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Author(s)	Ozaki, Noriatsu; Yamauchi, Takahiro; Kindaichi, Tomonori; Ohashi, Akiyoshi
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Relation	

Stormwater inflow loading of PAHs into urban domestic wastewater treatment plant for separate sewer system

Noriatsu OZAKI[1], Takahiro YAMAUCHI[1, 2], Tomonori KINDAICHI[1], Akiyoshi OHASHI[1]

1 Graduate school of Engineering, Hiroshima University, Higashihiroshima, 1-4-1, Higashihirohima, 739-8527, Japan
(E-mail: *ojaki@hiroshima-u.ac.jp*)
2 Now Kyushu electric power Co., Inc., Japan

Abstract
Polycyclic aromatic hydrocarbons (PAHs) are common contaminants present in wastewater, and determination of their sources is important for their management in the environment. In this study, stormwater loading of PAHs during rainfall periods was evaluated for sewage inflow into a wastewater treatment plant for separate sewer system. To accomplish this, sewage inflow volumes, suspended solid concentrations, and PAH concentrations were measured during eight rainfall events and on two no-rainfall days at the inlet of the plant. Based on a comparison between the rainfall and no-rainfall loading quantified by the measurements, excess PAH loadings with stormwater were evaluated for the rainfall events. The relationship between rainfall intensity and stormwater loading was then used to evaluate long-term stormwater loadings of water and PAHs. Their contributions to the sewage inflow were 0.7 % and 1.0 % for one-year for water and the sum of 16 measured PAHs, respectively. Our measurements and estimates demonstrate that direct stormwater inflow is not a primary source of PAHs to the plant for this separate sewer system.

INTRODUCTION

PAHs are a group of organic compounds that contain two or more fused benzene rings and have been linked to carcinogenicity and mutagenicity (IARC, 2018; Vinggard et al., 2000; Xue and Warshawsky, 2005). Polycyclic aromatic hydrocarbons are primarily generated during incomplete combustion processes, such as those that occur in vehicle engines, during the burning of coal, oil, or natural gas, and when cooking or conducting open burning, although they also originate from industrial processes, oil spills, and other sources. An important pathway for entry of PAHs into the water environment is discharge from wastewater treatment plants (WWTP) (Blanchard et al., 2004; Brändli et al., 2005; Pham and Proulx, 1997; Qiao et al., 2014). We previously estimated the load from WWTP discharge for a separate sewer system and evaluated the contribution of the sewer effluent loading of PAHs to the total PAH loading for an urban water area. We found that PAHs loading from WWTP effluent contributed up to one-tenth of total PAH loading (Ozaki et al., 2015b), indicating that the PAH loadings from WWTPs are substantial, even for separate sewer systems. In addition to the direct effluent, the emission pathway from the treatment of excess sludge is also important. The dewatered excess sludge was transferred to a composting plant, after which the products were used as fertilizer in agriculture. Hence, a large proportion of the PAHs flowing into the WWTP can be transferred to agricultural lands, after which they contaminate water environments. Further, metabolites of PAHs that might be produced in sewers, such as oxy-PAHs, may also contaminate water and soil environments (Lundstedt et al., 2007). To manage their risks, it is important to determine the sources of PAHs entering WWTPs for separate sewer systems.

The major sources of pollutants entering separate sewer systems are presumed to be private residences, industry, and road dust introduced with stormwater inflow. Depending on the location, landfill leachate from the disposal of domestic solid waste may also be the main source of pollutants including PAHs for wastewater pipelines (Ates and Argun, 2018). In our previous study, road dust with stormwater inflow was not considered to be a primary source because the water inflow from

rainfall was only 1 % of the total sewage inflow. However, this speculation has not been confirmed because the ratio of stormwater inflow to the total for pollutants is not necessarily proportional to that for water. Urban atmospheric deposition is generally considered to be an important pathway for PAHs in the environment (Cavalcante et al., 2009; Li, et al., 2009; Yunker et al., 2002; Zheng et al., 2014), and PAHs associated with atmospheric particulate matter, or more near-surface suspension (Al Ali et al., 2017) have been found to be deposited on urban surfaces and discharged to water environments with stormwater runoff. It is known that WWTPs for combined sewer systems receive pollutants from urban surfaces in stormwater runoff (Gasperi et al., 2010). In separate sewer systems, the sewer pipes are separated from the stormwater pipes, which is meant to stop entry of stormwater into the sewage system. However, stormwater is known to flow into and/or infiltrate into sewer pipelines (Belhadj et al., 1995; Itoyama and Kitatani, 2010; Karpf and Krebs, 2005; USEPA, 2014). Moreover, stormwater inflow due to the misconnection of pipelines is another possible route of entry. The problem of misconnection of sewer networks has mainly been discussed from the viewpoint of outflow to receiving aquatic systems (Revitt and Ellis, 2016; Chandler and Lerner, 2015; Panasiuk et al., 2015). At the same time, stormwater can flow into the sewage networks through these systems. Another possible way is from the pick holes of manholes which were originally sealed with rubber caps, which had likely corroded with time. As a result, they now provided access to sewer pipes for stormwater from the road during rainfall periods. Therefore, we considered nonpoint source pollutants, such as PAHs from urban surfaces, possibly enter the WWTPs through these open holes or misconnections.

In this study, the PAHs and sewage loadings were measured during rainfall periods at the inlet of a WWTP for a separate sewer system, and the inflows of stormwater and PAHs were estimated by comparing the loadings during rainfall and no-rainfall periods. Based on the relationship between rainfall and stormwater PAHs loading obtained from the measurements, the long-term contribution of stormwater PAHs to the WWTP was evaluated. Possible sources of PAHs for separate sewer systems are then discussed based on these results.

MATERIALS AND METHODS
Description of the WWTP
Samples were collected from a WWTP for separate sewer system located in Higashi-Hiroshima City in Japan (**Fig. 1**). The average yearly precipitation was 1,500 mm (1,100–2000 mm; for 2008–2018) and yearly average temperature was 14 ˚C. The monthly precipitation was higher in summer season and lower in winter season (ca. 100–400 mm/month in June and July, and ca. 20–100 mm/month in December and January). During the sampling campaign (Oct. 2013 – Aug. 2015), the monthly precipitation was 307 mm for the maximum (Aug. 2014) and the 39.5 mm for the minimum (Jan. 2014), and the mean value during the period was 127 mm. The treatment plant serves several discrete areas (1,300 ha in total) containing 50,480 people, and the inflow was 32,000 $m^3 d^{-1}$ (from April 1, 2015 – March 31, 2016; yearly average = 630 L $person^{-1} d^{-1}$). About half of the inflow comes from major industries and offices, while the rest is from private residences. The ratio of uncertain water was 6 %. Stormwater is discharged separately into stormwater pipes. The influent sewage biochemical oxygen demand and suspended solids (SS) were 180 ± 48 mg L^{-1} and 135 ± 35 mg L^{-1}, respectively.
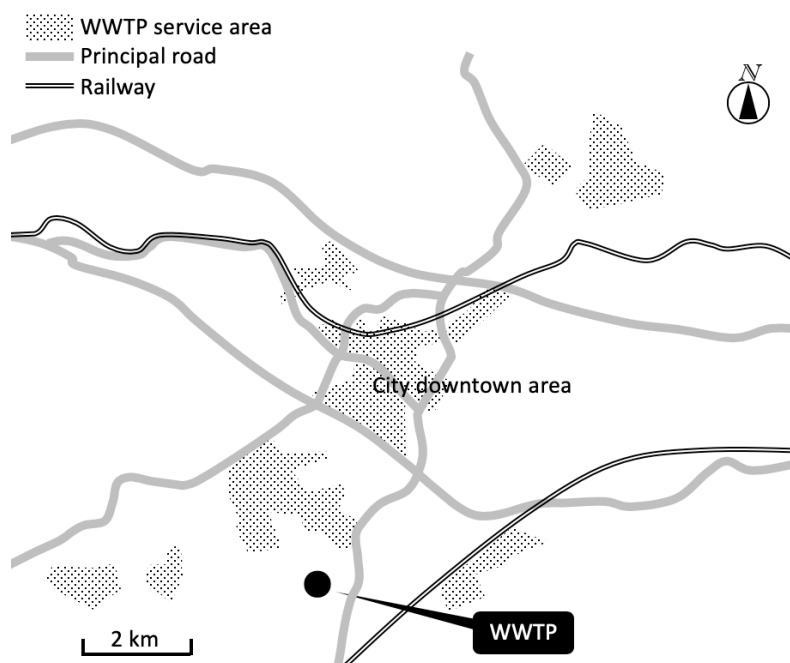
Fig. 1. Location of WWTP and its service areas

Sampling methods

Sampling was conducted twice for no-rainfall periods (**Table 1**) and eight times for rain periods at the inlet of the WWTP from October 2013 to August 2015 (**Table 2**). For the no-rainfall periods, sampling started at 0:00 and lasted for 24 h with a 2-h sampling interval. For the rainfall periods, sampling was performed intermittently with intervals ranging from 5 min to several hours. Samples were intended to be collected before raining, frequently during rainfall (5–30 minute intervals), and then after raining.

Table 1. Sampling during no-rain periods

Sampling date		Total rainfall during latest three days (mm)	SS* (mg L^{-1})	PAHs* (ng L^{-1})
`14/ 6/13(Friday)	0:00 − 24:00	2.0	131± 6	510±95
`15/ 2/ 3(Tuesday)	0:00 − 24:00	0.0	195±64	1060±96

n = 13 for each sampling
*mean±sd

Table 2. Rainfall samplings

Sampling rainfalls	SS* (mg L^{-1})	PAHs* (ng L^{-1})	Number of sampling	Total rainfall (mm)	Hourly maximum rainfall intensity (mm h^{-1})	Antcedent dry period (hr)
`13/10/05 05:00 -`13/10/ 6 3:00	104±12	166±53	7	48	10	551
`13/10/11 05:00 -`13/10/11 13:00	61±12	56±41	8	15	7.5	48
`13/10/23 13:00 -`13/10/26 22:00	155±47	518±195	10	165	11.5	4
`13/11/25 05:00 -`13/11/27 20:00	140±27	195±37	7	24	3	48
`14/01/08 14:00 -`14/ 1/ 8 22:00	175±13	229±68	8	18	4.5	255
`15/ 6/18 01:00 -`15/ 6/19 08:00	161±83	122±71	14 (26 for SS)	20	2.5	33
`15/ 7/13 23:00 -`15/ 7/14 18:00	136±85	149±76	12 (22 for SS)	23	15	121
`15/ 8/25 06:00 -`15/ 8/26 0:00	161±80	192±52	23 (25 for SS)	49	12	69

*mean±sd

For analysis, 1 L sewage samples were collected manually with a grab sampler, after which the

concentrations of suspended solid (SS) and 16 PAHs (**Table 3**) were measured. In addition, a field blank was processed once with every sampling event. The concentrations were below the detection limits for the field blanks.

Table 3. List of measured PAHs and their instrumental detection limits

Name	Abbreviation	IDL* (pg)	Name	Abbreviation	IDL* (pg)
Acenaphtylene	Acty	0.10	Chrysene	Chr	0.10
Acenaphthene	Acen	0.10	Benzo(b)fluoranthene	B(b)F	0.14
Fluorene	Flu	0.23	Benzo(k)fluoranthene	B(k)F	0.45
Phenanthrene	Phe	0.10	Benzo(e)pyrene	B(e)P	0.10
Anthracene	Ant	0.10	Benzo(a)pyrene	B(a)P	0.50
Fluoranthene	Flt	0.10	Dibenzo(ah)anthracene	D(ah)A	0.74
Pyrene	Pyr	0.10	Benzo(ghi)perylene	B(ghi)P	0.38
Benzo(a)anthracene	B(a)A	0.10	Indeno(123-cd)pyrene	Ind	0.43

*IDL: Instrument detection limit (pg injected)

Inlet water flow data were obtained from the WWTP office, and rainfall data were obtained from a weather station located approximately 800 m from the WWTP. The data collection interval was 1 h for both.

Pretreatment
Water samples were filtered using glass fiber filters (pore size: 0.7 μm, GF/F, Whatman Co. Ltd., Pittsburgh, Pennsylvania). Filters were dried for 3 days at room temperature in a desiccator in the dark. Sludge samples were freeze dried and stored at −4°C. Laboratory glassware used for analytical purposes was cleaned with dichloromethane (DCM) and then heated at 400°C for 1 h. All extractions were performed within 48 h of sampling. For filters and dried sludge, a sample was extracted with DCM in an ultrasonic water batch, after which the extract was concentrated into 2 mL with N_2-gas. For filtered water samples, an aliquot was percolated through a silica column cartridge (Waters Sep-Pak Plus tC18), which was conditioned with ethanol at a flow rate of 3 mL min^{-1}. The cartridge was subsequently dried under vacuum for 1 h. Following entrapment, DCM was passed through the column and concentrated to 2 mL by evaporation under N_2 gas. A mixture of acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12 was applied as internal standards for the correction of GC/MS peak detection sensitivity.

Instrumental analysis
The PAH concentration was analyzed by using a gas chromatograph equipped with a mass spectrometer (GC-17A/MS-QP5050; SHIMADZU) and operated in the single-ion monitoring mode. Injection was split with the detector, and the inlet temperature was set at 280°C. The initial temperature was 80°C held for 2 min, ramped at 30°C min^{-1} to 210°C, ramped at 5°C min^{-1} to 295°C, and ramped at 2°C min^{-1} to 315°C with 16 mL min^{-1} He as carrier gas. The mass transfer line and ion source were held at 250°C. Sixteen unsubstituted PAHs were measured (Table 3).

Quality control
The detection limit was set at the level of 3 in the SN ratio. Instrument detection limits (IDL) ranged from 0.1–1 pg for each species. Within this level, the coefficient of variation of each of the compounds was less than 20%. The quality of extraction was checked using dried marine sediments (HS-3B, National Research Council of Canada Institute for Marine Biosciences). The recovery averaged 50–80% and the repetition error was 5–10%.

Estimation of no-rain sewage inflow volume
No-rain sewage inflow at a rainfall time was evaluated as follows: an average was taken for the

values of sewage inflow volume at the same time of day from four weeks before until four weeks after the time, excluding the values at the time before which any rain was observed in 6 h. This averaged value was regarded as the sewage inflow excluding stormwater, i. e., no-rain sewage inflow. A 6 h period was selected because upstream runoff was thought to stop within 6 h. From the observation of rainfall data and sewage inflow data for clear rainfall events, the peak time shift was less or equal to 3 hours. Hence, longest residence time was evaluated to be 6 hours, as a double of that 3 hours.

The above calculation is valid if a clear diurnal fluctuation pattern exists for sewage inflow. In order to validate this, the diurnal fluctuation pattern of no-rain sewage inflow volume was calculated by averaging the sewage inflow for every hour for a year. For this calculation, sewage inflow data at the time of rainfall and in the 6 h before the rain occurred were similarly excluded.

Calculation of stormwater inflow for rainfall event

Stormwater inflow at a rainfall time was calculated by subtracting the no-rain sewage inflow, which was calculated as explained in previously, from the total sewage inflow at the time. The rainfall and stormwater inflow volume data were a 1-h interval, obtained as explained in the previous section.

Estimation of stormwater PAH loading

Stormwater PAH loading for a rainfall event was estimated by subtracting the no-rainfall PAH loading from the total PAH loading for a rainfall event using the following equations:

$$L_{Stormwater} = L_{Total} - L_{No-rain} \qquad (1)$$

$$L_{Total} = \sum_{i=1}^{n-1} \frac{C_{Total(i)} F_{Total(i)} + C_{Total(i+1)} F_{Total(i+1)}}{2} \Delta t \qquad (2)$$

$$L_{No-rain} = \sum_{i=1}^{n-1} \frac{C_{No-rain(i)} F_{No-rain(i)} + C_{No-rain(i+1)} F_{No-rain(i+1)}}{2} \Delta t \qquad (3)$$

where $L_{Stormwater}$ is the stormwater PAH loading (g event^{-1}); L_{Total} is the total PAH loading (g event^{-1}); $L_{No-rain}$ is the no-rain sewage PAH loading (g event^{-1}); F_{Total} is the total sewage inflow volume (m^3 hr^{-1}); C_{Total} is the inflow PAH concentration (g m^{-3}); $F_{No-rain}$ is the no-rain sewage inflow volume (m^3 hr^{-1}); $C_{No-rain}$ is the no-rain sewage PAH concentration (g m^{-3}); Δt is the sampling interval (hr); and i is the number of sampling, with $i = 1$ when a rainfall event starts and $i = n$ when the stormwater inflow of the rainfall event ends. The total PAH loading (L_{Total}) was determined directly from the samples taken during a rainfall event. To estimate the no-rainfall loading ($L_{No-rain}$), the no-rain sewage inflow ($F_{No-rain}$) and no-rain PAH concentration ($C_{No-rain}$) were required. The no-rain sewage inflow was presumed to have a stable diurnal fluctuation and calculated as explained in the previous section. For calculation of the no-rainfall loading during a rainfall event, $C_{No-rain}$ was presumed to be constant within a rainfall event.

RESULTS AND DISCUSSIONS

Sewage inflow during no-rainfall periods

To quantify the excess sewage inflow during a rainfall event, i. e., the stormwater inflow, the no-rain sewage inflow volume during a rainfall event was first estimated based on the 24-h fluctuation (**Fig. 2**). The fluctuation in a day was ~1000 m^3 hr^{-1} and the variation for a same time of a day for different days and ~200 m^3 hr^{-1}. The fluctuation in a day was higher than the variation for different days, indicating the existence of a clear diurnal cycle for no-rain sewage inflow.
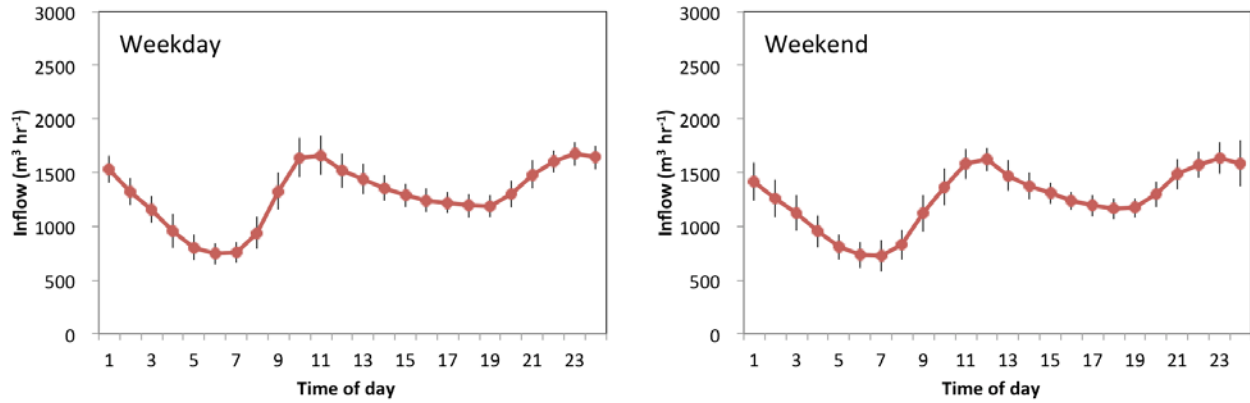
Fig. 2. Yearly mean of sewage inflow volume during no-rainfalls
(April 1, 2014–March 31, 2015, Total number of days: 181.4 days for weekday and 105.7 days for weekend, ±bars represent standard deviation)

Relationship between rainfall and stormwater inflow
The relationship between rainfall and stormwater inflow volume for every rainfall event is shown in **Fig. 3**. The total stormwater inflow increased with the total rainfall. This tendency is reasonable and the method of the prediction of no-rain inflow volume during a rainfall period was found to be appropriate.
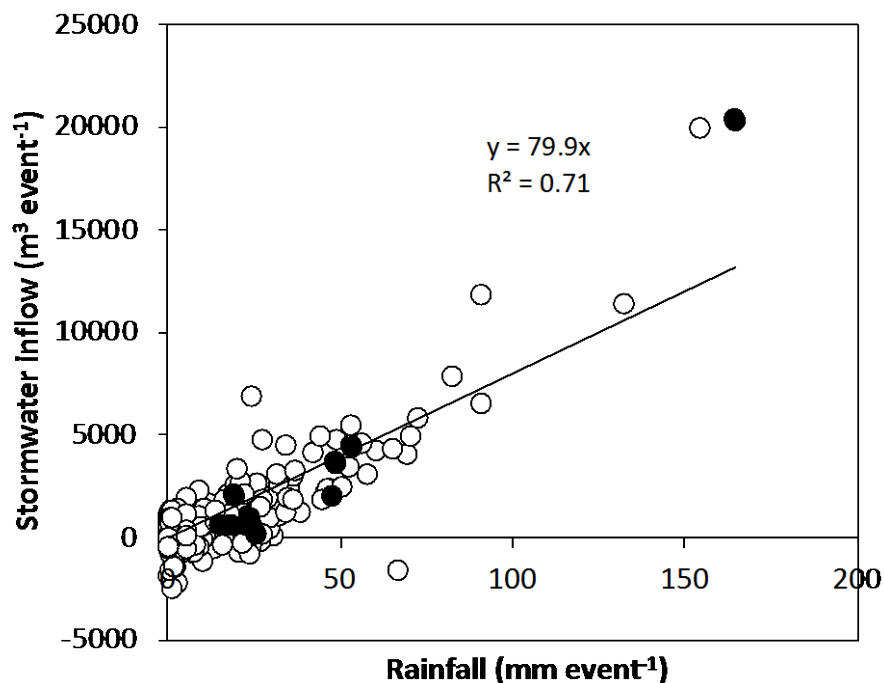

$$y = 79.9x$$
$$R^2 = 0.71$$

Fig. 3. Relationship between rainfall and stormwater inflow
(`12/04/01–`16/03/31; Filled circles denote the events of PAHs sampling)

PAHs inflow during no-rainfall periods
Figure 4 shows the results of the two 24-hour measurements of the inflow of the 16 PAHs during no-rainfall periods, along with the yearly mean of no-rain sewage inflow, obtained as described above. The total sewage inflow was consistent with the yearly mean values, suggesting both samplings can be regarded as typical no-rain period. The concentrations of the 16 PAHs were 510 ± 95 ng L^{-1} and 1060 ± 96 ng L^{-1} (mean $\pm$ sd) for the first and second measurements, respectively. Also, we took PAHs data for no-rainfall in the rainfall samplings resultantly, shown below (the data

at red arrows in Fig. 5). The value was 142 ± 129 ng L^{-1} (n = 11).

No clear diurnal fluctuation was observed for the PAHs concentration. However, the concentrations recorded on the different sampling days varied considerably. We previously measured the PAHs sewage inflow at the same WWTP for one year between 2005 and 2006 (Ozaki et al., 2015b). In that study, we also found the PAHs concentration was stable within one day but fluctuated greatly over the year (44–1028 ng L^{-1}; 219 ± 210 ng L^{-1} (n = 28), mean ± sd). On the basis of these findings, it can be inferred that the no-rain PAHs concentration changes day to day, even if there is no rainfall, and that it also remains stable for an entire day, or at least, for several hours. The mechanisms behind this contradictive tendency (short-term stability and long-term high fluctuation) were not clarified in this study; therefore, future studies to clarify these mechanisms are warranted.
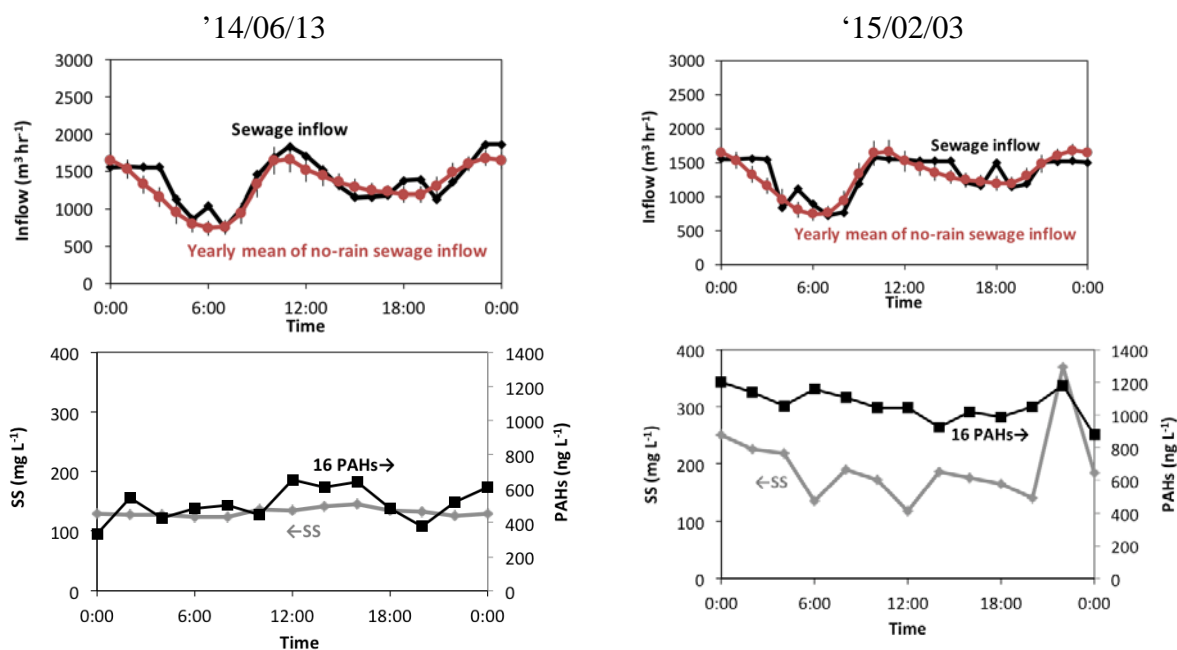

Fig. 4. Diurnal changes of Sewage inflow, SS, and 16 PAHs concentrations during no-rain periods

PAHs inflow during rainfall

To evaluate the stormwater loading of the 16 PAHs during rainfall, the total loading and no-rainfall loading were measured for the eight rainfall events (**Fig. 5**). The mean values for every events were shown in **Table 2**. The variation of PAHs concentration within each rainfall event was ~500 ng L^{-1}, which was substantially higher than that in no-rain days (~100 ng L^{-1}). From the measurements, the total and no-rain sewage loadings were calculated using Eqs. (2) and (3), and the difference was taken. To evaluate the baseline loading during rainfall (Eq. (3)), the no-rain sewage inflow volume ($F_{No-rain}$) was determined as explained above, and the no-rain PAHs concentration ($C_{No-rain}$) was presumed to be constant during rainfall. $C_{No-rain}$ was determined as the average of the PAHs concentrations before and/or after rainfall. **Figure 6** shows the results for the total and the predicted no-rain sewage loadings of the 16 PAHs for all the measured eight rainfall events. The total loading was higher than the predicted no-rain loading in most cases. This difference is defined as the stormwater loading of PAHs for every rainfall event. The relationships between stormwater volume and PAHs stormwater loading for the measured rainfall events are shown in **Fig. 7**. The PAHs inflow increased proportionally with the predicted stormwater inflow. Stormwater passed through the surface of the urban area and flows into the sewage pipeline from unintentional holes of manholes or pipe cracks. The PAHs are known to deposit on urban ground surfaces with high concentration (Aryal et al., 2013; Hassanien and Abdel-Latif, 2008; Liu et al., 2007; Ozaki et al., 2015a; Zhang et al., 2008), and hence, their flowing into the sewage pipeline would be one important PAHs sources for the measured excess PAHs loadings in this research.
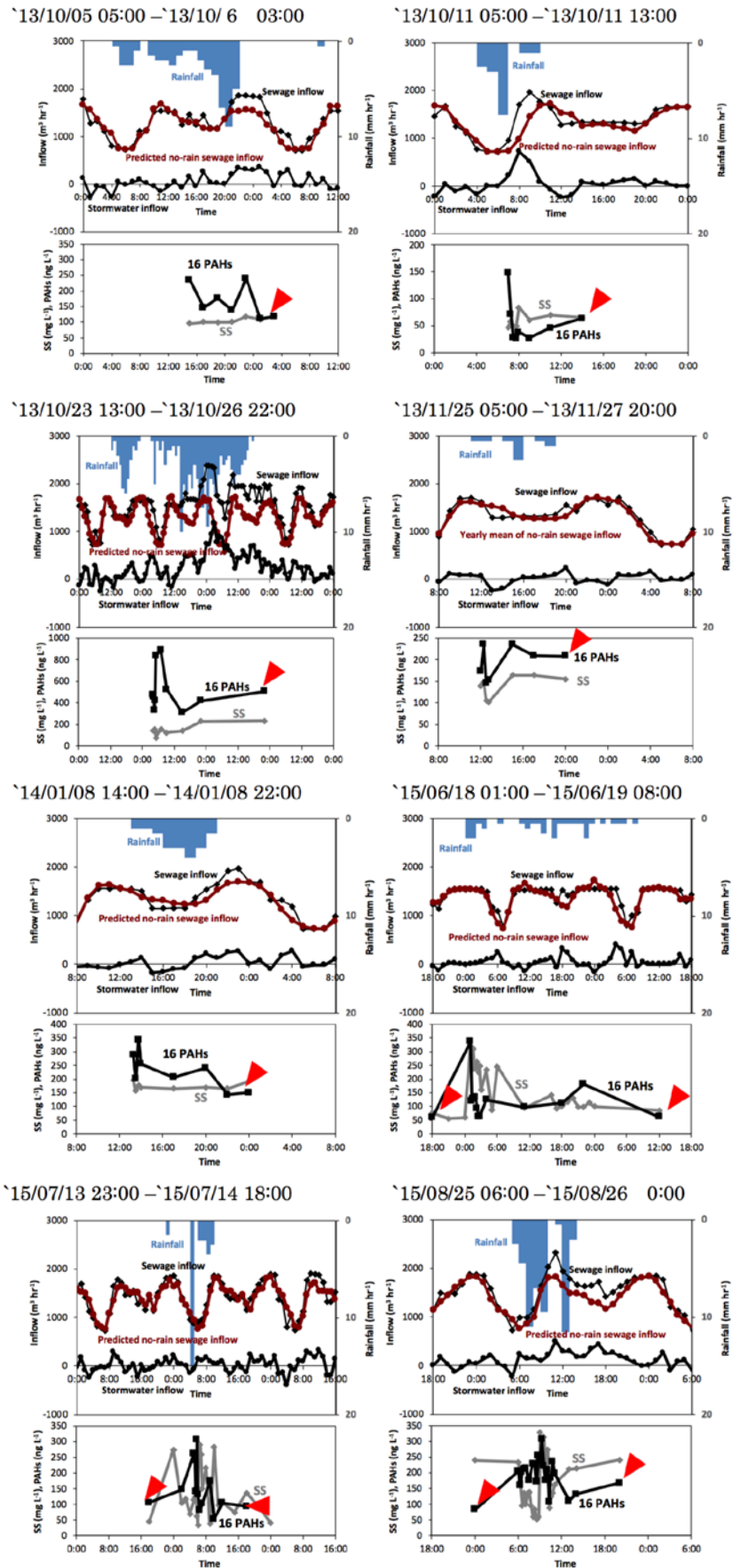
Fig. 5. Sewer inflows during rainfalls

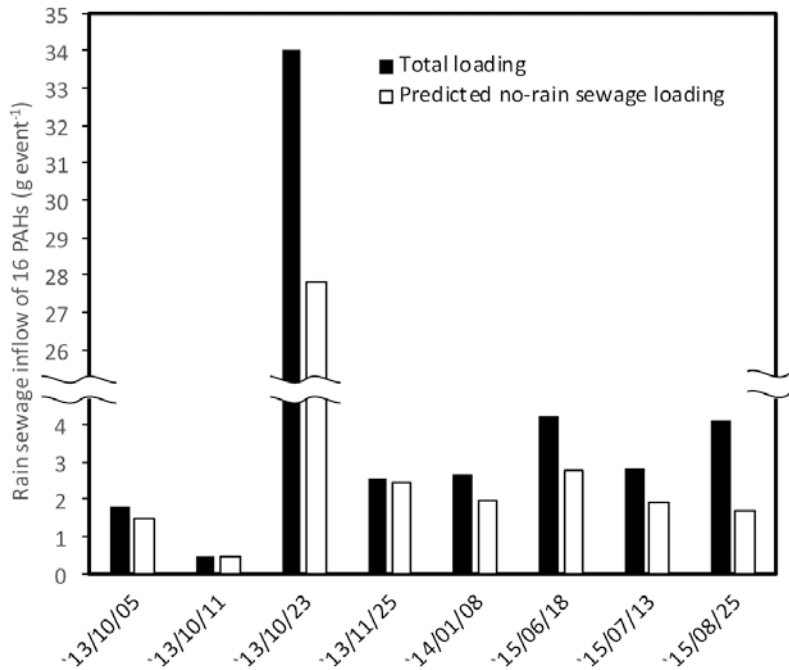(The arrows indicate the samplings that are used for no-rain PAHs concentrations for the rains)

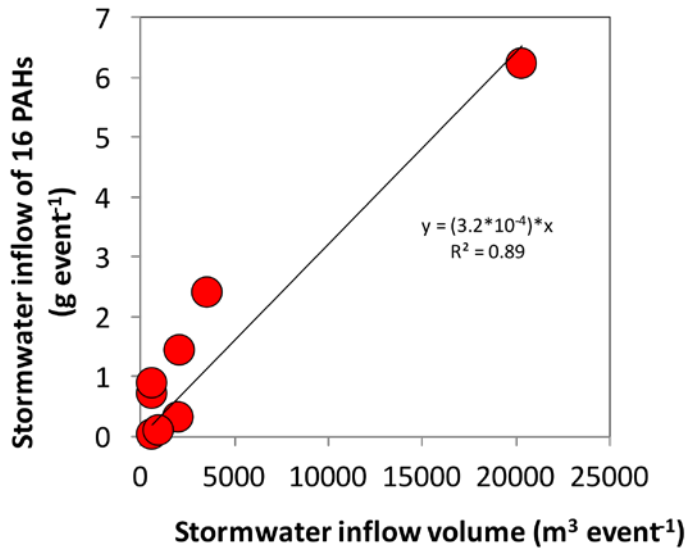**Fig. 6.** Sewage inflow loading of PAHs for each rainfall

Fig. 7. Relationship between stormwater inflow volume and 16 PAHs

Contribution of stormwater inflow of PAHs to WWTPs for separate sewer systems

Using the data obtained above, the total sewage inflow and stormwater inflow were evaluated for a year (April 1, 2014 – March 31, 2015; total precipitation: 1439.5 mm; number of rainfall events: 110) for sewage volume and PAHs. The total sewage inflow volume was 1.16×10^7 m^3 year^{-1}, and the stormwater inflow volume obtained by summing every rainfall event was 7.68×10^3 m^3 year^{-1}. Therefore, the contribution of stormwater inflow volume to the total was 0.70 %. The total sewage inflow of the 16 PAHs was subsequently estimated as the product of the total sewage inflow volume and the mean concentration. The yearly mean concentration was estimated for the same WWTP in

9

our earlier study (219 ± 81 ng L^{-1} (n = 28), mean ± 95 % confidence interval (CI); Ozaki et al., 2015b), and this value was used for the calculation in the present study. The data from another study (Ozaki et al. 2015b) were used because of their reliability. In our previous study, the influences of stormwater inflow were not considered as the main topic. From the present study, their effects were confirmed to be insignificant. Hence, since systematic measurements were performed over one-year in the previous study, we consider it to be better to use these values to calculate the annual averaged loading value. We calculated the total sewage inflow of the 16 PAHs to be 2544 g year^{-1}. In addition, the stormwater inflow of the 16 PAHs was estimated for every rainfall event from the relationship between the stormwater inflow volume and the 16 PAHs, and then summed for one year. The total was 26.5 g year^{-1}, which was only 1.04 % of the total sewage inflow. For a conservative approach, the possible maximum contribution for this calculation was predicted. First, the mean − 95 % CI (219 − 81 = 138 ng L^{-1}) was taken as the lowest yearly mean concentration. The mean + 95% CI of the slope of the equation describing the relationship of the stormwater inflow volume and 16 PAHs, which was 4.0×10^{-4} (g event^{-1})/(m^3 event^{-1}), was then taken as the highest slope of the stormwater inflow of the 16 PAHs/stormwater inflow volume. Using these values, the total sewage inflow and stormwater inflow of the 16 PAHs were calculated similarly as described above, and the contribution of the stormwater inflow to the total inflow was found to be up to less than 3 % (2.06 %), which was still considered low.

Another possible estimation error is the relationship between rainfall and stormwater inflow. Stormwater inflow was hypothesized to be proportional to rainfall. However, the minimum detection value of rainfall was 0.5 mm hr^{-1}, which may not be discharged to the pipelines; therefore, the hypothesis of this proportional relationship would be crude. In this case, the estimated rainfall stormwater contribution would be overestimated. During the period of calculation, the frequency of the precipitation equal to or less than 2 mm was 28 %, and the contribution for rainfall depth was 1.7 %, which was similarly low.

Road surfaces are also highly polluted in urban areas, and the inflow of PAHs from more polluted roads may be higher than that from less polluted roads. We previously measured the PAH concentrations and toxicities in excess sludge from several WWTPs for separate sewer systems, including the plant investigated in the present study (Ozaki et al., 2017). We found the PAH concentrations and toxicities tended to be higher in urban areas. For the WWTP in this study, direct inflow from urban surfaces was not considered a major source. However, PAH concentrations that were higher than those for the present WWTP were observed for a WWTP in a more urbanized residential area. In such areas, the contribution of urban surfaces cannot be ignored if substantial stormwater inflow occurs.

The effects of long-term in-pipe storage should also be considered. Since PAHs are generally hydrophobic and easily bond to solid matter, they can sediment with solids in pipelines. Such sedimentation has been considered for combined sewer systems (Ashley et al., 2002; Fan et al., 2003), but can also occur in separate sewer systems. If sedimentation occurs, the PAHs will not be discharged directly at the time of rainfall, and they will instead be retained within the pipeline until being discharged at a later time. In this case, the rainfall inflow and outflow would be balanced over the long term (if in-pipe decomposition is negligible). If this is the case, the observed levels of PAHs entering the WWTP from rain would be low, even with high stormwater inflow, and our estimation of no-rain contribution would be overestimation. In our no-rain measurements for PAHs, the observed contradictory tendency of short- and long-term may possibly be explained by this mechanism. Accordingly, the possibility of such sedimentation effects should be examined in future studies.

CONCLUSIONS

In this study, sewage inflows of 16 PAHs were measured at the inlet of a WWTP for a separate

sewer system during periods of rainfall, and stormwater inflow loadings of water and PAHs were evaluated. The contribution of the stormwater inflow to the total was 0.70 % and 1.04 % for water volume and PAHs, respectively, indicating that urban surface water runoff was not a primary source for the WWTP in this study. However, the PAHs that enter via rain inflow may make non-negligible contributions to the total inflow of PAHs into WWTPs when the stormwater inflow is 5 % – 10 %, which is possibly the general case for separate sewer systems in Japan.

In the WWTP for the present study, private residences and/or industries have considered a possible major source of PAHs in the WWTP from our previous study, as explained above. But from the present study, stormwater runoff from urban surfaces was not thought to be a primary source because the direct inflow from rain was very low. For future studies, private residences should be targeted to clarify the major sources of PAHs because it is easier to assume a more general situation for private residence loading than for the industries. Investigation of plants for purely residential areas, such as community plants, would be desirable for such a study.

ACKNOWLEDGEMENTS

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