is unique in the range of organic contaminants under

investigation and will provide vital information necessary to inform the development of a methodology and quality standards to improve confidence in the use of biosolids and other waste materials in agriculture.

Experimental Approach

Materials

The materials under investigation include biosolids (treated sewage sludge), meat and bone meal ash (MBMA), and poultry litter ash (PLA), representative of a range of recycled waste materials currently applied to agricultural land in the UK as sources of plant nutrients, and paper sludge ash (PSA), used as an agricultural liming agent. Additionally, compost like output (CLO) from the mechanical biological treatment (MBT) of municipal solid waste (MSW) is included as it has future potential as a source of plant nutrients in agriculture. A high degree of variability in the chemical composition of the materials was expected; hence, at least two examples within each waste category were collected where possible to increase the probability of finding a material containing the compounds of interest. The biosolids were collected from two of the UK's largest wastewater treatment plants. Both plants serve highly populated areas and accept combined sewage flows from domestic and industrial inputs, and historically have shown relatively high levels of contamination. Hence, these materials were selected as they represent potentially worst-case examples of biosolids chemical contamination. Both biosolids are produced from the mesophilic anaerobic digestion of the residual sludge from wastewater treatment and are dewatered and stockpiled for a period of several weeks before landspreading.

The waste materials have undergone routine physicochemical testing, and analysis for a selection of priority organic contaminants. The analysis of the materials enabled one example of each waste type to be selected for inclusion in the trials, which, in most cases, was the waste with the greatest concentrations of organic contaminants. However, a CLO containing relatively low concentrations of chemical contaminants was selected, for animal welfare reasons, because this CLO also had lower levels of physical (glass and plastic) contamination than the alternative CLO.

Crop trials

A protocol has been developed permitting reproducible testing of transfers of organic contaminants from waste-amended soil to vegetative barley shoots. A loamy sand is used in the bioassay, to minimise sorption to the soil complex and provide a reasonable worst-case of contaminant bioavailability. Barley is grown under controlled light and temperature conditions in a plant growth chamber for a period of 32 days, and the soil is maintained at a constant water content. A commercial slow release fertilizer incorporated into the soil supplies a balanced nutrient regime. To provide sufficient plant material sample for analysis, a separate plant growth trial is completed for: (1) unamended control soil, (2) biosolids amendment and (3) CLO amendment. Further controlled environment growth studies will focus on the transfer of contaminants from biosolids, CLO and ash (MBMA, PLA, PSA) – amended soil to carrot roots. Carrot roots have relatively high lipid content in their peel and represent a worst-case for assessing the potential significance of transfers of lipophilic and persistent organic contaminants to food crops and the human food chain via the plant uptake pathway. A field experiment with winter wheat was established during November 2014 on a loamy sand soil to investigate the transfer of organic contaminants from biosolids, CLO and ash (MBMA) to wheat grain. The crop will be harvested in September 2015, and the grain analysed to determine the extent of organic contaminant transfer from waste-amended soil.

Dairy cattle trials

The potential for organic contaminant transfer to milk, which is a highly sensitive dietary pathway for human food chain exposure, will be investigated in a series of controlled feeding ingestion studies with lactating cows. The ingestion of biosolids and other recycled waste products from soil or foliar contamination is simulated under controlled conditions. Soil ingestion levels of 5% of total feed dry matter (DM) intake are achieved by adding mixtures of soil and recycled wastes directly to a standard feed regime, based on a reasonable, worst-case upper mean soil ingestion rate for cattle (Thornton 1974). The wastes are first blended with the soil at rates equivalent to the maximum agronomic nutrient limits; the waste-soil mixtures are equilibrated for six weeks prior to the feeding experiment to simulate the minimum potential time period between sowing and grazing pasture. A similar rate of biosolids ingestion (5% of total feed DM) will be used for a treatment simulating the direct ingestion of biosolids without soil incorporation, representing a reasonable upper estimated sward contamination rate for pasture after surface application and a waiting period of 21 days (Smith, 1996), as required by the Code of Practice for Agricultural Use of Sewage Sludge (DoE, 1996). The control treatment will receive the same rate of soil addition in the feed to match the other experimental dietary treatments. There are four replicate animals per treatment, which are housed on straw bedding in pens. Each animal is electronically tagged to operate and access an individual gate controlled feeding trough allowing the specific feeding regime and intake of each animal to be monitored. The treatments will be fed for a period of three weeks, and feed intakes will be monitored and milk production measured. Milk samples are collected prior to feeding and on a weekly basis during the three week feeding period and during a four week withdrawal period following feeding. Organic contaminants will be measured in the milk collected prior to feeding, and at selected time points during the feeding period and at the end of the withdrawal period.

Results and Discussion

In general, the consignments of waste materials obtained for this investigation contained relatively low concentrations of many of the organic contaminant groups. Polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzodioxins/furans (PCDDs/Fs) and polychlorinated biphenyls (PCBs) are persistent organic pollutants that arise through incomplete combustion of fossil fuels and/or from environmental residues that enter urban wastewater through run-off (Smith, 2009). Notably, the concentrations of PAHs, PCDDs/Fs and PCBs present in biosolids, CLOs and ash were substantially lower than proposed and implemented limit values for these compounds across Europe for biosolids, composts and recycled ash materials where available. For example, the concentration, expressed as toxic equivalency (TEQ), of polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) in the biosolids samples was approximately 8-10 times less than a proposed EC (2000) limit for biosolids applied to agricultural land of 100 ng TEQ/kg DS. The concentration of polycyclic aromatic hydrocarbons (PAHs) in biosolids samples was also approximately 8-15 times less than a proposed limit of 6 mg/kg DS (EC, 2000), and the concentrations of polychlorinated biphenyls (PCBs) were approximately 17-42 times less than a proposed limit of 0.8 mg/kg DS (EC, 2000).

Polybrominated dibenzodioxins/furans (PBDD/Fs), polybrominated diphenyls (PBBs), mixed halogenated dibenzodioxins/furans (PXDD/Fs) and mixed halogenated diphenyls (PXBs) are related compounds to PCDDs/Fs and PCBs, but little is currently known about their presence in the environment and risk to human health. Polybrominated dibenzodioxins/furans were also detected in the biosolids and CLOs, but polybrominated biphenyls (PBBs), were generally only detected at negligible concentrations compared with PCBs. Only a relatively small sub-set of all the possible mixed polybrominated and chlorinated dibenzodioxins/furan (PXDD/F) and biphenyl (PXB) congeners can currently be analysed. However, these analyses indicated that PXBs were present at very low levels compared with PCBs in the organic waste materials. By contrast, the data for the PXDD/F congeners that were analysed indicated that PXDD/Fs are likely to be present in biosolids

and CLO at concentrations similar in magnitude to those of PCDD/Fs. Perfluoroalkyl compounds (PFCs), used, for example, in non-stick cookware and as a water and stain repellent treatment in textiles, were detected in the biosolids and CLO samples, and are of interest as they have a degree of water solubility, and therefore could potentially be taken up by crops. Brominated flame-retardants, such as polybrominated diphenyl ethers (PBDEs) were present in the biosolids and CLOs in low concentrations, but, as may be expected, were only detected at trace levels in some of the ash materials. A similar pattern was observed for polychlorinated naphthalenes (PCNs), which have been used in applications such as dielectric fluids, engine oil additives and lubricants, but can also be formed by combustion. The materials were also screened for a range of pharmaceutical compounds and these results, along with a detailed discussion of the full suite of organic contaminants analysed, are presented in the paper. The research programme will provide detailed information on the presence of organic contaminants in biosolids and other waste materials recycled in agriculture, and their transfer to milk or crops to improve the robustness of risk assessments. This is expected to enhance confidence in the use of these materials in agriculture, and help to establish improved or new guidelines to protect the food chain where necessary.

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