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Cadmium and lead in Dutch and imported cereals

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Abstract

In a survey investigation, grains of Dutch cereals were analysed for Cd and Pb. Additionally a limited number of samples from imported cereals were investigated. In wheat (n = 188), barley (n = 37) and rye (n = 4) average Cd concentrations of 0.067, 0.026 and 0.027 mg/kg, respectively, were found, while respective average concentrations for Pb were 0.08, 0.14 and 0.09 mg/kg. In imported cereals, similar Cd and Pb levels were found. The provisional Dutch legal limits for Cd and Pb in cereal grains, i.e. 0.15 and 0.5 mg/kg respectively, were exceeded in 3 samples of wheat for Cd and in 2 samples of wheat for Pb. The Cd concentrations found in Dutch cereals are in good agreement with data reported for other countries while the Pb levels found tend to be slightly higher, but nevertheless remain clearly within established safety limits.

Introduction

During the last decades an increasing amount of attention has been paid to the presence of contaminants in the food chain. It is now generally considered important to limit the uptake of environmental contaminants by man to the unavoidable minimum. In view of this, provisional legal limits for the Cd, Pb and Hg levels in foods have been established in the Netherlands (Van der Reijden et al., 1985). For Cd and Pb in grains of cereals legal limits of 0.15 mg/kg and 0.5 mg/kg respectively have been proposed. The government intends to legalize these provisional limits through the Commodities Act.

The uptake of metals like Cd and Pb by higher plants is a complex process. With respect to cereals genetic factors, soil and weather conditions, fertilization and the state of maturity at harvest time determine the final level of trace elements (Varo et al., 1980; Smilde et al., 1982; Vlamis et al., 1985; de Haan et al., 1986). The relation between environmental contamination and the metal levels in crops often is not clear. Therefore the consequences of the introduction of legal limits for the heavy metal concentrations in crops can be estimated only by means of a survey investigation of crops.

In 1976 and 1977, grains of cereals from the Netherlands were sampled and investigated for heavy metals and arsenic by Wiersma et al. (1986). In grains of wheat the limit values for Cd and Pb were exceeded in respectively 6 % and 2 % of the investigated samples (n = 84) and in barley in respectively 24 % and 7 % of the samples (n = 45). These results suggested that the introduction of the proposed legal limits could result in serious restrictions for the cultivation in the Netherlands of certain cereals for human consumption.

In order to obtain additional and more recent information on the Cd and Pb levels in Dutch cereals another survey investigation was carried out, in which both inland cereals and a limited number of samples from imported cereals were included. The results of this investigation are presented in this paper.

Materials and methods

Samples were collected in 1984 and 1985. The inland cereals originated from different parts of the Netherlands. The sampling frequency of the regions was according to the importance of these regions for the cereal production in The Netherlands. The major part of the collected wheat samples came from marine clay soils, including sandy clays, in the south-western (n = 77), central (n = 43) and northern parts (n = 41) of the Netherlands. Additionally, 20 samples of wheat came from sandy soils, two from river clay and five from reclaimed moor. The barley samples came from the south-western (n = 11), central (n = 10) and northern (n = 4) marine clay areas; 20 samples originated from sandy soils and one barley sample had been cultivated on river clay. The rye samples came from marine clay (n = 2) and sandy soils (n = 2). The investigated imported rice samples consisted of brown rice. Before analysis, samples were grounded but not dried.

Double distilled water and reagents of recognized analytical quality were used. Glassware and quartz vessels were soaked in hot diluted HNO_3 (2:1 v/v) and rinsed several times with water before use. Samples were digested using a dry-ashing procedure. Cd and Pb were determined with differential pulse anodic stripping voltammetry using a Metrohm E 506 polarographic analyser and a hanging-mercurydrop electrode. The method of standard additions was applied.

A more detailed description of the analytical procedure has been reported previously (Vos et al., 1987). All analyses were carried out at least in duplicate. Within each batch of samples a recovery test, a blank and a reference sample were run also. The recovery tests (n = 53), which implied the addition of a known quantity of Cd and Pb to a sample before digestion, yielded average recoveries of 101 ± 8 % for Cd and of 100 ± 8 % for Pb. For the analysis (n = 13) of NBS citrus leaves 1572 a Cd and Pb concentration of 0.043 ± 0.014 mg/kg (certified value: 0.03 ± 0.01 mg/kg) and 13.9 ± 1.2 mg/kg (certified value: 13.3 ± 2.4 mg/kg), respectively were found. Analysis (n = 44) of an internal reference sample (wheat) yielded average Cd and Pb concentrations of 0.101 ± 0.009 and 0.26 ± 0.02 mg/kg respectively.

Statistical methods used included methods for the comparison of two randomized groups, described by Snedecor (1964).

Results and discussion

The results of the present study on the Cd and Pb concentrations in inland and imported grains of cereals are summarized in Table 1 and 2. In inland samples of wheat, barley and rye, average Cd concentrations of 0.067 mg/kg, 0.026 mg/kg and 0.027 mg/kg, respectively, were found. The provisional Dutch legal limit for Cd in grains of cereals, i.e. 0.15 mg/kg, was exceeded in 3 samples of wheat. In wheat and rye from other countries similar or slightly lower Cd levels were found. The average Cd concentrations in brown rice originating from different countries varied between 0.007 mg/kg and 0.021 mg/kg. Due to the small number of investigated samples the data given for imported cereals may not be representative for cereals from these countries.

In samples of Dutch wheat, barley and rye, average Pb concentrations of 0.08 mg/kg, 0.14 mg/kg and 0.09 mg/kg, respectively, were found. The provisional Dutch legal limit for Pb in cereals, i.e. 0.5 mg/kg, was exceeded only in two samples of wheat, in which Pb concentrations of 0.69 mg/kg and 1.64 mg/kg were found. Similar or slightly lower Pb concentrations were found in foreign cereals. High Pb concentrations were found in a number of rice samples from Thailand.

The results of this study indicate that the introduction of the proposed limit values for Cd and Pb in cereals will probably not result in serious limitations with respect to the cultivation in the Netherlands of cereals for human consumption. Lowering of the limit value for Cd in cereals to 0.1 mg/kg would result in an exceeding frequency of about 10 % for wheat. In barley and rye no concentrations above 0.1 mg/kg were found.

Due to differences in soil characteristics, especially the pH, the mobility of metals like Cd and Pb is generally high in sandy soils compared to clay soils. Consequently the availability of metals for uptake is expected to be higher in sandy soils. In the present study no statistical significant relation was found between the Cd and Pb concentrations in cereals and the soil type on which these cereals had been cultivated, although Cd levels in cereals from sandy soils tended to be higher. With respect to this it must be stated that an accurate statistical evaluation of the relation between soil type and the metal concentrations in cereals was not possible due to the fact that only a limited number of the investigated samples originated from other than marine clay soils.

Regional differences could not be demonstrated for the Pb levels in cereals. However, the Cd levels found in grains of wheat from marine clays showed regional dependency. In the south-west of the Netherlands significantly ($P < 0.001$) higher Cd concentrations were found. The Cd concentrations for wheat from the south-western, central and northern marine clay areas were 0.074 ± 0.028 mg/kg, 0.051 ± 0.019 mg/kg and 0.052 ± 0.020 mg/kg, respectively.

Next to external influences, genetic factors may play a significant role in the metal uptake by plants. Andersson & Pettersson (1981) for instance found differ-

Table 1. Cadmium concentrations (in mg/kg fresh weight) found in grains of cereals.

Crop	Country	n	Range	Median	Mean ± s.d.
Wheat	The Netherlands	188	0.017-0.256	0.063	0.067 ± 0.028
	France	34	0.026-0.111	0.047	0.050 ± 0.020
	UK	8	0.032-0.054	0.042	0.043 ± 0.008
	Belgium	3	0.057-0.075	0.060	0.064 ± 0.010
	USA	11	0.030-0.105	0.042	0.055 ± 0.025
Barley	The Netherlands	37	0.010-0.054	0.024	0.026 ± 0.011
Rye	The Netherlands	4	0.011-0.043	0.028	0.027 ± 0.015
	Danmark	4	0.011-0.024	0.018	0.018 ± 0.005
	France	3	0.009-0.010	0.010	0.009 ± 0.002
	Canada	3	0.007-0.012	0.011	0.011 ± 0.002
Rice	Thailand	3	0.013-0.036	0.014	0.021 ± 0.013
	Surinam	5	0.007-0.014	0.009	0.010 ± 0.003
	Italy	4	0.040-0.084	0.048	0.055 ± 0.020
	Argentina	3	0.006-0.008	0.008	0.007 ± 0.001
	USA	5	0.031-0.078	0.052	0.053 ± 0.021

Table 2. Lead concentrations (in mg/kg fresh weight) found in grains of cereals.

Crop	Country	n	Range	Median	Mean ± s.d.
Wheat	The Netherlands	188	0.02-1.64	0.06	0.08 ± 0.13*
	France	34	<0.01-0.13	0.05	0.05 ± 0.03
	UK	8	<0.01-0.08	0.03	0.03 ± 0.02
	Belgium	3	0.02-0.05	0.04	0.04 ± 0.02
	USA	11	0.03-0.07	0.06	0.05 ± 0.02
Barley	The Netherlands	37	0.03-0.24	0.14	0.14 ± 0.05
Rye	The Netherlands	4	0.06-0.16	0.07	0.09 ± 0.05
	Danmark	4	0.03-0.06	0.05	0.05 ± 0.02
	France	3	0.03-0.04	0.03	0.03 ± 0.01
	Canada	3	0.02-0.04	0.03	0.03 ± 0.01
Rice	Thailand	3	0.09-2.81	0.34	1.08 ± 1.50**
	Surinam	5	0.03-0.08	0.05	0.06 ± 0.02
	Italy	4	0.04-0.09	0.05	0.06 ± 0.02
	Argentina	3	0.04-0.09	0.06	0.06 ± 0.03
	USA	5	0.03-0.08	0.05	0.05 ± 0.02

* Without high value of 1.64: 0.07 ± 0.06.
** Without high value of 2.81: 0.22 ± 0.18.

Table 3. Some literature data on the cadmium levels (in mg/kg fresh weight) in grains of cereals.

Crop	n	Mean	Range	Country	Reference
Wheat	188	0.067	0.017-0.256	The Netherlands	this study
	84	0.07	0.02 -0.35	The Netherlands	Wiersma et al., 1986
	211	0.043	0 -0.25	West Germany	Seibel & Ocker, 1979
	1082	0.057	0.008-0.85	West Germany	Ocker & Seibel, 1980
	113	0.056	0.006-0.126	Austria	Schindler, 1983
	49	0.096	0.053-0.171	Sweden*	Andersson & Pettersson, 1981
	58	0.066	0.028-0.132	Sweden**	Andersson & Pettersson, 1981
	85	0.050	0.017-0.085	Finland	Varo et al., 1980
	288	0.043	<0.017-0.207	USA	Wolnik et al., 1983
Barley	37	0.026	0.010-0.054	The Netherlands	this study
	45	0.13	0.01 -0.54	The Netherlands	Wiersma et al., 1986
	47	0.021	0.004-0.032	Finland	Varo et al., 1980
Rye	4	0.027	0.011-0.043	The Netherlands	this study
	97	0.012	0.003-0.079	West Germany	Seibel & Ocker, 1979
	267	0.012	0.003-0.080	West Germany	Ocker & Seibel, 1980
	50	0.013	0.004-0.037	Finland	Varo et al., 1980

* Southern Sweden.
** Central Sweden.

Table 4. Some literature data on the lead levels (in mg/kg fresh weight) in grains of cereals.

Crop	n	Mean	Range	Country	Reference
Wheat	188	0.08	0.02 -1.64	The Netherlands	this study
	84	0.16	0.03 -0.65	The Netherlands	Wiersma et al., 1986
	217	0.048	0 -0.43	West Germany	Seibel & Ocker, 1979
	998	0.045	0.01 -0.42	West Germany	Ocker & Seibel, 1980
	115	0.057	0.001-0.670	Austria	Schindler, 1983
	85	<0.050	<0.050-0.140	Finland	Varo et al., 1980
	288	0.037	0.008-0.716	USA	Wolnik et al., 1983
Barley	37	0.14	0.03 -0.24	The Netherlands	this study
	45	0.27	0.08 -0.71	The Netherlands	Wiersma et al., 1986
	47	0.063	0.010-0.140	Finland	Varo et al., 1980
Rye	4	0.09	0.06 -0.16	The Netherlands	this study
	97	0.077	0.019-0.359	West Germany	Seibel & Ocker, 1979
	267	0.081	0.019-0.61	West Germany	Ocker & Seibel, 1980
	50	0.048	0.010-0.170	Finland	Varo et al., 1980

ences in Cd uptake between different cultivars of winter wheat. In the present study no significant differences between the Cd and Pb levels in grains from different cultivars were observed. The results obtained for the samples collected in 1984 and 1985 are not significantly different.

Several authors (Andersson & Bingefors, 1985; Andersson & Pettersson, 1981; Kjellström et al., 1975) have reported on considerable annual variations in the metal concentrations in grains of cereals. These variations are attributed mainly to effects which are related to differences in precipitation (Andersson & Pettersson, 1981).

In Tables 3 and 4 the Cd and Pb levels found in Dutch cereals in the present investigation are compared with results from other studies. All data in these tables have been calculated to mg/kg fresh weight, assuming a dry matter content of 85 %. With respect to Cd the concentrations found in wheat in the present study are similar to those reported previously for Dutch wheat by Wiersma et al. (1986). The concentrations found are also in good agreement with data reported for other countries. The average Cd concentration found is close to the normal background concentration reported for wheat from the USA (Wolnik et al., 1983). The regional differences found by Andersson & Pettersson (1981) for the Cd levels in wheat in Sweden were attributed to soil factors, including Cd content, soil type, origin and formation.

In the present study an average Cd concentration of 0.026 was found in grains of barley. Wiersma et al. (1986) reported for Dutch barley an average Cd concentration of 0.13 mg/kg. The difference is too big to be explained by annual variations. The cause of this discrepancy is not clear. In both studies the same analytical procedure was used, while also no indications could be found that this difference is caused by differences in the selection of sample locations (soil type, region). The Cd concentrations reported for barley by Wiersma et al. (1986) are surprisingly high, also in view of the levels found in wheat. Smilde et al. (1982) demonstrated that the Cd accumulation in grain is higher in wheat than in barley. Consequently a random survey is expected to yield higher Cd concentrations for wheat. It is therefore rather surprising that Wiersma et al. (1986) found higher concentrations in barley.

Also for Pb in grains of wheat and barley Wiersma et al. (1986) found considerably higher concentrations compared to those found in the present study. Like for Cd the cause of this discrepancy is not known. The Pb concentrations found in the present study are slightly higher than those reported for cereals from other countries. This might be related to an elevated direct uptake of Pb from atmospheric deposition, due to the high traffic density in the Netherlands. The potential effect of motor traffic upon the Pb contamination of the environment is a well-known phenomenon. In 1978 the maximum permissible Pb concentration in petrol was set at 0.4 g/l. Before 1978 concentrations of ca 0.8 g/l were common. A further decrease of the Pb emission by motor traffic is expected in the near future.

In literature there has been some dispute about the question whether or not the Cd concentrations in cereal grains have increased during this century (Andersson & Bingefors, 1985; Kjellström et al., 1975; Jansson, 1980; Lorenz et al., 1986). The considerable annual fluctuations mentioned earlier strongly complicate the interpretation of data collected over long periods of time. In the Netherlands no data are available on the Cd and Pb levels in cereals from the first half of this century.

Conclusions

The Cd and Pb concentrations found in grains of cereals in the present study generally are low. For Cd in barley and Pb in barley and wheat the concentrations found are considerably lower than those found by Wiersma et al. (1986) in a previous Dutch survey investigation. The Cd and Pb levels in Dutch cereals are in good agreement with data from other countries, although the Pb concentrations in Dutch cereals tend to be slightly higher.

In general the concentrations found are well below the established provisional legal limits for Cd and Pb in cereal grains. Based on the results of this study it is not expected that the official introduction of these limit values, i.e. 0.15 mg/kg for Cd and 0.5 mg/kg for Pb, will lead to serious restrictions with respect to the cultivation in the Netherlands of cereals for human consumption.

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