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Tracking Toxaphene in the North American Great Lakes Basin. 2. A Strong Episodic Long-Range Transport Event

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In this paper we examine the modeled daily toxaphene air concentrations from September 9 to 13, 2000, during which air concentration levels were 2−3 orders of magnitude higher than those derived from in situ measurements around the Great Lakes during the same year and during the 1990s. Meteorological conditions revealed that a typical deformation flow system associated with a high-pressure system extending from the east coast of Canada to the southern United States was one of the critical elements that enabled the transport of toxaphene to the Great Lakes. Cloud bands seen on satellite imagery and the rain band predicted by an atmospheric forecast model indicate that the system also delivered warm and humid air from the Gulf of Mexico and the southern United States to the Great Lakes. This resulted in strong wet deposition of toxaphene to the lakes. Substantial increase in the air concentration of toxaphene over the Great Lakes in this short period contributed greatly to raising the annual average daily air concentration for all of 2000. The results suggest that such an episodic event could be a major pathway for atmospheric transport of toxaphene from the southern United States to the Great Lakes.

Introduction

In the accompanying paper (*2*) we examined the transport of toxaphene in the year 2000 from source regions in North America to the Great Lakes basin. In that study a regional-scale numerical model that included atmospheric transport and soil/air and water/air exchanges (*1*) was used to assess the effect of toxaphene residues in U.S. soils on the toxaphene budget over the Great Lakes basin. The highest monthly averaged air concentration over the basin for the year of 2000 was found in the month of September (*2*). Given the usual strong dependence of toxaphene air concentration and soil/air exchange on temperature (*3*−*5*), the abnormally high toxaphene air concentrations over the Great Lakes in September, a time of year when the temperature in the region was already on its way down from the summer values, was an unexpected result. It was found that the magnitudes of the modeled mean monthly toxaphene air concentrations at the first model level 1.5 m above ground (the model has 12 levels in the vertical from the surface to 7000 m) ranged from several tens to several hundreds of picograms per cubic meter over the lakes. These are 2−3 orders of magnitude higher than those derived from in situ measurements around the Great Lakes during September of the same year and during the 1990s (*2*, *6*). Clearly, the local sources of toxaphene in the northeast United States (see accompanying paper, *2*) with toxaphene soil residues of 1800 t in 2000 are not high enough to yield such high air concentrations. However, with toxaphene residues in soil of 22 700 t in the southeast United States, an order of magnitude higher than those in the northeast, long-range and trans-boundary atmospheric transport on a regional scale to the Great Lakes basin is a possibility (*2*).

A key finding reported in the accompanying paper (*2*) was that the impact of the largest toxaphene reservoir in the southeast United States on the Great Lakes ecosystem was more significant during spring and autumn than during summer, due primarily to interseasonal variations in the atmospheric circulation systems. This indicated that the contributions of toxaphene residue in soil in the different regions of the United States to its concentration levels in the atmosphere and deposition amount to the Great Lakes were season-dependent and underwent a pronounced shift, particularly in the winter/spring and summer/autumn transition periods. The modeling results demonstrated that the impact of the principal toxaphene sources in the southeast United States on the deposition of this insecticide to the Great Lakes was strongest in the autumn season (*2*), the season during which the main track of the jet stream and several common storm tracks originating in the Gulf of Mexico and the southern United States find their way across the United States northward up to the Great Lakes basin. Autumn is also one of the stormiest periods of the year in the region, leading to strong air mass exchange between low and high latitudes. In the winter season, although weather systems are very active and the large-scale atmospheric circulation favors long-range transport, lower air temperatures reduce the soil/air exchange and surface/air partitioning of toxaphene and other semivolatile organic compounds, thereby lowering their levels in the atmosphere and reducing their deposition to the Great Lakes.

In addition to favorable meteorological conditions, the occurrence of an extended episodic long-range transport of a pollutant also depends on the availability of the pollutant in the source region (*7*, *8*), atmospheric circulation with minimal dispersion, and limited chemical destruction (*9*) during transport. In this study toxaphene is assumed to be re-emitted to the air continuously from the contaminated soils in the United States, with the emission rate being determined by the compound's chemical−physical properties at prevailing environmental conditions. Also, with a relatively long half-life in the air (*6*), toxaphene is not subject to much chemical or photochemical degradation in the atmosphere during transport (*10*). Further, because the average atmospheric motion is zonal (west to east), a large-scale meridional (south to north) air mass exchange between the low- and high-latitude regions is most likely to be episodic and on a time scale of a few days to a week. This suggests that toxaphene re-emissions from the major sources in the southeast United States are most likely to impact the Great Lakes ecosystem at such times of episodic meridional long-range transport.

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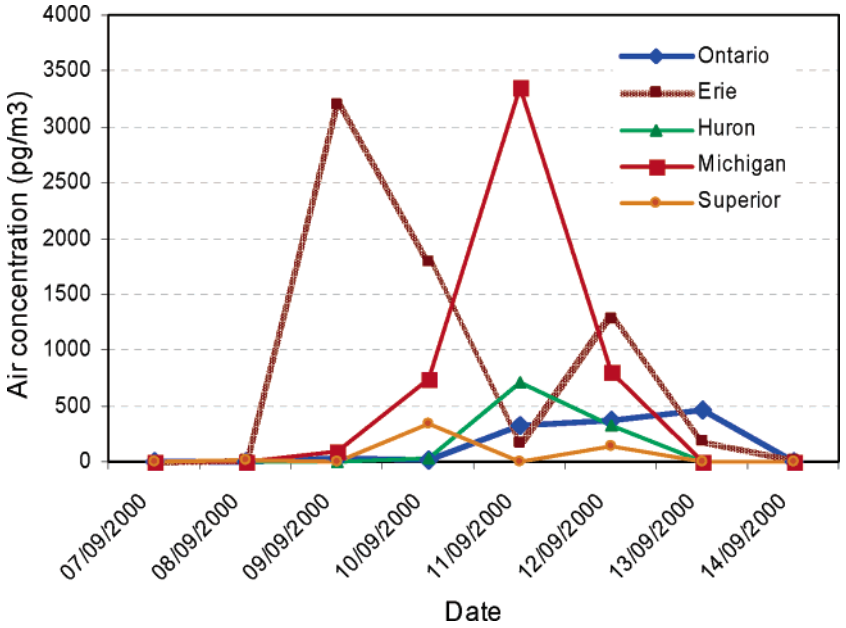

FIGURE 1. Modeled daily air concentration (picograms per cubic meter) at grid point near the center of each lake at 1.5 m height from September 7 to 14, 2000.

Episodic transport of persistent organic pollutants to the Great Lakes has been noted previously (*11*−*13*). Glassmeyer et al. (*13*) have reported observing an abnormally high toxaphene air concentration on September 9, 1996, on the coast of Lake Superior. There were also other high toxaphene air concentration events documented in field studies (*14*−*16*). Such episodic events occur on a relatively large spatial scale (over 1000 km) but their temporal scales are relatively short (less than a week). However, such events are seldom captured by observations because of the lack of sufficient spatial and temporal resolutions in the measurements. As a result, the details of the occurrence of episodic transport of pesticides from North American sources to the Great Lakes basin are still poorly understood. In this paper, we report on a September 2000 event of strong atmospheric transport of toxaphene from its major source region in the southeast United States to the Great Lakes basin. The type of atmospheric motion and weather systems (e.g., fronts) associated with such an event are quite common in the North American continent and also in other subtropical and mid-latitude areas around the world (*17*). The regional-scale model discussed in ref *1*, which includes atmospheric transport and soil/air and water/air exchange of pesticides, is used to perform the model simulations. The detailed modeling strategies and numerical experiments have been described in the accompanying paper (*2*). To identify quantitatively the contribution of different sources in the United States to the toxaphene budget over the Great Lakes, six different source loading scenarios were examined. The analysis in this paper is based on the modeling results from model scenario 1 that includes all toxaphene sources in the United States (*2*).

The September 2000 Event

The modeled daily air concentrations during the 5-day period of September 9−13, 2000, indicate a dramatic increase of toxaphene air concentrations occurring over the entire Great Lakes basin. Over the five-day period, the calculated maximum daily values at the model grids near the center of each of the Great Lakes were 463 pg m^{-3} over Lake Ontario, 3190 pg m^{-3} over Lake Erie, 716 pg m^{-3} over Lake Huron, 3360 pg m^{-3} over Lake Michigan, and 346 pg m^{-3} over Lake Superior. In the subsequent discussions, we will refer to this period of anomalous increase in the air concentrations of toxaphene over the Great Lakes as the *September event*. Modeled daily toxaphene air concentrations at 1.5 m height at these grid points from September 7 to 14, 2000, are illustrated in Figure 1 (note that the event started on September 9, but in the figure we also present the air concentrations on September 7 and 8 as the base values). The air concentration values during the September event are considerably higher than the 0.5−54 pg m^{-3} range measured in the rest of 2000 and cited in ref *2* (3.6 pg m^{-3} measured on September 21, sampled once per month) and also higher than the 1−100 pg m^{-3} range measured over the Great Lakes basin throughout the 1990s as noted in ref *6*. This sharply increased air concentration was also reflected in the monthly averaged air concentrations for September as shown in Figure 3 of the accompanying paper (*2*). Accordingly, the maximum monthly accumulated dry and wet depositions to the lakes occurring in September 2000 (*2*) were also associated with a sharp increase in daily depositions during the same period. For example, the maximum daily wet deposition to Lakes Erie and Ontario during the September event exceeds 2 kg (on September 12); and the maximum daily dry deposition to Lakes Erie and Michigan was 1 and 2 kg on September 9 and 11, respectively. These daily deposition values are even greater than monthly depositions to the lakes in the rest of the months in 2000. Table 1 lists the maximum of all daily air concentrations (picograms per cubic meter) at 1.5 m height averaged

TABLE 1. Standard Deviations of Daily Air Concentrations, Daily Dry Depositions, and Daily Wet Depositions Averaged over Each Lake throughout 2000[a]

	concn (pg m^{-3})		dry deposition (kg day^{-1})		wet deposition (kg day^{-1})	
Lakes	σ_c	C_{max}	σ_{Fd}	$F_{d,max}$	σ_{Fw}	$F_{w,max}$
Ontario	65.0	954	0.009	0.126	0.128	2.450
Erie	210.3	3721	0.054	0.991	0.144	2.287
Huron	55.2	849	0.019	0.271	0.093	1.760
Michigan	215.4	3771	0.121	2.094	0.013	0.206
Superior	23.9	417	0.029	0.546	0.004	0.005

[a] Maximum daily air concentrations (C_{max}) at 1.5 m height, maximum daily dry deposition flux ($F_{d,max}$), and maximum wet deposition flux ($F_{w,max}$) averaged over each lake for the period of the September event are also shown.

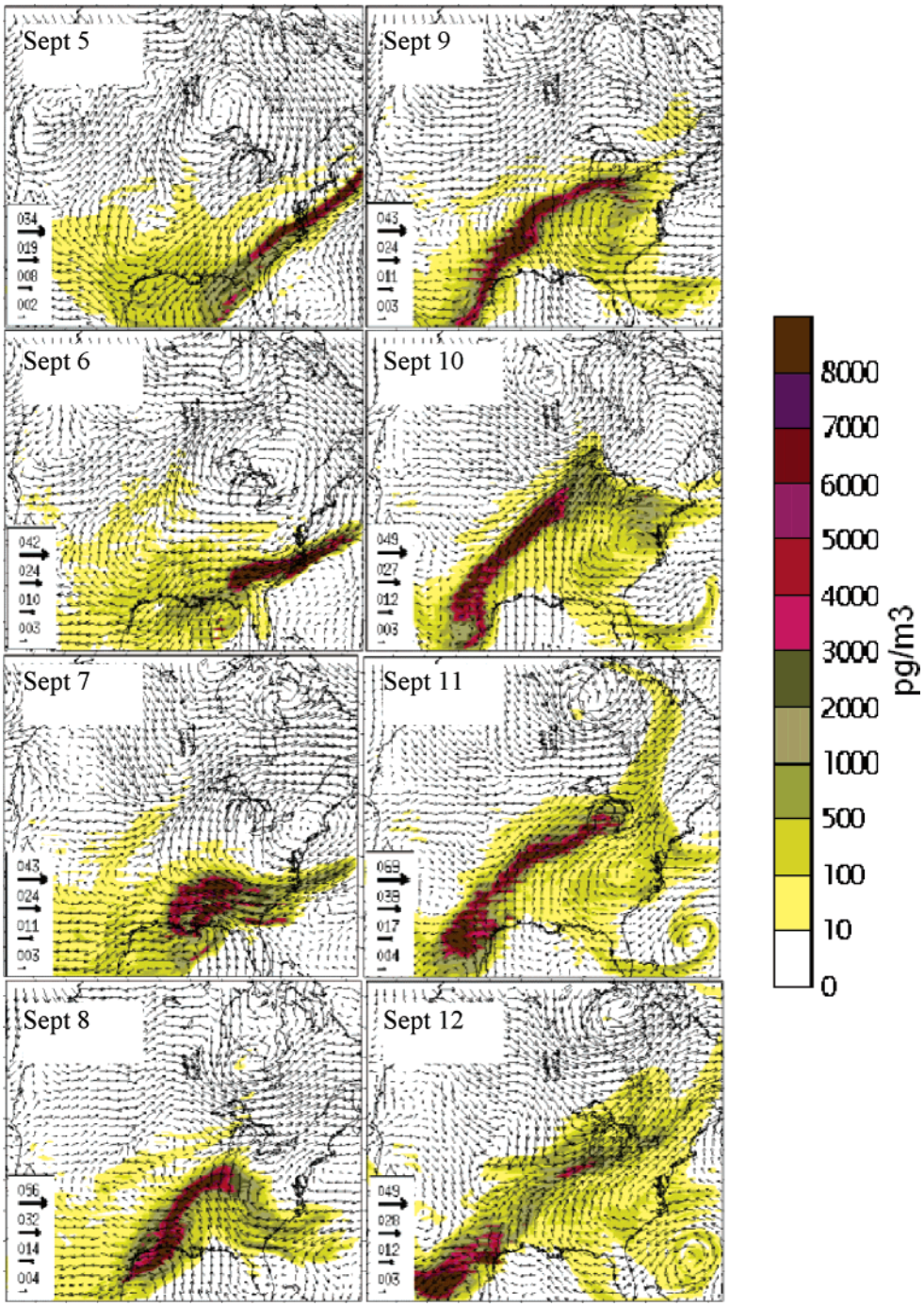

FIGURE 2. Modeled daily air concentration (picograms per cubic meter) overlaid by daily averaged vector winds at 1200 m height above the surface (the eighth model level) for the days of September 5−12, 2000. The bar at left low corner of each figure (knots) indicates magnitude of wind vector.

over each lake for the period of the September event and the standard deviation (picograms per cubic meter) of daily air concentrations averaged over each lake throughout 2000. Table 1 also shows the maximum of all daily dry and wet depositions (kilograms per day) to the lakes during the same period and the standard deviations (kilograms per day) of their respective daily depositions to the lakes throughout 2000. As seen, the maximum daily values of air concentration and depositions over each lake (except for wet deposition to Lake Superior) exceed, by a factor of 10, the standard deviation of their respective daily air concentration and depositions in 2000. It must be noted that the large values of the annual standard deviations are actually due primarily to the exceptionally high air concentrations observed during the September event. For example, if we excluded the daily air concentration for September 9−13, the standard deviation for Lake Erie would become 29 pg m^{-3}, as compared to 210 pg m^{-3} with those days included (Table 1). This suggests that this sporadic event, though occurring only for a few days, has a major contribution to the annual statistics of modeled daily toxaphene air concentrations over the Great Lakes basin.

This anomalous increase in air concentrations of toxaphene over the Great Lakes basin and depositions to the lakes during the September event is clearly an indication of occurrence of a strong long-range atmospheric transport from its major sources in the southern United States under a favorable atmospheric circulation pattern. Figure 2 illustrates

daily air concentration at 1200 m above the ground (the height of the eighth model level in the vertical; *1*) overlaid by daily averaged vector winds (knots) at the same height above the surface for the period September 5−12. It traces clearly the plume of high air concentrations of toxaphene from its principal source region in the southeast United States to the Great Lakes. The plume remains as a narrow band without much dispersion. Associated daily flow patterns indicated that, in early September (e.g., Sept 5 panel in Figure 2), relatively strong southwesterly winds carried the toxaphene from the southeast U.S. sources further east. Starting on September 6, this offshore flow changed direction from the southwest to the northeast, under the influence of an anticyclone (high pressure) centered near Lake Huron. It is interesting to note that this reversing flow carries the toxaphene air concentrations back over the major source region in the southeast United States, thus adding to the toxaphene being reemitted continually from those sources (see Sept 6 and 7 panels in Figure 2). On September 7, the anticyclone shifted eastward toward New England and then turned south on September 8 (Figure 2). The movement of the anticyclone was featured by strong easterly winds on the south side of the anticyclone (over southeast United States) and by southerly winds on the west side of the anticyclone, the southerly winds extending all the way from the southern United States to the Great Lakes as shown in Figure 2 (see Sept 8 panel in Figure 2).

For September 9−11, the anticyclone moved further southward and then remained in the U.S. south Atlantic region. The southerly winds in the west of the core of the anticyclone formed the atmospheric transport path for toxaphene from the southeast United States. On September 9, the plume reached the southern portion of the Great Lakes and the transport path extended further to northern Quebec, reaching as far as the Canadian arctic on September 11. The significant changes in the modeled daily air concentrations over Lakes Erie and Michigan (see Figure 1) are consistent with this transport path for toxaphene. These two lakes in the southern portion of the Great Lakes ecosystem also received the highest toxaphene among the five Great Lakes during the September event. From September 12, the anticyclone in the U.S.. south Atlantic region weakened and moved offshore, with strong northwest winds dominating the Great Lakes basin and carrying the high air concentrations quickly away from the basin (Figure 1).

It may be worthwhile to note that a hurricane can be also identified clearly over the tropical North Atlantic Ocean east of Florida in Figure 2. This is hurricane Florence. Florence did not make landfall during its lifetime but traveled northeastward off the east coast of North America. The model results indicated that relatively high air concentrations of toxaphene were trapped by this tropical storm (Figure 2). It is therefore possible that if toxaphene entered the convective cloud systems of the tropical storm, it would be carried to high levels of the atmosphere and then moved along its trajectory and subsequently deposited to the ground by heavy precipitation. A detailed analysis of the influence of the hurricane on toxaphene transport is beyond the scope of this paper.

Large values of depositional fluxes, especially dry deposition to the lakes, are associated strongly with and positively correlated with the exceptionally high toxaphene air concentrations during the September event. With a constant washout ratio of 1.23×10^4 (*2*), the wet deposition fluxes are determined by the precipitation fields produced by the GEM model. The significant increase in the daily wet depositions to the lake surfaces during the September event suggested that the changes in the atmospheric circulation during the early to mid September period not only caused this transport event but also produced strong precipitation over the Great Lakes basin. Further discussions are presented in following section.

Meteorological Basis for Transport to the Great Lakes Basin

The atmospheric transport pathway, which carries toxaphene rapidly from the southeast United States to the Great Lakes basin as shown in Figure 2, was realized through a unique set of atmospheric dynamics. The anticyclonic motion shown in Figure 2 is a reflection of this. Figure 3 illustrates the mean-sea-level pressure (in hectopascals) at 1200 UTC (Universal time) from September 7 to 12 overlaid by vector winds at 850 hPa. On September 7, a high-pressure system was located along the eastern seaboard of North America. This high-pressure system was subsequently split into two centers on September 9, with the northern one continuing to move northeastward while the southern one remained stationary or even moved slightly southward over the southern part of the U.S. eastern seaboard (see Sept 9−11 panels in Figure 3). At the same time there was a high-pressure system located in the northeastern Pacific off the west coast of North America and also a low-pressure system centered over the Canadian prairies sandwiched between the two high-pressure systems. This configuration is a classic textbook case for frontogenesis (*17*, *18*). The pressure patterns resulted in a strong deformation flow in the region between the low-pressure system and the high pressure along the southeastern United States. This deformation flow acted to increase the convergence, as did also the horizontal temperature gradient in this region, thus creating a strong front (*19*).

The deformation flow pattern can be also identified from Figure 2, which shows a belt of strong southerly to southwesterly winds extending from the southern United States to the Great Lakes basin. One of the axes of this deformation flow with its strong wind convergence zone aids the transport of the high air concentration from the southeast U.S. sources (Figure 2). The location of the front as discussed above is also indicated clearly by the cloud bands in the GOES-8 visible satellite imagery of September 10 at 18:15 UTC (Figure 4a). The convergent winds prevailed in the clear sky region to the west of the front. The relatively narrow, well-developed cloud band covering the region from the southeast United States to the Great Lakes is one that typically produces precipitation. Figure 4b shows the GEM-predicted total precipitation accumulated for September 9−12 and agrees fairly well with the cloud band displayed in the satellite imagery [Figure 4a; the presence of Hurricane Florence can be seen clearly in the satellite imagery (Figure 4a) and also by the precipitation distribution predicted by the GEM model for September 10 (Figure 4b)]. It is also interesting to note that, during the September event, the modeled wet deposition fluxes in the two upper lakes (Lakes Michigan and Superior) were lower than those in Lakes Erie, Ontario, and Huron (Table 1), correlating well with both the cloud and rain bands, which cross mostly the eastern portion of the Great Lakes basin (Figure 4).

The above analysis describes the impact of meteorological conditions in the continent on the toxaphene budget over the Great Lakes during the September event. The deformation and convergent winds associated with the surface pressure patterns formed a great conveyer belt for the transport of toxaphene from its major source region in the southeast United States to the Great Lakes. The surface level convergence, in turn, created a large-scale ascending motion. To gain more insight into this toxaphene conveyer belt in the atmosphere and the transfer of toxaphene by the large-scale ascending motion, in Figure 5 we illustrate the vertical profiles of modeled daily toxaphene air concentration for September 6−12 from the surface to 3000 m height (the 10th model level

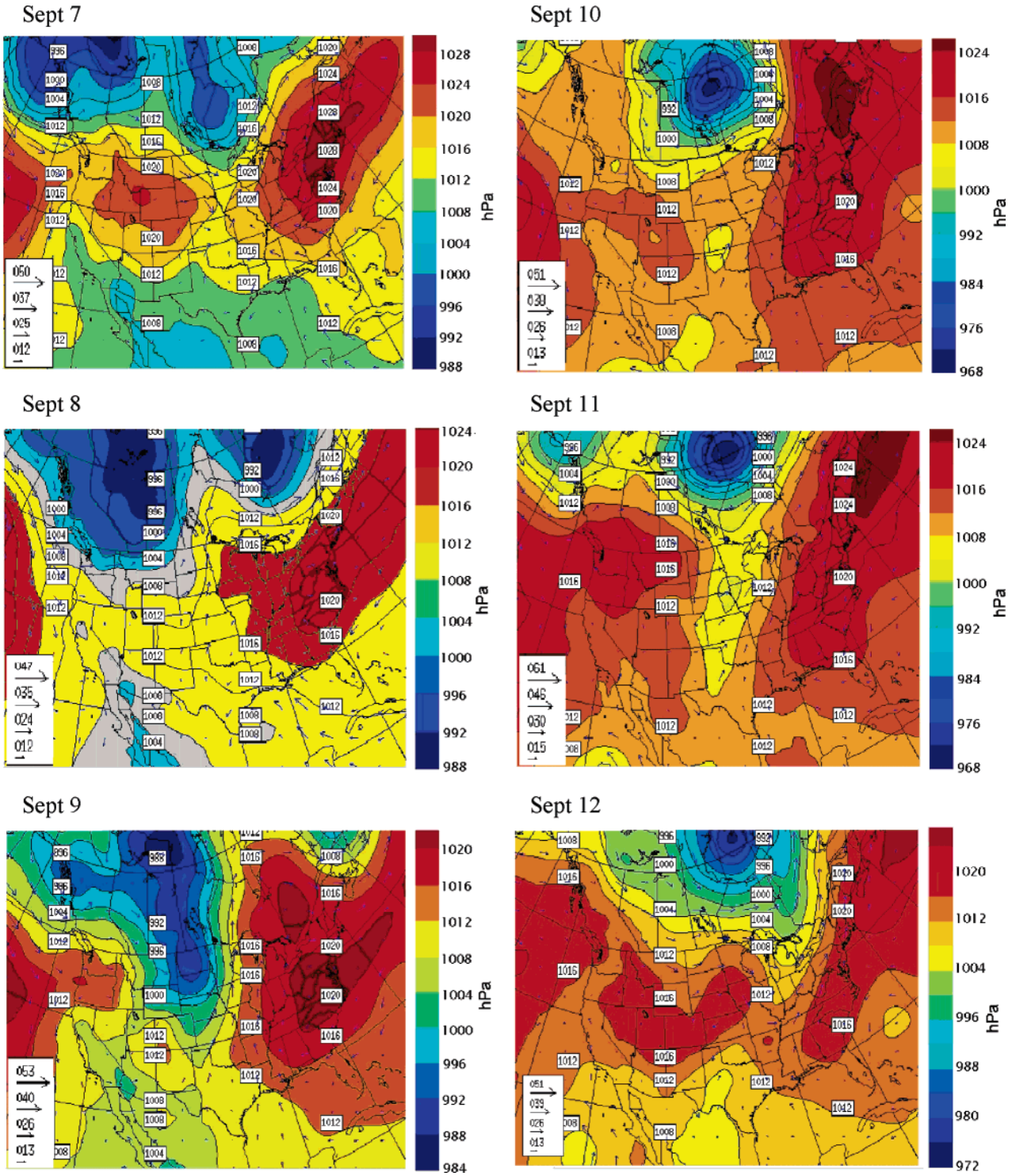

FIGURE 3. Mean-sea-level pressure (hectopascals) at 1200 UTC from September 7 to 12 overlaid by winds (denoted by arrows) at 850 hPa. Wind speed scale is in knots. Note that the domain is different from the model domain. Source: http://weatheroffice.ec.gc.ca/data.

in the vertical; *1*). These vertical profiles are along the blue line in Figure 4b, a line passing through grid point 150 (see Figure 2 of the accompanying paper). The value at a given point along this line is the average of the values between vertical lines 120 and 180 along the row passing through that point. That is, the vertical profile (along the blue line in Figure 4b) represents the average of the values along the belt bounded by the solid black lines in Figure 4b, a belt that includes the region from the southeast U.S. sources to the Great Lakes basin. Although the source of toxaphene was ground-based (from contaminated agricultural soils), model simulations showed the presence of high air concentrations of this insecticide at relatively high levels of the atmosphere throughout the September event, as seen in Figure 5. This is due primarily to the strong updrafts that followed the reverse flow (due to easterly winds), which brought the toxaphene that was already offshore back over the source region, thus enhancing the toxaphene air concentrations. The daily changes in the concentration profiles in the vertical further confirmed northward transport of the toxaphene in air from its principal sources in the southern United States during the September event. This transport occurred most

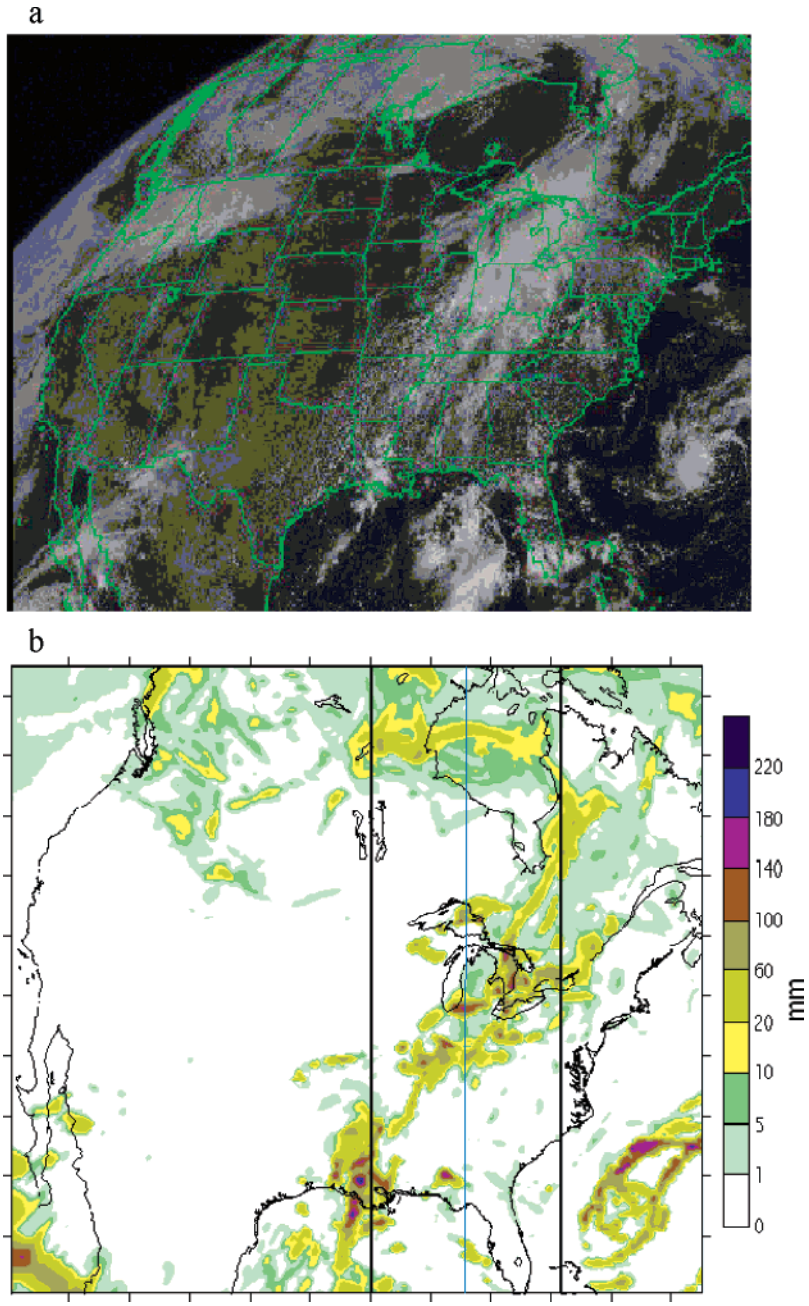

FIGURE 4. (a) GOES-8 visible satellite image on September 10, 2000, at 18:15 UTC. The image was made available through NOAA Satellite and Information at www.ncdc.noaa.gov/oc/ncdc.html. (b) Accumulated GEM predicted daily precipitation (millimeters) for September 9−12, 2000. Solid black and blue lines indicate cross-section and central line for vertical profiles of modeled daily air concentration as illustrated in Figure 5.

effectively at the higher levels in the atmosphere rather than near the surface, where strong surface friction and turbulence quickly dissipate any well-organized large-scale flow and weaken any transport of the pesticide.

In addition, this weather system creates a highway that transports warm and humid air from the southern United States and the Gulf of Mexico northward up to Hudson Bay. This can be seen clearly from the spatial pattern of the deformation flow (Figure 2) and also from the narrow cloud and rain bands (Figure 4). In other words, the warm and humid air originating from the lower latitudes combined with the large-scale convergence would become a primary source for the precipitation, which in turn determined the temporal and spatial distribution of the toxaphene wet deposition fluxes to the Great Lakes (Table 1).

Transport into Mexico

In the course of this study it was also seen (Figure 2) that there was significant transport of toxaphene from the major U.S. sources toward Mexico (as no toxaphene emissions inventory from Mexico was available, the model simulations did not include any emissions of toxaphene from Mexican sources). At the model grid point (54, 18) [see Figure 1a of the accompanying paper (*2*) for location] representing central Mexico, we found that the modeled toxaphene air concentration at 1.5 m height increased from 36 pg m^{-3} on September 6 to 720 pg m^{-3} on September 10 and then to 8824 pg m^{-3} on September 12. The evolution of the winds and sea-level pressure fields illustrated in Figures 2 and 3 suggests that the strong easterly winds in the southeast United States associated with southward movement of the anticyclone off the

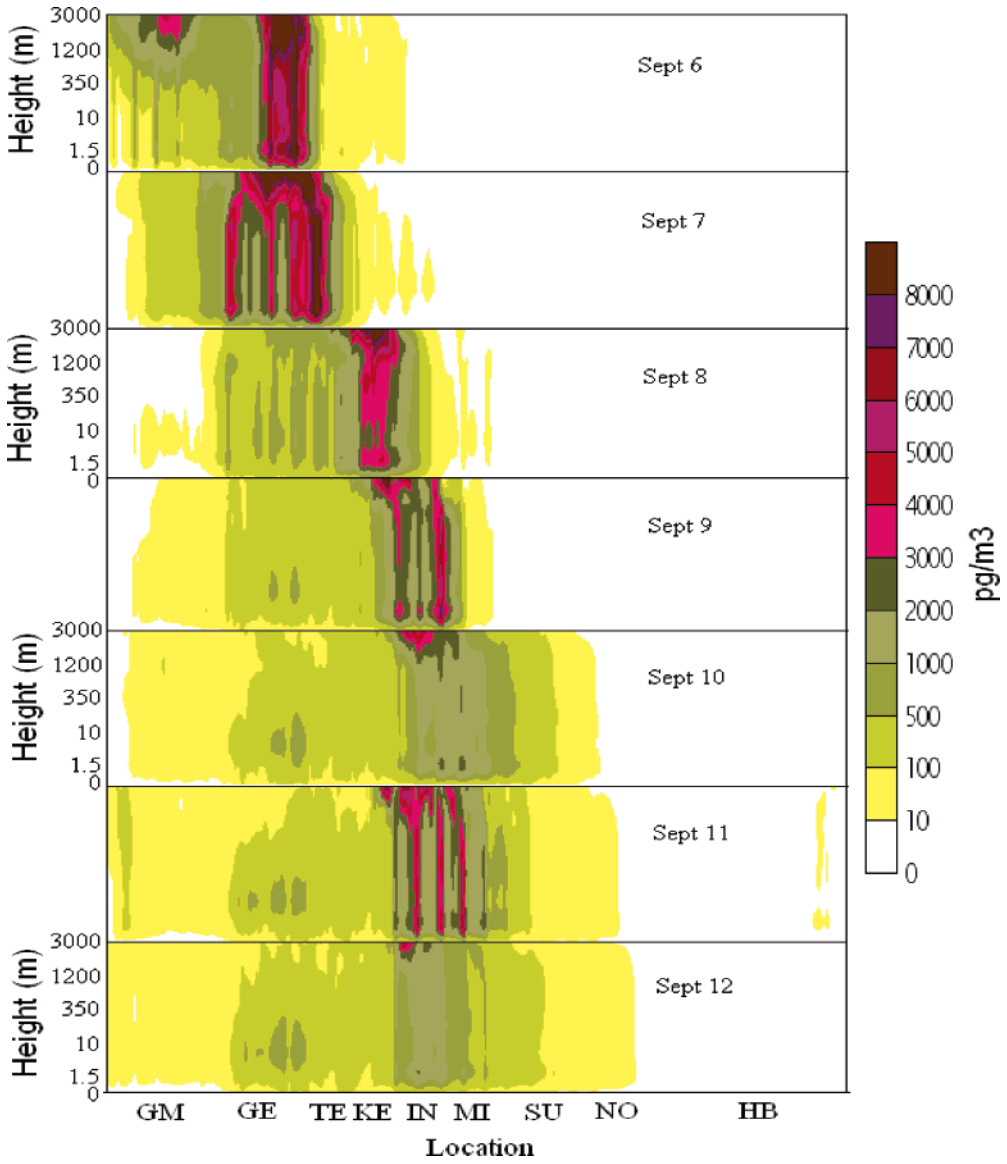

FIGURE 5. Meridional cross section (along the blue line in Figure 4b) of vertical profiles of modeled daily air concentration (picograms per cubic meter) from the surface to 3000 m height for September 6−12, 2000. The values along the blue line are zonally averaged values within the solid black lines in Figure 4b (grids 120−180 along *x*-axis). The location (abscissa) indicates regions across the central line of this belt (along grid 150, the central grid line between 120 and 180, as shown by the solid blue line in Figure 4b). These are GM, Gulf of Mexico; GE, Georgia; TE, Tennessee; KE, Kentucky; IN, Indiana; MI, Michigan; SU, Lake Superior; NO, northern Ontario; and HB, Hudson Bay.

eastern seaboard would deliver toxaphene air concentrations from the southeast United States to Mexico before September 8. Thereafter, the typical deformation flow, one component of which has winds directed toward the Great Lakes while the other pushes the toxaphene southwestward (see Sept 11 panel in Figure 2), played an important role in the continued transport of toxaphene to Mexico.

An Extended Memory of the September Event in Soil

Compared with the atmosphere, soil is a slowly varying medium. In our model, toxaphene enters the soil primarily via dry and wet depositions. It is then redistributed between the gaseous, dissolved, and adsorbed phases (*1*, *4*) within the soil. The much longer half-life of toxaphene in soil than in air implies that once the increased toxaphene air concentration leads to increased deposition, it results in increasing concentrations of this chemical in the soil, which then persist for a longer period than in the atmosphere. We would then expect that, although the episodic high air concentrations over the Great Lakes during the September event were observed for only a few days as shown in Figure 1, its impact on the environmental fate of toxaphene in the ecosystem is likely to persist for many days after the event. To gain insight into this extended memory of the September event in the soil, Figure 6 shows the modeled daily soil concentrations (micrograms per kilogram) at two selected model grid points in the Great Lakes basin. On the basis of the initial soil residue distribution as shown in Figure 1b of the accompanying paper (*2*), the model grid in southern Michigan [grid (155, 100), Figure 6a] can be regarded as a weak (local) source grid, and the other in western Ontario in Canada, as a receptor grid (167, 100) (Figure 6a). Temporal variation of soil concentration within the three soil layers of the model are examined for the source grid (Figure 6b) and for the receptor grid (Figure 6c). At the source grid, the soil concentrations (Figure 6b) in the deep "reservoir" layer (1−10 cm below the surface) defined in refs *2* and *4* decline

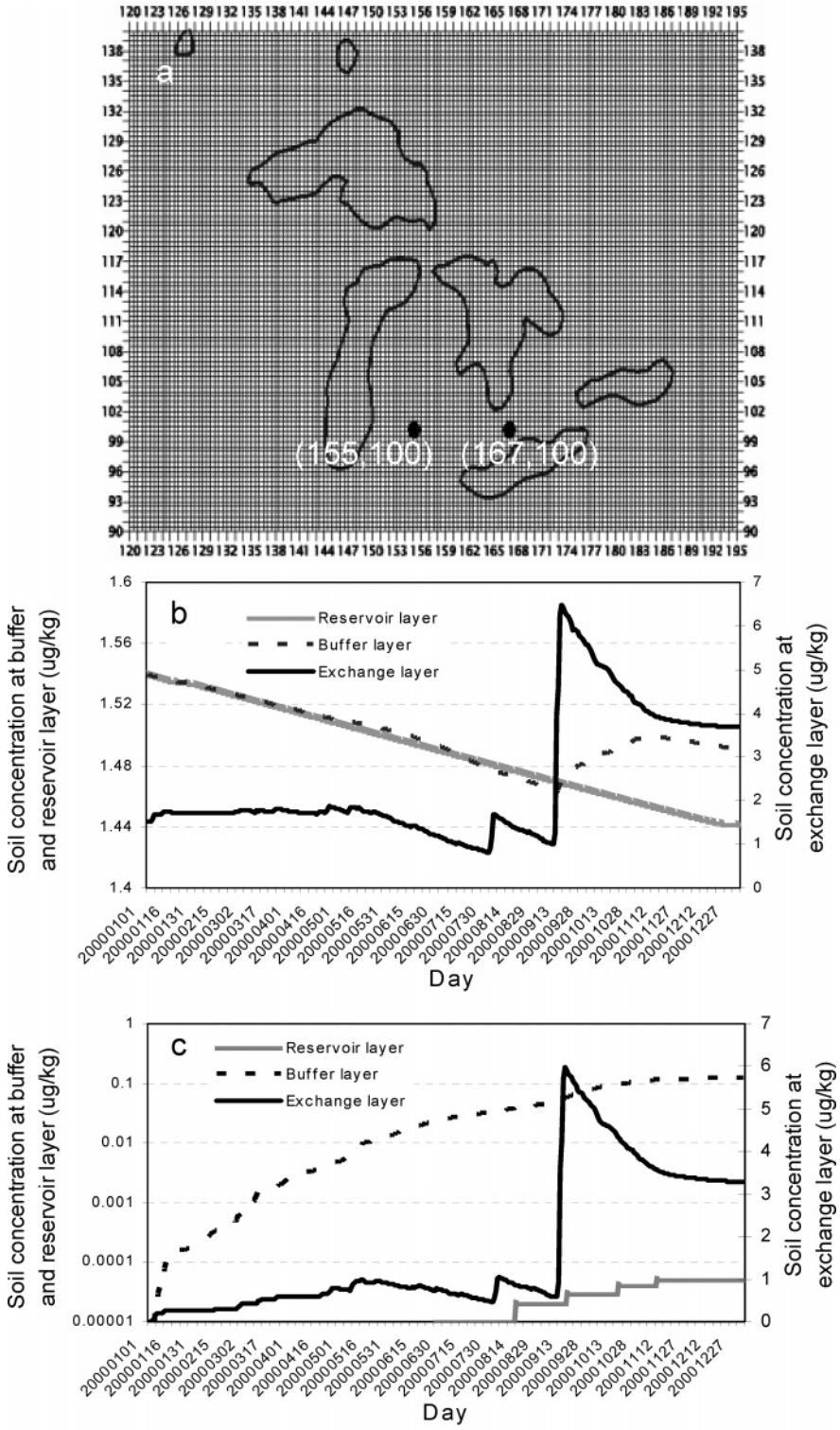

FIGURE 6. (a) Model grids in the Great Lakes basin; two grids indicating a weak source grid (155, 100) and a receptor grid (167, 100) are marked. (b) Modeled daily soil concentrations of toxaphene (grams per kilogram) (b) at a weak source grid (155, 100) and (c) at a receptor grid (167, 100) in the Great Lakes basin in 2000.

monotonically over the course of the year, due largely to degradation. In the relatively shallow "buffer" soil layer (0.1−1 cm), the temporal trend of soil concentrations is mostly identical to that in the "reservoir layer" (toxaphene was assumed to be well-mixed initially in the soil at the source grids) but increases substantially after September 10, indicating the strong soil/air exchange resulting from the rapid increase in air concentration and subsequent deposition. Because of the quick response of the top (exchange) soil layer (0−0.1 cm) to changes in air concentration, the greatest increase in the soil concentration occurred in this layer. The increase of the soil concentrations of toxaphene persists for two months, well into mid-November, as shown by the buffer layer soil concentrations in Figure 6b.

While the concentrations of toxaphene in the soil exchange layer (the top layer) varies through the summer months (June to August) at the selected source (Figure 6b) and receptor (Figure 6c) grids in the Great Lakes basin, the concentration changes are most pronounced during the September event (Figure 6b,c). Compared to the early summer (June), the soil concentrations in the exchange layer during the period of the September event increased by a factor of 5 at the weak source grid and by a factor of almost 10 at the receptor grid. The sharply increased concentrations in the soil exchange layer were then reflected in the buffer layer via leaching and diffusion. Clearly, the chemical exchange between the exchange and buffer layers in the soil is much slower than that between the air and the exchange layer of the soil. The sharp increase in the concentration levels in the exchange layer in fact leads to a reversal of the decreasing trend in the buffer layer at the weak source grid after September 9 (Figure 6b). At the receptor grid, the concentrations in the buffer layer are enhanced. Given these increases in the buffer layer concentrations, the impact on the Great Lakes ecosystem continues long after toxaphene in the atmosphere was cleared away from the Great Lakes basin by strong northwest winds after September 12. Also, given the fact that by September the air temperatures in the Great Lakes basin were already on their way down, emissions may be subject to global distillation/cold condensation effects (*20*), thus further prolonging the period for which the Great Lakes basin ecosystem may be impacted. It is very possible that the enhanced toxaphene soil residues may re-volatilize into the atmosphere during the spring/summer months the following year when the air temperatures are once again on the upswing.

It should be pointed out that both field measurements and modeling studies (*21*−*23*) have revealed that vegetation (forest, grass, etc.) canopies may play an important role in the fate of organochlorine pesticides. It has been reported (*21*, *22*) that forests tend to increase net atmospheric deposition, decrease air concentration, and speed up chemical transfer between air and soil. Toxaphene is a semivolatile compound that is likely affected by atmosphere−vegetation exchange processes (*23*). Such processes may include degradation loss, foliar uptake though dry particle deposition and gaseous uptake in canopies, absorption by vegetation, and plant/air partition. In the coupled modeling system (*1*) employed in this study, the effect of plant canopies on turbulent exchange of air concentrations is introduced through the surface roughness lengths for momentum and heat transfer near an underlying surface, which are defined on the basis of different surface properties (*24*). The soil/air exchange module considers explicitly the effect of plant canopies on dry particle deposition (*1*, *24*). However, other potential air/vegetation exchange processes (e.g., gaseous dry deposition) were neglected because of significant uncertainties that still exist in the parametrization of these processes. Although this could potentially result in an overestimate of atmospheric concentrations and an underestimate of soil concentrations (*23*), it is unlikely to change the direction of the exchange flux between the air and soil and hence the conclusions from this study.

The numerical modeling results presented in this study provide evidence of a strong episodic long-range and transboundary transport of toxaphene from major sources in the relatively low-latitude regions of the southern United States to the mid- and high-latitude regions in the Great Lakes basin and further north. It is worthwhile to point out that, to identify and study episodic long-range transport events with a time scale of several days to a week, daily meteorological conditions must be taken into account in any organochlorine pesticides model. Given that the mean atmospheric flows are essentially zonal (east−west), meridional atmospheric motion that determines south−north exchange of air mass and pollutants is in reality a perturbation with a time scale of less than a week over the mean atmospheric circulation. Such a perturbation is easily filtered out by monthly and annually averaged atmospheric circulations. The outcomes of the study could be beneficially applied to identify potential sources and pathways of some of the other toxic compounds detected in the Great Lakes ecosystem. It is logical to expect that, for those organochlorine pesticides that were used or are being used heavily in the southern United States (*4*, *14*−*16*, *22*), their transport to the Great Lakes basin is likely to be associated with similar episodic transport events. Given their relative intensity, such episodic events can have a large impact on the annual budget of toxic compounds over the Great Lakes ecosystem. Thus the episodic long-range transport process can be regarded as a major pathway for compounds in the Great Lakes basin. Considering that strong episodic transport of organochlorine pesticides may not readily be detected by existing in situ measurement systems, modeling may be a viable approach for the investigations of episodic transport of organochlorine pesticides into other ecologically sensitive environments such as the North American arctic.

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