Air Trajectory Pollution Climatology for the Lake Champlain Basin

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ABSTRACT

Detailed air mass history calculations from the CAPITA Monte Carlo model are being employed to investigate long-term, synoptic-scale meteorological conditions associated with ambient air quality and deposition measurements at several remote monitoring sites in (or near) the Lake Champlain Basin. The CAPITA model, with meteorological input from the National Climatic Center's Nested Grid Model (NGM) was run to calculate 5-day backward air mass histories, every 2 hours, for the years 1989 through 1996 for receptor sites surrounding the Lake Champlain watershed in Sutton, Que; Whiteface Mtn., NY; Underhill, VT and Lye Brook, VT. Preliminary results provide a probabilistic depiction of the Lake Champlain "airshed" (areas most likely to be upwind). This airshed has no fixed geographical boundaries, but varies with time (season, year, etc.), upwind meteorological conditions (pressure, wind speed, precipitation, etc.) and resultant pollutant concentrations. Over the long-term (8-year) period of record, areas which are persistently upwind prior to high concentrations or deposition of specific air pollutants at these background Champlain Basin sites are interpreted as predominant source regions for these pollutants.

INTRODUCTION

Lake Champlain's watershed is relatively undeveloped (89% forest and agricultural), large relative to the Lake's surface area (by a factor of 18:1), and is clearly defined by fixed geographical boundaries (mountain ridges). Environmental contaminants discharged to ground or surface waters anywhere within this large, rural watershed can eventually result in downstream consequences, including impacts in the Lake itself. In a similar way, contaminants discharged into the Lake Champlain "airshed" can result in downwind exposure to, or deposition of these contaminants to the Lake's surface or to upstream areas throughout the watershed. If airshed is taken to mean a geographic region which encompasses all emissions that occasionally have some (any) influence at a specified receptor area, then the airshed for any receptor may well extend to hemispheric or global scales. However, the likelihood or probability of impact from an upwind source is greatest near the receptor and decreases with distance, as modified by prevailing wind direction and other upwind meteorological conditions. For individual pollutants, the relative strength and spatial distribution of emissions sources can also modify the size and shape of the "effective" airshed. For certain short-lived pollutants like benzene, emitted primarily by motor vehicle-related activities, local urban centers like Burlington and Platsburgh may represent the predominant source areas for the Basin. Given the small sizes of these local urban areas and the relative absence of large industrial air pollution sources in the Lake Champlain watershed, the airshed for many longer-lived pollutants may extend hundreds or even thousands of kilometers beyond the watershed boundaries. Given the spatial and temporal variations of emission sources and meteorological conditions, the concept of airshed does not lend itself to a

fixed spatial definition. However, by combining multi-year sets of regional-scale meteorological data and local ambient pollution monitoring data a long-term or "climatological" description of airshed can be presented in "probabilistic" terms. We might ask questions like - which areas were most likely to be upwind of the Champlain basin: over the past several years, during the Winter, during precipitation events, or during pollution episodes of sulfate, ozone or arsenic, etc.?

METHODS

A backward air trajectory estimates the path of airmass motion prior to arrival at a specified "receptor" location at a specified arrival time. The authors have previously reported on ensemble backward air trajectory techniques - referred to as "residence-time analysis" - to explore predominant source regions of regional haze, particulate matter, trace elements and ozone for various receptor locations and time periods [Poirot and Wishinski, 1986, 1998; Wishinski and Poirot, 1986, 1998]. The general approach [after Ashbaugh, 1983] involves calculating large numbers of backward trajectories from a receptor site where concurrent ambient air monitoring data are available. A grid is used to track the multiple trajectory locations, and the gridded trajectories are sorted and/or aggregated as a function of the resultant ambient concentration at the receptor. The automated calculation of multiple trajectories for many arrival times requires a computerized trajectory model (such as the NOAA HY-SPLIT model [Draxler, 1992] or the CAPITA Monte Carlo model [Patterson et al., 1981; Schichtel and Husar, 1996]) and a multi-day, 3-dimentional meteorological database (such as from NGM [Rolph, 1996] or RAMS [McQueen et al., 1997]).

In a currently ongoing analysis for the US Forest Service, the authors are developing an "Air Mass History Pollution Climatology for Northeastern Forests and Parks" [Poirot et al., 1998]. The objective is to compare long-term, upwind meteorological characteristics with resultant air pollutant characteristics for a number of rural, regionally representative ambient monitoring sites in the Northeastern US and Eastern Canada. The 9 selected ambient monitoring sites displayed in Figure 1, generally include (at least) measurements of ozone, precipitation quantity and chemistry, and aerosol concentration and chemistry covering much (or all) of the 8-year period 1989 through 1996 - a period for which archived meteorological data suitable for calculation of airmass histories are also available.

The term "airmass history" as applied here includes the spatial information provided in a backward trajectory, but also incorporates other meteorological information along the trajectory path. For example, during the trajectory calculation, information on trajectory height, pressure, atmospheric mixing height, temperature, humidity, precipitation, wind speed, etc. can be calculated and retained for each location along the trajectory. The airmass histories for this study are calculated by the CAPITA Monte Carlo model and NGM meteorological fields. The Nested Grid Model (NGM) is routinely run by the National Meteorological Center to support meteorological forecasts on a variety of grid scales, including global. Input includes: radiosonde, satellite observations of temperature and cloud motion, and surface observations. A subset of the NGM data covering most of North America has been routinely archived since 1989 by the NOAA Air Resource Laboratory [Rolph, 1992]. These archived NGM data include estimates of about 20 meteorological variables with a 2-hour time step, horizontal grid resolution of about 180 km, and 10 vertical (sigma) layers ranging from about 150 m to 7000 m above the surface terrain. The routine archival of NGM data was discontinued in early 1997 (replaced by more sophisticated ETA model results). Hence the period 1989-1996 represents an ideal period for long-term analysis based on consistent methodologies.

The CAPITA Monte Carlo model acts upon the Eulerian NGM data to calculate a Lagrangian

airmass history, running (in this case) backward in time from a fixed receptor location and arrival time. A current version of the CAPITA model [Schichtel and Husar, 1995, 1996] is highly automated for PC operation, with options to run sequentially starting every 2 hours, retaining any or all of the NGM meteorological variables, at airmass positions every 2 hours backward in time for up to 5 days, for multiple receptor locations.

Several of the ambient monitoring sites in Figure 1 are "composite sites" - including measurements from 2 or more "nearby" locations. For 3 of the sites (Sutton, Lye Brook and Mt. Washington), ambient measurements are conducted at substantially different elevations, and 2 sets of air mass histories were calculated for different starting elevations - for a total of 12 sets of air mass history starting locations. The CAPITA model incorporates an estimate of vertical mixing by releasing 10 particles and allowing the vertical position of each to move randomly within the mixed layer - such that for each release time, 10 separate air mass histories are calculated for each starting location. In addition to the lat./long. (map) position of trajectory endpoints, various other aspects of the air mass histories are also retained - including trajectory pressure height, precipitation, mixing height, relative humidity, specific humidity, temperature, velocity, and age. So for each air mass history for each particle, values of about 12 meteorological variables are calculated and retained every 2 hours for 5 days backward in time. For each of 12 starting locations, 120 air mass histories are calculated each day for 8 years, such that a total of about 3 billion meteorological data points are calculated and retained. Example illustrations of some of the air-mass history information retained for individual release times for the Acadia, NP site are displayed in Figure 2. Note the influence of vertical mixing on the widely dispersed trajectories of particles arriving at the receptor on 8/4/95 at 8 AM, in contrast to the similar (horizontal and vertical) pathways traversed by all particles arriving on 8/1/95 at 10 AM. Note also the presence of precipitation along all trajectory paths just before arrival at Acadia on 8/4/95, and along several other locations further back along the particle pathways.

The individual air mass history information is tracked on a grid of 1,440 80x80 km squares, displayed with an example day's airmass history trajectories in Figure 3. To reduce data volume, and to facilitate direct comparison with ambient pollutant data (much of which is only available on a 24 hour basis), the air mass history data for each receptor site are converted to a 24 hour basis by aggregating all (120) air mass history results for each calendar day for each grid square. This aggregation assumes that the air mass history information for each of 10 particles released at each of 12 arrival times during a 24-hour period is equally representative of the upwind meteorological characteristics associated with pollutant concentrations measured during that 24-hour period. Rather than a single trajectory line, this approach identifies an upwind area (and associated meteorological parameters) as an upwind region of potential influence for each 24-hour measurement period. Examples of daily aggregate air mass history results (for the PANJNY site on 7/3/94 and 7/4/94) are displayed in Figure 4. In these examples, the spatial characteristics of all 120 airmass histories for each day are displayed as " residence-time probabilities", with 25% of the day's upwind residence-time hours included in each separately shaded area. Note that on 7/3/94, the upwind residence-time probability is relatively tightly constrained along a narrow upwind path - reflecting strong, persistent flow from the northwest of the receptor. On the following day, the upwind probabilities are more broadly distributed over nearby areas to the east and northeast of the receptor.

The Lye Brook/Bennington, VT site lies just beyond the southern end of the Lake Champlain watershed, while Sutton, Quebec (north), Whiteface Mtn., NY (west), Underhill/ Mt. Mansfield, VT (east) are located within, and distributed throughout, the Champlain Basin. The long-term airmass history results from these 4 sites provide a basis for exploring the complex concept of the "Lake Champlain Airshed".

RESULTS

Figure 5 displays "Everyday" upwind probability fields for the Sutton and Lye Brook sites. These are based on the daily airmass history data for every available day, 1989 through 1996. The tracking grid is employed to aggregate the "residence-time" hours that each airmass has spent over each grid square in its path en route to the specified receptor. The hours are summed for all dates, arrival times and grid squares, and the probability for an individual square is expressed as the fraction of total hours for that square divided by the total hours in all squares. Each separately shaded area in Figure 5 contains 20% of the residence-time probability for the total (approximately 10 million square kilometer) tracking grid, with isopleths bounding the smallest areas accounting for 20%, 40%, 60% and 80% of the total probability within the grid. The least probable 20% of upwind locations are included in the large, lightly shaded, outermost area - which represents about 70% of the total grid area, but represents a very low upwind probability for any specific location.

Similar everyday upwind probability fields result for the Whiteface Mtn., and Underhill sites. In Figure 6, the everyday probability fields from all 4 Champlain Basin sites are combined (averaged by upwind grid square) - to provide a best estimate of the Lake Champlain "airshed" in probabilistic terms. A cutoff at the 80% probability contour is arbitrary - the "total airshed" extends well beyond this (and beyond our tracking grid too), but the upwind probability for individual locations beyond the 80 % contour is relatively small. For example, an individual 80x80 km grid square on the outer edge of the 80% contour line is upwind of the Basin only 0.07% of the time - which is coincidentally equal to the percentage that would result if the probabilities were distributed randomly (equally) across all 1440 grid squares (1/1440 = 0.07%). The 80% contour also approximately corresponds to a transport time of 2 to 3 days (appropriate for secondary, reactive pollutants like ozone), and contains a number of large source regions which are upwind on a relatively frequent basis. These source regions, and certain aspects of their emissions characteristics are also displayed in Figure 6. The emissions of sulfur dioxide (SO_2), nitrogen oxide (NO_X) and volatile organic compounds (VOC) are taken from the 1986 NAPAP inventory, and are illustrative of predominant source types. They are plotted here with symbols sized relative to emission rates - with the exception that the SO_2 symbols are reduced by 50% for clarity (also, SO_2 emissions from both US and Canadian sources have been substantially reduced since 1985). The major source regions which are relatively close to, and likely to be periodically upwind of the Lake Champlain Basin include:

- Montreal & Boston Urban Areas (VOC and NO_X from area and mobile sources),

- East Coast Urban Corridor (VOC and NO_X from area and mobile sources),

- Ohio River Valley (SO_2 and NO_X from large utilities and industrial sources),

- Lower Great Lakes (mixed pollutants from urban & industrial sources),

- Canadian Smelters (SO_2 from a small number of large industrial sources).

These generalized source regions include a variety of other sources and pollutant emissions (trace elements, for example). As they are represent large emissions densities, are frequently upwind, and nearly surround the (low emissions density) Champlain basin, the air quality and deposition in the

Basin may be influenced by one or more of these source regions on a day to day and long-term basis. As they are located in many different directions from the Champlain Basin, and are characterized by different mixes of predominant source types, it should be possible to discern their impacts in the Basin through combinations of the airmass history and ambient pollutant data. If, for example, emissions from within the Champlain Basin and nearby Montreal and Boston are considered part of local "New England" sources, then the above mentioned 5 source regions are approximately equivalent to the 5 regional elemental "fingerprints" identified by Rahn and Lowenthal [1984, 1985] in aerosol samples at Underhill, VT during the early 1980's.

Seasonal differences in the upwind probability fields, from aggregation of the winter (DJF) and summer (JJA) quarters for the 4 Champlain Basin sites, are displayed in Figure 7. The airshed has a stronger northwesterly orientation in the winter, while areas to the southwest and south are relatively more likely to be upwind in the summer months.

Several different features of the Champlain airshed are displayed in Figure 8. In this case, the average pressure (left) and wind speed (right) are calculated as a function of prior upwind location for airmasses arriving at Underhill, VT. This plotting methodology differs from Figures 5-7 in that the frequency with which locations are upwind is not considered. Rather, an average value is calculated for each upwind grid square for all aimass histories which have passed over that square en route to the receptor. If the air passed over square X, then the average pressure (or speed) was Y. The pressure (representing the average pressure for all airmasses (particles) at each location upwind of Underhill) relates directly to the airmass height (above mean sea level) by the approximate relation:

$$\text{Height (meters above MSL)} = 10 \times [1{,}016 - \text{Pressure (mb)}]$$

Thus airmasses approaching the Champlain Basin from the northwest tend to be subsiding, having previously resided at higher altitude, whereas air near the Basin in all directions and for some distance from the south/southeast of the Basin tends to have been at relatively low altitude. This pattern represents an unfortunate, coincidental correspondence to the general nature of the emissions sources in these upwind regions. Emissions within the Basin and at relatively nearby Montreal, Boston and East Coast urban areas tend to be released primarily from ground-level sources; Midwestern utility stacks are moderate to tall, and the Canadian smelter stacks are among the tallest in North America.

Average wind speed for areas upwind of Underhill is displayed on the right side of Figure 8. Lowest wind speeds are typically associated with areas immediately surrounding the Basin and the south; while highest wind for long distances to speeds (on average) are typically associated (altitude), there is an with areas to the west. As with the pressure unfortunate coincidence with nearby urban areas (with the predominant source regions. Air approaching the Basin from large (stagnated) over these numbers of small, broadly distributed, ground level sources) tends to have moved slowly areas and along the relatively short transport route to the Basin. removal with the slow Opportunities for pollutant transport speeds are offset by the relatively more distant short transport distance. Transport speeds from the Midwestern and Smelter offsetting the regions (characterized by smaller numbers of larger sources) tend to be faster, potential for pollutant dispersion and deposition over the relatively long transport route to the Basin.

A strong seasonal variation wind speed is indicated in the Figure 9 comparison of average August (left) and December (right) wind speed as a function of upwind location. Upwind summer wind speeds are generally lower than winter, especially over large sections of the Eastern US - south of the lower Great Lakes. This is especially important for the build-up of secondary pollutants (formed in

the atmosphere) like sulfates and ozone. This secondary formation process is enhanced by temperature and solar radiation, and requires time - all of which are more abundant during the summer. Midwestern SO_2 emissions (leading to secondary sulfate formation) are highest in summer (air conditioning demands). Gasoline-related evaporative VOC emissions from urban areas and NOx emissions from utility boilers (two important precursors to secondary summer ozone formation) are also greatest in summer over (East Coast and Midwestern) source regions with greatest summer stagnation. During the winter, the relatively lowest wind speeds are over areas to the south and north of the Basin. The large nearby population centers in these (winter stagnating) directions experience a considerable seasonal increase in (oil furnace and woodstove) emissions associated with residential space heating. Smelter emissions tend to exhibit relatively little seasonal variation and are located in areas which tend to experience relatively high windspeeds in all seasons. This is not conducive to buildup and transport of secondary sulfate species from the northwest (and smelter emissions are relatively unimportant for ozone formation), but strong, persistent winds do represent an efficient mechanism for efficient delivery of primary emissions (emitted directly to the atmosphere) such as trace metals with minimal dispersion/deposition en route from a small number of tall stacks. Thus, the Lake Champlain airshed not only includes a variety of large source regions which are frequently upwind, but is also characterized by natural climatological features which seem ideally suited to maximizing the potential impact from these different upwind regions.

For three of the Lake Champlain Basin air monitoring sites - Whiteface, Underhill and Lye Brook - relatively long-term records of 24-hour fine particle (< 2.5 micron) concentration and composition (including trace elements) are available during the 1989-1996 period of available airmass histories, through the IMPROVE [Eldred et al., 1988] and NESCAUM [Flocchini et al., 1990] monitoring networks. The Lye Brook IMPROVE site collects fine particle samples 2 days/week, started operation in September, 1991 and is still operating (total 450 samples during 1989-96 period). The Whiteface and Underhill NESCAUM sites commenced operation in September, 1988, collecting 3 samples/week. The Whiteface NESCAUM site was discontinued in November, 1993 (total 676 samples) and the Underhill NESCAUM site was discontinued in May, 1995 (total 815 samples). Elemental analysis for both IMPROVE and NESCAUM samples has been conducted by Proton-Induced X-ray Analysis (PIXE) and X-Ray Fluorescence (XRF) at the Crocker Nuclear Laboratory, U. California at Davis [Eldred et al., 1988].

Figure 10 displays the upwind probabilities for airmasses which have resulted in "high" concentrations of the trace elements - selenium, nickel, arsenic and manganese - at the Lake Champlain sites. Each of these plots may be thought of as a probabilistic airshed, comparable to Figures 5-7, but constrained to the condition of high concentration of the individual pollutants. The "high" concentrations selected here are above a specified threshold or cut-point, for which we attempted to chose round numbers which represented approximately similar sample sizes and distribution percentiles across the 3 sites and 4 pollutants. In this case, the high subsets include 20 to 40 of the highest daily concentrations representing the highest 3 to 5% of the sample days for each pollutant at each site. Concentrations at or above these threshold levels might be expected to occur on roughly 10 to 20 days per year.

The upwind probability plots for high selenium in the top row of Figure 10 show a clear southwesterly orientation at all 3 receptor sites, pointing directly toward Midwestern (coal-burning) sources along the Ohio River Valley. While selenium has not been identified as a contaminant of concern in Lake Champlain sediments or at ambient aerosol concentrations in the Basin, it does appear to be an excellent tracer for influence from coal combustion [Rahn and Lowenthal, 1984], which in turn, is a predominant source of sulfate pollution in the Basin [Rahn and Lowenthal, 1984,

1985; Poirot and Wishinski, 1986; Wishinski and Poirot, 1986]. Aerosol sulfate compounds contribute about half of the regional fine particle mass concentrations and 60% of the regional haze in the Basin [Poirot et al, 1992], while sulfate deposition is a predominant cause of acidification in upland areas in the Basin [Scherbatskoy et al., 1998]. Coal combustion is also an important contributing source for deposition of other assorted trace metals, including mercury, of concern in the Champlain Basin [Scherbatskoy et al., 1998].

The upwind probability plots for high nickel in the second row of Figure 10 show a strong southerly orientation at all 3 sites, pointing directly toward the East Coast urban corridor. Atmospheric emissions of nickel are dominated by oil combustion - both residual oil (a common utility and industrial fuel in the East Coast urban corridor) and distillate oil (commonly used for residential and commercial space heating in the northeast corridor). While ambient air concentrations of nickel at Champlain Basin sites are relatively low compared to larger eastern urban areas, and are not currently considered to pose a direct health threat in the Basin, they do appear to be a useful tracer for influence in the Basin from larger urban centers to the south. Elevated nickel concentrations have also been noted in Lake Champlain sediments in Outer Malletts Bay [McIntosh and Watzin, 1998], although the causes of these sediment concentrations are unclear, and may have inconsequential impacts from atmospheric sources.

The upwind probability plots for high arsenic in the third row of Figure 10 have a strong northwesterly orientation at all 3 sites, pointing directly toward the Canadian smelter region. Locations of several large Canadian smelters are also identified in these high arsenic plots, with the Noranda smelter - identified as a green dot - appearing to be the most likely contributor. Previous "elemental fingerprint" analysis of aerosol samples at Underhill, VT during the early 1980's [Rahn and Lowenthal, 1984, 1985]apportioned approximately equal fractions (40%) of the ambient arsenic to Canadian smelters and Midwestern (coal burning) sources. While there is no indication in Figure 10 of a Midwestern influence, the apparent discrepancy with these earlier studies may be due in part to the focus here on only the very highest arsenic levels. Rahn and Lowenthal's analysis apportioned all the arsenic (including days with low and moderate concentrations) and was also based on analytical methods (neutron activation) with lower detection limits for Arsenic than the PIXE and XRF methods employed in the IMPROVE and NESCAUM networks. In any event, high arsenic levels in the current data set appear to be excellent tracers for influence in the Basin from the Canadian smelter region. Elevated levels of arsenic have been observed in sediments of outer Malletts Bay [McIntosh and Watzin, 1998], although the origin is unclear, and may have a minimal contribution from atmospheric sources. Ambient air arsenic concentrations in the Basin have declined markedly since 1989 (perhaps in association with acid rain-related improvements to smelter emissions control systems). Current concentrations remain of some concern, as they remain close to the level of Vermont's Hazardous Ambient Air Standards (0.23 ng/m^3 - intended to prevent a cancer risk of 10^{-6}) even at rural background sites.

The fine particle manganese concentrations used to determine "high" concentrations in the upwind probability plots in the bottom row of Figure 10 were reduced slightly (by about 10% on average) to remove the fraction of manganese assumed to result from natural soils. This adjustment was based on concurrent measurements of fine particle silicon, and an assumed Si:Mn ratio in soil of 277:1 (based on crustal composition estimates from Mason [1966]). The upwind probability plots for Mn are less directionally distinct than for the other 3 trace elements, and are less similar between 3 receptor sites. The 20% isopleths of highest probability are more uniformly distributed around each receptor. Lye Brook and Whiteface indicate areas of relatively high probability extending west along the US Canadian border. Whiteface also indicates an area of high probability to the north. For Underhill,

where peak Mn concentrations are higher than for the other 2 sites, the highest upwind probability is predominantly to the north. Extraordinarily high manganese levels have been observed in sediments in outer Malletts Bay, and have been associated with acute biological toxicity in sediment pore water tests [McIntosh and Watzin, 1998]. However the origin or cause of these high concentrations is unclear, and may be predominantly related to changes in redox chemistry related to periodically anaerobic conditions in deeper sections of the Bay. Ambient air Mn concentrations in the Basin are well below levels of concern from a human health perspective. However, potentially phytotoxic levels of Mn have been observed in Balsam Fir foliage on Roundtop Mtn. in Sutton, Quebec, where the patterns of foliar and soil Mn concentration suggest that foliar uptake from atmospheric exposures may be an important factor [Lin et al., 1995].

Atmospheric sources of Mn are not entirely clear. Methylcyclopentadienyl manganese tricarbonyl (MMT) was developed in the 1950s as an octane-enhancing fuel additive in leaded gasoline, but was banned by US EPA for use in unleaded gasoline in 1978. A similar ban was not imposed on MMT additive in Canada, where it has been used in unleaded gasoline since 1976. Wallace and Slonecker [1997] noted historical statistical associations between fine particle Mn and Pb at US monitoring sites that suggested leaded gasoline as a predominant source of airborne Mn prior to the elimination of leaded gasoline in the early 1990s. They also noted substantially higher Mn concentrations in more recent fine particle measurements throughout Canada compared to measurements at similar sites in the US - which they attributed to the influence of the MMT additive [Wallace and Slonecker, 1997]. The spatial patterns of upwind probabilities for high Mn concentrations at the Champlain Basin sites - showing highest probabilities to the north (Montreal) and west (along the US/Canada border) are consistent with an MMT source. However, manganese is also emitted by various industrial processes, including steel production (prevalent in the lower Great Lakes region on both sides of the border). A large manganese alloy production plant in Beauharmois, Quebec (about 20 miles southwest of Montreal) ceased operations in 1991, but may well have contributed to the northerly influence evident at the Whiteface and Underhill sites. Wood combustion has also been identified as an important source of manganese emissions in a recent US national toxics emission inventory [US EPA, 1997]. Additional analysis of the ambient data and airmass histories before and after the 1991 Beauharmois plant closing may shed additional light on these potential source influences. Similar analysis of future data may also prove illuminating, as a recent court ruling in the US [US Court of Appeals, 1995] has essentially allowed the use of MMT in US unleaded gasoline; while more recent Canadian legislation prohibiting the importation or intra-provincial sale of MMT is likely to lead to substantial reduction of MMT use in Canada.

CONCLUSIONS

The above results represent preliminary examples from an ongoing investigation of airmass histories and associated pollutant concentrations in the Northeastern US. Several of the selected data sets are directly relevant to exploration of regional-scale atmospheric impacts in the lake Champlain watershed. The Lake Champlain airshed is an illusive concept, which can't be defined in precise geographical terms, but which can be approximately described in probabilistic terms. A number of relatively large air pollution source regions, including nearby urban centers to the north and east, the East Coast urban corridor to the south, Ohio River Valley to the southwest, urban/industrial areas to the west, and Canadian smelters to the northwest are all periodically upwind of the Lake Champlain watershed on a relatively frequent basis, and within a transport time of 1 to 3 days. The potential impacts from these upwind source areas is in several cases enhanced by natural meteorological characteristics of airmasses passing over these source regions en route to the Champlain Basin. Airmass history analysis of selected trace elements suggests that distinctly different upwind source

regions are associated with high concentrations of selenium, nickel, arsenic and manganese in the Champlain Basin. Several of these trace elements exhibit (perhaps coincidentally) elevated concentrations in Lake Champlain sediments, and/or represent ambient air exposures of concern from a human health or ecological health perspective.

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Figure 5. Everyday Upwind Probabilities for Lye Brook, VT and Sutton Que.

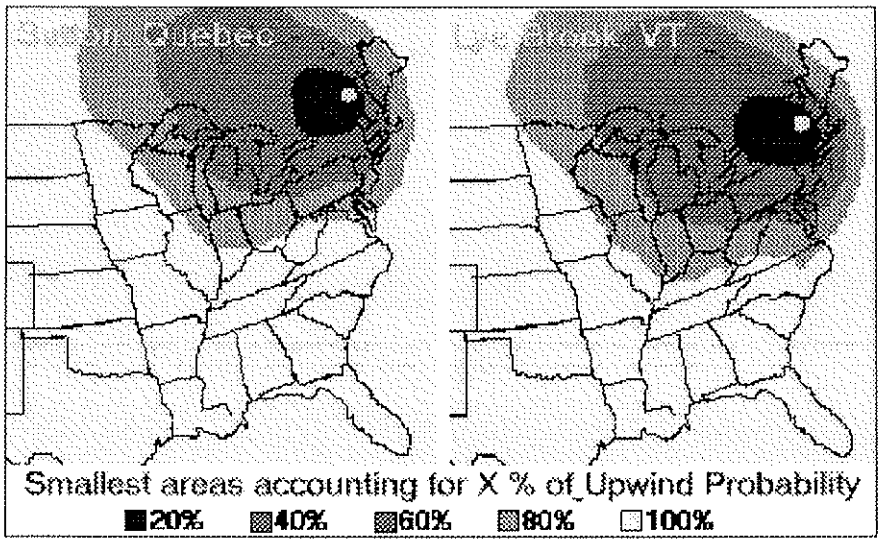


Figure 6. Probabilistic Lake Champlain Airshed (left) and "Nearby" Upwind Emissions Regions (Relative Magnitude of SO2, VOC and NOX Emissions (right) from 1985 NAPAP Inventory)

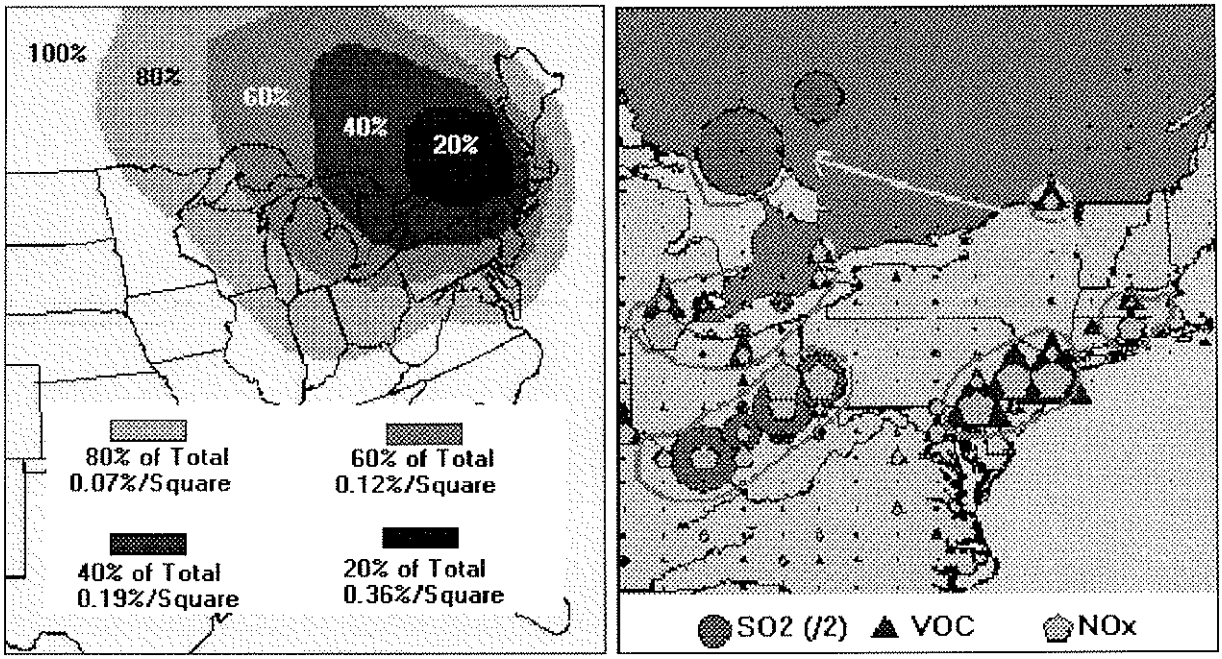


Figure 7. Seasonal Variation in Probabilistic Lake Champlain Airshed

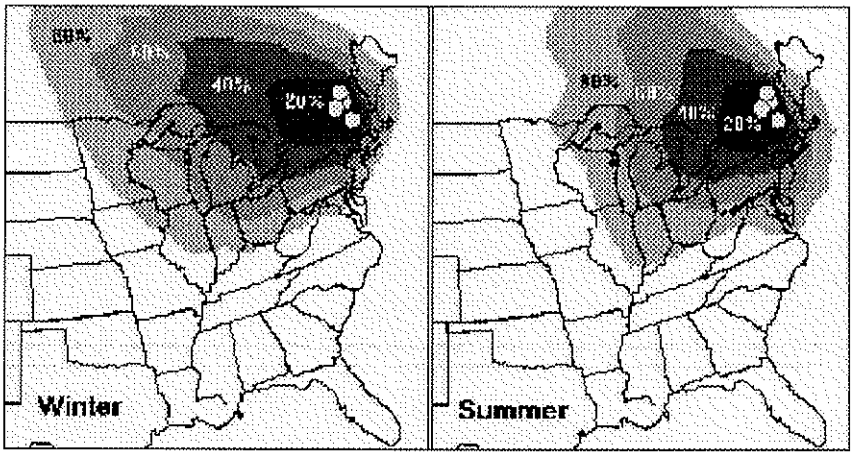


Figure 8. Average Atmospheric Pressure and Windspeed Upwind of Underhill, VT

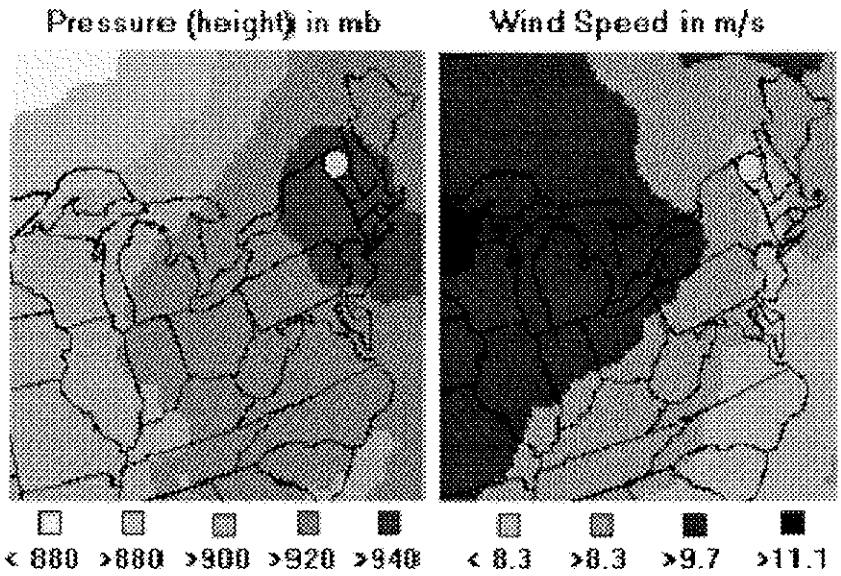


Figure 9. Monthly Average Windspeed (m/s) Upwind of Underhill, VT

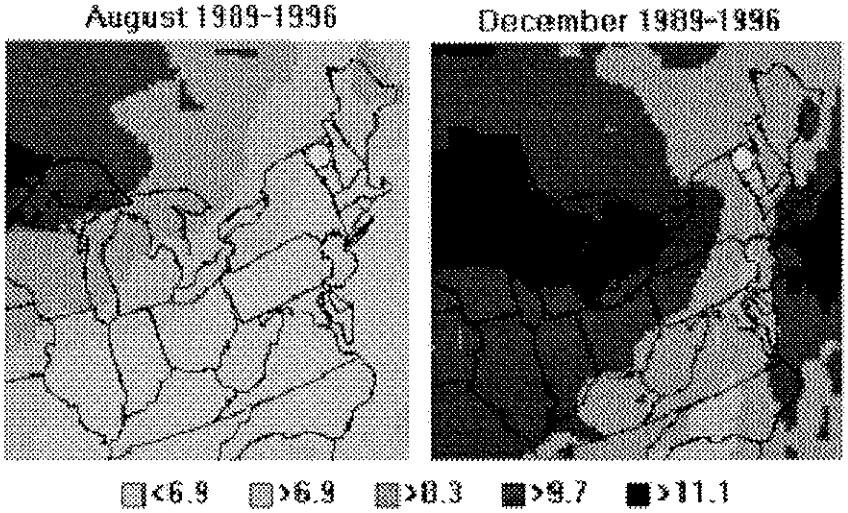
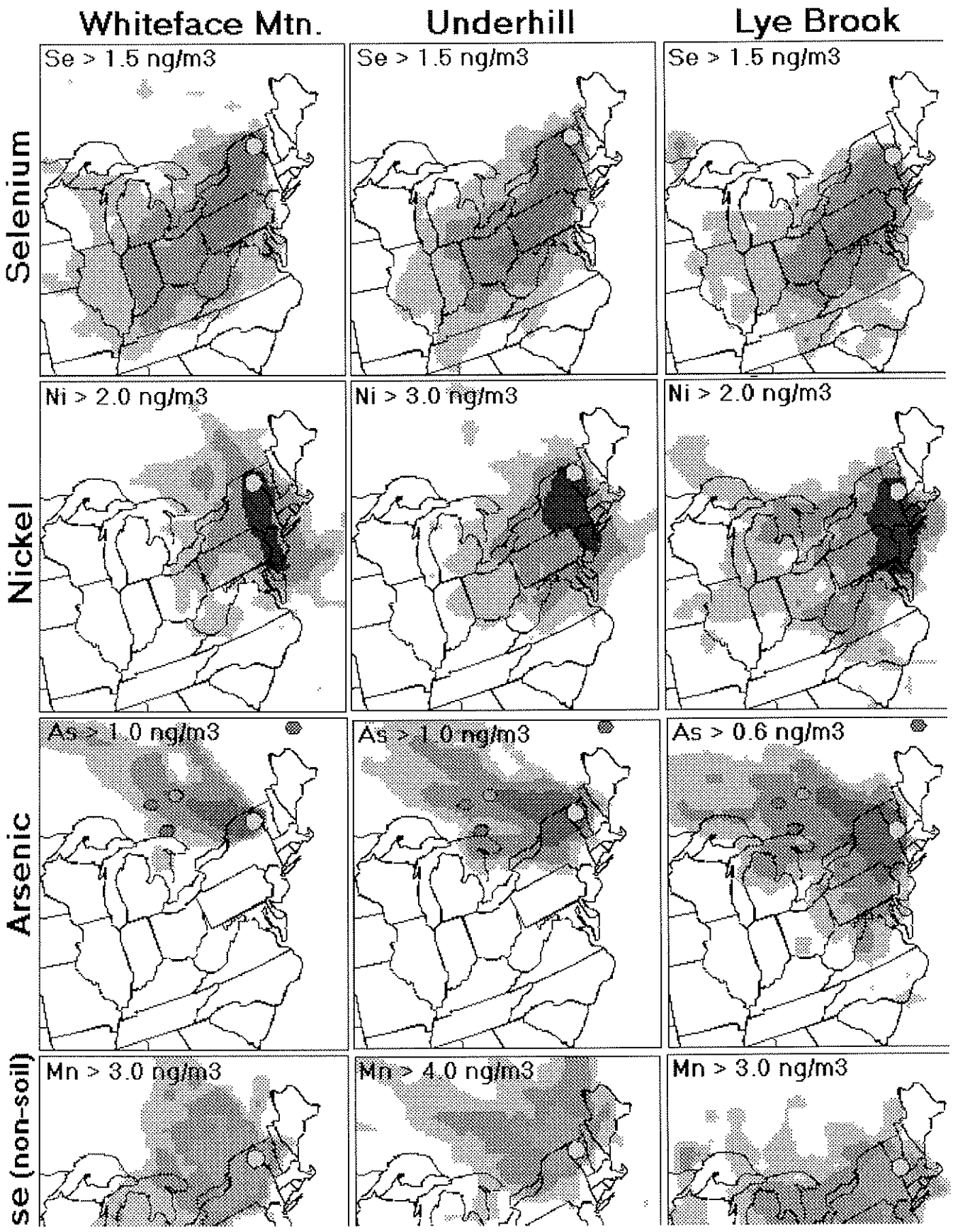
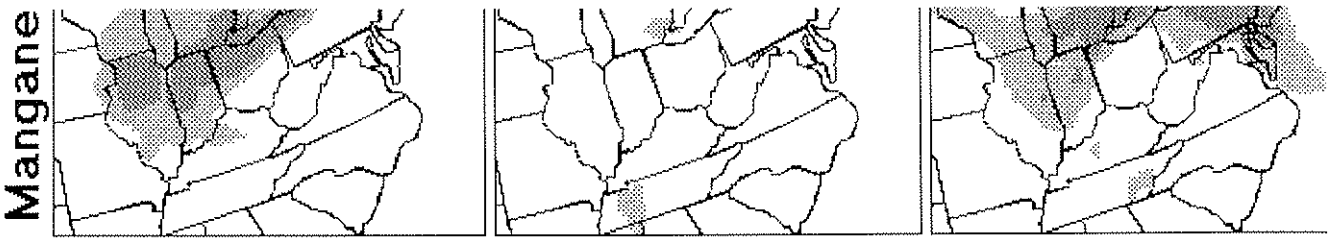


Figure 10. Upwind Probabilities for High Aerosol Se, Ni, As and Mn at 3 Champlain Basin Sites





Shaded Areas Show 20%, 40% and 60% of Upwind Probability on Highest Concentration Day