MODELING SOURCE-TO-RECEPTOR ATMOSPHERIC TRANSPORT: ATRAZINE, PCBS AND DIOXIN IN NORTH AMERICA

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Introduction

Persistant organic pollutants (POPs) emit into the atmosphere from near and distant sources, transport and deposit to land and marine surfaces, and enter the food chain through water, flora and fauna. Remedial policy must be directed to the reduction of emissions from the sources, but knowledge of source emissions, pollutant measurements, and distance alone is insufficient information to distinguish the sources most responsible for contamination. Numerical modeling of pollutant transport and environmental fate from individual sources to ecological receptors can close this information gap by taking into account weather patterns and the chemical characteristics of the pollutant affecting atmospheric destruction and deposition, but only when source-to-receptor information is preserved in the results. The approach we use with the HYSPLIT[1] atmospheric transport model can accomplish these objectives.

Methods

The Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT)[1] enables us to preserve source-to-receptor identity while simulating atmospheric transport, destruction and deposition of hypothetical emission puffs, from each source point to target receptor areas. The source-to-receptor relationship can be represented, as an output of HYSPLIT, by the ratio deposited to the receptor from one unit emission from the source, the atmospheric transport coefficient (ATC). When ATCs are mapped, the effect of weather patterns and pollutant chemical properties on transport can be evaluated. The product of the ATC and the quantity emitted is the amount deposited to the receptor from that source (ATC * Source Quantity Emitted = Receptor Deposition). Relative contributions of each source to receptor deposition can be determined and ranked. We are able to use this approach because we are modeling single-hop transport (no re-suspension; one-way deposition) and since the pollutants are transported in trace amounts. HYSPLIT algorithms use pollutant chemical properties to estimate vapor-particle partitioning, destruction (hydroxyl radical and photochemical reactions), dispersion and deposition.

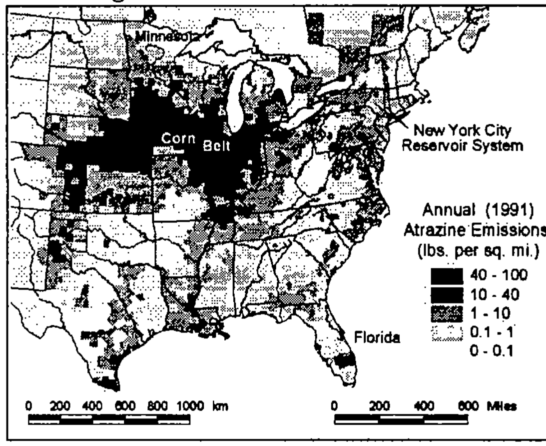
This method is made computationally feasible for a large number of sources and congeners by the use of a spatial and congener interpolation program, TRANSCO, which applies and emission inventory to a set of geographically represented standard point source-to-receptor HYSPLIT computer runs.

The atrazine emission inventory constructed in an earlier CBNS study[2] (Fig. 1) was spatially refined to localized farming areas according to satellite data, land use maps, and local experts (Fig. 2a). PCB emissions from the Hudson River were derived from USEPA measurements, hydrological modeling by Kevin Farley, Manhattan College and air concentration measurements.[3] Dioxin inventories originated from CBNS, USEPA, and Environment Canada.[4]

Results and Discussion

HYSPLIT estimated remote lakes in northern Minnesota and Lake Superior to be as contaminated by atmospheric transport of atrazine from the nearest sources (200km.) with high ATCs and moderate emissions as in the corn belt (up to 800 km.) with lower ATCs, but compensating greater emissions (Fig. 1).[2] HYSPLIT modeled transport to the New York City reservoir system found that it was far enough away from the corn belt that over 57-85% of deposition to the reservoirs can be reduced by restricting use within New York State.

Fig.1 Atrazine Emissions 1991



HYSPLIT modeled substantial deposition to some Florida Lakes, but not others only 40-80 kilometers away. This phenomenon is due to the combination of, rainy climate, atrazine's high water solubility and its efficiency of removal by rain and dry gas deposition, localized intense use of atrazine and the orientation of farms to lake receptors. The most intense use of atrazine in the U.S., on sugarcane around Lake Okeechobee, account for extraordinarily high deposition to Okeechobee and the greatest share of the deposition to the lake receptors with the most deposition (Fig. 2a), except for Lake Apopka. Lake Apopka receives 70% of its deposition from two counties bordering the lake, with a cluster of farms with high air transfer coefficients (Fig. 2a, 2b, 2c). The two lakes receiving the least contamination are slightly further away, receive the least amount of wet deposition and are more remote from local use. These modeling results are dependent on the fine spatial resolution of the emission sources and the meteorological data.

Fig. 2a Florida Emissions

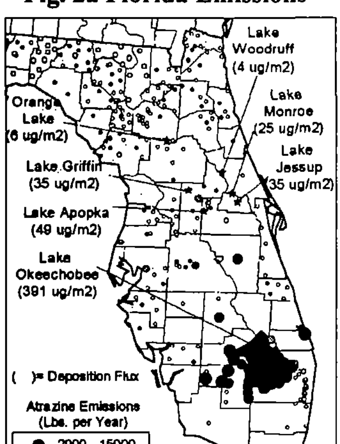


Fig. 2b Apopka ATCs

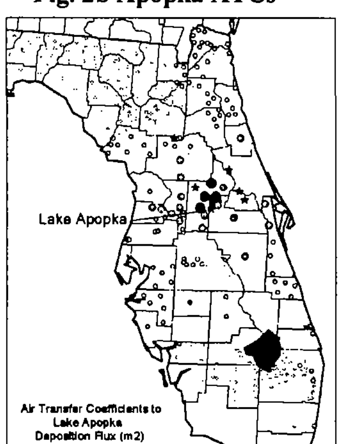
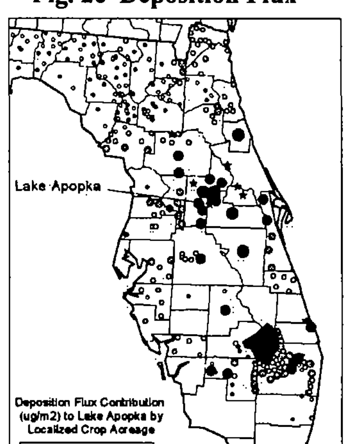


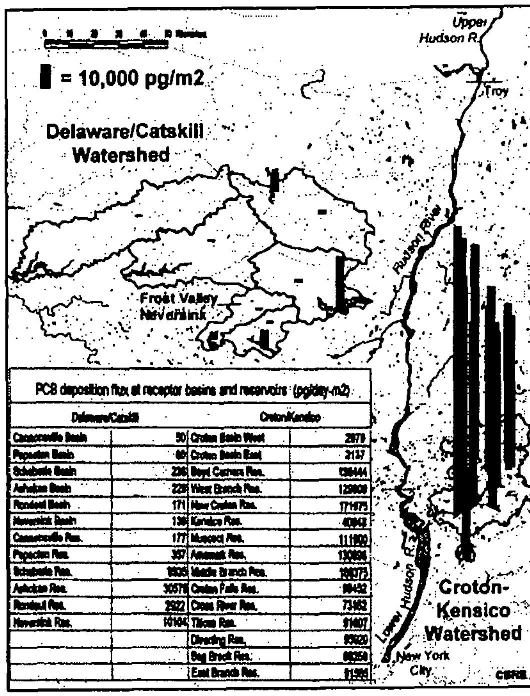
Fig. 2c Deposition Flux



Alligator developmental disorders measured by Louis Guillette, Florida University, Gainseville, were not present in the two lakes with the least deposition flux and the lake with the most, Okeechobee, but were found in the lakes with intermediate deposition flux.[5] A "U curve" endocrine effect has been hypothesized for environmental exposure. Further biological research and measurements of atrazine and other intensely used pesticides in Florida are needed.

We adapted HYSPLIT to model atmospheric transport of PCBs to find out if emissions from the Hudson River, contaminated by spills from GE with a distinctive PCB congener profile, could be a major source of PCBs to the New York City reservoir system. At this time we do not have a PCB emission inventory to compare relative contribution of this source to others, but were able to evaluate modeled predictions with air measurements made at a high elevation site in Frost Valley (Fig. 3). We were only able to account for 15% of the air concentration, but the higher chlorinated congener profile is close to the profile measured at the Hudson, indicating that the Catskill Mountains are a PCB sink from the river's historic emissions. The deposited higher chlorinated PCB congeners should accumulate more near the source of emission since they re-suspend and re-transport (the grasshopper effect) at a slower rate than the lower chlorinated congeners, which are predominantly in the vapor phase.

Fig. 3 PCB Deposition Flux Aug-Sept 1998



PCB deposition flux at receptor basins and reservoirs (pg/day-m2)			
Delaware/Catskill		Croton/Kensico	
Cannonsville Basin	50	Croton Basin West	2878
Pepacton Basin	60	Croton Basin East	2137
Schoharie Basin	236	Boyd Corners Res.	138444
Ashokan Basin	228	West Branch Res.	129808
Rondout Basin	171	New Croton Res.	171975
Neversink Basin	136	Kensico Res.	40843
Cannonsville Res.	177	Muscoot Res.	111809
Pepacton Res.	357	Amawalk Res.	130898
Schoharie Res.	1805	Middle Branch Res.	188073
Ashokan Res.	30578	Croton Falls Res.	98452
Rondout Res.	2922	Cross River Res.	73482
Neversink Res.	10104	Titicus Res.	91607
		Diverting Res.	95820
		Bog Brook Res.	88258
		East Branch Res.	91595

As expected, the mid-Hudson has the highest modeled ATCs (except for the most northern and southern reservoirs) and is responsible for the most deposition, despite the much higher PCB water concentrations in the upper Hudson near the spill sites. The predicted high levels of deposition to the reservoirs east of the Hudson was unexpected, but can be accounted for by the high receptivity of water to dry particle deposition and the more frequent and faster westerly winds. This results in higher PCB surface air concentration due to the limited time for dispersion. Nevertheless, there is a degree of model uncertainty of deposition of PCBs to surfaces, so more measurements are included in the next step of research. We plan to measure PCBs in tree bark at different distances and orientations from the river for transport patterns, and measure PCBs in different tree ages to identify historic deposition patterns. We plan to increase our model period to one year to take into account a fuller range of weather patterns.

Atmospheric modeling of dioxin from to the Arctic shows the importance of orientation to high emitting sources to the relative deposition onto receptors, as in the case of atrazine, but at much greater distances (fig. 4). Dioxin is much more stabile in the atmosphere, dioxin is primarily in the particle phase, and is less efficiently scavenged by rain, than atrazine. Only a small percentage of sources were modeled to be responsible for over one half of the emissions to Nunavut receptors.

Although there is model uncertainty, especially in regards to levels of deposition, HYSPLIT appears to be robust in the ranking of relative deposition to the arctic receptors and the ranking of sources. Wet and dry deposition ATCs are presented in Figure 5 for the Coral Harbour marine receptor. The Coral Harbour land receptor wet deposition, is the same, but has an order of magnitude lower dry deposition. Nevertheless, we found that the sources responsible for the most deposition to be nearly the same. Using a different methodology that lowered deposition to marine receptors when covered by snow and ice to be nearly the same as land receptors did not result in differences in the relative ranking of deposition between the eight Nunavut receptors. What is most uncertain in HYSPLIT modeling is the level of air concentration and deposition, not the relative ranking of receptor deposition or relative source rankings.

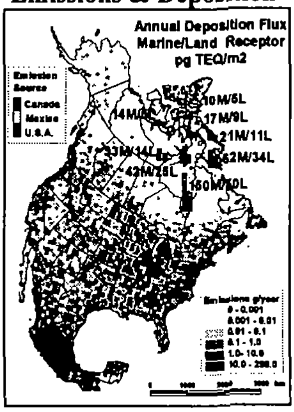


Fig. 4 PCDD/F 1996-97 Emissions & Deposition

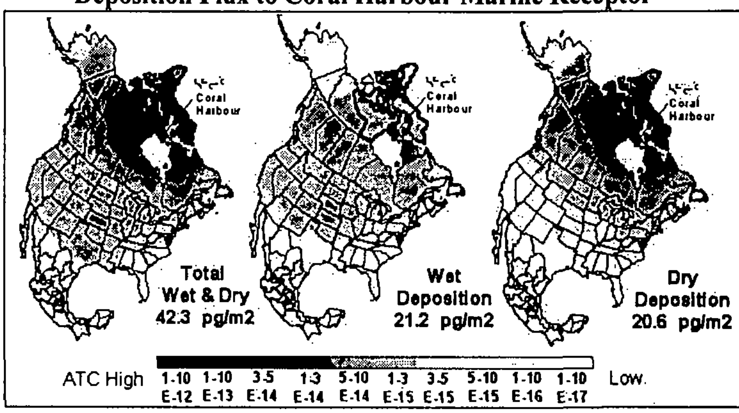


Fig. 5 Wet and Dry Deposition Air Transfer Coefficients & Deposition Flux to Coral Harbour Marine Receptor

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