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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 μg/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.

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-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrisene-d_{12}, and perylene-d_{12}) 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 (Na_2SO_4) 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 Na_2SO_4 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 Na_2SO_4 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 Na_2SO_4, 2 g of alumina, 5 g of activated silica gel, and 2 g of Na_2SO_4. 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-d_{14}, 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 (i.e., the mass of solids) is diminished by ultrasonic radiation. In this study, the disintegration of sludge after ultrasonic treatment was confirmed by observing changes in (1) the morphology of the sludge (e.g., cell walls and flocs) and (2) the levels of TSS and VSS. These observations commonly indicated that the disintegration of sludge was enhanced by increasing ultrasonic irradiation time as shown below.

Bacterial cell wall and floc destruction increased with increasing sonication time (Fig. S1 in the Supplementary Information). Consequently, the sludge sediment's volume decreased when the supernatant color changed from transparent to brown [29] owing to the materials leaching from the sheared microorganisms [30], resulting in an increased soluble chemical oxygen demand (SCOD) [31].

The levels of TSS and VSS decreased as the ultrasonic irradiation time increased (Fig. 1). The largest changes in these levels occurred from 15 min to 20 min with reduction ratios of 19.7% and 8.2%, respectively. Previous studies also reported that the levels of TSS and VSS decreased with ultrasonic treatment [20,30]. These results can be explained by the conversion of organic matter from insoluble to soluble after ultrasonic treatment [30].

3.2. Levels and patterns of PAHs in sludge without ultrasonic treatment

The profiles of the 16 priority PAHs in the supernatant and sediment of sludge without ultrasonic treatment are shown in Fig. 2. The total concentrations of PAHs ($\sum_{16}$ PAHs) in each phase (the sediment and the dissolved and particulate phases of the supernatant) were 2.10 µg/g, 1.87 µg/L, and 2.37 µg/L, respectively. The total concentration of PAHs in the sludge sediment (cake) in this study (2.10 µg/g) was comparable with those of previous worldwide studies (Table S1 in the Supplementary Information). This level is under Korea's guideline (4 µg/g of $\sum_6$ PAHs) [32], the proposed limit for use on land by the European Commission (6 µg/g of $\sum_9$ PAHs) [33], and the guidelines of several European countries (Denmark: 3 µg/g of $\sum_9$ PAHs, Sweden: 3 µg/g of $\sum_6$ PAHs, and France: 9.5 µg/g of $\sum_3$ PAHs) [6].

Among the 16 priority PAHs, Nap and Acy were dominant in the supernatant (both in the dissolved and particulate phases) and sediment, respectively (Fig. 2a). Flu and Phe, both with three rings, also were in relatively high concentrations in the supernatant, whereas the levels of high-molecular-weight (HMW) PAHs with five and six rings were relatively high in the sediment due to the hydrophobic nature of HMW PAHs. In other words, HMW PAHs have relatively high values of K_{OW} (octanol–water partitioning coefficient), thus, they showed high affinity to the sludge sediment with high contents of organic materials.

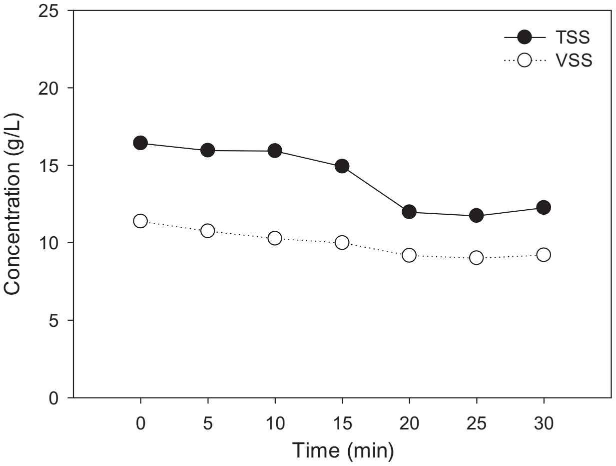


Fig. 1. Influence of ultrasonic irradiation time on the levels of TSS and VSS at an irradiation intensity of 600 W.

Overall, Nap, Acy, Phe, Ant, and DahA accounted for 59% of the total concentration of PAHs in the sludge sample with the highest fraction from Acy, followed by Nap (Fig. 2b). The fraction of Nap (PubChem CID: 931) was higher in the supernatant than it was in the sediment due to its high water solubility, whereas Acy (PubChem CID: 9161) was more partitioned to the sediment [34].

The profile of PAHs with high fractions of Nap and Acy in this study is very similar to that of sewage sludge collected from Poland [5]. The dominance of Nap was also found in wastewater sludge collected in Korea [4] and Tunisia [28]. As mentioned above, the main source of wastewater in this study is related to the production of terephthalate [35]. Phthalic acid is one terephthalate isomer that is produced by the catalytic oxidation of Nap derived from petroleum [36]. Therefore, the high fraction of Nap in the sludge sample seems to be related directly to the production of terephthalate and its isomers.

Meanwhile, the high fraction of Acy generally has not been found in other studies [3,7,37,38], which reported higher fractions of other compounds with three and four rings, especially Phe. Acy is commercially produced for use in pesticides, dye synthesis, and the manufacture of plastics [34]. Therefore, the high fraction of Acy in the sludge sample may be due to the commercial production of Acy as well as a release from raw materials or by-products of petrochemical/thermal processes. This inference, however, could not be confirmed due to limited data available from the chemical company.

3.3. Changes in PAH levels and patterns after ultrasonic treatment

The influence of ultrasonic treatment (600 W, 0–30 min) on the levels and patterns of PAHs with different numbers of rings in the three phases of sludge is displayed in Fig. 3. The concentrations of individual PAHs with and without 30 min of ultrasonic treatment are listed in Table S2. Generally, the levels of $\sum_{16}$PAHs in the supernatant increased with irradiation time (Fig. 3a and b), whereas those in the sediment slightly decreased (Fig. 3c). This result indicates that PAHs associated with the biomaterials of microorganisms were released into the supernatant by the ultrasonication. Biosorption of PAHs and other persistent organic pollutants is a well-known phenomenon, and even proteins as well as cell bodies have a strong affinity with aromatic pollutants [39].

The concentration of $\sum_{16}$ PAHs in the dissolved phase of the supernatant increased almost twofold after 15 min, and then it decreased and reached the level at 5 min (Fig. 3a). In contrast, the concentration of $\sum_{16}$ PAHs in the particulate phase of the supernatant was rather constant until 15 min, sharply increased at 20 min, and remained at approximately that level for the remaining time (ANOVA on rank, $p > 0.05$) (Fig. 3b). These time trends suggest that PAHs loosely bound to the microorganisms were released preferentially into the supernatant due to their low K_{OW} values until the full disintegration of the microorganisms. After substantial disintegration of the microorganisms, the PAHs strongly associated with the microorganisms instantly were leached into the supernatant in particulate form. This interpretation is supported by the time trend of TSS shown in Fig. 1.

The reasons for the unexpected decrease of $\sum_{16}$ PAHs in the dissolved phase after 20 min of ultrasonication are suggested as follows. First, an abrupt elution of biomaterials from disintegrated microbial cells may have induced a migration of dissolved PAHs into the particulate phase, i.e., less soluble biomaterials could act as the adsorption media. Second, a sampling artifact for dissolved

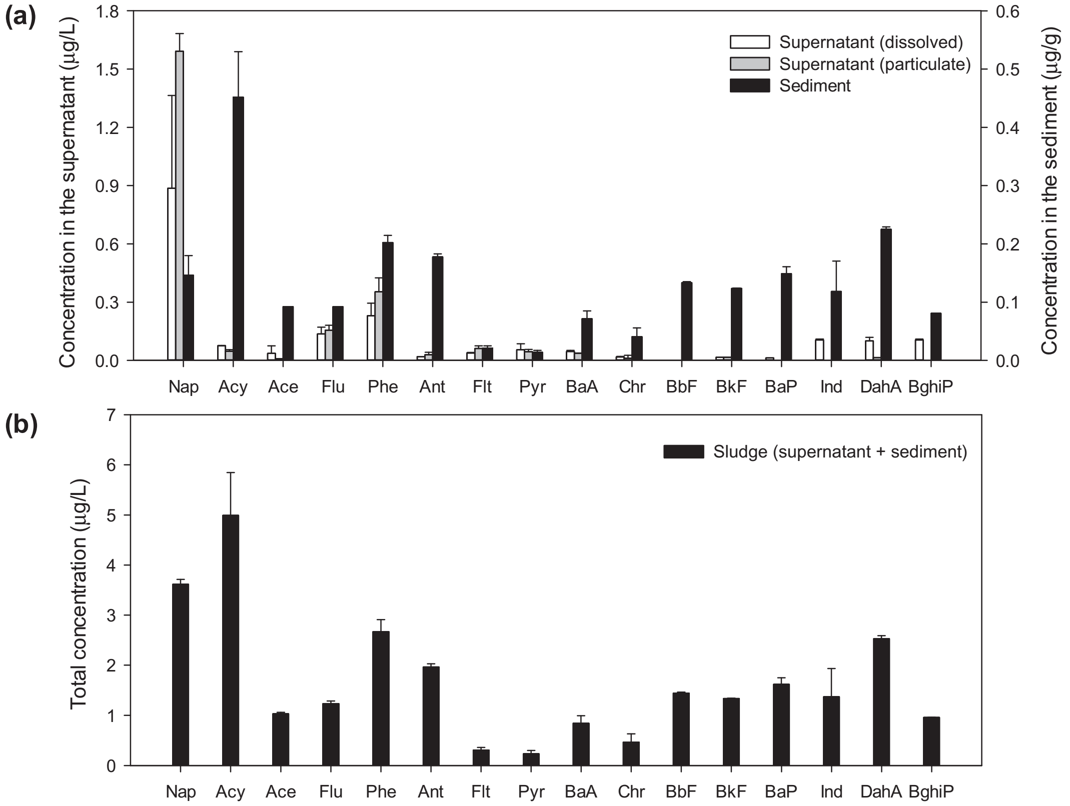


Fig. 2. Average profiles of individual PAHs without ultrasonic treatment in (a) the supernatant and sediment of sludge and (b) total sludge. Error bars represent standard deviations of triplicate data.

PAHs may exist. The pores of the GFFs seemed to become clogged with microbial cell debris as the sonication time increased. Therefore, the passage of dissolved PAHs through the filter may have been hindered by cell debris accumulation on the filter. Note that the decreasing trend of TSS with increased sonication time (Fig. 1) is opposite to the time trend of particulate PAHs (Fig. 3b).

The fractions of PAHs with different ring numbers can give insight into how the PAHs behaved after ultrasonic treatment. The fractions of PAHs in the sediment were constant during the differing sonication times (0–30 min) and dominated by 3- and 5-ring PAHs (Fig. 3c), implying that ultrasonic treatment did not significantly affect the major distribution pattern of the PAHs in the sediment. For the PAHs in the supernatant (both in the dissolved and particulate phases), Nap accounted for more than 50% of the $\sum_{16}$ PAHs, and increasing trends of Nap clearly were observed with increased sonication time (Fig. 3a and b). The PAHs with three rings also showed similar patterns to those of Nap. This result indicates that light and more water soluble PAHs were dominantly released to the liquid phase from the solid phase of sludge. The fractions of 2-, 3-, and 4-ring PAHs increased until 15 min, whereas 5- and 6-ring PAHs generally decreased in the dissolved phase, except at 15 min. It seems that heavier PAHs (those with five and six rings) with higher K_{OW} values in the dissolved phase quickly were adsorbed to particles. In the particulate phase of the supernatant, the levels of 2-, 3-, and 4-ring PAHs generally increased with sonication time. However, the levels of 5- and 6-ring PAHs were constant until 15 min due to their high K_{OW} values (i.e., they were strongly associated with microbial cells) and abruptly increased at 20 min by the substantial disintegration of the microorganisms, followed by a decreasing trend. This result suggests that heavy and more hydrophobic PAHs were bound strongly to particles, even after they were leached from the microbial cells.

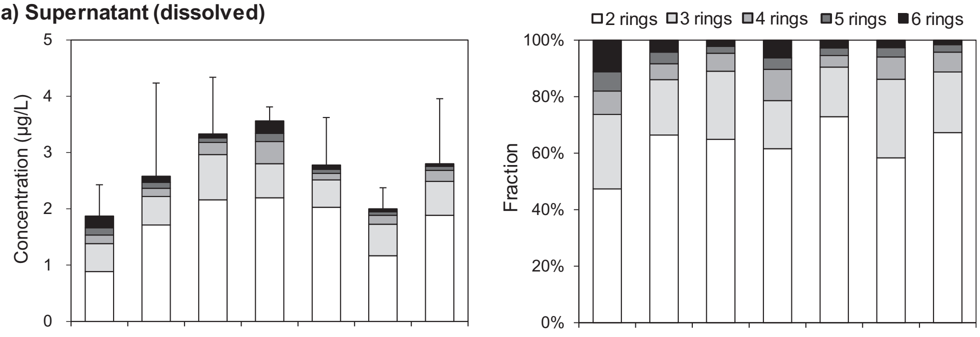
3.4. Mass balance of PAHs

The amounts and mass fractions of the $\sum_{16}$ PAHs for each phase of the sediment and the dissolved and particulate phases of the supernatant are shown in Figs. S2 and 4, respectively. The amounts of PAHs in each phase were calculated by multiplying the concentrations of PAHs by the volumes of each phase. The decreasing trend of PAH amounts in the sediment was clearly observed. Whereas the amounts of PAHs in the particulate phase of the supernatant increased with sonication time, those in the dissolved phase decreased after 20 min. As the total amounts of PAHs in the sludge samples were rather constant regardless of sonication time (one-way ANOVA, $p > 0.05$) with a relative standard deviation of 4% (Fig. S2d), it seems that there was no significant sonodegradation of PAHs. Therefore, we assumed that some of the PAHs in the sediment simply moved to the supernatant.

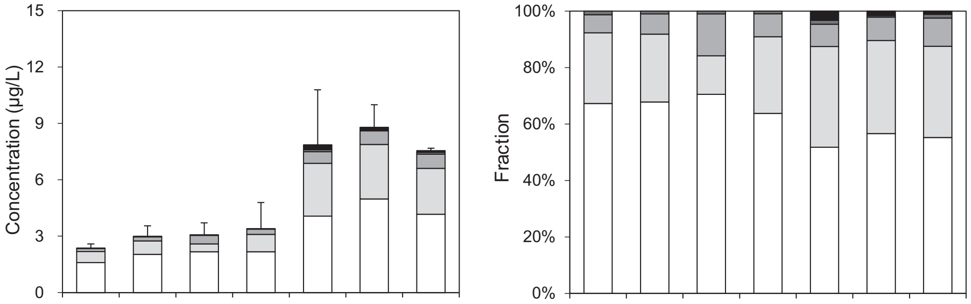
To confirm this assumption, we conducted an additional experiment for degradation of PAHs by using 500 mL of an acetone:water mixture (1:49, v/v); ultrasonic treatment at 600 W for 0, 10, and 20 min; and initial concentrations of individual PAH standards of 100 ng/L (i.e., 1.6 μg/L in total, which is similar to the level of $\sum_{16}$ PAHs in the dissolved phase of the supernatant). Other experimental conditions were the same as those of the sludge samples. According to the result of this degradation experiment ($n = 3$), there was no statistical difference in total ($\sum_{16}$ PAHs) and individual concentrations after the different sonication times (one-way ANOVA, $p > 0.05$). Namely, no discernible degradation of the PAHs was observed during ultrasonic treatment, even if sonodegradation occurred.

This result contrasts those in previous studies [25–27] that reported high removal efficiencies of PAHs (>90%) in water or wastewater with increased sonication time (up to 150 min). The

(a) Supernatant (dissolved)



(b) Supernatant (particulate)



(c) Sediment

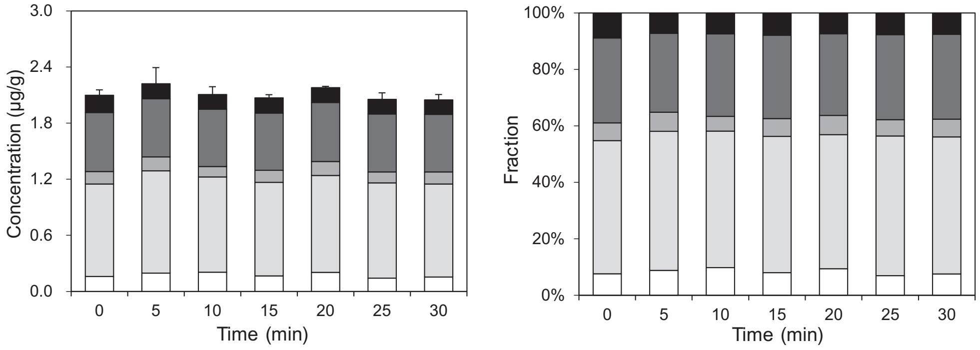


Fig. 3. Temporal changes in levels and patterns of PAHs with different ring numbers in the supernatant and sediment of sludge at different ultrasonic treatment times.

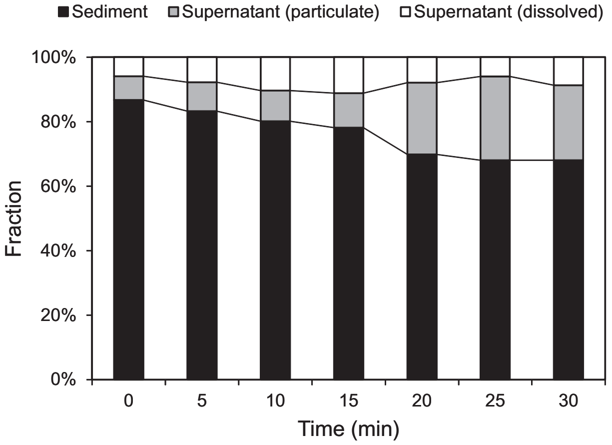


Fig. 4. Changes in fractions of $\sum_{16}$ PAHs in each phase of sludge at increasing sonication times.

levels of PAHs (e.g., 50, 100, and 126 μg/L of Phe) in these studies were much higher (approximately 1000 times) than those in our degradation experiment. Moreover, sonication times for the previous studies were longer than those in this study (i.e., up to 150 min and 30 min, respectively). These two factors seem to be major reasons for the lack of significant destruction of the PAHs in our experiment. The contradictory results could not be explained by the electric power input (W) and ultrasonic frequency (kHz). In addition, the matrix effect should be considered; high contents of microbial cells and other sludge materials could reduce the sonodegradation of the PAHs in the sludge samples.

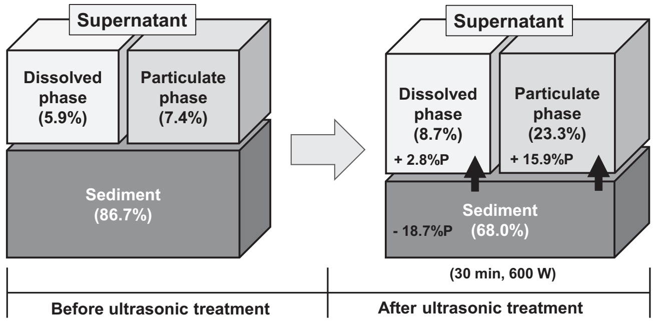


Fig. 5. Mass balances (i.e., relative fractions among the three phases) of the $\sum_{16}$ PAHs in the sludge samples before and 30 min after ultrasonic treatment.

Consequently, the sonodegradation of PAHs was not considered in the mass balance calculation before and 30 min after sonication (Fig. 5). The amounts of dissolved and particulate PAHs in the supernatant increased 1.5 times (from 5.9% to 8.7%) and 3.1 times (from 7.4% to 23.3%), respectively. Overall, 21% of the PAHs in the sediment moved to the supernatant. This result suggests that the fate of PAHs in wastewater can be influenced substantially by ultrasonic treatment.

4. Conclusions

In this study, we first reported the overall fate of PAHs in sludge after ultrasonic treatment by separately analyzing PAHs in each phase of sludge. The ultrasonication resulted in an increase and a decrease of PAHs in the supernatant and sludge sediment, respectively. The temporal change of the levels and patterns of PAHs caused by sonication suggested that light and more water soluble PAHs were released preferentially to the supernatant until the full disintegration of the microorganisms occurred. In contrast, heavy and more hydrophobic PAHs were bound strongly to particles and mainly leached after the substantial disintegration of the microorganisms.

Under our experimental conditions, the removal of PAHs by sonodegradation was negligible. According to the mass balance calculation, the reduced amounts of PAHs (21%) in the sludge sediment seemed to occur because those PAHs simply moved to the supernatant phases. This result indicates that not only dewatered sludge, but also the filtrate after ultrasonication, may contribute significantly to the loading of PAHs into aquatic environments. In conclusion, the fate of PAHs (and other trace pollutants) during the complete wastewater treatment process should be considered, especially for a WWTP equipped with an ultrasonication facility for dewatering and reduction of solid sludge in the final disposal stage.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ultsonch.2016.04.027.

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