

An Evaluation of EPA's ISCST -Version 3 Model Part 2. Deposition and Soil Concentration of Dioxins

Matthew Lorber¹, Randall Robinson², Alan Eschenroeder³

¹National Center for Environmental Assessment (8623D), US EPA, Washington, DC 20460; ²Region 5, US EPA, Chicago, ILL 60604-3507; ³Harvard School of Public Health, Dept. of Environmental Health, Boston, MA 02115-6069

Introduction

The central purpose of our study is to examine the performance of the United States Environmental Protection Agency's (US 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) near the Columbus Municipal Waste-to-Energy Facility (CMWTE) in Columbus, Ohio. The plant operated between June 1983 and December 1994 and was estimated to emit nearly a kilogram of dioxin toxic equivalents (TEQs) annually (TEQs calculated in this paper using the International TEF scheme¹ and not including dioxin-like PCBs). This compares with US estimates of total emissions of 12 kg TEQ in 1987 and 3 kg in 1995². A soil monitoring study conducted during 1995 and 1996 included 34 soil samples taken on-site and up to 12 km in all directions from the plant. An evaluation of this soil data clearly showed an imprint from the CMWTE, with concentrations decreasing as a function of distance from the stack, approaching a local background after about 3 km³. Two stack tests on dioxin emissions, taken in 1992 and 1994, were used in separate model evaluation exercises described herein. Annual average dry and wet deposition rates of particle-bound dioxins predicted by the ISCST3 model served as inputs to a simple soil reservoir model to predict dioxin soil concentrations. Complete descriptions of the stack and soil measurements conducted around the CMWTE are available in previous papers^{3,4,5} and not repeated here.

Modeling procedures

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. The short term version of the model used here relies on hourly wind speed, wind direction and stability for describing dispersion. Hourly precipitation data must be input in the present application to calculate wet deposition. The prediction of depositions of particle-bound CDD/Fs with ISCST3 relies on three types of data: (1) emissions data, (2) particle-specific (particle diameter, e.g.) and dioxin-specific (vapor/particle partitioning, e.g.) model parameters, and (3) meteorological data (wind speeds, directions, rainfall, e.g.). 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. Due to space considerations, details on parameter assignments for this modeling exercise cannot be provided in this abstract. Further detail on the ISCST3 can be found in EPA⁶.

The ISCST3 model was run on a "unitized" basis, meaning that deposition results were generated assuming that particle-bound contaminants were emitted at a rate of 1 g/sec. Deposition predictions for the 25 CDD/Fs (17 congeners of non-zero toxicity and 8 homologue groups) 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 (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/sec, 2) then, these particle-bound mass emission rates were multiplied by the unitized deposition predicted to occur at the receptor point to provide the compound-specific deposition rates.

Two stack tests were available to supply the critical source term for this exercise^{4,5}. 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 concentration (total = sum of the homolog group concentrations; dscm = dry standard cubic meter) and 976 g TEQ/yr mass emissions were found, leading to regulatory activities by the state and federal environmental agencies. Process modifications were undertaken for purposes of reducing dioxin emissions, and the CMWTE was retested in the spring of 1994. Total concentrations were reduced to 3685 ng/dscm and the mass TEQ emissions were reduced by about 75% to 250 g TEQ/yr. Rather than select one or the other stack emission tests 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.

Wet and dry depositions are summed and become the source term for a simple reservoir mixing model for predicting soil concentration C_s , as: $[F(1 - e^{-kt})]/kM$, where C_s is the soil concentration (pg/g); F is the annual total (wet + dry) deposition of dioxins as predicted by ISCST3 (pg/m²-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/m²). The dissipation rate assumed here for all dioxin compounds was 0.02772 yr⁻¹ (half-life of 25 years), a mid-range value selected to be between a value of 0.0693 (half-life of 10 years) often assumed for surficial dioxin residues⁷ and 0.00693 (half-life of 100 years) speculated to be an upper range for subsurface dioxin residues⁸. A time of operation, t , of 11.5 years was used, corresponding to the time of operation of the CMWTE. The soil mixing mass, M , equaled 112,500 g/m², which assumes a mid-range soil bulk density of 1.5 g/cm³ and the soil sampling depth of 7.5 cm.

Finally, the observed soil concentrations needed to be adjusted by subtracting the local background soil concentration. This was necessary in order to compare the predictions of the incremental impacts from the CMWTE with an appropriate set of observations. An analysis of this soil data in Lorber³ showed that concentrations decrease to the local soil background at about 3 km from the CMWTE, at a TEQ soil concentrations of 4.0 pg/g (ppt). The soil profile of CDD/Fs for this background provided in Lorber³ 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.

Results and Discussions

Table 1 provides results from this exercise, which are observed and predicted homologue and TEQ concentrations for 4 clusters of soil samples. These clusters were developed for purposes of displaying results from the soil monitoring study conducted around the CMWTE³, and generally correspond to increasing distance in all directions from the incinerator, as shown in their brief descriptions in Table 1. It should be noted that observed and predicted soil concentrations for soil samples taken on-site, the first cluster of Table 1, are shown for informational purposes only - it is not expected that the on-site soil samples represented long-term deposition trends. As discussed in Lorber³, the high soil concentrations found were speculated to have resulted in ash drift from piles

or trucks transporting the ash to nearby landfills rather than deposition. 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.). Some trends that may be observed from the results in Table 1 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, which also explains why the TEQ concentrations predicted using the 1992 stack test better match the observations compared to the TEQ soil concentrations predicted using the 1994 stack test. 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 CMWTE. The 1994 test occurred specifically after measures had been taken to reduce dioxin emissions. Because of process changes made to the CMWTE, 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 suggested⁹ 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, but still noticeable, extent with HpCDD; in other words, the model under-predicted the soil concentrations of these homologue groups. The companion paper to this one¹⁰, evaluating the capability of the ISCST3 to reproduce short-term (48 hr) air concentrations, found that the model 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.

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).

4) Besides the rate of dioxin emissions, there are numerous model uncertainties and deficiencies, in general and for this particular exercise. Some of these include: a) the lack of

consideration of plume depletion mechanisms including atmospheric degradation of either vapor or particle phase dioxins, vapor phase deposition, and vapor- and particle-phase vegetative capture; b) the representativeness of the meteorological data used in this exercise; and c) dioxin-specific fate parameters including vapor/particle partitioning of the CDD/Fs and soil half-lives. The meteorological data may have been responsible for some of the tendency for overprediction as the annual rainfall in 1989, 112 cm, somewhat exceeded the historical average of 94 cm. The dioxin fate parameter, soil half-life, may have been assigned too low a value (i.e., too long a half-life), hence leading to higher soil concentrations than observed. When reducing the half-life from 25 years to 10 years, model predictions of soil concentrations for all locations dropped by about 20%.

Concluding Remarks

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: multiple stack tests offering a full suite of dioxin homologue and congener data; a historically high emission rate and over 11 years of emissions such that a signal is left behind in the soil, and a reasonable approach to determining the local background of dioxin soil concentrations that could be subtracted from the total soil concentrations to characterize a “signal” of higher dioxin soil concentrations found near the incinerator. The deposition/soil modeling done here suggests that predicted soil concentrations are mostly within a factor of 10 of observations. Follow-up model testing of ISCST3 on dioxins should consider the mechanisms that deplete the plume indicated in the bullet 4a) 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. While the analysis in this paper as well as the companion paper on air dispersion testing¹⁰ 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.

References

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Table 1. 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 (soil concentrations in pg/g, 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).

Cluster->	On-site			Off-site			Urban			Urban Background		
Description of Cluster->	n = 3; on incinerator property			n = 5; just outside property, downwind within 500 m			n = 14; all directions within about 3 km.			n = 12; all directions from 3 to 12 km.		
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

