

-
- a completely-mixed adsorber at steady state. *Water Res.* 30 (6), 1495-1501.
- Villadsen, J.V., Stewart, W.E., 1967. Solution of boundary-value problems by orthogonal collocation. *Chem. Eng. Sci.* 22, 1483-1501.
- Weber Jr., W.J., Liang, S., 1983. Dual particle-diffusion model for porous adsorbents in fixed beds. *Environ. Progr.* 2 (3), 167-175.
- Yang, X.-Y., Al-Duri, B., 2001. Application of branched pore diffusion model in the adsorption of reactive dyes on activated carbon. *Chem. Eng. J.* 83 (1), 15-23.

Effect of rice-cooking water to the daily arsenic intake in Bangladesh: results of field surveys and rice-cooking experiments

K. Ohno, Y. Matsuo, T. Kimura, T. Yanase, M. H. Rahman, Y. Magara, T. Matsushita and Y. Matsui

ABSTRACT

The effect of rice-cooking water to the daily arsenic intake of Bangladeshi people was investigated. At the first field survey, uncooked rice and cooked rice of 29 families were collected. Their arsenic concentrations were 0.22 ± 0.11 and 0.26 ± 0.15 mg/kg dry wt, respectively. In 15 families, arsenic concentration in rice increased after cooking. Good correlation ($R^2 = 0.89$) was observed between arsenic in rice-cooking water and the difference of arsenic concentration in rice by cooking. In the second survey, we collected one-day duplicated food of 18 families. As a result, we estimated that six of 18 families likely used the arsenic contaminated water for cooking rice even they drank less arsenic-contaminated water for drinking purpose. We also conducted rice-cooking experiments in the laboratory, changing arsenic concentration in rice-cooking water. Clear linear relationships were obtained between the arsenic in rice-cooking water and the difference of arsenic concentration in rice by cooking. Factors that affect arsenic concentration in cooked rice are suggested as follows: (1) arsenic concentration in uncooked rice, (2) that in rice-cooking water, (3) difference in water content of rice before and after cooking, and (4) types of rice, especially, the difference between parboiled and non-parboiled rice.

Key words | arsenic, Bangladesh, cooking water, daily intake, duplicate portion sampling, rice

K. Ohno
Y. Matsuo
T. Kimura
T. Yanase
T. Matsushita
Y. Matsui
Department of Socio-Environmental Engineering,
Hokkaido University,
N13W8, Sapporo 060-8628,
Japan
E-mail: ohnok@eng.hokudai.ac.jp

M. H. Rahman
Department of Geology and Mining,
University of Rajshahi,
Rajshahi 6205,
Bangladesh

Y. Magara
Creative Research Initiative "Sousei",
Hokkaido University,
N21W10, Sapporo 001-0021,
Japan

INTRODUCTION

Arsenic contamination of groundwater in Bangladesh has been repeatedly reported (Nickson *et al.* 1998; Ohno *et al.* 2005). Arsenic poisoning posed by drinking arsenic-contaminated water is very severe and, for example, Hossain (2006) estimated that 85 million people were at risk in Bangladesh. In recent years, local people have become aware of the hazard of arsenic-contaminated water and begun to obtain drinking water from less contaminated sources as far as they can. However, arsenic is consumed not only via water but also via food. Regarding the arsenic intake via food, Roychowdhury *et al.* (2003) estimated the daily arsenic intake via water and food in West Bengal,

India, by market basket sampling methods. However, this sampling method cannot take into account of the effects of cooking process and cooking water. There may be the case that the safe drinking water sources may not be near enough to carry sufficient water home and the local people may still consume arsenic-contaminated water via indirect drinking such as cooking water, even if they drink safe water. In order to investigate the effects of cooking processes and cooking water, duplicated portion sampling method is necessary. Our research group has evaluated the arsenic intake via drinking water and food by the duplicated portion sampling method (Ohno *et al.* 2007). As a result of

the duplicated portion sampling, the mean arsenic intake from water and food was 0.15 ± 0.11 mg-As/day ($n = 18$), and the average contributions of each food category, which we defined in the paper, to the total daily arsenic intake were evaluated as follows: drinking water, 13%; liquid food, 4.4%; cooked rice, 56%; solid food, 11%; and cereals, 16% (Ohno *et al.* 2007). The contribution of cooked rice was the largest among the categories. Furthermore, we have found the following: (1) the local people were trying to find and drink less arsenic contaminated water because some of them were suffering from the symptoms regarding arsenic poisoning. (2) As a result, the arsenic intake via drinking water decreased drastically in many families; instead the contribution of cooked rice to the total arsenic intake was increased. (3) one third of the target families were suspected to be using arsenic-contaminated water for cooking although they tried to drink uncontaminated water. Therefore, arsenic concentration in rice seems important when people can obtain the drinking water that is less arsenic-contaminated. Moreover, using arsenic-contaminated water for cooking may increase the daily arsenic intake even if they are aware of the hazard of arsenic-contaminated drinking water. Especially, the effect of arsenic concentration in rice-cooking water is not well understood. Thus, the objective of this study is to investigate the effect of rice-cooking water to the daily arsenic intake. We conducted two kinds of field survey and laboratory experiments to investigate the relationship between arsenic concentrations in rice-cooking water and the difference of arsenic concentration in rice before and after cooking.

MATERIALS AND METHODS

Sample collection by field surveys

First field survey

First field survey was conducted in November, 2004. The survey area was the city of Nawabganj, Chapai Nawabganj district, Rajshahi division, Bangladesh. This area is one of the severely arsenic-contaminated areas and tube well waters contain as much as about 4 mg/L of arsenic (Ohno *et al.* 2005). Of course, local people were trying to avoid such highly contaminated water, but some people had

drunk the contaminated water that contained 0.1 mg/L or more of arsenic. We collected samples from 29 families in the area. We collected samples of uncooked white rice, cooked rice, and rice-cooking water from each house. The uncooked and cooked rice were collected in separate polyethylene sacks and the water was collected in a polyethylene bottle. Cooked rice was collected after the rice was cooled to room temperature. After the sampling, the weight of the cooked rice of each house was recorded, and then the rice was dried by a drier on the same day of the sampling. Finally, all the samples including the dried cooked rice were transported to the laboratory and kept in a cool and dark storage until analyzed.

Second field survey

In the second field survey, we collected one-day meal by the duplicate portion sampling methods from 18 families in one block in Chunalkhali village, Chapai Nawabganj district, Rajshahi division, Bangladesh. This sampling survey was conducted in June, 2005. The details of the location, the sampling methods, the pretreatment methods, and the analytical methods for the measurement of arsenic in the duplicated portion samples are published elsewhere (Ohno *et al.* 2007)

Laboratory experiments on rice-cooking

In order to elucidate the change of arsenic concentrations in rice during cooking rice, laboratory experiments were conducted. In Bangladesh, rice is generally cooked with excessive amount of water and the water that is not absorbed into rice grains during cooking is discarded (Bae *et al.* 2002; Rahman *et al.* 2006). We cooked rice in the Bangladeshi manner as follows. Fifty grams of raw white rice were put into a plastic bowl. The rice was washed three times for 30 seconds each with 100 mL of water. The washed rice was added to 250 mL of boiling water in a glass pot and the pot was covered with a lid. The rice was boiled in the pot for 10 minutes. Excessive hot water was then discarded and the boiled rice was kept in the pot with the lid for five more minutes to steam.

We used three types of uncooked white rice purchased at the market near the field survey areas. The names of rice

were Samsu China Variety (SCV), IRRI 28 Variety (IRRI), and Fine Variety Rice (FVR). In Bangladesh, there are two major kinds of rice depending on the way of polishing. One is parboiled rice, which is boiled before polishing, and the other is non-parboiled rice, which is polished without parboiling. Most of Bangladeshi people have been using parboiled rice for cooking (Rahman *et al.* 2006). SCV and IRRI are the parboiled rice, the common types that the local people eat. FVR is the non-parboiled rice, which is more expensive than other two types, and the local people seldom eat. Treated water collected before chlorination (after a rapid sand filtration process) from a water treatment plant in Japan was used as cooking water. This water contained only trace arsenic (0.0003 mg-As/L). For the preparation of cooking water that contains arsenic, the arsenic concentration was adjusted to the predetermined concentration (from 0.01 to 1.0 mg-As/L) by adding the solution of arsenic trioxide (Wako Pure Chemical Industries, Ltd., Osaka, Japan) to the water.

Pretreatment of rice samples

Rice samples were firstly freeze dried for a couple of days until the weight of the samples became stable, which means that water is sufficiently removed from the samples. Then, the samples were finely ground in a mill. The ground samples were digested using a microwave digestion system (ETHOS TC; Milestone S.r.l., Bergamo, Italy) by the following procedure. A portion of 0.5 g (dry weight) was weighed into a PTFE vessel and 4 mL of nitric acid and 1 mL of hydrogen peroxide (Ultra pure grade; Kanto Chemical Co., Inc., Tokyo, Japan) were added. The basic program of the microwave digester was as follows: increase the temperature from room temperature to 210°C over 30 min, remain at the temperature for 15 min and then cool down to room temperature over 10 min; maximum power was 1,000 W. Digested solution was made up to 50 mL with ultra pure water. Finally, it was filtered through 0.45- μ m membrane filter and used for determination of arsenic concentration.

Analytical method

Arsenic concentrations in water and in the pretreated solutions of rice and duplicated portion samples were

determined by inductively coupled plasma-mass spectrometer (ICP-MS; HP-4500; Agilent Technologies, Inc., Palo Alto, CA, USA). The instrumental parameters were as follows: RF power, 1,200 W; RF matching, 1.8 V; sample skimmer cone in Ni; monitoring masses; 75 (As) and 77 (ArCl⁺); plasma flow rate, 16 L/min; auxiliary flow rate, 1.1 L/min; nebulizer flow rate, 1.2 L/min. Yttrium (Y; $m/z = 89$) was used as the internal standard.

Validation of measurement

Standard Reference Material (SRM) of Rice Flour (SRM1568a) was purchased from National Institute of Standards and Technology (NIST), Gaithersburg, MD, USA, and the arsenic concentration of the SRM was measured in the same manner as rice samples. As a result, the certified values (0.29 ± 0.03 mg/kg dry wt.; mean \pm standard deviation (SD)) and the observed values (0.26 ± 0.01 mg/kg dry wt.; $n = 3$) of SRM were in good agreement.

RESULTS AND DISCUSSION

First field survey

The mean arsenic concentration of rice-cooking water in 29 families was 0.032 ± 0.067 mg/L. Seven of 29 families used the rice-cooking water that contained more than 0.05 mg/L of arsenic, which is the standard value for drinking water in Bangladesh. The mean arsenic concentration of uncooked and cooked rice were 0.22 ± 0.11 and 0.26 ± 0.15 mg-As/kg dry wt. ($n = 29$), respectively. The arsenic concentration in rice increased after cooking in 15 families. Regression analysis between the arsenic in rice-cooking water and the difference of arsenic concentration in rice before and after cooking was performed. As a result, good correlation ($R^2 = 0.89$) was observed (Figure 1). In this survey, we did not unify the types of rice and the methods of cooking rice, and arsenic concentration in uncooked rice was different house by house. The good correlation was observed regardless of these effects; this suggests that arsenic concentration in rice-cooking water gives strong influence to arsenic concentration in cooked rice. If people use arsenic contaminated

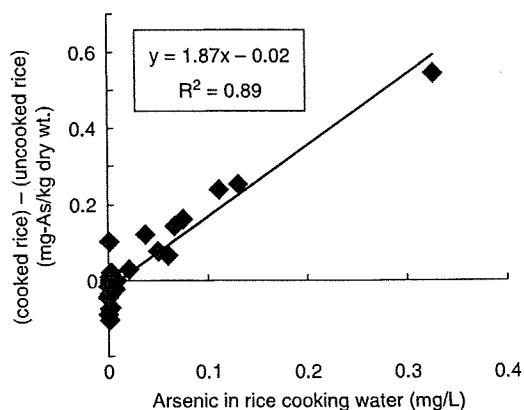


Figure 1 | Relationship between arsenic in rice-cooking water and the difference of arsenic concentration in rice before and after cooking ($n = 29$); results of the first field survey.

water for rice-cooking, their daily arsenic intake increases via cooked rice. On the other hand, the arsenic intake may decrease if they use water that contains low arsenic, which is about 0.01 mg/L or less (Figure 1). The arsenic in uncooked rice could be transferred from rice to water by cooking.

Rice-cooking experiments

In the experiments, we investigated the relationship between arsenic concentration in rice-cooking water and the arsenic concentration differences by cooking rice. As a result, very clear linear relationship was obtained within the same rice type (Figure 2). Determination coefficients were over 0.996 for all types of rice. When the arsenic concentration in cooking water was low, the

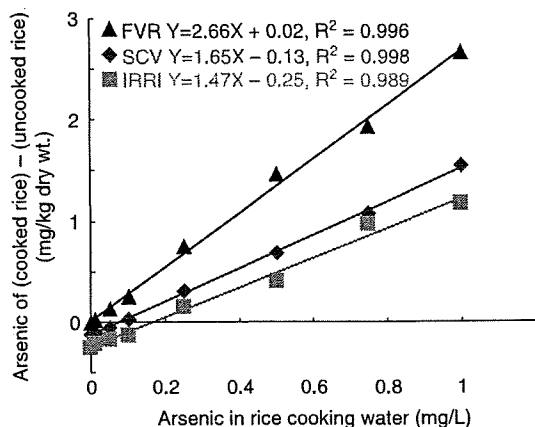


Figure 2 | Results of the rice-cooking experiments.

decrease in arsenic concentration in rice after cooking was observed in the experiments as well as the first field survey, except for the type of FVR. We measured the arsenic concentrations in the uncooked rice of FVR, SCV, and IRRI and they were 0.03, 0.21, and 0.36 mg-As/kg dry wt., respectively. Since the arsenic concentration in uncooked FVR rice was very low, the arsenic removal effect by the low arsenic in cooking water was not observed.

Slopes of regression lines were different among the types of rice (Figure 2). These differences are considered to be due to differences in the amount of water absorbed in the rice. The increases of water content (%) by cooking rice of FVR, SCV, and IRRI were 62%, 42%, and 48%, respectively. FVR type absorbs more water than the other two types, and the slope of the regression line is also larger. This result implies that arsenic may be absorbed in rice simply together with water, but the sorption mechanism seems more complicated and must be further investigated. Another big difference among the rice types is the method of polishing rice. As described above, FVR is non-parboiled rice, SCV and IRRI are parboiled rice. This difference may also cause the difference in the amount of arsenic sorbed while cooking. As the results of the first field survey and the rice-cooking experiments, we may suggest the factors that change the arsenic concentration in rice by cooking are as follows, though there may be other factors: (1) Arsenic concentration in uncooked rice, (2) that in rice-cooking water, (3) difference in water content before and after cooking, and (4) types of rice, especially, the difference between parboiled and non-parboiled rice.

Second field survey

In the second field survey, we collected the drinking water the 18 respondents drank at the survey period and also the water that the respondents had drunk before they changed the drinking water sources. Figure 3 shows arsenic concentrations in the previously drinking water sources and the water sources at the time of the survey. Previously, the people had drunk tube well water that contained high arsenic, but at the time of the survey, they drank tube well water that contained less arsenic, or water from dug well, which normally contained low arsenic. Accordingly, their arsenic intakes from drinking water were

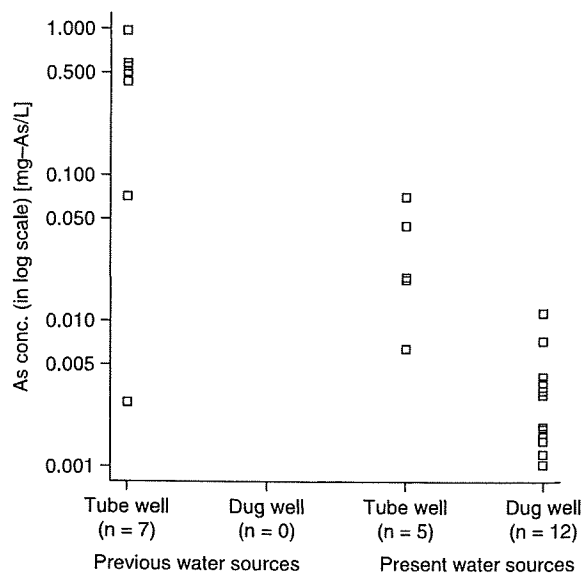


Figure 3 | Arsenic concentrations in previously drinking water sources and in the sources at the time of survey.

evaluated to decrease very much. Although dug well water does not always meet the water quality in terms of other chemical and biological standard values, The water quality issues other than arsenic are beyond the objective of this study.

Although they said they were drinking less arsenic-contaminated water, we estimated that six of 18 families likely used arsenic-contaminated water for cooking purpose (Ohno *et al.* 2007). In this study, we divided the families into two groups by cooking water sources: families using uncontaminated sources and those using contaminated sources. We compared the mean arsenic concentrations of drinking water and food categories between two groups (Table 1). The difference between two groups regarding

drinking water was not significant, but the differences regarding cooked rice and liquid food were significant. Arsenic in cooking water obviously gives direct influence on liquid food; it also gives distinct influence on cooked rice. Some of this deleterious effect by rice-cooking water was observed through the comparison among arsenic concentration in uncooked and cooked rice, and in drinking water (Figure 4). In the case of the first field survey, we collected the rice-cooking water and observed a good correlation (Figure 1). On the other hand, in the second field survey, we did not collect rice-cooking water but drinking water, and we observed some outliers (Figure 4). These outliers are considered to be due to the use of the arsenic-contaminated water for cooking purpose.

We also anticipated the significant differences between two groups regarding chapati-like cereal and solid food, because these kinds of food also need cooking water. Nonetheless, the differences were not significant. There are several possible reasons although we could not clarify them by the evidence in this study. With regard to the solid food, the amount of water used for cooking varied among the families. The amount was much less than that for cooking rice or liquid food. Therefore, the effect of arsenic from cooking water on the solid food may be less than that on rice and liquid food. One of other plausible reasons is that arsenic concentration in food itself varies among types of food (Roychowdhury *et al.* 2005; Smith *et al.* 2006), and the contents of solid food varied among the families. Thus, we could not distinguish the significant differences in arsenic concentrations in the solid foods between the two groups. We could not either find the significant differences in arsenic concentrations in the chapati-like cereals. One possible reason for this insignificance is due

Table 1 | The average concentrations of arsenic in drinking water and food

Cooking water source	Drinking water (mg/L)	Cooked rice	Chapati-like cereal (mg/kg dry wt.)	Solid food	Liquid food (soup) (mg/kg wet wt.)
All families ($n = 18$)	0.0078	0.46	0.20	0.44	0.038
Uncontaminated sources ($n = 12$)	0.0085	0.28	0.20	0.44	0.012
Contaminated sources ($n = 6$)	0.0063	0.83*	0.20	0.43	0.082 [†]

* $p < 0.1$ (Welch's one-sided t -test).

[†] $p < 0.05$ (Welch's one-sided t -test).

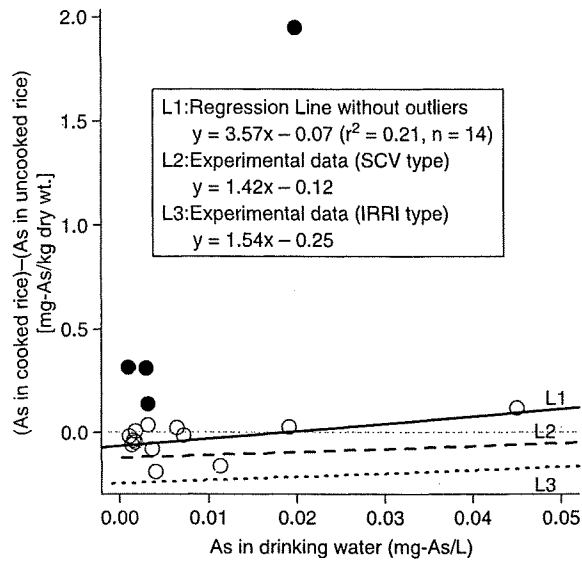


Figure 4 | Relationship between arsenic concentration in drinking water and the difference of arsenic concentration in rice by cooking. Circles indicate the samples ($n = 18$) and closed circles are the samples that are considered to be outliers.

to the large variation of water content, which was $35 \pm 20\%$ (Ohno *et al.* 2007). This variation is caused by differences in the cooking methods among families, which confounded the result. We may clarify the effect of using arsenic-contaminated cooking water for chapati-like cereals by conducting the cooking experiments that we have conducted to rice in this study.

We estimated daily arsenic intakes from water via direct drinking, liquid food, and rice-cooking water for all members of the target families (Figure 5). Arsenic intake from drinking water was estimated by multiplying the daily water consumption by arsenic concentration in drinking water of the corresponding family. Arsenic intake from liquid food was calculated by multiplying the daily consumption of liquid food by its arsenic concentration. Arsenic intake from rice-cooking water was estimated by multiplying the daily rice consumption by its arsenic concentration difference of rice before and after cooking (dry basis). We assigned zero to arsenic intake from rice-cooking water instead of allocating negative values when arsenic concentration in cooked rice is lower than that in uncooked rice. As a result, daily arsenic intake from drinking and cooking water was 0.069 mg/day in average. We also estimated the previous arsenic intake from water by applying the arsenic concentration of the previous drinking water source (Figure 3). In this case, the previous arsenic intake was calculated 1.6 mg/day in average, and about 90% of the intake was via direct drinking. The average arsenic intake via water has decreased by 96% by changing drinking water sources to less arsenic contaminated ones. Nonetheless, 12 of 65 people still took the daily arsenic intake more than 0.1 mg/day , which is the corresponding amount calculated by Bangladeshi drinking water quality standard value (Figure 5). Many of the people can reduce the arsenic



Figure 5 | Estimation of daily arsenic intake via drinking and cooking water ($n = 65$). Arsenic intake from food was excluded. Each vertical bar indicates one family member of target families, including children. Solid line at $y = 0.1$ and dashed line at $y = 0.02$ means the reference intakes. Arsenic intake of 0.1 mg/day is derived from the product of Bangladeshi drinking water standard value of 0.05 mg/L and general daily water intake of 2 L/d . Arsenic intake of 0.02 mg/day is derived from the product of WHO guideline value of 0.01 mg/L and the daily water intake.

intake by preventing from using arsenic-contaminated water for cooking purpose.

CONCLUSION

People may use arsenic-contaminated water for cooking purpose even after they can drink the water that contain less arsenic. In this study, we found that arsenic in rice-cooking water affected the arsenic concentration in rice after cooking. Therefore, the control and decrease of arsenic concentration in rice-cooking water would become important after the people could obtain the less arsenic-contaminated water for drinking purpose.

ACKNOWLEDGEMENTS

The authors thank Professor Bilkis Begum (Dept. of Fine Arts, University of Rajshahi, Bangladesh) for her work as an interviewer and translator during the surveys. This study was supported in part by research grants from the Ministry of Health, Labour and Welfare of the Government of Japan.

REFERENCES

- Bae, M., Watanabe, C. & Inaoka, T. 2002 Arsenic in cooked rice in Bangladesh. *Lancet* **360**, 1839–1840.
- Hossain, M. F. 2006 Arsenic contamination in Bangladesh—an overview. *Agric. Ecosyst. Environ.* **113**(1–4), 1–16.
- Nickson, R., McArthur, J., Burgess, W., Ahmed, K. M., Ravenscroft, P. & Rahman, M. 1998 Arsenic poisoning of Bangladesh groundwater. *Nature* **395**, 338.
- Ohno, K., Furukawa, A., Hayashi, K., Kamei, T. & Magara, Y. 2005 Arsenic contamination of groundwater in Nawabganj, Bangladesh, focusing on the relationship with other metals and ions. *Water Sci. Technol.* **52**(8), 87–94.
- Ohno, K., Tatsuya, Y., Matsuo, Y., Kimura, T., Rahman, M. H., Magara, Y. & Matsui, Y. 2007 Arsenic intake via water and food by a population living in an arsenic-affected area of Bangladesh. *Sci. Total Environ.* **381**, 68–76.
- Rahman, M. A., Hasegawa, H., Rahman, M. A., Rahman, M. M. & Miah, M. A. M. 2006 Influence of cooking method on arsenic retention in cooked rice related to dietary exposure. *Sci. Total Environ.* **370**(1), 51–60.
- Roychowdhury, T., Tokunaga, H. & Ando, M. 2003 Survey of arsenic and other heavy metals in food composites and drinking water and estimation of dietary intake by the villagers from an arsenic-affected area of west Bengal, India. *Sci. Total Environ.* **308**(1–3), 15–35.
- Smith, N. M., Lee, R., Heitkemper, D. T., DeNicola, C. K., Haque, A. & Henderson, A. K. 2006 Inorganic arsenic in cooked rice and vegetables from bangladeshi households. *Sci. Total Environ.* **370**, 294–301.

