

ATMOSPHERIC FATE AND TRANSPORT OF DIOXINS: LOCAL IMPACTS

Kristen Lohman and Christian Seigneur

Atmospheric & Environmental Research, Inc., 2682 Bishop Drive, Suite 120, San Ramon, CA, USA

Introduction

A major issue associated with emissions of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/F) is the zone of influence of major emission sources, which determines whether the source has predominantly local impacts (with potentially high PCDD/F concentrations) or contributes to a regional/global background (with dilute PCDD/F concentrations over a large region). Conversely, it is important to know whether PCDD/F deposition at a given location is due primarily to local sources or distant regional/global sources. Previous work has led to a wide range of results^{1,2,3,4}. The characteristic atmospheric travel distance of PCDD/F will depend on their gas/particle partitioning, the particle size distribution for particulate PCDD/F, and the deposition characteristics of the gaseous and particulate PCDD/F congeners. In addition, atmospheric deposition depends on precipitation, atmospheric turbulence, the effective height (i.e., the sum of the stack height and plume rise) of the emissions, and surface type.

A fair amount of work has been done on the local health impacts of PCDD/F emissions from point sources. Such studies typically have involved assumptions that tend to overestimate local deposition, e.g., all PCDD/F are assumed to be present in the particulate phase. A recent investigation into the sensitivity of ground-level concentrations of PCDD/F emitted from a municipal waste incinerator to various vapor/particle partitioning scenarios, particle sizes and densities, local terrain and emission data showed that PCDD/F ground-level concentrations are quite sensitive to these model inputs⁵.

We present here an analysis of the relative fractions of PCDD/F emissions that are deposited locally and transported beyond 100 km from the source. We consider several PCDD/F source categories and several locations with different terrain and meteorological characteristics. The sensitivity of the results to key input assumptions is also investigated.

Method

We reviewed the literature to identify those source categories that are estimated to contribute most to the total U.S. and European PCDD/F emission inventories. The emission source categories that we modeled include municipal solid waste combustors (MSW), medical waste incinerators (MWI), secondary copper smelters, hazardous waste burning cement kilns, sinter plants, and diesel trucks. MSW and MWI are among the major PCDD/F source categories in both the U.S. and Europe; secondary copper smelting and cement kilns are among the five largest source categories in the U.S. whereas sinter plants are the second largest (after MSW) source category in Europe^{6,7}. We also included diesel trucks as a non-industrial source in our study. Trucks do not produce as much PCDD/F as the other sources but they are located in all parts of the U.S. and Europe.

ENVIRONMENTAL FATE AND TRANSPORT

Moreover, their emissions are near ground level whereas the other sources considered here release their emissions from stacks.

Two of the selected source categories, MWI and MSW, were broken down further into subcategories. In nearly every country, MSW are the largest source of PCDD/F emissions. There are many different combustor designs for MSW. We selected two types here, a refuse-derived fuel facility (MSW-RDF) and a mass-burn waterwall facility (MSW-MB). Incineration of medical waste is also a large source of dioxins. Therefore, two types of MWI were modeled, one uncontrolled (MWI-UNC) facility and one controlled with an electrostatic precipitator (MWI-ESP).

Model simulations were conducted for six different geographic locations to account for the effect of meteorology and land use on atmospheric deposition. These locations are Seattle, Washington; Phoenix, Arizona; Chicago, Illinois; Pittsburgh, Pennsylvania; Long Island, New York; and Fort Myers, Florida. The conditions in these locations differ in terms of temperature, precipitation, wind and complexity of the local terrain. All these locations are within the U.S. because meteorological data are readily available from the National Climatic Data Center in a format compatible with the atmospheric dispersion model.

We used a model that includes treatment of plume rise, atmospheric transport and dispersion, transformation, wet deposition, and dry deposition. The plume model is based on the EPA-recommended Industrial Source Complex - Long Term (ISC-LT) dispersion model. Because it is important to simulate the fate and transport of individual congeners rather than that of a TCDD TEQ⁸, we modified ISC-LT to provide detailed treatment of PCDD/F transformation and deposition.

Results and Discussion

Model simulations were conducted for each source category in each one of the selected locations. Thus, 48 model simulations were initially conducted (8 source categories x 6 locations). In addition, several simulations were conducted to investigate the sensitivity of model simulation results to some key parameters, including the particle size distribution, the PCDD/F congener profile, the surface roughness length, local terrain, and atmospheric dispersion conditions (i.e., urban vs. rural).

Table 1. Fraction of PCDD/F emissions (as TEQ) transported beyond 100 km from the source.

Source Category	Location					
	Seattle, WA	Phoenix, AZ	Chicago, IL	Pittsburgh, PA	Long Island, NY	Fort Myers, FL
MSW-RDF	0.87	0.84	0.86	0.87	0.91	0.84
MSW-MB	0.89	0.86	0.88	0.89	0.92	0.85
MWI-UNC	0.83	0.77	0.81	0.82	0.88	0.60
MWI-ESP	0.80	0.74	0.79	0.80	0.86	0.43
Copper smelter	0.97	0.97	0.97	0.97	0.98	0.99
Cement kiln	0.95	0.94	0.95	0.95	0.96	0.97
Sinter plant	0.91	0.91	0.91	0.92	0.94	0.95
Diesel truck	0.91	0.88	0.90	0.91	0.93	0.74

Table 1 provides a summary of the results of the base case simulations. These results are presented in terms of a TCDD TEQ rather than total PCDD/F since the potential toxicity of PCDD/F is generally the quantity of interest. The model simulations predict that for all source categories, except MWI, more than 70% of the PCDD/F emissions were transported beyond 100 km from the source. Sources with a tall stack and/or high plume rise have a greater fraction of PCDD/F emissions being transported beyond 100 km from the source.

MWI have the smallest fraction of PCDD/F emissions transported beyond 100 km (between 43 and 88%); i.e., they have the largest fraction of emissions deposited locally. The MWI have relatively short stacks and small stack diameters compared to the other stationary sources.

Secondary copper smelters have the least local deposition among all the source categories considered here (less than 3%). This result is due in part to their tall stack height (154 m in the case study considered here) and their large buoyancy (stack diameter of 4.6 m and flue gas temperature of 494 K). In addition, secondary copper smelters have the largest fraction of TCDD and TCDF emissions (72% of total PCDD/F as TEQ) due to limited congener measurements. TCDD and TCDF are the two congeners that partition the most toward the gas phase. Since we made the assumption that deposition of gases was negligible, only a small fraction of PCDD/F emissions from copper smelters is subject to deposition.

Diesel trucks have a large fraction of their emissions transported beyond 100 km (from 74 to 93%) despite the fact that their emissions are near ground level. The reason for this low local deposition is the size of the diesel particles. Such particles are mostly fine (i.e., less than 2.5 μm in diameter) and, therefore, are not easily removed via wet or dry deposition processes.

Some clear patterns also appear among the different locations. Five locations were characterized by urban environments (i.e., urban dispersion option in ISC-LT and a surface roughness length of 1 m) and one location (Fort Myers, Florida) was characterized by a rural environment (i.e., rural dispersion option and a surface roughness length of 0.1 m).

Among the urban locations, Phoenix leads to more local deposition than the other locations. The higher ambient temperature in the Phoenix area (southwestern U.S. desert) limits the temperature gradient between the plume and the ambient air, and consequently leads to less plume rise. Lower plumes lead to more local deposition. Seattle, Chicago and Pittsburgh lead to similar results among them. Long Island, New York leads to the lowest local deposition among all the locations. This result is due to the fact that New York shows a different pattern of atmospheric stabilities (higher frequency of neutral stability and lower frequency of unstable and stable conditions) than the other three northern locations.

The rural environment in Florida leads to less atmospheric dispersion of the plume compared to the urban locations and, for a given ground-level concentration, less dry deposition because of a lower surface roughness length. Less atmospheric dispersion leads to different effects on plumes from tall stacks than on plumes from low stacks or surface releases. For a tall stack plume, less dispersion means that the plume remains elevated longer and that ground-level concentrations will remain negligible farther downwind. For a low plume, less dispersion means that the plume remains more concentrated and that ground-level concentrations are higher. As a result, a greater fraction of the emissions from source categories with tall stacks and/or higher buoyancy (i.e., copper smelter, cement kiln, sinter plant) is transported beyond 100 km in Florida than in the urban locations. On the other hand, for the source categories with moderate or low stack heights (i.e., MSW, MWI, diesel trucks), a lower fraction of the emissions is transported beyond 100 km in Florida than in urban environments. The lower surface roughness length also reduces the

particle dry deposition velocity compared to an urban environment, but the effect of atmospheric dispersion (i.e., rural vs. urban) dominates.

The results of the model simulations depend on the assumptions made when selecting model inputs. We investigated the sensitivity of model simulation results to the urban/rural dispersion option, the surface roughness length, the terrain elevation (complex vs. flat), the particle size distribution, the PCDD/F congener profile and the gaseous PCDD/F dry deposition velocity. The results were most sensitive to the urban/rural dispersion option and the particle size distribution. The implications are that (1) an atmospheric dispersion model such as the EPA-recommended ISC model should be used only for screening assessments and that more advanced dispersion models should be used for refined assessments and (2) measurements of the size distribution of particulate PCDD/F are needed. In this work, we used total particle size distributions because no PCDD/F – specific particle size distributions were available for emissions sources. PCDD/F may condense preferentially on smaller particles, in which case our model simulations will tend to overestimate the amount of PCDD/F being deposited locally.

The performance of the atmospheric fate and transport model should be evaluated with experimental data. Such field measurements are typically difficult to conduct because the signal from the source must be significantly greater than the noise of the variable background atmosphere^{9,10}. The design of a field program for model performance evaluation should include stack measurements of PCDD/F emissions including particle size distribution, ambient measurements of PCDD/F atmospheric concentrations upwind and downwind of the source with particle size distribution, on-site meteorological measurements near the ground and aloft, and, to the extent possible, deposition measurements.

Acknowledgments

This work was performed under contract with the Chlorine Chemistry Council of the Chemical Manufacturers Association. Thanks are due to Dr. James Franklin, Solvay, to Professor Donald Mackay, Trent University, and to three reviewers from Dow Chemical, for providing constructive comments on this work.

References

1. Bennett, D.H., T.E. McKone, M. Matthies and W.E. Kastenberg, 1998. *Environ. Sci. Technol.*, **32**, 4023-4030.
2. van Pul, W.A.J., F.A.A.M. de Leeuw, J.A. van Jaarsveld, M.A. van der Gaag and C.J. Sliggers, 1998. *Chemosphere*, **37**, 113-141.
3. Beyer, A., D. Mackay, M. Matthies, F. Wania and E. Webster, 2000. *Environ. Sci. Technol.*, **34**, 699-703.
4. Baker, J.I. and R.A. Hites, 1999. *Environ. Sci. Technol.*, **33**, 14-20.
5. Basham, J.P. and I. Whitwell, 1999. *Atmos. Environ.*, **33**, 3405-3416.
6. U.S. EPA, 1998a. The Inventory of Sources of Dioxin in the United States, EPA/600/P-98/002Aa, Office of Research and Development, U.S. Environmental Protection Agency, Washington, D.C.
7. UNEP, 1999. Dioxin and Furan Inventories: National and Regional Emissions of PCDD/PCDF, United Nations Environment Programme.
8. Lohman, L., C. Seigneur and L. Levin, 1998. *Organohalogen Compounds*, **36**, 445-447.
9. Eschenroeder, A. and M. Lorber, 1999. *Organohalogen Compounds*, **41**, 547-552.
10. Lorber, M., R. Robinson and A. Eschenroeder, 1999. *Organohalogen Compounds*, **41**, 541-545.