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https://doi.org/10.5109/2558883

出版情報：九州大学大学院農学研究院紀要. 65 (1), pp.9-14, 2020-02. Faculty of Agriculture, Kyushu University
バージョン：

J. Fac. Agr., Kyushu Univ., **65** (1), 9–14 (2020)

Contamination of the Summer and Winter Vegetables by Heavy Metals in a Multi–Industry District of Bangladesh

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(*Received October 10, 2019 and accepted November 14, 2019*)

Heavy metal (Cr, Cu, Zn, As, Cd, and Pb) contamination of the summer and winter vegetables were examined in a multi–industry district of Bangladesh. In this district, various kinds of industries discharged their wastewater into nearby irrigation canals, contaminating the vegetables that were cultivated by using the irrigation water with the heavy metals. Among the vegetables, the heavy metal concentrations were the highest in root vegetables, followed by leaf vegetables for both the summer and winter vegetables. Zn was the highest, while Cd was the lowest in concentrations throughout the vegetables. Every heavy metal concentration was lower in the summer than in the winter vegetables. The reason is probably that the concentrations of irrigation water and soil were diluted by rainfall during the rainy season when the summer vegetables were grown. The health risk index, which enables to assess the potential health risk due to the ingestion of vegetables, showed a high value in root and leaf vegetables, which indicated that the root and leaf vegetables grown in the district were found unsuitable for human ingestion.

Key words: Industrial wastewater, vegetables, contamination, season, health risk index

INTRODUCTION

Contamination of agricultural soil with heavy metals is a serious environmental concern in developing countries due to its acute and persistent toxicity (Ağca and Özdel, 2014). In recent decades, the anthropogenic activities like industrialization and urbanization have led to an increase in heavy metal and metalloid concentrations in water, soil and vegetables (Islam *et al.*, 2017). In Bangladesh, food safety is a prime issue for securing health, where various crops are assumed to be contaminated with carcinogenic (Pb, As, and Cd) and non–carcinogenic (Fe, Co, V, Cu, Zn, Cr, Mn, and Ni) heavy metals and are unsafe for human ingestion (Sultana *et al.*, 2017). Unplanned industrialization and urbanization in the country have led to discharge the heavy metal contaminated wastewater into water bodies and soil, and as a result, vegetables grown there became contaminated with heavy metals, and the vegetables were consumed by many people by transportation and retailing (Ikeda *et al.*, 2000). The contaminated crop products have caused a health concern due to its severe harmful impacts (Sultana *et al.*, 2014). When vegetables are produced in a contaminated soil, heavy metals can be stored in the edible parts of the vegetables that causes toxicity in the vegetables (Sharma *et al.*, 2009). Heavy metals are non–biodegradable in nature and once heavy metals are accumulated in organs of human body, it causes severe disorders in the organs (Duruibe *et al.*, 2007). According to the World Bank report, about one million populations are at a health risk in Bangladesh due to toxicity of heavy metal especially by lead (The Daily Star, 2018).

The farmland of Gazipur District, Bangladesh was selected for the present study, where various industries, including garment, textile, dyeing, ceramic, pharmaceutical, paint, and packaging industries, discharge their wastewater into nearby waterbodies without any treatment and which cause heavy metal contamination in the surrounding environment. Farmers use the heavy metal contaminated irrigation water for growing vegetables, which is the major cause of heavy metal contamination in vegetables in the district. Most of the produced vegetables are marketed in the capital Dhaka and are consumed by many people. The vegetables may be contaminated severely with heavy metals (Cr, Cu, Zn, Cd, As, Hg, Pd, Fe, and Ni) according to the earlier studies (Barakat, 2011; Sultana *et al.*, 2011). Here, the risk of intake of contaminated vegetables on human health was examined. Therefore, this study aimed to clarify the contamination levels of Cr, Cu, Zn, As, Cd and Pb for the summer and winter vegetables. Further, the potential health risk due to vegetables consumption was assessed by considering health risk index (HRI) of heavy metals. In this district, different types of the summer and winter vegetables are cultivated in the rainy and dry seasons, respectively.

MATERIALS AND METHODS

Study area

The study area of Gazipur District is a suburban industrial area located about 50 km away north from the capital Dhaka. The district has an area of 1,806 km^2 with a population of 3.4 million (BBS, 2011). The district has a flat lowland topography with an elevation of 4–24 m (Shapla *et al.*, 2015). The soils are acid basin clays with nutrient–poor characteristics, i.e., deficient in organic matter, phosphate, nitrogen and lime (UNDP/FAO, 1988). The annual average rainfall is 2,036 mm (Merkel, 2012), the rainy season is from April to October, and dry

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season from November to March. The annual average air temperature is 25.8°C (Merkel, 2012).

The target areas in the district are Banglabazar, Kashimpur, and Chandra, where various kinds of small–scale factories are located (Fig. 1). The main industries are different from area to area, i.e., in Banglabazar, textile, dye, battery, metallurgical and ceramics industries are located; in Kashimpur, plastic, garments and agrochemical industries, and in Chandra, pharmaceutical, agrochemical, fabric printing, poultry feed, and fish feed industries are located, respectively (Ahmed *et al.*, 2018; Ahmed *et al.*, 2019). In the respective areas, industrial wastewater is discharged from the factories into nearby irrigation canals throughout the year.

Sampling of vegetables

The sampling locations were shown in Fig. 1. The sampling location was the same in summer and winter vegetables. For the summer vegetables, taro (*Colocasia esculenta*), water spinach (*Ipomoea reptans*), helencha (*Enhydra fluctuans*), eggplant (*Solanum melongena*), and sponge gourd (*Luffa acutangula*) were sampled in June–July, 2015. While for the winter vegetables, radish (*Raphanus raphanistrum* subsp. *sativus*), amaranth (*Amaranthus lividus*), red amaranth (*Amaranthus gangeticus*), spinach (*Spinacia oleracea*), indian spinach (*Basella alba*), pumpkin (*Cucurbita moschata*), bottle gourd (*Lagenaria siceraria*), taro (*Colocasia esculenta*), and yard long bean (*Vigna sesquipedalis*) were sampled in November–December, 2017. Then the samples were cleaned with tap water followed by deionized water. The samples were then divided into root, stem, fruit and leaf parts and these parts were first air dried and then oven dried at 60°C for 48 hours and were milled uniformly with a grinder.

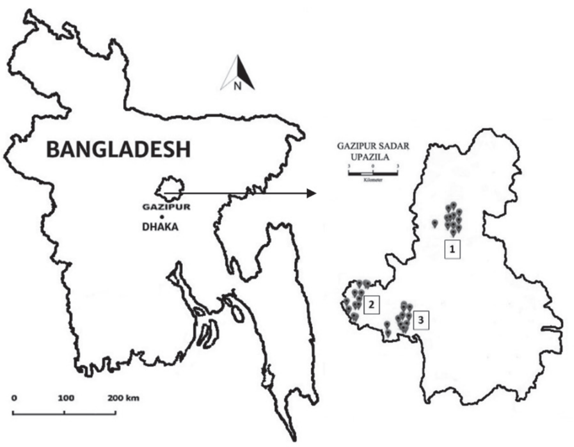

Fig. 1. Sampling locations of the summer and winter vegetables in Gazipur District, Bangladesh. 1. Banglabazar; 2. Chandra, 3. Kashimpur.

Sample analysis

For the digestion of vegetable samples, the UWLAB method (UWLAB, 2005) was applied. Oven dried 0.5 g of the samples was taken into 50 ml test tube. Five ml of concentrated nitric acid (65% HNO_3) was added to the samples and soaked at room temperature for 2–3 hours. Then the test tube was placed in the block heater and heated at 130°C for 14–16 hours. The mixture solution was heated again by adding 1 ml of 30% hydrogen peroxide (H_2O_2) for 20–30 minutes. Finally, the mixture was cooled, filtered and diluted with double deionized water to 50 ml. Heavy metal (Cr, Cu, Zn, As, Cd, and Pb) concentrations of digested solutions of vegetables were determined by inductively coupled plasma–mass spectrometry (ICP–MS; 7500 ce, Agilent Technologies, USA) at Kyushu University.

Daily Intake of Heavy Metals (DIM)

The daily intake of heavy metals through vegetables was estimated with the following equation.

$$\text{DIM}= \frac{\text{C metal} \times \text{C factor} \times \text{D food intake}}{\text{B average weight}}$$

Where C_{metal}, C_{factor}, $D_{food\ intake}$ and $B_{average\ weight}$ represent the heavy metal concentrations in the vegetables (mg/kg), conversion factor, daily intake of vegetables (Kg) and average body weight (Kg), respectively. According to Rattan *et al.* (2005), the fresh to dry weight conversion factor of green vegetables is 0.085. The daily intake of vegetables for adult persons was 0.1673 kg (BBS, 2016). The average body weight of adult person in Bangladesh was 66.5 kg (Khadem and Islam, 2014). These values were applied in the estimation.

Health Risk Index (HRI)

The health risk index is the quotient between daily intake of heavy metals (DIM) and their reference dose (R_fD), which is calculated as follows (Pierzynski *et al.*, 2000).

$$\text{HRI}= \frac{DIM}{RfD}$$

Where R_fD represents the oral reference doses, which are 0.003, 0.04, 0.3, 0.0003, 0.0005, and 0.004 mg/kg for Cr, Cu, Zn, As, Cd, and Pb, respectively (USEPA, 2007). HRI> 1 means that the exposed population is assumed to be at a potential health risk.

Data Analysis

The graphical presentation and calculations of data for the heavy metal concentrations in the vegetables were implemented by the Excel software (version 2016).

RESULTS AND DISCUSSION

Contamination level of heavy metals in different types of the summer and winter vegetables

Heavy metal concentrations of the vegetables varied widely with the type of vegetables and areas, as shown in Figs. 2 and 3. In the summer vegetables (Fig. 2), the highest Zn concentration (114.7 mg/kg) was observed in helencha of Banglabazar, followed by root (66.9 mg/kg) and shoot (61.1 mg/kg) of water spinach of the same area, and taro root (66.7 mg/kg) of Chandra. Taro leaf

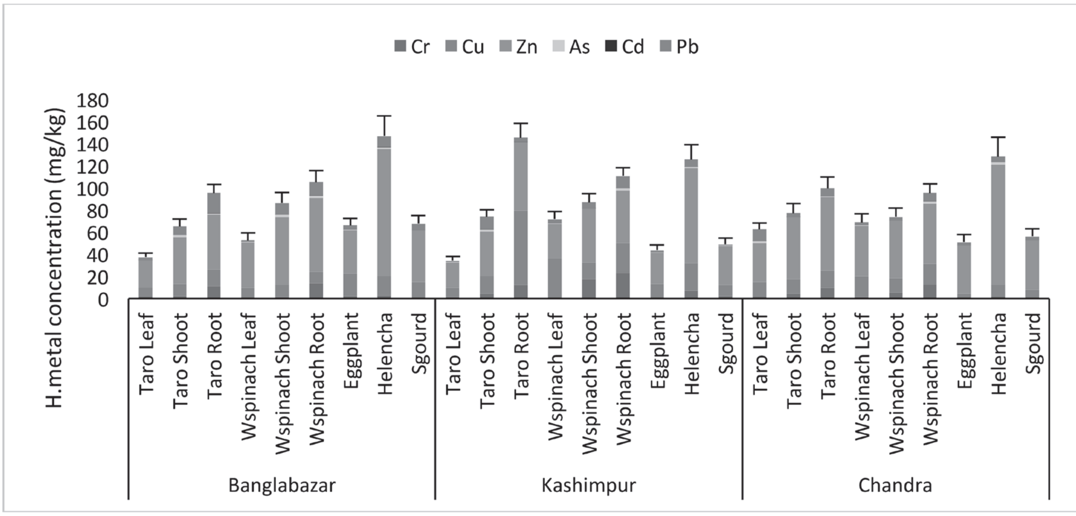

Fig. 2. Heavy metal concentrations of the summer vegetables in the respective areas.

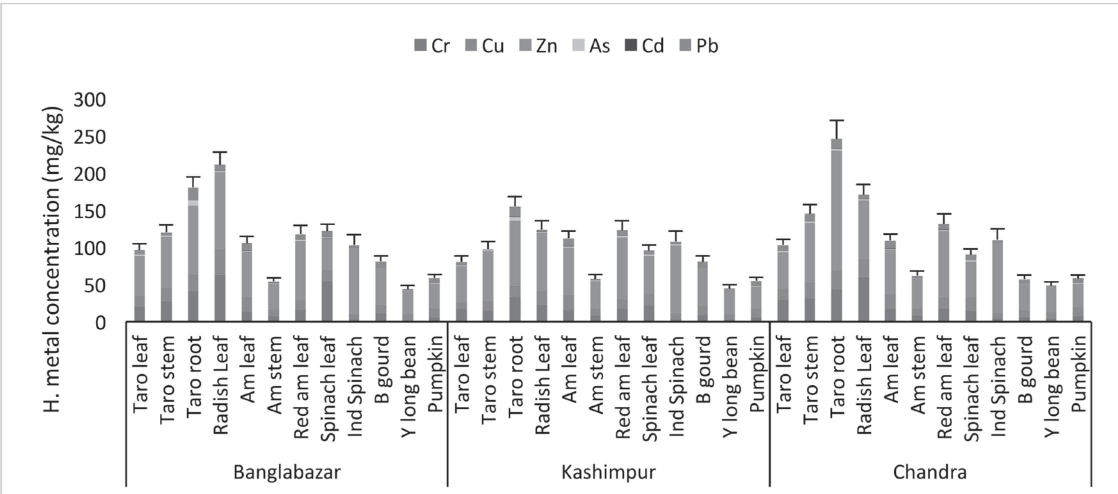

Fig. 3. Heavy metal concentrations of the winter vegetables in the respective areas.

contained the lowest Zn concentration (22.9 mg/kg) in the Kashimpur. The Cr and Cu concentrations in taro root and water spinach root were higher in Kashimpur; and the Pb and Cd concentrations were higher in Banglabazar than in the other areas. For eggplant, the concentrations of As and Cd were higher in Kashimpur and lower in Banglabazar than in the other areas. On the other hand, the Cr and Cu concentrations were high in eggplant in Banglabazar. Sponge gourd contained higher As and Cd concentrations in Chandra and contained lower As and Cd concentrations in Kashimpur, whereas the Cu and Pb concentrations of sponge gourd were higher in Banglabazar than in the other areas. Regarding the total concentration of all heavy metals, the highest was observed in helencha in all three areas; the second highest was in water spinach root; and very low concentrations were observed in sponge gourd, eggplant and taro leaf. The small standard error values of the concentrations in the upper part of the bar graph (Fig. 2) indicate no significant differences between the sampling locations.

For winter vegetables (Fig. 3), the highest Zn concentration (161.36 mg/kg) was observed in taro root of Chandra followed by Indian spinach (94.54 mg/kg), and by radish leaf (103.35 mg/kg) of Banglabazar. The lowest Zn concentration was observed in pumpkin (28.79 mg/kg) of Kashimpur. The highest Cr (63.55 mg/kg) and Cu (34.69 mg/kg) concentrations were observed in radish leaf of Banglabazar; in addition, the highest As (6.92 mg/kg) and Pb (17.23 mg/kg) concentrations were in taro root of Banglabazar. In Chandra, red amaranth leaf showed the highest Cd (1.11 mg/kg) concentration.

Regarding the total concentration of all heavy metals (dry season), taro root was the highest in the respective three areas, followed by radish leaf, red amaranth leaf, taro stem and Indian spinach, whereas yard long bean and pumpkin were the lowest in the respective areas. The small standard error values of the concentration in the upper part of the bar graph (Fig. 3) indicate no significant differences between the sampling locations.

In terms of the differences between root, leaf and fruit vegetables, the total heavy metal concentration was the highest in root vegetables, followed by leaf and then by fruit vegetables (Figs. 2 and 3). Root vegetables that were grown in contact with soil were thought to be more heavily contaminated than leaf and fruit vegetables. Roots of taro and water spinach grow in moist rich soils, making these vegetables to be highly contaminated with heavy metals. Soil moisture is provided by the contaminated irrigation water, and vegetable roots absorb the moisture from the soil. Heavy metals absorbed with the moisture are translocated in plant body. The absorption and translocation abilities depend on the kind of vegetables and which differentiate the heavy metal concentrations between vegetables (Islam and Hoque, 2014). The heavy metal concentrations in the summer vegetables were lower than the winter vegetables (according to paired t–test; t = 0.18–5.15, df = 26, $p < 0.05$), which may be due to the effect of rainfall in diluting irrigation water for the summer vegetables growth.

Estimated daily intake of heavy metals and health risk assessment

The degree of toxicity of heavy metals to human being depends upon their daily intake (Singh *et al.*, 2010). Long–term exposure to toxic heavy metals through the ingestion of contaminated vegetables could hamper biochemical processes (Anhwange *et al.*, 2013). Thus, assessment of the toxicity level of vegetables heavy metal concentrations is important. Therefore, daily intake of heavy metals (DIM) was estimated and health risk index (HRI) was calculated.

For the summer vegetables (Table 1), the highest intake of Cu, Cd and Pb were observed in taro. Helencha showed the highest intake of Zn and As, whereas water spinach exhibited the highest intake of Cr. The DIMs of Cr, Cu, Zn, As, Cd and Pb ranged from 0.0003 to 0.0019, from 0.0023 – 0.004, from 0.0079 – 0.022, from 0.00003 – 0.00035, from 0.000016 – 0.000043, and from 0.00059 – 0.00156, respectively. The order of heavy metal intake of all vegetables is Zn > Cu > Pb > Cr > As > Cd.

The health risk associated with the consumption of vegetables was estimated by calculated HRI values. Regarding HRI, taro indicated the highest HRI of Cu, Cd and Pb (Table 1). Helencha showed the highest HRI of Zn and As, whereas water spinach showed the highest HRI of Cr. The mean HRIs were found to be higher in As (0.53) followed by Cr (0.31), Cu (0.08) and Zn (0.04). The lowest HRI was observed in Cd (0.05). In total, the leaf and root vegetables exceeded the recommended HRI of larger than 1.

For the winter vegetables, the DIMs of Cr, Cu, Zn, As, Cd and Pb ranged from 0.0007 – 0.0105, from 0.0016 – 0.0055, from 0.0066 – 0.0196, from 0.00004 – 0.00048, from 0.000001 – 0.000229, and from 0.00008 – 0.00189 mg/kg per day, respectively (Table 2). Among different winter vegetables, radish showed the maximum intake of Cr and Cu, whereas Indian spinach and red amaranth showed the maximum intake of Zn and Cd, respectively. Taro indicated high intake of As and Pb. The mean DIM of the vegetables decreased in the order of Zn > Cr > Cu > Pb > As > Cd.

Regarding HRI (Table 2), radish showed the highest HRI of Cr and Cu. Taro exhibited the maximum HRI of As and Pb, whereas red amaranth and Indian spinach presented the maximum HRI of Cd and Zn. However, the highest HRI range was observed in Cr (from 0.29 – 3.49) followed by As (from 0.29 – 1.59), Cu (from 0.04 – 0.14) and Zn (from 0.02 – 0.07). The lowest mean HRI was found in Cd which ranged from 0.002 – 0.459 followed by Pb (from 0.02 – 0.47). In total, leaf and root vegetables exceeded the recommended HRI of larger than 1. The DIM and HRI for the summer vegetables were lower than those for the winter vegetables, which may be due to the lower heavy metal concentrations of the summer vegetables.

To assess the health risk of heavy metals, it is necessary to estimate the exposure level of heavy metals by quantifying their intake by the target organisms (Khan *et al.*, 2008). If people consumed the heavy metal contaminated crops, the people will face serious health risk. The vegetables produced in the study area are marketed in Gazipur District and Dhaka City. Therefore, the HRI for the respective vegetables still needs to be carefully monitored and assessed. The total HRI values (sum of individual heavy metal HRI) indicated much more health risk condition for the consumers when each vegetable is consumed. The total HRI for the vegetables were larger than the recommended value of HRI > 1, except for fruit vegetables in both the summer and winter vegetables. Therefore, potential health risk for root and leaf vegeta-

Table 1. DIM (mg/kg per day) and HRI for the respective summer vegetables

Division of vegetables	Name		Cr	Cu	Zn	As	Cd	Pb	Total
Root and Leaf	Taro (n = 9)	DIM	0.0012	0.0040	0.0095	0.00016	0.000043	0.00156	0.016
		HRI	0.40	0.10	0.03	0.54	0.09	0.39	1.55
	Water spinach (n = 9)	DIM	0.0019	0.0038	0.0107	0.00021	0.000019	0.00136	0.018
		HRI	0.63	0.09	0.04	0.69	0.04	0.34	1.83
	Helencha (n = 3)	DIM	0.0009	0.0038	0.0220	0.00035	0.000029	0.00151	0.029
		HRI	0.30	0.09	0.07	1.17	0.06	0.38	2.08
Fruit	Eggplant (n = 3)	DIM	0.0003	0.0026	0.0079	0.00003	0.000016	0.00059	0.011
		HRI	0.11	0.07	0.03	0.12	0.03	0.15	0.50
	Sponge gourd (n = 3)	DIM	0.0003	0.0023	0.0091	0.00003	0.000016	0.00069	0.012
		HRI	0.09	0.06	0.03	0.11	0.03	0.17	0.50

n: number of samples

Table 2. DIM (mg/kg per day) and HRI for the respective winter vegetables

Division of vegetables	Name		Cr	Cu	Zn	As	Cd	Pb	Total
Leaf	Radish Leaf (n = 6)	DIM	0.0105	0.0055	0.0186	0.00030	0.000125	0.00111	0.036
		HRI	3.49	0.14	0.06	1.00	0.250	0.28	5.22
	Amaranth (n = 12)	DIM	0.0026	0.0032	0.0104	0.00025	0.000111	0.00130	0.018
		HRI	0.86	0.08	0.03	0.85	0.223	0.33	2.37
	Red Amaranth (n = 6)	DIM	0.0038	0.0030	0.0179	0.00029	0.000229	0.00139	0.027
		HRI	1.26	0.07	0.06	0.98	0.459	0.35	3.18
	Spinach (n = 6)	DIM	0.0066	0.0035	0.0102	0.00029	0.000098	0.00136	0.022
		HRI	2.20	0.09	0.03	0.95	0.197	0.34	3.81
	Indian spinach (n = 6)	DIM	0.0009	0.0016	0.0196	0.00020	0.000036	0.00062	0.023
		HRI	0.29	0.04	0.07	0.67	0.072	0.15	1.29
	Pumpkin (n = 6)	DIM	0.0014	0.0027	0.0066	0.00009	0.000017	0.00155	0.012
		HRI	0.48	0.07	0.02	0.29	0.034	0.39	1.28
	Bottle gourd (n = 6)	DIM	0.0020	0.0024	0.0095	0.00009	0.000024	0.00170	0.016
		HRI	0.65	0.06	0.03	0.29	0.049	0.42	1.51
Root and leaf	Taro (n = 18)	DIM	0.0062	0.0037	0.0168	0.00048	0.000041	0.00189	0.029
		HRI	2.08	0.09	0.06	1.59	0.082	0.47	4.38
Fruit	Yard long bean (n = 6)	DIM	0.0007	0.0018	0.0072	0.00004	0.000001	0.00008	0.010
		HRI	0.22	0.05	0.02	0.12	0.002	0.02	0.43

n: number of samples

bles are of high health concern.

CONCLUSIONS

Heavy metal concentrations were higher in root and leaf vegetables than in fruit vegetables in both the summer and winter vegetables. Moreover, the heavy metal concentrations were lower in the summer than in the winter vegetables, for which dilution of irrigation water by rainfall during the summer vegetables growing period was perhaps responsible. The total HRIs for root and leaf vegetables were larger than 1, except for fruit vegetables in both the summer and winter vegetables. Therefore, the root and leaf vegetables produced from the study area are considered to have a high health risk and unsuitable for human ingestion.

ACKNOWLEDGEMENTS

The authors are grateful to the authorities of Bangabandhu Sheikh Mujibur Rahman Agricultural University, Bangladesh, and Kyushu University, Japan for providing laboratory facilities during the research period. Special thanks to Dr. Akinori Ozaki, Assistant Professor of the Institute of Tropical Agriculture, Kyushu University for his support of samples import from Bangladesh to Japan under the permission of Japanese Government.

AUTHOR CONTRIBUTIONS

Conceptualization and design, M. Ahmed and K. Kurosawa; Methodology and analysis and writing original draft, M. Ahmed; Investigation and supervision, K. Kurosawa and M. Matsumoto; Review and editing, M. Ahmed, M. Matsumoto and K. Kurosawa

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