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Review

Human health risk assessment of pharmaceuticals and personal care products in plant tissue due to biosolids and manure amendments, and wastewater irrigation

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ABSTRACT

Amending soil with biosolids or livestock manure provides essential nutrients in agriculture. Irrigation with wastewater allows for agriculture in regions where water resources are limited. However, biosolids, manure and wastewater have all been shown to contain pharmaceuticals and personal care products (PPCPs). Studies have shown that PPCPs can accumulate in the tissues of plants but the risk that accumulated residues may pose to humans via consumption of edible portions is not well documented. This study reviewed the literature for studies that reported residues of PPCPs in the edible tissue of plants grown in biosolids- or manure-amended soils or irrigated with wastewater. These residues were used to determine the estimated daily intake of PPCPs for an adult and toddler. Estimated daily intake values were compared to acceptable daily intakes to determine whether PPCPs in plant tissue pose a hazard to human health. For all three amendment practices, the majority of reported residues resulted in hazard quotients <0.1. Amendment with biosolids or manure resulted in hazard quotients ≥0.1 for carbamazepine, diphenhydramine, salbutamol, triclosan, and sulfamethazine. Irrigation with wastewater resulted in hazard quotients of ≥0.1 for flunixin, ketoprofen, lamotrigine, metoprolol, and sildenafil. Many of the residues that resulted in hazard quotients ≥0.1 were due to exposing plants to concentrations of PPCPs that would not be considered relevant based on concentrations reported in biosolids and manure or unrealistic methods of exposure, which lead to artificially elevated plant residues. Our assessment indicates that the majority of individual PPCPs in the edible tissue of plants due to biosolids or manure amendment or wastewater irrigation represent a *de minimis* risk to human health. Assuming additivity, the mixture of PPCPs could potentially present a hazard. Further work needs to be done to assess the risk of the mixture of PPCPs that may be present in edible tissue of plants grown under these three amendment practices.

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1. Introduction

Soil moisture and nutrient content are critical to plant growth. Therefore, modern agriculture is continually searching for innovative methods to supply these two critical requirements for plant growth in order to maintain or increase crop yields while reducing the cost of production. The ability to supply moisture and nutrients also allows agriculture to expand to regions that would not typically be used for crop cultivation. Amending soil with livestock manure or biosolids and irrigating soil with wastewater are practices that contribute to increased crop yields on land currently used for agriculture and allow for expansion of agriculture to land considered to be unsuitable (Cogger et al., 2013; Ilias et al., 2014; Oron et al., 2014; Singh et al., 2014). However, biosolids, manure and wastewater have been found to contain a number of contaminants of concern, including a variety of pharmaceuticals and personal care products (PPCPs) (Aust et al., 2008; Calderon-Preciado et al., 2011a; CCME, 2010; USEPA, 2009a).

The source and type of PPCPs found in biosolids, manure, and wastewater vary among the three matrices. Many PPCPs are disposed down the household drain. Consequently, PPCPs are frequently detected in the influent water of wastewater treatment facilities (Golovko et al., 2014; Karnjanapiboonwong et al., 2011; Kumar et al., 2010). Persistent and relatively hydrophobic PPCPs tend to partition into the solid portion of sewage, and therefore are found in biosolids (CCME, 2010; Clarke and Smith, 2011; USEPA, 2009a). For example, Heidler and Halden (2009) detected triclosan and triclocarban in influent water from 25 municipal wastewater treatment facilities across 18 US states. In facilities employing primary treatment with anaerobic digestion of sewage sludge, ≥50% of triclosan and triclocarban entering the facility can partition into biosolids (Heidler and Halden, 2009). PPCPs that are soluble in water and not degraded during the wastewater treatment process can be present in effluent water at relatively high concentrations (Golovko et al., 2014; Li et al., 2013b; Nakada et al., 2006; Writer et al., 2013). For example, Gao et al. (2012) observed that 50, 60, 61, 73, and 141% of doxycycline, lincomycin, oxytetracycline, sulfadiazine, and carbamazepine that enter a wastewater treatment facility are present in effluent water. This effluent water could then potentially be used to irrigate crops. Manure tends to contain a different set of PPCPs relative to biosolids and wastewater. In many jurisdictions, livestock are fed veterinary antibiotics at sub-therapeutic levels in order to promote growth. Therefore, the dominant group of PPCPs detected in livestock manure is veterinary antibiotics (Aust et al., 2008; Awad et al., 2014; Ben et al., 2013; Sura et al., 2014; Wang et al., 2014).

A number of studies have shown that crop plants can accumulate PPCPs present in biosolids, manure, and wastewater. Boxall et al. (2006) observed that the veterinary pharmaceuticals, florfenicol, levamisole, and trimethoprim in spiked soil can accumulate in lettuce plants, and enrofloxacin, florfenicol, and trimethoprim in carrot plants. Wu et al. (2013) investigated whether 20 PPCPs could accumulate in four plants species (i.e., lettuce, spinach, cucumber, and pepper) grown in a nutrient solution spiked with PPCPs. All of the PPCPs were detected in the roots of the plants and 13 of the 20 PPCPs were detected in the leaves of the plants. Collectively, these studies clearly demonstrate that plants have the capacity to accumulate PPCPs from the media in which they are growing.

PCPPs are present in biosolids, manure, and wastewater and if they are going to be used in agriculture, crop plants are going to be exposed to PPCPs. The ability of plants to accumulate PCPPs produces a potential pathway of exposure to humans. If plants grown in soil amended with biosolids or manure, or irrigated with wastewater contain PPCPs in the edible tissue, will these residues of PPCPs pose a risk to human health? The objective of this study was to perform a risk assessment to determine whether PPCPs in the edible tissue of plants grown in soil amended with biosolids or manure or irrigated with wastewater pose a risk to human health. The goal of this risk assessment is to provide insight on whether the risk due to exposure to PCPPs as a result of biosolids or manure amendment or irrigation with wastewater needs to be regulated.

2. Risk assessment approach

2.1. Collection of data on plant residues

The peer-reviewed literature was searched for studies that investigated the accumulation of pharmaceuticals and/or personal care products into plants from soil that was amended with either biosolids, manure, or irrigated with wastewater. For studies involving soil amendment with biosolids or manure, PPCPs analyzed in plant tissue, type of biosolids or manure, rate of amendment, concentration of PPCP in biosolids or manure, concentration of PPCP in soil at the initiation and conclusion of the test, plant species, type of tissue, and concentration of PPCP in plant tissue were tabulated (Supporting information Tables S1 & S2). For studies involving soil irrigated with wastewater, PCPPs analyzed in plant tissue, rate of irrigation, frequency of irrigation, duration of irrigation, concentration of PCPP in wastewater, concentration of PCPP in soil, plant species, type of tissue, and concentration of PCPP in plant tissue were tabulated (Table S3). Soil pH, which is of particular importance for ionogenic PPCPs, organic matter or carbon content, and irrigation water pH were also included. These parameters are critical to quantifying exposure to the plant and potential exposure to humans. Clearly defining the frequency, magnitude, and mode of exposure to the plant is also essential to determine whether the experimental design is relevant in an agricultural context. If the experimental design is not environmentally and agronomically relevant (e.g., unrealistic exposure concentrations, unrealistic amendment scenarios such as hydroponic studies), then the reported residues in the edible portion of plant should be considered carefully in the context of its use in a risk assessment. Therefore, in the present assessment, we excluded any studies that exposed plants to PPCPs through hydroponic systems or through spiking of soil. While these types of studies can be useful to characterize the ability of PPCPs to accumulate in plant tissue, they do not represent realistic exposure scenarios likely to occur under typical agricultural amendment practices.

2.2. Calculation of risk to human health

Acceptable daily intake (ADI) values were calculated for each PPCP reported in plant tissue. The ADI value is the amount of PPCP that can be consumed daily over a person's lifespan without evocating an adverse effect. The ADI values for pharmaceuticals used for treatment of humans were determined by dividing the lowest daily therapeutic dose for an adult (mg/day) by a safety factor of 1000 and a body weight of 70 kg (DEFRA, 2007; National Health and Medical Research Council of Australia, 2008; WHO, 2011) (Table 1). The safety factor of 1000 is composed of three factors of 10 applied to address differences in response between humans, potential sensitivity of subgroups of the population (i.e., children and infants), and the lowest daily therapeutic dose not being a level that represents no effect (DEFRA, 2007; National Health and Medical Research Council of Australia, 2008; WHO, 2011). An additional safety factor of 10 was added to compounds that exhibit activity with the endocrine system (e.g., progesterone, testosterone) (DEFRA, 2007; National Health and Medical Research Council of Australia, 2008). ADI values for triclocarban and triclosan were determined by applying a safety factor of 300 to a no observable adverse effect level (NOAEL) value of 25 mg/kg body weight (bw)/day (European Commission Health and Consumer Protection Directorate-General, 2005; Health Canada and Environment Canada, 2012) (Table 1). The NOAEL value for triclosan and triclocarban originated from a 90-day oral toxicity study with mice and a 2-year oral toxicity study with rats, respectively (European Commission Health and