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Heavy Metal Loads Accompanying Rainfall Runoff on Low Farmland around Ariake Bay

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Abstract

Discharges of zinc, arsenic, cadmium, and lead from farmland were examined based on field measurements conducted between June 15 and July 13, 2006. The study fields are located in a lowland area where the irrigation water was contaminated with water drained from upstream farmlands (cyclic irrigation). The area of the farmland is 11.2 ha, of which 7.6 ha and 3.6 ha had been used for rice and soybeans, respectively. Heavy metal loads accompanying the rainfall runoff were considered to become higher with the increase in rainfall. Seventy percent of the zinc load, 62% of the arsenic load, 70% of the cadmium load, and 69% of the lead load were discharged with the rainfall runoff even though the rainfall runoff time was only 21% of the observation period.

Discipline: Agricultural environment
Additional key words: creek canal, cyclic irrigation

Introduction

Low farmlands in the coastal area around Ariake Bay in Japan have creek canals. The farmlands through which the creek canals pass use the creek water as irrigation water, and discharge the drainage water back into the creek canal. Therefore, farmlands on the downstream side of the creek canal use water that contains the drainage water from the farmlands upstream (cyclic irrigation).

Cyclic irrigation involves the risk of increasing creek canal nutrient concentrations[9]. Total nitrogen (T-N) and total phosphorus (T-P) concentrations in the creek water around Ariake Bay tended to be higher than the environmental quality standard for water pollution (for lakes; agricultural use; T-N: 1 mg•L^{-1} or less, T-P: 0.1 mg•L^{-1} or less) and environmental standard for irrigation water quality (T-N: 1.0 mg•L^{-1} or less)[1,8,10].

In recent years, environmental problems such as heavy metal pollution have become important issues. Some heavy metals are necessary to support life, but other heavy metals form toxic chemical compounds and affect the ecosystem[2]. Since heavy metals tend to be discharged from farmlands where there is a mixture of paddy fields and upland fields even on days when there is no rainfall during the inundation period[4], cyclic irrigation involves the risk of increasing heavy metal concentrations. Excess heavy metal loads, one of the causes of the increase in heavy metal concentrations in the irrigation water, have become a serious problem. Especially, the drainage water from the farmland is considered to increase with rainfall. To our knowledge, the dynamics of heavy metal runoffs from farmland on rainy days has not yet been reported on.

Zinc (Zn), arsenic (As), cadmium (Cd), and lead (Pb) are listed in the environmental quality standard for water pollution in Japan and/or environmental standard for irrigation water quality in Japan (Table 1). These heavy metals have not only drawn attention in terms of their listing as part of the environmental standards, but it has also been pointed out that there is some level of toxicity with regard to plants. Cadmium has an inhibiting effect on the biosynthesis of chlorophyll[3] and there are concerns as to its negative effect on the growth of plants. Nitrate reduc-

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tase plays a key role in nitrogen assimilation by the body of the plant[6]. Since lead inhibits the supply of metabolic products that act as electron accepters, it tends to inhibit the activity of nitrate reductase[5]. It has been reported that concentrations of arsenic in the groundwater around the study field are high[7]. In this respect, it should be noted that there is a possibility that arsenic concentrations in the surface water are also high.

In this study, discharges of zinc, arsenic, cadmium, and lead from farmland were examined using field data for the summer of 2006. In particular, the study focused on the effects of rainfall runoff on the discharge of heavy metals from the farmland.

Methods

1. Observed Farmland

The farmland that was studied is located in a lowland coastal area of Ariake Bay, Kyushu Island, Japan (33°80'N, 130°26'E) where irrigation water partly contains the water drained from upstream farmlands (Fig. 1). A strip of farmland with a drainage canal flowing in a T-shaped pattern was selected for the observations. In the drainage canal in this farmland it is comparatively easy to observe the discharge at both the upstream and the downstream ends. The drainage canal has gates at the eastern and northern ends (upstream ends). The western end (downstream end) of the canal has no gate between the canal and the creek. The gate on the eastern end of the drainage canal is always closed, but slight leakage from the creek into the canal was sometimes observed when the water level in the creek was extremely high. The overflow water was usually observed at the overflow gate at the northern end of the canal.

Sta. A-1, Sta. A-2 and Sta. A-3 shown in Fig. 1c are the water sampling and discharge measurement points in

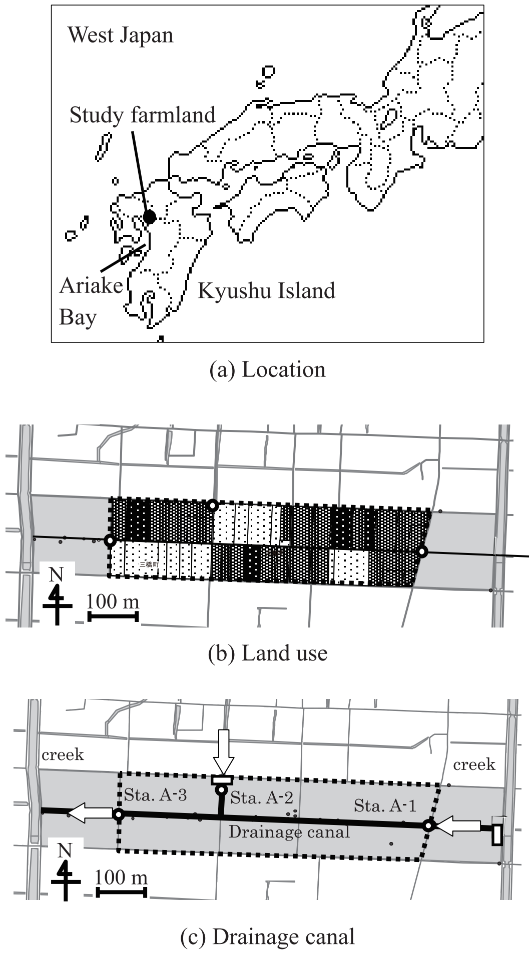

Fig. 1. Location and overview of the fields in the study
The area enclosed by the bold broken line is the farmland area of the study.
The arrows show the direction of the flow.
: Paddy fields (early ordinary rice, 62,000 m^2)
: Paddy fields (sticky rice, 14,000 m^2)
: Upland fields (soybean, 36,000 m^2)
: Sampling point and triangular measurement weir setting point (Stations).
: Gate.

Table 1. Heavy metal concentrations listed in water quality standards

Heavy Metals		Environmental quality standards for water pollution			Water quality standards for irrigation water (paddy field)
		Environmental quality standards for human health	Environmental quality standards for conservation of the living environment		
			Rivers and Lakes	Coastal water	
		(mg/L)	(mg/L)	(mg/L)	(mg/L)
Zinc	(Zn)		< 0.03	<0.01 or < 0.02*	< 0.5
Arsenic	(As)	< 0.01			< 0.05
Cadmium	(Cd)	< 0.01			
Lead	(Pb)	< 0.01			

*: Dependent on biological environment.

the drainage canal. Most of the inflow into the drainage canal was the flow through Sta. A-1 from the eastern side, the flow through Sta. A-2 from the northern side and drainage from the farmland.

Underdrains are installed to rice paddy fields and soybean upland fields. All underdrains of both rice paddy fields and soybean upland fields were closed during the observation period. Thus, the drainage from the farmlands to the drainage canal was considered to consist of the surface discharge and the horizontal infiltration.

However, surface discharges from the surrounding paved roads were observed and these flowed into the drainage canal with rainfall events, although the influent surface discharge was very low (around 2%) compared to the drainage from the farmland.

The area of the farmland is around 112,000 m^2 (170 m × 660 m), of which 76,000 m^2 and 36,000 m^2 are used as rice paddy fields and for soybean cultivation, respectively (Fig. 1). Yume-Tsukushi rice (early ordinary rice) and Hiyoku-Mochi rice (sticky rice) were planted in 62,000 m^2 and 14,000 m^2 of rice paddy fields, respectively. Since the upland soybean field was in rotation from prior use as a rice paddy field, the upland soybean field has a ridge around the field. The ridge around the upland soybean field blocked the surface drainage.

There was 26,000 m^2 of the farmland used as rice paddy fields and soybean upland fields on both sides of the drainage canal from the upstream end to Sta. A-1. The farmer village was located about 200 m upstream from Sta. A-2. The drainage water from more than 100,000 m^2 of the farmland flowed to the drainage canal between the farmer village and Sta. A-2.

2. Observation Periods and Intervals

Discharges were observed and water samples were collected from June 15 to July 13 (28 days) in 2006. Paddling and the transplanting were carried out in 14,000 m^2 of the paddy fields where Hiyoku-Mochi rice was being cultivated from June 16 to June 24. Other paddy fields had been planted with the Yume-Tsukushi rice before the observations were made. Fuku-Yutaka soybean was planted in the upland soybean field.

The sample water was collected at 3 points (Sta. A-1, Sta. A-2 and Sta. A-3) along the drainage canal every 6 hours. If rainfall was observed, the sample water was collected every hour from the beginning of the rainfall event to 5 hours after the end of the rainfall. The collected samples were kept in cold storage and then taken to the laboratory.

3. Flow Rate Observations

The discharge was observed at the upstream ends and the downstream end of the drainage canal (Sta. A-1, Sta. A-2 and Sta. A-3). The stream regime of each station was stable with enough discharge to measure flow rate using the triangular measurement weir at each station. Each station was suitable for the installation of the triangular measurement weirs. However, there was no discharge from the creek to the drainage canal through the closed eastern gate except at times of extremely high water levels in the creek, most of the time there was enough discharge to measure flow rate using the triangular measurement weir at Sta. A-1. This discharge maybe related to the water discharge from the farmland located between the canal and Sta. A-1.

The discharge was basically observed using the triangular measurement weirs. When the discharge deviated from the conditional tolerance of the triangular measurement weirs, the discharge was calculated from the discharge velocity and the cross-sectional area as measured in the stable flow regime site on the upstream side of the triangular measurement weirs. The discharge velocities and water levels were measured using a 2-dimensional electromagnetic flow meter (Kenek Corp., VP2000) and an automatic water level meter with a pressure gauge (STS Sensor Technik Sirnach AG, MC1100W), respectively.

4. Definition of rainfall runoff event

When the effect of rainfall on the load from the farmland is discussed, the definition of a rainfall runoff event is very important. The duration of the hydrographic peak of the rainfall runoff at Sta. A-3 was always longer than that for Sta. A-1 and Sta. A-2 in these observation periods. A single rainfall runoff event is defined as the period from the beginning of the increase to the end of the hydrographic peak in the discharge at Sta. A-3.

Figure 2 shows the calculation method for the discharge during a single rainfall event. The discharge

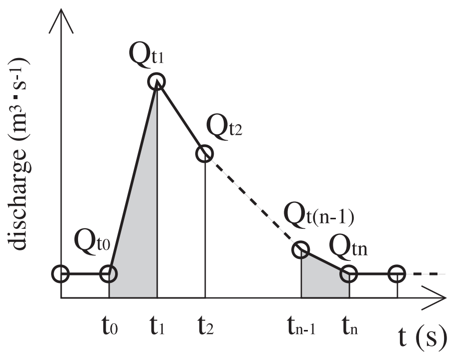

Fig. 2. Calculation method for the discharge during the rainfall events

through each station was calculated as the summation of the area shown in Fig. 2 from the beginning to the end of the rainfall runoff. The discharge between sampling times was linearly-interpolated. For example, the total discharge through Sta. A-1 was calculated as follows in equation 1:

$$D_{A-1} = \sum_{i=1}^{n} \frac{\{Q_{ti} + Q_{t(i-1)}\} \cdot (t_i - t_{i-1})}{2} \qquad (1)$$

where $D_{A\text{-}1}$ is the total discharge through Sta. A-1 during the single rainfall event [m^3], t_i is sampling time shown in Fig. 2 [s], and Q_{ti} is the discharge through Sta. A-1 in sampling on t_i shown in Fig. 2 [$m^3 \bullet s^{-1}$]. The drainage water from farmland was calculated as follows in equation 2:

$$D_{FL} = \frac{D_{A-3} - D_{A-2} - D_{A-1}}{1{,}000A} \qquad (2)$$

where D_{FL} is the drainage from farmland [mm], and A is the area of studied farmland [m^2].

The load was calculated as follows in equations similar to the calculation of discharge:

$$L_{A-1} = \sum_{i=1}^{n} \frac{\{Q_{ti}C_{ti} + Q_{t(i-1)}C_{t(i-1)}\} \cdot (t_i - t_{i-1})}{2} \qquad (3)$$

$$L_{FL} = \frac{L_{A-3} - L_{A-2} - L_{A-1}}{A} \qquad (4)$$

where $L_{A\text{-}1}$ is the total load through Sta. A-1 during the single rainfall event [mg], C_{ti} is the heavy metal concentration at Sta. A-1 on t_i [$mg \bullet m^{-3}$, $\mu m \bullet L^{-1}$] and L_{FL} is the heavy metal load from farmland [$mg \bullet m^{-2}$].

5. Heavy Metal Concentration Analysis

The concentrations of zinc, arsenic, cadmium, and lead were analyzed using inductively coupled plasma mass spectrometry (ICP-MS; Parkin Elmer, Inc., ELAN DRC-II) after decomposition. The total heavy metal concentration was estimated from the target isotope concentration and the content in natural conditions. The target isotope of each heavy metal element (^{52}Zn, ^{75}As, ^{114}Cd, and ^{207}Pb) was selected to ensure optimum sensitivity. Decomposition was carried out using a microwave oven (Milestone, ETHOS TC) employing the Easy Control Program No. B0031 (Milestone General). Detection limits of heavy metals by ICP-MS are; zinc: 0.075 $\mu g \bullet L^{-1}$, arsenic: 0.006 $\mu g \bullet L^{-1}$, cadmium: 0.001 $\mu g \bullet L^{-1}$, and lead: 0.004 $\mu g \bullet L^{-1}$, respectively. Residual standard deviations of heavy metal measurement by ICP-MS are; zinc: < 5.0, arsenic: < 10, cadmium: < 10, and lead: < 5.0, respectively.

Results and discussion

1. Discharge from the farmland

Figures 3a–3b show the precipitation and the observed discharge at each station in the drainage canal. The rainfall data represents the observed data in the Automated Meteorological Data Acquisition System in Yanagawa City (AMeDAS Yanagawa; 33°16'N, 130°40'E), which is the nearest weather station from the area of farmland studied. During the observation period 418 mm of rainfall was observed.

The fluctuations of the discharge at Sta. A-1, Sta. A-2 and Sta. A-3 are shown in Fig. 3b. The inflow from Sta. A-1 was small compared to Sta. A-2 and Sta. A-3. Without rainfall or a large water inflow from the upstream end, discharges in the drainage canal seem small. If the inflow from Sta. A-2 was large, the discharge at Sta. A-3 tended to also become large. While the rainfall runoff pattern depends on the properties of the rainfall, if the precipitation was above a certain level this caused obvious rainfall runoff.

Except in rainy periods, the discharge in the drainage canal was very small during the inundation period, and tended to be larger in the morning during the paddling and/or transplanting periods due to the large inflow from the upstream end. The average discharges in the periods without rainfall were 0.012 $m^3 \bullet s^{-1}$ at Sta. A-1, 0.024 $m^3 \bullet s^{-1}$ at Sta. A-2 and 0.042 $m^3 \bullet s^{-1}$ at Sta. A-3.

Discharges in the drainage canal tended to increase significantly with the rainfall during the observation periods. The maximum discharges in the rainfall periods were 0.048 $m^3 \bullet s^{-1}$ at Sta. A-1, 0.195 $m^3 \bullet s^{-1}$ at Sta. A-2 and 0.796 $m^3 \bullet s^{-1}$ at Sta. A-3, and these were 4.0, 8.1 and 19.0 times greater than the average discharges in the rainless periods. The average discharges in the rainfall periods were 0.013 $m^3 \bullet s^{-1}$ at Sta. A-1, 0.061 $m^3 \bullet s^{-1}$ at Sta. A-2 and 0.120 $m^3 \bullet s^{-1}$ at Sta. A-3, and were 1.1, 2.5 and 2.9 times larger than the average discharge in the rainless periods.

2. Heavy metal concentrations in the drainage canal

Figures 3c–3f show the fluctuation of the heavy metal concentrations in the drainage canal. Zinc, arsenic, cadmium, and lead concentrations tended to be high during the rainfall periods. The increases in the concentrations tended to be observed without reference to the amount of rainfall. The surface discharges from farmlands affected by rainfall are considered to be affected by not only the rainfall and its intensity, but also other factors. For example, the surface discharge from the paddy field doesn't drain until the water level in the paddy field exceeds the height of the end plate at the outlet point even if

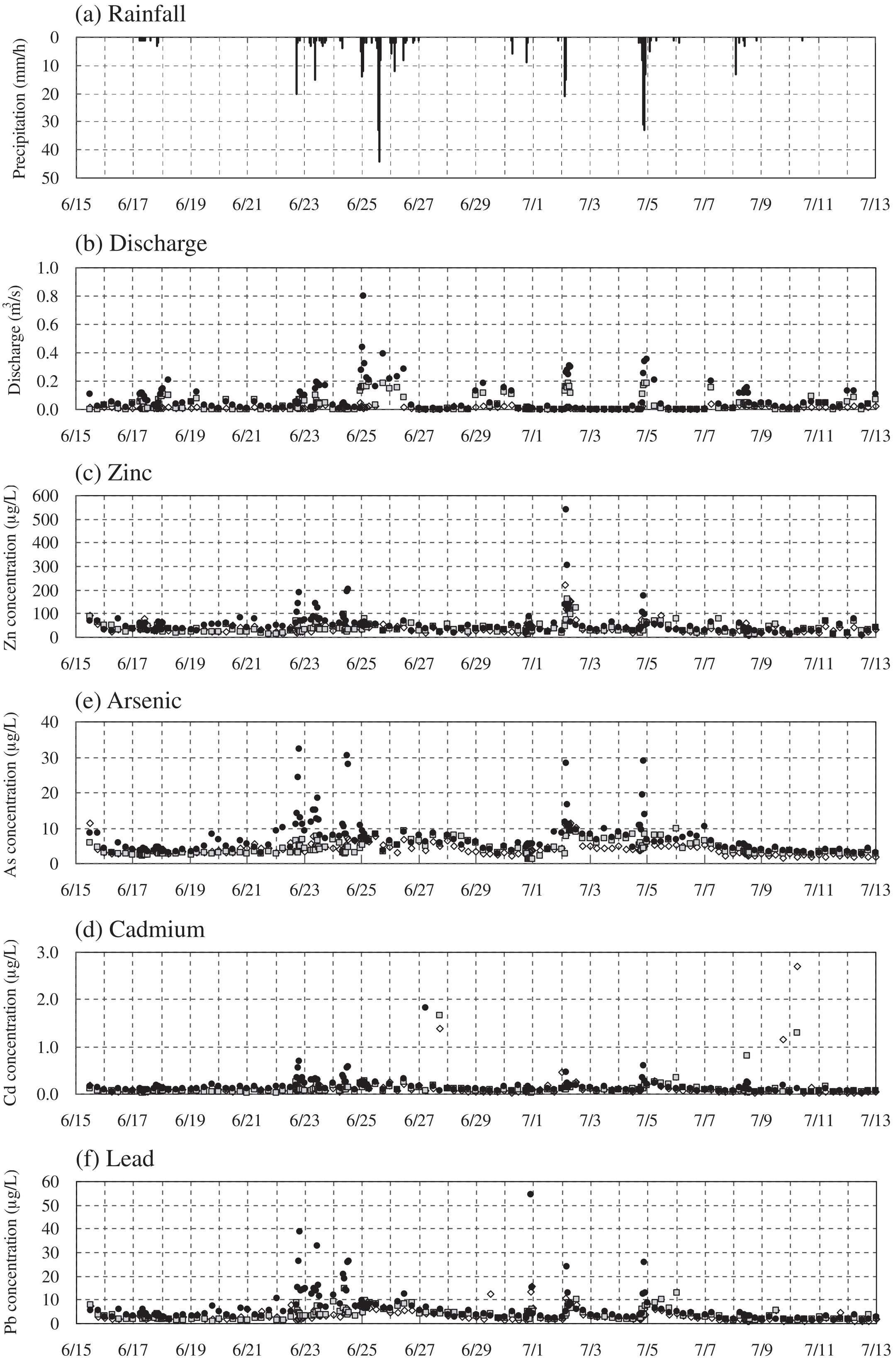

Fig. 3. Fluctuation of discharge and heavy metal concentrations in the drainage canal
◇: Sta. A-1, ▢: Sta. A-2, ●: Sta. A-3.

the water level rises by rain, and the surface discharge from the upland field doesn't drain until the rainfall intensity exceeds the intake rate at the surface of the soil. As typified by the rainfall event on June 25, however, when a large precipitation was observed, the heavy metal concentrations did not increase so much. The reasons for this phenomenon are not identified in this paper, but a dilution by rain water and a surface soil runoff from upland fields affected by rainfall intensity are considered to cause this phenomenon.

Zinc concentration is listed in the environmental standard for irrigation water quality in Japan; 0.5 mg•L^{-1} or less. The observed zinc concentrations were higher than the standard only once during rainfall events. The environmental standard for irrigation water quality in Japan lists the maximum concentrations. If plants are grown by hydroponic culture with higher concentrations than the maximum, there is some possibility that the plants will be damaged during growth. Therefore, it is not clear what the environmental standard for irrigation water quality in Japan applies to; the annual average, the daily average or the momentary value. Regardless of this, the observed zinc concentrations were not considered to be causing a problem since the observed zinc concentrations were sufficiently below the standard during the rainless periods and most of the observed zinc concentrations were lower than the environmental standard for irrigation water quality in Japan even during the rainfall periods. On the other hand, zinc concentration is listed in the environmental quality standards for conservation of living environment of ecosystem integrity (for rivers and lakes; 0.03 mg•L^{-1} or less). Zinc concentration in the creek water was comparatively higher than the standard of ecosystem integrity. It will be necessary to pay attention to it in the future.

Cadmium is not listed in the environmental standard for irrigation water quality in Japan but is listed in the environmental quality standard for water pollution in Japan (0.01 mg•L^{-1} or less). Cadmium concentration standards in foods are listed in food sanitation laws; for example, the cadmium concentration standard for rice is 0.04 ppm in milled rice in the Codex and 0.1 ppm in brown rice in the food sanitation law of Japan. Cadmium concentration in foods (plants) is considered to be affected by the planting environment including irrigation water. Thus, cadmium concentration in the surrounding water environment is very important. The observed cadmium concentrations were always lower than the standard even during rainfall events. Observed cadmium concentrations during the rainless periods were around the lower limit of the qualitative analysis.

Arsenic concentration is listed in the environmental standard for irrigation water quality in Japan; 0.05 mg•L^{-1} or lower. The observed arsenic concentrations were always lower than the standard even during rainfall events. However, arsenic concentrations in the groundwater around the study fields were high[7], although the observed arsenic concentrations were low and were not affected by the groundwater.

Lead is not listed in the environmental standard for irrigation water quality in Japan but is listed in the environmental quality standard for water pollution in Japan (0.01 mg•L^{-1} or less). The observed lead concentrations were lower than the standard during rainless periods, but sometimes higher than the standard during rainfall events. The environmental quality standard for water pollution applies to annual average concentrations. Therefore, the average over the long term should be used as a comparison with the standard. The average lead concentration during the observation period was 2.7 µg•L^{-1} and was below the standard.

3. Drainage water from the farmland during rainfall runoff events

Many rainfall events were observed during the observation period. Minor rainfall events that resulted in no rainfall runoff were ignored. The data collected during the rainfall periods on June 25–26 and July 5 is excluded since the discharge in the Sta. A-1 was too large to measure due to the heavy rain and the simultaneous increase in the water depth in the drainage canal. On June 30, since the background discharge was too high and the rainfall runoff was smaller than the background level, the rainfall runoff could not be evaluated. As a result, data on eight rainfall runoff events was collected during the observation period (Fig. 4a).

Figure 4b shows the discharges for the rainfall runoff events. The two graphs on the left side of Fig. 4 show the data collected during the paddling and/or transplanting periods, and the two graphs on the right side of Fig. 4 show the data collected during the inundation periods. Figures 4a–4b show that the discharges increased with the rainfall. With regard to rainfall events 1–5 and 7, excluding rainfall events 6 and 8, the inflows from Sta. A-2 increased and the outflows from Sta. A-3 increased in conjunction with the rainfall.

The pattern of the drainage pathway from the farmland is complex and consists of the surface drainage, the vertical infiltration drainage and the horizontal infiltration drainage. The in situ qualitative observation of each drainage route is too difficult. Thus, the drainage water from the farmland was estimated on the basis of the difference in the passage of the discharge between the flow at the upstream ends and that at the downstream end of

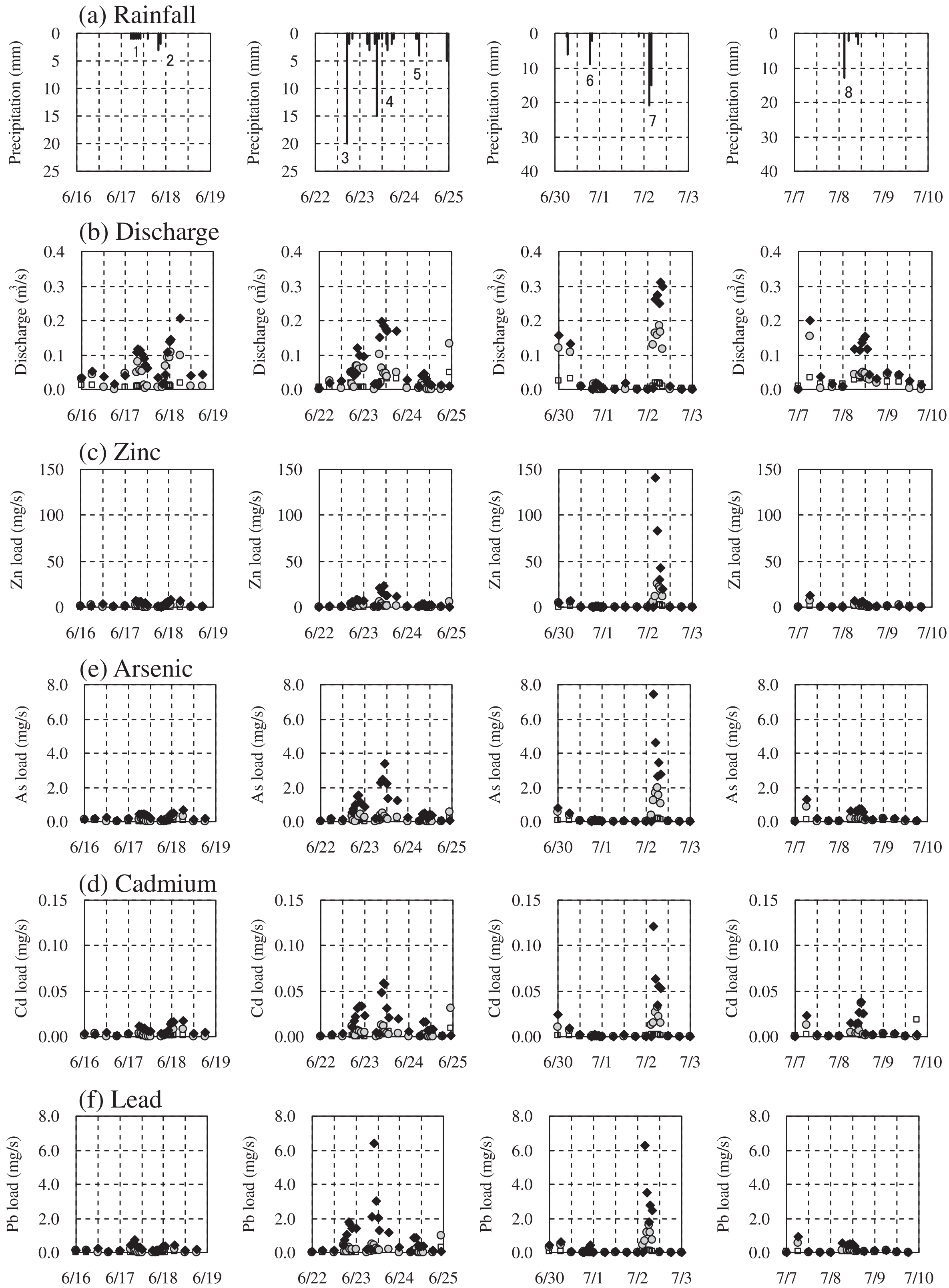

Fig. 4. Fluctuation of discharge and heavy metal loads in the drainage canal
□: Sta. A-1, ◯: Sta. A-2, ◆: Sta. A-3.

the drainage canal. This method makes it possible to estimate the total discharge from the farmland into the drainage canal. The discharge was calculated following equation 2.

Table 2 lists the rainfall, drainage water and runoff ratios on the left side. The rainfall values in Table 2 correspond to the eight numbered rainfall events shown in Fig. 4a. Table 2 indicates that the runoff ratio tended to be over 100% in the paddling and the transplanting periods, and the runoff ratios were less than 100% in the inundation periods. The condition of the end plate at the outlet point was considered to affect these phenomena. The surface discharge from the paddy field doesn't occur until the water level in the paddy field exceeds the height of the end plate at the outlet point even if the water level rises by rain, surface discharge from the paddy field did occur when the end plate at the outlet point was removed artificially. No distinct relationship between the rainfall, rainfall runoff and runoff ratios was found, but the rainfall runoff tended to be greater with an increase in rainfall.

4. Heavy metal loads from the farmland

Figures 4c–4f show the fluctuation of heavy metal loads at Sta. A-1, Sta. A-2 and Sta. A-3. The effect of rainfall on the heavy metal loads from the farmland was very substantial.

The increases in concentrations were affected not only by the discharged load from the farmland, but also by the inflows from Sta. A-1 and Sta. A-2. Therefore, the discharged heavy metal loads from the farmland were calculated as in equation 4.

The properties of the rainfall events and the discharged heavy metal loads from the farmland are listed in Table 2. Rainfall runoff accompanying the listed rainfall events was confirmed, except for rainfall event 6. In rainfall event 6, however, an increase in the discharge in the drainage canal was observed and the discharged load from the farmland was nearly 0. In other rainfall events, heavy metal loads with the rainfall runoff were considered to be higher with an increase in rainfall. A distinct relationship among rainfall, rainfall runoff and heavy metal loads accompanying the rainfall runoff was not found, because the conditions of paddy fields and the upland fields were different in each rainfall runoff event. The rainfall runoff and heavy metal loads accompanying the rainfall runoff were considered to be affected by rainfall in this study. Since the drainage from the farmland is affected by many factors, as described above, more observation data is required to clarify the relationship between rainfall and rainfall runoff.

The total heavy metal loads during this observation period of 600 h (except for June 25–26 and July 5) are; zinc: 23.0 mg•m^{-2}, arsenic: 2.59 mg•m^{-2}, cadmium: 47.3 μg•m^{-2}, and lead: 2.62 mg•m^{-2}, respectively. The total heavy metal loads during the 126 h of rainfall runoff events listed in Table 1 are; zinc: 16.2 mg•m^{-2}, arsenic: 1.60 mg•m^{-2}, cadmium: 33.2 μg•m^{-2}, and lead: 1.81 mg•m^{-2}, respectively. Therefore, 70% of the zinc load, 62% of the arsenic load, 70% of the cadmium load, and 69% of the lead load were discharged with the rainfall runoff even though the rainfall runoff time was only 21% of the observation period. In this study rainfall had a profound effect on heavy metal loads.

Conclusion

Discharges of the four heavy metals (zinc, arsenic, cadmium, and lead) from the low farmland to the drainage canal were examined based on field measurements conducted between June 15 and July 13, 2006. The following results were obtained in this study.

Table 2. Rainfall, drainage water, runoff ratios, and heavy metal loads during 8 rainfall events

Rain No. (Fig.4)	Rainfall (mm)	Drainage water (D_{FL}) (mm)	Runoff ratio (%)	Discharged load Zn (mg•m^{-2})	As (μg•m^{-2})	Cd (μg•m^{-2})	Pb (μg•m^{-2})
1	7	20	284	1.0	102	2.8	102
2	5	26	510	1.4	103	3.4	80
3	23	8	35	1.7	265	6.7	376
4	32	39	123	4.5	563	9.8	703
5	6	3	51	0.6	85	2.2	116
6	11	0.1	1	0.0	6	0.1	13
7	36	17	47	6.8	451	7.4	395
8	4	1	20	0.2	23	0.8	20

1) The concentrations of heavy metals in the drainage canal located in the area of low farmland met the environmental standards for water quality for human health and agricultural use.
2) Zinc concentration in the drainage canal was comparatively higher than the environmental quality standards for conservation of the living environment.
3) Heavy metal concentrations in the drainage canal were affected by rainfall.
4) Seventy percent of the zinc load, 62% of the arsenic load, 70% of the cadmium load, and 69% of the lead load were discharged with the rainfall runoff even though the rainfall runoff time was only 21% of the observation period.

A distinct relationship between rainfall, rainfall runoff and heavy metal loads accompanying the rainfall runoff was not found, but rainfall runoff and heavy metal loads were considered to be affected by rainfall in this study. Further observation is required to understand the mechanisms of the discharge of heavy metals from farmlands.

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