A multimedia fate and chemical transport modeling system for pesticides: II. Model evaluation

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Abstract
Pesticides have adverse health effects and can be transported over long distances to contaminate sensitive ecosystems. To address problems caused by environmental pesticides we developed a multimedia multi-pollutant modeling system, and here we present an evaluation of the model by comparing modeled results against measurements. The modeled toxaphene air concentrations for two sites, in Louisiana (LA) and Michigan (MI), are in good agreement with measurements (average concentrations agree to within a factor of 2). Because the residue inventory showed no soil residues at these two sites, resulting in no emissions, the concentrations must be caused by transport; the good agreement between the modeled and measured concentrations suggests that the model simulates atmospheric transport accurately. Compared to the LA and MI sites, the measured air concentrations at two other sites having toxaphene soil residues leading to emissions, in Indiana and Arkansas, showed more pronounced seasonal variability (higher in warmer months); this pattern was also captured by the model. The model-predicted toxaphene concentration fraction on particles (0.5–5%) agrees well with measurement-based estimates (3% or 6%). There is also good agreement between modeled and measured dry (1:1) and wet (within a factor of less than 2) depositions in Lake Ontario. Additionally this study identified erroneous soil residue data around a site in Texas in a published US toxaphene residue inventory, which led to very low modeled air concentrations at this site. Except for the erroneous soil residue data around this site, the good agreement between the modeled and observed results implies that both the US and Mexican toxaphene soil residue inventories are reasonably good. This agreement also suggests that the modeling system is capable of simulating the important physical and chemical processes in the multimedia compartments.

Keywords: pesticide modeling, multimedia model, pesticide emission, atmospheric transport and deposition, gas–particle partitioning, model evaluation

1. Introduction
Concern about the adverse effects of pesticides on human health, wildlife and the environment was largely absent until the publication of the book ‘Silent Spring’ [1]. The appearance of this publication, however, raised attention to the adverse effects of pesticides on the environment and spawned a new and vigorous branch of environmental pesticide research that continues to generate many publications. To address problems caused by environmental pesticides, we developed a multi-pollutant modeling system that can simulate agricultural practices as well as other important processes in multimedia compartments including the soil, atmosphere, and vegetation surface. To be more specific, the processes considered...
by the modeling system include pesticide application modes (i.e. spray application, soil incorporation, and seed treatment); spray interception of pesticide and precipitation by the vegetation canopy and subsequent volatilization and wash off; advection and diffusion of pesticides, moisture, and heat in the soil; soil tilling; emissions into the atmosphere; gas–particle partitioning and transport in the atmosphere; atmospheric chemistry; dry deposition and wet deposition. No other models, to our knowledge, can simulate all these processes.

The modeling system that we developed was tested by carrying out a study that investigates the environmental fate and transport of toxaphene, a toxic and highly persistent pesticide, in North America. The model development and implementation was described in the companion paper [2], and the primary objective of this paper is to evaluate the model performance by comparing modeled concentrations, and dry and wet depositions as well as their seasonal patterns against field measurements. In addition, this paper also attempts to assess a published US toxaphene soil residue inventory [3] since it has never been thoroughly evaluated.

2. Brief description of model simulations

As described by Li et al [2], the modeling system consists of a combination of PEM (Pesticide Emission Model) [4, 5] with the US EPA SMOKE/CMAQ Chemical Transport Modeling (CTM) system. The major components of the latter are the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system [6], the Community Multiscale Air Quality (CMAQ) modeling system [7, 8] and the Meteorology–Chemistry Interface Processor (MCIP) [9]. Both PEM and the CTM were driven by the Fifth-Generation Mesoscale Meteorology Model (MM5) [10, 11] using the Pleim–Xiu land surface model [12–14]. Two additional components were developed for the purposes of this study: a Meteorology–PEM Interface Processor (MPIP) and a PEM–SMOKE Interface Processor (PSIP) [2].

1999 US and 1995 Canadian criteria emission inventories were used in the simulations. The US inventories (other than biogenic) were obtained from the US EPA [15]; US and Canadian land use data for calculating biogenic emissions as well as Canadian criteria inventories for other sources (i.e. area, mobile, and point) were obtained from the Ontario Ministry of the Environment [16]. In addition, the 2000 Mexican and American toxaphene soil residue inventories [2] were also used to drive the model in this study.

For this study, the simulations were performed for a domain that covers southern Canada, the contiguous United States, and northern Mexico at a resolution of 36 km × 36 km. There are 22 vertical sigma layers in the atmosphere and 49 layers in the soil, with the highest resolution in both media near the air–soil interface. Only toxaphene air concentrations in the first atmospheric layer were used in the comparisons with surface measurements. The modeled hourly concentrations in the surface grid cell containing the measurement site were used to calculate daily, monthly and annual averages for the comparisons. Seasonal and annual values of the wet and dry depositions into Lake Ontario were also computed from the modeled hourly data. The evaluation was carried out for the period from 1 January, 2000 to 31 December, 2000 across the North America domain as shown in the following sections.

3. Comparisons of model predictions with measurements

We have compared the predictions of our multimedia model with observed daily average atmospheric toxaphene concentrations at five measurement sites across the United States as well as with measurement-based data of wet and dry depositions into Lake Ontario. We have also examined the predicted and measured distribution of toxaphene at locations of different distances from areas of application to separate emission dependences from transport issues.

3.1. Comparison of daily average air concentrations at five measuring sites

Figure 1 shows the locations of five measurement sites that produced daily average concentration data used for the comparisons in this study. The measurements on which the southern boundary condition was based (see [2]) were measured by Hoh and Hites [17] in 2002 at the site labeled LA (29°15′14″N, 90°39′4″W). The sampler at this site was located on the roof of Louisiana University’s two-story DeFelice Marine Center building, which is located only about 17 km from the Gulf of Mexico. The daily average toxaphene air concentration data at other four sites were collected by James and Hites [18] in 2000. The other four sites are: TX: the United States Department of Agriculture, Agricultural Research Service Cropping Systems Research Laboratory in Lubbock, Texas (33°34′33″N, 101°52′31″W); AR: the University of Arkansas Southeast Research and Extension Center at Rohwer, Arkansas (33°45′39″N, 91°16′32″W); IN: a porch adjacent to the School of Public and Environmental Affairs building,
Indiana University in Bloomington, Indiana (39°10′00″N, 86°31′17″W); and MI: the Integrated Atmospheric Deposition Network site on the north-eastern shore of Lake Michigan (44°48′47″N, 86°03′32″W). The measured gas phase toxaphene air concentration data at all the five locations will be used later for model-measurement comparisons.

Figure 2 shows the overall comparison of modeled daily average gas phase toxaphene air concentrations for the year 2000 with corresponding measurements made at the five sites indicated in figure 1. The CMAQ output (a mixing ratio in ppmV) was converted to pg m\(^{-3}\) using the MM5 temperature and pressure for the purpose of this comparison. The modeled concentrations are an average over the volume of the 36 km \(\times\) 36 km surface grid cell in the surface atmospheric layer at the measurement site.

The two dashed lines indicate deviations of plus and minus a factor of five from the 1:1 line (solid black). The large black symbols indicate the averages of all measured and modeled toxaphene concentrations at each of the five measuring sites. The average model results and field measurements agree to within a factor of 3.5 at all sites except TX, for which the model results are lower than the measurements by approximately a factor of 30. We believe this discrepancy is caused by incorrect soil residue data around the Texas site in the US toxaphene residue inventory and will provide further supporting analysis of this contention later. Figure 2 also shows that the modeled and measured toxaphene air concentrations may differ by 3–4 orders of magnitude between the sites, which illustrates the range over which the model provides accurate results.

Figure 3 shows the time series for the year 2000 of the modeled (green curve) and measured (red symbols) gas phase toxaphene air concentrations at the IN, MI, AR and LA sites. (Note that for the LA site, the model results refer to 2000, but...
the measurements were done in 2002; therefore only modeled and measured gas phase air concentrations were plotted for this site whereas modeled particle phase concentrations (blue lines) were also included for the other sites.) The measurements for IN, MI and AR are from James and Hites [18] and those for LA are by Hoh and Hites [17]. The blue curves are modeled particle phase toxaphene concentrations, using the toxaphene soil residues and criteria emission inventories including area, point, mobile, and biogenic sources, which range from about 0.5% to 5% of the total concentrations. While there are no measurements of particle phase toxaphene at the measurement sites, we can make a rough estimate of expected levels from the physical properties of toxaphene using the $K_{oa}$ approach.

At 20 °C, Henry’s law constant ($H$) for technical toxaphene is 0.30 Pa m$^3$ mol$^{-1}$ [19], and the measured log $K_{OW}$ (the octanol–water partition coefficient) is 5.50 [20]. Thus, using $K_{oa} = K_{OW} \times RT/H$, log $K_{oa}$ (the octanol–air partition coefficient) is found to be 9.41. Assuming $f_{om}$ (the organic fraction of particulate matter) is 0.2 [21], log $K_p$ (the particle/gas partitioning coefficient) is found to be [2, 22–24]:

$$\log K_p = \log K_{oa} + \log f_{om} - 11.92 = -3.19$$

and $K_p = 6.3 \times 10^{-4}$. Assuming the mass density of TSP (total suspended particles) is 40 μg m$^{-3}$ [21], the fraction of toxaphene on particles, $\phi$, is found to be about 3%, which is consistent with the results in figure 3.

Figure 3 shows good general agreement between modeled and measured daily average gas phase air concentrations at each of the four measurement sites. The modeled concentrations for IN and AR sites have a discernable seasonal variation (higher in warmer months), whereas this trend is much less obvious for the MI and LA sites. It is noted that during the simulation period the toxaphene residue inventory showed no toxaphene soil residues, resulting in no emissions, at the MI site or LA site, with part of the grid square in which the LA site is located being water in the Gulf of Mexico, and thus the modeled concentrations at these two sites, must be due to atmospheric transport only. It is noteworthy that the modeled toxaphene air concentrations caused by atmospheric transport alone can be as high as 508 pg m$^{-3}$ at the LA site, agreeing well with measurements. The good agreement between the modeled and measured daily average toxaphene concentrations at the MI and LA sites suggests that the modeling system simulates atmospheric transport accurately. There are soil residues in the AR and IN sites, so the air concentrations modeled at these two sites are due to both local emissions from the soil in the grid cells and atmospheric transport. Compared to the other measuring sites, the levels are noticeably lower at the MI site because it is the farthest from the locations of heaviest application in the southern United States and Mexico.

3.2. Discussion on the TX site

There was one exception to the general agreement of the model predictions with the available measurements. This occurred at the TX site. The relevant information is shown in figure 4. The top panel shows the measured gas phase concentrations (red symbols) [18], the corresponding model-predicted gas phase concentrations (green curve) and the modeled particle phase results (blue curve). The average gas phase model result underestimates the average gas phase measurements by about a factor of 30, whereas the average model-measurement differences at the other sites range from +1.2 to −3.5. The possible explanation for the large discrepancy at the TX site is in the information shown in the other panels of figure 4. Each of these is a section of 11 × 11 grid cells centered on the TX or AR measuring site, which is indicated by the star at the center of each panel.

The amount of historical toxaphene use and the exact locations where it was used are not known accurately, making residue distribution estimates somewhat uncertain. The US toxaphene residue inventory used in this work was estimated by Li et al [3], based on national historical usage of toxaphene in the United States, combined with cropland data—primarily cotton growing data because more than 85% of toxaphene was used for cotton. Annual emission factors were then applied to these inventories to estimate emission losses and hence residual toxaphene in the soil [3]. Since the soil residues are primarily a consequence of toxaphene use in agricultural land, we further examined the two agricultural datasets: cropland intensity data from Environment Canada [25] and agricultural land use data from the Ontario Ministry of the Environment (MOE) [16].

The Texas site, like the Arkansas site, with which it is most similar, was chosen for the field study by James and Hites [18] based on its proximity to cotton growing areas. The results of the measurements at these two sites are high. The agricultural land use around these sites is similar, a fact that was noted in the report of the field measurement by James and Hites [18]. Panels (b) and (c) of figure 4, however, show that this is not the case. They show, respectively, the percentage of land specified as cropland for TX and AR in the US inventory obtained from Environment Canada [25] and indicate that near the TX site, only about 4% is designated cropland, whereas this is about 63% at the Arkansas site. Panel (d) of figure 4 shows that the soil residues for the TX site are also very low. These low residues, of course, led to the erroneously low model prediction for the pesticide air concentrations. Conversely, the higher soil residue concentrations, consistent with agricultural land use around the AR site (panel (c)), lead to the higher modeled air concentrations, which are in agreement with measurements. We conclude, therefore, that the land use data and residue data for the TX site are incorrect.

We compared the cropland intensity dataset from Environment Canada, which is shown in panel (b) of figure 4 [25] with another dataset for the same location that was developed by the Ontario MOE [16]. The latter is shown in panel (e) of figure 4. The MOE data show that the agricultural land use at the TX site is similar to that for the AR site, which is shown in panel (c). Had these land use data been considered in estimating US toxaphene soil residue inventory, the resulting higher toxaphene residues would have led to higher modeled toxaphene air concentrations at the TX site, which would be expected to be also similar to the modeled concentrations at the AR site (annual average: 487 pg m$^{-3}$) and therefore to be in closer agreement with the measurements (average in 2000: 355 pg m$^{-3}$).
data and the toxaphene residue distribution around the Texas site may be incorrect. This conclusion is also consistent with the facts that toxaphene was primarily used on cotton growing land, and that the cotton production accounts for more than 80% of Lubbock’s crop production [26], where the TX site is located.

3.3. Comparisons of monthly/yearly average air concentrations in the Great Lakes region

Toxaphene in the Great Lakes region is a matter of increasing concern as the elevated toxaphene concentrations in fish has resulted in restrictions on fish consumption in the Great Lakes.
Table 1. Comparison of modeled and measured monthly/annual average toxaphene concentrations in the Great Lakes Region (Units: pg m\(^{-3}\)) \(^{(a)}\).

| Lake     | Model (average for 2000) | Model (period)      | Measured (period) | Notes |
|----------|-------------------------|---------------------|-------------------|-------|
| Ontario  | 13                      | 30 (June 2000)      | 25 ± 20 (June 2000) | (b)   |
|          |                         | 12 (July 2000)      | 19 ± 4 (July 1998) |       |
| Erie     | 15                      | N/A                 | N/A               |       |
| Huron    | 6                       | N/A                 | N/A               |       |
| Michigan | 7                       | 6.4 ± 2.2 (Sept. 1996 | to Dec. 1997)       |
| Superior | 3                       |                     |                   |       |

Notes:

\(^{(a)}\) Modeled results are given only for those months for which measurements were available.

\(^{(b)}\) Unpublished data collected by Jantunen and Bidleman reported by Burniston et al [28].

\(^{(c)}\) Averages from September 1996 through December 1997 at Eagle Harbor, Michigan, near Lake Superior [29].

Lakes system. Measurements of particle phase toxaphene in the Great Lakes region were conducted during 1997 and 1998 [27], which afford a rough comparison with the particle phase modeling results for the MI site. The comparison is complicated by the fact that for most of the measurements of the particle phase toxaphene concentrations were below the instrument detection limit of 0.25 pg m\(^{-3}\), but a range can be estimated using the following two limiting assumptions: all the concentrations below the detection limit are zero or all are just at the detection limit. The former assumption results in an average particle phase toxaphene concentration of 5.6%; the latter yields 6.5%. As mentioned in section 3.1, the model predictions range between 0.5% and 5.5% in the particle phase at the measurement sites. Since the modeled result also includes uncertainties in the prediction of such quantities as the fraction of organic matter in the aerosols and the mass density of total suspended particles, we consider this to be good agreement.

Additional comparisons of model predictions with averaged measurements made in the Great Lakes region are shown in table 1. Some of the measurement and modeling periods are not concurrent and no measurements were available for Lakes Erie and Huron, but the model predictions agree reasonably well with the available measurements. For the air concentrations at the center of Lake Ontario, the modeled monthly average concentrations for June and July of 2000 (30 pg m\(^{-3}\) and 12 pg m\(^{-3}\), respectively) are quite close to the observations over Lake Ontario, for June 2000 (25 ± 20 pg m\(^{-3}\)) and July 1998 (19 ± 4 pg m\(^{-3}\)), which are based on unpublished measurements of Jantunen and Bidleman that were reported by Burniston et al [28]. The modeled annual average concentration for the year 2000 at the center of Lake Ontario was 13 pg m\(^{-3}\). The modeled annual average toxaphene air concentrations close to the centers of Lake Michigan and Lake Superior (7 pg m\(^{-3}\) and 3 pg m\(^{-3}\), respectively) in the year 2000 also agree well with the average concentration of 6.4 ± 2.2 pg m\(^{-3}\), which was measured by Glassmeyer et al [29] at Eagle Harbor, Michigan (near Lake Superior) from September 1996 through December 1997.

3.4. Comparison of dry and wet deposition

We also compared the modeled toxaphene dry and wet depositions to Lake Ontario against field measurements. The annual toxaphene dry deposition into Lake Ontario for the year 2000 predicted in our study is 27 kg yr\(^{-1}\), which agrees well with an estimate of 26 kg yr\(^{-1}\) based on toxaphene air concentration measurements from a variety of studies reported by Burniston et al [28]. This agreement is much better than a previous study [30], which underestimated annual toxaphene dry deposition into Lake Ontario for the year by a factor of 39.4. Our modeled value for annual toxaphene wet deposition into Lake Ontario for 2000 is 23 kg yr\(^{-1}\), which, given the variability in precipitation as described later in this section, is in reasonable agreement with the toxaphene loadings for the years 1995 through 1998 (12, 17, 12, and 13 kg yr\(^{-1}\), respectively) that were summarized by Burniston et al [28] based on precipitation measurements.

The study of Burniston et al [28] also reports measured seasonal patterns of wet and dry deposition into Lake Ontario. Figure 5 shows a comparison of the measurement-based seasonal dry deposition estimates of toxaphene into Lake
Ontario with the corresponding modeled values (including both gas and particle phases). The seasons are defined as spring (March, April, May), summer (June, July, August), autumn (September, October, November), and winter (December, January, February). The respective modeled values are 8.6, 9.1, 5.7, and 3.9 kg/season, which agree quite well with the corresponding values based on measurements.

Figure 6 compares the seasonal wet depositions into Lake Ontario modeled for the year 2000 with those measured for 1995 through 1998 as reported in Burniston et al [28]. Unfortunately there are no measured data for the year 2000, but there is good qualitative agreement with the previous years for all seasons except spring. Precipitation data from the Northeast Regional Climate Center [31] shows that the Spring of 2000 was the second wettest in 106 years, with precipitation in New York state being 144% of the normal value and larger than the spring values for 1995 through 1998 inclusive. It is reasonable, therefore, to ascribe the discrepancy between the modeled value for spring of 2000 and the measured values for the other years to this difference in precipitation.

4. Discussion

The comparisons show that the modeled daily average toxaphene air concentrations at measurement sites in Indiana, Arkansas, Michigan, and Louisiana agree well with field measurements [17, 18]. Good agreement was also achieved for modeled and measured annual or monthly average toxaphene concentrations in the Great Lakes region. There was also reasonable agreement in both the annual total values and the seasonal patterns of modeled and measured dry and wet toxaphene depositions into Lake Ontario. These good agreements between model results and field measurements suggest that the modeling system is capable of simulating the important physical and chemical processes in the soil, in the atmosphere, and on vegetation surface as well as those related to agricultural practices.

Detailed scrutiny suggests that the origin of the large discrepancy (by a factor of 30) at the TX site is the anomalous toxaphene soil residue data in the published US toxaphene residue inventory around this location. Except the anomalous soil residues data around the Texas site, good agreements between model results and field measurements reported in this study also imply that the US and Mexican toxaphene soil residue inventories are generally reasonable.

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