論 文 の 内 容 の 要 旨

論文題目　Environmental fate modelling of PPCPs and microbial fecal indicators in Tokyo coastal area after rainfall events
(東京港沿岸域における降雨後の医薬品類及び糞便汚染指標微生物の消長モデル解析)

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Combined sewer overflows (CSOs) are considered as one of important pollution sources to the receiving water.　Different types of pollutants are presented in CSOs which can deteriorate the water quality and consequently pose risks to　human health through exposure to pathogens or toxic substances. In order to maintain water quality and safety, the　significance of studies on impact of CSOs to receiving water is emphasized. This study targeted fecal indicator bacteria, 2 types of bacteriophages and compounds in group of Pharmaceuticals and Personal Care Products　(PPCPs) as sewage markers to represent the diffusion and to evaluate the impact of CSOs in the receiving water. Odaiba seaside park is a popular waterfront area for recreational activities. It is located in Tokyo coastal area　surrounded by many sources of CSOs pollutions. In the populated Tokyo metropolitan area, around 800　overflow chambers and 30 pumping stations are presented. Thus, the waterfront consistently contaminated after heavy　rainfall events. Therefore, Tokyo coastal area is selected as study area to evaluate potential risks of fecal contamination in　this famous waterfront area after rainfall events for public health and safety. The overall objectives of this study were to evaluate the impact of CSO events and to investigate fates and behavior of sewage markers representing CSO pollutants in Tokyo coastal area. The specific objectives include:

1) To characterize sources of CSO pollutants and to evaluate effect of CSO discharge and tidal change condition on the distribution of sewage markers.

2) To investigate attenuation processes of PPCPs including biodegradation and photodegradation mechanisms and to determine kinetic constants of these processes considering situation after rainfall events.

3) To develop PPCPs fate model and to describe temporal behavior of PPCPs after rainfall events using simulation model that consider effect of hydrodynamics and degradation.

To achieve objective 1, several survey samplings were conducted. Surface water samples were collected to monitor the surface water quality from the upstream of the Sumida River to the coastal region of Tokyo. CSO discharge was estimated separately from pumping stations and overflow chambers. The major contribution from overflow chambers was found to be as high as 86 %–91 % of total discharge volume indicating their significance in controlling CSO pollution. In addition, major sources of CSO pollutants among urban rivers were pointed out. High concentrations of sewage marker were observed in a wide area due to CSO discharge of more than 30 hours. *E. coli* was found to be as high as 4.00–4.57 $\log_{10}$(CFU/100mL), showing a relatively similar concentration in most locations after a long period of CSO discharge. Monitoring results suggested the susceptibility of microbial markers to salinity. Meanwhile, caffeine was found to have the highest concentration of 2,105 ng/L among PPCPs. It was found to be a useful indicator of recent contamination that captured a unique spatial distribution tendency. On the other hand, crotamiton, a conservative PPCP, was found to be highly diluted and might not be appropriate for tracking pollutants under heavy rainfall events. The effect of CSO discharge pattern and tidal change on the distribution of sewage markers, including dispersion degree and pollutants travel time, were described. CSO pollutants were found to accumulate in the river mouth areas during high tide before discharged into the coastal area.

To achieve objective 2, attenuation mechanisms of PPCPs including biodegradation and photodegradation were investigated through laboratory studies. For biodegradation process, experiments were designed considering possible influencing factors on biodegradation rate including microbial community, microbial abundance, and salinity. FCM technique was introduced to monitor and control microbial abundance. In addition, bacteria inoculum preparation procedure was proposed, and bacteria was collected from different sources including raw sewage and coastal water. Raw sewage was used as source of nutrients and organic matter to imitate situation after CSO events. Labile PPCPs was found to be significantly degraded during 8 days of incubation. Acetaminophen (ACE) was found to be most readily biodegradable in every salinity level with half-life as fast as 0.4 days, under concentrated live cells condition. On the other hand, conservative PPCPs were persisted to biodegradation. Degradation of theophylline (THEO) and caffeine (CAF) was found to be delayed 2 – 4 days due to bacteria preferential degradation order. They were degraded when ACE was removed from the system. Microbial community from raw sewage contributed more to PPCPs degradation under CSO events. Overall results showed low salinity favored microbial activity resulting in faster degradation.

Photodegradation tests were conducted in simulated light chamber using environmental water collected in the study area. Several influencing factors on photodegradation rate were evaluated including salinity and raw sewage addition (CSO-simulated samples). Higher photodegradation rate was observed in samples collected from river, which have higher composition of secondary effluent. Presence of high refractory DOM was suggested as a reason. Positive salinity effect on photodegradation rate was observed for ACE and THEO only when raw sewage was added possibly due to introduction of higher DOM and the fraction with simple structure which is not presented in effluent. Moreover, addition of raw sewage could affect photodegradation rate in different ways either negative effect due to high competition of light absorption or positive effect under high salinity.

Possible range of kinetic constants were obtained, which was applied as model parameters to achieve the last objective. Based on their environmental fate and persistence, the three representative PPCPs with different features were selected including acetaminophen (ACE), caffeine (CAF) and crotamiton (CTMT) to be further investigated in model analysis.

To archive objective 3, PPCPs fate models were developed. Area-specific kinetic constants that were obtained from laboratory studies were applied as model parameters. Boundary conditions such as initial concentrations, concentrations in inflow rivers and concentrations in influent and effluent of sewage treatment plants (STPs) were carefully decided based on monitoring data. In addition, sensitivity analysis was conducted to understand how the parameters affect the concentrations of PPCPs. Model calibration was conducted by comparison with monitoring data. Temporal concentration change in surface water was monitored up to 8 days after rainfall events. Spatial-temporal distribution of CTMT was simulated. Results show high correspondence between simulated and monitored concentrations in most cases. Different tendency between *E. coli* and CTMT was illustrated. CTMT was found to mainly governed by the intensity of precipitation and tidal change condition. Under long-duration rainfalls, CTMT was found to be highly diluted on the 1st day after long-duration rainfalls whereas, *E. coli* increased significantly.