Development and Application of An Air Transport Model for Dioxins and Furans

Mark Cohen, Barry Commoner, Paul Woods Bartlett, Holger Eisl, Catherine Hill, Joyce Rosenthal; Center for the Biology of Natural Systems, Queens College, Flushing, NY 11367, USA.

Abstract

An air transport model has been used to estimate the amounts of dioxin and furan congeners which, emitted from numerous U.S. and Canadian sources, are deposited on each of the Great Lakes. It is also being used for corresponding estimates of dioxin and furan deposition on dairy farms and their occurrence in the farms' feed crops and milk.

Introduction

A large part of the general population's exposure to dioxins and furans comes from food, chiefly from milk, dairy products, beef, pork and chicken. The relevant animals absorb these pollutants from feed crops, which are in turn contaminated by the deposition of airborne dioxins and furans.[1] Since the sources of these airborne pollutants, for example incinerators, are often at a considerable distance from the farms on which the feed crops are raised, long-range air transport plays a crucial role in the overall path of human exposure.

Air transport models are a useful means of tracking the movement of airborne dioxins and furans from their numerous sources to important receptors such as dairy farms. This is especially true of models that are capable of ranking the individual sources with respect to their estimated contributions to the amount of airborne dioxins and furans deposited in receptor areas, so that remedial action can be appropriately targeted on the major sources. This paper describes the features and applicability of such a model.

Methods

The model is based on the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model developed by Roland Draxler at the National Oceanic and Atmospheric Administration.[2,3] In its original form, HYSPLIT is capable of estimating the air transfer coefficient (ATC) of gaseous or particulate material -- that is, the fraction of the material, emitted into the air at a given geographic location, that will be deposited to ground level at any other specified location. It incorporates weather data for the United States and southern Canada, tabulated at two-hour intervals for each year since 1988. The model is capable of estimating the advection, diffusion, deposition and photochemical destruction of material separately emitted by each source, recording its fate at hourly intervals over a one-year period. HYSPLIT has successfully undergone extensive validation tests.[4,5,6,7]

We have modified the original model to include the effects of vapor/particle partitioning and different particle sizes on the atmospheric transport and fate, separately, of the seventeen 2,3,7,8-substituted dioxin and furan congeners and eight overall homologue groups. We have also modified the model to estimate deposition on target areas -- as opposed to estimating the deposition flux intensity at discrete points -- to facilitate mass-balance accounting of the deposited material. Algorithms that estimate the uptake of the different dioxin congeners and homologue groups by vegetation can be added to this model. The structure of the model is summarized in Figure 1.

In order to estimate the ATC values for each of the dioxin congeners and homologue groups from the emissions of the numerous separate sources of dioxins, a data-processing program was also created: TRANSCO (Transfer Coefficient). The TRANSCO program includes two interpolation procedures that reduce the otherwise impractical computation time required for separate computer runs on 25 congener and homologue groups from each of thousands of emission sources to manageable dimensions.

First, a congener interpolation program was used to estimate ATC values for all 25 congeners and homologue groups from computer simulations of only several of them. This procedure was based on model runs on a series of the congeners that revealed a systematic relationship between their vapor/particle partitioning behavior and the ATC. This allowed an algorithm to be developed to estimate the ATC values of the entire set of congeners and homologue groups from interpolations based on their expected vapor/particle partitioning characteristics.

A second, spatial interpolation program enabled us to estimate ATC values for all of the numerous sites of emission from data generated by running the model for a limited number of "standard" emission locations. The spatial interpolation algorithm takes into account the relative distance of each of the four closest standard locations to the actual emission site and the relative angular orientation of the emission site with respect to the center-points of each of the target areas (in the case described below, each of the five Great Lakes). This procedure greatly reduces the computation time required, as explicit simulations do not need to be made for each actual source. In this application, 25 standard source locations were used to estimate the ATC values for thousands of actual sources of dioxin emission.

We have identified and estimated the dioxin air emissions, in 1993, of 1329 separate sources in the United States and Canada; 954 are individual facilities at identified locations and 375 sources are aggregated by state or province because they were too numerous or poorly identified to characterize separately.

Results and Discussion

The model has been used to estimate the air transfer of dioxins and furans from each of the identified sources in the U.S. and Canada to each of the Great Lakes for 1993. Figure 2 shows the effect of the distance of the sources from Lake Michigan on deposition. Sources contribute significantly to total deposition out to 2,000 km. The extent to which the different sources contribute to the dioxins and furans deposited in the Great Lakes depends not only on the amounts they emit and their distances from the lakes, but also on their geographic location relative to a given lake. A higher percentage of the dioxins and furans emitted from sources to the south and west of the lakes is deposited in them than from the sources to the north and east. A higher proportion of the emissions from sources southwest of the lakes is deposited in them

than emissions southeast of the lakes. Figure 3 illustrates these effects in the case of Lake Michigan.

These results are consistent with the prevailing weather pattern, which carries airborne pollutants more effectively to the Great Lakes in the west-to-east direction than in the reverse direction. This has been confirmed by mapping the values of the air transfer coefficient for emissions of dioxins from 20,000 hypothetical emission sites (sectors, each 270 square miles in area) covering all of the U.S. and southern Canada to each of the Great Lakes. Figure 4 is such a map showing the air transfer coefficients for 2,3,7,8-TCDD to Lake Michigan. It is characterized by a pronounced extension of the ATC values to the west and southwest. A corresponding map for OCDD shows that OCDD is more effectively transported to the lake over long distances than 2,3,7,8-TCDD. OCDD is almost entirely bound to particulates and thereby protected from photochemical degradation during transport; TCDD is partly in the vapor phase and therefore more vulnerable to photochemical degradation.

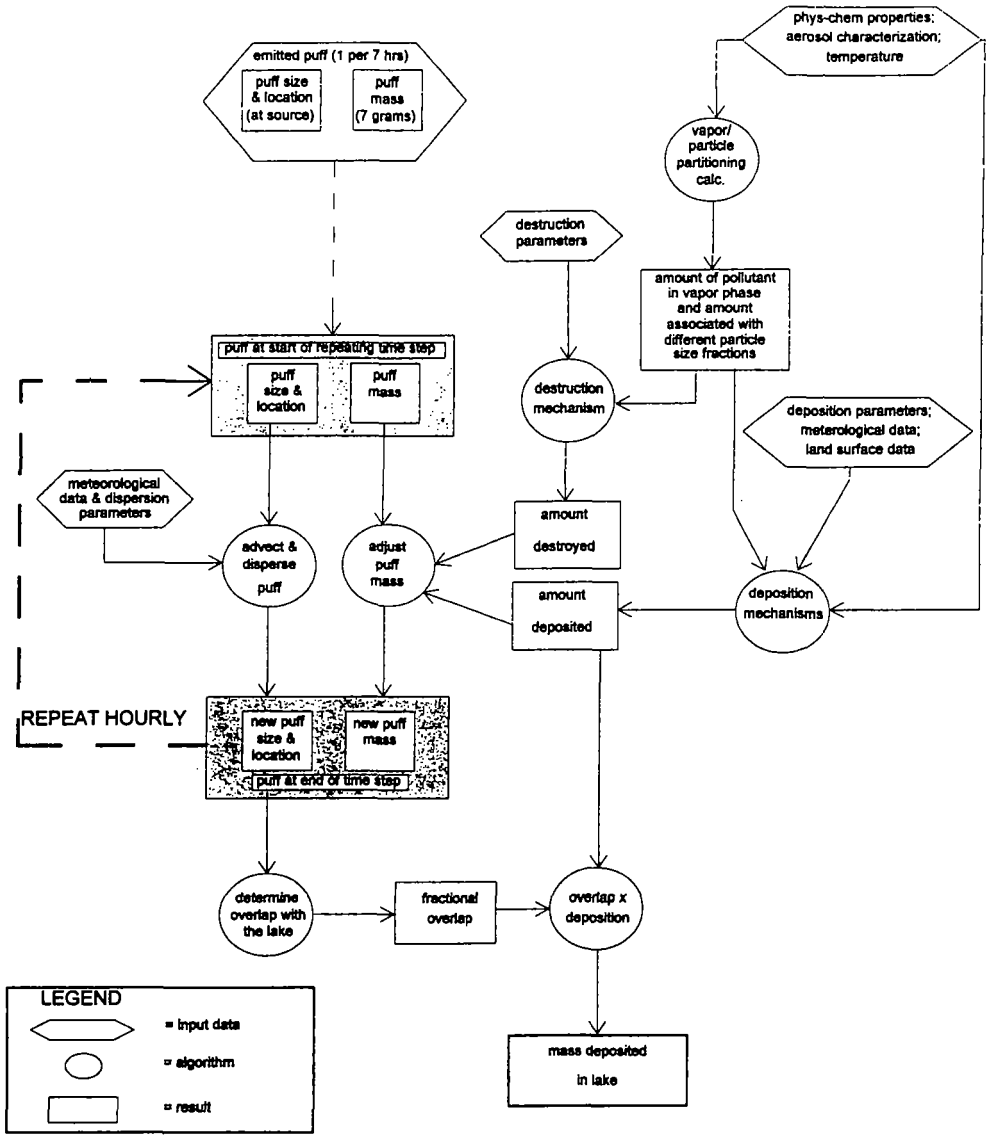
The data have been used to estimate the average flux of dioxins and furans deposited in each of the Great Lakes -- i.e., μg TEQ deposited per km^2 per year -- in 1993: Superior, 69; Michigan, 238; Huron, 145; Erie, 284; Ontario, 337.

Several procedures have been used to validate the HYSPLIT/TRANSCO program. First, in order to test the accuracy of the spatial interpolation procedure -- on which the model's ability to deal with numerous emission sites depends -- the ATC values for a number of sources were computed directly by running the model and then compared with the values generated by the interpolation procedure. Figure 5 shows the relationship between the two sets of values. The directly computed and interpolated values compare reasonably well over an approximately three-order-of-magnitude range. The average of the absolute values of the differences between the spatially interpolated values and the explicitly calculated values is 38%.

Finally, the validity of the overall HYSPLIT/TRANSCO model has been tested by comparing computed estimates of the concentrations of the 17 toxic dioxin and furan congeners at Dorset, Ontario, where two-day average ambient air concentrations were measured at monthly intervals.[8] As shown in Figure 6, the measured monthly values agree reasonably well with concurrent weekly average concentrations computed by the model. Because of the differing time period of the measurements (two days) and the model estimates (weekly), the two sets of data would not be expected to match precisely even if the estimates were "perfect." When average values for the entire year are compared, they are quite close: 3.28×10^{-15} g TEQ/m^3 for the actual measurements, and 3.40×10^{-15} g TEQ/m^3 for the model-estimated values. These and other results are described in more detail elsewhere.[9]

The model is also being used to estimate the contribution of U.S. and Canadian sources to the deposition of dioxins and furans on dairy farms in Wisconsin and Vermont (four farms in each state). Samples of air, feed crops and milk representative of dioxins and furans deposited over a 28-day period in August 1996 were collected and have been analyzed for dioxin and furan congeners. The data are being compared with model-estimated air concentrations at the farms and with additional algorithms that estimate the transfer of dioxins and furans from air to crop and from crop to milk in the dairy farm ecosystem.

FIGURE 1: Modified HYSPLIT Program



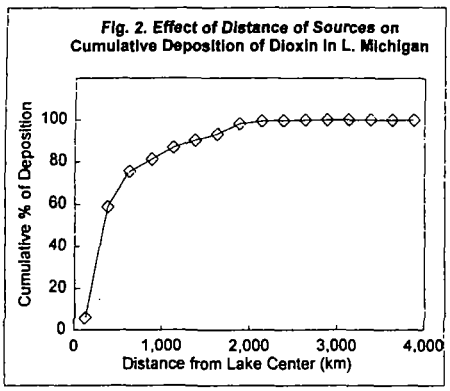


Fig. 2. Effect of Distance of Sources on Cumulative Deposition of Dioxin in L. Michigan
Cumulative % of Deposition
100
80
60
40
20
0
0
1,000
2,000
3,000
4,000
Distance from Lake Center (km)

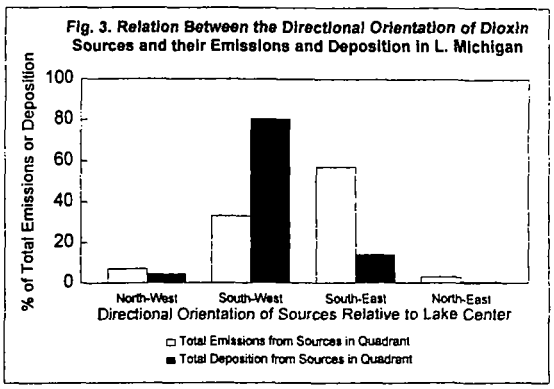


Fig. 3. Relation Between the Directional Orientation of Dioxin Sources and their Emissions and Deposition in L. Michigan
% of Total Emissions or Deposition
100
80
60
40
20
0
North-West
South-West
South-East
North-East
Directional Orientation of Sources Relative to Lake Center
Total Emissions from Sources in Quadrant
Total Deposition from Sources in Quadrant

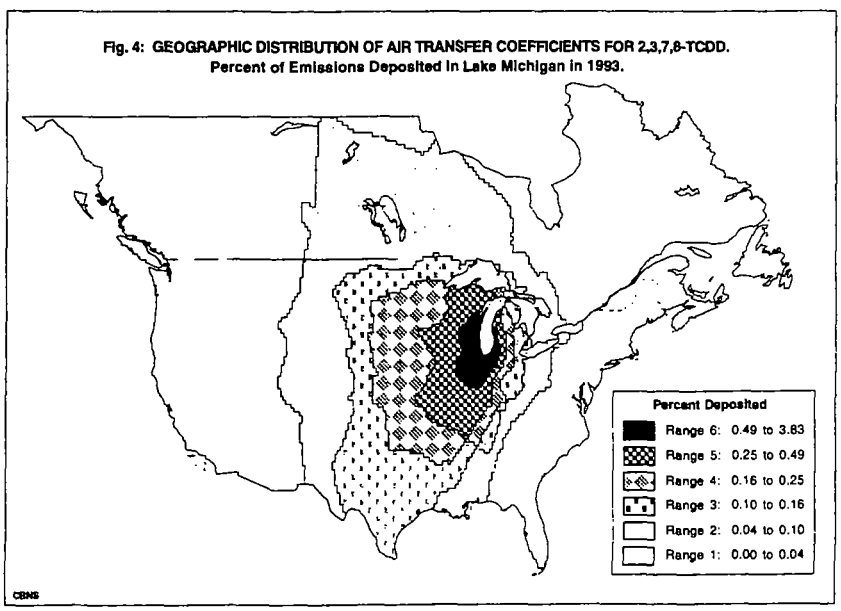
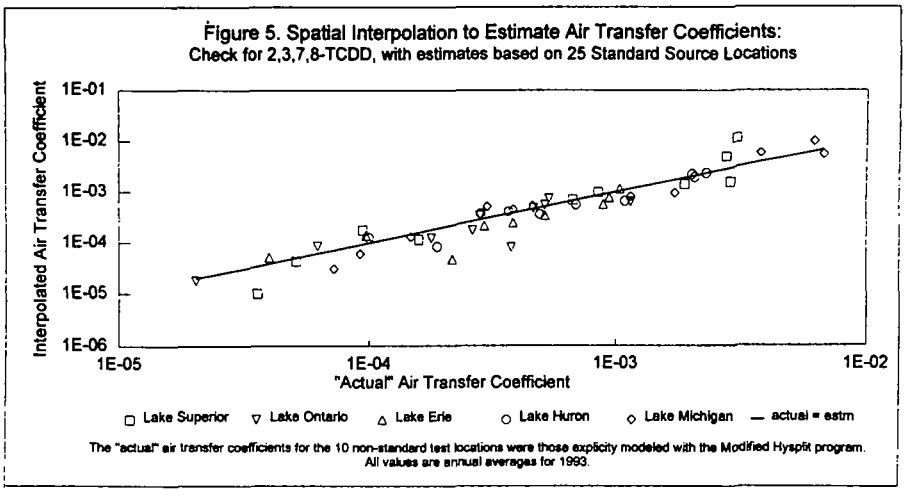
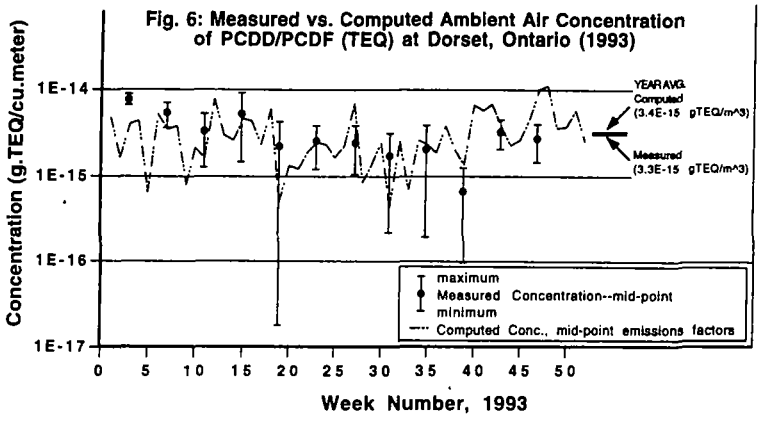


Fig. 4: GEOGRAPHIC DISTRIBUTION OF AIR TRANSFER COEFFICIENTS FOR 2,3,7,8-TCDD.
Percent of Emissions Deposited in Lake Michigan in 1993.
Percent Deposited
Range 6: 0.49 to 3.83
Range 5: 0.25 to 0.49
Range 4: 0.16 to 0.25
Range 3: 0.10 to 0.16
Range 2: 0.04 to 0.10
Range 1: 0.00 to 0.04
CBNS





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