Analysis of Environmental Behaviour of Sulfa-drugs in the Lake Biwa and Yodo River Basin by Using Multimedia Models

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Abstract. This study focuses on some sulfa-drugs that have been heavily used in livestock husbandry and emitted into the Lake Biwa and Yodo River Basin. Emissions of sulfa-drugs were estimated using the national statistical data on veterinary drug usage and livestock excrement in Japan. One-box and distributional type multimedia fate models were utilized with the estimated emissions in order to simulate the behaviour of sulfa-drugs in four environmental media (atmosphere, soil, water and sediment) in the study period from 2005 to 2015 in the basin. Based on the calculated results, sulfa-drugs emitted to the soil media were efficiently transported to water, which resulted in the largest amount in water among the environmental media and high concentrations in and around areas with large farmland proportion.

1. Introduction

Chemicals which are the active substances of pharmaceutical and personal care products (PPCPs) have been detected in trace amounts from the environment [1-2]. Veterinary medicines, as one of the PPCPs, are used in large quantities on agriculture industry. There is an increasing concern about the ecological consequences of chronic exposure to the chemicals. Sulfa-drugs, which are typical antibiotics, are frequently found in the environment media, especially water bodies, such as river [3-4] and lake [5].When a sulfa-drug is used for human beings, the chemical substances contained therein are metabolized in the body, and then released into the sewage as a metabolite. Those left without being treated in the sewage treatment plant are released into the river. On the other hand, when a veterinary medicine is used for livestock, after metabolized in their body, the chemical substances are discharged as manure. Most of manure is reused in farmland after composting treatment and liquid fertilization treatment while some of the manure are not subject to carbonization or incineration or wastewater purification treatment and even discharge to the environment directly such as field piling or digging. This study focuses on sulfa-drugs and chooses 2 typical substances which belong to sulfonamide type synthetic antibacterial agents, and are sold in large quantities among veterinary drugs.

2. 2. Model description

2.1. Multimedia model

In order to evaluate the environmental risk, it is necessary to estimate the concentration of chemical substances in the environment. One of the methods is numerical simulation based on a multimedia fate prediction model that expresses the behavior of chemical substances in and between plurality of environmental media. In this study, the kinetics of sulfa-drugs for livestock discharged into the environment were analyzed using one-box and distributional type multimedia models (OMM and DMM, respectively) for the Lake Biwa and Yodo River basin (LBYRB). In the multimedia model, the

environment is divided into four main media, atmosphere, soil, water and sediment and some sub-media. The relationship and the mass transfer between all media are shown in figure 1. The substances movement caused by advection in the air and water media from the adjacent area is took into the consideration. The atmosphere media consists of gas phase and particle phase, and is divided into the upper layer and the lower layer corresponding to the mixed layer. The water media consists of liquid phase and suspended solid (SS) phase, and considers outflow from soil and equilibrium with sediment.
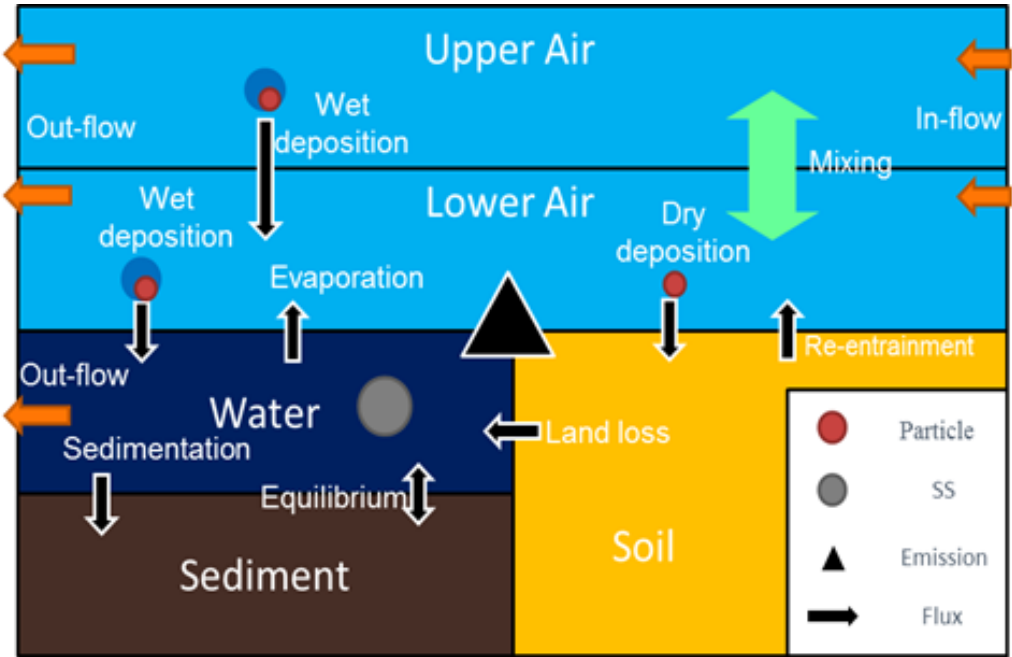

Figure 1. Structure of multimedia model.

2.2. One-box type model

The OMM assumed that the LBYRB is a huge three-dimension box which is shown in figure 2, and the concentration in each media was uniform. This imaginary box was about 111 km in the north-south direction, about 74 km in the east-west direction, and 2 km in the height direction. The inflow and outflow of air and water were considered as constant (with the average value of the whole LBYRB).
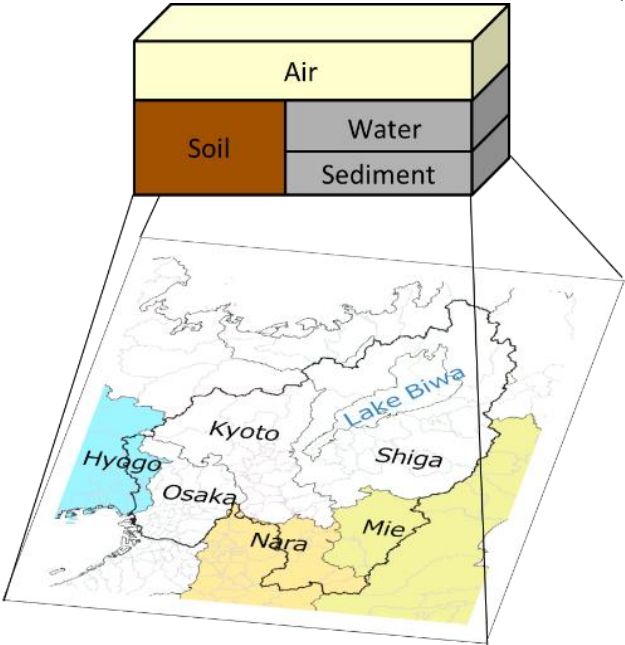
Figure 2. One-box multimedia model
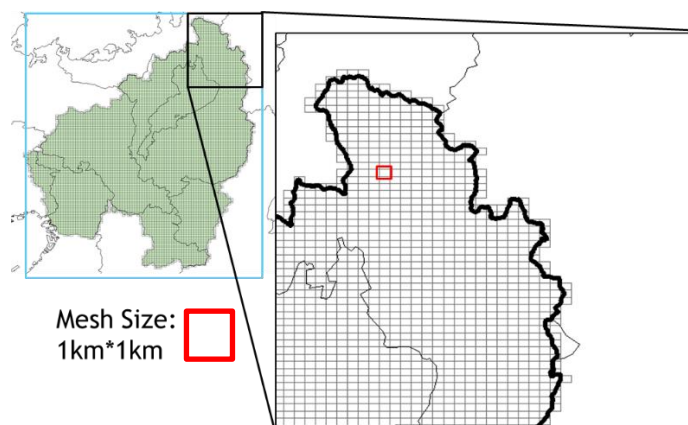
Figure 3. Distributional multimedia model

2.3. Distributional type model

In the OMM, the assumption of the uniform spatial distribution of chemicals leads to an uncertainty and an inaccuracy in simulated environmental fate. In order to interpret the spatial distribution of different chemicals in different environmental media, the DMM was developed as a mathematical tool. In the DMM, the LBYRB was divided into a grid with the size of 1 km (figure 3) and each of these grids was assumed as a three-dimensional box whose structure is the same as the one in OMM and the fluxes and mass transportation inside the compartment are affected by the 4 adjacent grids (if exist) in four directions (up down left right). The value of the inflow and outflow in each compartment which interact with the adjacent compartments were calculated dynamically using the meteorology and hydrology data.

3. Emission estimate

3.1. Target chemicals

This study focused on two kinds of sulfa-drugs, sulfamethoxazole (SMX) and sulfadimethoxine (Albon) which are widely used in Japan as an antibacterial agent for livestock production since they are synthetic drugs which show persistent degradability in nature compared to drugs made of natural extracts. According to statistical data by the Ministry of Agriculture, Forestry and Fisheries (MAFF), SMX is used in a larger amount than any other sulfur antimicrobial agents [3]

3.2. National usage

Japan began statistics on veterinary drug sales since 2005 based on the promulgation of the MAFF Ordinance No. 62 Animal Medicines Regulation Rules. In this research, the amount of 2 chemicals that were released into the environment was estimated based on the national sales amount of pure active substance sulfa-drug (an example shown in table 1) from 2005-2015 [3].

Table 1. Sales amount (active substance) and proportion by animal in 2005

Substance	Amount of pure active substance (kg)	Beef Cattle (%)	Dairy Cow (%)	Pig (%)	Broiler (%)	Layer (%)
SMX	60,223.7	0.0	0.0	79.7	16.1	4.2
Albon	18,092.8	17.0	17.8	40.9	9.8	7.2

3.2.2 LBYRB usage

A sulfa-drug administered to domestic animals is eventually discharged outside their body as excrement after organ metabolism. In this study, it was assumed that all sulfa-drugs administered were finally excreted into the environment, therewas no residual in livestock's organ and the usage of sulfa-drugs

and excrement volume per livestock were uniform across the whole country. Based on the ratio of 6 prefectures (Osaka, Kyoto, Hyogo, Shiga, Mie, Nara) included in LBYRB and national livestock excreta (table 2) [4-7], the sulfa-drug emission in the LBYRB, E_{lcoal} were calculated by using the equation (1).

$$E_{local} = \sum_{i=0}^{n} E_{i,all} \cdot \frac{S_{i,local}}{S_{i,all}} \qquad (1)$$

where i is the type of animal, E_{all} is national sulfa-drug emission, S_{all} and S_{local} are livestock excreta in the nationwide and LBYRB, respectively. The annual emissions in different prefectures are shown in figure 4.

Table 2. Excrement amount in different place (unit: 1000 ton)

|	Cattle	Cow	Pig	Hens
National	23570	24420	22380	7450
Mie	123	244	278	260
Shiga	56	168	12	18
Kyoto	108	64	26	85
Osaka	27	6	15	4
Hyogo	251	468	50	235
Nara	73	29	15	22
Local	638	979	396	624
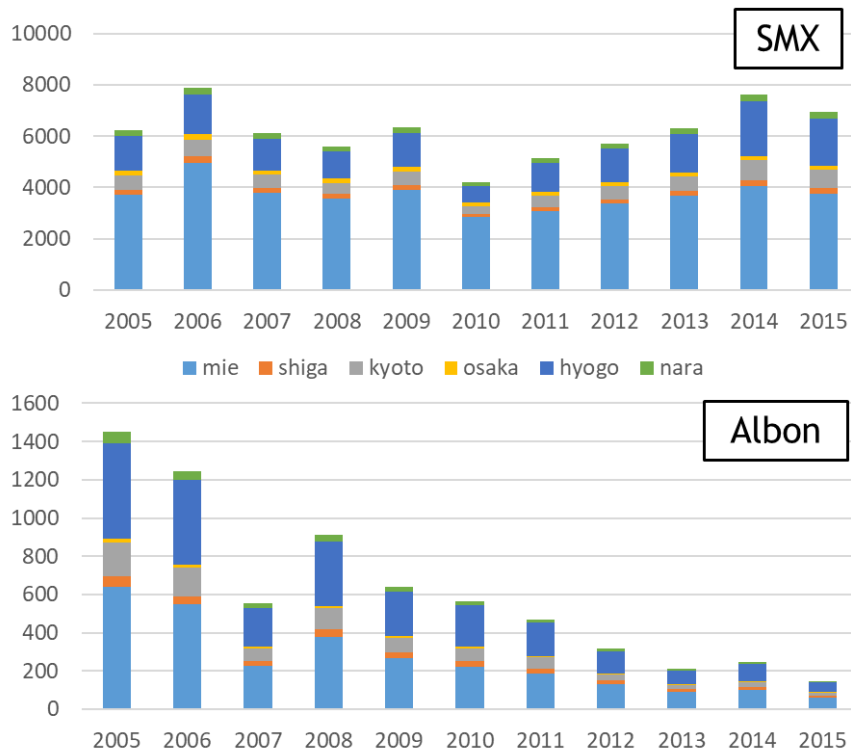
Figure 4. Estimated annual usage of sulfa-drug in 6 prefectures (unit: mol)

The spatial distribution of sulfa-drug emission in the LBYRB was estimated by using the formula (2), based on the farmland promotion according to the land use fragmented mesh data from the National Land Numerical Information System [8].

$$E_{mesh} = E_{all} \cdot P_{mesh} \qquad (2)$$

The spatial distribution (figure 5) shows that most emissions of the sulfa-drugs were concentrated in the east and south of the LBYRB. In terms of the temporal trend, the Albon was decreasing while SMX was basically the same.
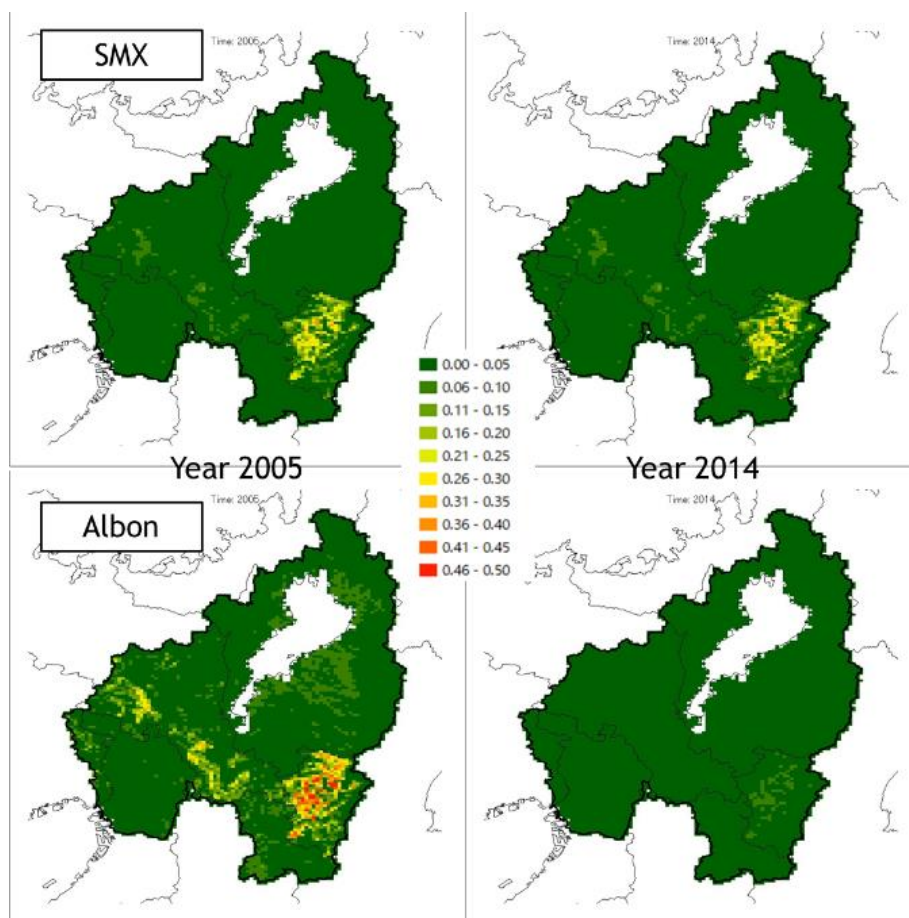
Figure 5. Distribution of estimated sulfa-drug emission in LBYRB (unit: mol)

4. Results

4.1. One-box type model

The results of the OMM show that the environment residual is only 7% of this year's emission and most of them, about 17% was absorbed in soil, 81% of them were dissolved in water. Meanwhile, considering the volume of the environment media, the annual average concentration of chemicals (figure 8) was around 18.5 ng/L in water, 9.1 ng/L in soil, 4.3 ng/L in sediment, which was a quite low-level pollution.
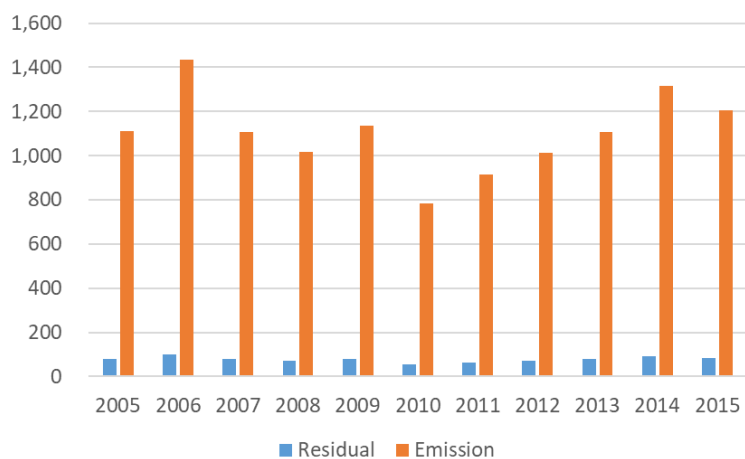
**Figure 6.** Amount of SMX emission and residual in environment (unit: mol)
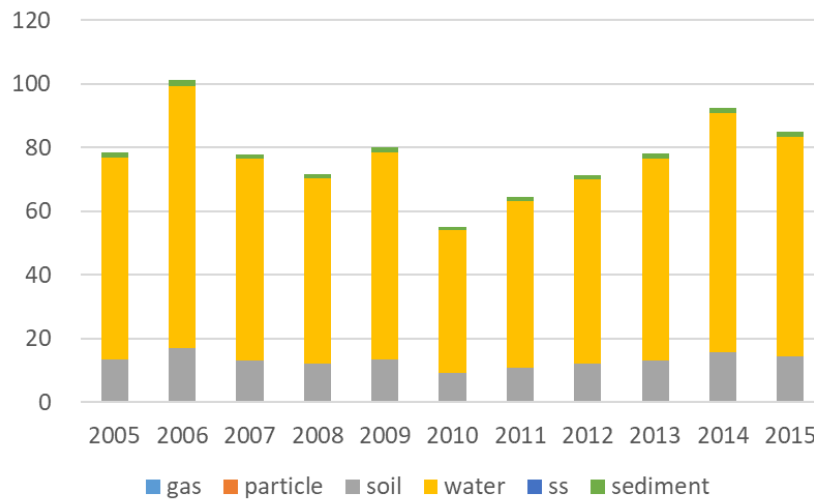
**Figure 7.** SMX residual in different environment media (unit: mol)
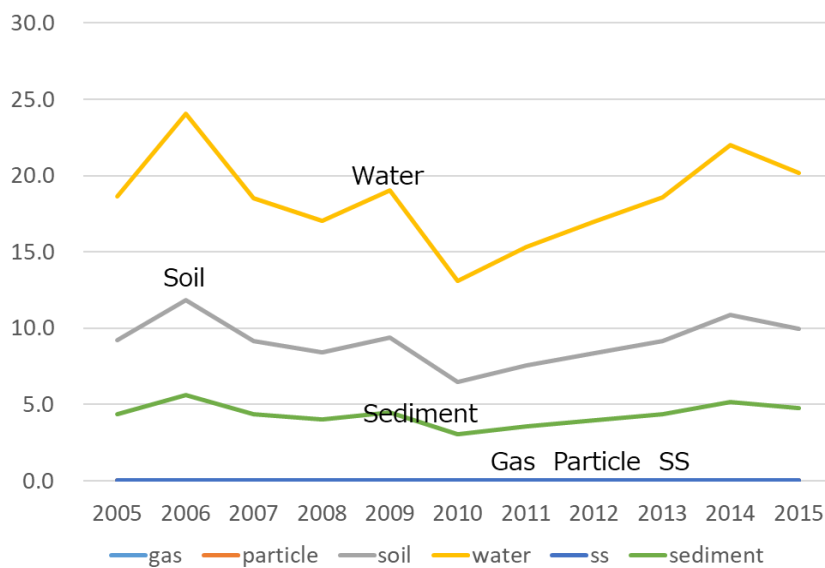
Figure 8. Concentration of SMX in different environment media (unit: ng/L)

4.2 Distributional type model

The spatial distribution of annual average concentration in soil media (figure 9) shows that the sulfa-drug was mainly concentrated in same place where the emission was concentrated as well, with the maximum SMX concentration of 600 ng/L and the maximum Albon concentration of 150 ng/L. Meanwhile, the distribution of annual average concentration in water media (figure 10) is different from that in soil. Although the emission was all assumed to be discharged into the soil, because of the fluxes between water and soil such as rainfall land loss, some chemicals move to the water media. Some mesh with large rivers had a lower level of the sulfa-drug concentration because the water flow was large, bringing a lot of clean water which dilute the chemicals. The annual concentration of Albon was decreasing while SMX was basically the same both in soil and water media that were consistent with the results of emissions estimation.
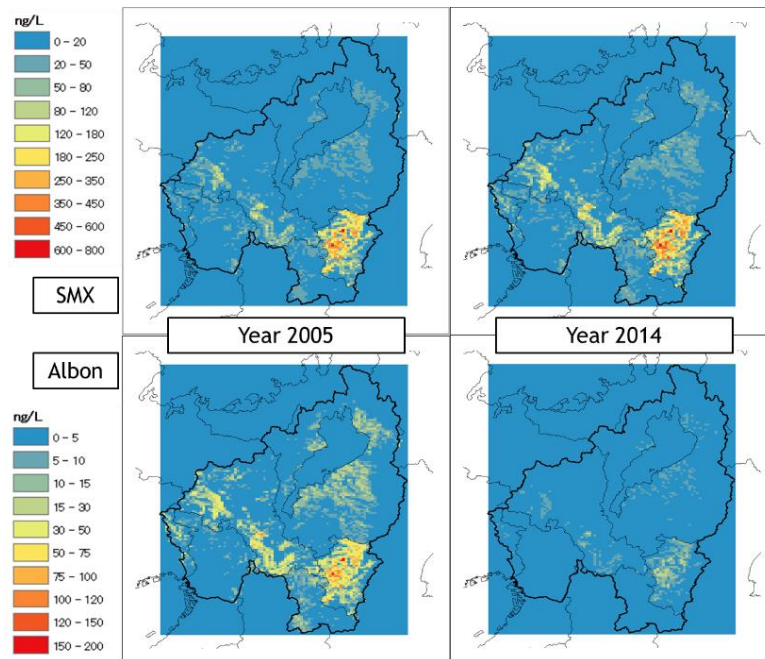
Figure 9. Distribution of sulfa-drag concentration in soil in LBYRB
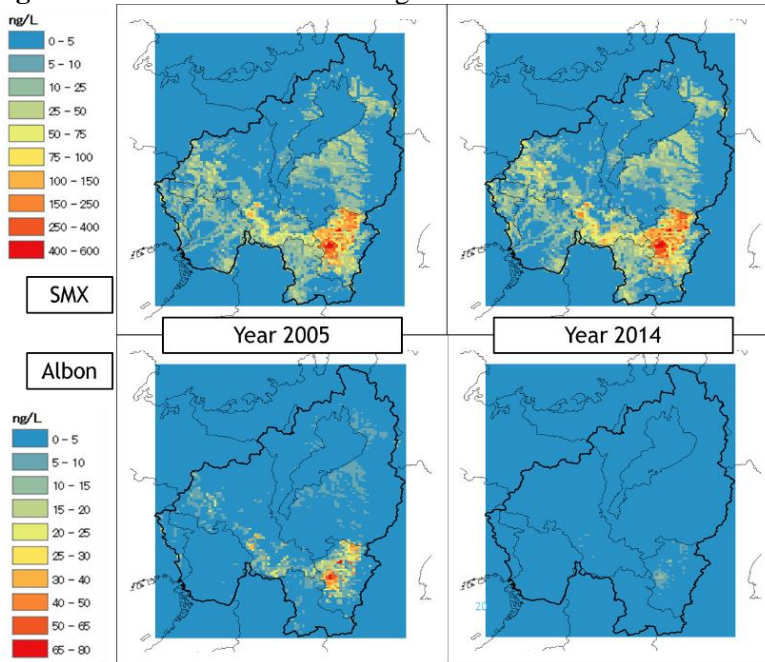
Figure 10. Distribution of sulfa-drag concentration in water in LBYRB

5. Conclusion

Two types of multimedia fate models (OMM and DMM) were successfully utilized in order to analysis the environmental behaviour of two kinds of sulfa-drugs (SMX and Albon) which are widely used in agriculture and husbandry. According to the result of the OMM simulation, around 93% of the sulfa-drugs emission in the Lake Biwa and Yodo River Basin (LBYRB) were quickly flowed out from the LBYRB to the ocean. Most of the residuals were dissolved in water, while the minority were absorbed in the soil or deposited in the sediment media. In the DMM simulation, according to the spatial distribution, most emission were occurred in the area where the farmland proportion are large. In both the simulations, the environmental concentration was correlated to the annual emission from the government statistics. Limitations of the current work that need to be solved include 1. Evaluating the

model with the observation data in the area; 2. Improving the accuracy of fundamental data which were used during the simulation; 3. Reducing the error caused by assumptions and simplifications in methodology. The above limitations require further studies in order to evaluate the potential risk of long-term exposure of such a low level concentration of these sulfa-drugs and judge if that can affect human health and leads to security problem. People need to be alert to the adverse environmental impacts of animal-use drugs discharged into the environment.

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