Cadmium Exposure of the Greek Population

S. Karavoltsos, A. Sakellari, M. Scoullos

University of Athens, Chemistry Department, Division III, Inorganic and Environmental Chemistry and Technology, Panepistimiopolis 157 71, Athens, Greece

Received: 22 February 2003/Accepted: 3 July 2003

Due to the toxicity of cadmium and its compounds and to their ability to bioaccumulate in the tissues of living organisms, numerous systematic efforts are recently carried out, in order to replace cadmium in products where it has been frequently used, such as cadmium pigments, stabilizers, Ni-Cd accumulators, electroplating and coating, cadmium alloys and so forth (ERL 1990; Roskil 1995).

An extended study on the cadmium cycle in EU countries was recently carried out in the framework of a project regarding the EU Policies on Heavy Metals called EUPHEMET (Scoullos et al. 2001). In reviewing the available data, it became apparent that in the EU, as is the case for the rest of the world, adults are exposed to cadmium predominantly through the consumption of food and drinking water, smoking and inhalation (FAO/WHO 1989; Van Assche and Ciarletta 1993). Moreover, it was shown from recent data that the average dietary cadmium intake for adults in European countries ranges considerably from 7 to 44 μ g/d, with the highest value provided for Greece, exceeding to a large extent the values of other European countries (RAR 1999).

It was therefore initially decided to investigate the dietary cadmium intake of the Greek population by reconsidering the actual cadmium content of foodstuffs from the Greek market, as well as that of the current Greek diet, which is no longer purely Mediterranean, but has incorporated elements of both more exotic origins and-mainly-features of the modern 'western' dietary patterns. In order to estimate the total quantity of cadmium absorbed daily by the Greek population and since no relevant data were found in literature, the contribution of smoking, of the consumption of drinking water, as well as of inhalation are also evaluated herewith.

MATERIALS AND METHODS

The calculation of the daily dietary cadmium intake is based on very recent analytical data of our team concerning the cadmium content in foodstuffs from the Greek market (Karavoltsos et al. 2002). Dietary data were derived from literature featuring sociodemographic, educational background and income data of sample groups of the Greek population (Trichopoulou and Lagiou 1997).

The evaluation of the absorption of cadmium through the consumption of drinking water in Greece was carried out by the measurements of drinking water samples originating from approximately 160 different sampling points distributed throughout the entire country. Samplings were carried out between March 2001 and December 2002, from frequently used household taps, municipal and communal water supplies, after a minimum 3 minute flow of water through the piping system. During the collection, the samples were placed in polyethylene vials of 50 mL volume, previously cleaned with nitric acid 65% and they were acidified with the addition of a few drops of nitric acid 65% suprapure. The determinations of cadmium were performed directly (without any intermediate treatment) by flameless Atomic Absorption Spectrometry Varian SpectrAA 640Z with Zeeman background correction, with the implementation of the calibration method. The accuracy and repeatability of the measurements were assured by intralaboratory quality assurance exercises as well as by the analysis of a reference material (BCR No 609, Ground water low contents), for which a recovery of 100±1% was measured. The detection limit was calculated at 0.010 µg/L, according to Miller and Miller (1993).

Determinations of the cadmium content of 15 cigarette brands both Greek and international, sold in the Greek market, were also carried out. Determinations were carried out exclusively in the tobacco of each cigarette, since the levels of cadmium in the filter and the cigarette paper were non detectable in all of the cigarette brands analysed. Six samples of each cigarette brand were analysed. The method selected as appropriate for the analyses was wet digestion. Analytical methods, as well as quality assurance exercises performed, are described in detail elsewhere (Karavoltsos et al. 2002).

The contribution of inhalation to the cadmium intake of the Greek population is based on recent analytical data concerning cadmium levels in the ambient air of the inner city of Athens (Bakeas et al. *unpublished data*) and Thessaloniki (Voutsa et al. 2002; Voutsa and Samara 2002), which are the two largest cities of Greece, since corresponding data for the rest of the country are limited. The measurements refer to PM_{2.5}, PM_{7.5}, and PM₁₀ particles, which constitute the major fraction of particles deposited in the lungs, responsible for adverse respiratory health effects (Schwartz et al. 1996).

RESULTS AND DISCUSSION

Based on quite recent available sociodemographic nutritional data (Trichopoulou and Lagiou 1997), the daily dietary cadmium intake of the Greek population is calculated at 13.1 μ g/d and 13.3 μ g/d in urban and semi-urban areas respectively and at 16.1 μ g/d in rural areas (Table 1), whereas according to nutritional data for groups of different educational background (Table 2) the daily dietary intake is calculated at 14.6 μ g/d for elementary completed groups of the Greek population, at 12.8 μ g/d for secondary completed groups and at 12.6 μ g/d for groups of university educational level. The differences observed could be accounted for by the higher consumption of agricultural products mainly bread, cereals and vegetables in rural areas, dominated by groups of lower educational level.

Table 1. Average daily dietary cadmium intake according to sociodemographic features of Greek population groups and contribution of different food groups

to the total dietary cadmium intake.

	URBAN		SEMI-URBAN		RURAL	
Food Category	g food	μg Cd	g food	μg Cd	g food	μg Cd
Cereals	248	4.5	272	4.9	328	6.0
Meat	154	0.1	152	0.1	164	0.2
Offal	5.8	0.1	6.7	0.1	6.5	0.1
Fish	32	0.3	36	0.4	43	0.5
Seafood	3.8	0.4	4.2	0.5	4.8	0.6
Eggs	0.4	< 0.1	0.5	< 0.1	0.7	< 0.1
Dairy products	325	0.5	277	0.4	306	0.5
Fats and oils	70	0.2	86	0.2	114	0.3
Potatoes	148	3.3	133	3.0	180	4.0
Leafy vegetables	44	1.2	43	1.2	44	1.2
Other vegetables	203	2.0	195	1.9	224	2.2
Pulses	12	< 0.1	15	0.1	24	0.1
Fruits	350	0.5	328	0.5	323	0.4
Alcoholic	14	< 0.1	20	< 0.1	29	< 0.1
beverages						
Total		13.1		13.3		16.1

Table 2. Average daily dietary cadmium intake according to the level of education of Greek population groups and contribution of different food groups to the total dietary cadmium intake.

	ELEMENTARY		SECONDARY		UNIVERSITY	
	COMPI	COMPLETED		COMPLETED		
Food Category	g food	μg Cd	g food	μg Cd	g food	μg Cd
Cereals	287	5.2	234	4.3	218	4.0
Meat	157	0.2	152	0.1	156	0.2
Offal	6.3	0.1	6.0	0.1	6.1	0.1
Fish	37	0.4	31	0.3	34	0.4
Seafood	4.3	0.5	3.7	0.4	3.8	0.4
Eggs	0.5	< 0.1	0.4	< 0.1	0.6	< 0.1
Dairy products	302	0.5	341	0.5	364	0.6
Fats and oils	90	0.2	69	0.2	63	0.2
Potatoes	163	3.6	146	3.3	134	3.0
Leafy vegetables	44	1.2	42	1.2	41	1.2
Other vegetables	213	2.1	197	1.9	194	1.9
Pulses	18	0.1	11	< 0.1	9.6	< 0.1
Fruits	321	0.5	364	0.5	404	0.6
Alcoholic	19	< 0.1	15	< 0.1	17	< 0.1
beverages						
Total		14.6		12.8		12.6

The mean value of daily dietary cadmium intake in Greece, calculated at 13.8 μ g/d in the present study is significantly lower than the values reported previously

ranging between 44.5 and 56.1 μ g/d (Tsoumbaris and Tsoukali-Papadopoulou 1994) and comparable to the relevant mean values of other European countries provided in literature, ranging between 2.4 and 30 μ g/d (Table 3).

Table 3. Comparison of the average daily dietary cadmium intake among

European countries.

European coun	urics.	
Country	Daily intake	Reference
	(µg Cd/d)	
Greece	13.8 (12.6-16.1)	Present study
	44.5-56.1	Tsoumbaris and Tsoukali-Papadopoulou 1994
Belgium	15	Nasreddine and Parent-Massin 2002
Denmark	15.0	Bro et al. 1990
Finland	13.0	Nasreddine and Parent-Massin 2002
France	30.0	Biego et al. 1998
Germany	2.4-27.6	Wilhelm et al. 2002
Ireland	22.6	EUR 17527 1997
Italy	14.8-20.1	Alberti-Fidanza et al. 2002
The Netherlands	21.0	Nasreddine and Parent-Massin 2002
Portugal	16.9	EUR 17527 1997
Sweden	11 (5.7-26)	Berglund et al. 1994
Spain	16-29	Cuadrado et al. 1995
<u>Û. K.</u>	14	Nasreddine and Parent-Massin 2002

Cadmium is absorbed by the gastrointestinal tract in a percentage equal to approximately 5% (Elinder and Jarup 1996). Considering this percentage, the average amount of cadmium absorbed through the consumption of food per person in Greece is estimated at $0.7 \,\mu\text{g/d}$, ranging from $0.6 \,\text{to} \, 0.8 \,\mu\text{g/d}$.

In approximately 500 samples of potable water analysed, the concentrations of cadmium range from non detectable levels to 0.84 μ g/L with a mean concentration calculated at 0.06 μ g/L and the 90th percentile value at 0.14 μ g/L. All the values detected were considerably lower than the maximum level imposed by legislation, set at 5.0 μ g/L (EEC 1998).

According to U.S. EPA (1988) recommendations, the estimation of the amount of cadmium ingested daily through the consumption of drinking water is based on the assumption of a reference body weight of 60 kg for adults and a reference value of 2 L/d for water consumption. Taking these data into consideration, the average cadmium daily intake from water consumption of the Greek population is calculated at 0.12 μ g/d, ranging from non detectable levels to 1.7 μ g/d. Considering an intestinal absorption of cadmium from drinking water equivalent to 5% (Elinder and Jarup 1996), a value which may increase in case of iron, calcium or protein deficiency (EC 2000), the mean net amount of cadmium absorbed daily is estimated at 0.01 μ g/d, varying from non detectable to 0.08 μ g/d.

The cadmium concentrations of both Greek and international cigarette brands sold in the Greek market were found to range from 0.2 to 1.6 μg/cigarette (Figure 1),

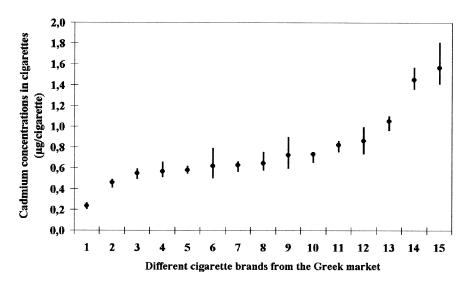


Figure 1. Cadmium concentrations (average, minimum and maximum) in various cigarette brands from the Greek market.

being comparable to the results of other studies (Elinder and Jarup 1996), as well as to those provided by Kalaitzoglou and Samara (1999) examining the cadmium content of numerous brands sold in the Greek market.

It has been suggested that approximately 10% of the cadmium contained in a cigarette is finally inhaled and it is estimated that 25-50% of the inhaled cadmium is finally absorbed (Sánchez et al. 1995). Assuming smoking a pack of 20 daily, the amount of cadmium inhaled by an average Greek ranges from 0.5 to 3.1 μ g/d, depending on the brands used, with a mean value of 1.5 μ g/d. Consequently, the net absorption of cadmium in body tissues is equal to 0.2 - 1.6 μ g/d, with a mean value of 0.8 μ g/d, i.e. an amount almost equivalent to that absorbed *via* food, in Greece.

The mean concentrations of cadmium in the ambient air of the two cities of Athens (Bakeas et al. *unpublished data*) and Thessaloniki (Voutsa et al. 2002; Voutsa and Samara 2002), has been calculated at 1.23 ng/m³, ranging from 0.10 to 7.2 ng/m³ referring to a total of 439 samples. According to U.S. EPA (1988) recommendations, a reference body weight of 60 kg for adults and a reference respiratory volume of 20 m³/d are assumed in order to calculate the cadmium intake from inhalation. The total absorption of cadmium includes the deposition rate, which varies between 10% and 50%, depending on particle size and the pulmonary absorption, which varies with the chemical nature of particles and for which an upper state estimate of 100% is a reasonable worst case assumption. For the calculations carried out in the present study, an average value of 50% for the total absorption of cadmium through inhalation was used (Van Assche and Ciarletta 1993; EC 2000). Consequently, the average contribution of inhalation to

the total cadmium daily intake of the Greek population is estimated at 0.02 $\mu g/d$, varying between 0.002 and 0.14 $\mu g/d$, imposing a lung absorption of cadmium varying between 0.001 and 0.07 $\mu g/d$, with a mean value equal to 0.01 $\mu g/d$.

In the current survey, the average total amount of cadmium ingested daily by the Greek population, through diet, drinking water consumption, smoking and inhalation pathways is calculated to range from 12.6 to 17.9 μ g/d for non smoking groups and from 13.1 to 21.0 μ g/d for smoking ones. Furthermore, assuming a body weight of 60 kg (EPA 1988), the total intake of cadmium for the Greek population is estimated to vary between 1.5 and 2.1 μ g/kg body wt/wk for non smokers and between 1.5 and 2.4 μ g/kg body wt/wk for smokers. Compared to the Provisional Tolerable Weekly Intake (PTWI) of cadmium set at 7 μ g/kg body wt/wk (FAO/WHO 1989) and comprising all the possible sources of the metal (Cuadrado et al. 1995), the previous values of cadmium uptake are lower for both smokers and non smokers. The average total amount of cadmium absorbed by the Greek population is estimated to range from 0.6 to 1.0 μ g/d for non smokers and from 0.8 to 2.5 μ g/d for smokers (Table 4).

Table 4. Estimation of the amount of cadmium absorbed daily in Greece, in $\mu g/d$, (% of total).

	1 1	•	Manage	nalrina manu	lation		
Sme	oking populat		Non si				
Low	Average	High	Low	Average	High		
(%)	(%)	(%)	(%)	(%)	(%)		
0.6	0.7	0.8	0.6	0.7	0.8		
(72)	(47)	(32)	(100)	(97)	(84)		
n.d.	0.01	0.08	n.d.	0.01	0.08		
	(0.6)	(3.2)		(1.5)	(8.4)		
0.2	0.8	1.6	-	-	-		
(28)	(52)	(62)					
0.001	0.01	0.07	0.001	0.01	0.07		
(<0.1)	(0.4)	(2.8)	(<0.1)	(1.5)	(7.6)		
0.83	1.49	2.52	0.60	0.72	0.95		
	Sme (%) 0.6 (72) n.d. 0.2 (28) 0.001 (<0.1)	Smoking popular Low Average (%) (%) 0.6 0.7 (72) (47) n.d. 0.01 (0.6) 0.2 0.8 (52) 0.001 0.01 (<0.1)	Smoking population Low Average High (%) (%) (%) 0.6 0.7 0.8 (72) (47) (32) n.d. 0.01 0.08 (0.6) (3.2) 0.2 0.8 1.6 (28) (52) (62) 0.001 0.01 0.07 (<0.1)	Smoking population Non sr Low Average High Low (%) (%) (%) (%) 0.6 0.7 0.8 0.6 (72) (47) (32) (100) n.d. 0.01 0.08 n.d. (0.6) (3.2) 0.2 0.8 1.6 - (28) (52) (62) 0.001 0.001 0.001 (<0.01)	Low Average (%) High (%) Low (%) Average (%) (%) (%) (%) (%) (%) 0.6 0.7 0.8 0.6 0.7 (72) (47) (32) (100) (97) n.d. 0.01 0.08 n.d. 0.01 (0.6) (3.2) (1.5) 0.2 0.8 1.6 - - (28) (52) (62) 0.001 0.01 0.07 0.001 0.01 (<0.1)		

In the case of smoking population, nutrition and smoking constitute the main sources of cadmium, while under average circumstances the contributions of these two pathways are almost equivalent. However, under high uptake circumstances, the inhalation of cigarette smoke may represent the dominant factor for the absorbed amount of cadmium. For the non smoking population, the uptake of cadmium through the consumption of food contributes the largest amount to the total daily dose. In all cases, cadmium uptake *via* inhalation of ambient air contributes less than 8% to the total absorbed cadmium daily dose.

Acknowledgments. This research has been carried out in the framework of the project 'Towards an Integrated EU Policy for HEavy METals' (EUPHEMET), European Commission (contract no. ENV4-CT97-0614). The authors greatly acknowledge Assistant Professor M. Dassenakis for his support. Professor P.

Siskos and Dr. E. Bakeas for kindly providing unpublished analytical data on ambient air.

REFERENCES

- Alberti-Fidanza A, Burini G, Perriello G (2002) Trace elements in foods and meals consumed by students attending the faculty cafeteria. Sci Total Environ 287:133-140.
- Berglund M, Akessson A, Nermell B, Vahter M (1994) Intestinal Absorption of Dietary Cadmium in Women depends on Body Iron Stores and Fibre Intake. Environ Health Perspect 102:1058-1066.
- Biego GH, Joyeux M, Hartemann P, Debry G (1998) Daily intake of essential minerals and metallic micropollutants from foods in France. Sci Total Environ 217:27-36.
- Bro S, Sandström B, Heydorrn K (1990) Intake of essential and toxic trace elements in a random sample of Danish men as determined by the duplicate portion sampling technique. J Trace Elem Electr Health Dis 4:147-155.
- Cuadrado C, Kumpulainen J, Moreiras O (1995) Lead, cadmium and mercury contents in average Spanish market basket diets from Galicia, Valencia, Andalucía and Madrid. Food Addit Contam 12:107-118.
- EC (2000) Ambient air pollution by As, Cd and Ni compounds. Working Group on Arsenic, Cadmium and Nickel Compounds, European Commission, DG Environment, Position Paper, final version, October.
- EEC (1998) Council directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. Official Journal L 330: 32-54.
- Elinder CG, Jarup L (1996) Cadmium exposure and health risks: recent findings. Ambio 25: 370-373.
- EPA (1988) Recommendations and Documentation of Biological Values for Use in Risk Assessment. U.S. Environmental Protection Agency, Washington, DC, USA.
- ERL (1990) Environmental evaluation of the sources of human and environmental contamination by cadmium. Commission of the European Communities, Directorate General for Environment, Consumer Protection and Nuclear Safety. Final Report.
- EUR 17527 (1997) Dietary exposure to cadmium. European Commission, DG 3. Office for Official Publications of the European Communities, Luxembourg.
- FAO/WHO (1989) Evaluation of certain food additives and contaminants. Thirty-third report of the joint FAO/WHO expert committee on food additives. World Health Organization, Technical Report Series 776, WHO, Geneva.
- Kalaitzoglou M, Samara C (1999) Yields of cadmium, tar, nicotine and carbon monoxide in mainstream smoke of Greek cigarettes: a comparative study. Beitr Tabakforsch Int 18:235-244.
- Karavoltsos S, Sakellari A, Dimopoulos M, Dasenakis M, Scoullos M (2002) Cadmium content in foodstuffs from the Greek market. Food Addit Contam 19:954-962.
- Miller JC, Miller JN (1993) Statistics for analytical chemistry 3rd ed. Ellis Horwood PTR Prentice-Hall, New York.

- Nasreddine L, Parent-Massin D (2002) Food contamination by metals and pesticides in the European Union. Should we worry? Toxicol Lett 127:29-41.
- RAR (1999) Risk assessment cadmium. CAS-No.: 7440-43-9. Draft text.
- Roskill Information Services Ltd (1995) Cadmium market update, analysis and outlook. Roskill, London.
- Sánchez FG, Diaz AN, Arbaizar A (1995) Determination of cadmium in tobacco smoke and zinc in tap water by solvent extraction atomic absorption spectrometry. Mikrochim Acta 118:265-272.
- Scoullos M, Vonkeman G, Thornton I, Makuch Z (2001) Cadmium. In: Scoullos M (ed) Mercury- Cadmium Lead, Handbook for Sustainable Heavy Metals Policy and Regulation. Kluwer Academic Publishers, Dordrecht, p 71.
- Schwartz J, Dockery DW, Neas LM (1996) Is daily mortality associated specifically with fine particles? J Air Waste Manage Assoc 46:927-939.
- Trichopoulou A, Lagiou P (1997) Methodology for the exploitation of HBS food data and results on food availability in 5 European countries, Vol 1. DG for Science, Research and Development, EUR 17909, European Commission.
- Tsoumbaris P, Tsoukali-Papadopoulou H (1994) Heavy metals in common foodstuffs: Daily intake. Bull Environ Contam Toxicol 53:61-66.
- Van Assche F, Ciarletta P (1993) Environmental exposure to cadmium in Belgium: decreasing trends during the 1980s. In: Heavy metals in the environment. CEP consultants, Publ. Edinburg, United Kingdom.
- Voutsa D, Samara C (2002) Labile and bioaccessible fractions of heavy metals in the airborne particulate matter from urban and industrial areas. Atmos Environ 36:3583-3590.
- Voutsa D, Samara C, Kouimtzis Th, Ochsenkühn K (2002) Elemental composition of airborne particulate matter in the multi-impacted urban area of Thessaloniki, Greece. Atmos Environ 36:4453-4462.
- Wilhelm M, Wittsiepe J, Schrey P, Budde U, Idel H (2002) Dietary intake of cadmium by clhidren and adults from Germany using duplicate portion sampling. Sci Total Environ 285:11-19.