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Contents of Pollutant and Nutrient Elements in Rice and Wheat Grown on the Neighboring Fields

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ABSTRACT

Because of recent agricultural policy to suppress rice production, a rather rare situation occurred in one prefecture in Japan that rice and winter wheat were grown in fields neighboring each other, rice being grown from May to October, and wheat from November to June of the next year. Grains of such rice and wheat were analyzed for cadmium (Cd) and lead (Pb) by atomic absorption spectrometry, and eight nutrient minerals by inductively coupled plasma emission spectrometry. Concentrations of nutrient minerals were higher in wheat grains than in unpolished rice grains (without husk), and similar trends were observed also for Cd and Pb. Flour obtained by milling of the wheat grains had significantly less Cd and nutrient minerals than the mother grains, and such reduction was also observed by treatment of unpolished rice to polished grain. Pb concentration was also reduced by the polishing of rice. Pb in wheat flour appeared to be higher than that in whole grain. Comparisons between the final edible forms of the two cereals showed that K and Cu were higher in polished rice than in wheat flour and that such may also be the case for Cd, whereas the reverse was the case for Ca and Fe, and possibly for Pb.

Index Entries: Cadmium; dietary intake; lead; nutrient minerals; pollutant elements; rice; wheat.

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INTRODUCTION

Rice and wheat are the most popular sources of energy for the daily life of many people in the world. For example, 1988 statistics showed that Japanese people obtained 701 kcal every day from rice, whereas Americans and Germans gained 564 and 522 kcal/d, respectively, from wheat *(1)*. It is well known, however, that both cereals are not only important sources of nutrients, but are also significant sources of pollutant elements, such as cadmium (Cd) and lead (Pb) *(2–4)*.

In the present study, the concentrations of the two pollutant metals (Cd and Pb) together with eight nutrient elements are compared between rice and wheat samples that were grown on neighboring agricultural fields in the same village.

MATERIALS AND METHODS

Sample Collection

The collection of samples was conducted in a farming village in western Japan in November 1994. Seven ripe unpolished rice samples (without husk) and eight ripe winter wheat grain samples were obtained from seven farmers in a single village (thus, one farmer out of the seven offered two wheat samples because he produced two types of wheat). Selection of farmers was by chance depending on their good will, and no rigid sampling strategy could be taken. It should be stressed, however, that their fields were in a group, so that the two sample cereals were all grown in the fields neighboring each other as will be discussed later, although the harvest seasons were different (i.e., October for rice and June for wheat).

Analysis for Elements

Unpolished rice was treated in the laboratory to obtain polished rice with a small-scale rice polisher "PARASET" for research purposes (a product from Kett Science, Tokyo, Japan). The recovery of the polished rice grains after 1 min of treatment of 10 g unpolished rice grains was about 96% (by weight). Wheat grains were ground in a kitchen mill "MILLSER" (a product from Iwatani Sangyo, Tokyo, Japan). Grains (40 g) were treated for 15 s to rough whole grain powder, and 6 g of rough whole grain powder were sampled. The remainder was filtered through a 1000-μm nylon sieve to obtain the brown bran fraction. The filtrate was milled for another 15 s, and the fine powder thus obtained was passed through a 105-μm nylon sieve to obtain white flour.

A 6-g portion of each sample was taken in a Teflon tube and wet-ashed by heating (up to 160°C for 3 h) in a block digester *(5)* in the presence of nitric acid (5 mL) + sulfuric acid (0.1 mL). Each sample was then

Table 1
Instruments, Wavelength, and Detection Limits for the Measurement of 10 Elements in Rice and Wheat

Element	Instrument[a]	Wavelength (nm)	Dilution[b]	Detection limit
Ca	ICPES	317.933	x 6	1 µg/g
Cd	AAS	228.8	x 1	0.1 ng/g
Cu	ICPES	324.754	x 24	0.01 µg/g
Fe	ICPES	259.940	x 24	0.2 µg/g
K	ICPES	766.490	x 6	10 µg/g
Mg	ICPES	285.213	x 24	0.5 µg/g
Na	ICPES	588.995	x 6	5 µg/g
P	ICPES	214.914	x 24	1 µg/g
Pb	AAS	283.3	x 1	4 ng/g
Zn	ICPES	213.856	x 24	0.1 µg/g

[a] ICPES, inductively coupled plasma emission spectrometry. AAS, graphite furnace atomic absorption spectrometry.

[b] Dilution of the wet ash before application to the instrument.

cooled down to room temperature, and then nitric acid (10 mL) and perchloric acid (2 ml) were added and reheated (up to 160°C for 3 h) to complete digestion.

One portion of the wet ash was analyzed for Cd and Pb with a graphite furnace atomic absorption spectrometer (Hitachi Model Z-8100, Tokyo, Japan) with an automated liquid sampler (Hitachi Model SSC-200, Tokyo, Japan), taking advantage of the Zeeman effect for background correction. Analytical conditions were as previously detailed *(6)*. Ammonium phosphate and ammonium nitrate were employed as matrix modifiers for Cd and Pb analysis, respectively. Another portion of the digest was diluted 6–24 times with deionized water and then subjected to nutrient element analysis *(7)* with an inductively coupled plasma emission spectrometer (ICPES: Model SPS 7000A with a glass concentric nebulizer: Seiko Instruments, Tokyo, Japan) connected with an automated liquid sampler (Model C83, Seiko Instruments, Tokyo, Japan). The power was suppled at 1 kW (frequency: 27.12 MHz), and the operation was in PSM mode, except for Cu for which DPM mode was used. Slit width for the light path was set at 20 µm at the entrance, and at 40 µm at the exit. Ar was employed as plasma gas (at a flow rate of 16 L/min), carrier gas (0.5 L/min), and chamber gas (0.5 L/min). Two-point linear calibration was employed for standardization, and the observation height was 10 mm. Samples were run three times. Wavelength used for the measurements, dilution, ratios, and respective detection limits are listed in Table 1.

Statistical Analysis

It was observed in a previous study that Cd and Pb concentrations in rice distribute log-normally rather than normally *(6)*, whereas nutrient concentrations distribute normally *(8)*. Accordingly, both pollutant and nutrient element concentrations were expressed in terms of arithmetic mean (AM) and arithmetic standard deviation (ASD). In addition, the two pollutant element concentrations were expressed also as geometric mean (GM) and geometric standard deviation (GSD). Student's *t*-test, either paired or unpaired as indicated, analysis of variance (ANOVA), and multiple comparison test after Scheffe were employed to detect possible differences between (or among) means. The tests were applied after log-normal conversion of the measures when a log-normal distribution was assumed. The difference was considered to be insignificant when $P > 0.10$.

RESULTS

Possible Metal Contamination During Milling Process of Wheat Grains

Because the mill "MILLSER" has blind metal blades to crush grains, possibility of metal contamination during milling was examined. For this purpose, rice grains rather than wheat grains were selected; Pb contents were lower in rice grains than in wheat grains, and therefore testing with rice grains should be more sensitive to detect possible Pb contamination than wheat grains. Accordingly, metal concentrations in rice grains before and after milling were compared.

When 380-g rice grains were treated with "MILLSER" and rice powder thus obtained was filtered through the 1000-μm nylon sieve, 50.1-g filtrate and 328.6-g residue were obtained. Pb contents in the rice powder, the filtrate, and the residue were 21.5, 82.1, and 12.9 ng/g, respectively. The quantitative evaluation showed that the Pb amounts in the filtrate and the residue were 4114 ng (82.1 ng/g × 50.1 g) and 4239 ng (12.9 ng/g × 328.6 g), respectively. The sum of Pb in the two fractions was 8353 ng, or 102.6% of the estimate from the weight and Pb content of the rice powder (8142 ng, or 21.5 ng/g × [328.6 + 50.1] g).

When tested with the rice polisher "PARASET," the Pb contents in rice grain (20.3 ng/g as GM of 5 determinations) and the rice powder (21.5 ng/g) did not differ from each other as expected, because no metal parts contact with the sample in the case of the rice polisher. Thus, processes of powdering and filtration were considered not to be associated with Pb contamination.

No changes were induced in Cd levels by milling or polishing of rice grains. It was also confirmed that the levels of the eight nutrient minerals were not affected by milling procedure.

Levels of Cd, Pb, and Eight Elements in Unpolished Rice and Wheat Grain

When the concentrations of Cd and Pb in rice and wheat were calculated for AM and ASD, ASD was often in excess of 30% of AM, suggesting that a normal distribution might not be applicable as expected (*7*). Accordingly, GM and GSD were also calculated for Cd and Pb with an assumption of a long-normal distribution. The results are summarized in Table 2. Levels of eight nutrient minerals in rice and wheat are summarized in Table 3 in terms of AM and ASD, where ASDs were < 30% of corresponding AMs. Na concentration was also measured, but the concentrations were all below the detection limit of 5 μg/g cereal.

Although Cd and Pb levels in wheat grains tended to be somewhat higher than the counterpart values in unpolished rice, they did not differ significantly when compared after or without logarithmic conversion (Table 2). In cases of nutrient minerals (Table 3), unpolished rice contained significantly ($P < 0.01$) less Ca, P, Fe, and Zn, and more K than wheat grains. The levels of Mg and Cu were comparable between the two cereal grains.

Effects of Polishing or Milling on Cd and Pb Concentrations

Comparison between the seven pairs of unpolished rice samples and those after polish (i.e., polished rice samples) showed that Cd concentrations tended to be reduced by the treatment; the difference was significant ($P < 0.01$) when assayed by paired *t*-test after logarithmic conversion. The high Cd content in the bran sample appeared to be on line with the reduction in rice after polish.

A similar trend was observed also for Pb in rice, but the changes were of borderline significance ($P < 0.10$). Because the instrument was not designed for recovery of rice bran, it was not possible technically to collect bran separately from each rice samples. Thus, only one pooled bran sample was available from all of the seven unpolished rice samples. The analysis of the bran sample showed much higher Cd and Pb content than the levels in polished rice (Table 2).

Effects of Polishing and Milling on Nutrient Mineral Concentrations

Concentrations of seven nutrient minerals after polishing of the rice grains were all significantly ($P < 0.01$, except Ca for which $P < 0.05$) lower than the concentrations before the treatment when assayed by paired *t*-test (Table 3). In agreement with such reductions, the element concentrations in rice bran were all higher than the corresponding average values for polished or unpolished rice samples. It was not possible to make a statistical analysis, because only one bran sample was available.

Table 2
Cd and Pb Contents in Rice and Wheat Harvested in the Neighboring Agricultural Fields

Cereal	Cadmium (ng/g)		Lead (ng/g)	
	AM ± ASD	GM (GSD)	AM ± ASD	GM (GSD)
Rice (7 samples)				
a. Unpolished rice	170.0±96.6	142.3(1.97)	11.0±9.4	7.9(2.41)
b. After polish	150.7±89.8^{BS}	121.8(2.13)^{ns}	6.2±3.3	5.4(1.65)
c. Bran	317.7		88.8	
P by paired *t*-test (a vs. b)		**		BS
Wheat (8 samples)				
d. Whole grain	202.1±44.0^{ns}	196.9(1.29)^{ns}	16.6±10.1^{ns}	13.6(2.03)^{ns}
e. Milled whole grain	193.8±47.1	187.9(1.32)	24.8±10.0	22.7(1.60)
f. Flour	86.3±17.3	84.6(1.24)	79.6±37.1$^{\circ\circ}$	72.5(1.60)$^{\circ\circ}$
g. Bran	209.7±49.0	202.9(1.35)	9.9±7.2	8.0(1.95)
P by paired *t*-test (d vs. e)		ns		*
Ibid. (e vs. f)		**		**
Ibid. (f vs. g)		**		**
Ibid. (e vs. g)		ns		**

Values are AM ± ASD in ng/g wet wt, or GM in ng/g wet wt (GSD). Circles (°°) show that the value is significantly ($P < 0.01$) larger than the counterpart value when comparison is made between polished rice and wheat flour. Asterisks show significant (** for $P < 0.01$, * for $P < 0.05$) difference in ANOVA. In the cases of assay with paired *t*-test, asterisks are given to the values that are significantly (** for $P < 0.01$, * for $P < 0.05$) larger than the counterpart. "BS" means borderline significance (i.e., $0.05 < P < 0.10$). "ns" means not significant.

Table 3
Nutrient Mineral Contents in Rice and Wheat Harvested in the Neighboring Agricultural Fields

Cereal	Ca	Mg	K	P	Fe	Cu	Zn	Na
Rice (7 samples)								
a. Unpolished rice	92.5±13.2	1078±66	958.9±65.1††	2894±213	10.9±1.0	3.43±0.70	24.0±4.3	ND[a]
b. After polish	46.3±5.5	377±150^{ns}	595.4±132.6°°	1235±352^{ns}	2.7±1.1	2.79±0.66°°	17.0±4.2^{ns}	ND
c. Bran[b]	505.1	6732	4812.7	16560	95.9	10.78	127.9	ND
P by paired t-test (a vs. b)	*	**	**	**	**	**	**	
Wheat (8 samples)								
d. Whole grain	270.1±9.1††	1104±38^{ns}	484.8±56.9	3456±184††	45.0±1.9††	3.94±0.24^{ns}	43.1±8.3††	ND
e. Milled whole grain	271.1±11.2	1133±49	502.7±31.2	3493±198	45.7±3.3	4.23±0.46	43.0±6.9	ND
f. Flour	117.5±7.6°°	221±20	433.0±14.8	1081±85	20.0±6.5°°	1.33±0.12	10.7±2.2	ND
g. Bran	301.5±11.7	1471±141	506.3±20.6	4290±374	52.2±3.4	5.05±0.53	50.7±8.0	ND
P by ANOVA (among e, f & g)	**	**	**	**	**	**	**	
P by paired t-test (d vs. e)	ns	ns	ns	ns	ns	BS	ns	
Ibid. (e vs. f)	**	**	**	**	**	**	**	
Ibid. (f vs. g)	**	**	**	**	**	**	**	
Ibid. (e vs. g)	**	**	ns	**	**	**	**	

Values are AM ± ASD in μg/g wet wt. Daggers (††) show that the value indicated is significantly ($P < 0.01$) larger than the counterpart value when comparison is made between unpolished rice and wheat grain. Circles (°°) show that the value is significantly ($P < 0.01$) larger than the counterpart value when comparison is made between polished rice and wheat flour. Asterisks show show significant difference (** for $P < 0.01$, * for $P < 0.05$) in ANOVA. In the case of assay with paired t-test, asterisks are given to the values that are significantly (** for $P < 0.01$, * for $P < 0.05$) larger than the counterpart. "BS" means borderline significance (i.e., $0.05 < P < 0.10$). "ns" means not significant. [a]Not detectable (i.e., below the detection limit of 5 μg/g). [b]One determination only.

Very similarly, flour separated from bran after milling of wheat grains contained significantly ($P < 0.01$) less elements than original wheat grains or milled grains when assayed by paired *t*-test. Conversely, the bran fraction contained significantly ($P < 0.01$) more elements than milled grains with the exception of K.

Comparison of Edible Preparation of Rice and Wheat

The most common edible forms of rice and wheat are polished rice and wheat flour, respectively. Accordingly, contents of Cd, Pb, and seven nutrient minerals were compared between polished rice samples and wheat flour samples. Regarding the two toxic metals, polished rice tended to contain more Cd (although the difference was statistically insignificant; Table 2) and probably less Pb. In the cases of nutrient minerals, polished rice contained more K and Cu ($P < 0.01$ for both), and less Ca and Fe ($P < 0.01$ for both). The contents of Mg, P, and Zn did not significantly differ between the two cereal preparations.

DISCUSSION

Historically in Japan, rice and winter wheat had been grown in sequence in the same agricultural fields as crops for warmer and colder seasons of the same year, respectively. In several decades, however, interest of farmers was concentrated on production of economically more efficient high-quality rice, and production of winter wheat was almost discontinued because seedlings of high-quality rice request planting in the paddy before winter wheat becomes ripe. Very recently, further changes have been taking place in some parts of Japan, especially in the western half, which has enabled winter wheat production to come back. This revival is related to the governmental agricultural policy of rice harvest curbing in fear of excess rice production. Winter wheat now is a choice of some farmers as nonrice crops.

Taking advantage of such local changes in recent years, the present study was conducted in a farming village where farmers use a part of their fields for winter wheat production on dry land and the remaining major parts for rice production in the paddy, with changes of fields for wheat and rice depending on the choice of the year. Thus, wheat fields are surrounded by a rice paddy, and two types of fields (one unit commonly being about 1000 m^2) are neighboring each other with different harvest times—late spring for winter wheat and late autumn for rice.

It has been considered that the application of Cd-contaminated fertilizer is one of the leading sources of Cd in soil *(9)*, although natural sources, such as volcanic ash, may also play an important role *(10)* in soil contamination with Cd, whereas Pb contamination is primarily in relation to human activities *(11)*, such as addition of organic Pb in automobile gasoline. The present study suggests that Cd and Pb contents tend to be higher (although the difference is statistically insignificant) in wheat grains than unpolished rice grains in the two cereals harvested essentially from the same fields. Nutrient minerals are also more abundant in wheat grain than in unpolished rice with the exception of K.

Thus, it is likely that the levels of minerals, including Cd and Pb, independent of physiological roles, are generally higher in wheat grains than in rice grains from the same soil. It is known that only a small portion of the metals absorbed via roots will reach grains of wheat *(12,13)* and rice *(14–16)*. Nevertheless, the present observation of Cd and Pb is on line with the previous publication *(17)* that Cd in rice grain is higher when water over paddy soil is removed early in the earing period than late when the grains are nearly ripe, because the metal in the soil is in reductive forms (and less efficiently absorbed into roots) when the soil is under water cover (i.e., as paddy). It is in oxidative forms and readily absorbed, however, when soil is dry. Winter wheat is grown on dry soil throughout its life, and a linear correlation was observed between Cd in soil and Cd in wheat grains grown on the soil *(18)*.

Such is also in general agreement with the findings in southern India by Srikumar *(19)* that both Cd and Pb are higher in wheat than in rice. In fact, Cd in grains of plants grown on soils spiked with 1000 ppm Cd oxide was more than 10 times higher in wheat grain than in rice grain *(20)*. Similarly, when soil was added 1000 ppm PbO, Pb in wheat grains was several times higher than that in rice *(20)*.

From public health viewpoints, the bioavailability of minerals, such as Fe and Ca *(21,22)*, in cereals is known to be lower than that in foods of animal origin, and more attention is paid to Cd and Pb contents because they are well-known insidious environmental pollutants and intoxicants *(23–26)*. Rice is in fact the leading source of Cd burden among rice-eating populations *(27)*. Rice grains are edible once the husk is removed, but commonly cooked after it is polished for better taste. In contrast, wheat is seldom consumed as grains, but is usually milled to flour. The observation that both Cd and Pb are twice or more times higher in rice bran than in polished rice (which is predominantly albumen) is in agreement with the report by Yoshikawa et al. *(28)* as cited by Kitagishi and Obata *(29)* that Cd level is twice as high in the outermost layer of the rice grain than in albumen. Thus, a significant amount of Cd may be removed by polishing rice grains. It should be noted that rice grains after polish nevertheless tend to contain more Cd than wheat flour.

Milling of wheat grains to flour also induced a significant reduction in Cd, i.e., reduction from about 200 ng Cd/g whole grains to 86 ng Cd/g flour. Moberg et al. *(30)* described that Cd contents in feed experimentally prepared for animals were higher in the bran (63 ng/g) than in the flour (32 ng/g). Wing *(31)* similarly observed that Cd content in whole wheat grains (71 ng/g) was higher than that the flour from the grains (31 ng/g). In wheat grown on soils amended with sewage sludge *(32)*, Cd content was almost three times higher in bran (1.20 µg/g) than in flour (0.41 µg/g). In contrast, Ahmad et al. *(33)* did not find a significant difference in Cd and Pb concentrations in wheat grains and flour in Pakistan. Ahn et al. *(34)* reported in their study on metal contents in livestock feed in Korea that the Cd level in wheat was comparable to that in wheat bran and that the same was also the case for Pb. It is highly possible in the latter two reports, however, that the flour (or the bran) and the grains analyzed were from different batches of wheat.

The reverse may be the case for Pb. The Pb concentration in wheat flour (79.6 ng/g) was much higher than the concentration in polished rice (6.2 ng/g). It appears that the milling of wheat grains to flour is not associated with the reduction in Pb content. Comparison of Pb in unmilled whole grain wheat (13.6 ng/g as GM) with that in milled whole grain (22.7 ng/g) suggests at first that the milling process employed may induce an increment in Pb concentration by 9 ng/g, although this is highly unlikely as previously tested with rice grains (*see* the Results section for details). Suppose the Pb in flour (72.5 ng/g) is mostly attributable to the Pb contamination during the milling process. Then the contamination should take place almost exclusively to the flour fraction and essentially nil to the bran fraction, because Pb in the bran is low (8.0 ng/g). Such is also quite unlikely to occur. The apparent increase may be attributable to technical difficulty in measuring Pb at levels close to the detection limit.

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