Levels and trends of PCDD/Fs in human blood and milk of residents in the vicinity of a modern municipal solid waste incinerator near to Lisbon

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Introduction

Possible health hazard from human exposure to municipal solid waste (MSW) incinerator emissions, which can include chlorinated persistent organic compounds such as polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDDs), is of great public concern. The needed rigorous control of these emissions should be reflected by not different time trends in the PCDD/Fs levels of both the populations residing in the vicinity of incinerators and others selected as control, similar in socio-demographic and other factors potentially influencing those levels.

To check the possibility of enhanced body burdens due to MSW emissions, well-designed studies have to be performed to determine human exposure to PCDD/Fs over time. Biological (human) monitoring of these chemicals has several advantages over environmental monitoring, since it is able to measure human body burden and accounts for exposure from all sources, environmental pathways and routes of absorption. However, it is practically unable to identify specific sources, routes and pathways, important issues for risk management purposes[1].

Since 1999, a modern municipal solid waste incinerator is operating in the Metropolitan Area of North-Lisbon, Portugal. In order to get a reliable data basis on the body burden of PCDD/Fs in the population residing in the vicinity of the plant, susceptible of identifying time trends and possible regional differences, a biomonitoring program was implemented in the ambit of the VALORSUL Environmental Health Survey, referred elsewhere[2]. The present paper describes results of this biomonitoring program, which includes repeated cross-sectional studies and, as much as possible, regular examinations of blood and breast milk samples, to determine PCDD/F levels.

Materials and methods

Study group: Apparently healthy adults, not-known occupationally exposed to dioxins, living at residing area for more than 1 year volunteered, after written informed consent was given, to participate in blood and breast milk biomonitoring studies. Participants were classified as exposed and controls, depending on specific living area at a distance lesser or farther than 5 km from the MSW facility. Similar relevant socio-demographic characteristics were ensured, in order to avoid

between-group bias. For breast milk monitoring, only primiparous and/or women breast-feeding first child or, at least, 3 years after breast-feeding the last child were included.

Sample and data collection: Blood and milk samples have been collected every two years, starting on 1999 until 2002 or 2003, for blood and milk, respectively, meaning that to establish baseline levels and to monitor space and time trends two and three observational periods have been considered.
At working place, fasting participants gave both venous blood samples (about 80 ml), collected by health professionals, and, via questionnaire, data on individual characteristics, smoking and drinking habits, specific dietary habits (mainly consumption of meat, fish, dairy and local food), professional activity and specific hobbies, residential and work conditions, physical activity, past history of diseases and treatments.
Breast milk samples (about 40 ml) were obtained at women's residence, four weeks after delivery. Questionnaires were also administered to gather relevant socio-demographic and health data on mothers and newborns, as well as information on parity, number of breastfed children and breastfeeding duration of previous children.

Analytical procedures: Frozen blood and breast milk samples were delivered to ERGO Dioxin Laboratory, Hamburg, Germany, to determine concentration of seven PCDDs and ten PCDFs, via high-resolution gas chromatography and high-resolution mass spectrometry (HRGC/HRMS), described in detail elsewhere[3]. PCDD/F-levels were converted into TEQ-levels using WHO-TEFs[4].

Statistical analysis: Data base management was performed using Microsoft Access 2000 (9.0.3821 SR-1) and, for the statistical analyses, SPSS software version 12.0 for Windows was used. Significance level was generally fixed at α=0,05. Numerical variables were described by their arithmetic means and 95% confidence intervals or medians, percentage of results above them and variation intervals. Appropriate tests (t-Student, Mann-Whitney, Chi-square and Fisher exact) were used to compare means, medians and proportions across the two areas of residence and between age and other relevant related groups. Single and subsequent multiple linear regression analyses were used to identify determinants of dioxins body burden among individual characteristics and environmental factors.

Results and discussion

Study group: In relation to the specific living area of the participants and considering both population groups, differences in the studied variables such as age, main professional activity, relevant dietary and smoking habits generally were not statistically significant. The only few exceptions are related to variables with high percentage of missing information.
Concerning mean age, differences along the period of study had no statistical significance, either for the general population (ranging between 41 and 45 years, for men and women) or breastfeeding women (assuming values from 29 to 31 years). These findings led to the conclusion that the results to be obtained from the global study are not likely to be confounded by a selection bias.
PCDD/Fs in human blood: A total of 118 adults from general population were studied for determination of PCDD/Fs in individual blood samples during the two cross-sectional studies already performed. In Table 1, results of the analyses are summarized for every period, stratified by

living area, sex and age of the participants. As can be seen, in relation to the first two factors and over time, controls and women, when compared with exposed and men, show blood PCDD/F mean levels slightly higher, although differences are not statistically significant. Therefore, two conclusions can be drawn: a) PCDD/F levels in the global population did not change significantly (p=0,329) from baseline to first potential specific impact of MSW incinerator; and b) studied population can be characterized by a dioxin body burden spread over a range from 5 to 34 pg/g WHO-TEQ/g fat, with mean (median) values around 16 (15) pg/g WHO-TEQ/g fat. As described by others[5], significant positive correlations between age and PCDD/F concentrations in blood for both periods have also been observed.

Table 1 - PCDD/F levels in human blood in relation to study period, living area, sex and age of participants

Factor	Group	TEQ-WHO (pg/g fat)	T0[a]	T1[b]
Living area	Exposed	N	10	50
		Mean	14,9	16,8
		Median	13,6	15,3
		Standard deviation	5,6	5,8
		Min.-Max.	8,2-26,2	4,0-34,3
	Control	N	23	35
		Mean	15,5	16,1
		Median	15,6	15,7
		Standard deviation	4,8	6,0
		Min.-Max	6,3-25,0	4,6-32,3
		p [c]	0,603	0,605
Sex	Male	N	11	39
		Mean	14,9	15,7
		Median	13,8	14,9
		Standard deviation	4,5	4,7
		Min.-Max	8,2-23,7	4,0-24,8
	Female	N	22	43
		Mean	15,5	17,5
		Median	15,4	16,5
		Standard deviation	5,3	6,8
		Min.-Max	6,3-26,2	4,6-34,3
		p [c]	0,778	0,235
Age (years)		**r[d] (p)**	0,56 (**0,002**)	0,42 (< **0,001**)
Global population		N	33	85
		Mean	15,3	16,5
		Median	15,1	15,4
		Standard deviation	5,0	5,9
		Min.-Max	6,3-26,2	4,0-34,3

a, b – 1st (T0) and 2nd (T1) study periods, respectively; c – Mann-Whitney test; d – Spearman coefficient correlation

PCDD/Fs in human breast milk: Altogether, 123 women have been studied for determination of PCDD/F levels in individual breast milk samples, along the first 5-year operation period of the VALORSUL MSW facility. In relation to specific living area (Table 2), results of the analyses, shown for each of the three periods of study, suggest that differences between groups are not statistically significant. Moreover, PCDD/F levels for the global group did not change significantly (p=0,822) over time, meaning that dioxin body burden, as estimated by PCDD/F concentrations on breast milk samples, were relatively constant during the whole monitoring period. Being dioxin

levels in human milk one of the best indicators of environmental human exposure to these chemicals, it can be considered that found levels are a good estimate of local *background* exposure to PCDD/F.
Although for a more reduced age range than for adults from general population, women also show significant positive correlations between age and PCDD/F concentrations in breast milk for the three periods considered in the study.

Table 2 - PCDD/F levels in breast milk in relation to study period, living area and age of mothers

Factor	Group	TEQ-WHO (pg/g fat)	T0[a]	T1[b]	T2[c]
Living area	Exposed	N	9	27	15
		Mean	10,4	10,9	10,9
		Median	9,4	10,4	11,0
		Standard deviation	3,4	4,1	2,9
		Min.-Max.	5,7-16,2	4,4-20,9	5,9-15,6
	Control	N	43	14	15
		Mean	10,9	9,8	10,2
		Median	10,8	10,5	10,4
		Standard deviation	2,9	2,5	3,0
		Min.-Max	4,8-19,9	5,4-12,7	5,5-16,9
		p [d]	0,536	0,541	0,512
Age (years)		**r[d] (p)**	0,45 (**0,001**)	0,43 (**0,005**)	0,56 (**0,001**)
Global population		N	52	41	30
		Mean	10,8	10,5	10,5
		Median	10,8	10,5	10,7
		Standard deviation	2,9	3,6	2,9
		Min.-Max	4,8-19,9	4,4-20,9	5,5-16,9

a, b, c –1st (T0), 2nd (T1) and 3rd (T2) study periods, respectively; d - Mann-Whitney test; e - Spearman coefficient correlation

Other determinants of PCDD/F levels in blood and breast milk: Several other personal and environmental variables have been analysed for contribution to the concentration of PCDD/Fs in blood and breast milk of the study populations. However, in single regression, either for general population or breast-feeding women, age was the only identified determinant, confirming above analysis.

Congener profile of PCDD/Fs in human blood and milk: Either in human blood and milk, the profile of the single congeners for PCDD/Fs was quite similar to those generally observed in industrialized countries [6]. Most contributors to the TEQs were, by descending order, 12378-PCDD, 23478-PCDF, 123678-HCDD and 2378-TCDD. Altogether, these individual congeners accounted for approximately 80% of the total identified dioxin body burden in the studied groups.

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