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Probabilistic determination of the ecological risk from OTNE in aquatic and terrestrial compartments based on US-wide monitoring data

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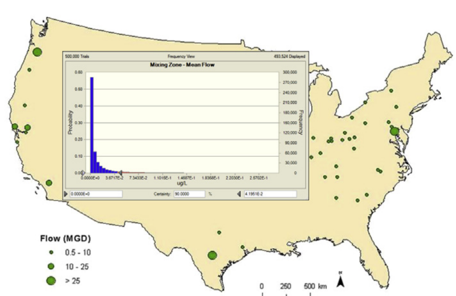
HIGHLIGHTS

- US-wide OTNE wastewater treatment plant effluent and sludge monitoring data.
- US-wide OTNE probabilistic risk assessment in WWTP mixing zones.
- US-wide OTNE probabilistic risk assessment in sludge amended soils.
- OTNE poses negligible risk to aquatic, sediment, and terrestrial dwelling organisms.

GRAPHICAL ABSTRACT



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ABSTRACT

OTNE [1-(1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethyl-2-naphthyl)ethan-1-one; trade name Iso E Super] is a fragrance ingredient commonly used in consumer products which are disposed down the drain. This research measured effluent and sludge concentrations of OTNE at 44 US wastewater treatment plants (WWTP). The mean effluent and sludge concentrations were 0.69 ± 0.65 μg/L and 20.6 ± 33.8 mg/kg dw respectively. Distribution of OTNE effluent concentrations and dilution factors were used to predict surface water and sediment concentrations and distributions of OTNE sludge concentrations and loading rates were used to predict terrestrial concentrations. The 90th percentile concentration of OTNE in US WWTP mixing zones was predicted to be 0.04 and 0.85 μg/L under mean and 7Q10 low flow (lowest river flow occurring over a 7 day period every 10 years) conditions respectively. The 90th percentile sediment concentrations under mean and 7Q10 low flow conditions were predicted to be 0.081 and 1.6 mg/kg dw respectively. Based on current US sludge application practices, the 90th percentile OTNE terrestrial concentration was 1.38 mg/kg dw. The probability of OTNE concentrations being below the predicted no effect concentration (PNEC) for the aquatic and sediment compartments was greater than 99%. For the terrestrial compartment, the probability of OTNE concentrations being lower than the PNEC was 97% for current US sludge application practices. Based on the results of this study, OTNE concentrations in US WWTP effluent and sludge do not pose an ecological risk to aquatic, sediment and terrestrial organisms.

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1. Introduction

OTNE [1-(1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethyl-2-naphthyl)ethan-1-one; trade name Iso E Super] is a widely used fragrance ingredient in consumer products such as cosmetics, soaps, household cleaners and detergents. These consumer products are most commonly disposed down the drain, thus the major route of OTNE into the environment is through wastewater treatment plant (WWTP) effluent and sewage sludge. OTNE has been shown to be highly removed during wastewater treatment. Simonich et al. (2002) reported >91% removal for US activated sludge WWTPs and Bester et al. (2008) reported removals ranging from 56 to 84% for activated sludge treatment plants in Germany. Despite its widespread use, limited environmental monitoring data are available for OTNE in US WWTP effluents and sludges. Simonich et al. (2000) measured OTNE concentrations in 3 day composite effluent samples from 2 US WWTPs and reported concentrations of 0.110 μg/L for an activated sludge plant and 0.334 μg/L for a trickling filter plant. In a continuation of the first study, Simonich et al. (2002) measured effluent concentrations of OTNE from 12 US WWTPs and reported concentrations ranging from 0.028 to 0.672 μg/L. Di Francesco et al. (2004) reported OTNE levels in anaerobically digested and dewatered sludge from 2 activated sludge WWTPs in Delaware of 7.3 and 30.7 mg/kg dw. Continued wide dispersive use of OTNE in consumer products justifies the need to generate high tier exposure data from monitoring WWTP effluent and sludge in order to confirm environmental safety in the aquatic, sediment and terrestrial compartments.

Recent research has presented new approaches for conducting US-wide aquatic, sediment, and terrestrial risk assessments for consumer product chemicals disposed of down the drain (Simonich et al., 2013; Federle et al., 2014; Kapo et al., 2015). This current study builds on the methodology presented by Federle et al. (2014) in their HHCB probabilistic environmental risk assessment in US mixing zones, sediments and sludge amended soils. Specifically, HHCB effluent and sludge concentrations from 40 US WWTPs were used as statistical distributions to evaluate loadings to the aquatic and terrestrial environments. The distributions of effluent concentrations were combined with distributions of US effluent-in-river water dilution factors (DF) immediately below wastewater treatment facilities under mean and 7Q10 low flow (lowest river flow occurring over a 7 day period every 10 years) conditions to forecast the distribution of concentrations in US mixing zones. The sludge distributions were combined with sludge loading rates to soil in order to predict concentrations in sludge amended soils. The forecasted HHCB concentrations were then compared to predicted no effects concentrations (PNEC) in order to evaluate environmental safety in the aquatic, sediment, and terrestrial compartments.

A suite of high tier toxicity studies exist for OTNE in the aquatic, sediment, and soil compartments. These studies were all conducted using GLP techniques with analytical confirmation of dose and exposure. In the aquatic compartment OTNE chronic toxicity to algae (*Scenedesmus subspicatus*), *Daphnia magna*, and zebra fish (*Danio rerio*) was evaluated (IFF, 1998; RIFM, 2002; RIFM, 2001). For the sediment compartment, OTNE chronic toxicity data is available for *Chironomous riparius*, *Hyalella azteca*, and *Lumbriculus variegatus* (IFF, 2005a,b,c). And for the terrestrial compartment, OTNE toxicity to microorganisms (OECD 216 Soil Microorganisms Nitrogen Transformation Test), earthworms (*Eisenia fetida*) and 6 different plant species was previously evaluated (IFF, 2016a,b,c). These studies were leveraged in combination with the exposure data generated in this research in order to evaluate ecological risk in the aquatic, sediment and terrestrial compartments.

The objective of this research was to measure OTNE effluent and sludge concentrations from 44 US WWTPs. The distribution of effluent concentrations was combined with mixing zone dilution factors and modeled to predict aquatic and sediment OTNE concentrations. The distribution of sludge concentrations were combined with information on sludge loading rates and modeled to predict OTNE levels in US sludge amended soils. The predicted environmental concentrations (PECs) were evaluated against OTNE PNECs in order to quantify ecological risk in each compartment.

2. Materials and methods

2.1. Chemicals

OTNE (CAS # 54464-57-2, EC # 915-730-3) was provided by International Flavors and Fragrances, IFF (lot # RG05074698) and had a purity of 95%. An internal standard (IS) d_3-OTNE was synthesized at Procter and Gamble (from the IFF starting material) and was confirmed to be >99% labeled material via LC/MS/MS and to have the same component composition (GC/MS). ACS grade formalin was used in sample preservation. HPLC grade methanol (MeOH) and hexane and Ultrapure water (UPW, Milli-Q Unit, Millipore, USA) were used for sample preparation and analysis.

2.2. Field sampling

Simonich et al. (2000) studied influent and effluent concentrations of fragrance materials over a 24 h period and found that while influent levels are highly variable, effluent levels are diurnally stable, therefore this research focused on grab effluent concentrations in order to evaluate OTNE levels entering the aquatic environment from WWTP discharges. Kapo et al. (2015) reported that 86% of US wastewater (by volume) is treated by activated sludge, 12% by trickling filter or lagoon processes and 2% by rotating biological contactor, therefore for this research activated sludge wastewater treatment plants were sampled as it is by far the predominant form of treatment in the US. From July through December 2013, effluent and sludge samples were collected from 44 US municipal WWTPs. (Fig. 1). The WWTPs sampled receive greater than 90% domestic wastewater which allows evaluation of maximum loading from consumer use. This sampling strategy has previously been used by Simonich et al. (2013) for 1,4 dioxane and Sun et al. (2014) for polycyclic musks.

Duplicate 100 mL grab effluent samples were collected at the discharge point of each treatment plant using amber glass I-Chem containers and spiked with 0.05 μg of d_3-OTNE IS. The effluent samples were preserved with 3 mL of formalin (Simonich et al., 2000), placed on ice and transported to the lab for analysis. Effluent samples were stored in the lab at 4 °C prior to analysis (analysis completed within 3 weeks of sampling). An effluent sample was also collected for analysis of ammonia and total suspended solids (TSS) concentrations which can provide information regarding the efficacy of the treatment processes while sampling. Specifically the physical treatment processes (evaluated via TSS) can impact sorption and the biological treatment efficacy (evaluated via ammonia levels in the effluent) can impact biotransformation. Both of these processes contribute to removal of OTNE during wastewater treatment (Bester et al., 2008; Procter and Gamble, 2000; RIFM, 2009; Simonich et al., 2000 & 2002). Duplicate 200 g sludge samples were collected in 24 oz. WHIRL-PAK® bags, frozen on dry ice for shipment, and stored at −80 °C until analysis (within 3 weeks of sampling). Information on the processing of sludge at each WWTP prior to collection is contained in Table S3.