IMPACT OF IRON AND STEEL INDUSTRY AND WASTE INCINERATORS ON HUMAN EXPOSURE TO DIOXINS, PCBs AND HEAVY METALS: RESULTS OF A CROSS-SECTIONAL STUDY IN BELGIUM

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ABSTRACT

We evaluated the impact of two iron and steel plants and two municipal solid waste incinerators (MSWI) in Wallonia (Belgium) on the exposure of residents to dioxins, PCBs, and heavy metals. A total of 142 volunteers living around these facilities were recruited and compared with 63 referents from a rural area with no industrial source of pollution. Information about smoking habits, dietary habits, anthropometric characteristics, residential history and health status was obtained from a self-administered questionnaire. The volunteers provided blood under fasting conditions in order to evaluate the body burden of dioxins (17 PCDD/Fs congeners) and PCBs. Samples of blood and urine were also taken for the determination of cadmium, mercury and lead. After adjustment for covariates, concentrations of cadmium, mercury and lead in urine or blood were not increased in subjects living in the vicinity of MSWIs or sinter plants by comparison with referents. Residents around the sinter plants and the MSWI located in the industrial area had concentrations of dioxins and PCBs in serum similar to that of referents. By contrast, subjects living in the vicinity of the MSWI in the rural area showed significantly higher serum levels of dioxins (geometric mean, 38 vs 24 pg TEQ/g fat, $p<0.0001$) and coplanar PCBs (geometric mean, 10.8 vs 7.0 pg TEQ/g fat, $p<0.05$). Whereas age-adjusted dioxin levels in referents did not vary with local animal fat consumption, concentrations of dioxins in subjects living around the incinerators correlated positively with their intake of local animal fat, with almost a doubling in subjects with the highest fat intake. These results indicate that dioxins and coplanar PCBs emitted by MSWIs can indeed accumulate in the body of residents who regularly consume animal products of local origin.

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INTRODUCTION

Emissions from industries contribute to global air pollution by emitting a large range of pollutants. Municipal solid waste incinerators (MSWIs) and iron and steel plants represent major sources of atmospheric pollution by dioxins (polychlorinated dibenzo-*p*-dioxins/dibenzofurans) (Everaert and Baeyens, 2002; Gilpin et al., 2003; Zook and Rappe, 1994). Since the mid-nineties, stringent emission standards have been imposed to MSWIs in order to achieve dioxin emission levels below 0.1 ng TEQ/Nm³. With the progressive abatement of emissions from MSWIs, the iron and steel industry is now becoming the major contributor to the global atmospheric pollution by dioxins (Anderson and Fisher, 2002; Buekens et al., 2001). The human impact of these pollutants is still of concern because of their ability to bio-accumulate in the food chain and then in human fatty tissues (Startin and Rose, 2003). Incinerators are potential sources of other pollutants, especially heavy metals which can also bio-accumulate in the food chain (Lisk, 1988; Schuhmacher et al., 2002).

In order to assess the human exposure impact of these pollutants, we carried out an epidemiological study to compare the concentrations of dioxins, polychlorinated biphenyls, cadmium, mercury and lead in blood and urine of subjects living in the vicinity of two sinter plants and two MSWIs, with levels found in referent subjects recruited in a rural area with no industrial source of pollution.

MATERIALS AND METHODS

After approval by the University Ethics Committee, a total of 142 volunteers living around either a MSWI or an iron and steel facility were recruited. Fifty-one subjects aged 21 to 80 years were living within a distance of 2 km from the MSWI of Thumaide (MSWI 1), in a rural area. This MSWI was constructed in 1980 and has a capacity of 12.4 tons/hour. Thirty-three subjects aged 33 to 65 years were recruited within a distance of 2 km from the MSWI of Pont-de-Loup (MSWI 2), in an industrial area. This second MSWI was constructed in 1978 and its capacity is 15.5 tons/hour. Fifty-eight subjects aged 25 to 67 years were living within a distance of 4 km from two iron and steel plants of the Cockerill-Sambre company (Usinor group), one plant located in the suburbs of Liège (n=12) and the other in the suburbs of Charleroi (n=46). These subjects were compared with 63 referents recruited in three villages (Bertrix, Daverdisse and Nassogne) situated in rural areas in the Ardenne (South of Belgium), with no known local source of pollution. After having given their informed consent, the volunteers provided approximately 200 ml of fasting blood and a urine sample. Information about smoking habits, dietary habits, anthropometric characteristics, residential history and health status was obtained via a self-administered questionnaire. Total fat intake was calculated from the questionnaire, on the basis of consumption of poultry, bovine and swine products. The local animal fat intake was calculated from the consumption of poultry and bovine products only. For each type of food, the amount produced locally was estimated by asking in the questionnaire the proportion produced locally by the participants themselves (like poultry products) or purchased in the farms located in the vicinity of the place of residence. Swine products were not considered for local fat intake because of the higher dependence of pigs to manufactured feeds, and their lower ability to accumulate dioxins (Bernard et al., 2002). Fish consumption information was obtained by the questionnaire. We quantified serum concentrations of the seventeen 2,3,7,8- substituted polychlorinated dibenzodioxin / dibenzofuran congeners (PCDD/Fs or dioxins), coplanar PCBs (IUPAC n° 77, 81, 126 and 169) and 12 PCB markers (IUPAC n° 3, 8, 28, 52, 101, 118, 138, 153, 180, 194, 206 and 209) by gas chromatography/high-resolution mass spectrometry (GC-HRMS) (Focant et al., 2001; Focant and De Pauw, 2002). The dioxin and coplanar PCB concentrations were expressed in toxic equivalent (WHO-TEQ) (Van den Berg et al., 1998). The concentrations of cadmium and mercury in urine and the blood lead concentrations were determined by atomic absorption spectrometry (Cardenas et al., 1993).

All variables except age and body mass index (BMI) were normalized by log transformation. Differences between groups were assessed by analysis of variance (ANOVA) followed by Dunnett's multiple-comparison post-hoc tests. Chi-square tests were used to compare homogeneity of prevalence between exposed groups and referents. Determinants of dioxin, PCB and heavy metal concentrations were identified by stepwise multiple linear regression analysis. All analyses were carried out with SAS version 8.0, Enterprise Guide version 2.0 (SAS Institute Inc., Cary, NC).

RESULTS

The characteristics of the studied population are presented in Table 1. The four groups were comparable with respect to the sex ratios (51 to 61% of women) and age, with the exception of the group around MSWI 2 who were slightly younger than the other groups. There were no statistically significant differences in the smoking habits between the groups. The total animal fat consumption was rather similar between the four groups but as expected, the groups around MSWI 2 and the sinters plants, localized in industrial areas, consumed significantly less animal fat from local productions.

Table 1. Characteristics of the studied population.

	Referents	**MSWI 1**	**MSWI 2**	**Sinter plants**
N	63	51	33	58
Number of women	34 (54%)	26 (51%)	20 (61%)	32 (55%)
Number of men	29 (46%)	25 (49%)	13 (39%)	26 (45%)
Age (years)[a]	52.9 (7.8)	53.3 (12.5)	46.1 (8.4)*	52.0 (10.3)
BMI (kg/m^2)[b]	25.3 [24.4-26.1]	27.4 [26.1-28.7]*	27.2 [25.7-28.8]*	25.9 [24.9-27.0]
Tobacco consumption				
Number of smokers	10 (16%)	7 (14%)	5 (15%)	12 (21%)
Consumption (pack-years)	9.0 [3.9-21.1]	23.8 [9.8-57.9]	15.3 [10.5-22.2]	20.4 [10.2-40.7]
Number of past-smokers	14 (22%)	18 (35%)	4 (12%)	14 (24%)
Consumption (pack-years)	13.5 [7.8-23.6]	24.2 [14.2-41.2]	24.8 [11.9-51.8]	13.9 [7.9-24.2]
Alcohol consumption				
Number of consumers	48 (76%)	45 (88%)	23 (70%)	42 (76%)
Consumption (drinks/week)	6.1 [4.7-7.9]	9.6 [7.2-12.9]*	4.4 [3.2-6.1]	7.3 [5.7-9.3]
Animal fat consumption (terrestrial food chain)				
Total animal fat (g/week)	285 [256-316]	272 [244-304]	254 [225-288]	292 [245-347]
Local animal fat				
Number of consumers	46 (73%)	50 (98%)*	22 (66%)	22 (38%)*
Local animal fat (g/week)	99 [77-127]	109 [90-133]	66 [43-101]*	47 [33-66]*
Fish consumption				
Number of consumers	49 (79%)	49 (96%)*	28 (85%)	52 (91%)
Consumption (g/week)[c]	254 [212-302]	174 [157-192]*	184 [137-248]	283 [225-355]

Data are geometric mean [95% Confidence Interval], except otherwise stated. [a]Arithmetic mean (SD). [b]Harmonic mean [95 % CI]. BMI = Body mass index. [c]Fresh weight. *$p<0.05$ for ANOVA with Dunnett's post-hoc tests or Chi² tests in case of prevalence.

The mean concentrations of pollutants in blood or urine in the different groups are compared in Table 2. Residents around the sinter plants showed concentrations of dioxins, PCBs and lead in blood that were similar to that of referents. The concentrations of cadmium and mercury in urine of these residents were also not increased. There was no increase in the concentrations of these pollutants in the group living around the MSWI in the industrial area (MSWI 2). The only group to show statistically significant increases in studied biomarkers was the group of subjects living around the MSWI in the rural area (MSWI 1). These subjects showed a significant increase of the serum levels of dioxins and coplanar PCBs compared to referents as well as with the rest of the population. Their concentrations of heavy metals in the urine or blood were however not significantly increased, although this group had the highest average urinary excretion of cadmium, even after adjustment for smoking status.

All these observations were confirmed by stepwise multiple linear regression analyses testing age, gender, BMI, animal fat consumption, fish consumption, alcohol consumption, tobacco smoking and place of residence as independent variables. The accumulation of dioxin was significantly influenced by age, residence around MSWI 1 (partial $r^2=0.12$; $p<0.0001$), animal fat consumption and BMI while the body burden of coplanar PCBs was determined by residence around MSWI 1 (partial $r^2=0.20$; $p<0.0001$), age, BMI and fish consumption. After adjustment for covariates, the increases of dioxins and coplanar PCBs in the serum of residents around MSWI 1 averaged 54% and 49%, respectively. As illustrated in Figure 1, the adjusted dioxin concentrations were significantly correlated with the intake of local animal fat in residents around the two MSWIs, with almost a doubling in subjects with the highest fat intake. Such a relationship was not found in referents, reflecting the absence of contamination of local food chain in the control area.

Table 2. Concentrations of pollutants in blood or urine.

	Referents n=63	MSWI 1 n=51	MSWI 2 n=33	Sinter plants n=58	Total n=205
Dioxins and polychlorinated biphenyls					
PCDD/Fs (pg TEQ/g lipids)	23.9 [21.4-26.6]	37.9* [32.8-43.8]	24.1 [20.2-28.9]	23.8 [20.8-27.1]	26.8 [24.9-28.8]
Coplanar PCBs (pg TEQ/g lipids)	7.0 [6.1-8.0]	10.8* [9.2-12.8]	6.4 [5.1-7.9]	6.3 [5.3-7.6]	7.5 [6.8-8.2]
PCDD/Fs+cPCBs (pg TEQ/g lipids)	31.3 [28.2-34.8]	49.0* [42.4-56.7]	30.6 [25.6-36.7]	30.7 [26.8-35.2]	34.7 [32.3-37.8]
Σ12 PCB markers (ng/g lipids)	416 [385-450]	465 [406-530]	375 [320-440]	402 [358-451]	416 [393-441]
Heavy metals					
Cd urine (µg/g creatinine)	0.49 [0.41-0.59]	0.62 [0.53-0.74]	0.43 [0.33-0.58]	0.49 [0.40-0.61]	0.51 [0.46-0.57]
Hg urine (µg/g creatinine)	1.95 [1.75-2.16]	1.80 [1.53-2.11]	2.11 [1.75-2.55]	1.79 [1.55-2.06]	1.89 [1.76-2.02]
Pb blood (µg/L)	45.8 [39.4-53.2]	43.3 [36.1-51.9]	39.4 [33.3-46.6]	42.2 [36.1-49.3]	43.1 [39.7-46.7]

Data are geometric mean [95% Confidence Interval]. *p<0.05 (ANOVA with Dunnett's post hoc test).

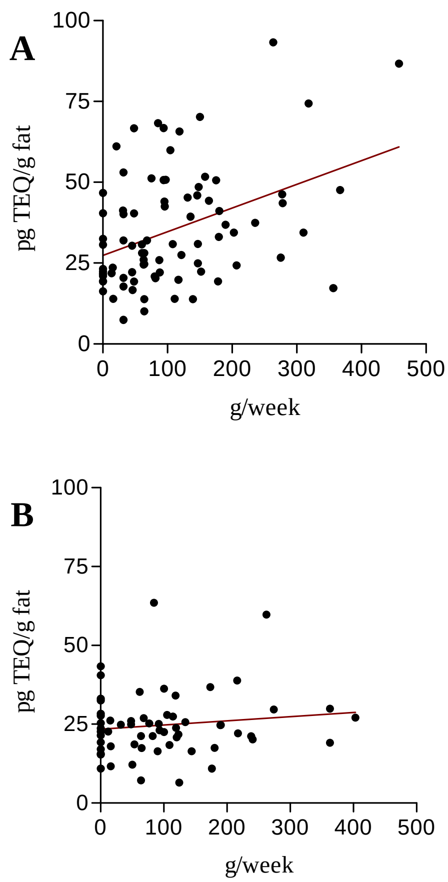

Figure 1. Correlation between dioxin serum levels and consumption of locally produced animal fat. A. Residents around the two MSWIs (n=84; Spearman r=0.37; p=0.0006). B. Referents (n=63; Spearman r=0.06; p=0.64). Dioxin values are adjusted for age, BMI and total fat consumption.

Stepwise regression analyses confirmed also the absence of significant impact of the MSWIs or of the sinter plants on the exposure of residents to heavy metals. The determinants of urinary cadmium were age, tobacco smoking and gender (higher in women). Blood lead correlated with age, alcohol consumption and gender (higher in men) whereas urinary mercury correlated only with BMI and gender (higher in women).

DISCUSSION AND CONCLUSION

The present study shows that dioxins and coplanar PCBs emitted by MSWIs can accumulate in the body of people living around these facilities. The accumulation requires however a regular consumption of local animal products contaminated by relatively high emissions of dioxins. The two studied MSWIs indeed had been in activity since the early 80s and emitted, during many years, large amounts of dioxins (>50 ng TEQ/ TEQ/Nm^3). This resulted in high levels of dioxins in the food chain and in particular in cow's milk in nearby farms, some samples having concentrations up to 38.9 pg TEQ/g fat. For these reasons, it is unlikely that the increased dioxin body burden found in the present study occurs around MSWIs complying with emission standards currently in force in most countries. Although dioxin emissions of the MSWI in the rural area resulted in a significant contamination of residents, with almost a doubling of the body burden in subjects with the highest local fat intake, the observed values were comparable to values prevailing in the 80s in Belgium and most industrialized countries (Schecter et al., 1994; Wittsiepe et al., 2000). The values remained also lower than current levels found in populations regularly eating seafood (Kiviranta et al., 2000).

By contrast with MSWIs, emissions from the sinter plants were not associated with an increased dioxin body burden of residents. One possible explanation is that the dioxin emissions from the two studied plants were not sufficiently high to contaminate the local food chain. The lower local animal fat consumption of residents around these facilities has probably also contributed to decrease the risks of exposure, like also the fact that dioxins emissions from this industry are dominated by furans (PCDFs), known to be less persistent in the environment and farm animals than dioxin congeners (PCDDs) (Bernard et al., 2002).

In conclusion, although iron and steel industry and waste incinerators contribute to global air pollution, it appears that they significantly increase the exposure of residents to persistent pollutants like dioxins only when high emission levels are coupled with a regular consumption of locally produced food.

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