

Appendix A: Application of Trajectory Analysis Methods to Sulfate Source Attribution Studies in the Northeast U.S.

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ABSTRACT

Back trajectories have been calculated (8 per day) for the five-year period including 2000 through 2004 using the HYSPLIT modeling system for 13 Eastern sites including 10 rural locations near Class 1 areas subject to EPA's Regional Haze Rule and 7 urban location which have annual average $PM_{2.5}$ concentrations above or near the National Ambient Air Quality Standard (NAAQS). The back trajectories have been clustered based on 3-dimensional similarity to identify the predominant meteorological pathways influencing each site. Trajectories have also been associated with the nearest temporal value of 24-hr average concentration of $PM_{2.5}$ and IMPROVE sulfate values measured at or near each site. Individual trajectories and clustered probability fields are utilized in a variety of ways to apportion geographic contribution to meteorological transport of sulfate. Results are compared with quantitative methods of apportioning state-specific contributions to observed sulfate and serve as an independent check on contribution assessments developed through these alternative techniques.

Appendix A: Application of Trajectory Analysis Methods to Sulfate Source Attribution Studies in the Northeast U.S.

A.1. Introduction

The 1999 Regional Haze Rule (RHR) contains requirements for a site-specific pollution apportionment as part of each mandatory Federal Class I area's long-term emissions management strategy. A variety of techniques have been explored for conducting such pollution apportionments, but tagged chemical transport modeling is one of the few techniques which provide quantitative assessments of individual state or regional contributions to ambient concentrations. Given the importance of accurate pollution apportionment assessments, it is highly desirable to have several independent techniques provide confirmation of transport model results.

Traditional trajectory analyses that associate an ambient measurement of air quality with the geographical region upwind prior to the observation are limited in that they demonstrate the relationship between ambient air quality and the integrated path along the length of a back trajectory. It is difficult to distinguish the contribution of a specific point along a single back trajectory from the contribution of other points along that path. Large numbers of back trajectories have been used in a variety of ways to try to "triangulate" by taking advantage of the variation in meteorology and paths that an ensemble of back trajectories offers.¹⁻⁵ Combining results from multiple receptor sites offers a more robust method of triangulation and can yield very specific source regions associated with unique chemical signatures available with source apportionment techniques. These methods – which include ensemble trajectory analysis and factor analysis methods – all select based on chemical signatures. That is, they identify geographic regions on the basis of a unique chemical characteristic, such as high levels of an observed pollutant or high levels of a specific combination of pollutants.

Moody et al.,⁶ following methods of Dorling,⁷ have applied the Patterns in Atmospheric Transport History (PATH) clustering algorithm, to large numbers of back trajectories in order to group trajectories by three-dimensional similarity. Calculation of average pollution levels corresponding to the members of a cluster of back trajectories of similar three-dimensional structure provides a robust technique of associating air pollutants with typical meteorological pathways,^{8,9} but remains limited in its ability to distinguish individual points along an atmospheric pathway defined by a cluster of back trajectories. The cluster methods provide a way of identifying geographic regions based primarily on meteorological – rather than chemical – characteristics. The meteorologically selected regions can then be analyzed with respect to their chemical associations.

The definition of an individual cluster of back trajectories in PATH analysis is dependent on a subjective choice of the "Radius of Proximity." This threshold defines the limiting difference between the three-dimensional coordinates of two back trajectories and determines if they are in the same cluster or different clusters. Selection of a smaller radius of proximity, in effect, will split clusters into component sub-clusters. Thus in the limiting case (radius of proximity = 0) the analysis reverts to a traditional trajectory

analysis with each trajectory representing its own cluster. In this sense, PATH analysis offers a trade-off between uncertainty and fine scale structure of a trajectory analysis. By defining a radius of proximity and back-trajectory length, 10,000+ back trajectories for each site over the 5 year period can be represented by approximately 10-20 clusters.

Using these trajectory and cluster data bases, as well as the associated receptor-based pollution data, we have carried out analyses using the incremental probability method, and two PATH-derived techniques method for attributing pollution transport to geographic areas.

These qualitative methods support and enhance the conclusions derived through alternative methods (independent of back-trajectories) of associating emissions with downwind air quality impacts (i.e. the use of chemical transport models, or *source* modeling, rather than the *receptor* based approaches used in trajectory analysis). Here we use our large database of back trajectories and corresponding air pollution measurements to develop multiple metrics related to Wishinski and Poirot's "incremental probability"¹⁰ which is reflective of the increase in probability – relative to the everyday probability – of a geographic region being associated with a predominant meteorological pathway for sulfate transport (as opposed to a source region itself). The results are used to bolster the conceptual model of visibility impairment at Class I sites presented in Chapter 2.

A.2. Trajectory Approaches

The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model^{11,12} was used to calculate back trajectories for 13 sites in the Northeast and Eastern U.S. The locations correspond to Class I areas subject to the RHR as well as several sites where potential nonattainment issues with the PM_{2.5} NAAQS warranted analysis. Results are presented primarily for three rural sites which geographically span the MANE-VU class I domain: Acadia National Park, Maine, Lye Brook Wilderness Area, Vermont, and Brigantine Wilderness Area, New Jersey.

Back trajectories were calculated eight times per day for starting heights of 200, 500 and 1000m above ground level using meteorological wind fields for the five year period 2000-2004. NOAA ARL archives analyzed meteorological products for use with the HYSPLIT model including the Eta Data Assimilation System (EDAS) wind fields, which cover North America with an 80 km spatial resolution and are based on 3-hourly variational analyses.¹³ These back-trajectories -- and the underlying wind fields used to generate them -- provide the primary dataset used to explore meteorological patterns in the Northeast. Figure A-1 shows a single EDAS back-trajectory representing the path that a particle of air has traveled prior to arriving at Acadia National Park.

The back trajectories with corresponding pollutant measurements and emissions data can be combined in a number of ways to yield various metrics which may help identify specific meteorological pathways that are more likely than others to contribute to sulfate transport to a specific receptor site.

Figure A-1. Single HYSPLIT Calculated back-trajectory, Acadia National Park

A common method of analysis requires the calculation of residence-time probabilities, which are a measure of the time spent in a specific grid cell relative to the total time spent in any grid cell.¹⁴ When calculated for all trajectories considered in an analysis, this defines the *everyday* probability as shown in Equation 1.

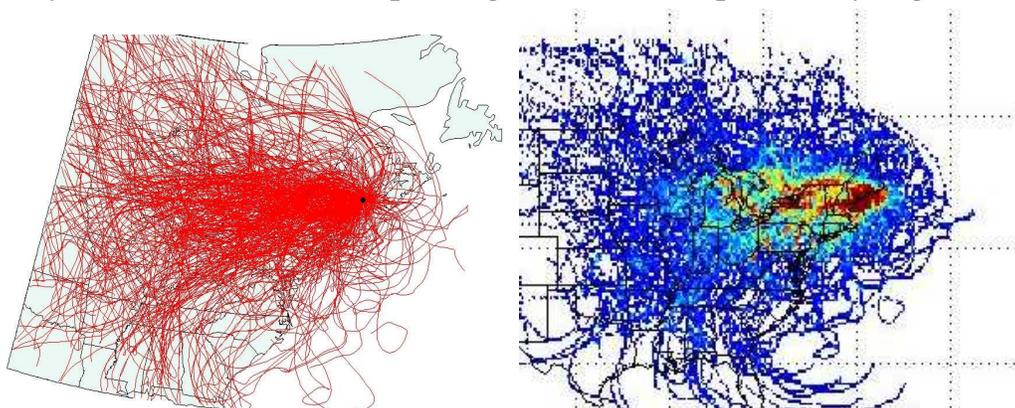
Equation 1. Everyday Residence-time Probability

$$EP = \left(\frac{n_{ij}}{N} \right)$$

n_{ij} = total endpoints passing through grid cell i, j

N = total endpoints passing through all grid cells from all trajectories

These residence time probabilities provide an indication of the fractional time spent in a specific grid cell relative to time spent anywhere in the domain by air parcels that eventually pass over a given receptor. They can be calculated for any subset of trajectories, including all the days within a time frame, yielding the “everyday” probability, or a subset corresponding to days with high measured concentrations of pollutants at a receptor site. Figure A-2 shows trajectories from the 10% of days with the highest sulfate concentrations during the 2000-2004 time period. The plot on the left shows the actual trajectories while on the plot on the right shows the resulting residence-time probability field for high sulfate days.

Figure A-2. Trajectories from the highest sulfate days at Acadia National Park. The highest 10% sulfate trajectories (left) and corresponding residence-time probability (right) are shown.

When the residence-time probability field is calculated for the subset of days corresponding to high concentrations of pollutants (as shown in Figure A-2) the result is called a *high-day* probability as shown in Equation 2.

Equation 2. High Day Residence-time Probability

$$HP = \left(\frac{m_{ij}}{M} \right)$$

m_{ij} = total high day endpoints passing through grid cell i, j

M = total high day endpoints passing through all grid cells from high day trajectories

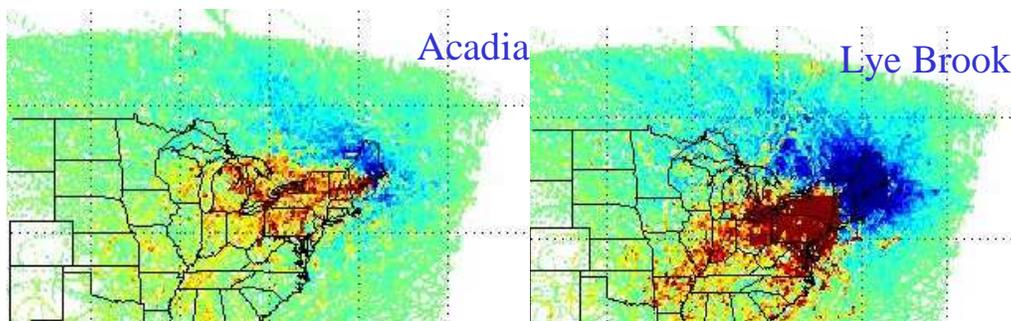
The difference between the everyday probability (Equation 1, all days during 2000-2004) and the high day probability (Equation 2, high pollution days during 2000-2004) has been referred to as the *incremental* probability and identifies areas where the probability of poor air quality is greater than the average probability associated with typical meteorological patterns (see Equation 3).

Equation 3. Incremental Probability

$$IP = HP - EP$$

When calculated in this way for each trajectory with its individual pollutant concentration value, a standard incremental probability analysis results and can serve as a basis for comparison to alternate metrics. Figure A-3 shows the 5-year incremental probability field for days corresponding to the top 10 percent of observed sulfate values at two sites: Acadia National Park in Maine and Lye Brook Wilderness in Vermont. These figures are based on available 500 meter start height HYSPLIT-EDAS back trajectories and monitoring data for those sites. Incremental probabilities have been calculated for 7 sites in the MANE-VU region and 3 nearby sites in the VISTAS region. Plots for all sites (and several different percentile groupings) are presented at the end of this Appendix.

Figure A-3. Incremental Probability (Top 10% observed sulfate days) at Acadia, ME and Lye Brook, VT during 2000-2004.

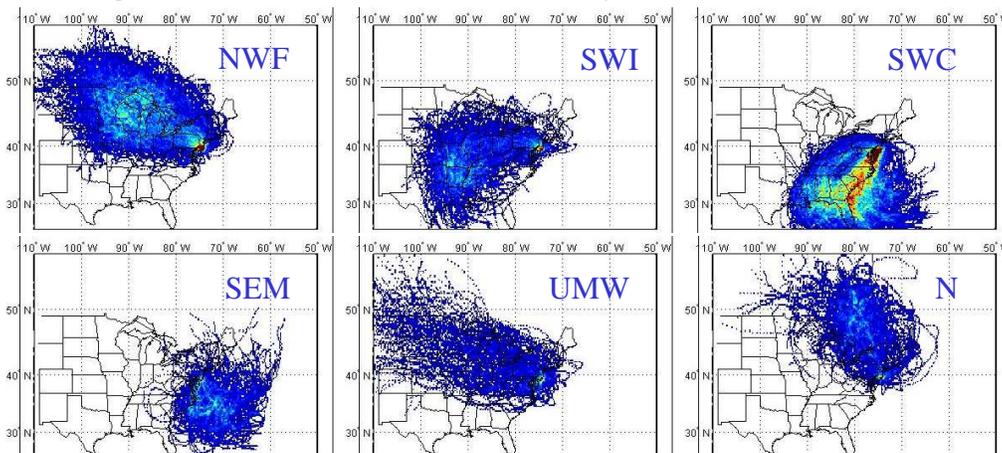


A.3. Cluster-Based Approaches

Trajectories can be used individually or grouped to create another dataset based on the PATH approach.⁶ Trajectory clusters are based on the calculated Euclidean distance between three-dimensional normalized coordinates of the respective trajectories. These clusters are formed by finding the “central” trajectory which has the greatest number of neighboring trajectories within a subjectively selected “radius of proximity.” There is a trade-off between the “resolution” of various modes of atmospheric transport identified by PATH and the number of clusters. Using a narrow radius of proximity, more defined clusters that are easily associated with a specific class of meteorological transport result (e.g. fast flow from the Northwest, shallow coastal flow, etc.); however, it also results in a large number of clusters at each site.

In order to better define specific meteorological pathways that might be associated with pollutant transport, we used a radius of proximity of 12 (this is a unit-less value since the coordinates have all been normalized prior to clustering) and a back-trajectory time of 120 hours. This typically resulted in approximately 100-200 frequency based clusters at each site, but relatively few clusters contain the majority of the trajectory population. Figure A-4 shows typical meteorological patterns among the most frequent clusters calculated for Brigantine Wilderness Area, New Jersey. Results are plotted as a residence-time probability for each cluster (see Equation 1). Clusters in the figure have been associated with specific atmospheric “modes” or meteorological patterns that are commonly observed at multiple sites. The modes pictured correspond to Northwest Fast flow (NWF), Southwest Interior (SWI), Southwest Coastal (SWC), Southeast Maritime (SEM), Upper Midwest (UMW), and Northerly flow (N). Whereas each cluster is unique to a specific site and the specific group of trajectories clustered, the modes represent patterns which are often observed year after year, and from site to site. In this sense, the clusters provide a means of identifying modes of transport which can be used for identifying pollution transport mechanisms.

Figure A-4. Residence-time probability for 6 frequency-based clusters observed at Brigantine Wilderness Area, New Jersey between 2000 and 2004.

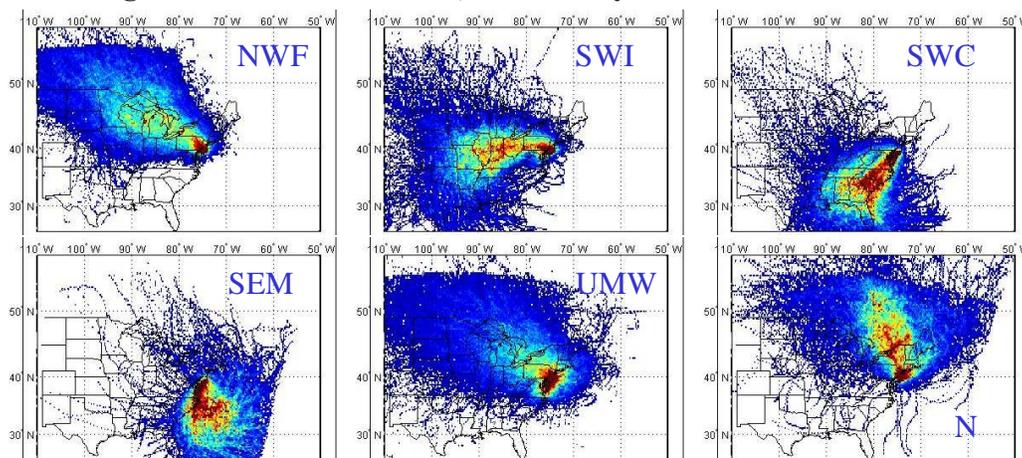


Note: Codes reflect atmospheric “modes” or typical patterns of transport which consistently appear at multiple sites.

While these frequency-based clusters are useful, a variant method of grouping these trajectories was developed to redistribute trajectories into the most common modes. This alternate method of clustering relies on the frequency-based cluster groups described in the preceding paragraphs, but forms trajectory groups based on proximity rather than frequency. In the first step, the frequency-based approach is used to identify the central trajectories that represent the most populated frequency-based clusters (approximately 10 clusters typically contain at least 98 percent of the trajectories in the dataset using R=12 and 120 hour back-trajectory time). These 10 central trajectories are then used to develop 10 proximity-based clusters by assigning every trajectory in the dataset to its nearest central trajectories (calculated back to 72 hours). The main advantage of these proximity-based clusters is that they more uniformly distribute the population of trajectories amongst the most frequently observed atmospheric modes and result in more defined clusters than the frequency-based approach. Figure A-5 shows the proximity-based clusters resulting from the frequency-based clusters shown in Figure A-1. In this proximity-based approach, trajectories are more evenly distributed, as the entire trajectory dataset are assigned to only 10 modes.

Each trajectory, frequency-based cluster, and proximity-based cluster was associated with corresponding monitoring data (or averaged monitoring data in the case of clusters) measured as close in time as possible to the “start” time of the back trajectory calculation. Associations were made for PM_{2.5}, sulfate ion, organic carbon, and aerosol extinction as PM components routinely measured as part of the IMPROVE program, although results are presented here only for 24-hr integrated sulfate ion mass.

Figure A-5. Residence-time probability for 6 proximity-based clusters observed at Brigantine Wilderness Area, New Jersey between 2000 and 2004.



Note: Codes reflect atmospheric “modes” or typical patterns of transport which consistently appear at multiple sites.

A.3.1. Cluster-weighted Probability

Everyday probabilities can be calculated for any subset of trajectories. When calculated for the trajectories within a given cluster, that cluster’s everyday probability results. This is essentially a normalized version of the residence-time probabilities shown in Figure A-4 and Figure A-5. Everyday probabilities for clusters at 13 sites are

presented at the conclusion of this appendix. The everyday probabilities for clusters at an individual site and the associated average pollution information corresponding to each cluster is combined through a method identified here as the cluster-weighted probability (CWP).

Each PATH-derived cluster's residence-time probability is weighted by the average sulfate value for any measurements corresponding to a trajectory which is a member of that cluster. The weighted residence-time probability is summed over *all* clusters calculated for a site (as opposed to just the highest probability for a specific pollutant as was the case with the incremental probability metrics). The everyday probability is subtracted from the sum of cluster-weighted probabilities to identify areas of increased (or in the case of negative values, decreased) probability of being associated with a meteorological pathway for pollutant transport. Equation 5 presents the cluster-weighted probability.

Equation 5. Cluster-weighted Probability

$$CWP = \frac{1}{\bar{C}} \left(\sum_{i=1}^L (\bar{C})_i \cdot RP_i - \bar{C} \cdot EP \right)$$

L = total number of clusters calculated

$(\bar{C})_i$ = Average pollutant concentration (based on observations associated with cluster i)

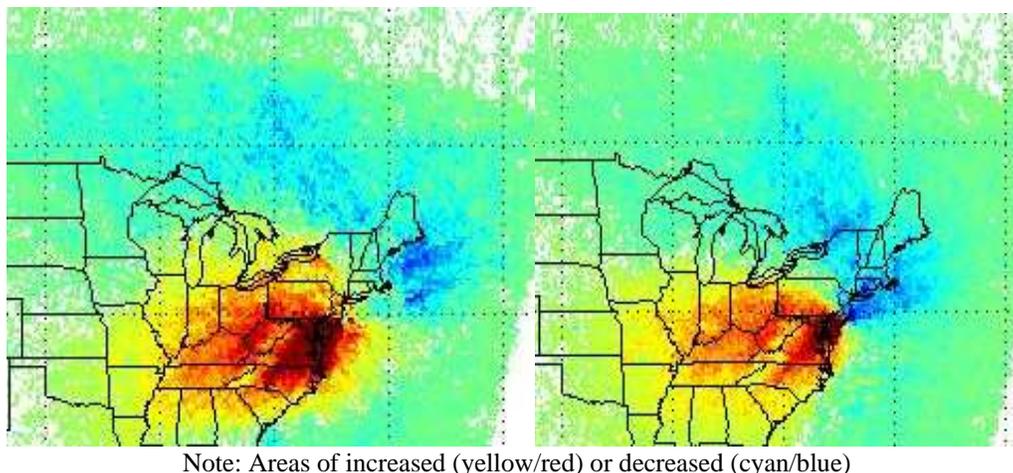
\bar{C} = Average pollutant concentration (based on all days)

Here, $(\bar{C})_i$ represents the average sulfate value for all trajectories within cluster i which had an associated SO_4^{2-} measurement (roughly 25-30 percent, given 1-in-3 day sampling schedules). By weighting the residence-time probability for cluster i by this quantity, we are implicitly assuming that similar trajectories (i.e. traversing similar source regions under similar meteorological conditions) will have similar resulting ambient concentrations at the receptor. The quantity (\bar{C}) represents the average value for sulfate measurements associated with trajectories in any of the clusters and acts to normalize the sum of the residence-time probabilities.

As we have examined two alternate methods for assigning trajectories to clusters – frequency-based clustering and proximity-based clustering – two methods for calculating cluster weighted probabilities exist. The first uses the frequency-based clusters as the dataset, while the second uses the more distributed, proximity-based clusters as the dataset. Figure A-6 shows the CWP calculated for both methods at Brigantine Wilderness Area, New Jersey.

While calculated from the same underlying trajectory files and sulfate observations, this metric is calculated in a fundamentally different way than the incremental probability approach presented. The CWP uses trajectory clusters (as opposed individual trajectories) to develop a weighted probability field which is then compared to the everyday probability. The fact that the results are similar to those calculated for the IP technique provides reassurance that these approaches are reasonable for identifying areas that are associated with sulfate transport.

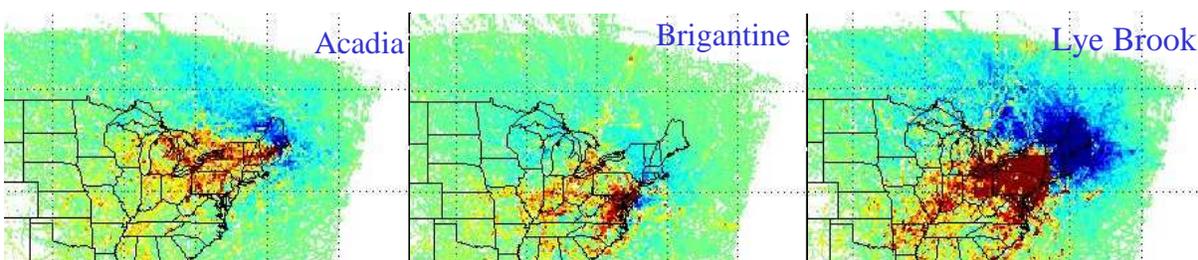
Figure A-6. Cluster-weighted Probability methods associated to sulfate transport using frequency based clusters on left and proximity-based clusters on the right at Brigantine Wilderness Area, New Jersey.



A.4. Results

Regions of transport to different sites can be determined using the trajectory (non-clustered) technique, incremental probability. In this method, pollution values are assigned to individual trajectories (as opposed to clusters) and regions associated with high sulfate transport are determined by looking at the difference of the high day trajectory probabilities from the everyday trajectory probabilities. Figure A-7 shows the top 10% sulfate incremental probability at three sites, Acadia, Brigantine, and Lye Brook.

Figure A-7. Incremental Probability (Top 10% Sulfate) at three sites, Acadia, Brigantine and Lye Brook 2000-2004.

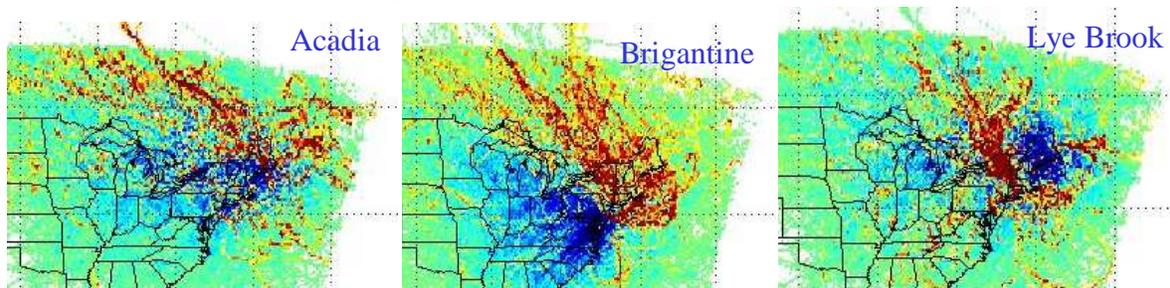


In these incremental probability plots, trajectories with higher pollution concentrations generally follow a path from the Midwest across the Northeast region prior to arriving at Acadia. Transport paths from the Southeast and the Midwest are evident at Brigantine, and a combination of Midwest transport and transport from the Southeast along the industrial I-95 Corridor affect Lye Brook.

Incremental probability can also be used to look at the regions contributing to the best pollution days at a site. Figure A-8 shows the 10% lowest sulfate incremental probability. Best-day incremental probability shows that trajectories arriving to the sites

on days with low sulfate levels tend to follow a path from the north in Canada or off the Atlantic Ocean. This is an expected result as fewer sources are located in this region.

Figure A-8. Incremental Probability (Bottom 10% Sulfate) at three sites, Acadia, Brigantine and Lye Brook 2000-2004.



By examining pollution levels associated with different clusters, we can gain insight into which atmospheric modes are associated with transport to sites in the MANE-VU region. Figure A-9 compares the proximity-based cluster with the highest sulfate value at three sites, Lye Brook Wilderness Area, Brigantine Wilderness Area, and Acadia National Park.

Figure A-9 demonstrates that the clusters showing the highest pollution values show similar meteorological characteristics at different sites. The three sites show that in the higher average pollution clusters, the air follows the path from the Midwest to the site in question. Lye Brook shows an east coast component contributing as an area of transport on the worst average sulfate days. Using this method, we have grouped trajectories purely based on meteorological characteristics and can examine pollution levels within these groupings. Thus agreement between this method and the incremental probability results shown earlier suggest that whether you select trajectories based on meteorology or chemistry, similar regions are identified as being associated with sulfate transport. Figures at the end of this appendix show the clusters and average pollution values for all the sites in the region analyzed.

Figure A-10 displays the proximity-based clusters with the lowest average sulfate concentrations. They show which meteorological mode contributes to the best pollution days at three different sites, Acadia, Lye Brook, and Brigantine. This provides a method that is comparable to Figure A-8 where incremental probability for the lowest sulfate trajectories is shown. Again, the comparison is made between this clustering technique where trajectories are grouped solely on meteorological characteristics and the incremental probability technique that looks at trajectories with similar pollution characteristics.

Much like the atmospheric modes that contribute to the highest sulfate levels at our three sites, very similar atmospheric patterns contribute to the best sulfate days across multiple sites in the region. Air masses moving in from the north and off the Atlantic Ocean contribute to the best days at Acadia, Brigantine, and Lye Brook. Similar modes affect the best days at many other sites, seen in the section following this appendix.

Figure A-9. Proximity based cluster with the highest associated sulfate value for three sites in the MANE-VU region, Acadia (sulf=3.19 $\mu\text{g}/\text{m}^3$), Brigantine (sulf=6.79 $\mu\text{g}/\text{m}^3$), and Lye Brook (sulf=4.56 $\mu\text{g}/\text{m}^3$).

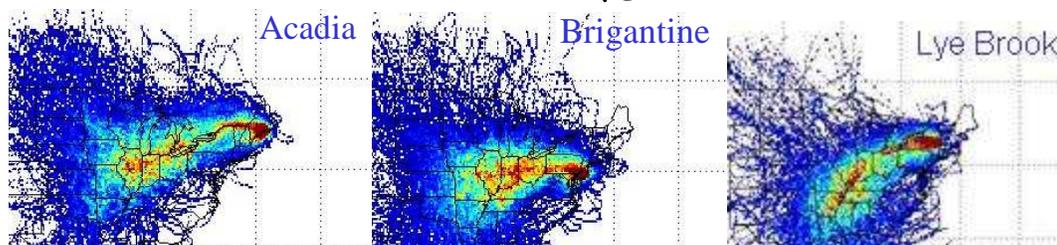
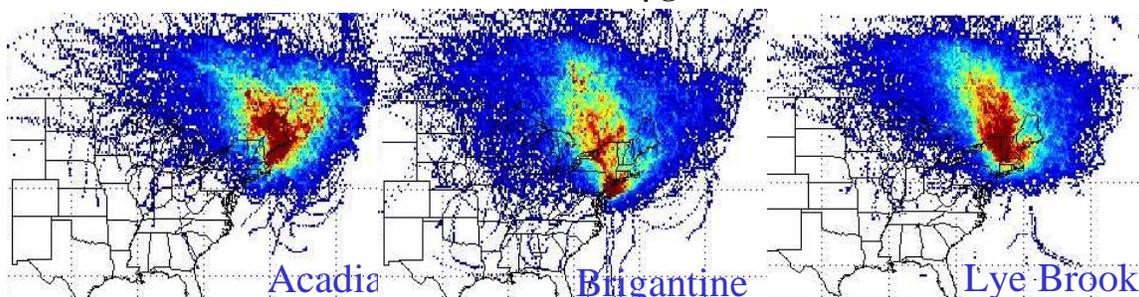
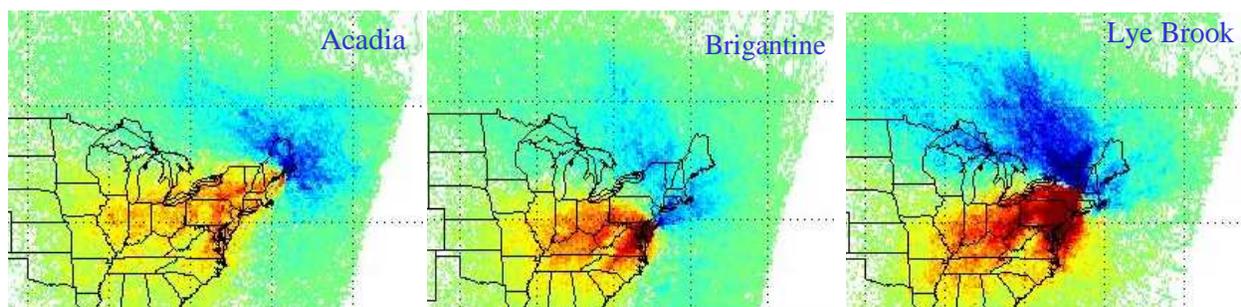


Figure A-10. Proximity based cluster with the lowest associated sulfate value for three sites in the MANE-VU region, Acadia (sulf=1.41 $\mu\text{g}/\text{m}^3$), Brigantine (sulf=2.23 $\mu\text{g}/\text{m}^3$), and Lye Brook (sulf=1.16 $\mu\text{g}/\text{m}^3$).



Cluster weighted probabilities are used to combine cluster information with pollution information and the following CWP plots show regions with greater probability of sulfate transport in red/yellow and regions of decreased probability of sulfate transport in blue. We have presented the CWP based on proximity-based clusters in Figure A-11, which shows results similar to Figure A-9 since the clusters weighted by the highest pollution values tend to enhance the regions they cover, consistent with a greater probability of contributing to sulfate transport. However, the CWP plots in Figure A-11 now contain information from all the clusters (and thus all the trajectories) in the dataset.

Figure A-11. Cluster Weighted Probability at three sites, Acadia, Brigantine and Lye Brook 2000-2004.



By averaging the various probability fields across sites, we can gain insight into which regions are most likely to be associated with sulfate transport to multiple sites in the MANE-VU region. A region that is associated with transport to several sites in different locations is far more likely to be associated with large emission sources. Figure A-12 show a multi-site averages of incremental probability for the top 10 percent worst days at five northern sites, Acadia, Lye Brook, Camp Dodge, Moosehorn, and Mohawk Mountain. Figure A-13 shows the multi-site average CWP which includes all proximity-based clusters for the same sites.

Figure A-12. Multi-site average probabilities showing areas of increased (yellow/red) or decreased (cyan/blue) probability of being associated with sulfate transport to Acadia, Lye Brook, Camp Dodge, Moosehorn, and Mohawk Mountain on the ten percent worst sulfate days.

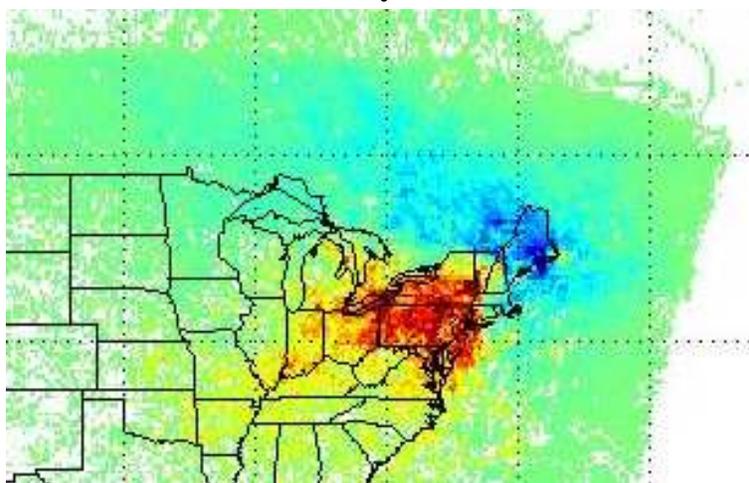
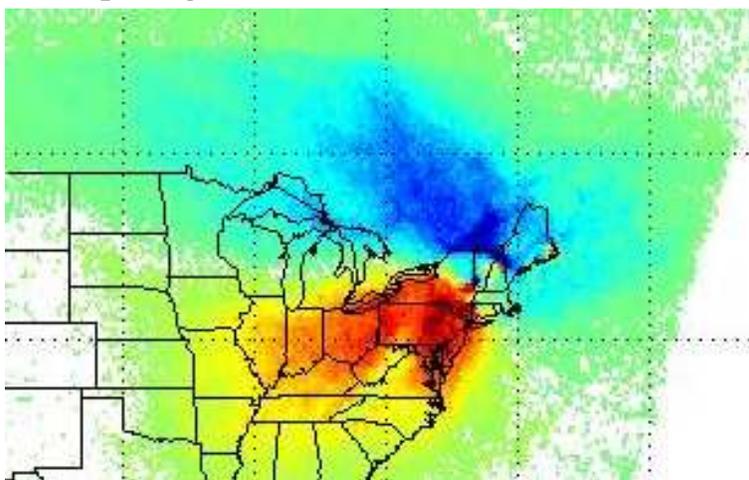


Figure A-13. Multi-site average CWP showing areas of increased (yellow/red) or decreased (cyan/blue) probability of being associated with sulfate transport to Acadia, Lye Brook, Camp Dodge, Moosehorn, and Mohawk Mountain.



From a qualitative perspective, the two metrics (IP and CWP) are quite similar showing significant sulfate transport (on an annual average basis) along the Eastern corridor from Virginia up through Maryland and Eastern Pennsylvania. A second area of influence along the Ohio River valley between Ohio, Pennsylvania and West Virginia seems to play a significant role as well.

These averages can also be applied to different geographical regions as well. Figure A-14 shows the average IP while Figure A-15 shows the average CWP for four southern sites, Brigantine, Shenandoah, Dolly Sods, and Great Smokey Mountains. The figure highlights specific regions of higher probability of sulfate transport affecting a large area along the east coast.

Figure A-14. Multi-site average IP showing areas of increased (yellow/red) or decreased (cyan/blue) probability of being associated with sulfate transport to Brigantine, Shenandoah, Dolly Sods, and Great Smokey Mountains.

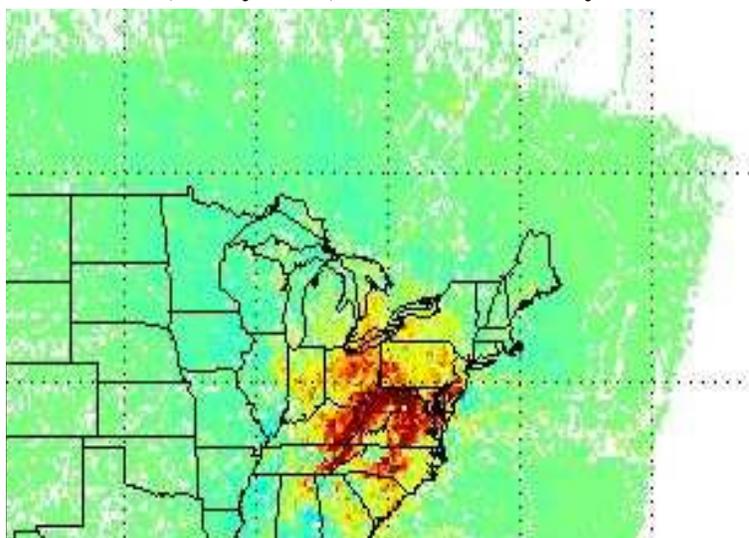
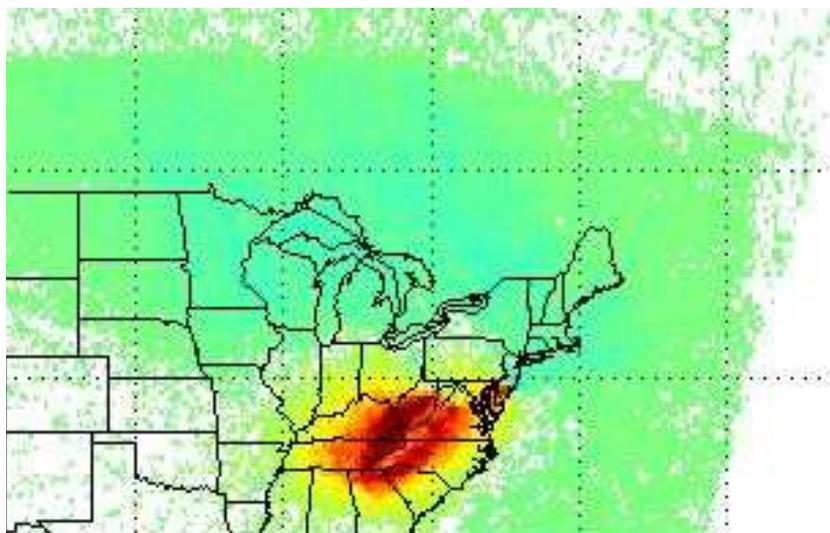


Figure A-15. Multi-site average CWP showing areas of increased (yellow/red) or decreased (cyan/blue) probability of being associated with sulfate transport to Brigantine, Shenandoah, Dolly Sods, and Great Smokey Mountains.



A.5. Conclusions

A large database of back trajectories and corresponding air pollution measurements have been used with trajectory cluster analysis techniques to apportion observed sulfate mass concentrations as an independent check on source modeling results. Clusters have been used here to develop metrics related to Wishinski and Poirot's "incremental probability"¹⁰ which provides the relative increase in probability of a geographic region being associated with a predominant meteorological pathway associated with pollutant transport.

Several techniques have identified state-specific contributions of SO₂ emission sources to sulfate formation. Results indicate that cluster-based trajectory techniques can provide a semi-quantitative check on chemical transport model results. This analysis demonstrates a potentially novel way of identifying regions that play a role in pollutant transport (and may host source emissions as well). Combined with meteorological information and source apportionment model results, the approach may yield a more comprehensive picture of source emissions and the circumstances under which they are transported to specific receptor sites.

A.5.1. Acknowledgements

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