



Leaching of polycyclic aromatic hydrocarbons (PAHs) from industrial wastewater sludge by ultrasonic treatment



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ABSTRACT

Ultrasonic treatment for sludge reduction in wastewater treatment plants (WWTPs) can substantially affect the fate of trace pollutants. However, their fates in the different phases of sludge and mass balances have rarely been reported. In this study, wastewater sludge samples were ultrasonicated at 600 W for 0–30 min. Then, the leaching of the 16 priority polycyclic aromatic hydrocarbons (PAHs) from the sludge solids (sediment) to the liquid phase (supernatant) was investigated. The total concentration of PAHs (\sum_{16} PAHs) in the sludge sediment (2.10 $\mu\text{g/g}$) was comparable with those of previous worldwide studies. Among the 16 PAHs, naphthalene and acenaphthylene were dominant. The total concentrations of PAHs in the supernatant generally increased with sonication time, indicating that PAHs associated with sludge materials, such as microorganisms, were released into the supernatant. Lighter and more water soluble PAHs were released preferentially into the supernatant in dissolved form, whereas heavier and more hydrophobic PAHs were strongly bound to particles. According to mass balance calculations, 21% of the PAHs in the sludge sediment moved to the supernatant without discernible sonodegradation. An additional experiment for degradation of PAHs supported this interpretation, and several reasons for the no significant sonodegradation were discussed. This result suggests that leaching trace pollutants may significantly contaminate the sludge filtrate after ultrasonic treatment, and therefore their fates should be investigated.

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1. Introduction

Sludge is generally produced from wastewater treatment processes, and its amount has increased with the rapid growth of industrialization and urbanization [1]. Sludge contains high fractions of water and solids containing microorganisms and their extracellular polymeric substances (EPS) such as proteins, humic compounds, and carbohydrates [2]. It also contains trace-level toxic organic chemicals such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) [3,4]. These trace pollutants are an important issue for both agricultural recycling of sludge [5,6] and incineration [7].

As the London Convention has prohibited ocean dumping of sludge, efficient reduction of sludge has received much attention. Various technologies for sludge reduction have been investigated: thermal treatment [8,9], biological treatment [10], acid and surfactant treatment [11], alkali treatment [12], ultrasonic treatment [8],

high-pressure homogenization [13], ozone treatment [8], electrochemical treatment [14], electro-flotation [15], and microwave treatment [16]. Among them, ultrasonic treatment has been used widely because of its relatively low processing cost and high efficiency of reduction [17–20].

Sludge reduction after ultrasonic treatment is based on ultrasound cavitation phenomenon that destroys microorganism flocs and microbial cell walls [1,20,21]. Ultrasonic pretreatment can enhance not only direct reduction of sludge, but also digestion of biosolids in sludge [22]. Furthermore, dewaterability of sludge can be improved [17], resulting in cost reduction for the final disposal of sludge through landfilling, incineration, and ocean dumping. Previously, studies have been carried out to find optimum processing conditions for high removal efficiencies of sludge. For example, both ultrasonic irradiation time and intensity significantly improve removal efficiencies of sludge [21,23,24].

Studies have shown that PAHs in water [25,26] and wastewater [27] are degraded with ultrasonic treatment with stronger irradiation intensity and longer irradiation time. However, these studies did not investigate the degradation of PAHs in sludge. Furthermore, individual concentrations and amounts of PAHs in the distinct

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phases of sludge (i.e., supernatant and sediment) rarely have been reported in previous studies [3,4,28]. For this reason, the mass balance of PAHs and their fate in sludge before and after ultrasonic treatment are not clearly understood. For instance, to the best of our knowledge, there is no information on whether PAHs in sludge are physically degraded or simply moved from the solid phase (sediment) to the liquid phase (supernatant).

The purpose of this study is to understand the overall fate of PAHs in sludge after ultrasonic treatment. We collected industrial wastewater sludge and ultrasonicated it under different conditions. Then, the concentrations and amounts of PAHs in the supernatant and sediment were analyzed separately, and the characteristics of sludge contamination with PAHs were investigated. Mass balance calculations and degradation experiments were also conducted. To our understanding, this is the first study to investigate the mass balance of PAHs in the different phases (the sediment and the dissolved and particulate phases of the supernatant) of wastewater sludge before and after ultrasonic treatment.

2. Materials and methods

2.1. Sludge sampling

Sludge samples were collected from the wastewater treatment plant (WWTP) of a petrochemical company located in Ulsan, Korea. This WWTP treats wastewater (150 ton/d) generated from chemical processes that produce terephthalate (50%), polyester chip (35%), and others (15%). In the first stage of the WWTP, a pressure flotation process eliminates suspended solids. After aeration and flocculation, solid and liquid of wastewater are separated in a settling tank. Then, sludge is generated by mixing the suspended solids in the wastewater with sediment in a pressure flotation tank and a settling tank. The collected sludge was transferred (approximately 30 min in transit) to a laboratory and stored in opaque glass bottles at 4 °C until the experiments were performed.

2.2. Sludge sample ultrasonic treatment and preparation for chemical analyses

Individual sludge samples (750 mL) were placed in 1000 mL beakers and treated with a horn-type ultrasonicator (Sonosmasher, ULH-700S, 20 kHz, Sibata, Japan) at 600 W for 0, 5, 10, 15, 20, 25, and 30 min. An ultrasonic intensity of 600 W was selected as the default option based on our prior study [29], which reported sludge reduction over a range of irradiation intensities (100–600 W). The sonicated sludge samples were separated into supernatant and sediment by a centrifuge (Combi-514R, HANIL Science Industrial, Korea) at 3000 rpm for 30 min.

Before centrifugation, total suspended solids (TSS) and volatile suspended solids (VSS) were measured as an index for sludge reduction resulting in a release of soluble organic carbon and extracellular enzymes [30]. Suspended solids were filtered through a glass fiber filter (GFF) (47 mm diameter, 0.7 µm pore size, Whatman, England), dried for 2 h at 110 °C, and then the VSS were determined after ignition in an electric oven for 15 min at 400 °C.

2.3. Instrumental analysis of PAHs

The PAHs in the three phases of sludge (the sediment and the dissolved and particulate phases of the supernatant) were analyzed separately. The supernatant samples were diluted five times with distilled water, and an aliquot of 100 mL of the final dilution was used for extraction. The supernatant was separated into two phases by a glass microfiber filter (GFF: 47 mm diameter, 0.7 µm

pore size, Whatman, England). Note that there were suspended solids in the supernatant even after centrifugation. Before extraction, surrogate standards (100 ng each of naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂) were added to all the samples.

The PAHs in the dissolved phase of the supernatant (i.e., filtrate) were liquid–liquid extracted for 10 min with 100 mL of a mixed solvent (dichloromethane:methanol = 9:1, v/v). This extraction procedure was repeated three times. The extract was concentrated to 5 mL by a Turbo Vap II (Biotage, Sweden) and cleaned up using a column consisting of 10 g of anhydrous sodium sulfate (Na₂SO₄) to remove moisture from the samples. Finally, the samples were concentrated to 1 mL by a nitrogen evaporator (MGS-2200, Eyela, Japan).

The PAHs in the particulate phase of the supernatant (i.e., GFF) were extracted using 20 mL of n-hexane/acetone (9:1, v/v) mixture in an ultrasonic bath operated for 15 min. This was repeated three times. The extract was concentrated to 5 mL by the Turbo Vap II and purified using a column consisting of 5 g each of silica gel and Na₂SO₄ with 60 mL of dichloromethane/hexane (1:1, v/v). Finally, the purified samples were concentrated to 1 mL by the nitrogen evaporator.

For the analysis of the PAHs in the sludge sediment, 0.5 g of dried samples were mixed with 2 g of Na₂SO₄ and then Soxhlet extracted for 20 h with 350 mL of n-hexane/acetone (9:1, v/v). The extract was concentrated to 5 mL by the Turbo Vap II and cleaned up using a silica gel column with 2 g of Na₂SO₄, 2 g of alumina, 5 g of activated silica gel, and 2 g of Na₂SO₄. The samples were eluted with 120 mL of n-hexane/dichloromethane (3:1, v/v). Finally, the purified samples were concentrated and transferred to gas chromatography (GC) vials.

Before analysis, an internal standard (*p*-terphenyl-d₁₄, 100 ng) was injected into the GC vials. The final samples were analyzed for the 16 priority PAHs: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (Ind), dibenzo(a,h)anthracene (DahA), and benzo(g,h,i)perylene (BghiP). A gas chromatograph (Agilent 7890A, USA) equipped with a mass spectrometer (Agilent 5975C, USA) was used under the following conditions: injection temperature of 280 °C; splitless injection mode; helium carrier gas at 1.0 mL/min; DB-5MS column (30 m × 0.25 mm i.d., 0.25 µm film thickness); oven temperature of 50 °C for 1 min, then increased by 10 °C/min to 300 °C, which was held for 5 min; ionization voltage of 70 eV; and EI/SIM mode.

Analytical data were corrected by method blanks analyzed by the same method as was used for the samples. The recoveries of surrogate standards ranged from 61% to 115%. Method detection limits (MDL) were calculated by multiplying the standard deviations of seven replicates of the MDL standard analyzed with the same procedure as the samples and the Student's *t* value (3.14) for a 99% confidence level. Data below detection limits were represented as non-detects (ND).

3. Results and discussion

3.1. Physical change of sludge after ultrasonic treatment

Ultrasonication induces sludge disintegration by deagglomerating biological flocs and breaking down large particles into smaller particles [1]. In addition, bacterial cell walls are broken down by the high-pressure-induced shear force, thus releasing intracellular materials into the liquid phase [1]. Therefore, the amount of sludge

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