Ecotoxicology and Environmental Safety 158 (2018) 28–36

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Migration and health risks of nonylphenol and bisphenol a in soil-winter wheat systems with long-term reclaimed water irrigation

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ARTICLE INFO

Keywords:
Reclaimed water irrigation
Nonylphenol (NP)
Bisphenol A (BPA)
Migration
Soil-winter wheat systems
Health risk

ABSTRACT

Reclaimed water reuse has become an important means of alleviating agricultural water shortage worldwide. However, the presence of endocrine disrupters has roused up considerable attention. Barrel test in farmland was conducted to investigate the migration of nonylphenol (NP) and bisphenol A (BPA) in soil-winter wheat system simulating reclaimed water irrigation. Additionally, the health risks on humans were assessed based on US EPA risk assessment model. The migration of NP and BPA decreased from the soil to the winter wheat; the biological concentration factors (BCFs) of NP and BPA in roots, stems, leaves, and grains all decreased with their added concentrations in soils. The BCFs of NP and BPA in roots were greatest (0.60–5.80 and 0.063–1.45, respectively). The average BCFs of NP and BPA in winter wheat showed negative exponential relations to their concentrations in soil. The amounts of NP and BPA in soil-winter wheat system accounted for 8.99–28.24% and 2.35–4.95%, respectively, of the initial amounts added into the soils. The hazard quotient (HQ) for children and adults ranged between 10^{-6} and 1, so carcinogenic risks could be induced by ingesting winter wheat grains under long-term reclaimed water irrigation.

1. Introduction

Global water resource shortages are becoming increasingly serious, and reclaimed water irrigation has emerged as an important means of alleviating agricultural water shortages (FAO, 2012). However, in reclaimed water, there exist all kinds of pollutants, such as heavy metals, microorganisms and organic pollutants, which can induce risks to human health through inhalation, dermal contact, and ingestion (EC, 2001; Rajaganapathy et al., 2011; Liu et al., 2017). Endocrine disrupters, especially, are raising public concern. Nonylphenol (NP) and bisphenol A (BPA) are two endocrine disrupters that are frequently detected in reclaimed water (Petrovic et al., 2002; Lu and Gan, 2014a, 2014b; Wang et al., 2015). These two substances can severely affect living organisms' endocrine systems, causing strong carcinogenic, teratogenic, and mutagenic effects (Woo1 et al., 2016; Tabassum et al., 2017), which have drawn intense scrutiny.

NP is composed of multiple isomers with a low water solubility of 5.42 mg L^{-1} and an octanol-water partition coefficient of 4.48 (Lu and Gan, 2014a, 2014b). It can be very easily adsorbed by organic matter such as biodetritus, soil minerals, and colloidal substances. NP mainly comes from the degradation of nonylphenol ethoxylates (NPnEO), which are typically used in consumer and commercial cleaning products such as surfactants. Then they are discharged in wastewater influent, treated in the plants, which results in degradation of some fraction of the NPEOs to NP (Petrovic et al., 2002; Ömeroğlua et al., 2015). The concentration of NP in reclaimed water generally ranges from 0.05 to 8 μg L^{-1} (Campbell et al., 2006; Hao et al., 2006; Wang et al., 2015; Diao et al., 2017), and the concentration of NP in groundwater is no higher than 3.85 μg L^{-1} (Loos et al., 2010; Félix-Cañedo et al., 2013; Luo et al., 2014; Wang et al., 2015). Landfill leachates, sewage irrigation, and septic-tank leachates are the primary sources of NP in groundwater (Luo et al., 2014). The concentration of NP in sewage spray fields and fertilization farmlands ranges from 0.01 to 27,882 μg kg^{-1} (Vikelsøe et al., 2002; Dong et al., 2015; Diao et al., 2017; Liu et al., 2017).

BPA has a water solubility of 120–300 μg L^{-1}, and it has been commonly used industrially as a plasticizer that subsequently can migrate into consumer products and liquids that are in contact with those plastic products. It frequently ends up in wastewater influent as a result (J. Lu et al., 2015; Z. Lu et al., 2015; Liu et al., 2016). BPA is frequently

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https://doi.org/10.1016/j.ecoenv.2018.03.082
Received 31 July 2017; Received in revised form 28 March 2018; Accepted 28 March 2018
0147-6513/

detected in farmlands irrigated with reclaimed water, and soils or groundwater near refuse landfills (Félix-Cañedo et al., 2013; Michałowicz, 2014; Diao et al., 2017). The concentrations of BPA in soils, groundwater, and reclaimed water are 0.55–147 μg kg^{-1}, 0.001–20 μg L^{-1}, and 0.15–2 μg L^{-1}, respectively (Campbell et al., 2006; Kinney et al., 2008; Gibson et al., 2010; Luo et al., 2014; Sidhu et al., 2015; Diao et al., 2017).

Currently, relevant studies mainly focus on NP and BPA in water, sediments, and soils, and have been less concerned with the migration of NP and BPA in soil-plant systems (Ferguson et al., 2001; Petrovic et al., 2002; Ömeroğlua et al., 2015; Liu et al., 2016). Investigation on soybeans, wheat, tomatoes, rice, and sorghum revealed that NP and BPA have been detected in plant tissues (Mortensen and Kure, 2003; Imadeddin et al., 2004; J. Lu et al., 2015; Z. Lu et al., 2015). The distributions of NP and BPA in plants are not only related to their physicochemical properties and the types of plants, but also affected by many other factors such as the chemical forms of NP and BPA, soil pH, soil organic matter content, microorganisms around the root systems, redox potential, and the activity of enzymes in plants (Montgomery-Brown et al., 2003; Dodgen et al., 2013). Under aerobic conditions, NP and BPA can be degraded in soils. Microorganisms are a key factor affecting the degradation of NP and BPA (Ike et al., 2002; Zhang et al., 2009; Rozalska et al., 2010a, 2010b), and also NP and BPA can undergo biological metabolism in plants (Bokern et al., 1996; Schmidt and Schuphan, 2002).

Wheat is a globally cultivated crop; however, few studies have been conducted on the migration of NP and BPA in soil-wheat system under long-term reclaimed water irrigation. This study aims to evaluate (1) the distribution of NP and BPA in different tissues of winter wheat with different simulated irrigation time; (2) the migration and accumulation of NP and BPA from soil to winter wheat; (3) the health risks induced by ingesting winter wheat grains with accumulated NP and BPA with different irrigation times.

2. Materials and methods

2.1. Research area

The research area is located in Pang Gezhuang Experimental Station of the National Center for Efficient Irrigation Engineering and Technology Research-Beijing,China (39°36'N, 116°21'E). The study area is in a warm temperate zone with a semi-humid continental monsoon climate. It is hot and rainy in summer and cold and dry in winter. The annual mean temperature is 14.0 °C; specifically, the temperature in summer ranges from 28 °C to 32 °C and in winter it ranges from − 5°C to 10 °C. The precipitation shows great inter-annual variations, and the rainy season is from June to September, with an annual average precipitation of 554.5 mm. The research area was located in the lower reaches of an alluvial-proluvial fan, and the soils at the depth of 0–40 cm were composed of 27.5% sand, 67.5% silt, and 5% clay. The background soil concentration of NP and BPA were 37 μg kg^{-1} and below the method detection limit (MDL) respectively. The background concentrations of NP and BPA in groundwater were 16.64 ng L^{-1} and 38.12 ng L^{-1}, respectively.

2.2. Materials

NP isomers standard, purity of 100%, 0.25 g, CAS:25154-52-3; BPA standard, purity of 99.8%, 0.25 g, CAS:80-05-7, both purchased from Dr. Ehrenstorfer GmbH; Diethyl phthalate-3,4,5,6-d4 (PAE-d4) served as surrogate, purity of 99.3%, CAS:93962-12-6, purchased from Sigma-Aldrich; Hexane, CAS:110-54-3,density:0.6594 g mL^{-1}; Methanol, CAS:67-56-1, density:0.7913 g mL^{-1}; Acetone, CAS: 67-64-1, density:0.7857 g mL^{-1}, all solvents were HPLC grade, purchased from Honeywell.

2.3. Experimental design

The highest concentrations of NP and BPA in reclaimed water are 8 μg $L^{-1}$and 2 μg L^{-1}, respectively (Hao et al., 2006; Sidhu et al., 2015; Diao et al., 2017), and the irrigation quota for crops are 0.6 m^3/m^2 annually. We calculated annual input loadings of NP and BPA irrigated to soils with the aforementioned highest concentrations in reclaimed water and irrigation quota. Five treatments, all measured in dry weight, of 0.467 mg kg^{-1} (NP_{T1}), 0.897 mg kg^{-1} (NP_{T2}), 1.327 mg kg^{-1} (NP_{T3}), 1.757 mg kg^{-1} (NP_{T4}), 2.617 mg kg^{-1} (NP_{T5}) were set up to simulate different NP initial concentrations in soils respectively according to NP input loadings under the five scenarios where soils were irrigated for 50, 100, 150, 200, and 300 years, ignoring attenuation. Five treatments of 0.107 mg kg^{-1} (BPA_{T1}), 0.215 mg kg^{-1} (BPA_{T2}), 0.323 mg kg^{-1} (BPA_{T3}), 0.430 mg kg^{-1}(BPA_{T4}), and 0.645 mg kg^{-1} (BPA_{T5}) were set up to simulate soil BPA initial concentrations respectively to BPA input loadings with the aforementioned five irrigation times. The stainless steel barrels used in the experiments were 45 cm in height, in which the soil was 40 cm in depth. The inner diameter was 40 cm and the effective volume was 50 L. There was a hole at the bottom of the barrel, and a plate 20 cm in diameter and 4 cm in depth was fixed below to collect the leaking water during irrigation. There were larger barrels outside to protect the plate from the soil around. NP and BPA were first dissolved in methyl alcohol (volume < 5%), and methyl alcohol was dissolved in ultrapure water. The bulk density of the field soil was 1.4 g/cm^3. The soils were first dug out and 5 mm-sieved to ensure uniformity, layered thinly and then NP and BPA solutions were sprayed on them respectively according to the five treatments. After application, they were mixed together completely with the soils. Meanwhile, a blank control denoted as CK (the control check) with no NP or BPA was set up for comparison. Each treatment was performed in triplicate. Finally, the sieved soil was backfilled into the barrels, 10 cm per layer at one time, using a hairbrush to roughen the surface. And the soils were 70 kg in each barrel. Then the barrels were put back into the holes where the soil was removed and the soil in the barrels were leveled the same with the ground to ensure the same growing conditions for winter wheats. Amounts of 2 g compound fertilizers (produced by Shandong Aolindan Feritilizer Co., Ltd. (N(26%)-P (6%)-K (8%)) were applied in each barrel and then 5 g of wheat seeds (Taishan No. 5) were sown. The soil was irrigated with groundwater for the first time. Then, according to the meteorology, the second, third and the fourth time irrigations were applied during the tillering, green and jointing periods, respectively. Each barrel was irrigated with 8 L groundwater each time. The growth period of winter wheat started on October 13, 2014 and ended on June 13, 2015 (240 days). During this time, it rained 3 times and snowed twice.

2.4. Sample collection and treatment

After the winter wheat harvest, soils were collected at different depths from the top, 10 cm, 25 cm, bottom of the barrels. The soils were freeze-dried at − 20 °C and then 0.9 mm-sieved. The roots, stems, leaves and grains were also separately collected. 264 samples were collected in total. The roots were cleaned in ultrapure water, sonicating 3 times, each time for half an hour, then naturally air dried and, at last, weighed. After the wheat hulls were removed, the wheat seeds were weighed. The stems and leaves were also separated and weighed for pre-processing.

After air drying, all the roots, stems, leaves, and grains of the winter wheat were weighed and grounded using a pulverizer (Tianjin Taisete Instrument Co., Ltd., China) and then 0.9 mm-sieved. Next, 2 g of roots, 2 g of stems, 2 g of leaves, and 5 g of grains (dry weight) were added to 8 μL PAE-d4 standard solution (with a concentration of 25 μg L^{-1}). After soxhlet extracted with 220 mL n-hexane, the extracted solutions were dried with anhydrous sodium sulfate, concentrated using rotary evaporation (RE-52AA, Shanghai Yarong Biochemical Instrument Plant,