Environmental impacts evaluation of onsite sanitation system using wastewater pharmaceutical indicators

（家庭排水中の医薬品成分を指標とするオンサイトサニテーションの環境影響評価）

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Abstract

Rudimentary on-site sanitation systems (OSSs) are extensively used in low-middle income countries (LMICs) despite the risk to water environment as well as human health and ecology. The first part of this study aimed to investigate the sanitary situation in LMICs and address the potential impact to environment. Pharmaceutical active compounds (PhACs) was applied as the innovative indicators due to its advantages over the traditional indicator such as fecal bacteria. The second part of this study focus on the unsaturated zone that plays an important role in pollutant removal mechanisms from the OSSs. The sub-objectives are to determine the mobility and biodegradation of PhACs in different soil types.

In the first part, field study at selected representative research site – Galle erea of Sri Lanka has provided the overall picture of sanitary situation at both urban and rural area of this developing country. Based on screening test result, a list of 8 pharmaceutical (acetaminophen, caffeine, carbamazepine, cotinine, sulfamethoxazole, sulfapyridine, atenolol, and acesulfame) were selected as sewage markers. In surface water matrix, acetaminophen was dominant over selected markers in hospital discharge (70.2–123.6 μg/L) while caffeine was the largest contributor to STP influent (16.2–68.7 μg/L) and surface drainage (0.95–21.73 μg/L). A set of criteria including removal efficiency, concentration magnitude, excretion rate, and wastewater burden were applied to consider PhACs occurrence and source. On the other hand, the detection of 3 pharmaceuticals (caffeine, carbamazepine and sulfamethoxazole) at trace levels in groundwater evidenced wastewater impact from OSSs. Caffeine was detected in 89% of well water samples, indicating the impact of fecal pollution. Carbamazepine was detected in 42% of the samples with a concentration of up to 6.9 ng/L, whereas sulfamethoxazole was detected in only 2 samples. The presence of carbamazepine and sulfamethoxazole was also consistent with recorded drug use of the residents. *Escherichia coli* showed a moderate positive correlation with caffeine concentration (Kendall's $\tau = 0.38$, $p = 0.017$), indicating concurring short-lifetime fecal bacteria and labile wastewater organic compounds. Nitrate showed a significant correlation with carbamazepine concentration ($\tau = 0.39$, $p = 0.016$). Fecal bacteria and nitrate can be used in screening for micropollutants in domestic wells impacted by OSSs.

In the case study in Galle area, we found ACT, CAF, CBZ, ACS as good marker of domestic wastewater in surface water while CAF, CBZ showed their applicability in groundwater. The case study in Sri Lanka emphasizes the potential impact of poor sanitation on aquatic environment and the necessity of sanitation improvement in achieving Sustainable Development Goals in low-middle income countries.

The mechanism of organic pollutants by sanitation – soil system was thought to consist of (1) Biodegradation of contamination within the pit; (2) Sorption onto the material of unsaturated zone; (3) Dilution of the aquifer. Part 2 of this study provided an insight look into the behavior of pharmaceuticals in three different standard soils (loamy sand, sandy loam, loam). Experiment design were based on the OECD guideline Test No. 106 for adsorption/desorption study and test no. 307 for biodegradation study. Chapter 6 investigated the mobility of pharmaceuticals in unsaturated zone through a series of adsorption and desorption experiment of pharmaceuticals on soil matrix. The result indicated that the removal efficiency of OSSs primarily depends on the physicochemical characteristics of the nature soil such as clay fraction, organic carbon and CEC. Most of the test compounds performed the moderate mobility to high mobility, especially in case of sandy soil type. The result emphasizes the risk of emerging pollutant leaching from OSSs to the shallow aquifer. The desorption process might release the loosely sorbed compound such as CBZ, CAF, COT. Chapter 7 consider the biodegradation of selected pharmaceuticals in soil under different conditions. Microbial degradation of six target PhACs (ACT, CAF, CBZ, COT, SFM, SFP) on sandy loam soil was thoroughly investigated. The result from inoculum solution presented a slight hydrolysis of ACT, COT and SFM. ACT and CAF are favorable substrates for microbial degradation, especially in aerated condition. The calculated kinetic constant k of ACT and CAF in aeration bottle were at 0.67 and 1.3 day^{-1}, respectively.

The sanitary situattion in low-income countries need to be improve in order to achieve the SDGs. Pharmaceutical wastewater indicators can be integrated to predict the impact of domestic wastewater via onsite sanitary systems. It is suggested to integrate several common PhACs (ACT, CAF, CBZ) into the groundwater monitoring program. Taken that the soil characteristic is the key factor determine the effect of unsaturated soil to protect the aquifer, before the construction of any OSSs, geological information is strictly required.