

# Dispersion Modeling as a Dioxin Exposure Indicator in the Vicinity of a Municipal Solid Waste Incinerator: A Validation Study

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Whether low environmental doses of dioxin affect the general population is the matter of intense debate and controversy. In a previous study, we found a 2.3-fold risk for non-Hodgkin lymphoma associated with residence in areas classified as highly exposed to dioxin emitted from a municipal solid waste incinerator (MSWI) (Besançon, France). The main limitation lay within the use of a first-generation Gaussian-type dispersion model as a proxy for dioxin exposure, since its accuracy had not been assessed before. The aim of this study was to validate this geographic-based exposure through PCDD/F measurements from soil samples. PCDD/F concentration, pH, organic carbon concentration, cation exchange capacity, and geomorphology and ecology features were assessed for 75 sampling points. In simple terrain (i.e. northeast of the MSWI), a significant association was highlighted between modeled dioxin ground-level air concentrations and log-transformed measured dioxin soil concentrations with a strong gradient across exposure categories. Conversely, in a complex topography situation (i.e. southwest of the MSWI), the model overpredicted ground-level air concentrations, particularly in the high exposure zone. First-generation modeling provided a reliable proxy for dioxin exposure in simple terrain, reinforcing the results of our case-control study. However, a more advanced atmospheric diffusion model should have been used for refined assessment in complex terrain.

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## Introduction

Among combustion byproducts of municipal solid waste incinerators (MSWIs), polychlorinated dibenzo-*p*-dioxins (PCDDs), and polychlorinated dibenzofurans (PCDFs) have generated the most public concern. Because of their notably toxic properties, PCDD/Fs have received prolonged attention by environmental regulators (1). According to the International Agency for Research on Cancer, one of them, the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (the so-called Seveso dioxin), is carcinogenic to humans (2). In this assessment, human carcinogenicity data came mainly from cohort studies of industrial populations. While PCDD/Fs were detected for the first time in MSWI emissions in the 1970s in The Netherlands (3), a lot of controversial issues related to this subject still persist, especially vis-à-vis their impact on public health. In this respect, whether low environmental doses of dioxin affect the general population, particularly in the vicinity of a municipal solid waste incinerator, is debated.

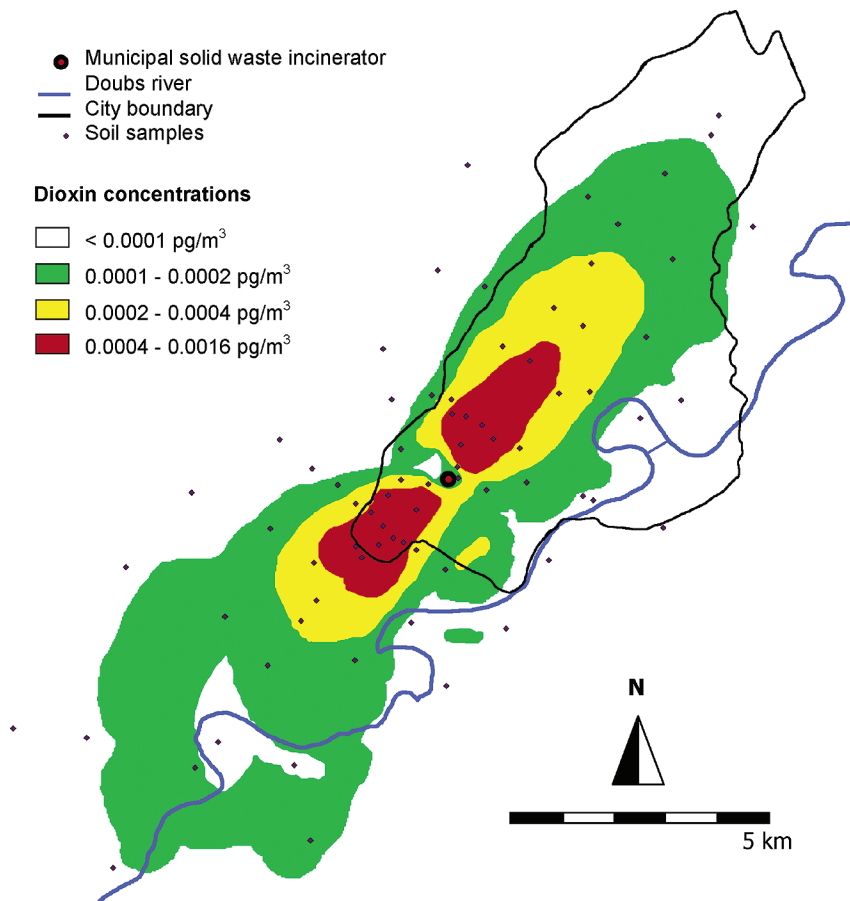
In France, in the past, dioxins have not been properly controlled in old plants. In 1998, the French Ministry of the Environment revealed that of 71 MSWIs processing more than 6 metric tons of material per hour, 15 had dioxin emissions which were above 10 ng international toxic equivalent (I-TEQ)/m<sup>3</sup>, whereas the European guide value is 0.1 ng I-TEQ/m<sup>3</sup> (4).

Our team recently examined the spatial distribution of non-Hodgkin lymphomas (NHL) around one of these polluting MSWIs (Besançon, eastern France). Using a spatial scan statistic we found evidence for a cluster, composed of two electoral wards (one of them containing the MSWI), with a standardized incidence ratio of 1.3 (95% confidence interval (CI), 1.1–1.4) (5). A subsequent study, focusing on the city of Besançon, aimed at comparing the incidence of NHL between 1980 and 1995 with that in controls randomly selected from the 1990 population. Ground-level air dioxin concentrations were modeled with a Gaussian-type dispersion model, yielding four exposure categories. The latter were linked to individual places of residence using GIS technology. Predicted concentrations of PCDD/Fs from an air dispersion model were therefore used to classify exposure in the study population. We found a 2.3-fold risk for non-Hodgkin lymphoma associated with residence in the highest dioxin concentration area (95% CI, 1.4–3.8) (6). In our opinion, these findings lend support to an environmental dioxin risk for NHL in the vicinity of a MSWI for the surrounding population. The main limitation of this study lay within the use of dispersion modeling as a proxy for dioxin exposure, since its validity or appropriateness had not been assessed before (and cannot be inferred from successfully associating effect and exposure).

Obviously, the only definitive method of confirming the accuracy of a dispersion model is to collect local monitoring data. Since PCDD/F emissions result in subsequent aerial deposition onto soil, this matrix is considered an adequate environmental monitor for assessing long-term exposure to PCDD/Fs (7). The aim of this study was therefore to validate geographic-based exposure categories (derived from isopleths of predicted ground-level air concentrations from a diffusion model) through PCDD/F measurements from soil samples.

## Materials and Methods

**Study Area.** The MSWI under investigation is situated near the southwest boundary of Besançon, 4 km from the city center. A main road with heavy traffic runs near the plant. The surrounding region exhibits a complex pattern. On the



**FIGURE 1. Modeled average ground-level dioxin concentrations and soil samples locations around the municipal solid waste incinerator of Besançon, France. Reprinted with permission from Floret, N.; Mauny, F.; Challier, B.; Arveux, P.; Cahn, J.-Y.; Viel, J. F. *Epidemiology* 2003, 14, 392–398. Copyright 2003 Lippincott Williams & Wilkins.**

northeast side, the site is a mixed commercial/urban area with gentle hills of moderate slope, while on the southwest side, the terrain is complex, with more pronounced hills and valleys, predominantly covered by natural prairie grasses interspersed with forest and urban patches.

As for the physical description of the plant, the stack is 40 m high. Combustion chambers 1 and 2 (each with a capacity of 2.1 metric tons per hour) were put into service in 1971. In 1976, a third combustion chamber was opened (with a capacity of 3 metric tons per hour). In 1998, approximately 67 000 metric tons of waste were processed. Some legal guidelines for incinerator emissions have not been followed at this location. For example, in 1997, dust and hydrogen emission levels were higher than prescribed, and exhaust gases were not maintained at temperatures of more than 850 °C for the legal time ( $>2$  s), allowing dioxins to be emitted. The first time that the dioxin concentration of an exhaust gas was ever measured (in December 1997), it was found to be 16.3 ng I-TEQ/m<sup>3</sup> (4). With such conditions, it could be expected that in the last two decades remarkable amounts of PCDD/Fs were released into the environment. Combustion chamber 1 (the most polluting) was shut down on December 31, 1998. To comply with the European guideline, combustion chamber 2 was replaced by a new one with up-to-date pollution controls, combustion chamber 4, which started operation in late 2003. Combustion chamber 3 remained unchanged.

**Dioxin Exposure Modeling.** Naive use of simple exposure models (e.g. distance-only models) can lead to erroneous conclusions, since it is clear that differential exposure may occur with a change in distance and direction, particularly around air pollution sources such as an incinerator stack.

Hence, some degree of sophistication in exposure modeling is required. We took advantage of a dioxin dispersion model performed in 1999 with the APC3 software (Aria Technologies, Colombes, France). This subcontracting company intervened in the framework of an environmental impact statement (supervised by the district council) to predict the future impact of dioxin emissions from combustion chambers 3 and 4. APC3, a first-generation Gaussian-type dispersion model, allowed the modeling of the transport and dispersion of dioxin emissions. The model took into account meteorological data (5 years of data for wind speed, wind direction, pressure, temperature, and Pasquill atmospheric stability classes), simplified surface topography, plume rise, stack characteristics, and future dioxin emission rate from the MSWI. It assessed average concentrations in hundreds of meteorological conditions (one Gaussian plume for each particular meteorological condition). The respective contours of these modeled ground-level air concentrations, a priori determined by the subcontracting company ( $<0.0001 \text{ pg/m}^3$ ,  $0.0001\text{--}0.0002 \text{ pg/m}^3$ ,  $0.0002\text{--}0.0004 \text{ pg/m}^3$ ,  $0.0004\text{--}0.0016 \text{ pg/m}^3$ ), were digitalized and contoured onto the surface of the map (Figure 1). In our case-control study, we assumed that contour shapes, as derived from the prediction model were reliable estimates of past dioxin exposure profiles, provided relative figures rather than absolute figures were used. Hence, the contours were classified as very low (modeled ground-level air concentrations:  $<0.0001 \text{ pg/m}^3$ ), low (modeled ground-level air concentrations:  $0.0001\text{--}0.0002 \text{ pg/m}^3$ ), intermediate (modeled ground-level air concentrations:  $0.0002\text{--}0.0004 \text{ pg/m}^3$ ), and high (modeled ground-level air concentrations:  $0.0004\text{--}0.0016 \text{ pg/m}^3$ ) exposure areas.

**Selection of Sampling Sites.** Sampling points were selected based on the prediction of the time-average emission plume obtained from the Gaussian model. The study design employed a stratified random selection process, involving the four quadrants around the incinerator with an emphasis on sampling in the quadrants that were historically downwind from the incinerator: the northeast and the southwest quadrants. Accurate position of each sampling site was determined in relation to homogeneous geological and topographical conditions and vegetation. The following conditions were sought during site selection: (1) level, undisturbed soil, (2) away from trees, (3) not adjacent to roads or railway lines, and (4) not known or suspected to have high dioxin concentrations for any other reason. In July 2002, 75 soil samples were collected in the vicinity of the facility, at between 97 m to 12 km from the stack (Figure 1).

**Soil Sampling Procedure.** Each sample site consisted of an area of 10 m × 10 m. Aliquots were collected at each corner and at the center. A stainless steel auger was used to extract cylindrical samples of soil. Soil samples were taken from the upper 10 cm of soil, without vegetation. All the borehole aliquots collected in the same sampling site were mixed to obtain a composite sample of about 500 g.

At each sampling place, ellipsoidal geographic coordinates WGS84, expressed in degree: minute: second, were recorded. A standardized questionnaire consisting of parameters describing the sampling point (altitude, vegetation type, and geologic characteristics) was filled in. Morphological features of soil profile (structure, texture, stoniness) were also described. Hard calcareous rocks (that are fissured and well drained) were classified permeable; other soils (marls, silts, alluvial deposits) were classified impermeable. The same geologist assessed all the geomorphology and ecology features of soil samples. As a proxy for terrain elevation and land cover heterogeneity (which was superimposed on the modeled ground-level air concentration symmetry), a topography complexity index was described for each sampling point. A point located below the northwest-southeast diagonal passing through the MSWI site (i.e. the southwest side) was assigned a "complex" topography, and a "simple" topography was assigned to points on the northeast side.

**Preparation, Extraction, and Analysis of Soils.** In the laboratory, any overlying vegetation was trimmed from the cores. Soil samples were dried in a sweating room overnight and then sieved through a 9-mesh (2 mm) sieve to obtain a more homogeneous and fine grain distribution. Samples were packed into glass containers and stored in obscurity at room temperature until analysis.

The pH of each soil sample was assessed. Since the retention of PCDD/Fs in soil is considered to be a function of both the load of PCDD/Fs deposited onto the soils and the organic carbon content ( $\delta$ ), the organic carbon concentration and cation exchange capacity (CEC) were measured. The organic carbon content was quantified after removing inorganic carbon (carbonates) and expressed in g/kg of soil. CEC was measured in neutral 1 M  $\text{NH}_4\text{CO}_2\text{CH}_3$  following the method of Metson and expressed as centimoles of positive charge per kg of soil ( $\text{cmol}_c/\text{kg}$ ) (9).

The extraction and cleanup procedures as well as the analytical determination of PCDD/Fs were carried out by the Pasteur Institute of Lille (France) according to the U.S. Environmental Protection Agency (EPA) method (10). About 50 g (dry weight) was used for analytical purposes. Samples were analyzed for each of the five chlorinated dibenzo-*p*-dioxin and dibenzofuran congener groups (with four to eight chlorines) by HRGC/HRMS. Analyses of PCDD/Fs in soil samples were carried out using an Agilent 6890 coupled with a MAT 95 XL Finnigan (EI and multiple ion determination mode, resolution 10 000). Two columns were used: DB-5 MS for heptaCDD and octaCDD/CDF and RTX-2330 for tetra-

through hexaCDD/CDF, including 2,3,7,8-substituted isomers. For detection, at least two masses ( $m^+$  and  $m^{2+}$ ;  $m^{2+}$  and  $m^{4+}$ , respectively) were used for each native and labeled dioxin and furan homologue group.

Quantitative determinations of PCDD/Fs were performed by an isotope dilution method using relative response factors previously obtained from five calibration standard solutions (Wellington Laboratories Inc., Ontario, Canada).  $^{13}\text{C}$ -Labeled analogues of the PCDD/Fs congeners were added as internal standards before extraction of samples. A blank sample was added and analyzed for every batch of eleven samples. Blank samples did not contain any PCDD/F concentrations above the detection limit, which was between 20 and 150 pg/kg for the congeners tetra- to octachloro dibenzodioxins and dibenzofurans. Recoveries of internal standards usually varied between 80% and 110%. The relative deviation of the method was around 15%.

Soil samples were analyzed for each of the 7 chlorinated dibenzo-*p*-dioxin and 10 dibenzofuran congener groups and expressed in WHO-TEQ (weighted sum of the 17 dioxin and furan congeners) using toxic equivalency factors defined by the World Health Organization (WHO) (11). In the case of values under the detection limit, the congener was assumed to be absent. Concentrations are reported in pg/g of dry matter.

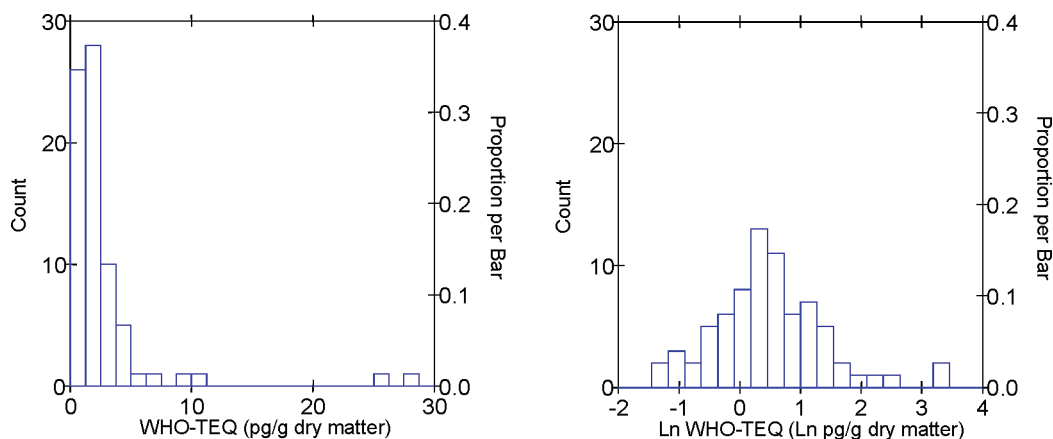
**Data Analysis.** The dispersion model was computed to predict the impact from a new combustion chamber that had not yet commenced operation. Conversely, the soil samples reflected past emissions. With such an inadequate time lag, comparing modeled concentrations in soil with those measured at the same locations would have been meaningless. Our approach, rather, was to check whether the modeled exposure areas were characterized by different mean dioxin concentrations in their respective soils, while adjusting for confounding factors.

Geomorphology and ecology features were compared across dioxin exposure areas under the plume, by means of exact Chi-Square and Kruskal & Wallis tests.

PCDD/F congeners found in environmental samples show a composition that is characteristic of their sources and their subsequent environmentally included decomposition reactions. To evaluate possible similarities and/or differences in the congener profiles of the PCDD/Fs and, therefore, to determine whether more than one potential emission source could explain the presence of PCDD/Fs in soil samples, a principal component analysis (PCA) was carried out. It consisted in deriving a few new components as the linear combination of the 17 PCDD/F congener concentrations.

Simple and multiple regression analyses were carried out to model the relation of the continuous dependent variable, the natural logarithm ( $\ln$ ) of WHO-TEQ concentration (a log transformation forced the concentration data to follow a normal distribution), with explanatory variables. Dioxin exposure derived from the dispersion model was entered into the regression model as an ordinal scale variable (very low/low/intermediate/high). Two sets of confounding factors were considered as well. Soil parameters, all continuous, consisted of pH, organic carbon (g/kg), and cation exchange capacity ( $\text{cmol}_c/\text{kg}$ ). Geomorphology and ecology parameters were either continuous (altitude in m), dichotomous (vegetation type: agriculture (farmland, grassland, pasture and lawn)/forest, permeability: yes/no, topography complexity: yes/no), or ordinal (terrain slope: 0–10%/10–30%/>30%). Dummy variables were generated for ordinal variables, taking the lowest category as the reference group.

Univariate analyses were performed for each independent variable in turn. Independent variables were included in the multivariate analysis if they had a *P*-value of 0.20 or less in the univariate analysis. As a result, average values of  $\ln$  WHO-TEQ concentration for each level of modeled dioxin exposure,



**FIGURE 2.** Histograms of dioxin concentrations and natural logarithm of dioxin concentrations (World Health Organization toxic equivalent, 75 sampling points, Besançon, France).

**TABLE 1.** Associations between In WHO-TEQ<sup>a</sup> Value and Independent Variables in Simple Regression Models (75 Sampling Points, Besançon, France)

variable	regression coefficient	95% CI	P-value
modeled dioxin exposure			
very low			
low	0.431	-0.082, 0.943	0.10
intermediate	0.866	0.327, 1.405	0.01
high	0.949	0.399, 1.498	0.001
soil parameters			
pH	0.134	-0.091, 0.360	0.24
organic carbon concentration	0.017	0.001, 0.034	0.04
cation exchange capacity	0.003	-0.032, 0.039	0.85
geomorphology and ecology parameters			
altitude	-0.007	-0.013, -0.002	0.01
vegetation	0.324	-0.991, 1.640	0.62
permeability	0.093	-0.507, 0.693	0.76
topography complexity	-0.775	-1.159, -0.391	0.0001
terrain slope			
0-10%			
10-30%	-0.133	-0.629, 0.364	0.60
>30%	-0.436	-1.773, 0.901	0.52

<sup>a</sup> World Health Organization toxic equivalent.

adjusted for other covariates, were therefore calculated to remove their variability due to these covariates.

All statistical analyses were performed with StatXact 5 (CYTEL Software Corporation, MA, U.S.A.) or SYSTAT 10 software (SPSS Inc., IL, U.S.A.).

## Results

Among the 75 soil samples analyzed for dioxin concentrations, 25 samples were collected in the very low exposure area, 19 in the low exposure area, 16 in the intermediate exposure area, and 15 in the high exposure area under the plume. Locations of sampling sites are plotted in Figure 1.

No statistically significant difference was found across the four dioxin exposure areas for geomorphology and ecology parameters of samples, except for altitude ( $p = 0.03$ ).

PCDD/F soil concentrations range from 0.25 to 28.06 pg WHO-TEQ/g dry matter (mean: 2.70 pg WHO-TEQ/g, median: 1.48 pg WHO-TEQ/g, standard deviation: 4.47 pg WHO-TEQ/g). As Figure 2 shows, the WHO-TEQ distribution exhibits a marked positive skewness, countered by logarithmic transformation.

The PCA provided a one-dimensional model (a unique eigenvalue was greater than 1.0). The first principal component explained 88% of the variance (eigenvalue = 14.94, total variance = 17). It was strongly and positively correlated with all congeners (component loadings ranged from 0.74 to 0.99).

Univariate regression results are reported in Table 1. Regarding classification of dioxin exposure (based on ground-level air concentrations), the regression coefficients represented contrasts between the mean values of log-transformed measured dioxin concentrations for categories "low, intermediate, high", and that for category "very low" (the higher the coefficient, the larger the contrast). An upward trend was noticeable, the upper two categories exhibiting a significant increase ( $p < 0.01$ , and  $< 0.001$ , respectively). Three further variables met the criterion for inclusion in the multivariate model ( $p < 0.20$ ): organic carbon concentration ( $p = 0.04$ ), altitude ( $p = 0.01$ ), and topography complexity ( $p = 0.0001$ ).

The resulting multivariate model, therefore, included the four above-mentioned factors, which explained 47.5% of the variance in the data. Results remained globally similar, except the organic carbon concentration which was no more significantly associated with the log-transformed dioxin concentration (Table 2). When testing first-order interactions, the interaction between dioxin exposure and topography complexity yielded a  $P$ -value of 0.12 ( $r^2 = 52.0\%$ ) (results not shown). This prompted us to carry out separate analyses by topography complexity category. Organic carbon concentration and altitude regression estimates remained quite stable in both models (Table 3). However, these subanalyses revealed an unexpected and clear-cut picture. In the complex topography situation ( $r^2 = 30.5\%$ ), only the intermediate

**TABLE 2. Association between ln WHO-TEQ<sup>a</sup> Value and Independent Variables in a Multiple Regression Model ( $R^2 = 47.5\%$ ) (75 Sampling Points, Besançon, France)**

variable	regression coefficient	95% CI	P-value
modeled dioxin exposure			
very low			
low	0.339	-0.086, 0.764	0.12
intermediate	0.687	0.229, 1.144	0.01
high	0.751	0.274, 1.228	0.01
soil parameters			
organic carbon concentration	0.010	-0.005, 0.024	0.19
geomorphology parameters			
altitude	-0.008	-0.013, -0.004	0.001
topography complexity	-0.816	-1.159, -0.474	0.0001

<sup>a</sup> World Health Organization toxic equivalent.

**TABLE 3. Association between ln WHO-TEQ<sup>a</sup> Value and Independent Variables in Multiple Regression Models According to the Topography Complexity (75 Sampling Points, Besançon, France)**

variable	regression coefficient	95% CI	P-value
<b>Complex Topography<sup>b</sup></b>			
modeled dioxin exposure			
very low			
low	0.490	-0.216, 1.197	0.17
intermediate	0.870	0.148, 1.592	0.02
high	0.472	-0.246, 1.191	0.19
soil parameters			
organic carbon concentration	0.014	-0.010, 0.038	0.23
geomorphology parameters			
altitude	-0.008	-0.016, 0.000	0.05
<b>Simple Topography<sup>c</sup></b>			
modeled dioxin exposure			
very low			
low	0.229	-0.324, 0.782	0.40
intermediate	0.560	-0.066, 1.185	0.08
high	1.215	0.56, 1.873	0.001
soil parameters			
organic carbon concentration	0.009	-0.011, 0.029	0.35
geomorphology parameters			
altitude	-0.008	-0.014, -0.002	0.01

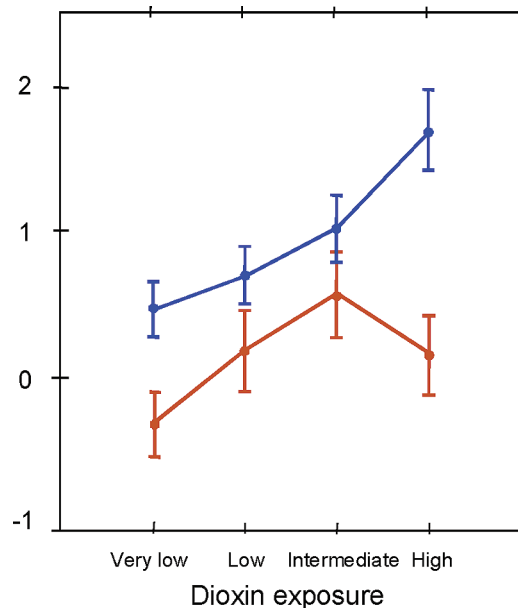
<sup>a</sup> World Health Organization toxic equivalent. <sup>b</sup> Southwest of the municipal solid waste incinerator,  $r^2 = 30.5\%$ . <sup>c</sup> Northeast of the municipal solid waste incinerator,  $r^2 = 52.2\%$ .

exposure was significantly linked to the dependent variable. Conversely, in the simple topography situation ( $r^2 = 52.2\%$ ), a strong gradient across exposure areas was highlighted with a significant association between modeled dioxin exposure and log-transformed dioxin concentration for the upper category ( $p = 0.001$ ). Adjusted means of log-transformed WHO-TEQ concentration, split by dioxin exposure and topography complexity categories, clearly illustrate both the main effects and the first-order interaction (Figure 3).

## Discussion

We found a clear gradient in the soil results, with increasing average log-transformed WHO-TEQ concentration as a function of increasing predicted ground-level air concentration (used as a surrogate for exposure and assessed by a first-generation Gaussian-type dispersion model), in simple terrain.

To our knowledge, this is the only study ever performed with as large a number ( $n = 75$ ) of soil samples, yielding fairly representative and precise estimates. Soil acts as a conservative matrix, reflecting a cumulative PCDD/F deposition during rather long periods of time. After deposition, PCDD/Fs' movement into soils is controlled by the equi-



**FIGURE 3. Adjusted means of log-transformed dioxin concentration per modeled dioxin exposure category (World Health Organization toxic equivalent, 75 sampling points, Besançon, France): (a) blue line, simple topography (northeast of the municipal solid waste incinerator) and (b) red line, complex topography (southwest of the municipal solid waste incinerator).**

librium of sorption/desorption processes between the soil compartments (air, water, mineral, and organic matter). Degradation of PCDD/Fs in soils is considered to be slow or nonexistent under natural conditions (8). Half-life of PCDD/Fs in soils is thought to be over 10 years (7), while other estimates are in the range of 25–100 years in the subsurface and 9–15 years in the top 1 mm of soil (8).

Metrological readings were taken by the same geologist, and quantitative determinations of PCDD/Fs were performed by a single laboratory, so no measurement bias is to be feared. Only about 50% of variance was explained by the linear models, suggesting the role of unmeasured or unknown variables. However, the similarity between geomorphology and ecology profiles across the exposure areas, makes confounding by soil-related factors unlikely.

Keeping in mind that soil concentrations in various locations around a MSWI must be compared with caution (the dispersion of PCDD/F emissions in the atmosphere and their deposition onto soil are governed by numerous factors: MSWI capacity, annual mass emissions, stack characteristics, presence of an air pollution control system, vapor-particle partitioning, ambient air temperature, etc.), PCDD/F concentrations in soil samples at the Besançon site are comparable to levels found in other MSWI sites (12–18).

MSWIs are usually installed in broad industrial parks where other factories are susceptible to taking part in dioxin emissions into the atmosphere, making the sources of PCDD/Fs difficult to discern (19). The studied site is very specific in this respect. There are no adjacent industrial sources of exposure of combustion-effluents; polluting industries were replaced two decades ago by small-scale advanced technologies. Before that time, the main factory (producing synthetic textiles) was located 5 km east of the city in the very low exposure area. No cement kilns, iron or steel works, or foundries were located in this area. To avoid dioxin contamination resulting from automobile emissions (resulting anyway in diffuse, nonspatially organized emissions), all the sampling points were chosen at a distance from main roads. In this respect, the PCA delivered a clear-cut picture. The one-dimensional model reflected high similarities in the

congener profiles of the PCDD/Fs in soil samples. No other additional sources of PCDD/F contamination than the MSWI was, therefore, to be feared.

In no instance was actual individual exposure measured or calculated in this study but estimated through exposure zones based on predicted ground-level air concentrations using a dispersion model. However, the two assumptions required to use geographic exposure indicators (concentrations should differ between areas in the manner expected, and pollutant levels within an area should be relatively uniform) (20) were met to the northeast of the MSWI (Figure 3). Therefore, although modeled ground-level air concentrations were used to assess exposure for a temporal pattern for which they were not designed, we were right to assume that dispersion modeling was heavily influenced by factors that are stable over time (mean meteorological conditions, terrain elevations, stack height) and could adequately reflect past exposure (6).

On the other hand, the standard Gaussian plume straight-line model revealed inappropriate for assessment of exposure on the southwest side (a complex topographic environment). Compared with simple terrain results, the model overpredicted ground-level air concentrations, and if a gradient is discernible across the first three categories, overprediction was more pronounced in the high exposure zone (Figure 3). This bias toward overprediction of ground-level air concentrations on the southwest side is in line with the results of a paper by Hanna et al. (21), showing that ISC3 (a first-generation model similar to APC3) typically overpredicted compared to second-generation models (ADMS or AERMOD). Several limitations of APC3 software can explain these results. First, only a simplified topography has been introduced, ignoring complex terrain with hills and channels. Second, the model assumed that turbulence generated in one place tends not to persist for any significant distance downwind (the so-called equilibrium of turbulence), not accounting for the turbulence boundary layer between surface and air. Third, surface roughness, which affects the vertical profiles of wind and temperature and the dispersion rates in the surface layer, was not accounted for. A short roughness length (0.2 m for open grassland, 2–3 m for arable crops, compared to 5–10 m typical of urbanized areas) (22) leads to significant decreases in particle deposition velocities and, therefore, lower local deposition of PCDD/Fs (23). Moreover, the stack shortness (40 m) represents a further variable making the fraction of PCDD/F emissions that is deposited locally very sensitive to the treatment of dispersion (23). Most of these factors occurred at a relatively higher magnitude in the high exposure zone (the closest to the MSWI and the most sloping) and could explain the more pronounced overprediction in this area.

The subsequent question is whether this overprediction challenges the findings of our case-control study, since it entails a misclassification bias (although nondifferential) for people living to the southwest of the MSWI. We were fortunate that only 10.5% of cases and 9.3% of controls were concerned. Moreover, a logistic regression restricted to cases and controls residing on the northeast side (where the dispersion model appears appropriate) yielded a slightly increased OR in the highest dioxin exposure area (OR = 2.5, 95% CI, 1.4–4.5), compared to our initial finding (OR = 2.3, 95% CI, 1.4–3.8).

This study confirms that first-generation modeling around the local MSWI provided a powerful tool for dioxin relative classification of exposure in simple terrain, reinforcing the results of our case-control study. However, a more advanced atmospheric diffusion model should have been used for refined assessment in the complex hilly terrain located southwest of the MSWI. The local contrasted situation by its very nature is unique. The validation we provide, therefore, does not challenge the fundamental rule of environmental

modeling, which is to not transfer use of a model from one geographic region to another without validating it with measurement data from the new study area (24).

For dioxins, an indirect exposure pathway (ingestion) is considered to account for a very high fraction (90%) of total intake in the general human population (2). Since most of the meats and dairy products are not produced locally but have been transported hundreds or thousands of kilometers, the majority of dioxin exposure does not come from local dioxin sources. Previous studies did not report an increased dioxin body burden in residents around incinerators (25–27). However, local communities whose diets consist significantly of foodstuffs grown/reared in the vicinity of an incinerator may have significantly elevated serum PCDD/Fs levels. Goldman et al. showed that the consumption of both home-produced eggs and meat for 2–15 years was associated with a significant 2- to 6-fold increases in serum levels of dioxin-like chemicals (28). In another study, concentrations of PCDD/Fs in subjects living around two old incinerators in Belgium increased proportionally to their intake of local animal fat, with almost a doubling in subjects with a fat intake higher than 150 g of fat per week (29). Fruit and vegetables grown locally can also become contaminated from incinerator emissions. To our knowledge, no study has attempted to determine whether dioxins emitted by incinerators increase the contaminant body burden of residents consuming locally produced vegetal foods. However, after analyzing fruit (apple) and vegetable (courgette, lettuce, potato) samples, Lovett et al. concluded that consuming these foodstuffs would represent an additional 8% of the normal dietary intake for PCDD/Fs (30).

According to these results, we assume that exposure to air-borne dioxins via the food chain (rather than direct inhalation) might represent a significant pathway for people residing near the MSWI of Besançon. In this semirural setting, the fraction of local foods, resulting from the shopping in popular open markets where locally raised products are sold, can be elevated relative to more urban areas. However, whether this indirect exposure would induce a meaningful risk remains to be determined. To this aim, a survey of PCDD/F congeners in vegetables and eggs and a case-control study in which dioxins are measured in blood are currently underway.

## Acknowledgments

This research was supported by the French Ministry of Health (Grant # 2001-1485). We are grateful to Robert Schwint (former mayor of Besançon) for transmitting the dioxin diffusion model, to Tamara Lindner for assistance with manuscript preparation, and to Amandine Buzzeto for the field sampling.

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Received for review November 16, 2005. Revised manuscript received February 2, 2006. Accepted February 2, 2006.

ES052309U