



Testing the USA EPA's ISCST-Version 3 model on dioxins: a comparison of predicted and observed air and soil concentrations

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Abstract

The central purpose of our study was to examine the performance of the United States Environmental Protection Agency's (EPA) nonreactive Gaussian air quality dispersion model, the Industrial Source Complex Short-Term Model (ISCST3) Version 98226, in predicting polychlorinated dibenzodioxins and polychlorinated dibenzofurans concentrations (subsequently referred to as dioxins and furans, or CDD/Fs) in both air and soil near the Columbus Municipal Solid Waste-to-Energy Facility (CMSWTE) in Columbus, OH. During its 11 yr operation, the CMSWTE was estimated to be emitting nearly 1 kg of CDD/F Toxic Equivalents (TEQs) per year, making it one of the highest single emitters of dioxin in the United States during its operation. An ambient air-monitoring study conducted in 1994, prior to its shutdown in December of 1994, clearly identified high dioxin air concentration in the downwind direction during two sampling events. In one of the events, the CMSWTE stack was concurrently monitored for dioxins. A soil sampling study conducted in 1995/1996 was similarly able to identify an area of impacted soil extending mainly in the predominant downwind direction up to 3 km from the CMSWTE. Site-specific information, including meteorological data, stack parameters and emission rates, and terrain descriptions, were input into ISCST3 to predict ground-level 48-h concentrations which could be compared with the 48-h measured air concentrations. Predicted annual average dry and wet deposition of particle-bound dioxins were input into a simple soil reservoir model to predict soil concentrations that would be present after 11.5 yr of emissions, which were compared to measured concentrations. Both soil- and air-predicted concentrations were generally within a factor of 10 of observations, and judged to be reasonable given the small number of observations and the uncertainties of the exercise. Principal uncertainties identified and discussed include: source characterization (stack emission rates of dioxins), meteorological data, and atmospheric and soil fate and transformation processes of the dioxins © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: ISCST3; Atmospheric fate modeling; Dioxins; Furans

1. Introduction

The Columbus Municipal Solid Waste-to-Energy (CMSWTE) in Columbus, OH, operated between June

1983 and December 1994, and processed an average of 1600 metric tons of solid waste per day during its operation. A stack test taken in 1992 (EERC, 1992) indicated that the annual emission rate of dioxin toxic equivalents (TEQs) was 984 g (TEQs calculated in this paper using the International TEF scheme (EPA, 1989) without including dioxin-like PCBs). Measures were taken to reduce dioxin emissions by the operators of CMSWTE. A second stack test was taken in 1994 (EMC, 1994) to

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evaluate the effectiveness of these dioxin reduction measures. The rate of emission from this test was calculated at 267 g TEQ yr⁻¹, indicating about a 75% reduction in dioxin emissions. These rates of emission can be compared against United States estimates of total annual emissions from all known sources of dioxin release of 12 kg TEQ in 1987 and 3 kg in 1995 (Cleverly et al., 1999).

An ambient air-monitoring study undertaken by the Ohio Environmental Protection Agency (OEPA) included two rounds of sampling in 1994, one during a concurrent stack test, and one round in 1995 after the incinerator had shut down (OEPA, 1994, 1995). For both sampling events in 1994, the concentration was highest in the air monitor (1 of 6 total monitors; 5 operational for each sampling event) located in the downwind direction (southeast) from the CMSWTE. Also, the profile of dioxins in the air matched the stack emission profile much more closely than the other air samples, which had lower and more typical urban air concentrations. A soil-monitoring study conducted during 1995 and 1996 included 34 soil samples taken on-site and up to 8 km in all directions from the plant (Lorber et al., 1998). An evaluation of these soil data clearly showed an imprint from the CMSWTE, with concentrations decreasing as a function of distance from the stack, approaching a local background after about 3 km.

Complete descriptions of the stack, air, and soil measurements conducted around the CMSWTE are available in previous papers (EERC, 1992; EMC, 1994; Ohio EPA, 1994, 1995; Lorber et al., 1998) and only summarized here.

The ISCST3 was run twice, once to obtain predicted concentrations over the 48-h period corresponding to the two ambient air-monitoring events in 1994, and once to obtain annual average wet and dry deposition of sorbed dioxins to input into a simple soil reservoir mixing model to predict dioxin concentrations. The objective of this exercise was to use current EPA guidance on the use of ISCST3 for air dispersion/deposition modeling of dioxins (EPA, 1994–1996), coupled with a soil concentration model, and after doing so, determine how well the model was able to reproduce observed air and soil concentrations. This might be described as a “model validation” exercise because it has these characteristics: (1) all available site-specific information – stack emission rates, meteorological data, stack parameters, and others – are input into the model, (2) all other parameters for which no site-specific data is available – soil half-lives, atmospheric particle densities and mass fractions, and others – are input into the model using best available information with no attempts at “calibration” in order to make the model results best match observations, and (3) model results including predicted air and soil concentrations are compared against corresponding monitored concentrations.

On the other hand, it is recognized that this exercise falls short of a rigorous model validation exercise for ISCST3. The observed ambient air data set includes only two monitoring dates, with five ambient measurements for one date and four for the other date. Actual stack measurements of emissions are available for one of those dates, so a comparison of measured and predicted air concentrations for the second date does not qualify as a “validation” exercise. It must be assumed that emissions were the same for this second air-sampling date. This is admittedly a small data set and a resulting rigorous test for ISCST3 dispersion model testing. Also, ISCST3 (and similar Gaussian dispersion models) is expected to perform better for longer averaging periods (e.g., annual) than for short-term events. Expectations for the “success” or characterization of the “failure” of the ISCST3 dispersion algorithms have to be tempered by these considerations.

The input of the average predicted depositions of dioxins into a simple soil-mixing model to predict soil concentrations, and then comparing those to observed soil concentrations, is an exercise that may come closer to being a “model validation” exercise. In that case, the ISCST3 is applied in an hourly short-term mode over one year’s worth of meteorological data to predict average long-term depositions to soil. These depositions are assumed to occur over the 11.5 yr of operation of the CMSWTE. They are input into a soil-mixing model to predict soil concentrations, and then the model results are compared with observations of soil concentrations. Unlike 48-h air measurements, soil impacts are “long term”, particularly since dioxins are known to accumulate in soils over time and not undergo very meaningful dissipation. Also, there are 34 soil measurements around the CMSWTE available, and this number allowed for a reasonable characterization of the elevation of dioxin concentrations near the facility and the decline of concentration with distance.

In any case, discrepancies between predictions and observations in both the air and soil model comparison exercises were examined in order to gain insight on the capability of the ISCST3 model to predict ambient air and soil impacts of emitted dioxins, and to gain insight on potential issues for further study of atmospheric and soil fate for dioxins.

2. Brief description of dioxin-like compounds

Concentrations of 25 CDD/F compounds were available for use in these exercises: 17 individual dioxin and furan compounds, commonly known as congeners, which have been assigned non-zero toxicity, and 8 homologue groups. Congeners of non-zero toxicity are those with chlorine substitutions in, at least, the 2, 3, 7, and 8 positions, and have been described as “dioxin-like”. The 17

congeners can be combined in a single “toxic equivalent”, or TEQ, concentration by multiplying the concentration of each congener by its corresponding “toxicity equivalency factor”, or TEF, and then summing the resulting products from this calculation for all 17 congeners. The TEF scheme used in this paper is known as the “International” scheme (EPA, 1989). Recently, an updated scheme was proposed by the World Health Organization (Van den Berg et al., 1998). A check on the differences in observed TEQ concentrations for air and soil concentrations of this data set showed them to be within 10% of each other; the older TEF scheme is used here since the data were generated and most of the ISCST3 work done before the newer scheme was available. A homologue group concentration is the total concentration obtained when summing the individual CDD/F congener concentrations with the same chlorine number, whether the congeners are dioxin-like or not. For example, a TCDD concentration is the sum of 22 individual congeners that have four chlorines – only 2,3,7,8-TCDD is considered to have dioxin-like toxicity. “Total concentrations” of CDD/Fs are defined as the sum of homologue group concentrations in this paper.

Modeling efforts in this paper focused on the TEQ concentration and the homologue group concentrations. TEQ concentrations were generated by simulating results for all 17 congeners, and then combining them to calculate the TEQ. Results were generated for the 8 homologue group concentrations – TCDD and TCDF (abbreviated TCDD/F; these homologue groups are characterized by four chlorines), PCDD/F (five chlorines), HxCDD/F (six chlorines), and HpCDD/F (seven chlorines), as well as the single compounds, OCDD and OCDF (eight chlorines), for a total of 10 concentrations defined by their number of chlorine atoms. Fate and transport for the dioxins are often described as a function of their chlorine number, which is why the focus in this paper is on the homologue groups and not the individual CDD/F congeners. For example, lower chlorinated CDD/F compounds partition more into the vapor phase in the ambient air, are more soluble (but still considerably sorbed with $\log K_{ow}$ above 6.0), and more volatile (but still considered “semi-volatile” in the open literature with Henry’s Constants in the range of 10^{-5} atm m³ mol⁻¹) compared to the higher chlorinated CDD/Fs.

3. Modeling procedures

3.1. ISCST3 modeling

ISCST3 is a Gaussian plume model, which accepts a variety of source geometries and emissions schedules in order to compute ambient air concentrations and surface deposition fluxes at specified receptor points. Two applications of ISCST3 were conducted for this effort. In

one, the air dispersion algorithms alone were run, and meteorological data requirements included hourly wind speed, wind direction and stability for describing dispersion. These runs were conducted for the purpose of predicting 48-h air concentrations, to compare with the 48-h ambient air measurements. For the other, the particle-phase deposition algorithms were employed and dioxins were depleted from the plume by an amount equal to that depositing as the plume moved outward from the CMSWTE. The key output from these runs were long term average dry and wet deposition of particle-bound dioxins, which were used for predicting soil concentrations of dioxins, to compare with the soil measurements.

For the dispersion model test, there were, in fact, two separate tests of the air dispersion algorithm – each test had a different meteorological data set: an “airport” set and an “on-site” meteorological data set. The airport set includes surface data (wind speed, wind direction and atmospheric stability) from the Columbus, OH airport and upper air data for the mixing height from Dayton, OH, airport (BEEline Software, Inc. ISCST3 driver diskette 1998). The airport data was applied only for the first of two air-monitoring events. On-site data for both of the 1994 measurement periods include wind speed and wind direction (Ohio Environmental Protection Agency, private communication, 1998). The Columbus stability and Dayton mixing height were used in the on-site set. The purpose of obtaining and testing two meteorological data sets was to be able to evaluate the importance and uncertainty associated with key input stream. Besides meteorological data, other required inputs for modeling dispersion alone included: (1) building configuration data, (2) emissions data, and (3) receptor data. These dispersion model runs omitted particle-phase deposition, plume depletion, and chemical decay in the air. The dioxins were modeled as if the entire emission were in the form of a conservative pollutant, with no differentiation in fate of the individual compounds as a function of vapor/particle partitioning behavior, or atmospheric degradation or transformations. These simplifications are supported by the facts that there was no rain during either sampling event and the monitors were located reasonably close, less than 3 km, from the CMSWTE.

For the deposition application of ISCST3, wet and dry deposition of particle-bound dioxins were modeled and then input to a separate soil-mixing model to predict soil concentrations. Therefore, additional meteorological data required were precipitation data. The prediction of depositions of particle-bound CDD/Fs with ISCST3 relies on particle-specific (e.g. particle diameter) and dioxin-specific (e.g. vapor/particle partitioning) parameters which are not required for dispersion modeling. The ISCST3 model estimates deposition flux values by multiplying the pollutant concentration in airborne particles by a deposition velocity. The deposition velocity is calculated considering gravitational settling velocities

and atmospheric resistance. Annual average depositions were predicted using a single year of meteorological data from 1989; modeling from 1983 to 1994 would obviously have been preferable, but only one year of data was available. Meteorological data was provided by the National Climatic Data Center and from EPA's Support Center for Regulatory Air Modeling internet page (www.epa.gov/ttn/scram). The surface and precipitation data were collected from the Columbus, Ohio Weather Service Office. Atmospheric mixing heights were determined using upper air data collected at the Dayton, Ohio Weather Service Station. An examination of the meteorological data from 1989 compared with historical averages showed that the wind speed and direction were very similar to historical means, and the precipitation was slightly above normal for 1989 (111 cm for 1989 compared to an historical average for Columbus, OH of 96 cm yr⁻¹). The ISCST3 was run in plume depletion mode, meaning that dioxins were depleted from the plume moving away from the incinerator by an amount equal to the dioxins depositing by dry and wet particle-phase deposition.

Like the air dispersion tests of ISCST3, two sets of outputs were generated for soil concentration modeling. There were two stack tests available, and it was unclear as to which would better characterize long-term emissions of dioxins from CMSWTE. Both were used to predict soil concentrations. This is described in more detail in the next section below on source characterization.

The ISCST3 model was run on a "unitized" basis for both dispersion and deposition simulations, meaning that ambient air concentrations and deposition results were generated for an emission rate of 1 g s⁻¹. For the dispersion-only runs, the individual total emission rates of all 25 CDD/Fs (17 congeners on non-zero toxicity and 8 homologue groups) were multiplied by the predicted unit concentration to give the predicted ambient concentrations at the receptor points. Deposition predictions for the CDD/Fs were generated using this two-step procedure: (1) the total amount of the CDD/F emitted was assumed to partition into vapor and particle fractions according to ambient conditions at 20°C (in contrast to partitioning assuming conditions at the stack exit); this step allowed for an estimation of dioxin-specific particle-bound emission rates in g s⁻¹, (2) then, these particle-bound mass emission rates were multiplied by the unitized dry and wet deposition rates predicted to occur at the receptor point to provide the compound-specific deposition rates.

All model parameters for both runs, with the exception of the details on receptor locations (air- and soil-monitoring locations around the CMSWTE) are provided in Table 1. Further detail on modeling algorithms for the ISCST3 can be found in EPA (1995).

3.2. Source term characterization

Two stack tests were available to supply the critical source term for this exercise (EERC, 1992; EMC, 1994). The first was conducted in 1992 by the Ohio Environmental Protection Agency (OEPA) for purposes of permit renewal. High dioxin emissions at 6799 ng total dscm⁻¹ concentration (total = sum of the homologue group concentrations; dscm = dry standard cubic meter) and 976 g TEQ yr⁻¹ (when extrapolating the results from 1 stack to the 3 stacks at CMSWTE and assuming historical average operation times for the CMSWTE) mass emissions were found, leading to regulatory actions by the state and federal environmental agencies. Process modifications were undertaken for purposes of reducing dioxin emissions, and the CMSWTE was retested in March 1994. Total concentrations were reduced to 3685 ng dscm⁻¹ and the mass TEQ emissions were reduced by about 75% to 267 g TEQ yr⁻¹ (estimated using the same historical CMSWTE operation practices).

This second stack test occurred during 16–18 March 1994. This corresponds closely to the time that the OEPA was sampling the air for dioxins – on the 15–17th of March. Therefore, the air dispersion model tests for March used the March stack test results. Unfortunately, the CMSWTE was not stack-tested during the April air-sampling events. It was necessary to use the March stack test results for the April dispersion model tests, and then, of course, to assume that the April emissions were similar to the March stack test emissions.

For deposition modeling, a decision also needed to be made regarding characterization of long-term emission rates. Rather than select either the 1992 or the 1994 stack emission test for this evaluation, or an average of the two, to represent long-term dioxin emission rates, results were generated for both emission tests to demonstrate the importance of this critical and uncertain term in the modeling procedure.

3.3. Soil concentration modeling

Wet and dry depositions are summed and become the source term for a simple reservoir mixing model for predicting soil concentration C_s , as

$$C_s = \frac{F(1 - e^{-kt})}{kM},$$

where C_s is the soil concentration (pg g⁻¹), F is the annual total (wet + dry) deposition of dioxins as predicted by ISCST3 (pg m⁻² yr), k is the first-order annual soil dissipation rate (yr⁻¹), t is the time during which deposition occurs (yr), and M is the soil mixing mass (g m⁻²).

The dissipation rate assumed here for all dioxin compounds was 0.02772 yr⁻¹ (half-life of 25 yr), a mid-range value selected to be between a value of 0.0693 (half-life of

Table 1
ISCST3 and soil model input assumptions and parameters

Description	Parameter value and comments				
I. ISCST3 model inputs					
Source characterization	1992 stack test: 6799 ng m ⁻³ total; 136 ng/m ⁻³ TEQ emission concentration from tested stack; <i>extrapolated to:</i> 3.12 × 10 ⁻⁵ g TEQ s ⁻¹ emission rate considering three stacks, equal to 985 g TEQ yr ⁻¹ 1994 stack test: 3685 ng m ⁻³ total; 64 ng/m ⁻³ TEQ emission concentration from tested stacks; <i>extrapolated to:</i> 8.47 × 10 ⁻⁶ g TEQ sec ⁻¹ emission rate, equal to 267 g TEQ yr ⁻¹				
Dispersion coefficients	Rural				
Terrain	Flat				
Regulatory default option	Yes				
Stack tip downwash	Yes				
Final plume rise	Yes				
Buoyancy induced dispersion	Yes				
Wind profile exponents	Regulatory defaults				
Calm winds processing	Calm hours not included in conc. calculations				
Vertical potential temp. gradient	Regulatory defaults				
Decay coefficient	0 (no decay of contaminant in plume)				
Building wake effects	Building dimensions were input to the model				
Wind speed/stability category	Regulatory defaults				
Wet/dry particle-phase deposition	Yes				
Wet/dry vapor-phase deposition	No				
Plume depletion by deposition	Yes				
Building height/stack height	36 m, 83 m				
Stack temperature	434 K				
Exit velocity	5.5 m s ⁻¹				
For deposition modeling only	Diam. (μm)	Mass fraction	Density (g/cm ³)	Scav. coef. (liq) 1/(s-mm hr ⁻¹)	Scav. coef. (ice) 1/(s-mm hr ⁻¹)
Particle Category 1	1.00	0.88	1.4	0.000	0.00014
Category 2	6.78	0.09	1.4	0.0046	0.0016
Category 3	20.0	0.03	1.4	0.0066	0.0022
II. Soil modeling inputs					
Soil half-life (yr)	All homologue groups assume 25 yr half-life				
Particle fraction	TCDD: 0.49; PCDD: 0.87; HxCDD: 0.97; HpCDD: 0.99; OCDD: 0.998				
(vapor fraction = 1-particle fraction)	TCDF: 0.53; PCDF: 0.80; HxCDF: 0.945; HpCDF: 0.985; OCDF: 0.998				

10 yr) often assumed for surficial dioxin residues (EPA, 1994) and 0.00693 (half-life of 100 yr) speculated to be an upper range for subsurface dioxin residues (Paustenbach et al., 1992). The best justification for this choice of half-lives for all dioxin congeners comes from McLachlan et al. (1996), who reported on an analysis of dioxin soil concentration measurements from experimental plots which had been amended with sewage sludge in 1968 and sampled in 1972, 1976, 1981, 1985, and 1990. These archived samples were analyzed for all 17 dioxin-like CDD/Fs, and based on an analysis of results, McLachlan and coworkers concluded that half-lives were on the order of 20 yr, with dioxin removal from the plots being mainly physical removal processes (overland runoff, wind erosion). Furthermore, their results suggested that all

congeners had been removed at roughly the same rate, which is why they concluded that removal processes were mainly physical and very little in situ degradation appeared to be occurring.

A time of operation, *t*, of 11.5 yr was used, corresponding to the time of operation of the CMSWTE. The soil mixing mass, *M*, equaled 112,500 g m⁻², which assumes a mid-range soil bulk density of 1.5 g cm⁻³ and the soil sampling depth of 7.5 cm.

3.4. Description of the measured air and soil concentrations

Ambient air monitoring was conducted by the Ohio Environmental Protection Agency (OEPA) in 1994 to

evaluate ambient air concentrations after process modifications reduced dioxin emissions from the CMSWTE. General Metal Works model PS-1 high-volume samplers were used to collect 48-h samples. Concentrations were, therefore, the sum of vapor + particle phase concentrations. Six monitors were in the city of Columbus between 1.8 and 3.0 km from the site, mostly in the historical downwind direction, northeast, but one in the upwind southwest direction. Two of the samplers were co-located (for purposes of quality control), so results from these two samplers were averaged to represent one sampling point. A seventh sampler was located 45 km southwest of the facility in a rural “background” setting; results from this sampler were not used in this modeling study. Five samples (4 sampling locations; the co-located samples were averaged) were taken in March and 6 samples (5 locations) were taken in April, 1994. The March set, taken on the 15–17th of the month, occurred at nearly the precise time that the March 1994 stack testing occurred, on the 16–18th. The April sampling event occurred during 19–21 April. Exact starting and stopping times of the air monitors were not available for this test. For purposes of air dispersion modeling, the starting and stopping times were assumed to be the mid-day of the beginning and ending days of each sampling periods. In all, there were nine urban air concentration measurements available during 1994 (again, 4 in March and 5 in April) that comprise the “observed” air concentration data set used in this modeling study. Wind rose data for the March and April sampling periods were also available, and the provided insights into the expected impact patterns. A final round of air samples from the seven air monitors was taken in 1995 after the CMSWTE had shut down. The purpose of this data set was to evaluate the air quality now that the CMSWTE was no longer operating. Full details on the air-monitoring studies, including analytical methodologies, quality control, and final results, are described in OEPA (1994, 1995).

A first phase of soil sampling was conducted by the United States Environmental Protection Agency (EPA) in December of 1995. Sampling in this round included 4 samples on the site of the incinerator, 18 samples within about 3 km of the incinerator in the city of Columbus, and 3 samples at a background site 45 km from the CMSWTE. This background site was the same as the air-monitoring background site. The study design for this phase employed a stratified random selection process, involving sites in the four major quadrants around the incinerator (northeast, southwest, etc.) with an emphasis of sampling in the quadrant which was historically downwind from the incinerator, the northeast quadrant. The following conditions were sought during site selection: (1) level, undisturbed soils, (2) away from trees, (3) not adjacent to roads, (4) not near pressure treated wood, and (5) not known or suspected to have high dioxin concentrations for any other reason. All samples were collected

using pre-cleaned equipment dedicated to each sampling location. Each sample site consisted of an area of 1.5 m × 1.5 m. A grid of 25 sections was established at each site and used for random selection of aliquot sample sites. Four random aliquots were collected for each sample. A “sample” for this study was, therefore, a composite of four aliquots. Aliquots were collected using a stainless steel tulip bulb planting device. This device removed a plug approximately 7.5 cm in diameter to a depth of about 7.5 cm.

A second phase of soil sampling was undertaken in August of 1996. Thirteen samples were taken from about 2 km away from the incinerator to about 8 km distant. The purpose of this second phase was to ascertain whether a background concentration for the city of Columbus could be determined. A similar selection criteria for sample sites was employed in this second phase.

Altogether, there were 4 soil samples on the incinerator property, 31 samples in the city of Columbus taken from right outside the incinerator to upwards of 8 km away, and 3 background samples taken 45 km away, for a total of 38 soil samples. This modeling used 34 of the samples – it did not have use for the 3 background samples, and 1 of the remaining sample was found to be contaminated by a local source not associated with the CMSWTE. Full details of the soil-monitoring study can be found in Lorber et al. (1998).

Fig. 1 shows the location of the CMSWTE in relation to the 32 soil samples in Columbus and the five urban air-sampling locations. Not shown in this figure are 3 of the 4 soil samples taken on the site of the incinerator, and the background site in which 3 soil samples were taken and the 1 background air sampler was located. This figure identifies the groupings of the soil samples, as described Section 4.

3.5. Subtracting local background concentrations from measured concentrations

The ISCST3 and soil-mixing models used in this effort will predict only the increments of dioxin concentration in the air and soil that are due to emissions from the CMSWTE. Therefore, a procedure had to be developed to subtract a local “background” of dioxins from both the air and soil-observed data. Only then can one appropriately compare predicted and observed concentrations.

The average of six air measurements taken in 1995 after the CMSWTE shut down was assumed to represent the background dioxin air concentrations for this site. The average total concentration from 1995 was 2870 fg m⁻³, with a range of 2030–4760 fg m⁻³. The 1995 average concentrations of each dioxin-like congener as well as those of the homologue groups were subtracted from each of the March and April 1994 corresponding measurements. When such a subtraction resulted in a concentration less than 0, the concentration was assumed to be 0 for purposes of this exercise.

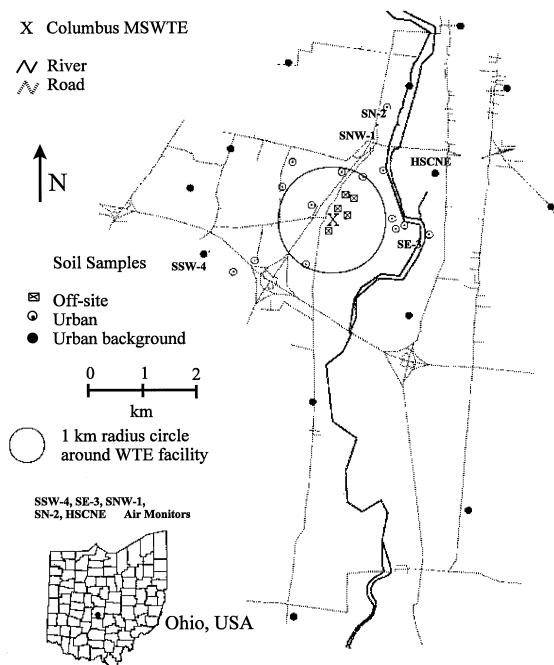


Fig. 1. Schematic of the Columbus MSWTE showing location of the incinerator, soil- and air-monitoring sites.

An analysis of the observed soil data in Lorber et al. (1998) showed that concentrations decrease to the local soil background at about 3 km from the CMSWTE, at a TEQ soil concentrations of 4.0 pg g^{-1} (ppt). The soil profile of CDD/Fs for this background provided in Lorber et al. (1998) was subtracted from each of the 34 observed soil measurements; when this subtraction resulted in a concentration less than 0, the concentration was set to 0.

For both soil and air, the possibility exists that there is a very localized source leading to elevations in some of the presumed “background measurements”. This may be particularly true for the air measurements. As noted, there was a range of concentrations between 2000 and nearly 5000 fg m^{-3} observed in 1995. To simply average the six air measurements and subtract the average from all 1994 measurements neglects the possibility that some of the higher measurements in this range could reflect nearby sources. Therefore, while it is necessary to subtract a “background” dioxin concentrations in air and soil measurements in order to characterize the signature of dioxins that can be attributed to the CMSWTE, the procedures used here may not have been ideal.

3.6. Procedures for evaluating the performance of the models

The paucity of the observed data, particularly the air measurements, makes a rigorous “goodness-of-fit” stat-

istical comparison of predicted versus observed inappropriate. Rather, tabular summaries of predicted and observed concentrations are utilized, and simple qualitative discussions address the goodness-of-fit. For the air dispersion comparisons, predicted air concentration quantities associated with a 48-h air-monitoring event are compared with the appropriate observed quantities. For the deposition comparisons, soil samples are “clustered” and simple mean concentrations are generated for both modeled and observed concentrations. Four clusters which are displayed include: (1) “on-site” – 3 soil samples taken on the site on the CMSWTE, (2) “off-site” – 5 samples just off-site and in the historical downwind direction, northeast, within 500 m of the incinerator, (3) “urban” – 14 samples taken from about 500 m to about 3 km, and (4) “urban background” – 12 samples taken from about 3 to about 8 km. As discussed in Lorber et al. (1998), the high soil concentrations found in the on-site cluster were speculated to have resulted in ash drift from piles or trucks transporting the ash to nearby landfills rather than deposition. Therefore, a comparison of predicted and observed concentrations for this on-site cluster are displayed for information purposes only, not to be considered in the context of model testing. Otherwise, all observed soil samples, and clusters, can be considered to represent long-term deposition trends as the monitoring study protocols insured that they were in relatively flat, undisturbed locations away from any nearby potential dioxin sources (roadways, PCP treated wood, etc.). The predicted and observed concentration quantities which are displayed include: (1) homologue group concentrations, (2) total concentrations, which are sum of the 10 homologue group concentrations, and (3) TEQ concentrations. These terms were defined above.

In addition to tabular summaries, isoline figures were generated. These are lines of equal concentration around the CMSWTE, either air or soil concentrations, that were generated using ArcView® – a desktop GIS package. First the point data, measured or modeled concentrations, are brought into ArcView® as point coverages. Then, using the ArcView® kriging routine, surfaces of the concentrations are generated using the exponential function to estimate the semivariogram. For the air and soil concentration isoline generation, modeled concentrations were generated for 250 m intervals to about 3 km in all directions, and these were input as point coverages into ArcView®. There were too few observed air concentration measurements, so isolines could not be generated for these. Instead, measured air concentrations were overlain on the predicted isolines. For soil concentration, there was judged to be sufficient coverage with 34 soil samples to generate “observed” isolines to compare with predicted isolines.

Finally, it is reiterated that all “observed” concentrations, both soil and air, were generated by subtracting out background concentrations in the procedure

described above. Therefore, all tabular or figure notations of a “0” observed concentration means that, if subtracting out the background concentrations from the measured concentrations resulted in a negative concentration, the measured concentration was set to zero for purposes here.

4. Results and discussions

4.1. Air dispersion modeling

Even before air dispersion modeling was undertaken, examination of the data revealed clear trends. Analyses of on-site wind roses for the March and April 1994 sampling dates reveal that there is one dioxin monitoring station likely to have been influenced by the CMSWTE. This station, termed SE-3 by the Ohio Environmental Protection Agency (OEPA) was about 2 km east of the source and was downwind from the source approximately 53% of the time during the sampling period in March 1994 and 78% of the time in April 1994 (OEPA, 1994). In contrast, none of the other 5 stations was downwind for time fractions approaching those of SE-3. The measurements confirmed that SE-3 was the most impacted of the samplers, with TEQ measurements of 168 fg TEQ m⁻³ in March and 353 fg TEQ m⁻³ in April. The average of the measurements from the other 5 samplers over the two dates (a total of 8 samples; one sampler was not operational for both events) was 52 fg TEQ m⁻³, with a range of 10–98 fg TEQ m⁻³ (*note*: background not subtracted out for these observations). Lorber et al. (1998) examined this trend further, showing also that the profile of CDD/Fs found in March and April in SE-3 matched the stack emission profile of CDD/Fs more closely than the other ambient air samples, which displayed profiles more typical of background air.

Table 2 compares the observed total concentrations at each reporting monitoring station with the model predictions for both meteorological data sets, the “on-site” and

“airport” sets. As noted earlier, two important considerations for evaluating the comparison of predicted and measured air concentrations are: (1) having 4 and 5 air measurements for sample dates in March and April, respectively, is a small sample size, and (2) one can expect the ISCST3 to perform better for longer averaging times as compared to shorter averaging times. It would be fair to conclude that the paired comparisons of predicted and observed 48-h air concentrations are severe tests of model performance, and Table 2 shows the large scatter expected from this test.

Still, some meaningful observations might be possible from Table 2. First, it does not appear that either of the meteorological data sets provides a superior fit between observed and predicted air concentrations for the March sampling event. The model runs using on-site meteorology appeared to better identify SE-3 as the monitor of most impact, and also to identify the sampler SN-2 as having some impact, but not as much impact as SE-3. The model runs using airport meteorology set appeared to show a significant impact to SN-2, but not as much of an impact for SE-3. On the other hand, the simulations using on-site meteorological data identified SNW-1 as having the highest concentrations, while the simulations using the airport data correctly modeled this site perhaps more correctly as having little impact. Model runs using both meteorological data sets correctly identified SSW-4 as the monitor which showed no impact.

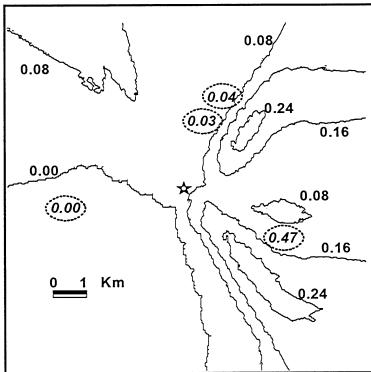
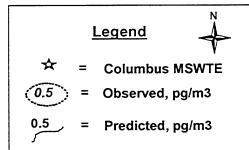
For the April sampling date, the on-site meteorological data correctly identified SE-3 and SSW-4 as monitors having some impact, with little or no impact for the other three monitors. However, SE-3 was not simulated to have the most impact, as was found.

The difference between using the on-site and air meteorological data was further examined using isoline figures. Six such isoline drawings, with observed concentrations overlain, are shown in Fig. 2. These include TCDD, OCDD, and TEQ predicted and observed results from March for simulations using the on-site and the

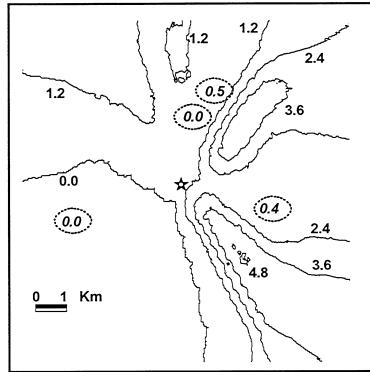
Table 2

Comparison of observed and modeled total CDD/F concentration increments at the urban monitoring stations (total = sum of homologue group concentrations; on-site, airport = model results generated using on-site and airport meteorological data; NA = not available)

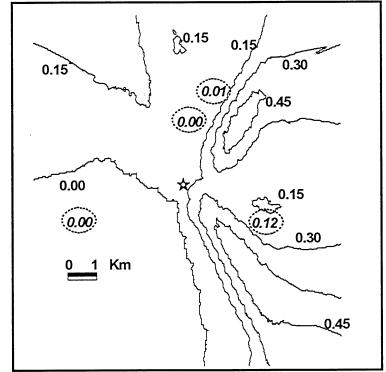
Station	March 94 Sampling, fg m ⁻³			April 94 Sampling, fg m ⁻³	
	Observed	On-site	Airport	Observed	On-site
SN-2	1321	6606	20833	0	0
SE-3	6368	8181	2388	16105	8994
SNW-1	0	8943	1270	557	0
SSW-4	0	0	0	3682	8638
HSCNE	NA	NA	NA	1493	8028



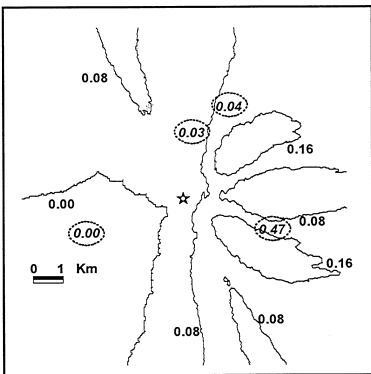
(a) TCDD, March, On-site



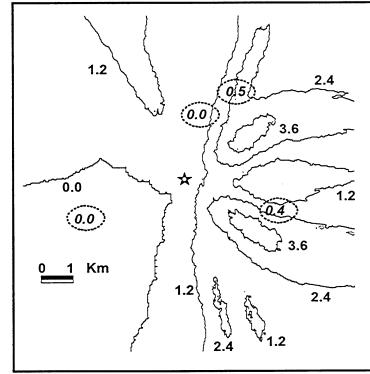
(b) OCDD, March, On-site



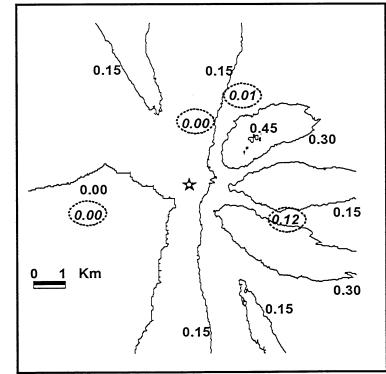
(c) TEQ, March, On-site



(d) TCDD, March, Airport



(e) OCDD, March, Airport



(f) TEQ, March, Airport

Fig. 2. Isoline figures of predicted air concentrations overlain by measured air concentrations of TCDD, OCDD, and TEQ (pg m^{-3}) when using the “on-site” meteorological data set figures (a-c) and when using the “airport” meteorological data set (d-f).

airport meteorological data sets. Observations from this figure include:

- (1) ISCST3 modeling runs using both meteorological data sets appear to have correctly identified the western quadrants (northwest and southwest) as being areas of little impact. The observed “0” concentration in the southwest quadrant (sampler SSW-4) is consistent with this trend, as are wind rose that are displayed and discussed in OEPA (1994). Both figures appear to have identified the northeast and southeast as areas of principal impact, with little impact due

- east. The two observed air measurements in the northeast quadrant (samplers SN-2 and SNW-1) do, in fact, suggest low impact due north with increasing impact as one moves in the northeast direction. The observed air measurement in the southeast quadrant (sampler SE-3) may, in fact, have missed areas of higher impact during the two days, which the model runs suggest are either further north or further south.
- (2) The discussion above comparing the measured point estimates with the modeled point estimates suggests that there may be significant differences in the way the two meteorological data sets simulated impacts.

Looking at Fig. 2, however, the differences do not appear that meaningful. The on-site simulations seemed to push the plume a little more northeast and southeast compared to the airport runs, which showed more impact due north and south. Sampler SN-2 was simulated to be in a zone of important impact according to the airport meteorological data set, while sampler SNW-1 was simulated to approach this zone more so with the on-site meteorological data set. These types of trends emphasize the potential problems and misinterpretations that can occur when one attempts a validation exercise with ISCST3 with short-term data and a limited number of air measurements.

- (3) The biggest discrepancy for the three dioxin quantities compared in Fig. 2 is for OCDD. It appears as though much higher concentrations, ranging from 1 to almost 5 pg m^{-3} in the northeast and southeast were modeled, while measurements of 0.5 pg m^{-3} and less were observed. These high-modeled OCDD concentrations were the main reason that the ISCST3 modeled much higher observed total concentrations than were measured (see Table 2). For the other quantities, TCDD and TEQ, while the location of high impacts may not have been perfectly identified, at least the magnitude of the high-measurements were in the range of the high modeled concentrations. This important trend is discussed in more detail below.

As mentioned above, station SE-3 stands out in both the March and April sampling as having the highest impact of all stations. Thus, the data from this station have the best chance of avoiding the uncertainties introduced by background fluctuations. Predicted and observed homologue group concentrations for SE-3 for the

both sampling dates are compared in Table 3. These results were generated using on-site meteorological data. Table 3 also shows the CMSWTE stack emission rate of these homologue groups.

Being only an exercise in air dispersion modeling (no wet/dry deposition; no stack speciation; no atmospheric chemical reactions), there is a perfect correlation between the homologue profile of the emissions and air concentration predictions for both March and April. The observed air concentrations clearly do not have this stack emission profile, however. Specifically, the speciation pattern from source to receptor has shifted in these ways: (1) the lower chlorinated tetra and penta CDD/Fs have greatly magnified in importance in the ambient air profile as compared to the stack profile, and (2) conversely, the hexa through octa homologues, with the exception of OCDF, have been reduced in importance in the ambient air profile as compared to the stack profile. Said another way, the model predicted lower concentrations for the lower chlorinated CDD/Fs than were measured, and higher concentrations for the higher chlorinated CDD/Fs. The total concentration predictions were, however, within about a factor of two of observations. Not that it has meaning with regard to fate and transport considerations, but the TEQ concentrations were comparable: 125 and $309 \text{ fg TEQ m}^{-3}$ measured during March and April compared with 142 and $156 \text{ fg TEQ m}^{-3}$ modeled for SE-3.

Three possible explanations are offered to explain why the model did not predict the measured shift in homologue profile between the stack and field:

- (1) It is known that CDD/Fs with fewer chlorines have higher vapor-to-particle (V/P) ratios (Bidleman, 1988; EPA, 1994); indeed, high temperatures in the stack

Table 3

Comparison of observed and modeled homologue and TEQ concentrations at station SE-3 using on-site meteorological data for model input

Homologue group	Stack emission rate (ng dscm^{-1})	SE-3, March 94 fg m^{-3}		SE-3, April 94 fg m^{-3}	
		Observed	Modeled	Observed	Modeled
TCDD	32	490	71	851	78
PCDD	97	594	215	1144	236
HxCDD	300	543	666	1402	732
HpCDD	508	424	1126	1378	1237
OCDD	578	384	1281	1575	1408
TCDF	293	904	651	1976	716
PCDF	439	1226	977	2982	1074
HxCDF	648	951	1439	2518	1582
HpCDF	616	718	1366	1846	1502
OCDF	170	134	391	433	429
Total	3681	6368	8181	16105	8994
TEQ	64	125	144	309	156

could generate even higher V/P ratios (Eschenroeder et al., 1994). If stack sampling methods underestimate the amount of vapor pollutant being emitted, then the lower chlorinated dioxin emission rates are being underestimated – an error that would be exacerbated by the even higher V/P ratios in the high temperature stack gas. The PS-1 samplers capturing both vapor and particle-phase CDD/Fs in ambient air are well tested and not expected to have caused error in the characterization of total ambient air concentrations of dioxins. There has been some speculation that PS-1 samplers may overestimate the vapor fraction of dioxins (EPA, 1994), but this would not affect their characterization total concentrations (sum of vapor and particle phase concentrations).

(2) Running the air dispersion algorithms of ISCST3 alone did not account for particle deposition, yet some of the higher chlorinated CDD/Fs, expected to be sorbed to ambient air particles or fly ash, may have deposited by dry deposition prior to the air-sampling locations (since no rain fell during the sampling events, wet deposition could not be an explanation for the results). The results for the deposition modeling described below support this hypothesis, at least for the dioxins – less so for the furans. It compares model predictions of soil concentration with measured soil concentrations. One clear trend was that the model consistently underpredicted the soil concentration of the hepta and octa dioxin

homologue groups. This result, combined with the observation that the higher chlorinated dioxins were the most overpredicted in air concentrations in this paper, suggests that the plume is being depleted of higher chlorinated dioxins by deposition. However, this trend was not duplicated by the higher chlorinated furans. There, modeled soil concentrations were more nearly consistent with measured soil concentrations, with a small degree of overprediction.

(3) Another possible physical explanation is that dechlorination may occur between the emission point and the ambient measuring station a kilometer or two downwind. Workers at Monsanto Laboratories (Orth et al., 1989) and at the Agro-Environmental Science Institute in Japan (Koshioka et al., 1989) have observed photolysis of TCDD. Generally, polychlorinated organic compounds easily experience photochemical loss of chlorine atoms. If the higher chlorinated CDD/Fs dechlorinated to form lower chlorinated CDD/Fs in the atmosphere, than more lower chlorinated CDD/Fs would have arrived at the ambient air-monitoring stations to cause the distinct ambient air profile.

4.2. Deposition and soil concentration modeling

Table 4 provides results from this exercise, which are observed and predicted homologue and TEQ concentrations for four clusters of soil samples. These clusters were

Table 4

Results of ISCST3 deposition and soil prediction modeling, comparing measured concentrations for clusters of soil samples with modeled concentrations assuming either the 1992 or the 1994 stack tests^a

Cluster ->	On-site			Off-site			Urban			Urban		
	Description of cluster ->			Description of cluster ->			Description of cluster ->			Description of cluster ->		
Homologue	Obs	'92	'94	Obs	'92	'94	Obs	'92	'94	Obs	'92	'94
TCDD	1118	265	19	98	93	7	19	38	3	< 1	9	< 1
PCDD	1820	815	102	64	286	35	13	117	15	2	29	4
HxCDD	1885	1202	351	150	421	123	43	173	51	4	43	13
HpCDD	1666	781	606	654	273	212	154	112	87	20	28	21
OCDD	1431	445	696	2901	156	243	613	64	100	150	16	25
TCDF	2147	1304	187	153	457	66	35	188	27	2	47	7
PCDF	255	2335	425	194	818	149	33	336	61	5	83	15
HxCDF	1195	2769	740	116	970	259	22	399	107	3	99	26
HpCDF	1183	1079	732	193	378	256	37	155	105	5	39	26
OCDF	222	274	212	88	96	74	15	40	31	3	10	8
Total	12922	11269	4070	4611	3948	1424	984	1622	587	194	403	146
TEQ	466	236	69	45	83	24	9	34	10	< 1	8	2

^aSoil concentrations in pg g⁻¹, obs = observed; '92, '94 = ISCST3 results using 1992 and 1994 stack test data; "on-site" observed data not expected to represent deposition trends — see text for more details.

developed for purposes of displaying results from the soil-monitoring study conducted around the CMSWTE (Lorber et al., 1998), and generally correspond to increasing distance in all directions from the incinerator. As discussed above, the observed and predicted soil concentrations for soil samples taken on-site, the first cluster of Table 4, are shown for informational purposes only; it is not expected that the on-site soil samples represented long-term deposition trends. Some trends that may be observed from the results in Table 4 include:

- (1) Since emission rates between the 1994 and 1992 stack tests differed by about a factor of 4, subsequent predictions of soil concentration made with each stack emission rate also differed by this factor of 4. Generally, the 1994 stack test predictions appear to better match the observed soil concentrations compared to the 1992 stack test with all homologue groups except TCDD; the TCDD predictions using the 1992 stack test are a better match. Most of the time, however, both sets of predicted homologue group soil concentrations were higher than observed soil concentrations, sometimes by more than a factor of 10 when using the 1992 stack test.

The question that this study is unable to answer is which stack test is more likely to have been representative of long-term emission trends from the CMSWTE. The 1994 test occurred specifically after measures had been taken to reduce dioxin emissions. Because of process changes made to the CMSWTE, it would be reasonable to assume that the 1994 test is not representative of long-term emissions. On the other hand, the 1992 test was occurring during heavy rainfall, which soaked the refuse to be burned. Data on the refuse moisture content showed that the average moisture content of the refuse burned in 1992 was about 10% higher than in 1994 – it was about 38% during the 1992 test compared to 28% in 1994. Some have suggested (K. Jones, personal communication, Zephyr Consulting, Seattle, WA) that wetter refuse may result in higher dioxin emissions, although this hypothesis is unproven and the moisture content of feed materials is not considered to be a principal factor in predicting dioxin emissions – factors such as feedstock content, combustion efficiency, pollution control device, and pollution control inlet gas temperature are more often cited as the critical factors.

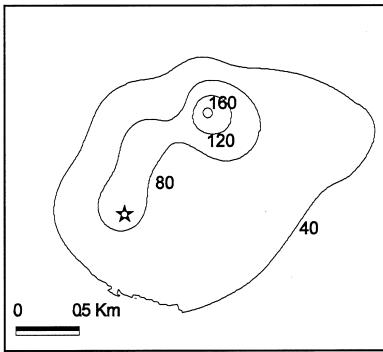
- (2) Noteworthy for results with both stack tests is that much more OCDD is found in the soil than predicted, and the same is true but to a lesser, although still noticeable, extent with HpCDD; in other words, the model under-predicted the soil concentrations of these homologue groups. As noted above in the description of air dispersion results, the ISCST3 was found to have greatly over-predicted OCDD and HpCDD ambient air concentrations. Taken together,

these trends suggest that OCDD and HpCDD deposited near the incinerator to a much greater extent than was modeled. Since both dioxin homologue groups exist in the atmosphere principally sorbed to particles, this may reflect inappropriate parameter assignments relating to particle phase deposition algorithms, or possibly inappropriate deposition algorithms in general. However, the model appears to *overpredict* OCDF and HpCDF, and like OCDD/HpCDD, OCDF and HpCDF are also tightly sorbed to airborne particles, so perhaps the model's treatment of particle fate may not be the cause of significant underprediction of OCDD. Another explanation could be that the particles to which HpCDD/OCDD are attached (fly ash?) are simply distinct from the particles to which HpCDF/OCDF are attached.

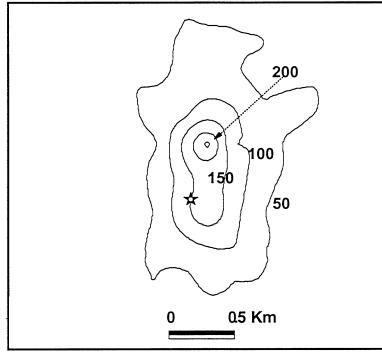
- (3) With both stack test results, the model would appear to proportionally overpredict most congeners (not OCDD/HpCDD) to a greater degree the further downwind one gets. This suggests that more dioxin mass is being removed from the plume as it disperses downwind than ISCST3 is able to simulate. Removal mechanisms include particle and vapor-phase deposition, plant capture, and atmospheric degradation (photolysis and photooxidation).

Fig. 3 shows a series of nine isoline maps crafted to additionally display the trends of the measured versus the modeled soil concentration. Each group of three isoline maps pertains to one CDD/F compound; there are three isoline figures each for TCDD, OCDD, and TEQ concentrations. The first in the sequence of three are isoline maps drawn from the measured data, and the next two are maps drawn from using the 1992 and then the 1994 stack test. Observations from Fig. 3 include:

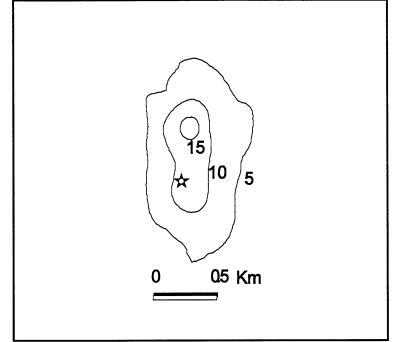
- (1) The shape of the isoline figures developed using the 1992 stack emission test will be the same as those developed using the 1994 stack emission test, because they were all developed from the same unitized simulation – the only difference will be the mass of particle-bound emissions as a function of the compound and stack test.
- (2) The observed maximum soil concentration appears to occur in the northeast quadrant about a kilometer away. The predicted maximum soil concentrations are also found in the northeast quadrant, but they are a bit closer, at about $\frac{1}{2}$ kilometer away. Also, the isolines drawn from model simulations seem to suggest that the maximum will occur more due to north of the CMSWTE as compared to isolines drawn from measured data.
- (3) As was noted above, these isolines suggest much higher OCDD concentrations, in the thousands of parts per trillion (or equivalently, parts per billion),



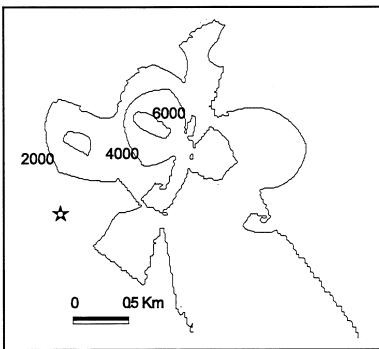
(a) TCDD, Observed



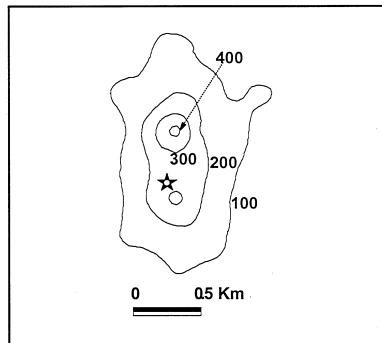
(b) TCDD, '92 Stack Test



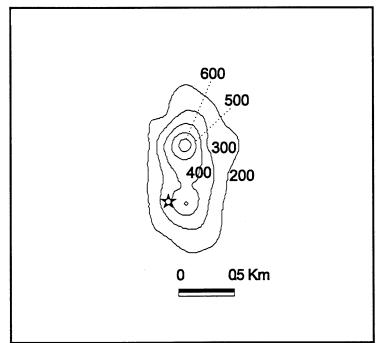
(c) TCDD, '94 Stack Test



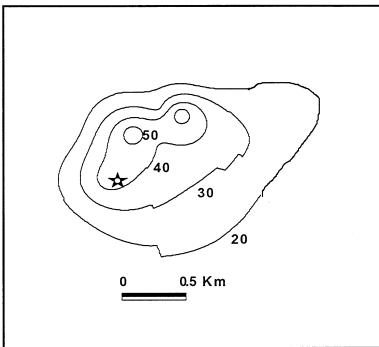
(d) OCDD, Observed



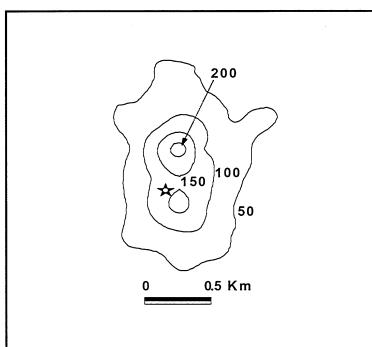
(e) OCDD, '92 Stack Test



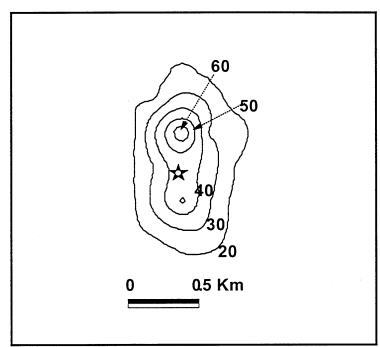
(f) OCDD, '94 Stack Test



(g) TEQ, Observed



(h) TEQ, '92 Stack Test



(i) TEQ, '94 Stack Test

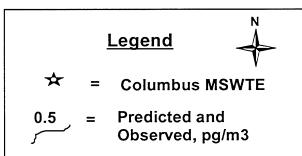


Fig. 3. Isoline figures of predicted soil concentrations of TCDD, OCDD, and TEQ (a, d and g) compared against isoline figures of measured soil concentrations using the 1992 stack emission test (b, e and h) and the 1994 stack emission test (c, f and i).

are found near the CMSWTE, as compared to modeled OCDD concentrations, which are in the hundreds of parts per trillion. It is noted that smooth isolines could not be drawn from the observed OCDD data because of the inhomogeneity of the results. Specifically, of the 8 highest soil samples nearest the CMSWTE, 5 had observed concentrations of OCDD above 1900 pg g^{-1} , ranging from 1930 to 6651 pg g^{-1} , but the other 3 measurements were less than 1000 pg g^{-1} , ranging from 309 to 731 pg g^{-1} . The observation that average OCDD was elevated in soils well above 1000 pg g^{-1} is supported by the data, despite oddly shaped isolines. The observed TCDD concentrations in the vicinity of the CMSWTE appear in range from 40 to a peak of 160 ppt, which is close to the range of 50–200 ppt modeled when using the 1992 stack emission test. However, when using the 1994 stack emission, the elevation in TCDD is only suggested to be in the 5–20 ppt range. Although not meaningful with regard to fate and transport, *per se*, the 1994 stack test appears to better duplicate the observed range of elevated TEQ concentrations — between 20 and 50 ppt, while the 1992 stack test simulations suggest elevations as high as 200 ppt TEQ.

5. Discussion and concluding remarks

Caution was expressed in the opening paragraphs that this exercise should not be characterized as a “model validation” exercise, mainly because of the weaknesses and uncertainties in the data and model parameters. To reiterate, some of those weaknesses/uncertainties include: (a) a very small number of observed air-monitoring data points (and the lack of precise information on when the air monitors were turned on and off, which can be important for short-term air dispersion testing), and a relatively small number of soil measurements, (b) the lack of consideration of all possible plume depletion mechanisms in the dispersion and deposition modeling. For the deposition modeling, the plume depletion by particle-phase deposition was considered, but other plume depletion mechanisms include atmospheric degradation of either vapor- or particle-phase dioxins, vapor-phase deposition, and vapor- and particle-phase vegetative capture, (c) a reasonable but still possibly flawed means to subtract “background” concentrations from measured air and soil concentrations, (d) uncertainties in dioxin-specific fate parameters including vapor/particle partitioning of the CDD/Fs and soil half-lives, and (e) uncertainties and/or lack of representativeness in the important source term, the rate of dioxin emissions from the stack, and the equally important meteorological data used to drive the model simulations.

These latter uncertainties in source term and meteorological data were evaluated by using different data sets.

Specifically, two meteorological data sets were used in the dispersion modeling exercise — an “on-site” meteorological data set supplied by Ohio EPA (who took the air samples), and a publicly available data set from a nearby airport. Two possible stack tests were used to characterize long term emission rates for deposition and soil concentration modeling. As discussed above, there were no clear “superior” choices in either meteorological data set or stack emission test. While it was clear that air- and soil-concentration results differed when using both data sets, in fact it was also clear that both data sets seemed to predict some quantities better than the counterpart data set.

With these cautions, it may be fair nonetheless, to make these statements regarding the ability of the ISCST3 to model the impact of dioxin emissions from the CMWSTE:

- (1) Elevations of dioxins in air and soil due to emissions from the CMWSTE were clearly identified in the sampling programs, and they were also clearly modeled by ISCST3. Predicted and measured dioxin elevations in air and soil appear to generally be within a factor of 10 of each other, with both under and over predictions identified above. These elevations appear to be restricted to only within a few kilometers, 2–3 km, and this was also found in the dispersion and deposition modeling. The ISCST3 correctly identified the north/northeast quadrant as being the one with elevated soil concentrations.
- (2) The small number of air concentration measurements made the comparison of predicted and measured air concentrations a tenuous exercise — the model didn’t always correctly identify locations of high and low air concentrations. Use of isolines was helpful in displaying model performance: it showed how no impacts were modeled for the western quadrants (only one air monitor was able to verify this trend), and how two air samplers appeared to be right on the border of where air impacts may have been occurring. From the soil-modeling exercise, it appears as though the model overpredicted soil concentrations to a greater degree the further downwind one went. This suggests that the plume was being depleted by dioxins in a manner that was not duplicated by the ISCST3 modeling.
- (3) It is clear from the analysis in this paper that the stack emission profile of CDD/Fs is very different from the profiles measured in the soil and in the air. This could be explained by “changes” in the profile at some point between the stack and both air and soil measurement sites, or it could be that there were problems in the measurement of CDD/Fs in either the stack or the environmental media. Assuming no major problems with measurement, it can be said that these trends cannot be duplicated in ISCST3 without the input of

congener-specific atmospheric degradation rates, and/or congener-specific soil dissipation rates. One hypothesis offered to explain the change in the dioxin profile from stack emission to air measurement was that some dechlorination might be occurring – the higher chlorinated CDD/Fs may be dechlorinating to form lower chlorinated CDD/Fs. If so, and if attempting to duplicate this trend, the ISCST3 model would need additional algorithms to model these transformations.

While admittedly a limited field test of deposition and soil concentration models, the data used here had these important features, which are not readily (if at all) available for similar model testing of ISCST3 with CDD/Fs: multiple stack tests offering a full suite of dioxin homologue and congener data; a historically high-emission rate and over 11 yr of emissions such that a signal is left behind in the soil and an imprint in the ambient air while monitoring was occurring, and a reasonable approach to determining the local background of dioxin soil and air concentrations that could be subtracted from the total measured soil and air concentrations to characterize a “signal” of higher dioxin concentrations found near the incinerator.

Follow-up model testing of ISCST3 on dioxins should consider the following: (1) model refinements or assignment of degradation rates for dispersing dioxins to consider mechanisms that deplete the plume indicated (discussed above), as the tendency to overpredict proportionally higher soil concentrations for further downwind locations can only be attributed to the air-modeling portion of this exercise, not the soil-modeling portion, and (2) similar sensitivity exercises which can test if particle deposition or photolytic dechlorination could account for the observed homologue shift between the stack and field.

While the analyses in this paper are not intended as rigorous model evaluations, they will hopefully stimulate interest in conducting coordinated model runs, source tests and field ambient measurements to better understand the processes that influence the fate and transport of dioxins emitted from tall stacks.

6. Disclaimer

The views expressed in this article are those of the authors and do not necessarily reflect the view or policies of US Government Agencies.

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References

- Bidleman, T.F., 1988. Atmospheric processes wet and dry deposition of organic compounds are controlled by their vapor-particle partitioning. *Environmental Science and Technology* 22 (4), 361–367.
- Cleverly, D., Schaum, J., Winters, D., Schweer, G., O'Rourke, K., 1999. Inventory of sources and releases of dioxin-like compounds in United States. *Organohalogen Compounds* 41, 467–471.
- EERC, 1992. Energy and environmental research corporation. Permit Compliance Test Report, Columbus Solid Waste Reduction Facility. 18 Mason, Irvine, CA 92718. Conducted for Ohio EPA September 11, 1992.
- EMC, 1994. EMC Analytical, Inc. Emission Study Performed for Solid Waste Authority of Central Ohio, Project No: 35 001, April 18, 1994. P.O. Box 149. Gilberts, IL 60136.
- EPA, 1989. Interim procedures for estimating risks associated with exposures to mixtures of chlorinated dibenzo-*p*-dioxins and -dibenzofurans (CDDS and CDFs) and the 1989 Update. United States Environmental Protection Agency, Risk Assessment Forum, Washington, DC, EPA/625/3-89/016.
- EPA, 1994. Estimating exposure to dioxin-like compounds. United States Environmental Protection Agency, Office of Research and Development, National Center for Environmental Assessment. Review Draft. EPA/600/6-88/005Ca,b,c.
- EPA, 1995. User's Guide for the Industrial Source Complex (ISC3) Dispersion Models, Vol. I – User Instructions and Vol. II – Description of Model Algorithms. United States Environmental Protection Agency. Office of Air Quality Planning and Standards, September, EPA-454/B-95-003a,b.
- EPA, 1996. 40 CFR 51, Appendix W. Guideline on Air Quality Models (Revised). EPA 450/2-78-027R.
- Eschenroeder, A., Cullen, A. C., von Stackelberg, K., 1994. Gas/particle partitioning effects on dioxin exposures from incinerators. A paper presented at the US Environmental Protection Agency Ninth Annual Regional Risk Assessment Conference, April 25–27.
- Koshioka, M., Yamada, T., Knazawa, J., Murai, T., 1989. Photolysis of tetrachlorodibenzo-*p*-dioxins. *Chemosphere* 19, 681–684.
- Lorber, M., Pinsky, P., Gehring, P., Braverman, C., Winters, D., Sovocool, W., 1998. Relationships between dioxins in soil, air, ash, and emissions from a municipal solid waste incinerator emitting large amounts of dioxins. *Chemosphere* 37, 2173–2197.
- McLachlan, M.S., Sewart, A.P., Bacon, J.R., Jones, K.C., 1996. Persistence of PCDD/Fs in a sludge-amended soil. *Environmental Science and Technology* 30, 2567–2571.
- Ohio EPA. 1994. Franklin county ambient air monitoring study for dioxins and dibenzofurans. Ohio Environmental Protection Agency, Division of Air Pollution Control. P.O. Box 1049, 1800 Watermark Dr., Columbus, OH 43266-0149, July 1994.

- Ohio EPA, 1995. Dioxin Monitoring Study 1995 Franklin County, Ohio. Ohio Environmental Protection Agency, Division of Air Pollution Control. P.O. Box 1049, 1800 Watermark Dr., Columbus, OH 43266-0149, September 1995.
- Orth, R.O., Ritchie, C., Hileman, F., 1989. Measurement of the photoinduced loss of vapor phase TCDD. *Chemosphere* 18, 1275–1282.
- Paustenbach, D.J., Wenning, R.J., Lau, V., Harrington, N.W., Rennix, D.K., Parsons, A.H., 1992. Recent developments on the hazards posed by 2,3,7,8-tetrachlorodibenzo-*p*-dioxin in soil: implications for setting risk-based cleanup levels at residential and industrial sites. *Journal of Toxicology and Environmental Health* 36, 103–149.
- Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X.R., Liem, A.K.D., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Warn, F., Zacharewski, T., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environmental Health Perspectives* 106, 775–792.