González-Muñoz, M.J., Peña, A. and Meseguer, I. (2008) 'Monitoring heavy metal contents in food and hair in a sample of young Spanish subjects', Food and chemical toxicology, 46(9), pp. 3048–3052.

Available at https://doi.org/10.1016/j.fct.2008.06.004

Monitoring heavy metal contents in food and hair in a sample of young Spanish subjects

M.J. González-Muñoz *, A. Peña, I. Meseguer

Departamento Nutrición, Bromatología y Toxicología, Facultad de Farmacia, Universidad de Alcalá, Crta, Madrid – Barcelona Km. 33.6, 28871 Alcalá de Henares, Madrid, Spain.

ABSTRACT

For most people the main route of exposure to the toxic elements is through the diet. Consequently, information concerning dietary intake is of the utmost importance in being able to assess risks to human health. The goal of this study was to intend to assess the usefulness of hair as a biomonitor of the mineral status in young adults. Daily intakes of selected toxic and essential mineral elements were evaluated using a food frequency questionnaire. In addition, the levels of these same elements in hair samples were measured by inductively coupled plasma mass spectrometry and inductively coupled plasma atomic emission spectrometry. The contents of the essential elements in the study population were all well above Spanish recommendations for adult males and females. The estimated intakes of toxic elements were appreciably below the respective PTWIs, indicating that these intake levels do not pose a health concern for this group. Significant differences in hair metal levels were observed between the men and the women, who were in the same age group. Interestingly, no correlation was found between trace element intakes and the corresponding levels in the hair. In conclusion, hair is only limited usefulness as a means of estimating the nutritional status of the essential and toxic elements considered.

INTRODUCTION

Some metals and their compounds are essential to human health (i.e. Fe, Zn, Cr), although they are potentially harmful if consumed in large quantities. Other metals may be harmful to health, for example, As, Pb, Cd and Hg have no known beneficial biological function and long-term exposure may be toxic even at low doses. Although some individuals are exposed to toxic elements chiefly in the workplace, for most people the main route of exposure to the toxic elements is through the diet. Consequently, information concerning dietary intake is of the utmost importance in being able to assess risks to human health (Leblanc et al., 2000).

This type of information is usually compiled by means of previously validated food surveys. The mineral status of individuals has conventionally been determined by analysis of biological samples, basically blood samples.

However, human scalp hair has gained considerable ground in recent years for use as a biomonitor of trace elements to estimate environmental exposure levels and assess nutritional status, as well as for use as a means of diagnosing illness (Sanna et al., 2003; Dunicz-

Sokolowska et al., 2006). As a metabolically inactive tissue, hair has become well established, especially for investigating levels of and changes in many trace elements that accumulate in the body (Druyan et al., 1998). Furthermore, hair levels are less sensitive to immediate intake and could therefore also be a good biological indicator of the nutritional status of certain elements.

However, alongside the potential advantages, hair analysis also has a number of disadvantages, for example, contamination by dust and sweat or the influence of age, sex, and place of residence (Benes et al., 2003).

The aim of the present study is triple: (a) to monitor the dietary food mineral content; thus, to assess the mineral consumption; (b) to monitor the hair mineral content; and (c) to study the possible relationship between both mineral contents.

Materials and methods

The study population consisted of 350 students between 20 and 24 years of age from different towns in the Autonomous Community of Madrid (Spain) attending the University of Alcalá. Subjects were selected by means of cluster sampling. The study was performed following guidelines of Helsinki Declaration.

Participation was restricted to healthy non-smokers who did not use any hair treatment (stains, fixers, permanents waves, etc.), given the influence of these fac- tors on the utility of hair as a biomonitor (Sukumar, 2002).

Dietary mineral content and consumption

To assess the mineral content obtained by the study population from the diet, subjects were administered a semi-quantitative food frequency questionnaire de- signed specifically for this study, which called for detailed answers by subjects on the types of foods and quantities eaten over the course of a week. The food frequency questionnaire included estimates of small, medium and large portion sizes, contained 110 foods and was previously validated and involved 24-hour re- call carried out on alternate days, three days a week.

The daily intake of the elements considered in the different food groups was calculated taking into account food content and consumption. Toxic element concentrations in the different food groups were taken from the specialized literature (Moreiras and Cuadrado, 1992; Llobet et al., 2003). Information on the nutrients Fe, Cr, and Zn came from the Spanish Food Composition Tables (Moreiras et al., 2006) and from The Composition of Food (Mc Cance, 1980) and Food Composition and Nutrition Tables (Souci et al., 1981).

Mineral element concentrations in hair samples

Samples collection

Scalp hair samples weighing approximately 1.0 g were taken from the occipital region of the head, using a stainless steel scissors without vanadium, and stored in plastic bags. To prepare the samples for analysis, the hairs were cut into 5-mm pieces and washed according to the procedure recommended by the IAEA Advisory Group (acetone, three times water, acetone) to remove surface dirt and grease (Nowak, 1998). The washed samples were dried at about 50 ºC to constant weight (Schlegel-Zawadska, 1992).

Analytical methods

The mineral elements considered in this study were Fe, Zn, Pb, Sn, Cr, Mn, Ni, Cd, Hg, and As.

The mineral content was measured by inductively coupled plasma mass spectrometry (ICP-MS) and by inductively coupled plasma atomic emission spectrometry (ICP-AES) following wet ashing of the organic matter according to the method of Granero et al. (2004). Each sample (2.5 ml) was digested in 2 ml of 65% nitric acid (Suprapur, Merck, Darmstadt, Germany) at room temperature in Teflon bombs for 8 h and subsequently heated at 100 ºC for 12 h. After cooling, the solutions were filtered and made up to 25 ml with deionized water. The accuracy of the instrumental methods was validated by replicating all samples as well as by taking measurements of a reference material (lobster hepatopancreas, NRC Canada TORT-2) every ten samples. Quantification was based on the most abundant isotope of each element free of analytical interference. Mean recovery rates were between 90% and 95%.

The Fe and Zn contents were measured by ICP-AES (Perkin Elmer Optima 3200 RL) using the Fe emission line of 259.939 nm and the Zn emission line of 213.857 nm. The levels of Mn, Cr, Pb, Sn, Ni, Cd, Hg, and As were measured by ICP-MS (Perkin Elmer Elan 6000) using the isotopes Mn55, Cr52, Pb208, Sn118, Ni60, Cd114, Hg202, and As75. The emission lines and isotopes used are free of spectral interference for this matrix type, and therefore, have not been necessary to perform any corrections.

Statistical analysis

The statistical significance of the data was computed by one-way analysis of variance (ANOVA). The Kolmogorov–Smirnov test was used to confirm that the values were normally distributed, while homogeneity of the variances was assessed using Snedecor’s F-test. In addition, the LSD multiple rank tests was used to deter- mine which means differed significantly from the others using a significance level of 0.05% or less.

Pearson’s correlation coefficient was employed to provide a technical description of the correlations between data obtained by different methods of measurement.

Results and discussion

Table 1 summarizes the anthropometrical characteristics and energy and macronutrient intakes of the studied population. These individuals followed a low-carbohydrates (~42%), high-protein (~20%), and high-fat (~38%) diet, as is usual in industrialized countries.

Higher energy intakes were observed in men, mainly because they ate larger amounts of carbohydrates and fats, although the data were not statistically different.

Mineral element intake through the diet

The intake of the essential elements (Table 3) in the studied population was in all cases well above the recommended values for adult males and females in Spain (Moreiras et al., 2006). These levels were higher to the values reported by Bocio et al. (2005) for the general population of Tarragona (Spain): Cr, 88.3 lg/d; Mn, 2

421.4 lg/d; Ni, 138.3 lg/d; and Sn, 34.6 lg/d. The differences could be related to substantial differences in the quantity and quality of dietary food among the different Spanish regions (Moreiras and Cuadrado, 1992).

Significantly different intakes between the men and the women were recorded for Cd, Mn, Hg, and Sn. Men exhibited significantly higher intake of Cd than women ($p < 0.01$). On examining food groups consumption levels (Table 2), men can be seen to eat significantly ($p < 0.05$) more cereals than women; cereals are the food group that contributed the most to the dietary intake of Cd (Llobet et al., 2003). Similarly, since cereal grains, nuts, fruits and tea are sources of Mn, the higher cereal consumption could also account for the higher Mn intake in men.

Fish and shellfish are the main food group responsible for intake of the heavy metal Hg, which is directly related to fish consump- tion (Moreiras and Cuadrado, 1992). Men ate significantly ($p < 0.05$) more fish than women, which could account for the higher intake of this heavy metal in men.

Sn intake depends basically (98%) on eating food preserved in tin cans (Biego et al., 1999). The higher intake of this metal in men could therefore be indicative of higher consumption of this type of foods by men.

By way of assessing the health risks attaching to the above in- take levels, the values estimated were compared to the current provisional tolerable weekly intakes (PTWI) for these elements (FAO/WHO, 1993). The daily intake estimates of the non-essential (toxic) elements As, Sn, Cd, Hg, and Pb were similar to or somewhat lower than their respective PTWIs and showed good agreement with other country studies.

As intake was higher in men at 220.85 lg/d, compared with 192.43 lg/d in women. The PTWI for inorganic As is 15 lg/kg of body weight/week, or 128 lg/d (FAO/WHO, 1993), and these levels were found to be exceeded in the present study. According to the US food and drug administration (FDA) total diet study (Adams et al., 1994) food contributes 93% of the total As intake, with sea- food contributing 90 % of that amount. The percentage of inorganic As in fish and shellfish to be between 0.02% and 11% (Muñoz et al., 2000), and the maximum acceptable daily load of As to be 3000 lg for a subject weighing 60 kg (FAO/WHO, 1989), the estimated As intake in the present study in fact turns out to be lower than the PTWI. The total As intake was lower than that in traditional fish- consuming regions as the Basque Country in Spain (Jalón et al., 1997) and Japan (Tsuda et al., 1995), where it attained values of 297 lg/d and 280 lg/d, respectively. In another studies on Spain, the dietary intake of As was similar: 223.6 lg/d in Catalonia (Llobet et al., 2003) and 221 lg/d (Delgado-Andrade et al., 2003).

The PTWI for Cd is 60 lg/d for an individual weighing 60 kg. In the present survey the Cd intake ranged between 27.8% of the PTWI in men and 20.63% of the PTWI in women. Other studies have reported the following Cd intakes: 14.3 lg/day (Bocio et al., 2005) for subjects living in Catalonia; 12 lg/d in Britain (Ysart et al., 2000); 11–29 lg/d in the Basque Country (Jalón et al., 1997); 3.6 lg/d in France (Noel et al., 2003) 15 lg/d in Denmark (Larsen et al., 2002); and 22.2 lg/d in Germany (Wilhelm et al., 2002).

The average consumption of Hg often tends to vary more widely in regions where pollution is low. A PTWI of 5 lg/kg of body weight was established for Hg by FAO/WHO (1993) (43 lg/d/60 kg.) In the current study men's intake represented the 55.19% of PTWI and women's intake the 42.82% of the PTWI. Intake levels were higher than those reported for other industrialized countries, e.g., 4.8 lg/d in the United Kingdom (Ysart et al., 2000), 3 lg/d in

Denmark (Larsen et al., 2002), 15.3 lg/d in Japan (Iwasaki et al., 2003), and 9 lg/d in France (Noel et al., 2003) a and similar to the levels found by the Tarragona study (Llobet et al., 2003).

The Pb intake was 11.26% of the PTWI in men and 9.57% of the PTWI in women (25 lg/d/60 kg). This average Pb intake is similar to or lower than the levels recently reported for other parts of Europe: 26 lg/d in the United Kingdom (Ysart et al., 2000), 52 lg/d in France (Leblanc et al., 2000) and 18 lg/d in Denmark (Larsen et al., 2002). The daily amount of lead in the diet in different parts of Spain has been reported to range between 37 and 521 lg/d, (Llobet et al., 2003; Bocio et al., 2005; Jalón et al., 1997). Specially, Madrid area had the highest average intake of this

metal, exceeding the PTWI, because of the vegetables and cereals consumed (Cuadrado et al., 2005).

For the rest of the elements considered, intake levels were sub- stantially below the normal PTWIs. Accordingly, the estimated intakes for all the elements consid- ered in this study were substantially lower than the respective PTWIs, which indicates that these intakes do not represent a health concern for this population.

Hair levels

The values for the mineral elements recorded in the hair samples from the study population (Table 4) were within what can be considered the normal range of values. This is an important finding, since according to Dunicz-Sokolowska et al. (2006a), bioelement concentration values in hair outside the reference ranges in young people could be a sign of disturbances leading to various diseases. Thus, deficiencies in essential trace metals and high levels of toxic metals could play a role in the developments of heart dis- ease. Likewise, toxic metals could also reduce the absorption of essential elements (Afridi et al., 2006).

The results for the levels of toxic elements in the hair yielded mean hair As levels of 0.011 ppm in women and low levels below the detection threshold in men. These values were within the range of allowable values (0.08–0.25 mg As/kg hair) and were somewhat lower than in most developed countries. Fish consumption was inversely related to As consumption (0.31 lg/g) in Pakistan (Anwar, 2005) as was the case in our study, suggesting other sources of As exposure among the population (e.g, beans, main in Pakistan).

Cd levels were lower than those reported for developing countries, including Japan and Pakistan (0.08 lg/g), while the Pb levels observed in the present study were similar to those reported for developed countries (Anwar, 2005).

The US Environmental Protection Agency and the National Academy of Sciences recommend keeping hair Hg levels at <1.0 lg/g, corresponding to a reference dose (RfD) of 0.1 lg/kg body weight per day (Senofonte et al., 2000). In the present study, the values exceed the RfD, although they were below the values re- ported by others, e.g., 2.0–89 lg/g in the US (Hightower and Moore, 2003). Hair Hg levels have been associated with age and

fish consumption frequency (McDowell et al., 2004).

Zn levels in hair were similar to those found by Schlegel- Zawadzka et al. (2002) in healthy youngsters (182.98 lg/g in boys and 203.82 lg/g in girls). Gender did not influence this parameter, in similar way that has been referred for Italian population (Bertazzo et al., 1996). Therefore, the relationship of the Zn content of food, food frequency, and the concentration in the different tis- sues of healthy children could be influenced by a variety of internal and external factors (Schlegel-Zawadzka et al., 2002).

Looking at the influence of gender on mineral element concentrations in the hair revealed higher levels in the women compared to the men, with the differences being significant for Pb ($p < 0.05$), Sn ($p < 0.001$), Cr ($p < 0.001$), Mn ($p < 0.05$), Ni ($p < 0.05$), and As ($p < 0.001$). In contrast, the Cd content was significantly higher in men ($p < 0.01$). Significant differences in the hair metal contents in girls and boys of 10 and 20 years were observed by Dunicz- Sokolowska et al. (2006b) in a Polish population and Benes et al. (2003) in the Czech Republic. This finding cannot yet be conclusively explained; perhaps, it is caused by still unknown hormonal influences. In a study of the Pakistani population ranging in age be- tween 3 and 100, Khalique et al. (2005) found that the hair from females exhibited higher levels of all the metals considered except Fe and Co as compared with their male counterparts. This study also showed that in the case of males, except for Cu, Co, and Cr, metal concentrations decreased with age. However in females, hair metal levels rose with age. This accumulation over time could help explain the higher levels recorded for the women here.

On the other hand, metals levels in hair suggest not to be affected by body fat of participants because there were no significant correlations between both parameters (data not shown). Usually, these elements tender to accumulate in liver, kidney, blood and bones and therefore, fat does not constitute a target organ.

It should be noted that the mineral element concentrations obtained were not in keeping with the intake levels recorded previously. Men had higher intake levels for most of the elements studied but lower levels of these same elements in their hair. In fact, on examining the correlation between intake levels and hair levels (Table 5), correlations were only found for Cd (-0.457). Consequently, the mineral element contents in the hair do not reflect

the intake of these elements in the food. Element concentrations in the hair are highly variable on account of a variety of factors, e.g., age, income, dietary habits, and environmental status (e.g. food, air, water, soil) (Nowak and Chmielnicka, 2000). Interestingly, no correlation was observed between the intake of trace elements and the corresponding serum values in a one year study of trace element intake and status performed on Italian subjects. It was concluded that trace element intake should be assessed by chemical analysis of the food (Alberti-Fidanza et al., 2003).

Ca, Cu, Fe, Mg, P, Se, and Zn concentrations in the hair of Japanese do not significantly correlate with those relative concentrations in the internal organs (Yoshinaga et al., 1990). These findings indicate the limited usefulness of hair as a means of estimating the nutritional status of the essential elements examined.

On the other hand, correlation analysis yielded significant ($p < 0.05$) synergistic interactions between the bioelements Zn– Mn (0.944), Zn–Ni (0.972), Mn–Ni (0.917), and Ni–Fe (0.922). There

was a significant ($p < 0.05$) antagonistic interaction of the bioelement Cr with the toxic metal Hg (-0.916), while other bioelements and toxic elements had significant synergistic relationships ($p < 0.05$), i.e., Cd–Ni (0.967), Cd–Mn (0.879), Cd–Zn (0.986), Zn–Pb (0.944), Pb–Cd (0.985), Pb–Ni (0.937), and Pb–Cr (0.900). These interactions probably indicate that mineral balances in the body are regulated by major homeostatic mechanisms in which toxic elements compete with the essential metals, even at low levels of metal exposure. Knowledge of these correlations may be basic to gaining an understanding of the kinetic interactions between metals and their repercussions on the trace metal metabolism (Lopez Alonso et al., 2004).

Conclusion

The intake of essential trace elements in total diet samples of young adults living in the Madrid region in Spain fulfilled recommended dietary levels. The intake of As and the toxic heavy metals Pb, Cd, and Hg do not represent a health concern for this population.

Mineral element levels in the hair of this study population were within the considered normal range, and significant differences were observed between both sexes. An analysis of the correlations between mineral element intake and hair levels indicated that hair levels cannot be regarded as a suitable biological indicator of the mineral nutritional status and exposure to toxic elements through the diet. However, more studies are needed to assess the useful- ness of hair as a marker of mineral consumption.

Conflict of interest statement

The authors declare that there are no conflicts of interest.

References

Adams, M.A., Bolger, P.M., Gunderson, E.L., 1994. Dietary intake and hazards of arsenic. In: Chappell, W.R., Abernathy, C.O., Calderon, R.A. (Eds.), Arsenic: Exposure and Health, Science and Technology Letters. Northwood, UK. pp. 51– 58.

Afridi, H.I., Kazi, T.G., Kazi, G.H., Jamali, M.K., Shar, G.Q., 2006. Essential trace and toxic element distribution in the scalp hair of Pakistani myocardial patients and controls. Biol. Trace Elem. Res. 113, 19–34.

Alberti-Fidanza, A., Burini, G., Perriello, G., Fidanza, F., 2003. Trace element intake and status of Italian subjects living in the Gubbio area. Environ. Res. 91, 71–77. Anwar, M., 2005. Arsenic, cadmium and lead levels in hair and toe nail samples in

Pakistan. Environ. Sci. 12, 71–86.

Benes, B., Sladka, J., Spevackova, V., Smid, J., 2003. Determination of normal concentration levels of Cd, Cr, Cu, Hg, Pb, Se and Zn in hair of the child population in the Czech Republic. Cent. Eur. J. Publ. Health 4, 184–186.

Bertazzo, A., Costa, C., Biasiolo, M., Allegri, G., Cirrincione, G., Presti, G., 1996. Determination of copper and zinc levels in human hair: influence of sex, age, and hair pigmentation. Biol. Trace Elem. Res. 52, 37–53.

Biego, G.H., Joyeux, M., Hartemann, P., Debry, G., 1999. Determination of dietary tin intake in an adult French citizen. Arch. Environ. Contam. Toxicol. 36, 227–232.

Bocio, A., Nadal, M., Domingo, J.L., 2005. Human exposure to metals through the diet in Tarragona, Spain: temporal trend. Biol. Trace Elem. Res. 104, 193–201.

Cuadrado, C., Kumpulainen, J., Moreiras, O., 2005. Lead, cadmium and mercury contents in average Spanish market basket diets from Galicia, Valencia, Andalucia and Madrid. Food Addit. Contam 12, 107–118.

Delgado-Andrade, C., Navarro, M., López, H., López, M.C., 2003. Determination of total arsenic levels by hydride generation atomic absorption spectrometry in foods from South-East Spain: estimation of daily dietary intake. Food. Addit. Contam. 20, 923–932.

Druyan, M.E., Bass, D., Puchyr, R., 1998. Determination of reference ranges for elements in human scalp hair. Biol. Trace Elem. Res. 62, 183–187.

Dunicz-Sokolowska, A., Radomska, K., Dlugaszek, M., Graczyk, A., 2006a. Contents of bioelements and toxic metals in the Polish population determined by hair analysis. Part 1. Children aged 1–10 years. Magnes. Res. 19, 35–45.

Dunicz-Sokolowska, A., Graczyk, A., Radomska, K., Dlugaszek, M., Wlazlak, E., Surkont, G., 2006b. Contents of bioelements and toxic metals in a Polish population determined by hair analysis. Part 2. Young persons aged 10–20 years. Magnes. Res. 19, 167–179.

FAO/WHO, 1989. Evaluation of certain food additives and contaminants. Technical Report Series 759. World Health Organization, Geneva, Switzerland.

FAO/WHO, 1993. Evaluation of certain food additives and contaminants. Technical Report Series 837. World Health Organization, Geneva, Switzerland.

Granero, S., Vicente, M., Aguilar, V., Martínez-Para, M.C., Domingo, J.L., 2004. Effects of beer as a source of dietary silicon on aluminium absorption and retention in mice. Trace Elem. Electrol. 21 (1), 28–32.

Hightower, J.M., Moore, D., 2003. Mercury levels in high-end consumers of fish.

Environ. Health Perspect. 111, 604–608.

Iwasaki, Y., Sakamoto, M., Nakai, K., Oka, T., Dakeishi, M., Iwata, T., Satoh, H., Murata, K., 2003. Estimation of daily mercury intake from seafood in Japanese women: Akita cross-sectional study. Tohoku J. Exp. Med. 200, 67–73.

Jalón, M., Urieta, I., Macho, M.L., Azpiri, M.O., 1997. Metales pesados y arsénico. In: Vigilancia de la contaminación química de los alimentos en la Comunidad Autónoma del País Vasco 1990–1995. Servicio Central de Publicaciones del Gobierno Vasco, Vitoria, Spain.

Khalique, A., Ahmad, S., Anjum, T., Jaffar, M., Shah, M.H., Shaheen, N., Tariq, S.R., Manzoor, S., 2005. A comparative study based on gender and age dependence of selected metals in scalp hair. Environ. Monit. Assess. 104, 45–57.

Larsen, E.H., Andersen, N.L., Moller, A., Petersen, A., Mortensen, G.K., Petersen, J., 2002. Monitoring the content and intake of trace elements from food in Denmark. Food Addit. Contam. 19, 33–46.

Leblanc, J.C., Malmauret, L., Guérin, T., Bordet, F., Boursier, B., Verger, P., 2000. Estimation of the dietary intake of pesticide residues, lead, cadmium, arsenic and radionuclides in France. Food Addit. Contam. 17, 925–932.

Llobet, J.M., Falcó, G., Casas, C., Teixidó, A., Domingo, J.L., 2003. Concentrations of arsenic, cadmium, mercury and lead in common foods and estimated daily intake by children, adolescents, adults and seniors of Catalonia, Spain. J. Agric. Food Chem. 51, 838–842.

Lopez Alonso, M., Prieto Montana, F., Miranda, M., Castillo, C., Hernandez, J., Luis Benedito, J., 2004. Interactions between toxic (As, Cd, Hg and Pb) and nutritional essential (Ca, Co, Cr, Cu, Fe, Mn, Mo, Ni, Se, Zn) elements in the tissues of cattle from NW Spain. Biometals 17, 389–397.

Mc Cance, Widdwson, 1980. First Supplement of the Composition of Foods. Elsevier, North-Holland Biomedical.

McDowell, M.A., Dillon, C.F., Osterloh, J., Bolger, P.M., Pellizzari, E., Fernando, R., Montes de Oca, R., Schober, S.E., Links, T., Jones, R.L., Mahaffey, K.R., 2004. Hair mercury levels in US children and women of childbearing age: reference range data from NHANES 1999–2000. Environ. Health Perspect. 112, 1165– 1171.

Moreiras, O., Cuadrado, C., 1992. Theoretical study of the intake of trace elements (nutrients and contaminants) via total diet in some geographical areas of Spain. Biol. Trace Elem. Res. 32, 93–103.

Moreiras, O., Carvajal, A., Cabrera, L., Cuadrado, C., 2006. Tablas de composición de alimentos, tenth ed. Ed. Pirámide, Madrid.

Muñoz, O., Devesa, V., Suñer, M.A., Velez, D., Montoro, R., Urieta, I., Macho, M.L., Jalón, M., 2000. Total and inorganic arsenic in fresh and processed fish products.

J. Agric. Food Chem. 48, 4369–4376.

Noel, L., Leblanc, J.C., Guerin, T., 2003. Determination of several elements in duplicate meals from catering establishments using closed vessel microwave

digestion with inductively coupled plasma mass spectrometry detection: estimation of daily dietary intake. Food Addit. Contam. 20, 44–56.

Nowak, B., 1998. Contents and relationship of elements in human hair for a non- industrialised population in Poland. Sci. Total Environ. 8, 59–68.

Nowak, B., Chmielnicka, J., 2000. Relationship of lead and cadmium to essential elements in hair, teeth, and nails of environmentally exposed people. Ecotoxicol. Environ. Saf. 46, 265–274.

Sanna, E., Liguori, A., Palmas, L., Soro, M.R., Floris, G., 2003. Blood and hair lead levels in boys and girls living in two Sardinian towns at different risks of lead pollution. Ecotoxicol. Environ. Saf. 55, 293–299.

Schlegel-Zawadska, M., 1992. Chromium content in the hair of children and students in Southern Poland. Biol. Trace Elem. Res. 32, 79–83.

Schlegel-Zawadzka, M., Zachwieja, Z., Huzior-Baajewicz, A., Pietrzyk, J.J., 2002. Comparative analysis of zinc status, food products frequency intake and food habits of 11-year-old healthy children. Food Addit. Contam. 19, 963–968.

Senofonte, A., Violante, N., Caroli, S., 2000. Assessment of reference values for elements in human hair of urban schoolboys. J. Trace Elem. Med. Biol. 14, 6–13. Souci, S.W., Fachmann, W., Kraut, H., 1981. Food Composition and Nutrition Tables

1981–1982. Wissenschaftliche Verlagsgesellschaft, Stuttgart.

Sukumar, A., 2002. Factors influencing levels of trace elements in human hair. Rev.

Environ. Contam. Toxicol. 175, 47–78.

Tsuda, T., Inoue, T., Kojima, M., Auki, S., 1995. Market basket and duplicate portion estimation of dietary intakes of cadmium, mercury, arsenic, copper, manganese and zinc by Japanese adults. J. AOAC Int. 78, 1363–1368.

Wilhelm, M., Wittsiepe, J., Schrey, P., Budde, U., Idel, H., 2002. Dietary intake of cadmium by children and adults from Germany using duplicate portion sampling. Sci. Total Environ. 285, 11–19.

Yoshinaga, J., Imai, H., Nakazawa, M., Suzuki, T., Morita, M., 1990. Lack of significantly positive correlations between elemental concentrations in hair and in organs. Sci. Total Environ. 99, 125–135.

Ysart, G., Miller, P., Croasdale, M., Crews, H., Robb, P., Baxter, M., de l'Argy, C., Harrison, N., 2000. UK total diet study-dietary exposures to aluminium, arsenic, cadmium, chromium, copper, lead, mercury, nickel, selenium, tin and zinc. Food Addit. Contam. 17, 775–786.