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Current Knowledge of Air Pollution and Air Resource Issues in the Lake Champlain Basin

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ABSTRACT

The Lake Champlain basin, although predominately rural, is exposed to a variety of atmospheric pollutants and related environmental stresses, largely originating outside the basin. These include acid rain, dry deposition of sulfur and nitrogen compounds, organic and inorganic toxic substances in gaseous and particulate forms, tropospheric ozone, ultraviolet radiation, and climate change. The relatively large land area of the basin (18:1 land:lake area) is a sink for airborne pollutants which can be captured and transferred in the basin, producing direct and indirect effects on the lake and surrounding ecosystems. Because of the growing human population in this region, complex and diverse ecosystems, and multiple ecosystem management issues, it is important to assess the status and impacts of atmospheric contaminants in the basin. This report summarizes the relevant information on regional air quality during the past two decades, collected at several monitoring sites in northern Vermont, New York and Quebec. The data show patterns and trends for concentration or deposition of acidifying compounds, mercury, toxic compounds, fine particulates and ozone. Exposure to many of these contaminants continues to threaten human and ecosystem health in the basin, despite increased regulation of air pollutants. Analysis of regional meteorology and application of air transport models to these data show the probabilistic extent of the airshed affecting the basin and the contaminant source regions. Finally, information gaps, research needs and management issues are examined to provide guidance for future work to improve understanding and control of air pollution issues in the basin.

INTRODUCTION

> "The smoke of coal pits covered the land. The trees were swept away as if some gigantic scythe bearer had mowed them over. One may travel now for miles in that region and not find a tree large enough to make a respectable fish pole."
>
> - L.E. Chittenden, around 1890

As this quote demonstrates, air pollution in the Lake Champlain basin is not a new problem, but certainly the form and extent of pollution has changed substantially in the past 100 years. Today, pollution and its effects in the basin are likely to be comparatively subtle

and largely invisible to the average observer. Despite the absence of gross water or air pollution, however, the public has become increasingly aware of and concerned about the degradation of Lake Champlain ecosystems by pollutants being transported into the basin by the atmosphere. Concern about mercury contamination of fish in Lake Champlain and the possible effects of acid rain on forests in the basin are but two examples of this awareness.

The Lake Champlain basin (Figure 1) is a region that is particularly exposed to air pollution in the eastern United States by virtue of its location "downwind" of many pollutant sources and its relatively large catchment area. The basin has a large (18:1) ratio of watershed to lake surface area, so the capture and processing of atmospheric pollutants by the terrestrial portions of the basin may be important in this region. Approximately 89% of the basin is forested or agricultural, with the lake itself representing only 5% of the total basin area (Table 1). This is in sharp contrast to the Great Lakes, for example, which themselves represent a large proportion of their watersheds. These characteristics and the relatively mountainous, high elevation terrain favor the capture and accumulation of air pollutants in the basin by processes including wet and dry deposition, foliar wash-off, leaf litter-fall, spring snow-melt and runoff, and stream transport, as well as direct deposition to surface waters.

Historical Development of Air Pollution Management in the Basin

Air pollution management and measurements in the Lake Champlain basin were initiated on a "routine" basis in the late 1960's and early 1970's, primarily in response to the 1963 Clean Air Act and its 1970 amendments. The federal 1963 legislation was motivated to a large extent by a growing public perception that air pollution was a serious "local" problem in many large urban and industrial areas. Pollution control efforts focused on the largest, most poorly controlled sources or source categories. In the Lake Champlain basin these included open burning at dumps, asphalt plants, wood burning boilers and a few large industrial sources. Early ambient measurements were conducted predominantly in urban areas or in the vicinity of large sources, with primitive methods including lead sulfation plates (for sulfur dioxide), folded rubber strips (for ozone) and dust-fall buckets (for particulate matter). The 1970 amendments required establishment of national ambient air quality standards for a number of common "criteria" pollutants (carbon monoxide, nitrogen dioxide, sulfur dioxide, particulate matter, hydrocarbons and photochemical oxidants). Improved measurement methods were developed, and routine monitoring was required to determine compliance with the ambient standards. Given the effectiveness of early control efforts at reducing the most obvious "smoking" sources, and the relatively small size of urban areas and industrial sources in the Champlain Basin, few exceedances of national standards were recorded, and the general public perception was of pristine air quality - rivaling the clarity of Lake Champlain's sparkling waters. During this period similar awareness and regulatory attention were given to surface waters in the U.S., although only recently has Lake Champlain water quality been recognized to be at risk.

The "discovery" of "acid rain" by the popular press in the late 1970's, dramatically changed the public perception of air pollution in the region. It introduced the concepts of "long-range transport", and "effects of air pollutants in other environmental media" - like

rainfall, surface waters and forest soils. Significant responses by the scientific community ensued, and the upland Adirondack and Green Mountain sections of the Champlain Basin became centers of major monitoring and research efforts to address questions of acid rain effects. The first air pollution monitoring effort to address basin-wide questions began in 1982 with the establishment of an acid rain monitoring station at the University of Vermont Proctor Maple Research Center in Underhill Center, Vermont. During the next decade a dozen more air quality monitoring studies were added at this site. Also during this period, acid rain monitoring and related air quality studies were upgraded at Whiteface, NY (where a MAP3S precipitation chemistry station had been established in 1976) and Sutton, Quebec (where the CHEF Program had been established in 1985). Added to these sites, air quality monitoring for criteria pollutants and assorted research measurements at several locations in Vermont, New York and Quebec, plus the network of National Weather Service (NWS) primary and cooperative stations were deployed to provide the basis for a modern monitoring infrastructure in the basin (Figure 2).

During the 1980s, new methods were developed and implemented to sample the chemistry of precipitation and cloud water [Galloway *et al.* 1978, Scherbatskoy and Bliss 1984]. Routine measurements of criteria pollutants were extended to rural areas, where surprisingly, the region's highest ozone concentrations were observed on the remote top of Whiteface Mountain. Accumulation of heavy metals was observed in forest soils and lake sediments in the "pristine" Adirondacks and Green Mountains [Friedland *et al.* 1984]. During this period, inter-governmental agreements were developed for more efficient management of shared air resources, and by the late 1980's, New York, Vermont and Quebec had each committed to reducing and maintaining emissions of acidifying sulfur emissions. In 1988, these three jurisdictions also signed a Memorandum of Understanding on environmental cooperation on the management of Lake Champlain. In concert with the rising awareness of the ecological interconnectedness among air, land and water resources, inter-disciplinary approaches were developed and applied to environmental measurement programs, leading to the formation in 1990 of new coordinating institutions such as the Lake Champlain Research Consortium and the Vermont Monitoring Cooperative.

The 1990 Clean Air Act Amendments included requirements for phased reductions in acidifying sulfur emissions from electric utility sources in areas upwind of the Lake Champlain basin (with similar emissions reduction commitments from Canadian sources). Also under the 1990 Amendments, New York and Vermont were included in the multi-state Ozone Transport Region - composed of 11 "Northeast Corridor" States from Virginia to Maine. Lake Champlain was specifically included, along with the Great Lakes and Chesapeake Bay, as one of the "Great Waters", for which additional monitoring and research was required to understand the extent, effects and causes of atmospheric deposition of hazardous air pollutants to these waters. In 1990, the Lake Champlain Special Designation Act provided a range of new inter-governmental management structures, and included direct support for needed monitoring and research programs. State-sponsored "air toxics" monitoring programs were initiated in the Vermont, New York and Quebec sections of the Lake Champlain Basin, and monitoring and research programs to assess concentrations, deposition, sources and ecological processing of airborne mercury were also established. Measurements of ultraviolet radiation, and additional monitoring and research of local meteorology and climate conditions were initiated, reflecting public and scientific concerns over emerging global air pollution issues.

Current Air Resource Issues in the Basin

Currently, media attention, public hearings and our experience suggest that public concern about air pollution issues in the Lake Champlain basin appears to be focused on mercury contamination of fish, global climate change, and acid rain, in that order of importance. Although not so widely perceived by the public, exposures to air toxics, fine particulates and tropospheric ozone are currently receiving scientific attention as significant contributors to environmental quality in the basin. As discussed further at the end of this report, public perception of immediate and obvious impacts on human health and economics often drives decisions about resource management and research priorities, sometimes independently of their scientific importance. Mercury contamination of waters in the basin captures public attention in this way. Questions about the effects of acid rain on the health of forest ecosystems in the basin have never been fully answered and recent findings of nutrient depletion [Likens *et al.* 1996], episodic acidification and nitrogen saturation [U.S. EPA 1995] in northeastern forests have stimulated renewed attention on additional acid rain controls. Although ozone concentrations have not exceeded federal standards in the basin in the past several years, there continues to be persistent exposures at levels of concern to forest ecologists. The recently revised, stricter ozone standards may increase public perception of this issue.

In light of this background, the purpose of this report is to discuss the recent history of air pollution monitoring in the Lake Champlain basin and to document the long term trends and current status of the major air pollutants affecting the lake and basin, including mercury, acid deposition, tropospheric ozone, air toxics and fine particulates. In addition, analysis of air transport patterns for these pollutants is used to show the probable geographic extent of the airshed of the basin.

AIR POLLUTANTS IN THE LAKE CHAMPLAIN BASIN

Acid Deposition

Acidic deposition in precipitation has been monitored at three sites in the basin since 1984 under the National Atmospheric Deposition Program National Trends Network (NADP/NTN) at the Vermont Monitoring Cooperative monitoring station in Underhill, VT (400 m elevation), at Whiteface Mountain, NY (620 m) and at Sutton, Quebec (244 m). Weekly data for the U.S. sites are available at the NADP Internet Web site [NADP 1998] including meta-data and wet-only precipitation amount, pH, and concentration and deposition of major cations and anions. Dry deposition of acidifying compounds has been monitored in the basin since 1992 at the VMC site in Underhill, VT and since 1984 at the Whiteface site. Using weekly filter-pack samplers, air concentrations of sulfur dioxide, sulfate aerosol, nitric acid vapor and nitrate aerosol are measured and the corresponding deposition of sulfur and nitrogen is calculated by inferential methods [Hicks *et al.* 1991].

Nationally, SO_2 emissions have been declining slowly since the early 1970s, with a sharp drop in 1994-95 in electric utility emissions due to compliance with Title IV of the Clean

Air Act Amendments. These trends are apparent in the SO_4 concentration in precipitation for the Lake Champlain basin as well as many other northeastern locations. Figure 3 shows pH and volume-weighted seasonally averaged concentrations for sulfate and nitrate in precipitation at Underhill and Whiteface. The long-term downward trend for sulfate and absence of trend for nitrate are obvious in these data. In addition, pH appears to be increasing only slightly, sulfate concentration is typically highest and nitrate the lowest in the summer period, and sulfate concentrations tend to be slightly higher at Whiteface, while nitrate concentrations are slightly higher at Underhill. During the summer seasons of both 1993 and 1994, sulfate concentration was noticeably elevated at both sites due to very high sulfate concentrations in June and August. Another interesting observation is that calcium concentration in precipitation at both sites has been declining at a rate of about 0.3% annually (data not shown). Reasons for this are obscure, but concern about the effect of this trend on forest ecosystem health has been expressed in other studies [Likens *et al.* 1996].

Figure 4 shows the concentration patterns of the compounds in the basin that dominate dry deposition of sulfur and nitrogen as gasses (SO_2 and HNO_3) and particulates (SO_4^{2-} and NO_3^-). Note that the concentration of reduced (SO_2) and oxidized (SO_4^{2-}) sulfur compounds alternates seasonally, with sulfur dioxide concentration being greatest in the winter months and sulfate being highest in the warmer months (as for precipitation). Elevated sulfate concentrations during summer 1994 are also apparent, suggesting a possible relationship between particulate and precipitation sulfur.

Annual wet and dry deposition rates of sulfur and nitrogen compounds at Underhill and Whiteface are shown in Table 2. Over this period, annual wet deposition rates at Underhill averaged 32% higher for nitrogen and 10% higher for sulfur than at Whiteface, suggesting possible reduced influence of nitrogen sources at Whiteface. These data indicate that annual deposition of sulfur and nitrogen in the basin occasionally exceeds the annual critical loading rate of approximately 8 kg S/ha and 8.4 kg N/ha, levels at which changes in vegetation composition (due to to nitrogen) or soil aluminum mobilization (due to sulfur) can be expected in this region [DeVries 1993].

The NOAA Air Resources Laboratory (ARL) measures daily precipitation chemistry at the Underhill, VT site as part of the AIRMoN Network [AIRMoN 1998]. This site began operation in 1992, shrtly after the termination of the Whiteface, NY MAP3S station, and is the only long-term daily monitoring station in New England. Daily measurements allow a much clearer evaluation of meteorological conditions than is possible with the weekly NADP samples. ARL also maintains meteorological data and data analysis service via its "READY"(Real-time Environmental Analysis and Display sYstem) Internet site [NOAA 1998]. Among other things, the READY site provides access to the HY-SPLIT (HYbrid Single Particle Lagrangian Integrated Trajectories) trajectory model [Draxler and Hess 1997]. The HY-Split trajectories can be calculated backward (or forward) in time to track the path of airmass motion for air arriving at (or leaving from) a specified receptor location. Trajectory Cluster Analysis [Stunder and Artz1996] is a powerful statistical technique which allows large numbers of trajectories to be grouped into a much smaller number of trajectory "clusters", for which trajectories have common spatial characteristics which are similar within each cluster and different from those in other clusters.

In a recent analysis of the Underhill site AIRMoN rain events for 1995, backward HY-SPLIT 36-hour trajectories were calculated for 88 of 99 precipitation events for which chemical analysis was available. For 11 cases trajectories could not be computed because

of missing meteorological data. This analysis resulted in 5 clusters (Figure 5) representing the general paths of air masses resulting in the precipitation events occurring at Underhill in 1995. Using the AIRMoN data, the chemical characteristics associated with the trajectories in each cluster can be examined (Table 3). According to this analysis, precipitation events associated with the Southwest Long cluster (cluster #1), accounted for about 24% of the total 1995 precipitation volume at Underhill, but contributed about a third of the total sulfate and hydrogen ion deposition, and over 40% of the nitrate ion deposition. Together, the three West-Southwest clusters (clusters # 1, 2 and 4) accounted for 69% of the precipitation events, 77% of the precipitation volume and more than 85% of the 1995 deposition of sulfate, nitrate and hydrogen ion. Although the northwest sector is typically the source of fair weather and clean air in this region, the concentration of sulfate and hydrogen ion in precipitation from this sector in 1995 was nearly as great as any other. Because this sector contributed only a small quantity of the total precipitation, the total wet deposition of pollutants from these storms was small (5%). This analysis underscores the fact that precipitation events arriving in the Champlain Basin are both frequent and polluted. Fortunately, future reductions in sulfur and nitrogen emissions from Midwestern electric utilities are anticipated from the 1990 Clean Air Act Amendments, and from EPA's recently proposed nitrogen oxide reductions based on recommendations from the Ozone Transport Assessment Group [OTAG 1998].

Ozone

Tropospheric ozone is a federally regulated air pollutant, harmful to all animal respiratory systems and plant life. It is routinely monitored at three locations in the Lake Champlain basin, at 394 m elevation in Underhill, VT, at 620 m and 1142 m at Whiteface Mt., NY, and at several elevations in Sutton, Quebec. Ozone is monitored by continuous photometric analysis and is reported as hourly average concentrations.

Ozone pollution is of concern for potential effects on human health, forest ecosystems and agricultural crops in the Lake Champlain basin. The health-based federal standard for ozone (formerly based on maximum 1-hour concentration of 125 parts per billion [ppb]) has recently been changed to protect against exposures at a lower threshold (85 ppb) over a longer averaging time (8 hours). Hourly data from Whiteface - which typically experiences among the highest concentrations in the Basin - indicate no exceedances of the 1-hour standard in the past five years (Figure 6, top panel), although this threshold is frequently approached, and a number of exceedances were recorded there in the late 1980's and early 1990's. The annual pattern of ozone concentration seen at Whiteface during the past five years is fairly representative of ozone behavior at the other monitoring sites in the basin. Analysis of historical data indicates that ozone concentrations in the basin would have been close to exceeding the new ozone standard on several occasions, and the new standard will be more stringent (more easily exceeded) than the 1-hour standard.

Forest health and agricultural scientists are more concerned with exposures exceeding 40 to 60 ppb, which occur quite frequently at all sites in the basin. As indicated in Figure 6 (top panel), regional ozone exposures clearly peak during the summer growing season and also exhibit distinct diurnal patterns and elevational gradients (Figure 6, bottom panel). At lower elevation sites like Bennington, VT (just south of the Lake Champlain basin), summer

ozone levels typically fall off dramatically at night, as ozone formation requires sunlight and ozone destruction by chemical reaction and physical deposition takes place predominantly at or near the earth's surface. At higher elevations, the nocturnal destruction is minimal, such that mountain forests in the basin are chronically exposed to higher ozone exposures than low elevation counterparts [Poirot 1993]

Fine Particles and Air Toxics

Particulate matter pollution is of current concern for its potential effects on human health and visibility, and for its contribution to the deposition of acidifying compounds and toxic metals and organic contaminants in the Basin. The federal standards for particulate matter, based on particles of less than 10 micrometer (μm) diameter (PM-10), are currently being revised to focus on smaller particles, less than 2.5 μm (PM-2.5). These smaller particles penetrate and reside at further depths of the respiratory tract and tend to be composed of inherently injurious substances, including acidic sulfates, and potentially toxic organic compounds and trace metals. Analysis of historical data suggests that ambient fine particle concentrations in the basin may be relatively close to the new PM-2.5 standard of an annual average not to exceed 15 micrograms per cubic meter ($\mu g/m^3$).

Fine particles and air toxics have been monitored in the basin at various urban and rural sites, including Burlington, Barre, Rutland and Underhill in Vermont, Glens Falls, Whiteface and Willsboro in New York, and Sutton, Quebec. Methods for collection of fine particles typically involve the use of a size-selective inlet to remove large particles and weekly or bi-monthly collection of a 24-hour air sample on a filter which is then subjected to various analysis by spectrometry, fluorescence or other techniques. Gaseous air toxics are typically sampled in canisters or absorbent cartridges with subsequent analysis by gas or liquid chromatography.

PM-2.5 data from 1993 in the Burlington, VT urban area and at two nearby, remote "background" sites at Whiteface Mtn., NY and Underhill, VT are reported in Figure 7. These data indicate that average background fine particle mass concentrations are about half the level of the proposed standard, with about half of the background concentration composed of sulfate compounds. As these sulfate particles result primarily from distant sources, their concentration does not increase at the urban Burlington site. Organic compounds and elemental carbon (soot) are substantially higher at the urban site (adding about 3 $\mu g/m^3$ to the background levels) indicating significant local source contributions for these fine particle species.

Local urban sources are also important (predominant) contributors to elevated local concentrations of several toxic gaseous organic compounds, including formaldehyde, benzene and 1,3-butadiene. As indicated in Figure 8, these compounds slightly exceed Vermont's health-based toxic standards at the remote background site in Underhill, and increase by a factor of 2 to 4 in nearby suburban and urban areas. The Vermont toxic standards for these 3 contaminants are based on an estimated cancer risk of 10^{-6} (1 cancer death per million population) such that an ambient concentration of 10 times greater than the standard would represent an estimated risk of 10^{-5}. Local emissions of these contaminants result primarily from mobile sources (gasoline and diesel vehicles and associated refueling), and a reduction in the concentration of these toxic compounds will

require some combination of cleaner (or alternative) transportation and/or major lifestyle changes for the basin's inhabitants.

Mercury

Mercury (Hg) contamination of fresh waters and their biota has become a widespread and serious problem in many parts of the world, including northeastern North America. Health advisories restricting consumption of some mercury contaminated fish in Lake Champlain and other waters in the basin and throughout the Northeast have focused public and scientific attention on this problem. There is concern that atmospheric sources of mercury are responsible for increasing Hg burdens in Lake Champlain [Scherbatskoy *et al.* 1997, Watzin 1992, Vasu and McCullough 1994], but mercury sources, transport mechanisms and accumulation are not yet well understood. There is growing evidence that forested ecosystems have an important and complex role in the transport of atmospheric Hg to aquatic systems [Bishop *et al.* 1995]. Recent studies have identified several processes affecting Hg movement in ecosystems, including organic complexation in soils and water, accumulation in plant tissues, foliar capture of atmospheric Hg, and a large watershed:lake area ratio [Schuster 1991, Rea *et al.* 1996, Swain *et al.* 1992]. The Lake Champlain basin is susceptible to mercury pollution because of its large proportion of forested lands where these processes occur and its proximity to relatively large Hg emissions in the region [NESCAUM 1998].

Atmospheric mercury has been studied at two locations in the basin: for precipitation, vapor and particulate forms of Hg since 1992 in Underhill, VT [Scherbatskoy *et al.* 1998] and for vapor and particulate forms in 1992-94 in Willsboro, NY [Olmez *et al.* 1996]. The Underhill site is the longest running continuous monitoring program for all three forms of atmospheric Hg in the world, and provides an important long-term and regional reference for other studies that have recently begun in other parts of the Northeast. The Underhill Hg research program is a collaborative effort between the Vermont Monitoring Cooperative and the University of Michigan Air Quality Laboratory. Precipitation for Hg and trace metal analysis is collected at this site on an event basis in a modified wet-only precipitation collector, and vapor and fine particulate (2.5 μm) air samples are collected for 24 hours on gold-coated sand vapor traps and glass-fiber filters, respectively. Sampling and analysis procedures are described in detail by Burke *et al.* [1995]. At Willsboro similar methods were used for Hg vapor and particulate sampling, using charcoal vapor traps and Teflon filters, respectively [Olmez *et al.* 1996].

The long-term patterns of Hg concentration and deposition in precipitation, based on the data of Keeler *et al.* [this volume], are shown in Figure 9. The magnitude and pattern of seasonal increase in Hg concentration during the summer is similar to that seen at other sites in the northern U.S. Because of greater concentration and precipitation in the summer months, Hg deposition in the basin is greatest during this period. Annual wet deposition of Hg at this site averages 87 mg/ha. Dry deposition of Hg vapor to the forest ecosystem is large during the warmer months, but because of large uncertainties about the re-emission of Hg from forests, it is difficult at this time to accurately fix the total wet plus dry net deposition. Recent analysis by Rea [1998] indicates that 50-120% of dry deposited Hg may be re-emitted to the atmosphere from forests. Nonetheless, according to Shanley *et al.* [this volume] about 92% of the measured wet and estimated dry Hg deposition is retained in the

terrestrial ecosystem (with some unknown amount returning to the atmosphere by re-emission), and only 8% of this deposition is transported out of the ecosystem in stream water.

Descriptive statistics for concentrations of precipitation, vapor and particulate forms of Hg at Underhill are presented in Table 4. These values are generally similar to those for other sites in the northern U.S. There do not appear to be any significant trends in concentration at this site, except perhaps for Hg vapor which has declined from around 2 ng/m^3 in 1993 to around 1.6 ng/m^3 recently (Figure 10). Comparable values for Hg vapor were measured by Olmez *et al.* [1996] at Willsboro, NY during 1992-1994, except for two periods (winters of 1992 and 1993) when concentrations were approximately twice as high as Underhill.

Meteorology and Ultraviolet Radiation

Two other important issues related to atmospheric resources in the Lake Champlain basin are global climate change and increasing solar ultraviolet radiation (UV-B). While these are both potentially important environmental factors in the Lake Champlain basin and elsewhere, our data and understanding of their effects are very limited. Meteorological monitoring in the basin has been enhanced in recent years by the addition of automated weather monitoring stations located at regional airports, and by research weather stations operated by the Vermont Monitoring Cooperative at Colchester Reef on Lake Champlain and at several locations on Mount Mansfield. Data from these stations, plus data from the National Weather Service cooperative weather stations throughout Vermont are available through the Vermont Monitoring Cooperative Data Library. In addition, an assessment of precipitation patterns in the Lake Champlain basin has been recently conducted by A.R.D. Inc. [B. Hegman, pers. comm. 1998) for use in watershed pollutant loading studies. These data are valuable for interpreting recent and historical climate and ecological processes.

Ultraviolet radiation has been monitored in the basin since 1993 at the Vermont Monitoring Cooperative's forest research site in Underhill, VT using a broad-band pyranometer (Yankee Environmental Systems UVB-1). In 1996, the Underhill site became part of the U.S. Department of Agriculture UV-B Monitoring Program, a climatological network of 25 sites nationwide using several broadband instruments. Information about and data from this network are available from the program's Web site [USDA 1998]. Because of the limited data for this site, no trend or effects information is available at this time, but the availability of these data for the basin is important for future environmental research.

THE LAKE CHAMPLAIN AIRSHED

While many pollutants affecting the Lake Champlain basin are of predominantly local origin (see Figures 7, 8), others are of more distant origin and subjected to long-range transport. Toxic metals, for example, are a current concern for their potential effects on human or ecological health in the basin. Certain metals (like mercury) in our ambient air and precipitation have significant local source contributions such as commercial incinerators, while others (nickel, arsenic, selenium) may result predominantly from more distant sources. It is valuable to identify source regions for these contaminants (as for acid deposition,

above), and to determine the extent of the airshed, the geographic region with significant sources potentially affecting the basin. Poirot *et al.* [this volume] examined the concept of the synoptic-scale "Lake Champlain airshed", employing 8 years of (12 per day) backward "air mass histories" to derive a long-term "probabalistic" definition of "areas most likely to be upwind" of the Lake Champlain basin. They found this probabilistic airshed varied in size and shape as a function of season, meteorological condition, and pollutant of concern. For high local concentrations of fine particle arsenic, the basin's air was likely to have previously resided to the northwest over a Canadian region of large primary smelter emissions. For high nickel concentrations, transport from East Coast metropolitan areas was indicated, while high selenium concentrations were associated with transport from the Ohio River Valley.

Similar methods are employed and shown here in Plate 1 to explore the "airshed" for 3 Lake Champlain basin monitoring sites on an "everyday" basis, and for high concentrations of ozone, PM-2.5 mass and PM-2.5 sulfur. The top row in Plate 1 displays "everyday upwind probability fields" for sites at Whiteface Mtn., NY, Underhill, VT and Lye Brook, VT (outside but near the basin boundary in southeastern Vermont). These are based on all available trajectories for these sites over an 8-year (1989-96) period. The shaded areas depict the smallest areas accounting for 20%, 40% and 60% of the trajectory "residence time hours" on the map. The patterns are similar for all 3 sites, and are generally oriented in a westerly (or WNW) direction from the receptors. The second row displays similar upwind probability fields, but in this case, only trajectories on days with high 8-hour ozone concentrations at the receptors were included. While there is a strong southwesterly orientation on high ozone days at all sites, note there is also a strong southerly influence (East Coast urban corridor) on the Lye Brook site. Lye Brook also experiences somewhat more southerly influence than the northern sites for fine particle mass and sulfur - which generally show a dominant southwesterly influence at all the sites. Similar patterns for fine particle mass and sulfur are not surprising, since half or more of the fine particle mass at these rural sites is sulfate). Sulfur in the fine particle phase can range from pure sulfuric acid to fully neutralized ammonium sulfate. All of these sulfate compounds efficiently absorb water from the atmosphere, which increases their light scattering efficiency and makes them the most important causes (accounting for 60-70%) of regional haze in the Champlain Basin. The results here for fine particle sulfur are quite consistent with the NOAA ARL cluster analysis results for Underhill precipitation sulfate (Figure 5). Hence, a common upwind source region is a major contributor to acid deposition, fine particle pollution, regional haze, and ozone pollution in the Basin, and is a key component of the Lake Champlain Airshed.

AIR RESOURCE ISSUES IN THE FUTURE

Perception, Policy and Management

While our understanding of air pollution, and of the priority air resource issues in the Lake Champlain basin is evolving and far from complete, much has been learned over the past 30 years since routine measurement programs were initiated. The management of environmental resources can be viewed as a policy response to public perception of environmental issues of concern. Environmental measurement, through routine monitoring

and more focused research efforts, can provide critical scientific guidance for effective management decisions by clarifying the nature, severity and causes of environmental problems. Over time, environmental measurements can lead to changes in public perception, and provide the means to assess environmental management decisions.

Specific issues facing air resource managers and the public in the Lake Champlain basin include acid deposition, mercury, fine particles, air toxics, greenhouse gasses, UV-B and climate change. There are numerous complex possible impacts of these stressors on human and ecosystem health, which may be caused by direct single-factor and multiple exposures, and by indirect processes. Effective responses to and management of these problems must be guided by good quality information. Access to data such as those presented in this report, however, faces several challenges. There have been several proposed cuts and actual lapses in funding for several monitoring programs described in this report, even during the period when air pollution levels were expected to respond to new regulations in the Clean Air Act. Only through continued monitoring of acid rain constituents were we able to see that indeed these regulations were effective and that additional controls were still needed. In addition, data access itself can be a problem for managers and policy-makers. In order to improve the relationship between air pollution science and management, easy and rapid access to monitoring data, is helpful. Finally, it is important to recognize that our perceptions of environmental problems change, and today's issues in the basin will evolve and new issues arise in the future. Good communication among stakeholders, scientists, managers and policy makers is necessary to ensure dynamic and accurate responses to perceived environmental problems. Thus, support for long-term monitoring, good data access, and integrated communications are critical components of effective air resource management in the Lake Champlain basin in the years ahead.

Air Resource Management Needs and Recommendations

Although we have good information about some aspects of air resource issues in the basin, there are a number of areas where improvements and additional data are needed. Broadly, there are three type of processes to pay attention to when addressing air resource issues in the basin. *Atmospheric exposure and deposition* of acidifying substances, toxic contaminants, and ozone are important atmospheric environmental stressors in the basin. *Ecosystem processes,* including element and energy cycling, population dynamics, and changing landscape structure, underlay the interactions between these stressors and ecosystems. Finally, *climate change processes* are probably having significant direct and indirect effects on the ecosystems of the basin and their responses to atmospheric stressors and other factors.

At a recent workshop of the Lake Champlain Research Consortium [McIntosh 1998], two broad areas of research and monitoring were identified to improve our understanding of air resource issues in the basin. These were to assess *source-receptor relationships* for atmospheric contaminants entering the basin, and to clarify the *transport mechanisms* for hazardous air pollutants within the basin. In addition, a number of strategic recommendations were made, including increased attention to air issues in the basin, under the clean air regulations of the state and the Clean Air Act Amendments (section 112m).

While many of the air pollutants and environmental stressors affecting the basin originate

outside its borders, there are important local sources of some contaminants such as mercury and some volatile organic compounds. A better inventory of these emissions and an analysis of their contribution to pollutant exposures in the basin are needed. This represents a significant gap in our knowledge and an impediment to effective management of air resources in the basin.

Transport and deposition of regional and local air pollutants are strongly affected by regional and local weather patterns. As seen in this paper, regional and synoptic analysis of meteorology and air pollution data allows assessment of transport patterns and some understanding of source-receptor relationships. Local-scale processes, however, are poorly understood, particularly as they affect deposition patterns and movement of locally generated pollutants. Most of the modeling of deposition has been conducted at coarse scale resolutions (>100 km), while meso-scale modeling (<10 km resolution) is needed to assess deposition processes and source-receptor relationships within the basin. The NOAA Air Resources Laboratory, as well as several other federal and university research groups, have the capability to model wind-fields and pollutant deposition at this finer scale. Transferring this technology to the Lake Champlain basin would benefit our understanding and management of air resource issues in the basin.

Although this report documents much useful information about air contaminants in the basin, there remain a number of concerns that need specific attention. To achieve effective air resource management in the basin in the foreseeable future, the following additional needs must be addressed. *1. Support long-term monitoring.* Only with continuous, high-quality monitoring at the appropriate frequency will we be able to provide reliable information capable of detecting patterns and trends in spatial and temporal environmental data. *2. Assess source-receptor relationships.* Further analysis of air transport and high resolution chemical deposition data is needed to better identify regional and local sources of air contaminants in the basin. *3. Conduct elemental speciation of fine particulates.* Although new regulations set standards for fine particulate (2.5 μm) matter, there is currently inadequate information on the concentration of toxic elements in this material to evaluate specific health risks and identify sources. *4. Clarify watershed transport and cycling processes.* There are still important gaps in our understanding of how forested and agricultural watersheds affect the movement of air contaminants in the basin, particularly with respect to the cycling of mercury and land use effects on nutrients and contaminants. *5. Assess environmental and ecological effects of climate change.* Although this is complex, collection of environmental data and development of ecological models are needed that help illuminate the direct, indirect, and interactive effects of climate change in the ecosystems of the basin.

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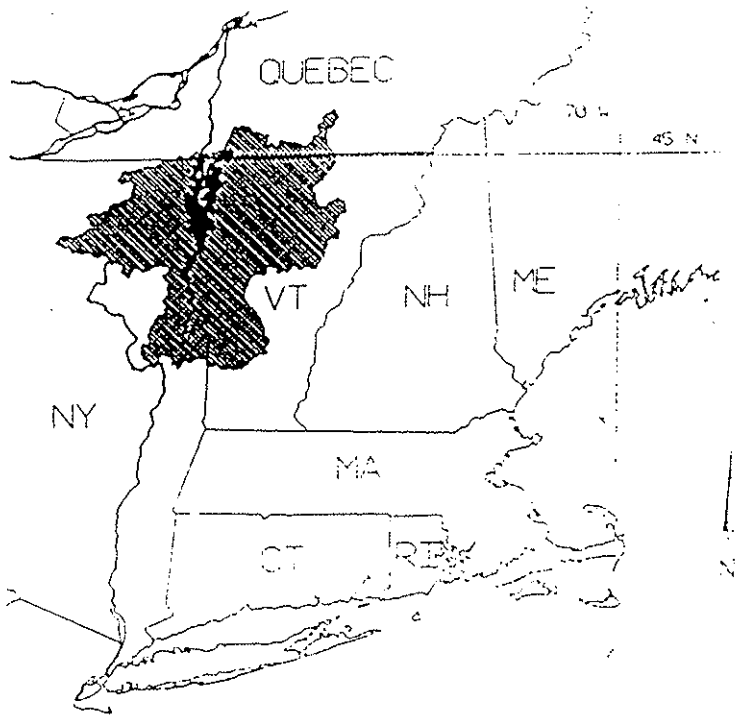

FIGURE 1 Map of the Lake Champlain Basin in the New England region

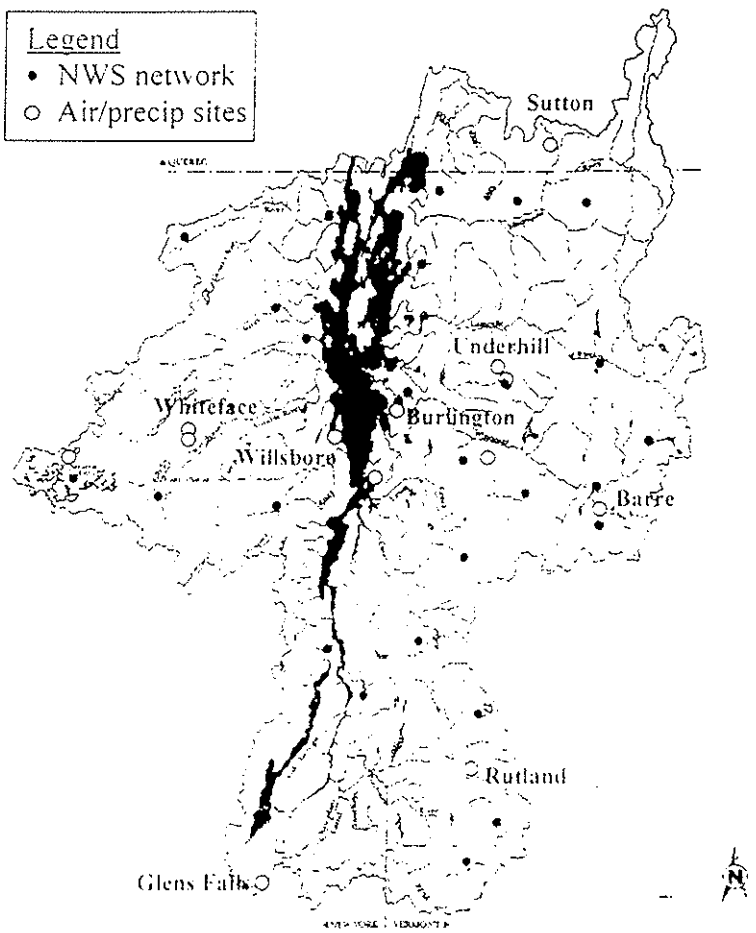

FIGURE 2. Map of the Lake Champlain basin showing locations of air and precipitation monitoring stations and weather stations.

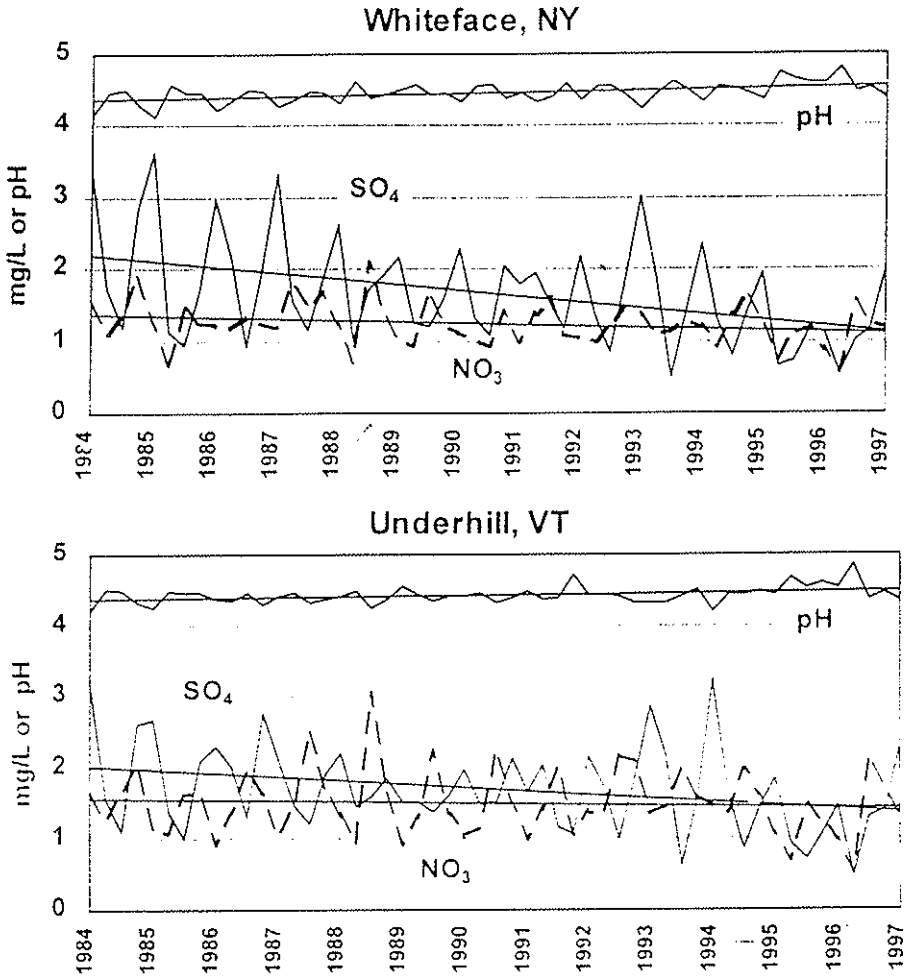

FIGURE 3. Trends in seasonal (quarterly) volume weighted average pH (upper line) and concentration of sulfate (solid line) and nitrate (dashed line) in precipitation in the Lake Champlain Basin at Whiteface Mountain, NY and Underhill, VT, from NADP weekly samples in 1984 - 1997, with linear regression trend lines.

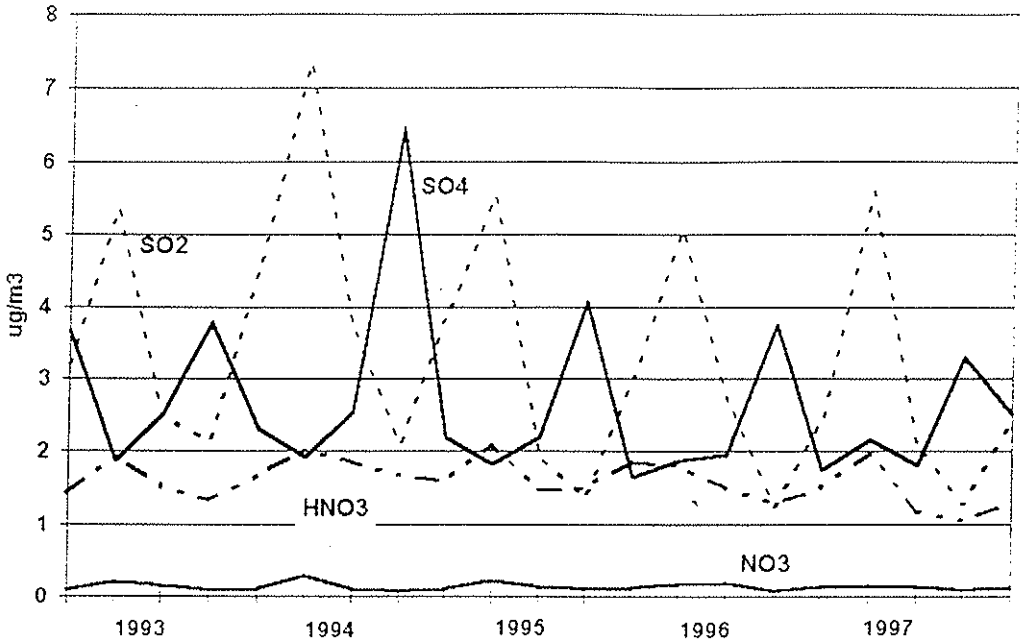

FIGURE 4. Seasonal (quarterly) average concentrations of ambient vapor and particulate sulfur and nitrogen compounds in the Lake Champlain basin at Underhill, VT, from NOAA AIRMoN weekly samples in 1992 - 1997.

TABLE 1. Area and percent of total basin area of major land use classes in the Lake Champlain basin, based on aerial photography in 1973 (Budd and Meals 1994).

Class:	Forest & Wetland	Agriculture	Urban	Surface Waters	Lake Champlain	Total Non-Lake	Total Basin
Hectares:	1,322,506	567,262	55,840	75,111	113,000	2,020,719	2,133,719
Percent:	62%	27%	3%	4%	5%	--	--

TABLE 2. Annual wet and dry deposition as kg/ha of total sulfur (SO_4^{2-} S and SO_2 S) and total nitrogen (NO_3^- N, NH_4^+ N and HNO_3 N) at Underhill, VT and Whiteface, NY.

Year	S. wet		S. dry		S. total		N. wet		N. dry		N. total	
	VT	NY	VT	NY	VT	NY	VT	NY	VT	NY	VT	NY
1985	7.34	7.34		2.03		9.37	6.30	5.21		1.69		6.90
1986	7.90	7.68		1.45		9.13	6.32	4.77		1.58		6.35
1987	7.04	7.24		1.46		8.70	6.76	5.62		1.50		7.12
1988	6.53	5.73		1.83		7.56	5.49	3.85		1.96		5.81
1989	6.15	7.09		1.94		9.03	6.85	5.77		2.29		8.06
1990	8.49	7.03		1.96		8.99	7.72	5.70		2.97		8.67
1991	6.52	6.24		2.10		8.34	5.94	4.53		3.55		8.08
1992	4.64	4.89		1.42		6.31	4.78	3.57		2.42		5.99
1993	7.87	7.49	2.05	1.44	9.92	8.93	7.64	5.18	1.82	2.55	9.46	7.73
1994	7.70	5.50	2.34	2.23	10.00	7.73	7.31	4.64	2.08	3.25	9.39	7.89
1995	5.27	4.00	1.53	1.00	6.81	5.00	5.70	4.43	1.75	2.28	7.45	6.71
1996	4.27	3.89	1.40	0.89	5.67	4.78	5.30	4.48	1.79	2.10	7.09	6.58
1997	5.79	4.86	1.32	1.41	7.11	6.27	5.79	4.60	1.31	3.45	7.10	8.05
Mean:	6.58	6.08	1.73	1.63	7.90	7.70	6.30	4.80	1.75	2.43	8.10	7.23

TABLE 3. Contributions of each trajectory cluster shown in Figure 5 to the precipitation chemistry at Underhill, VT in 1995.

Cluster #	1	2	3	4	5
Cluster Name	SW Long	W	NW	SW Short	NE
# of Trajectories per Cluster	32	17	12	20	7
Total Precipitation (cm)	12.2	9.2	2.4	17.9	9.8
VWM [a] SO_4 Concentration (μeq L)	31.9	29.3	31.3	20.0	10.5
% of Total SO_4 Deposition	33	23	5	30	9
VWM NO_3 Concentration (μeq L)	29.7	21.2	18.4	10.6	5.1
% of Total NO_3 Deposition	43	23	5	22	6
VWM H^+ Concentration (μeq/L)	44.3	32.7	38.0	24.6	13.4
% of Total H^+ Deposition	36	20	6	29	9

[a] Volume Weighted Mean

TABLE 4. Descriptive statistics for atmospheric Hg concentrations in Underhill, VT for the period December 1992 through December 1997.

	Mean	Std. Dev.	n	Minimum	Maximum	Median
Vapor Hg (ng/m^3) [a]	1.67	0.49	324	1.01	7.31	1.59
Particulate Hg (pg/m^3) [a]	9.24	6.32	356	0	42.0	7.79
Precipitation Hg (ng/L) [b]	8.23	8.48	553	1.15	89.8	8.03

[a] Corrected to Standard Temperature (25 C) and Pressure (1015.3 mb)

[b] Volume Weighted Mean

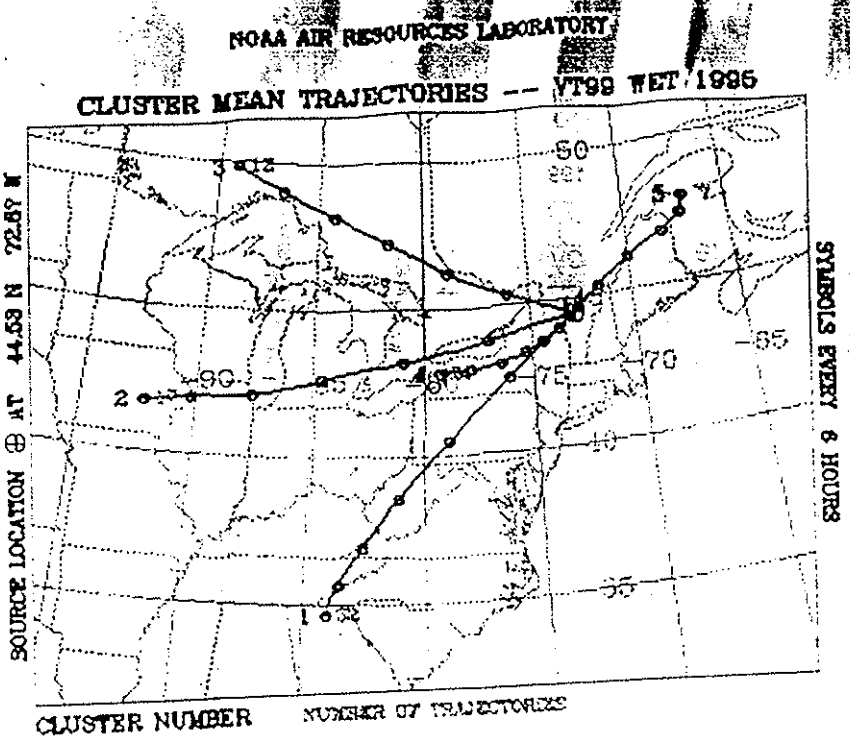

FIGURE 5. Mean trajectories of clusters of air trajectories for precipitation events arriving at Underhill, VT during 1995. Dots along trajectories represent 6-hour increments. Numbers in larger type at the end of each trajectory is the trajectory cluster label (see Table 3), and numbers in smaller type is the number of trajectories in the cluster.

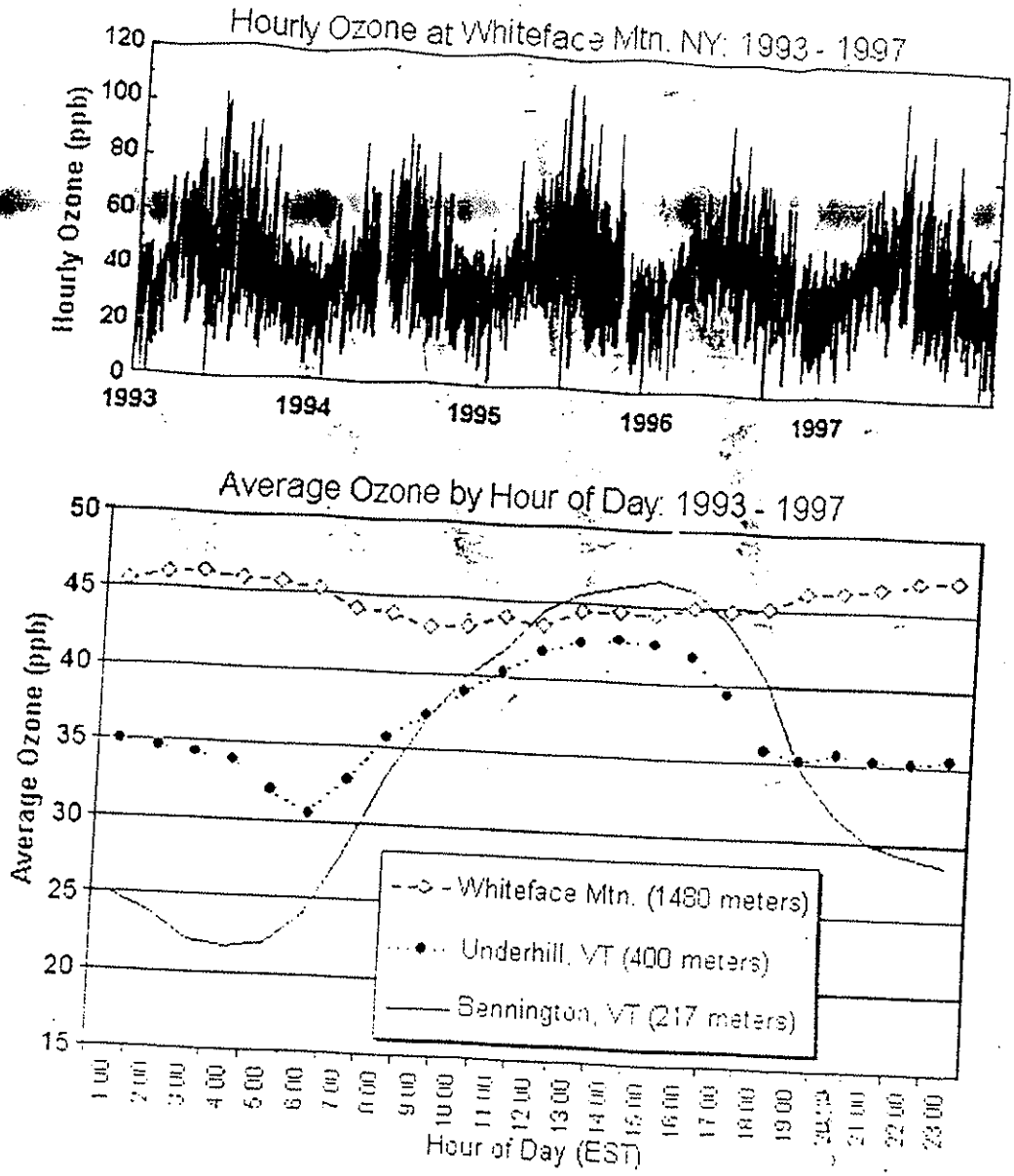

FIGURE 6. Trend in tropospheric ozone concentration at Whiteface Mountain, NY (top) and diurnal average hourly ozone concentration patterns for three sites in and near the Lake Champlain basin (bottom), based on hourly average ozone concentrations monitored in 1993 - 1997.

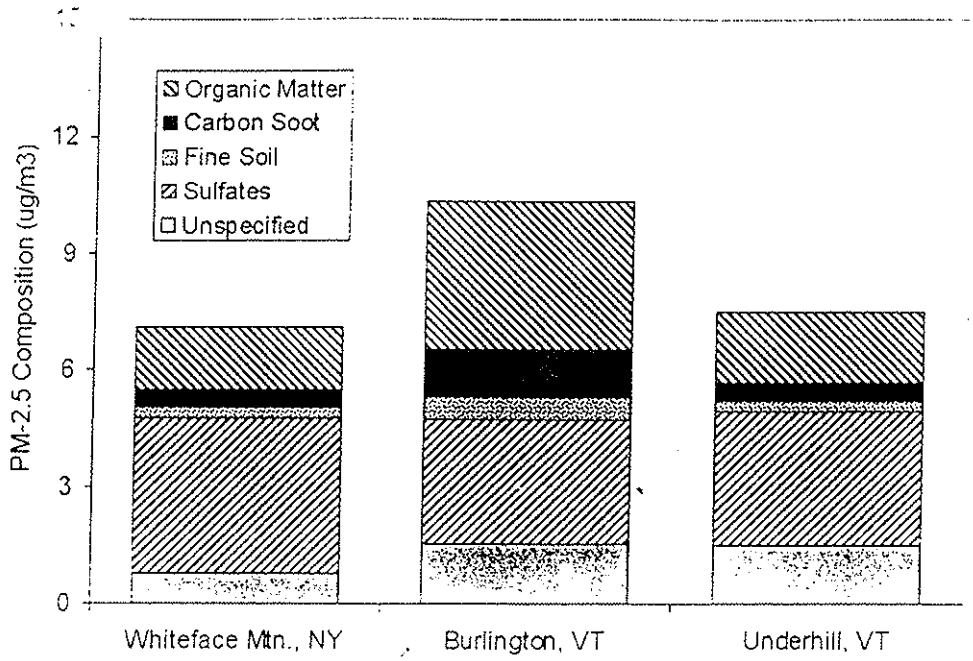

FIGURE 7. Fine particle (2.5 μm) composition and concentration at three sites in the Lake Champlain basin, calculated from every-sixth-day 24-hour samples in 1993.

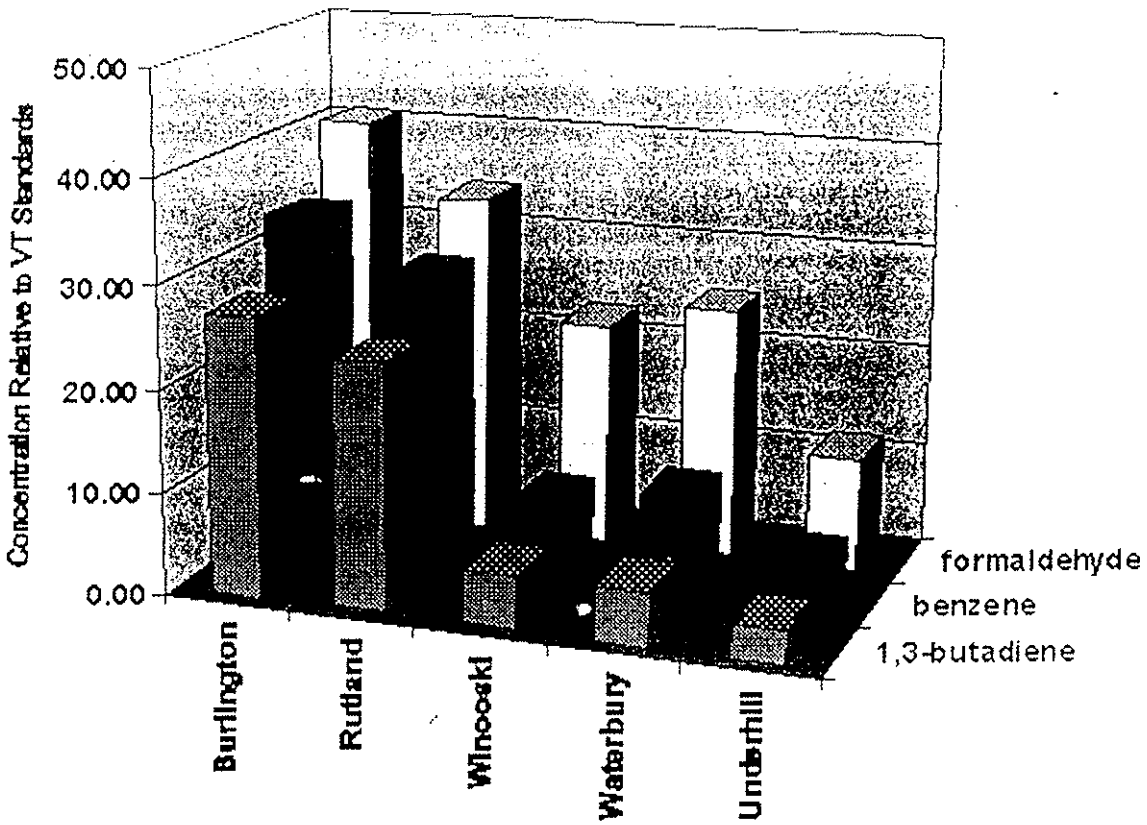

FIGURE 8. Relative concentrations of three toxic air contaminants at five sites in the Lake Champlain basin, calculated from 24-hour samples collected every 12 days in 1994

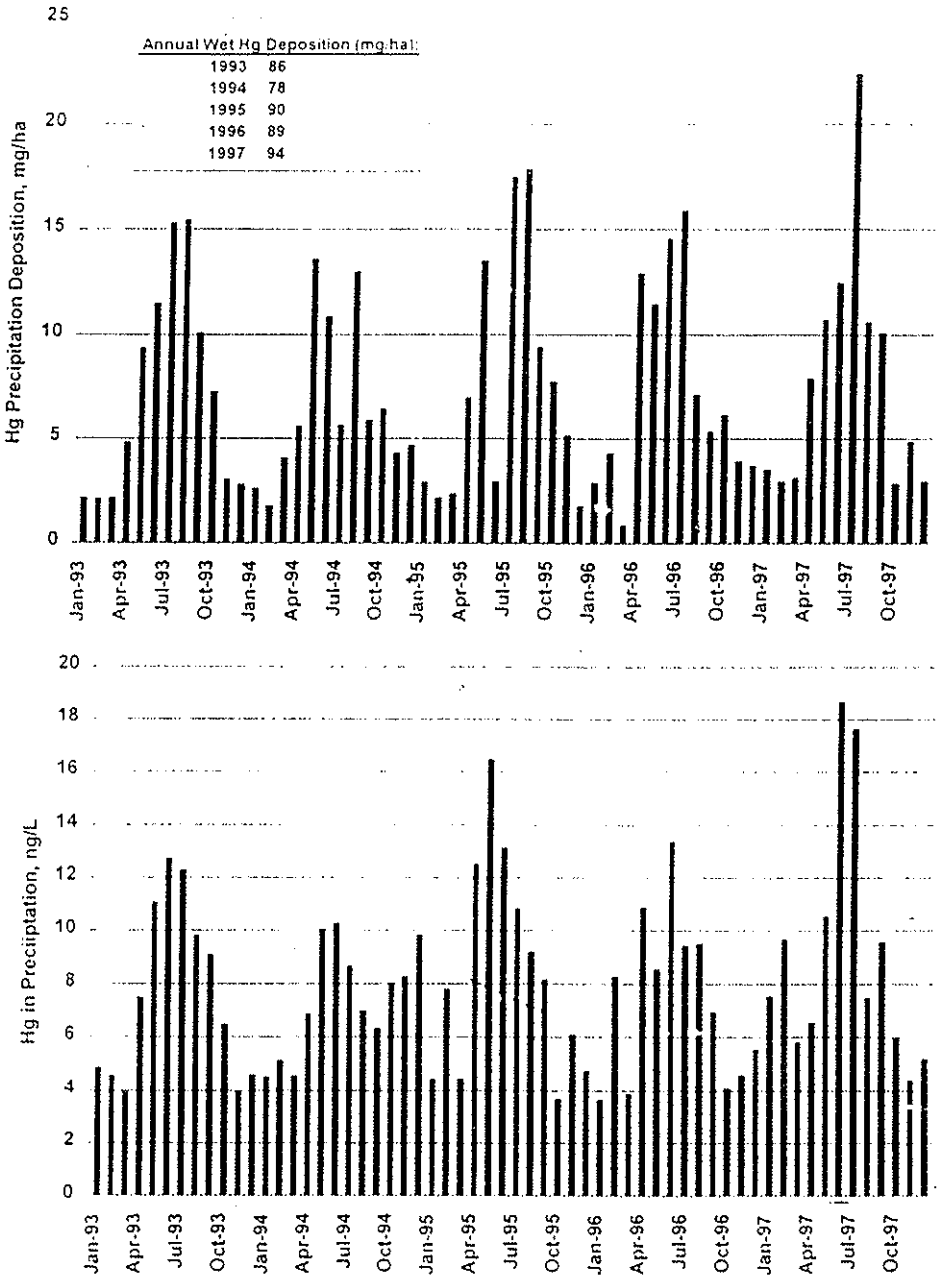

FIGURE 9. Monthly mercury deposition (top) and volume weighted monthly mean mercury in precipitation at Underhill, VT in the Lake Champlain basin, based on event samples in 1993 - 1997.

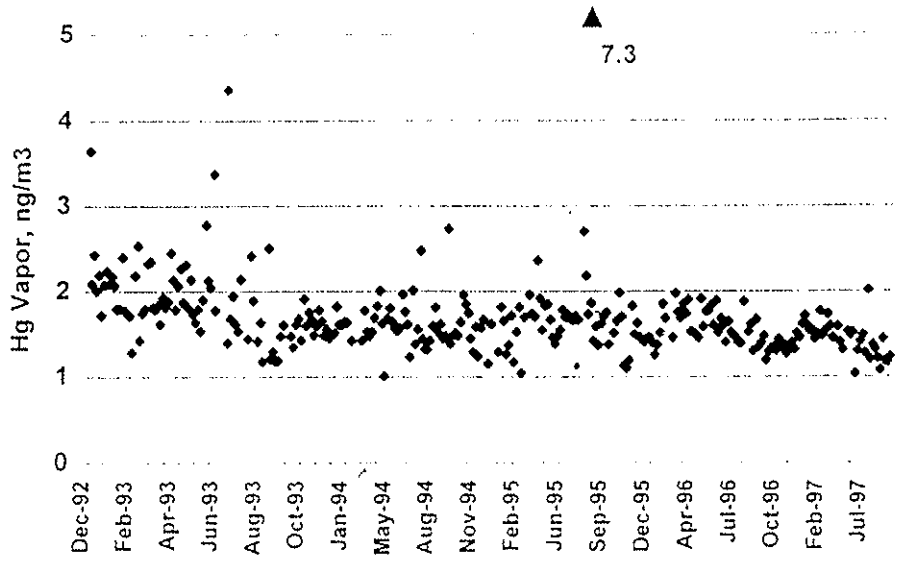

FIGURE 10. Ambient mercury vapor concentration at Underhill, VT in the Lake Champlain basin, based on every-sixth-day 24-hour samples in 1993 - 1997.

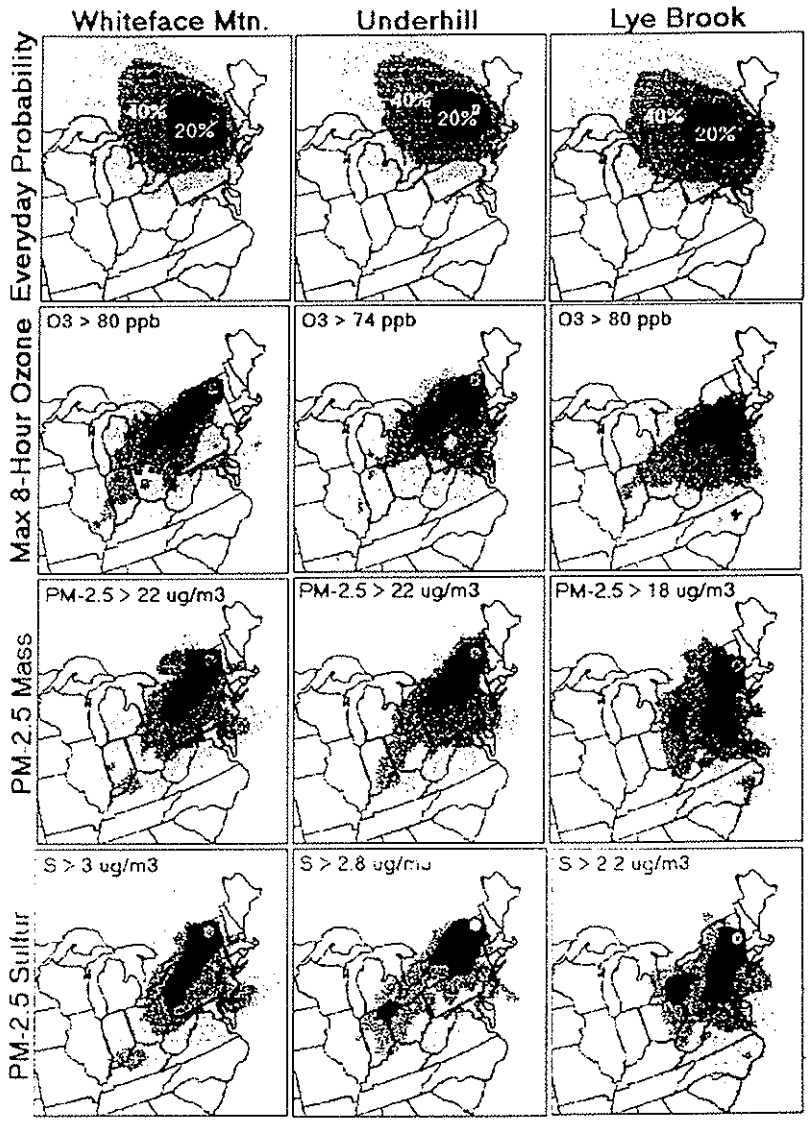

PLATE 1. Probability regions for the Lake Champlain airshed for three sites in or near the basin based on meteorological data for 1989 - 1996 for days of elevated sulfur, fine particle, and ozone, and for all days ("everyday"). For each receptor site, the shading shows the area of probability that the air mass originated in the region with a probability of 20% (black), 40% (dark gray), or 60% (light gray).

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