学　位　論　文　要　旨

A comprehensive modeling to bridge across laboratory, field and basin data for exposure assessment of paddy pesticide in aquatic environment
水稲用農薬の水環境中暴露評価に用いる実験室，圃場および流域データをつなぐための包括的モデリング

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In Japan, paddy pesticide applied to paddy field is more prone to runoff to the outsides of the fields, and therefore one of the major concerns for the pollution of aquatic environment. Although the pesticide safety in the environment has been rigorously screened under the standard scenarios in the registration, this approach cannot cover the regionally-variated actual field condition where the monitoring study is conducted as the exposure assessment in post-registration process. Therefore, this study aimed to develop a comprehensive modeling of paddy pesticide to assess the regional exposure characteristics of paddy pesticides.

Four-year experiments were conducted to compare the dissipation patterns of a total of 20 pesticides in various formulations applied by submerged application, nursery-box application and foliar application in flooded lysimeters (lysimeters) and paddy fields with two soil types. The similarities of the dissipation data between test plots were assessed by the simple kinetic modeling to derive DT50. For submerged application, although the lysimeters could simulate nearly half of the decreasing phase of dissipation with granular formulations in paddy fields, the accuracy of the detection level was low. This tendency was consistent for flowable formulation. For the case of nursery-box and foliar application cases, the detection levels were comparable between lysimeters and paddy fields. From these results, the submerged application scenario had the highest possibility to variate the pesticide dissipation patterns between lysimeters and paddy fields.

For more detailed analysis, an inverse analysis procedure of paddy pesticide

dissipation was developed using the mathematical model (PCPF-1R model) and open software R packages. The developed procedure was verified using the dissipation data of simetryn and molinate applied in the lysimeters and the paddy fields. The model calibration was performed by the global and local sensitivity analyses and Markov Chain Monte Carlo (MCMC) technique. From the calibrated simulations of simetryn and molinate showed that the current experimental deign of the lysimeters might underestimate the paddy fields mainly due to the faster daily percolation setting in the lysimeter. However, this problem was successfully improved by modifying experimental design of lysimeter through the case study.

To clarify the pesticide behavior in soil and interface between paddy water and soil, a laboratory container tests for flooded soils applying four herbicides were conducted. The results were subjected to in-laboratory inverse analysis using PCPF-LR model. Then, the calibrated parameters were exported to analyze the outdoor experimental data with flowable and granular formulations by in-field inverse analysis using PCPF-$1R_{v1.1}$ model. The PCPF-LR model accurately simulated the concentrations in water and soil as well as apparent sorption in the laboratory data. The calibrated simulations of the PCPF-$1R_{v1.1}$ model reasonably represented the outdoor experimental data. It was found that initial partitioning in outdoor experiment was highly affected by the physical effects rather than formulation types. Furthermore, persistence indicator ($DegT_{50}$) was consistent regardless of formulation types although DT_{50} was significantly different.

For the reginal-based pesticide exposure assessment, the improved basin scale model (PCPF-B/DRAFT 2.0 model) was proposed as the distributed hydrologic-hydraulic model by introducing a new hydrologic module. Then, a GIS processing to construct the hydrological cascading system representing the basin properties was developed. Finally, the model was tested to simulate the monitoring results of paddy herbicide (pretilachlor) in Oppe River Basin conducted as the Ministry of Environment's monitoring study in 2017. For water flow simulation, flow condition in Oppe River was evaluated regarding both discharge and water level. The simulated pretilachlor concentrations at assessment point were greatly sensitive to the behavior of pretilachlor at neighboring tributaries because of low specific discharge in Oppe River. The result of case study showed that the pretilachlor exposure in this basin could be mitigated by rigorous implementation of 7-day water holding practice after pretilachlor application.

The developed modeling approach could be useful to access or extract the quantitative characteristics of paddy pesticide by manipulating the regional uncertainties and variabilities as well as the experimental constraints. Furthermore, all experiment applied in this study were designed based on the test guidelines for the pesticide registration in Japan, and thus this approach can be also applied to the regional-based exposure assessment by using the registrant submitted data.