

# ATMOSPHERIC DISPERSION STUDY OF DEICING SALT APPLIED TO ROADS



**PART II FINAL REPORT FOR PERIOD JULY 2002 TO JUNE 2004  
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<p>This report describes results from a project to characterize the atmospheric emission and deposition of deicing road salt composed mainly of sodium chloride (NaCl). The focus of the effort has been to determine the airborne concentrations and deposition of roadsalt aerosol due to the proposed construction of a roadway, FAP 340, which would join Interstate 55 (I-55) and Interstate 80 (I-80) southwest of Chicago. Five monitoring sites were established to characterize the road salt deposition. The report gives an overview of the sampling sites, presents data from the different instruments deployed at the sampling sites, and describes a model developed for the study to simulate salt deposition</p> <p>The roadsalt model is the combination of an emissions model and a deposition model. The deposition parameterization is based on measurements following several snow events of salt collected in snow samples at different distances along I-55, which show the salt deposition to essentially vary inversely with the distance from the roadway. The emissions model is based on an assumed exponential decay with time of emissions from a roadway for which the start time and decay rate of emissions were determined from wind-directed measurements of the salt deposition at the different sampling sites. The initial emission rate is determined by applying the emission model and recorded wind direction measurements with time for the periods following the snow events after which the snow samples were collected. Finally, the annual deposition in the roadsalt model is computed as the average deposition pattern from different snow events using the above parameterization and recorded measurements of the wind direction with time scaled to the annual number of snow events. Results of the model are compared to measurements of deposition at the 5 sampling sites and the model is applied to simulate the deposition patterns following construction of FAP 340.</p>			
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# **Atmospheric Dispersion Study of Deicing Salt Applied to Roads: Part II Final Report for Period July, 2002 to June, 2004**

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## **Introduction**

This report describes results from a project to characterize the atmospheric emission and deposition of deicing road salt composed mainly of sodium chloride (NaCl). The focus of the effort has been to determine the airborne concentrations and deposition of roadsalt aerosol due to the proposed construction of a roadway, FAP 340, which would join Interstate 55 (I-55) and Interstate 80 (I-80) southwest of Chicago. Five monitoring sites were established to both characterize the road salt deposition prior to the construction of the proposed roadway and to measure the increase in deposition once the new roadway is built. Results for the contract period, July 1, 2002 to June 30, 2004 are summarized from measurements at the five monitoring sites established during this project. These sites will be referred to as the Lemont sites. Measurements to characterize the timing and duration of roadsalt emission, from a trailer sampling platform which was placed at the Leverett Road salt facility located about 5 miles north of Champaign, IL, will also be discussed. Results of measurements at the Lemont sites from 1996 to spring 2002 were reported in previous progress reports for this contract.

The report gives an overview of the sampling sites and presents data from the different instruments deployed at the sampling sites. A detailed description of the road salt model developed as a part of this effort is also described. The roadsalt model and supporting files are included on a CD as part of the report. Appendix A is a user guide explaining the procedures necessary to run the model. Appendices B and C contain copies of two papers expanding aspects of the project. The paper in Appendix B was presented at the 2004 meeting of the Air and Waste Management Association (AWMA) in Indianapolis, IN and the paper in Appendix C is a preprint of a paper to be presented at the 2005 AWMA meeting in Minneapolis, MN. Rather than including multiple copies of figures in the text, those figures cited in the write-up that are also included in the papers will be referenced according to the citation in the Appendix. So, for example, Figure 1 of Appendix C will be cited as Figure C-1.

From the standpoint of model development, a key advance has been the formulation of the emission source term with regard to its strength, decay, duration, and timing. From analysis of the wind-directed data at the Lemont sites we have been able to formulate a representation of the salt emission source. The basic procedure was to make a simple emissions model and perform sensitivity experiments varying the start time and duration of emissions using measured hourly wind data for each of several sampling periods for all five sites. The relative amounts of salt at each wind-directed sampler could best be explained by a scenario where the salt emission begins at the end of snowfall and the emission lasts for 2 days (48 hours). The model is based on the 48-hour emission scenario and the spatial

distribution of salt concentration measured in snow samples downwind of I-55. The model is incorporated into Geographical Information Systems (GIS) ArcGIS 8.3 software. The user must prepare input files using common ArcGIS procedures and supplemental files included with the model. The roadsalt model runs inside ArcGIS and from the input files computes a map of the annual salt deposition at the selected site 5 km x 5 km site. With the GIS basis for the presentation of the model, the salt deposition can be layered on other GIS maps as an aid to interpretation. Therefore, the salt deposition can be estimated for any areas that have the proper geographic projection.

## **Description of Baseline Monitoring Sites**

The proposed roadway, FAP 340, is a 12-mile road section that would join I-55 at Bolingbrook and would join I-80 at New Lenox (see Figure C-1). A bridge approximately 1 mile long would span the Des Plaines River Valley crossing the river, the Sanitary and Ship Canal, and the I&M Canal. Figure C-2 is an aerial view of the area showing the approximate position of the proposed roadway.

The primary strategy behind locating the sampling sites was to provide a method for determining the road salt deposition from the proposed new road. Essentially, the sites were positioned in areas that are somewhat isolated and that would be affected by salt deposition from the new roadway. The project goal was to contrast measurements of the salt deposition at the sites before and after construction of the roadway. As shown in Figure C-2, all 5 sites were close to the proposed roadway. Sites 1 and 2 were in the river valley, in an area that would be below the proposed bridge, at two different (downwind) distances toward the east. Measurements from dry deposition and aerosol samplers should show larger values adjacent to the bridge compared to lower values at a farther distance away. Both the magnitude and rate of falloff with distance from the source are important parameters for characterizing the impact of the new road. Sites 3 and 4 were located similarly to sites 1 and 2 at different distances east of the proposed roadway. These sites were outside the river valley in an at-grade position with respect to the new roadway.

A wind-directed sampling strategy was a consideration for site selection. Collecting samples when the wind is carrying salt directly from the roadway to the sampling site, without the interference of salt material originating elsewhere, helps in determination of the impact of emissions from the roadway. In the case of sites 3, 4, and 5, where site 5 was on the opposite side of the proposed roadway from sites 3 and 4, the wind-directed measurements can be used to determine the amount of salt in the background from which the amount added by the new roadway can be more easily determined. This was considered to be especially important with westerly winds that could carry salt emissions from I-55, Joliet Road, and International Parkway to site 5 and could increase the background at sites 3 and 4.

Additional considerations for site locations were the practical aspect of acquiring permission from land-owners to establish monitoring sites on their land. Sites 1 and 2 were along a portion of the historic I&M Canal on land managed by the City of Lemont. Site 4 was in the DuPage County Forest Preserve, and Sites 3 and 5 were on properties of Citizens Utilities Corporation. Several aspects of the sites raise questions for the interpretation of the data presented here. The salt deposition at site 5 should be influenced by I-55, although the effect of buildings and other obstructions shown in Figure C-3 are complicating factors that potentially affect the salt deposition pattern. A considerable portion of the warehouse construction between site 5 and I-55 has been done since the beginning of the study and there has been significant new construction since the photograph in Figure C-3 was taken. There is

no direct pathway between I-55 and site 5 that was unobstructed by warehouse buildings. International Parkway (IP) is within 30 m (100 ft.) of site 5, and the impact of the IP salt source is potentially important. Joliet Road, which lies between site 5 and I-55, is also a potentially strong salt aerosol source for site 5 and possibly sites 3 and 4. It is a divided highway that joins I-55 and carries heavy rush hour traffic.

Sites 3 and 4, while less obstructed from I-55 emissions by the buildings, were located in wooded areas and were obstructed by trees. Figure C-4 provides an even more detailed aerial photograph of sites 3 and 4. Site 3 was clearly in the middle of a woods and salt aerosol originating from any direction could be filtered by the trees before arriving at site 3. Site 4 was obstructed by the same woods from the south and east. Two open fields separated by a line of trees lie to the north and east of site 4. There were no obvious local sources to sites 3 and 4 such as IP or Joliet Road that may affect salt deposition at site 5. Their seclusion in wooded areas should serve to decrease the salt deposition in comparison to the other sites because of distance from local sources and the potential filtering effect of the trees.

Sites 1 and 2 were isolated from I-55 by larger distance as well as buildings and trees, and they have the additional complicating effect of the intervening river valley. The Village of Lemont lies immediately to the south of sites 1 and 2. As shown in C-5 New Avenue runs in an east-west direction passing both sites at a distance of approximately 0.25 mi to the south, and is a potential source of salt aerosol. The bridge crossing the canal area at Lemont Road is located about a mile to the east of site 2. The elevated source of the bridge on Lemont Road, as well as the proposed bridge, may be of some importance for salt emission. Like Joliet Road, Lemont Road connects with I-55 and has a considerable rush-hour traffic load.

Although the primary design consideration for placement of the sites was to characterize the proposed new roadway, since the construction was not undertaken it was decided to focus the data toward characterizing the background salt deposition and air concentration values and to develop a model to simulate salt emission and deposition. Essential data for a model include representation of the emission source. Measurements of the salt deposited in snow adjacent to I-55 for 4 different events have shown that the deposition occurred mainly south of the roadway and that the prevailing winds were such that the deposition patterns were inconsistent with emission occurring during the snow event. Based on the snow samples and the associated wind patterns we consider dry emission to be the dominant emission mechanism. In addition the deposition was found to falloff approximately as the inverse of the distance from the roadway and to have dropped to fairly close to the background in a 1 km or so from the roadway. Since the sampling sites were all more than 1 km from I-55, which is considered to be the major roadsalt source in the area, the site placement is not ideal for determining its source characteristics. The sites were not only too far from I-55 to obtain a strong signal, there were intervening buildings and other construction as described. Also, the uneven terrain is a complicating aspect of the site locations that is difficult to quantify but undoubtedly affects the salt aerosol deposition. The Des Plaines River Valley is approximately 600 ft elevation above sea level as were sites 1 and 2. Site 3 is at approximately 650' elevation and surrounded by woods. Site 4 which is only about 350 m northwest of site 3, is at approximately 700' elevation the same as site 5. Bluff Road, depicted in Figure C-2, is at approximately 600' elevation and marks the northern boundary of the River valley. North of Bluff Road the elevation increases rapidly to 650. So sites 1 and 2 were in the flat valley floor, site 3 was on the hillside, and sites 4 and 5 were in a flat upland area.

## Overview of Sampling Procedures

The main instruments used for sampling the airborne road salt include dichotomous aerosol samplers (dichots) and high-volume aerosol samplers (hivols). Aerochem wet-dry precipitation samplers, to be identified as “aerochem samplers”, were used to collect the salt from dry deposition as well as that contained in precipitation. The wind-directed aerosol sampler was designed to sample the same particle sizes as the hivol based on a commercial inlet with aerodynamic characteristics similar to a hivol sampler. The dichot collects particles in the two diameter (D) ranges of  $D < 2.5$  micrometers ( $\mu\text{m}$ ) and  $2.5\mu\text{m} < D < 10\mu\text{m}$ . The hivol collects all particles with approximately  $D < 30 \mu\text{m}$ . The wind-directed instrument monitors wind speed and direction at the top of a 20 ft tower at each site and, depending on the direction of the wind, opens one of 4 filter-sampling valves. Thus each of the 4 filters represent material collected when the wind, averaged over 15 minutes, was from a particular sector. The inlet nozzle is also at the top of the tower, and the sample flow train extends to filters located at the bottom of the tower. Table 1 of Appendix C (Table C1) summarizes the aerosol size classes and the associated instruments used to measure them.

Dry deposition of Na has been measured at the five Lemont sites using automatic wet/dry precipitation samplers ( Aerochem Metrics Model 301) throughout the course of the study. A picture of the sampler at site 2 is on the web: (<http://www.sws.uiuc.edu/atmos/deposition/deposit2.asp?sz=bgr&pn=2>). The Aerochem sampler is used at the sites in the National Atmospheric Deposition Network/National Trends Network (NADP/NTN is described at <http://nadp.sws.uiuc.edu> ). The Aerochem holds two 14-liter plastic buckets, with a lid that is automatically positioned over the wet-side bucket during periods without precipitation and over the dry-side bucket during precipitation events. A sensor for rain or snow particles activates a motor that moves the lid between the two bucket positions. The sample buckets were cleaned with deionized water and stored in plastic bags for transport to and from the field sites. The rain and snow captured in the wet-side buckets is filtered and analyzed by ion chromatography. For the dry-side buckets, 250 milliliters of deionized water was added to the bucket, and after a minimum of 24 hours for soaking, the dry-side sample is handled like a wet-side sample. The Aerochem sensor works very well for rain events. It is common for some of the snow to end up in the dry-side bucket, especially during cold temperatures and when snowflakes are small, because the sensor has not detected the snowflakes and so not moved the lid to cover the dry-side bucket.

There were Aerochem, hivol, and wind-directed samplers at each site, and dichot samplers at sites 3, 4, and 5. To prevent sample overload but yet collect a representative sample during the course of the approximately 1-month sampling periods, the hivol and dichot samplers were operated for 10 minutes at the beginning of each hour.

The wind-directed sampling sectors were depicted by 4 points that were defined by roadway intersections and can be seen in Figure C-2. The “north” sector was determined by the intersection of I-55 and Joliet Road, on one hand, and the intersection of I-55 and Lemont Road on the other. This means that the “north” direction was slightly different for each site. The point of such a definition is that emission from the same segment of I-55 would be collected at each site for the “north” directional sampling. The two other points defining the different sampling segments were the intersection of Lemont Road and New Avenue, and the intersection of Joliet Road and Route 53. The principle local source in the “east” direction was Lemont Road including the bridge which crosses the river and canals just north of New Avenue. The “east” segment may have also picked up emissions from Chicago. The “south” direction included emissions from New Avenue, Lemont as a whole, and a wide area to the

southwest which should have low local sources. Finally, the “west” direction included emissions both from a segment of I-55 and Joliet Road. Wind-directed inlets at all 5 sites were programmed to sample from a particular sector when the wind is from a direction included in the angle from the receptor to these intersection points. The actual angles will be slightly different for each site with the result that winds from directions defining the sector boundaries can be regarded as from different sectors depending on the sampling site. For example, a northerly wind with a slight easterly component, while satisfying the criteria of being from the northerly sector at site 4 may be regarded as being from the easterly sector at sites 1 and 2.

## Measurements at the Lemont Sites

Discussion of the Aerochem results is presented in the papers in the attached as Appendixes. In addition to the data from the Lemont sampling sites, the papers analyze Aerochem data collected at the Argonne National Laboratory (see Figure C-2) over a 18 year period. In particular it is shown that dry deposition of roadsalt is much higher than wet deposition. A relationship is developed between the Aerochem dry deposition and the amount of salt applied. The magnitude of the salt deposition at the five sampling sites is discussed. The salt deposition at site 5 is statistically higher than the other sites as expected because of the proximity of IP to site 5. Also it was found that the salt deposition at site 1 is statistically higher than at site 4. This suggests that New Avenue is the dominant source in the canal region of sites 1 and 2 rather than I-55.

A summary of mass and speciation measurements for each of the aerosol size classes during snow and non-snow periods is shown in Table 1. For the snow periods 77% of the salt mass is in particles larger than 10 micrometers ( $\mu\text{m}$ ) diameter, and 96 % of the salt mass is in particles above 2.5  $\mu\text{m}$  diameter. However, salt makes up only about 5 % of the total mass above 10  $\mu\text{m}$  diameter and only about 6% of the mass above 2.5  $\mu\text{m}$  diameter. Thus the salt is concentrated in the larger particles and even during snow periods it remains a small portion of the total particle mass.

Figure 1 shows the annual variation found for Chloride concentration over the period 1996-2002. In both 1997 and 1998 the salt aerosol concentration was significantly higher than the other years. The monthly average concentration over the same time period is shown in Figure 2. As expected the salt aerosol concentration is higher in the winter than in the summer. The relative concentrations at each of the sites of the different particle size classes measured are contrasted for the different sampling sites in Figures 3 and 4. As seen in the Aerochem data, site 5 has the highest concentrations in all aerosol size ranges. The aerosol concentrations at the canal sites (sites 1 and 2) are slightly higher than the upland sites 3 and 4.

## Trailer Experiments

Attempts were made for both the winters of 2002 and 2003 to characterize the timing and magnitude of the emission from I-57. In these experiments a trailer was located near I-57 at the Leverett Road maintenance facility north of Champaign. The idea of the measurements was to monitor the aerosol particle size distribution (PSD) with optical particle counters and to simultaneously collect wind-directed filter samples. Since the optical measurements are made quickly they can possibly be used to observe the evolution of the salt aerosol source with time following a snow event. The filter measurements, although requiring several hours to collect sufficient mass for analysis, can be used to confirm that if periods of high particle

counts occur during periods of high salt concentrations. Even though the monthly average salt aerosol concentration makes up only 5% or so of the total aerosol mass as discussed above, if the salt emission is episodic such that it is high for just a few days of the month, then during those days it could account for a more significant portion of the aerosol mass. In that case one would expect to see high concentrations of aerosol following a snow event.

The experiments were frustrated by equipment problems and the data did not turn out as well as hoped. However, there did not appear to be a discernable difference in the size distribution following snow events. During periods when emissions of salt should have been high there was no discernable difference in the size distribution downwind of I-57 as compared to other wind directions.

An unexpected result of the trailer experiments were occasional very large spikes lasting a few hours, but none following a snow event. The largest such period was at a time after the snow had been on the ground for several weeks and the temperature had warmed enough to melt the snow. During the whole time the roads were clear. A possible interpretation is that as the temperature rises after a prolonged cold period during which salt was applied to the highway numerous times, water is wicked from the porous concrete on the highway surface bringing salt to the surface where it can be eroded by vehicle traffic.

## **ROADSALT MODEL**

### ***Introduction***

The development of a roadsalt deposition model has previously been challenging by lack of information on the emission source. The trailer experiments were designed to help characterize the source, but due to instrumental problems and the inherent difficulty of identifying whether the aerosol measured over a short time interval was predominantly NaCl or other aerosol, the data is of limited utility. However, the wind-directed monitoring at the permanent sites has yielded data from which source characteristics can be extracted. The essential questions are 1) what is the source strength, 2) when does the emission start, and 3) how long does it last. Considering the salt deposition at a point due to a nearby road source, no matter what the emission source strength if the wind is not from the road to the target point there will be no deposition at the target point. Also, there are questions as to when the source starts relative to the snow event and how the source, once started, decays with time. If the wind is from the road to the target point before the source starts or after it has sufficiently decayed the deposition will be small. Data from the snow sampling northwest and southeast of I-55 in the Romeoville area showed the deposition in each of four different cases to be largely southeast of the roadway with comparatively very little deposition to the northwest of the road. That this happened in all four cases raises the possibility that the deposition predominantly occurs during periods of northwesterly winds. Timing of the emission and the direction of the wind during emission are critical factors determining the emission scenario.

The strategy employed here is to construct an emission model that depends on the start time of emissions with respect to the end of snowfall and the duration of the emission source. The model is then optimized to find the start time and duration parameters that yield the best comparison between the model output and the salt loading on the wind-directed filters. The essential goal of the emission analysis is to define a consistent scenario describing when, after a snow event, the emission starts and how long it lasts.

An important aspect of parameterization of the salt deposition is the observation of the falloff of salt as a function of distance from a treated roadway. Our previous analysis of

the observed salt deposition patterns in snow indicates the deposition decreases approximately as the inverse of distance from the roadway (see Figure 5). Measurement of salt deposition in the snow near an interstate highway in cases where the wind changed direction following the end of precipitation has led to the conclusion that the negligible emission occurs during the snow event. Most of the emission occurs after the roadway has had a chance to become dry. We therefore consider dry emission to be the dominant pathway for salt aerosol emission.

At this stage an annual average deposition pattern is desired rather than a deposition pattern for each snow event. Although more salt is applied for larger snow events, more snow and applied salt are plowed to the side of the roadway during roadway clearing operations, so it is not known how the amount of dried salt on the road is related to snow amount. Records are available for the amount of salt applied. Figure C-8 is a plot of the salt applied to I-55 near the Argonne Aerochem sampler versus the Aerochem sodium deposition. The amount of salt applied has a significantly higher correlation to the sodium deposition than does the total snow amount. To be the most effective, salt must be applied prior to the snow event so application is somewhat based on the prediction of snowfall. It is therefore reasonable that the salt application is related to the number of snow events. To proceed with the model we will compute a deposition pattern representative of an average snow event and multiply by the number of events to get the annual average.

Parameterization of the annual emission can be used in a general way to estimate the deposition resulting from an arbitrary roadway network. By integrating the parameterization of the annual emission into a format consistent with the Geographical Information System (GIS), which is widely used by IDOT, we can achieve the capability of applying the emission model to any roadway network. This would aid in interpretation of the salt emission data since other GIS overlays, such as maps of sensitive biological areas that might be impacted by salt deposition, can be displayed in conjunction with the estimated salt deposition.

### ***Emissions Model***

The emissions model is based on the assumption that the emission from a roadway starts at its highest level and decays exponentially. Accordingly, the emission rate,  $E$  (the mass emitted per length of road per unit time), can be expressed as

$$E = E_0 \exp(-t/T) \quad (1)$$

Here  $E_0$  represents the initial emission rate,  $t$  the time since start of emission.  $T$ , the time for the emission to be reduced to  $1/e$  of its original value, is representative of the duration of emission. The amount of salt emitted over a time interval is determined by integrating equation 1 from the start to the end of the interval. For the wind-directed data set the average wind direction is determined over 15 minute intervals and the filter corresponding to the indicated direction is loaded for a fraction of that period. The integral of equation 1 over a 15 minute interval is

$$E_0T [\exp -t/T - \exp -(t+15)/T]. \quad (2)$$

If the integral is taken over all time the result is  $E_0T$ , describing the total emission possible from the model, so the emission for each 15 minute increment divided by the total emission represents the fraction of the total emitted during that time interval. By normalizing the

emission in this way,  $E_0$  cancels out of the ratio and the fraction,  $F$ , of the total emission for each time interval can be determined from the time  $t$ , and the characteristic time  $T$  as

$$F = [\exp -t/T - \exp -(t+15)/T]. \quad (3)$$

Experimentally, the timing of the end of the snow fall for the site sampling was determined from a Belfort rain gauge modified to electronically monitor the 15-minute bucket weights that increase as precipitation falls into the bucket. Belfort gauges were placed at two of the 5 sites to determine the number and timing of precipitation events. With this data and supplementary data from nearby O'Hare and Midway Airports, judgments were made on a case by case basis whether individual events were rain or snow. As shown in Figure 6, the Belfort data shows quite accurately when precipitation ended in each case.

The wind-directed instrumentation consisted of an anemometer and wind vane to determine wind speed and direction. These readings were also recorded and 15-minute averages determined. At the end of each 15-minute interval the wind direction so determined was used to trigger sampling on the appropriate filter. Therefore an electronic record was available from which sampling direction was selected at each site for each 15-minute interval.

The emissions model describes source emission independent of source strength as given by equation 1. It is assumed that each source starts emitting salt at the same time following a snow event, and that each source decays similarly, emitting the fraction  $F$  of the total emission from that source in each 15-minute time interval. If the scenario is correctly specified so the timing of the emission and wind direction approximate the actual case, then the scenario predicted deposition should correlate with the actual filter loading. The comparison is made for the different cases by varying  $T$  in equation 3 and by varying the emission start time with respect to the measured end of snowfall.

Figure 7 shows plots for the case for loading of the wind-directed filters at sites 1 and 2 under easterly winds from sector b. In this scenario the amount of sodium measured on the filters is contrasted with the computed fractional loadings  $I_{4}$ ,  $I_{8}$ ,  $I_{12}$ , and  $I_{48}$  assuming the emission starts at the end of snowfall and lasts for durations of  $T = 4, 8, 12,$  and  $48$  hours respectively. As can be seen, the measured loading and the computed fractional emission correlate most highly for a duration of  $T = 48$  hours. The algorithm describing the emissions essentially has two aspects. First, each case considered follows the same wind pattern corresponding to the measured wind direction following the end of snowfall. Also, for a given hour after the end of snowfall the wind is always from the same sector. Therefore, the wind direction and open or closed condition of the wind-directed sampler is the same for each of the plots in Figure 7. The second aspect of the algorithm is that the exponential decay of emissions according to equation 1 is different for each plot in the figure. The model is essentially an exponential numerical filter operating on a time series. In the case shown, the  $T = 48$  hours duration gives the best fit between the measured sodium values and the estimated fractions. The scenarios examined include  $I_{2}$ ,  $I_{4}$ ,  $I_{8}$ ,  $I_{12}$ ,  $I_{24}$ , and  $I_{48}$  which represent emission for 2, 4, 8, 12, 24, and 48 hours after the end of snowfall. Also scenarios described as  $I_{12\_2}$ ,  $I_{12\_4}$ ,  $I_{12\_8}$ ,  $I_{24\_2}$ ,  $I_{24\_4}$ ,  $I_{24\_8}$ ,  $I_{48\_2}$ ,  $I_{48\_4}$ , and  $I_{48\_8}$  were examined. Here the first number of the pair is the duration of the emission and the second number is the number of hours after the snow ends that the emission starts. Paired  $t$ -tests of each scenario predicted filter loading with the measured filter loadings show that  $I_{48}$  gives the best match. The  $I_{48}$  scenario then describes the emission to start immediately

after the end of snowfall and to decrease according to  $\exp(-t/T)$  where  $T = 48$  hours. This defines the emission scenario used for the model.

### ***Deposition Model***

The deposition model is based on the source strength and falloff with distance from the roadway as measured in the snow sample adjacent to I-55 and shown in Figure 5. The best fit equation shown in the figure is for the case of measurements taken at various distances from a line source. The approach in the model will be to determine the length of road in each model grid cell, a computation that can be easily performed in GIS, and approximate the variation of deposition with distance as proportional to  $1/D$ . The approach, then, is to represent the emissions as a series of point sources instead of a single line source, so one would not necessarily expect the best fit curve shown in Figure 5 to hold.

For a point at a distance  $D$  from an emission source, the time variation of the source will be assumed to follow equation 1 and the spatial variation of the deposition assumed to vary as  $1/D$ . So the deposition per unit of time per unit length of road will be taken as

$$f(D,t) = (K_0/D) \exp(-t/48) \quad (4)$$

where  $K_0$  is a constant. Integrating equation 4 over distance from  $D1$  to  $D2$ , corresponding to the size of a grid cell at an arbitrary location in the grid, and over 1 hour, the time step of the wind record, yields

$$\text{Dep} = 48 K_0 \ln(D1/D2) [\exp(-t/48) - \exp(-(t+1)/48)] \quad (5)$$

A model based on equation 5 was developed and the constant  $K_0$  determined by comparison of the results to the snow sample data.

The method used is to specify a rectangular grid as the model domain and designate a single cell located at the center of the grid as an emission source corresponding to a unit length of road. Since the distance and direction from the center cell to each other cell in the domain is known, then for a specified wind direction the grid cells downwind from the source for that hour can be determined. The amount of hourly emissions is distributed over the downwind cells according to the  $1/D$  variation. By repeating the process for each hour after the end of snowfall for the duration of 48 hours, the total emission in each cell for the event can be determined. For the purposes of calibration the roadway network can be taken as a segment of I-55 beside which the snow samples were collected. The wind records for the sampling period are known. Comparison of the computed deposition and the snow sample measurements will allow quantification of the parameter  $K_0$ .

The deposition pattern for the grid with a single source at its center can be computed for whatever wind direction sequence desired and the resulting salt amount in each grid cell computed. Since the calculation is specific to the wind pattern and not to the roadway network, it only has to be computed for each calibration sequence and for the average annual deposition pattern. The results can be stored in a “kernel” file and accessed whenever needed. The procedure to compute the actual deposition corresponding to a roadway network starts by determining which grids of the domain contain road segments that emit salt. The kernel can then be translated to the center of the emitting cell and the kernel deposition imposed. The deposition pattern from the emitting cell is then the length of the road segment times the deposition pattern of the kernel. By sequentially translating the kernel center to every cell in

the domain that contains a road length segment and in each case summing the resulting deposition over the domain, the total deposition pattern corresponding to the initial roadway network can be determined.

An additional assumption made in the model development addresses the question of some roads being more intense emitters than others. Besides the length of road, the traffic vehicle use density for each road segment is used to scale the emission strength. Thus, the calibration is performed on I-55 and the emission from another road to which the model is to be applied is scaled according to its traffic volume and the traffic volume on I-55.

Whether calibrating the model or during normal use a particular roadway network must be chosen. The method employed here is to use a roadway file of Illinois roads supplied by IDOT. This file dictates the GIS projection and insures that the projection used is consistent with IDOT use. A model grid consisting of 100 cells on a side of 50 x 50 m size was chosen. This produces a domain which is 5 km on a side. The grid is projected in a GIS ArcMap model along with the roadway network. The grid being at much smaller scale than the state roadway network can be moved inside the roadway network using the ArcMap "Editor" function. Once positioned, the road and grid can be "intersected" with the ArcMap "Geoprocessing Wizard". This produces a projection the same size as the grid but with roadway properties for each grid cell including the length of road and traffic density. These are multiplied to produce a single variable, and a "raster" file is produced containing that product for each grid cell. The raster file can then be accessed and processed by a Visual Basic for Applications (VBA) program written for the model. The VBA program operates within ArcMap similar to a common Macro. The VBA program reads the kernel and the road intersection file and performs the process of translating the center of the kernel to each emitting cell in turn and computing the resulting deposition over the whole domain. The result of the VBA calculation is another raster file that can project the emissions spatially over the grid.

## **Model Calibration**

A plot of the modeled concentration adjacent to I-55 for the January 14, 1998 snow sample experiment is shown in Figure 8. The concentration pattern is seen to fall off monotonically with distance from the road. Different ArcMap tools in the "Spatial Analyst" were used to make raster maps of the distance and direction of each point in the grid from the roadway. These files and the concentration file were placed in an ArcMap Geodatabase that has the advantage of being accessible through a database program. Microsoft ACCESS was then used to group the data to find the distance of each concentration reading from the road. The computed deposition as a function of distance from the road was summarized keeping only those points on the map lying south of I-55. Figure 9 is the computed and measured concentration vs. distance for snow samples collected on Jan 14, 1998 and Figure 10 represents similar data for samples collected on Jan 17, 1998. The computed data actually represent the second iteration at the process, where the value of  $K_0$  had already been set, so the computed and measured values already match in the cases presented and no further calibration is needed.

## ***Annual winds***

The hourly wind fields following snow events were extracted from a 6 year data record of hourly meteorology at O'Hare International Airport. A total of 51 snow events were identified and the wind direction for each hour after end of snowfall recorded. Hourly

averages of the wind direction are shown in Figure 11 of each hour up to 48 hours. The data shows that the wind starts at around 260 degrees (winds from the southwest) and stays nearly from that direction for 24 hours. It then shifts to around 200 degrees for the remainder of the time. The average wind field would not yield a very realistic wind pattern to describe annual emissions since it is confined to such a limited angular range. The method used for preparing the kernel file for the model was to compute separate deposition patterns over for each of the 51 wind fields and then average the deposition pattern. This provides a deposition pattern for a single snow event. It was assumed that there are 17 snow events per year for which road salt is applied.

The wind data are provided on an hourly basis so the practical modeling question arises as to whether to tabulate the emissions according to fixed winds every hour. The procedure used was to consider the wind directions for consecutive hours as being the hourly range and that the wind is equally distributed over that angular range.

## ***Model Results***

The database of Illinois roads provided by IDOT for the salt deposition study is under revision and unfortunately some of the newly constructed roads in the study area are not yet reflected in the database. In the vicinity of sites 3, 4, and 5, International Parkway (IP) is not in the database and Davey Road, which was in large part replaced by IP, is different in several respects. This is a key factor from the standpoint of the salt deposition at sites 3, 4, and 5 because of the possibility of emissions from IP dominating the salt deposition. We know this to be true at site 5 and the dominant sources at sites 3 and 4 are an open question. The road network in the study area was therefore changed for this study by building IP into the database.

Figure 12 is an example of the model output for the region where Joliet Road intersects I-55. IP also crosses Joliet Road as indicated in the figure. The concentration plot shows that the prevailing wind carries more salt southeast of I-55 than to the Northwest. As I-55 and Joliet Road are near their intersection at the top of the frame the emission sources are so close together they behave as a single source. As they are separated at the bottom the deposition patterns are separate. The impact of IP is also evident. As noted the strategy for handling emissions from different roads was to use the calibrated source strength and scale the particular roads according to their traffic volume. The daily traffic volume for I-55 in the road segment shown is 121,400 whereas that for Joliet Road it is 25,000. The daily traffic volume for IP was estimated at 15,000.

Simulation of the annual salt deposition over the study area has been carried out for two cases corresponding to different roadway networks in the study area. For the first case the only change was to add IP as stated. In the second case proposed highway FAP 340 is constructed in the database to simulate the effect it would have on salt deposition. The exact location of the roadway is estimated based on available data and the traffic volume is assumed to equal 104,100 which is representative of existing I-355 north of I-55. To construct the case studies the different simulations are overlapped to make a mosaic depicting the salt deposition over a wider area than the 5 km x 5 km domain of the model output. Care must be taken in this process with the current version of the model because sources just outside the model domain will not contribute to the deposition pattern inside the domain. If a simulation is performed with a source just outside the domain, and then a simulation performed with the source inside the domain, there will be points common to both simulations differing in value. In no instance can the domain positioning cause spuriously high values, but spuriously low values can occur if a source is excluded from the domain.

The continuity of the concentration contours over multiple model domains is an indication that the overlapping procedure is working properly.

The combined deposition pattern for approximately 20 simulations of the model were used to construct Figure 13 which is the model estimate of the salt deposition in the study area under current conditions. The deposition pattern in the neighborhood of each monitoring site is of particular interest from the standpoint of model validation. From the figure it is difficult to make a case for emissions from I-55 affecting any of the sites. Sites 1 and 2 lie in the shadow of potential emissions from New Avenue. Site 5 is clearly dominated by IP. Deposition at sites 3 and 4 is more complex. When a prevailing wind direction is along a roadway the simulated concentrations can become much higher than when the wind is across the road. The associated deposition pattern becomes more apparent when the road changes direction. This is the case with the simulation in the neighborhood of sites 3 and 4 where IP in its east-west orientation lines up approximately with sites 3 and 4 and then turns to the north.

Table 2 lists the measured and computed annual salt deposition values at each of the monitoring sites. The annual median salt deposition values compare quite well with the measured values. The poorest agreement is at site 5 where the model over-predicts the observed value. In this case the sampling site is within a single model grid cell from the roadway so the spatial resolution is beyond the model capability. For each of the other sites the model is about 30 % lower than the observations.

Since the traffic volume for IP was not available in the database and was estimated at 15,000, further simulations were done to check the sensitivity of the deposition at sites 3 and 4 to the traffic volume on IP. If the traffic volume is reduced to 10,000 the deposition at sites 3 and 4 decrease by almost an order of magnitude. If it is increased to 20,000 the deposition very closely matches the observed values. The traffic volume on Joliet Road in this area is 24,000. Clearly Joliet Road has more traffic than IP and the 20,000 estimate for IP seems too high. The value of 10,000 traffic volume on IP seems like a reasonable estimate, but the resulting model simulated deposition is much too low. An aspect of potential importance is that IP has a disproportionate amount of truck traffic because of the large number of warehouses along IP. If the emission per vehicle is proportional to the number of axels, an "18 wheeler" truck would produce over 4 times the salt emission than an automobile.

The model simulation for emissions over the study area after the completion of FAP 340 is shown in Figure 14 and the concentration values evaluated at the sampling sites are shown in Table 2. The model estimates large increases in the deposition at the sampling sites if FAP 340 is constructed. This is partly by experimental design as sites 1 and 4 are very close to the expected alignment. The annual deposition at these sites is projected to be 5 g/m<sup>2</sup> representing twenty-fold increases from the present values. Sites 2 and 3 which are further from the alignment are projected to increase to 1.2 and 2 g/m<sup>2</sup> respectively which is only moderately higher than the present modeled value at site 5. The model simulation at site 5 is relatively unchanged by the presence of FAP 340.

### ***Model Limitations***

The underpinnings of the model are substantially based on the observed regular decrease of salt concentration in snow samples measured adjacent to I-55. The model was calibrated based on the snow sample measurements. Application of the model is largely based on the formulation of an annual average deposition pattern which is then applied to different roadway networks. The model results compare very well with experimental results at the sampling sites.

The model is a first effort at simulating salt deposition. The total effort involved developing not only a deposition model but an emissions model. The events that could affect emissions are extremely complex. However, to within the accuracy of the experimental evidence from the wind-directed monitoring, the assumed exponential decrease of salt emission with time after the end of snowfall seems to provide satisfactory results.

The model is very simple and non-sophisticated in an important way. It does not consider atmospheric transport of the salt aerosol, but uses instead observed deposition patterns. Wind speed is not considered at all in the model since the observed deposition pattern was not developed as a function of wind speed. The meteorological information having to do with wind direction and timing of the end of a snow event are all formulated in the “kernel” file which is then applied to different roadwork networks.

	Non-Snow Periods			Percentiles			Snow Periods			Percentiles		
	N	Mean	Std. Error	25%	50%	75%	N	Mean	Std. Error	25%	50%	75%
PM-10+ Mass	8	43.20	8.02	29.75	34.39	64.44	16	28.00	4.60	16.03	21.75	36.85
PM-2.5+ Mass	15	11.36	1.32	6.33	11.89	15.31	24	8.52	0.81	5.99	7.46	10.01
PM-2.5 Mass	15	11.78	0.76	9.86	11.96	13.41	27	12.90	2.12	12.07	14.59	17.14
PM-10+ Na	7	0.40	0.13	0.21	0.23	0.48	16	1.06	0.20	0.54	0.71	1.34
PM-2.5+ Na	15	0.03	0.00	0.02	0.03	0.04	24	0.25	0.05	0.06	0.20	0.39
PM-2.5 Na	15	0.03	0.00	0.02	0.03	0.04	25	0.05	0.01	0.02	0.05	0.06
PM-10+ Cl	7	0.32	0.09	0.11	0.28	0.62	15	1.30	0.30	0.36	0.96	1.59
PM-2.5+ Cl	15	0.05	0.01	0.03	0.05	0.07	24	0.32	0.06	0.08	0.24	0.43
PM-2.5 Cl	15	0.01	0.00	0.00	0.00	0.02	27	0.09	0.02	0.03	0.06	0.11
PM10+ Ca	7	0.91	0.12	0.69	0.98	1.13	16	0.79	0.09	0.53	0.71	1.05
PM-2.5+ Ca	15	0.36	0.04	0.29	0.33	0.39	24	0.31	0.03	0.23	0.29	0.38
PM-2.5 Ca	16	0.06	0.01	0.04	0.06	0.08	24	0.05	0.00	0.03	0.05	0.07
PM-10+ K	6	0.13	0.04	0.04	0.11	0.21	16	0.07	0.01	0.05	0.07	0.08
PM-2.5+ K	15	0.02	0.00	0.02	0.03	0.04	24	0.02	0.00	0.01	0.01	0.02
PM-2.5 K	15	0.09	0.01	0.06	0.08	0.11	26	0.08	0.01	0.05	0.08	0.10
PM-10+ SO4	5	3.20	0.41	2.28	3.53	3.96	10	2.92	0.42	1.98	2.84	4.00
PM-2.5+ SO4	15	0.34	0.04	0.19	0.31	0.44	24	0.39	0.04	0.26	0.35	0.45
PM-2.5 SO4	16	3.12	0.37	2.15	2.90	4.12	27	2.47	0.21	1.63	2.45	3.17

Table 1. Super-coarse (PM-10+), Coarse (PM-2.5+), and Fine (PM-2.5) Measured Mass Concentrations ( g/m3) for Snow and Non-snow periods.

Table 2. Comparison of annual salt deposition (mg/m<sup>2</sup>/yr) for Aerochem measurements, model output with the present roadway network, and the projected deposition at the sites after construction of FAP 340.

<b>Site</b>	<b>Aerochem</b>	<b>Model present</b>	<b>Model with 355</b>
1	0.33	0.23	5.00
2	0.25	0.17	1.20
3	0.18	0.14	2.00
4	0.29	0.20	5.00
5	0.52	0.80	1.20

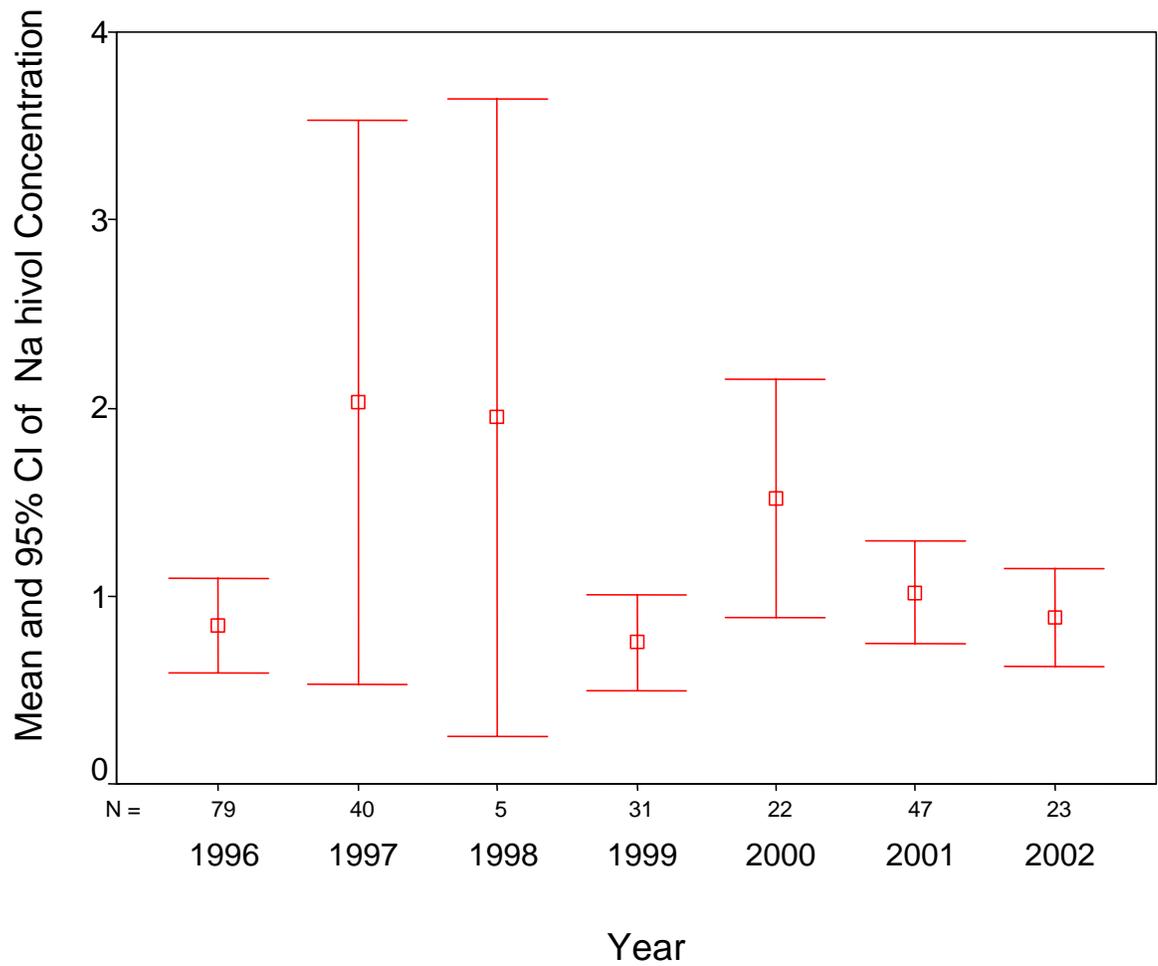


Figure 1. Mean ( squares) and 95% confidence interval (CI) about mean (whiskers) of Na hivol concentration ( $\mu\text{g}/\text{m}^3$ ) for 1996 through 2002. N = annual sample number.

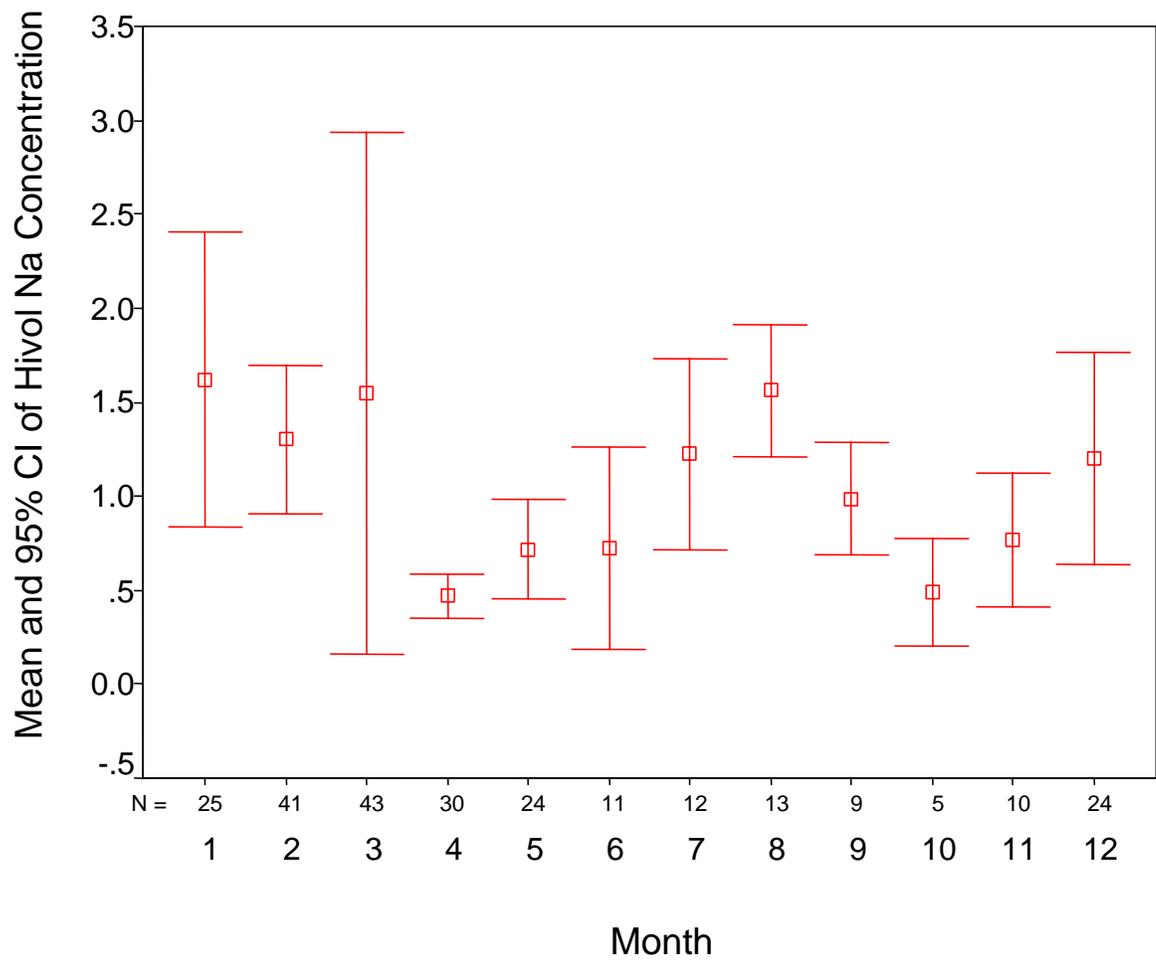


Figure 2. Mean (squares) and 95% confidence interval (CI) about mean (whiskers) of monthly Na hivol concentrations ( g/m<sup>3</sup>) for 1996 through 2002. N = monthly sample number.

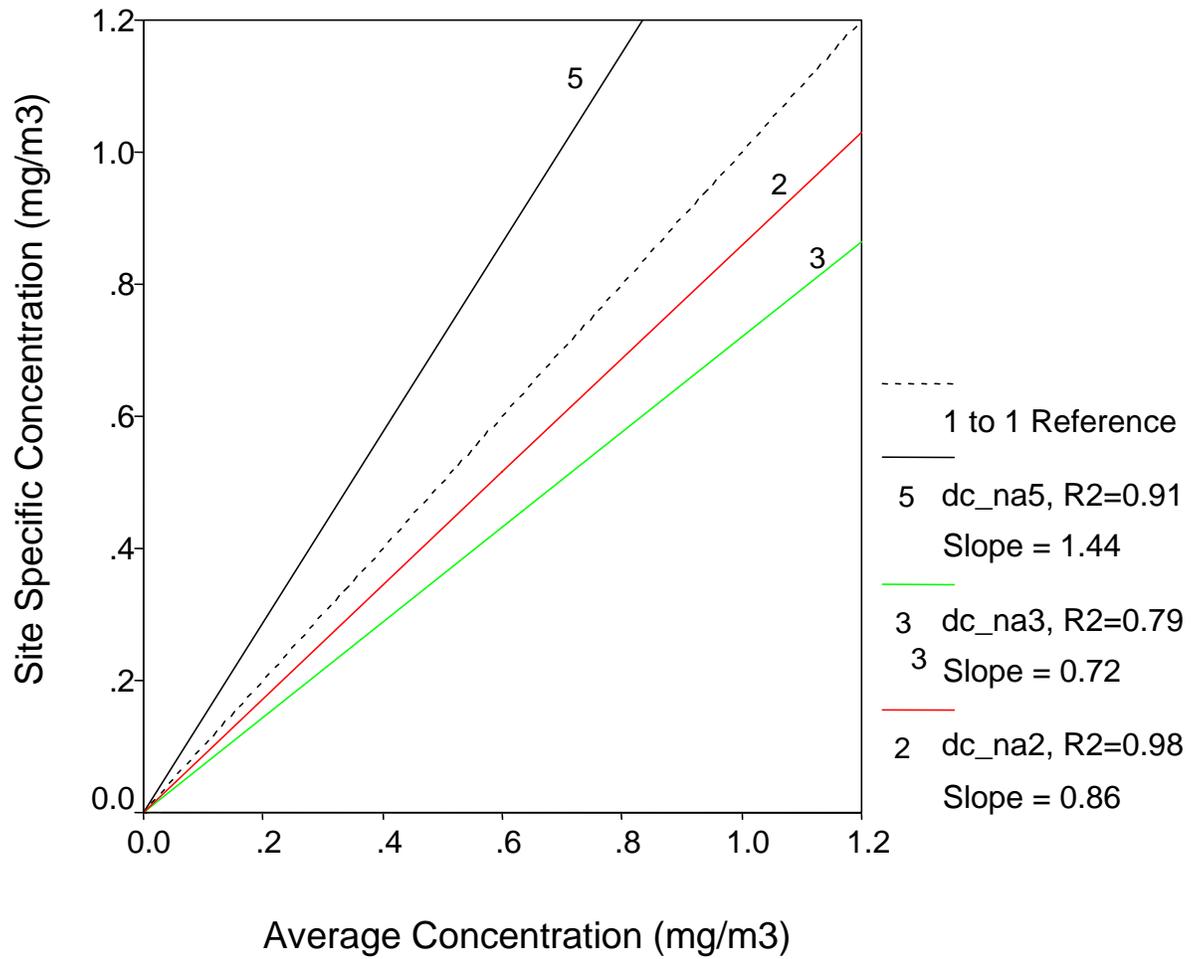


Figure 3. Comparison of each of the dichot coarse particle concentrations at sites 2, 3, and 5 to the average of sites 2, 3, and 5.

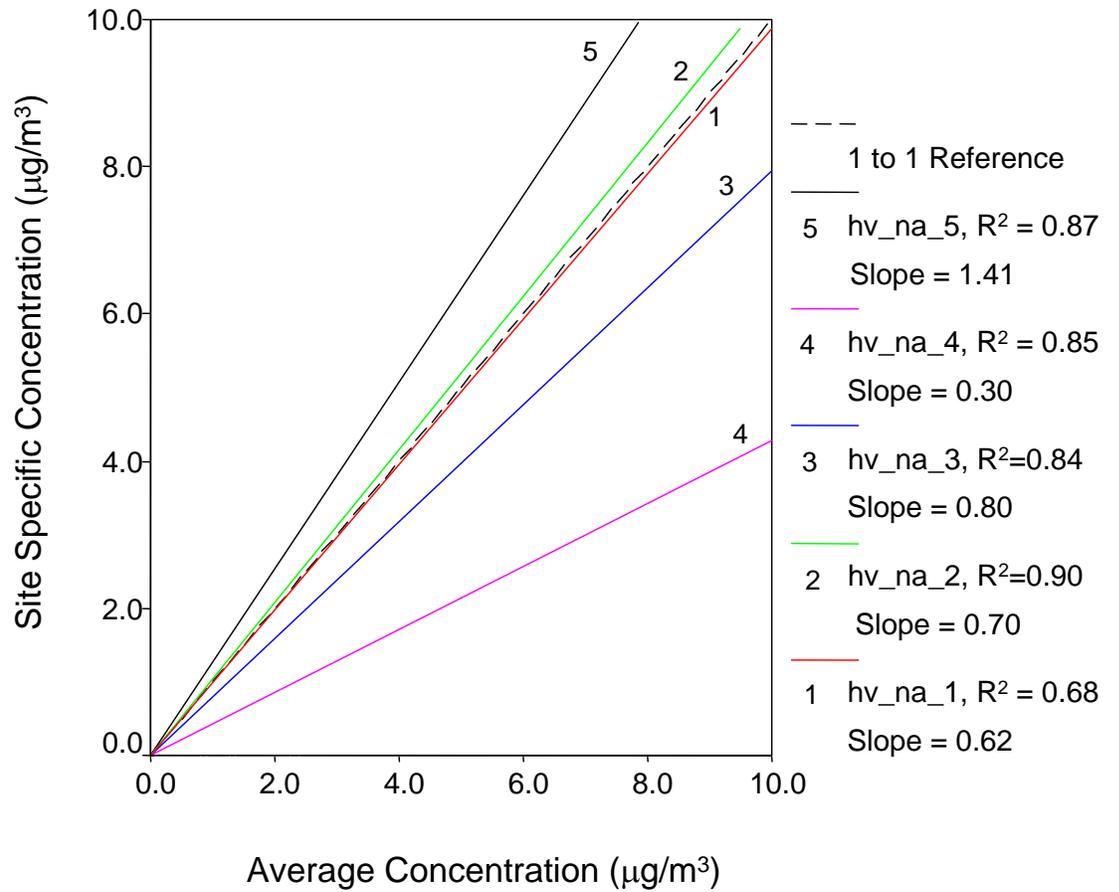


Figure 4. Comparison of each of the hivol particle concentrations at sites 1,2, 3, 4, and 5 to the average hivol particle concentrations at sites 1, 2, 3, 4, and 5. Dotted line is 1 to 1 reference line.

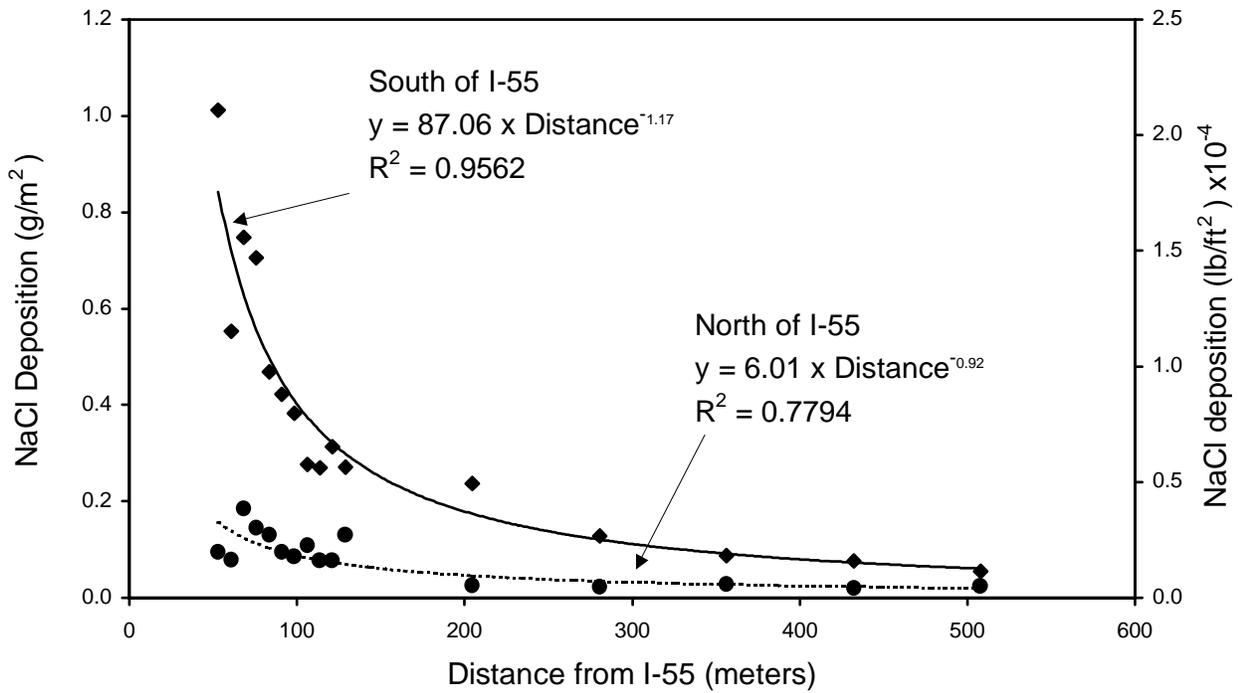


Figure 5. Average of snow deposition amounts for samples collected north and south of I-55. Regressions are valid in the approximate range 50 m < D < 500 m.

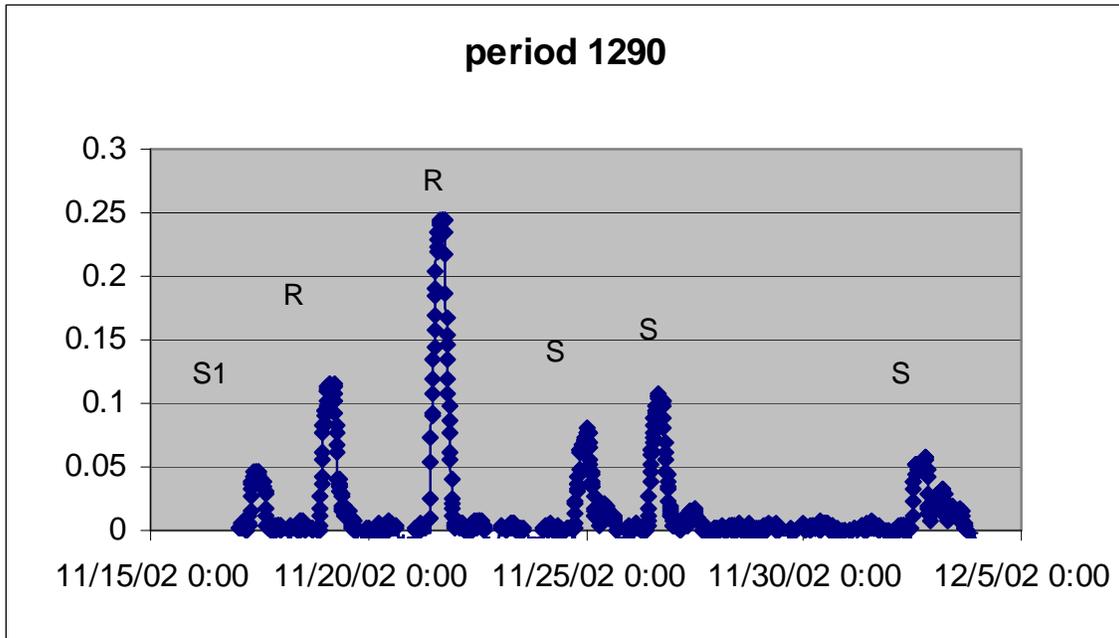


Figure 6. Belfort rain gauge data showing precipitation in the sampling period 1290 which was from 11/15/02-12/05/02. The determinations between rain (R) and snow (S) were made independently. The time that snow ends can be estimated from the data.

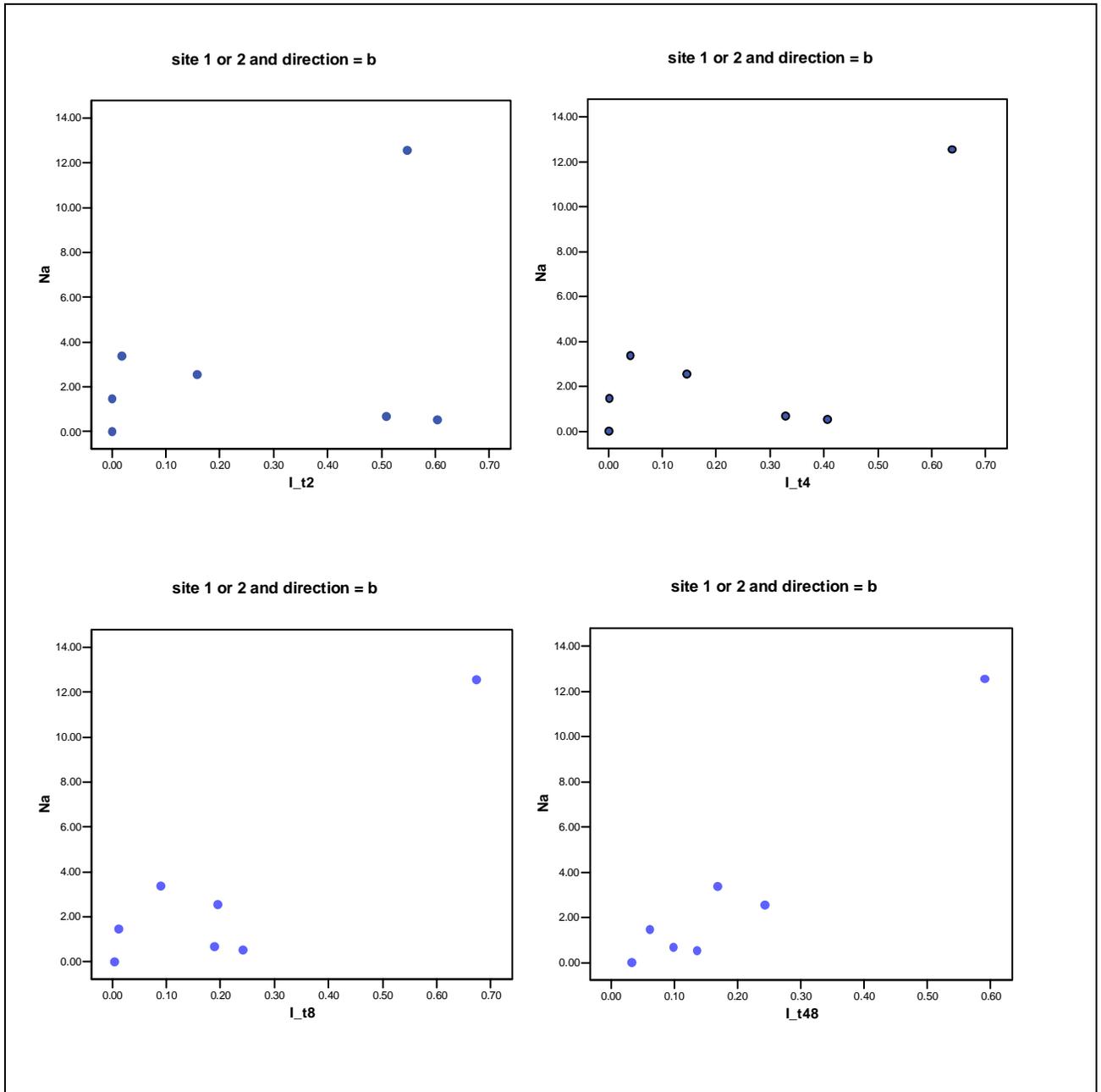


Figure 7. Sodium collected on wind-directed filters vs. the computed fraction of salt emitted (see equation 3) for time periods starting at end-of -snow and extending for different time periods of 2, 4, 8, and 48 hours. Data is for sites 1 and 2 when the wind was from the east.

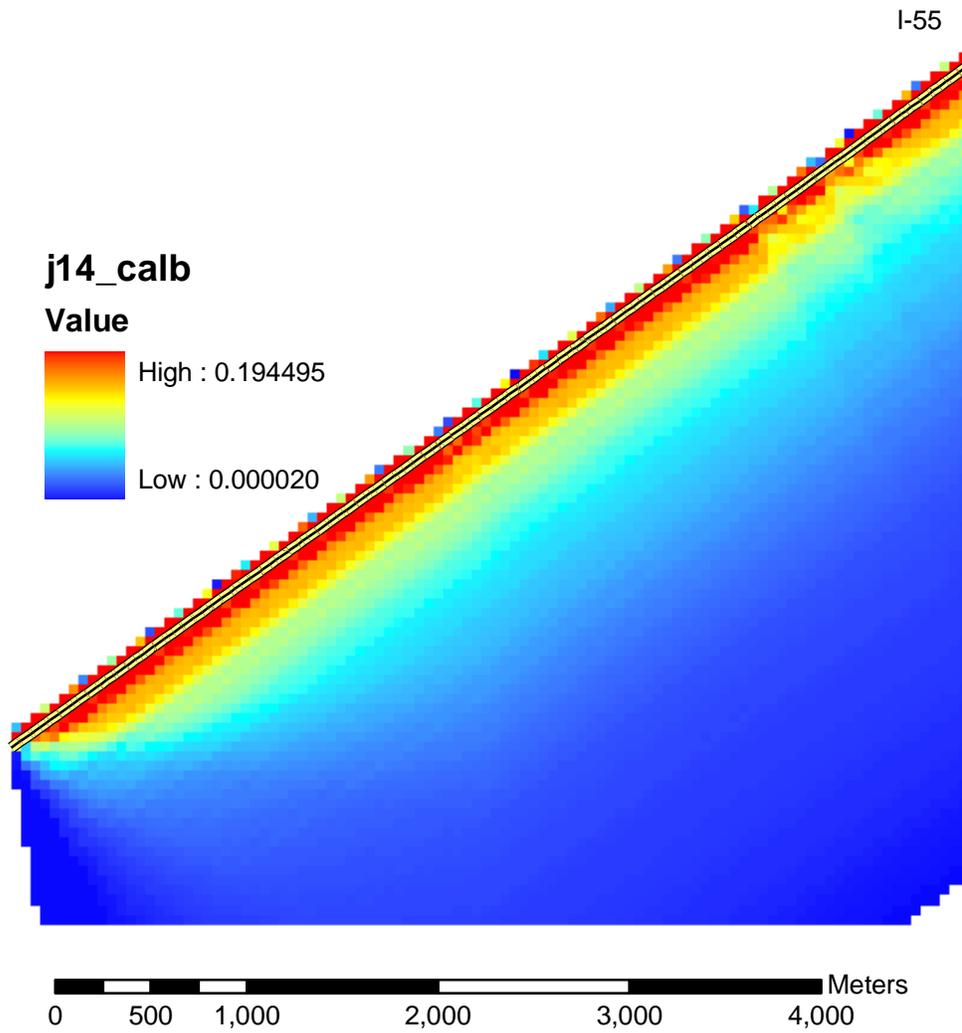


Figure 8. Simulation of salt deposition ( $\text{g/m}^2$ ) from I-55 for the Jan. 14, 1998 snow sampling. Samples were collected in a line perpendicular to the road out to a distance of 500 m.

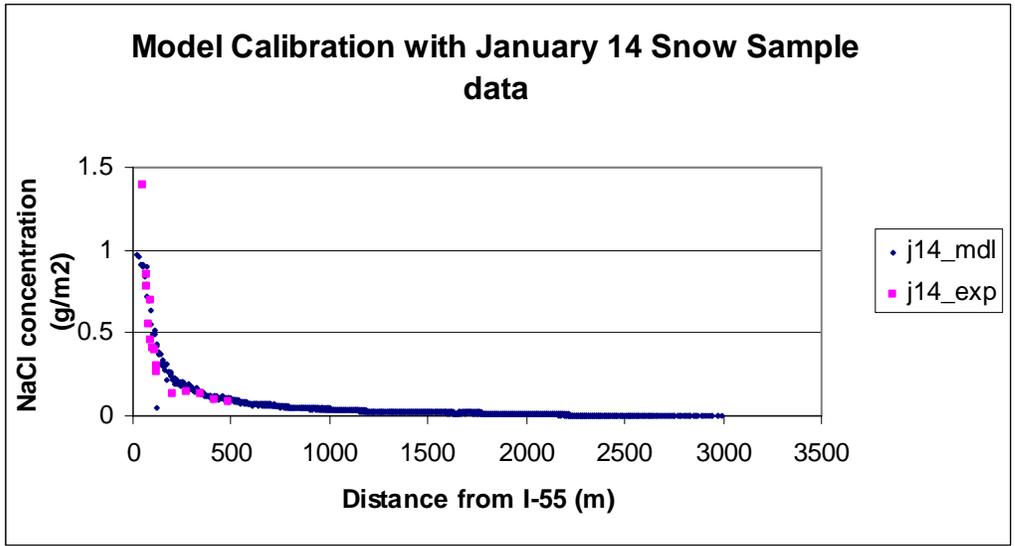


Figure 9. Comparison of model simulation and salt concentration measurements for snow samples collected on January 14, 1998.

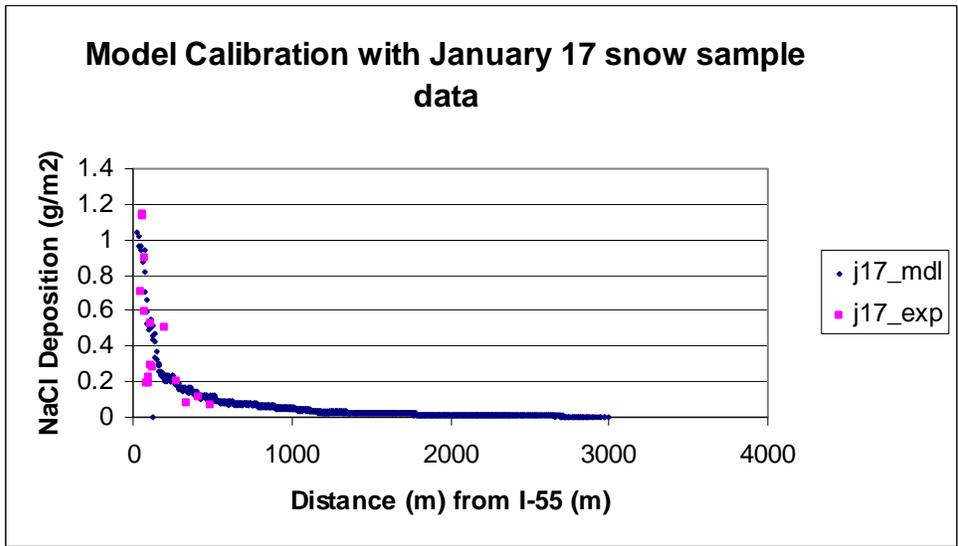


Figure 10. Comparison of model simulation and salt concentration measurements for snow samples collected on January 17, 1998.

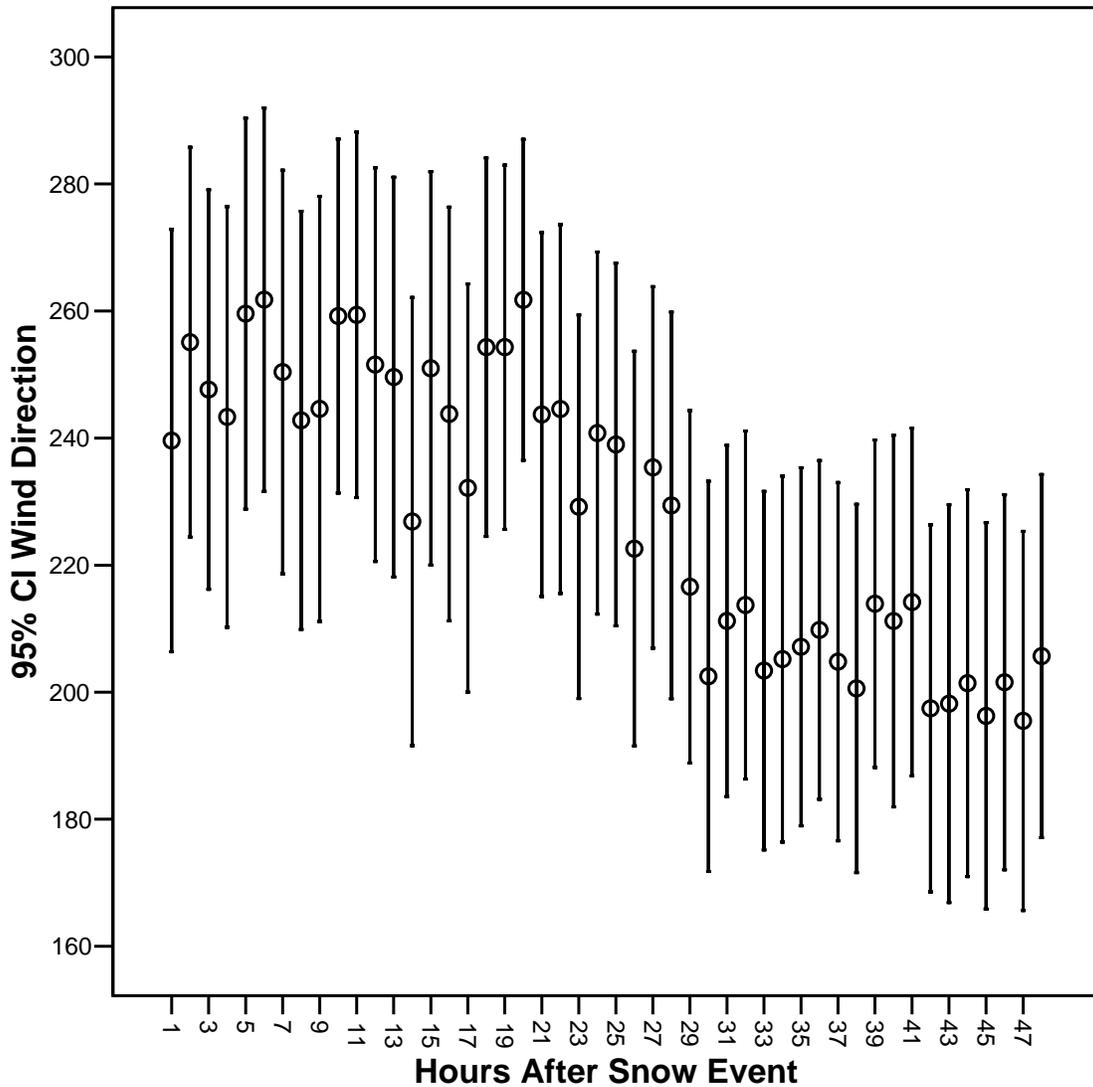


Figure 11. Average of measured wind direction following end of snowfall for the 51 snow events during the winters of events for 51 cases

### Salt Deposition (g per sq m)

<VALUE>

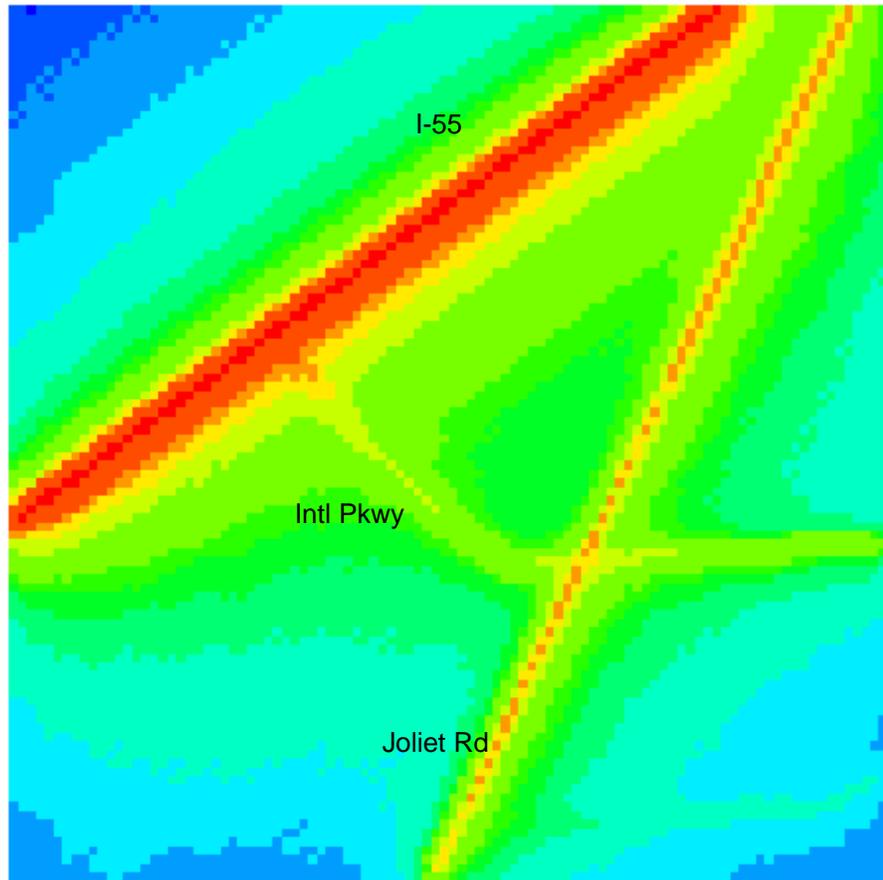
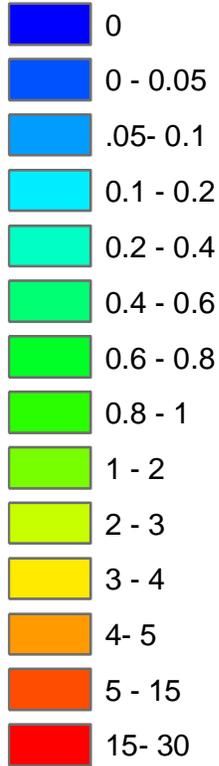


Figure 12. Example Model Output Showing Simulated Annual Salt Deposition Near the Intersection of Joliet Road and I-55.

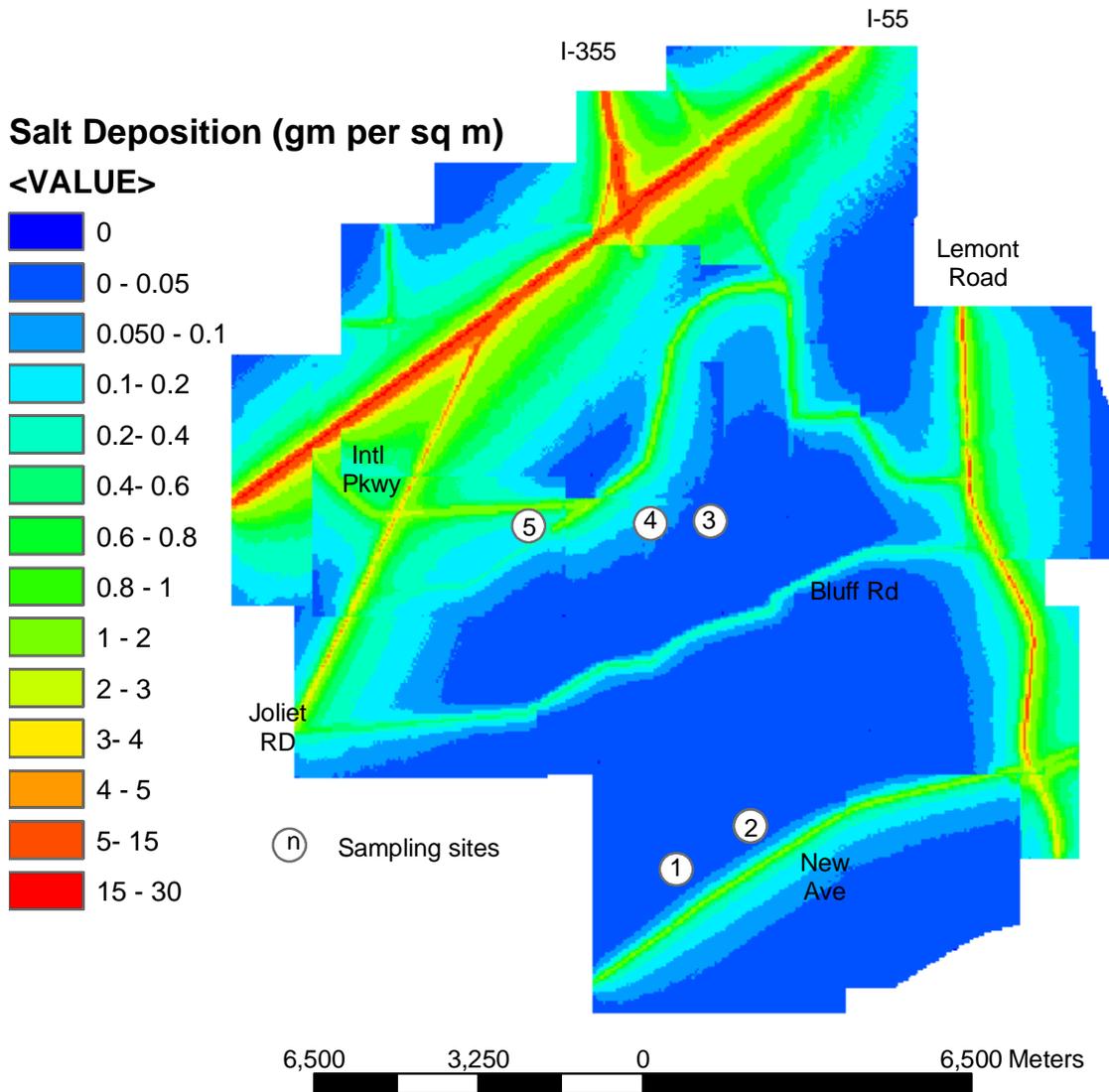


Figure 13. Model Simulations of Salt Deposition Near the Sampling Sites

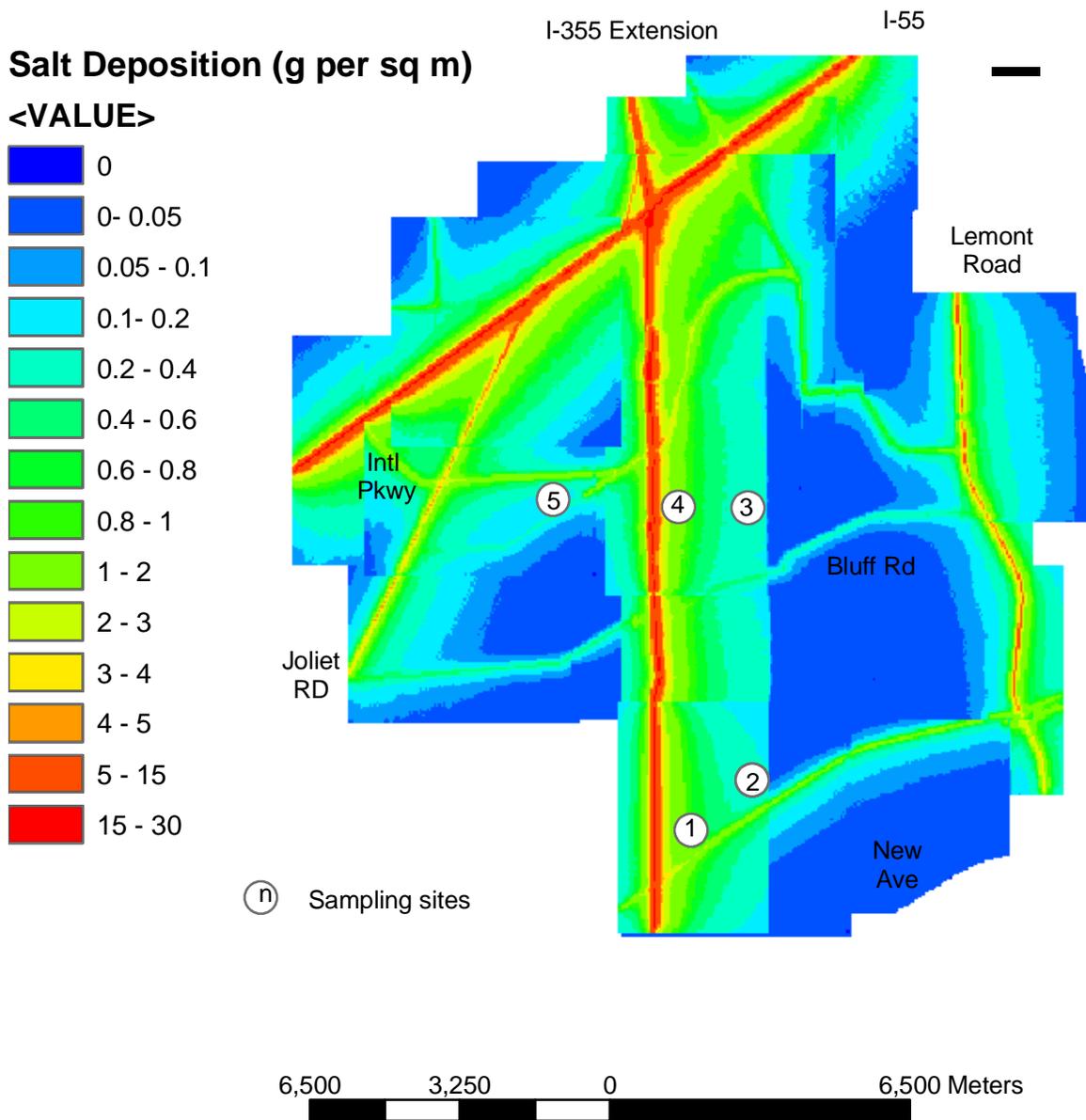


Figure 14. Model projection of salt deposition in study area after construction of FAP 340.

## **Appendix A.**

### ***Model User Guide***

#### ***Background Information***

The roadsalt model described here is designed to compute the annual salt deposition over a 25 km<sup>2</sup> area selected from a supplied roadway map. The supplied map is intended to be projected using Graphical Information Systems (GIS) tools, and the desired area for computation of salt deposition visually selected. The process involves using GIS tools to produce suitable input files and submitting the files to a processor developed within GIS to compute the deposition patterns. This User Guide describes the essential aspects of preparing model inputs, running the model, and displaying results.

The model is a Visual Basic for Applications (VBA) program. The input file is constructed using procedures in version 8.3 of ArcMap, ArcCatalog, and ArcToolbox, and the VBA model is launched from inside ArcMap. The Basic algorithm used in the model for describing annual salt emissions from a treated roadway is described in the final report for the project. The essential process describes the salt deposition from a treated road segment as a function of distance and direction from the treated segment. The algorithm is implemented in the model in terms of an input file or “kernel” that uses a model domain 5 km on a side broken into 10,000 cells. The domain is thus composed of 100 x 100 or 10,000 cells each representing a distance of 50 meters on a side. The kernel file represents the deposition pattern from a target cell located in the center of the domain at coordinates (50,50) containing a unit length (1 meter) of road. A roadway file giving the location and traffic volume of the roadway network for which deposition is required must be provided in an input file. During the computation, the kernel is numerically translated to be centered at each cell of the domain where a roadway segment occurs, and taking into account the length of road and traffic volume for the corresponding road segment, the deposition to each other cell in the domain is computed. The final computed deposition pattern is the accumulation of the deposition at each cell from the different roadway segments.

Preparation of the roadway input file is accomplished by user manipulation of two input files: the model grid and a roadway network file. Accompanying the model is a shapefile ‘il-roads’ supplied courtesy of IDOT. This file of Illinois roads is in a NAD 1983, State Plane West coordinate system commonly used by IDOT. Another file supplied with the model is the domain grid file ‘grid’, which was developed as a coverage in ArcInfo and converted to a shape file and projected into the same coordinate system as ‘il-roads’. The il-roads file can be updated to newer versions as more complete information becomes available. Besides the projection the file must contain information on road length and traffic volume.

#### ***Instructions***

The basic input files for the model are the roadway network in a file named “Il-roads” and the model “grid” file. The provided CD contains a geodatabase with multiple copies of the il-roads and grid files in the NAD 1983 projection. In addition an ArcMap document “Roadsalt.mxd” is included where the grid and road files can be projected. When initially opened in ArcMap, il-roads and grid files can be projected onto a visual map. The first step then is to move the grid over the roads for which the salt deposition is desired. The road and grid are then intersected using an ArcMap utility. The intersected file is then joined back to the original grid, a field is added and computed as the product of the road segment length and

traffic volume, and the file is converted to a raster format. With this input the model can be run as a VBA program from within ArcMap. The model creates an output raster file of the annual salt deposition pattern inside the 5 km x 5 km model domain. Using additional ArcMap utilities the output file can be transformed to appropriate representation for visualization and tables created giving deposition amounts.

The procedures requiring description are 1) the grid translation, 2) the intersection, 3) the join back to grid, 4) create and compute new variable, 5) make input raster, 6) the model run, and 7) post processing. A description of these procedures is given below.

**Grid Translation** Open the supplied ArcMap document 'roadsalt\_model.mxd' or start a new ArcMap document and add the 'il\_roads' and 'grid' files from the geodatabase. It is assumed that the ArcMap projection shows the two files in a 'Layers' or table of contents (TOC) section on the screen and that the rest of the screen is available to show the projected maps. If the box beside 'il\_roads' is checked inside the 'Layers' section in ArcMap a projection should appear. Selecting 'il\_roads' by clicking the right mouse button (right clicking il\_roads) and selecting the option 'zoom to layer' from the menu that appears will produce a map of the roads in the whole state, the shape of the state being clearly outlined. Selecting only 'grid' in the layers menu and zooming to layer shows the grid at a visible resolution. If from this point the box beside il\_roads is checked so as to make the road map again visible only a small portion of roads in the Chicago area will be displayed as the default location of the grid is in the Chicago area. If a layer is highlighted in the TOC right clicking on it and selecting "zoom to layer" usually shows the figure in a reasonable projection. Using these procedures produce an appropriate projection showing the grid and the place it is to be moved.

The grid will be translated on the map using the ArcMap 'Editor' utility. Before starting the Editor choose 'Selection' from the main menu and choose 'Set Selectable Layers' from the dropdown box. Check 'grid' and uncheck 'il\_roads'. This assures that the il\_roads file will not be disturbed by moving the grid over it. Once the projection is satisfactory and the grid is the only selectable layer, start the 'Editor' which is located on the Editor toolbar. If the editor toolbar is not visible it can be selected from the list produced by selecting 'view' from the main toolbar then selecting 'toolbar'. Once the editor is started choose grid as the target from the editor toolbar. Also from the editor toolbar choose 'select features using area' from the task option. The grid, projected on top of the roads, can now be moved. It is selected by depressing the left mouse button and dragging it over the grid. The area shown to be selected must include all of the grid and none of the roads outside the grid. Once selected the color of the grid changes. In order to move the grid, place the mouse cursor on the grid and hold down the left mouse button. The color of the grid will again change. Keeping the left mouse button depressed drag the cursor, which will now have the shape of the grid, to the desired location on the il-roads background and release the button at the desired grid location. With multiple iterations of the move and with zoom adjustment the grid can be placed precisely.

Once the grid is moved to the desired location, again select Editor and choose 'stop editing'. A prompt will appear asking whether to save the edits. Answering to the affirmative will change the grid file. At this stage the new grid file should be saved to the geodatabase.

Grid translation summary:

Selection – set selectable layers to only the grid file

Start editor

Target-grid file, not roads file

Tasks – select using area

-Move grid to desired location on roads map

Editor – stop editing

Save changes – Upon saving the edits the grid file is now changed.

**Intersection** Once the grid is placed at the desired position on the il\_roads projection, the grid and roads should be intersected using the Geoprocessing Wizard. The order of the intersection is roads first and grid second. If the output file is here defined with reference to the geodatabase, the fields of the intersection fields will be automatically calculated. Naming the intersection “roads\_x\_grid”, its projection should show only that portion of il\_roads that are inside the 5 km grid. The road\_x\_grid table will contain entries for the length of road in each cell. The intersection is accomplished by opening the Geoprocessing wizard and choosing the intersection option. The subsequent dialog box asks for the file names. Save the output as a personal database feature class. The intersection may take a few minutes. The working dialog box should indicate that the intersection is being processed.

Tools – GeoProcessing Wizard – intersect

Road first - grid second

Save output as type personal database feature class

Example name is road\_x\_grid

### **Join**

Two new calculated fields in the intersection file will be used: “Road\_Gri\_1” which is the number the cells from top right by row and “Shape\_Length” which is the length of road in each grid where a road segment occurs. These fields should already be calculated as a geodatabase feature. To start the join, right click the grid listing in the layer menu and select ‘joins and relates’ from the dialog box. The join type is ‘join attributes from a table’. The field on which the join is based is road\_Gri\_1 and the table to join to the layer is road\_x\_grid using the same field road\_Gri\_1. The join process is very fast.

### **Create and compute new variable**

To create the traffic volume variable save the joined files into a shape file, open its attribute table, and select options-Add field. Type the name for the new field and set its type as double. Now open the editor, start editing, and select the shape file for editing. Open the attribute table (it may still be open) and move to the newly defined field. Right click on the field name and choose calculate values. A calculation box comes up ready to calculate values for the new field. Click the field name AADT (traffic volume) then the \* (multiply) operator and finally the field Shape\_Le\_5 (this field now contains the road segment lengths). Now select ok and the field should be calculated in a few seconds. Finally select stop editing and answer yes to the prompt to save edits. Saving edits will place the newly create file and its new values into the starting shape file (which is the join of the grid and the road-grid intersection). It will also prompt if you want to have it on screen.

### **Make input raster**

The final step in file preparation is to make a raster file that the VBA code can read from the file just edited. Highlight the file in the table of contents and select “spatial analyst”.

From inside the spatial analyst select convert-files to raster. The dialog box should show the file being converted. Select the newly created field as the 'field' entry, change cell size to 50, pick a name and path for the file, and save it. The name and path of this file will later need to be entered when the VB code is started.

### **Running VB code**

At this point the raster input file should have been created. From ArcMap select tools-Macro-visual basic editor. From inside the editor choose view-project explorer if the project explorer is not already open. From the project explorer dialog select the project "roadsalt\_model" and click forms - salt\_deposition. Either the form or code should come up. You can select between them in the project explorer dialog. To run the model, select the form option, then click the 'run' symbol in the toolbar. Select 'start ' from the model dialog. The model dialog box should request first the path then the file name of the input raster. It will then ask for the path and name of the output raster file. Finally it will ask for the path of the kernel file called "kern\_out.txt". When the code starts the model form box should be visible with the ArcMap view instead of the Visual Basic Editor visible in the background. When the run finishes the form box will appear again only back in the Visual Basic Editor.

If the program has trouble it will usually be a file name or path problem. Once the job is ready to submit again make sure the output file was not created in the previous attempt. If it was created either delete it or specify a new file name for the output raster. Deleting the file can sometimes be difficult. If done from ArcCatalog it may prompt that the file is in use. If so, save and close ArcMap. Sometimes even that does not work and both ArcMap and ArcCatalog will have to be closed and the file (which at this point appears as a directory containing files) can be deleted with Windows Explorer.

If it becomes necessary or desirable to change the default input and output paths and filenames in the model, this can be easily done by editing the code. From the project explorer select code. The prompt for the filenames etc. is at the top of the code. This must be done carefully including quotation marks around the filenames and paths as they were originally. It would be a good idea to export the new VB file under a new name.

### **Post Processing**

Production of maps to display model results depends to a large degree on their use, and considerable flexibility is available from the general GIS procedures. A useful procedure is to add colors corresponding to concentration levels to a model result. First different color schemes must be added to ArcMap. From the ArcMap menu Tools – Styles – Style Manager different color schemes can be added from a wide selection. Then from the Layers menu for the projection to which the color scheme is to be applied, right click the projection and select "Properties". The "Symbology" selection of the Layer Properties menu provides an option for selecting a color ramp from those added with the Style Manager. Also, from Symbology selection the number of classes to be viewed can be specified. The "Classify" option of the Symbology selection provides a means to specify the concentration range for each color class. Finally in the Symbology selection the label value, which would ordinarily correspond to the values set in "Classify" can be set. Working with these procedures can produce a suitable graphic representation. The sample results included with the electronic version of the model, when loaded into ArcMap, should have styles selected and the settings in the Symbology section should correspond to the maps produced in Figures 12-14 of this report. Once a map is produced it can be saved as a layer file (filename.lyr) by right clicking the

listing from the ArcMap layers menu and choosing the “Save as layer file...” option. The intricacies of the color map classification and concentration settings are preserved in the layer file representation.

## **Appendix B**

# **Atmospheric Emission and Deposition of Deicing Salt Applied to Highways in the Chicago Area**

Paper 698

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### **ABSTRACT**

This paper reviews wet and dry deposition of aerosol sodium measured from 1980 to 1998 at a suburban site at Argonne National Laboratory, about 40 kilometers southwest of downtown Chicago. The deposition samples were obtained with a wet/dry deposition sampler operated as part of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN)<sup>1</sup>. Dry deposition data were for eight-week samples and wet deposition data were for weekly samples. Air quality measurements made with dichotomous and high-volume aerosol samplers at five sites 1-4 km southwest of the Argonne site showed that most of the sodium aerosol mass was in particles greater than 10  $\mu\text{m}$  in diameter. Thus one can expect that the simple bucket measurement of dry deposition is a valid sampling approach for the relatively large sodium particles. Dry and wet depositions of the road salt aerosol in the November to April period were about 103 and 14  $\text{mg}/\text{m}^2$ , respectively. For the May to October period of warm months, dry and wet depositions of sodium were both about 24  $\text{mg}/\text{m}^2$ . Road deicing salt is the primary source of the increased sodium in the cold months. The daily application rate of road deicing salt is known for the period 1987 to 2002 for 22 individual areas comprising the Chicago area. A linear correlation analysis of dry deposition of sodium versus road salt applied showed that about 75% of the variance of sodium at the suburban Chicago site could be explained with the linear model. Winter total (wet plus dry) deposition of sodium greatly exceeded summer total deposition, and for the winter months dry deposition greatly exceeded wet deposition.

### **INTRODUCTION**

This paper addresses the issue of deposition of deicing salt from the atmosphere to the ground. Results from 18 years of deposition measurements at a suburban site about 40 km southwest of downtown Chicago will be discussed. The sodium deposition record will be compared to the deicing salt applied to state roads and expressways in the area.

Many deicing substances are applied to roads but due to its relatively low cost, sodium chloride is the dominant deicing material used in the USA<sup>2</sup>. For this paper the term salt or deicing salt refers to sodium chloride. The sodium chloride used in the United States

as a highway deicing salt rose steadily from about one million tons per year in the early 1950's to the fluctuating level of 8-11 million tons per year from 1968 to 1992<sup>3</sup> and then to the fluctuating level of 15-20 million tons per year from 1993 to 2001 (Salt Institute, <http://www.saltinstitute.org/33.html#hwysale>).

Salt and other highway deicers has been the subject of hundreds of research papers and reports in the last 50 years. Studies of the impacts on motor vehicles, highway infrastructure, human health, soils, surface water, groundwater, vegetation and biological communities (in soils and surface waters) have been reviewed<sup>4,5,6</sup>. The field studies, where impacts of deicing salt have been examined, have generally focused on distances within a couple hundred meters of the road emission source. In these impact studies salt deposition rates were sometimes quantified, using bulk collectors (containers continuously exposed to the atmosphere to collect precipitation (wet deposition) and dry deposition of salt aerosols) or by washing salt deposits that had accumulated on vegetative surfaces or by measuring salt levels in soil samples at various distances from the road sources. Occasionally road deicing salt has been identified as one of many emission sources that influence atmospheric studies of air quality, precipitation quality, or wet and dry deposition. Gélinas and Schmit<sup>7</sup> carried out two years of bulk sampling at sites in Montreal (2 sites) and in a small watershed about 30 kilometers away from the city (14 sites). Site location and seasonality of the data led the authors to identify road salt as one of the sources influencing their bulk deposition data. Dasch and Cadle<sup>8,9</sup> collected dry deposition and wet deposition samples for two years at an urban site in the Detroit area and at a rural site north of Detroit. Here the patterns in sodium deposition were attributed to road salt emissions. Two bulk sampling studies have been done in the Chicago area to quantify the airborne transport and deposition of road salt. Kelsey and Hootman<sup>10</sup> measured bulk deposition at the Morton Arboretum for two winters, at distances from 15 meters to 1018 meters from nearby expressways. A winter 1999/2000 study near South Elgin, IL included measurements of bulk deposition for one winter at 19 sites at distances from 8 to 285 meters from the road<sup>11</sup>. Williams and Stensland<sup>12,13</sup> described an ongoing air quality and deposition study investigating road salt levels at five sites 40 kilometers southwest of downtown Chicago, near Lemont, IL. This last study, completing field measurements in April, 2004, is the first to include ambient aerosol sampling equipment to determine airborne concentrations and size distributions for road deicing salt ambient aerosols.

## **METHODS**

### **Conceptual Model for Emission, Transport and Deposition of Deicing Salt**

The tires of vehicles traveling on salted roads inject salt particles into the air. The emission process will be referred to as wet emission when the tires are thrusting salt droplets into the air from a wet/slushy road and will be referred to as dry emission when the tires are thrusting solid salt particles into the air from a dry road. In either case, turbulence caused by the moving vehicles will rapidly mix the emitted particles into a vertical column behind the vehicles and the wind will transport the particles away from the road. The particles will be deposited onto the ground by wet deposition and by dry deposition. The wet deposition process refers to precipitation (i.e. raindrops and

snowflakes) falling from a cloud containing a background level of salt (from in-cloud scavenging of particles) that will be increased as the precipitation falls through a deicing salt plume (i.e. below-cloud scavenging of particles). Dry deposition of deicing salt particles refers to the gravitation and inertial impaction processes that result in the salt droplets and solid salt particles being removed from the atmosphere, during periods when precipitation is not occurring. Impaction processes result in deicing salt being deposited onto vegetation while gravitational settling results in deicing salt being deposited into sampling vessels or onto the ground that is sometimes covered by a snow pack. Large traffic generated salt drops, referred to as splash drops, fall out close to the road. Arbitrarily the strip of ground from the edge of the road out to 15 meters will be referred to as the splash deposition zone. Smaller drops, referred to as droplets or spray, are deposited in the spray and solid particle deposition zone, which begins 15 meters from the road.

Particles carried by the wind and settling out due to gravity, in a non-turbulent atmosphere, would follow smooth parabolic trajectories. Fluctuating wind speed and direction, i.e. turbulence, is generally present in the atmosphere, and results in some salt particles being lofted higher in the air than particles following a simple gravitational settling trajectory and thus allows some fraction of the emitted deicing particles to travel long distances from the emission sources. Likewise turbulence will deflect some particles downward, toward the ground, causing them to dry deposit at shorter distances than would be predicted by gravitational settling trajectories. Results from gravitational settling calculations, in an atmosphere without turbulent eddies, for particles with a density of one, are shown in Table 1, for three wind speeds, three emission heights, and three distances from the road. For particles less than 100  $\mu\text{m}$  in diameter the terminal settling velocities were calculated by assuming spherical particles with negligible slip.<sup>14</sup> For particles greater than or equal to 100  $\mu\text{m}$ , the empirically determined terminal settling velocities of Gunn and Kinzer<sup>15</sup> were used. Beard and Pruppacher<sup>16</sup> verified the accuracy of the Gunn and Kinzer results. To illustrate the results in Table 1 consider a scenario where the wind speed is 11.2 mps and the emission height is 200 centimeters, as representative for deicing salt particles being emitted from a road. Particles larger than 238  $\mu\text{m}$  would be deposited within 15 meters of the edge of the road; particles smaller than 238  $\mu\text{m}$  would travel more than 15 meters from the road; particles smaller than 60  $\mu\text{m}$  would travel more than 100 meters from the road; and particles smaller than 26  $\mu\text{m}$  would travel more than 500 meters from the road.

It is useful to consider the implication of the results in Table 1 for the sampling of airborne particles. Suppose a road being studied is emitting a salt spray with particle diameters in the range 5-500  $\mu\text{m}$ . Also suppose a high volume air sampler with a known particle size cutoff of about 40  $\mu\text{m}$  is being used to sample the airborne deicing particles. For simplicity assume that the 40  $\mu\text{m}$  cutoff for the high volume sampler is independent of wind speed, which is not true. Wherever the deicing particles sizes shown in Table 1 exceed 40, there will be incomplete sampling of the airborne aerosol by the high volume sampler. So at a downwind distance of 500 meters, the incomplete sampling would occur only when the H is 600 cm and V is greater than about 11 mps. At a downwind distance of 100 meters, the sampling would begin to be incomplete for the two higher wind speeds

with all three emissions heights. At 15 meters the high volume sampling would be incomplete for most conditions shown in Table 1.

What is the size of the road salt aerosol in the immediate vicinity of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) site operated by Argonne National Laboratory. This site is also known as IL19. The major source, I-55, is 1.9 km or more from the IL19 site, depending on wind direction. Dichotomous aerosol samplers and standard high volume air samplers have been operated for several years at five sites 1-4 kilometers southwest of IL19<sup>13,17</sup> With the dichotomous sampler, fine ( $D < 2.5 \mu\text{m}$ ) aerosols are collected on one filter and course ( $2.5 < D < 10 \mu\text{m}$ ) aerosols on a second filter. As was already noted, the high volume sampler collects aerosols up to about  $40 \mu\text{m}$  on a filter. Results from measurements with these devices are shown in Table 2 for winter sampling periods during which snowfall occurred sometime during the sampling period. The mean values for sodium show that very little sodium is found in the fine mode, and the course and super-course modes have about 18% and 78% of the sodium, respectively. No air quality measurements were made to determine if the road salt aerosol at these 5 sites has a significant component greater than  $40 \mu\text{m}$ . These data should be representative of the IL19 location and tend to be consistent with simple calculations in Table 1.

Table 1. Summary of trajectory calculations for deicing salt splash and spray particles. Listed in the table are the diameters of the particles, in  $\mu\text{m}$ , that settle to the ground due to gravity, at downwind distances of 15, 100, and 500 meters from the road. Results for three wind speeds (V), three emission heights (H) are shown.

		Downwind Distance (Meters)	H = 100 cm	H = 200 cm
H = 600 cm				
V = 1.3 mps (2.9 mph)	500 m	-----	6-----	10-----
	100 m	-----	14-----	20-----
	15 m	-----	37-----	51-----
Road Location-----				
-----				
V = 11.2 mps	500 m	-----	19-----	26-----
	15 m	-----	46-----	

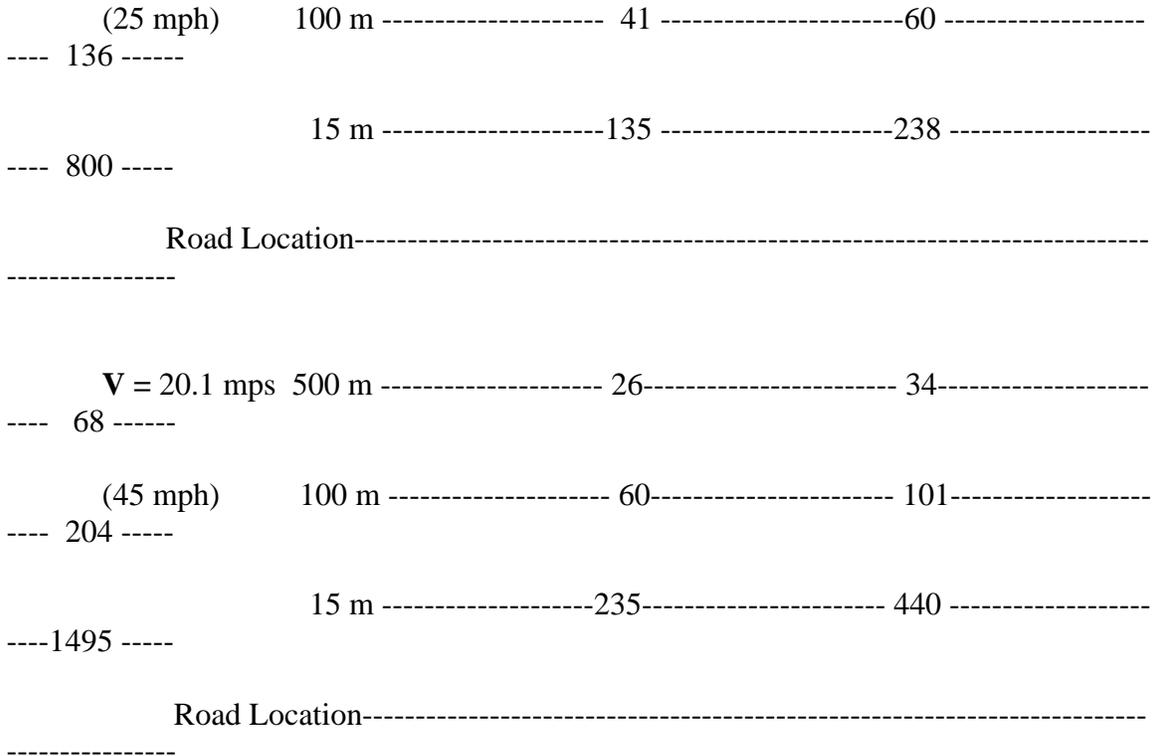


Table 2. Sodium average size distribution for sampling periods when snow occurred, for five sites a few kilometers west of the IL19 site.

Size Range ( $\mu\text{m}$ )	N	Mean ( $\text{ug}/\text{m}^3$ )	Std. Error ( $\text{ug}/\text{m}^3$ )	25%	50% Percentiles	75%
10 to ~40	16	1.06	.20	.54	.71	1.34
2.5 to 10	24	.25	.05	.06	.20	.39
0 to 2.5	25	.05	.01	.02	.05	.06

Dry deposition can be calculated by multiplying the aerosol concentration by the dry deposition velocity. With the data in Table 2 showing that most of the aerosol mass is in particles sizes greater than 10  $\mu\text{m}$ , one can assume that the dry deposition velocity for the road salt aerosols is equal, to a good approximation, to the terminal settling velocity. To implement this dry deposition measurement approach effectively one would need to know the aerosol concentration as a function of particle size, where the size classes should probably not be more than 3-4  $\mu\text{m}$  in width. Accurate sampling of the road salt particle size distribution is difficult because the large momentum of the particles means it is difficult to keep them from impacting onto surfaces of the sampler other than the actual collection media. The sampling difficulty for large particles was dramatically demonstrated in a “shootout” type of equipment intercomparison by May et al.<sup>18</sup> in which 17 devices were compared in collecting 20, 30, and 50  $\mu\text{m}$  test aerosols during about 15 tests under field conditions. The intercomparison demonstrated that large sampling biases can occur when attempting to sample large particles. The greatest bias occurred with a

modified Anderson cascade impactor that only collected 4%, 7% and 12% of the 50, 30, and 20  $\mu\text{m}$  test aerosols, respectively. Gélinas et al.<sup>19</sup> used the bulk method to determine total deposition but commented “Dry and bulk deposition fluxes are more reliably determined using airborne concentrations along with a dry deposition velocity model.” We agree in principal with this statement but are not aware of a study for large sized aerosols like road salt aerosols that demonstrates the practicality of the statement. Considering the challenges in properly sampling large particles and the fact that multiple sites are usually necessary, it appears that resources will seldom be available to determine deposition over long periods of time without resorting to something like bulk or dry-side/wet-side sampling with surrogate surfaces.

An alternative method to determine dry deposition is to place clean sampling containers in the ambient air for sampling intervals of 1 to 30 days. This approach has been very controversial but everyone recognizes that the method is most likely to be useful in providing deposition estimates for very large sized aerosols, which includes the road salt aerosol according to the data in Table 2. A second concern, the influence of contaminants in biasing the bulk sampling results, will be discussed below.

In the winter period 1999/2000 a field study in South Elgin, IL<sup>11</sup> biweekly bulk sampling was done to look for road salt deposition decreasing with distance from a highway with a traffic flow of about 12,000 vehicles per day (vpd). In one part of the experiment seven plastic sampling buckets were deployed at seven sites from 13 to 136 meters from the road. During each of seven two-week sampling periods when deicing salt was used on the highway, the deposition of sodium decreased with distance from the highway as was expected. The consistency of the data qualitatively supports the bulk sampling approach for wintertime road salt aerosols.

### **The IL19 Site**

This paper presents wet and dry deposition results for sodium from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN)<sup>1</sup> site operated by Argonne National Laboratory (ANL). Reports on methods and on QA/QC are available on the NADP/NTN web site. The NADP/NTN site operated by ANL, known as the IL19 site, is located about 40 km southwest of Navy Pier, which is on the edge of Lake Michigan near downtown Chicago. The deposition sampler is in the southwest portion of the ANL property in a grass-covered area and is far enough away from

obstacles such as buildings or trees to meet the NADP siting criteria (refer to photos at <http://nadp.sws.uiuc.edu/sites/siteinfo.asp?net=NTN&id=IL19>).<sup>20</sup> When the wind is perpendicular to the road the site is 1.9 km southeast/downwind of the I-55 salt source, and 0.8 km downwind of Lemont Road salt source with a west wind. Lemont Road is a heavily used 4-lane road with traffic typically moving at about 45 miles per hour. Traffic rates are about 145,000 vpd on I-55 and 15,000 on Lemont Road. The I-55 expressway is hypothesized to be the dominant sources of sodium deposition measured at IL19 in the cold season months, with smaller deposition components from Lemont Road and from a more general suburban background level.

The special features of the IL19 site that make it useful for looking at road deicing salt characteristics is the length of its dry deposition record, 18 years, and its suburban location. There are approximately 260 NADP sites in operation but since this primary objective of the network is to collect regionally representative data there are only a few sites in urban or suburban areas. Only IL19 and a site in the suburbs of Boston are (a) in the immediate vicinity where large amounts of road deicing materials are applied and (b) have long dry deposition records, making these sites potentially useful to look for temporal signals of airborne road salt. However a road deicing salt signal in data for the Boston area site would be difficult to identify since the sea salt aerosol component will be very large at this location compared to the IL19 location.

### **Dry-side Bucket Procedures Used in NADP/NTN**

NADP/NTN sites collect wet deposition samples with a wet/dry sampler, the model 301 Aerochem Metrics wet/dry sampler often referred to simply as the “Aerochem sampler”<sup>21</sup> The Aerochem sampler has been the primary sampler used by USA researchers and monitoring networks since the early 1980s. The Aerochem holds two 14 liter plastic buckets, with a lid covering one of the buckets at all times. An Aerochem sensor detects periods of precipitation, activating a motor that moves the lid from one bucket to the other bucket, thereby exposing the wet-side bucket to the falling rain and snow. During non-precipitating periods the lid covers the wet-side bucket, leaving the dry-side bucket exposed. When precipitation leads to the exposure of the wet-side bucket, a heater in the grid sensor is activated to dry out the sensor quickly after the end of precipitation, thus minimizing how long the wet-side bucket is exposed to dry deposition and thus minimizing the collection of dryfall in the wetfall bucket.

The amount of precipitation is measured at the NADP/NTN sites with a Belfort B5-780 recording raingage. The Belfort gage is a spring-scale device with a recording chart drum. An ink pen on the raingage records, on the chart mounted on the rotating drum, the amount and timing of precipitation. A second ink pen on the raingage records the duration of the exposure of the wet-side buckets on the nearby Aerochem. Besides determining which wet-side samples are the desired wet-only samples, the raingage also provides a standard precipitation value that can be compared to the volume of rain and snow captured in the wet-side sample. On average, for weeks without equipment failure and with only rainfall occurring, the rainfall captured by the Aerochem is a few percent less than the rainfall measured by the Belfort raingage. The Aerochem sensor does not

work as well in detecting snowfall, especially “dry” snow. Thus in the winter it is common to find some snowfall in the dry-side buckets.

For dry-side samples, 250 mL of very pure water was added to the buckets at the Central Analytical Laboratory (CAL). After soaking for at least 24 hours the 250 mL aliquot was filtered and analyzed by flame atomic absorption spectrometry. Annual QA reports for the CAL are available (e.g. see Rothert, 2002<sup>22</sup>). If some precipitation was in the dry-side bucket, the same extraction procedure was followed and the ion concentrations were corrected to those for a 250 mL sample.

From 1978 to 1984 dry-side buckets at NADP/NTN sites were changed either bimonthly or every eight weeks and analyzed by the CAL. From 1984 to 1998 it was optional for sites to collect the dry-side samples and only a small percentage of the sites including those in IL and NC, continued to do the dry-side sampling. Since 1998 no dry-side samples have been collected by NADP/NTN.

In early 1980's a debate was occurring within NADP/NTN as to whether the dry-side bucket data provided credible and useful data for the various analytes. At that time the primary interest was in determining deposition of sulfur and nitrogen species. To determine the true dry deposition of something like sulfur it was argued that one needed to measure separately the deposition of gaseous species (i.e. sulfur dioxide), fine sulfate particles ( $D < 2.5 \mu\text{m}$ ), coarse sulfate particles ( $2.5 < D < 10 \mu\text{m}$ ) and super-coarse particles ( $D > 10 \mu\text{m}$ ), and that one should not assume that sulfate ion measured in dry-side bucket extractions was likely to represent or be well correlated with the sum of these four components. For crustal aerosols like calcium, which have a mass median diameter greater than  $5 \mu\text{m}$ , it was noted that the data from dry-side buckets was more representative.

A concern with the dry-side samples has been that they are quite frequently contaminated by bird droppings and thus might not be useful for scientific studies. Hicks (1986)<sup>23</sup> referred to the problem as a severe limitation but did not provide support for the conclusion. Feely et al. 1985<sup>24</sup> collected six dry-side samples daily for 23 summer days. Comparison of the visible contamination recorded for each sample and the ion concentrations in the extraction solutions led Feely et al.<sup>24</sup> to conclude that bird droppings were the only visible contaminant that seemed to significantly affect the chemical composition. Gatz et al.<sup>25</sup> examined the contamination comments recorded for the samples in the NADP dry-side data set and found that the median dry deposition values calculated for ions such as orthophosphate, potassium and ammonium were greatly affected by bird

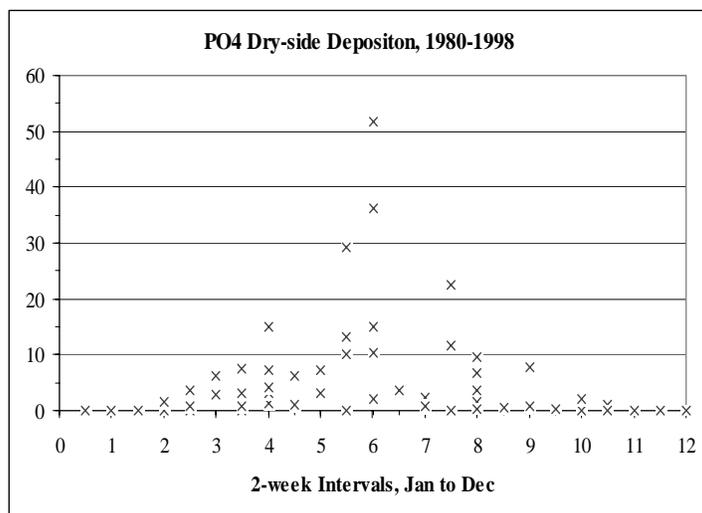


Figure 1. Dry-side deposition of orthophosphate at the NADP/NTN IL19 (Argonne) site from 1980-1998,  $\text{mg/m}^2/\text{month}$ .

droppings while the dry deposition calculations for ions such as sodium and calcium were not strongly affected. The contamination comments in the database were observations by laboratory personnel, and to some extent by field personnel, of the visible state of the dry-side bucket sample. Such comments could not be 100% reliable in identifying bird contaminated samples because shipment of the sample through the mail from site to CAL could stir the sample and thus sometimes mask the bird contamination. If one were going to attempt to examine patterns of dry deposition for an ion such as potassium through the use of such dry-side bucket data, one would certainly need to have a means to screen out those samples affected by contamination.

The orthophosphate ion concentration for the dry-side bucket samples from IL19 is shown in Figure 1. Elevated values are bunched in the warm months when birds numbers are greater and thus Figure 1 is as expected from the studies such as that of Gatz et al.<sup>26</sup> If such contamination were the major source of sodium in dry-side bucket samples, then one would find a sodium maximum in the warm months. Data in the next section

will show that such a summer peak does not occur.

Records of the amount of road salt applied daily in the 1987-2002 period were obtained from the Illinois Department of Transportation (IDOT). The records are for the IDOT District 1, the Chicago area, composed of Cook County and the 5 counties that share boundaries with Cook County. Twenty two winter operations teams, distributed across the area, apply most of the deicing salt for IDOT in District 1. IDOT salts all the major highways and expressways in the Chicago area. Other road salting occurs in the area, including salting of city streets by the city of Chicago and by other municipalities in the area. By comparing the salting activity by one team and with the salting activity of nearby teams it was observed that the salting patterns are well

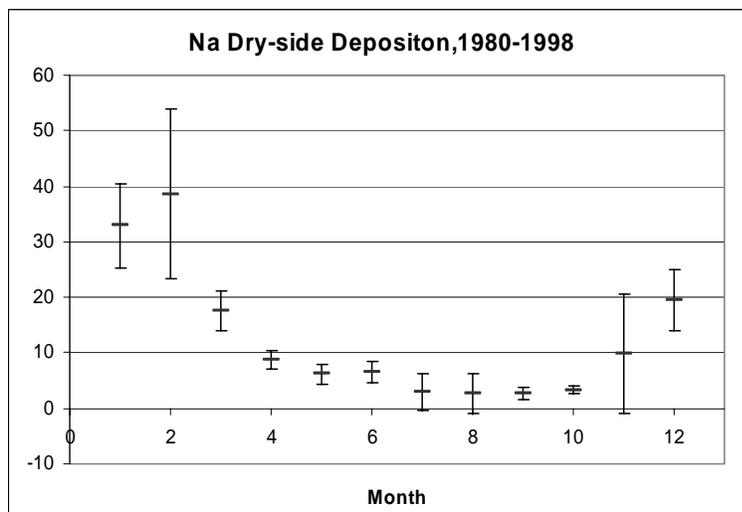


Figure 2. Median and 95% confidence interval of dry-side deposition of sodium at the NADP/NTN IL19 (Argonne) site from 1980-1998, mg/m<sup>2</sup>/month.

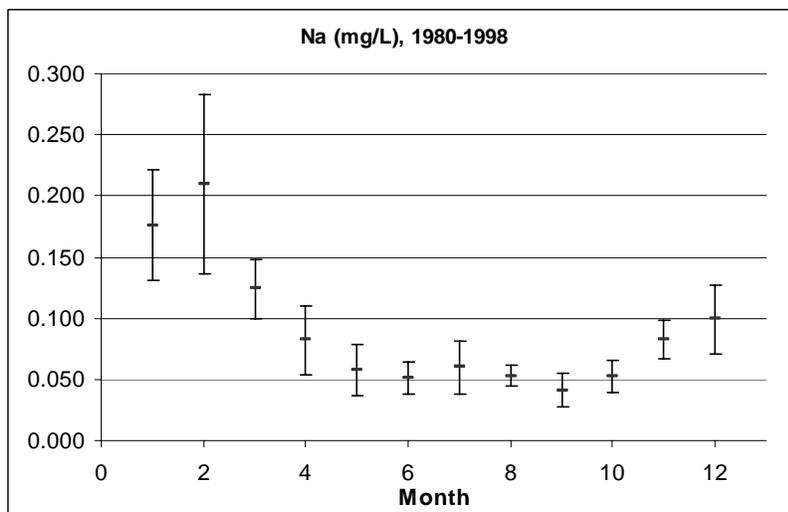


Figure 3. Median and 95% confidence interval of wet-side concentration of sodium at the NADP/NTN IL19 (Argonne) site from 1980-1998, mg/L.

correlated. That is, when a snow event occurs, all the IDOT teams and municipal teams will be simultaneously salting streets and roads in their jurisdiction.

## RESULTS and DISCUSSION

A total of 93 dry-side Aerochem samples were collected at the IL19 site from 1980 until early 1998 and were analyzed for a suite of ions including sodium. The mid-value of a sampling interval was used to assign the samples to months. The number of dry-side samples by month was 8,8,7,9,7,7,7,8,6,8,5, and 13 for January through December, respectively. Since wet-side samples are weekly in duration, the number of wet-side samples is approximately 8 times the number of dry-side samples. Wet-side deposition data are still being collected at IL19 but only data through 1998 were used. Median dry deposition values at IL19 for sodium are shown in Figure 2. Dry deposition is being operationally defined as that amount collected with the Aerochem dry-side sampling protocol. For each month the median and the +/- 95% confidence intervals<sup>26</sup> are shown.

The sodium concentration data for the wet-side Aerochem samples are shown in Figure 3 and the wet-side deposition data are shown in Figure 4. Wet deposition is calculated as the product of the weighted average ion concentration for a month times the precipitation amount for the month, where the later is determined with the Belfort precipitation gage at the NADP/NTN sites and not from the volume of water in the Aerochem buckets. The sodium wet deposition values have a maximum in April and a minimum in September, and are relatively uniform throughout the year when compared to the dry deposition pattern shown in Figure 2 where the maximum values are about 7-8 times the minimum values. Data from Figures 2 and 4 are combined to calculate the percent that dry deposition of sodium is of total deposition (wet plus dry), with the result being shown in Figure 5. For December-March, the months with the largest dry deposition, the percent dry deposition values are greater than 70%; for Jan-Feb, with the really large dry

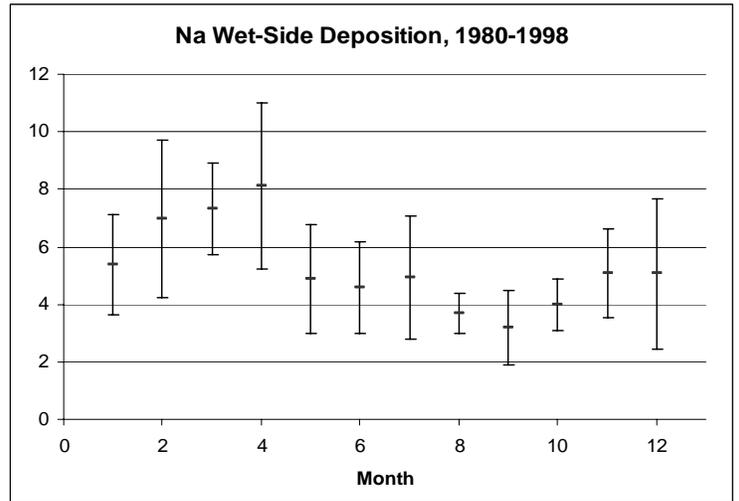


Figure 4. Median and 95% confidence interval of wet-side deposition of sodium at the NADP/NTN IL19 (Argonne) site from 1980-1998, mg/m<sup>2</sup> month

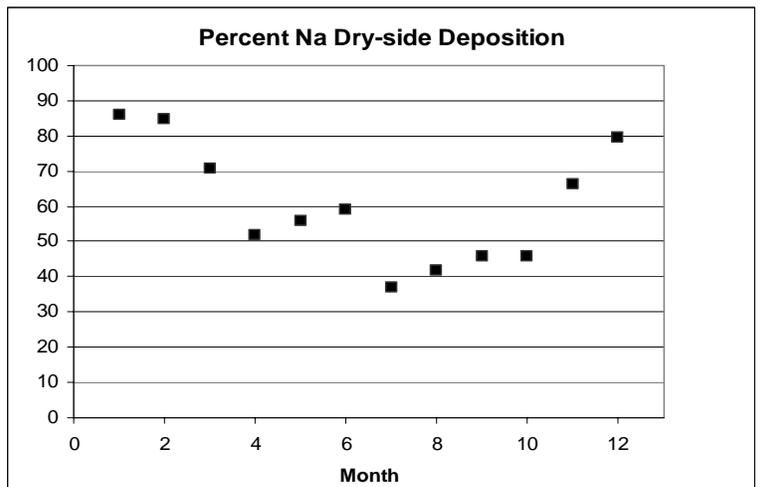


Figure 5. Percent dry-side deposition of sodium = 100 (dry-side deposition)/(dry-side deposition + wet-side deposition).

deposition values for sodium, the percent dry deposition is about 85%. The sodium deposition data in Figures 2 and 4 show that for the warm months May to October, when road salting is not occurring, the average wet and dry deposition values of sodium are both about  $4 \text{ mg/m}^2/\text{month}$ . Soil dust and sea salt aerosols transported into the Chicago area are likely sources for this warm period deposition of sodium, with industrial emissions and demolition and construction activities also being possible sources. We assume these background values are the same during the November to April cold months, when road salt aerosols are present. Subtracting these background levels from the cold month values in Figures 2 and 4, and summing over the six cold months gives an estimate for total salt aerosol deposition due to road salt of about  $103 \text{ mg/m}^2$  per winter for dry deposition and  $14 \text{ mg/m}^2$  per winter for wet deposition. For the six warm months, May – October, the dry and wet deposition values of sodium are both about  $24 \text{ mg/m}^2$ .

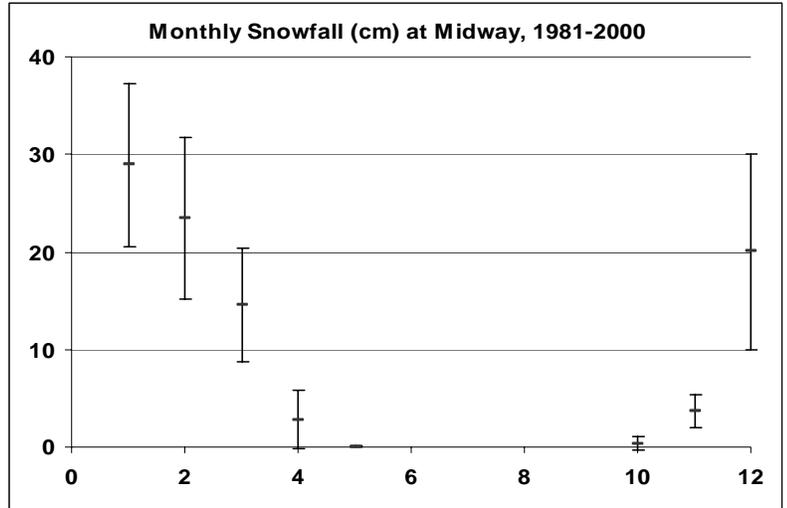


Figure 6. Median and 95% confidence interval of monthly snowfall (cm) at the National Weather Service Cooperative Station at Chicago Midway Airport for the period 1981 – 2000.

The monthly snowfall data for 1980-1998 for the closest National Weather Service Cooperative Observer Site, near Midway Airport, are summarized in Figure 6. The monthly precipitation data (rain plus snow) for 1980-1998, for the raingage at IL19, are shown in Figure 7. The largest precipitation values are in the warm months and the largest snowfall values are in January and February. It is reasonable to hypothesize that the quantity of deicing salt applied to roads in the area will have a temporal pattern similar to that shown in Figure 6 for monthly snowfall. Thus the road salt emissions should be the largest in January and February, the two snowiest months, and the dry deposition should also be the largest in January and February, as is observed in Figure 2.

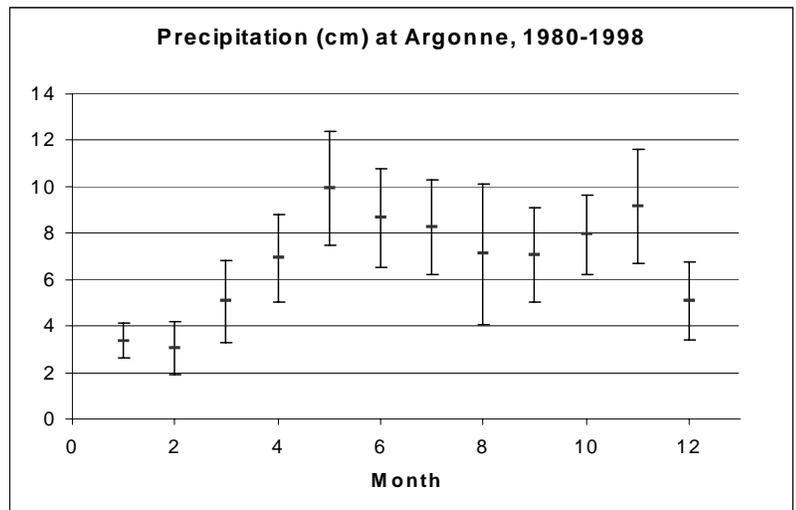


Figure 7. Median and 95% confidence interval of monthly precipitation (cm) at the National Weather Service Cooperative Station at Chicago Midway Airport for the period 1981 – 2000.

IDOT Team 135 applies the salt in the vicinity of site IL19 (on I-55 and within about 20 km of site IL19).

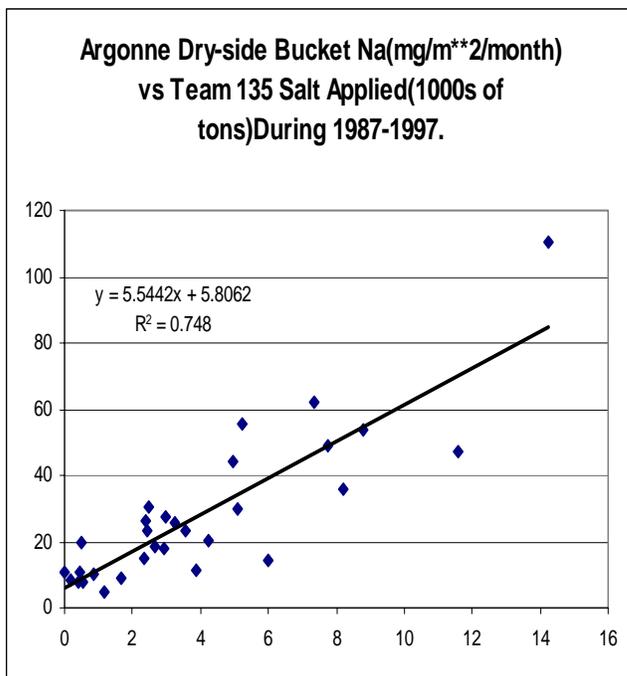


Figure 8. NADP/NTN IL19 site dry-side deposition of sodium versus deicing salt applied by team 135 from 1987-1998.

The Team 135 daily salt applied values were summed into time periods corresponding to the exposure periods (about 8 weeks) of the IL19 dry-side bucket samples. A scatter plot of Team 135 salt applied versus the corresponding sodium deposition determined from the dry-side buckets is shown in Figure 8. Dry-side samples that included exposure only during the months of May through October, when no deicing salt was applied, are not included. From Figure 8 it is seen that the dry deposition of wintertime sodium particles at the IL19 site is well predicted by the road deicing salt applied in the Team 135 area, with a linear fit giving  $R^2=0.748$ . Precipitation chemistry data (i.e. wet-side data) are often examined to gain insight and make qualitative

comments on features of the emission sources. One examines both the geographical patterns and the temporal patterns of the constituent of interest. For wet-side Aerochem data, the question is always whether one should expect concentration data or deposition data to better reflect characteristics of the emission sources. For sodium aerosols at site IL19, we expect the road salt emission source to have the shape of the snowfall pattern in Figure 6 with the highest road salt emissions in January and February, the second highest emissions in December and March, and the third highest emissions in November and April. This is exactly the pattern in wet-side concentration, in Figure 3, and very different from the pattern for wet-side deposition, in Figure 4. Therefore the wet-side concentration, and not wet-side deposition, should be examined to learn something about the temporal pattern of the aerosol emission source.

The estimated dry-side plus wet-side deposition of road deicing sodium at the suburban site IL19 site for the six cold months is  $117 \text{ mg/m}^2/6\text{-cold months}$ . This value is relatively small compared to values at sites very close to the ocean. The NADP/NTN year 2000 wet-side deposition values for sodium<sup>27</sup> for the Cape Cod site (MA01) and the Everglades site (FL11) are 184 and 55  $\text{mg/m}^2/\text{month}$ , respectively. These values, multiplied by 6, are much greater than the IL19 value, even without adding on the dry deposition component. For coastal sites sea spray is the dominant source of sodium being deposited.

## Conclusions

In previous studies road deicing salt deposition has been measured by bulk sampling, close to treated roads, for one or two years. The Kelsey and Hootman study was different

in that their sampling included one site about a kilometer from the treated roads. The dataset for this study is unique in that (1) it is from a site quite far (about two km) from the major road source, (2) both wet and dry deposition samples were collected, and (3) the data record is long (18 years). Conclusions are:

1. Sodium bearing aerosols in the vicinity of site IL19, are large sized, with about 4% of the mass in sizes from 0-2.5  $\mu\text{m}$ , 18% from 2.5-10  $\mu\text{m}$ , and 78% greater than 10  $\mu\text{m}$ . With this relatively large sized sodium aerosol the dry-side “bucket” sampling technique is a reasonable approach. Contamination of the dry-side samples by bird droppings is not a significant issue for sodium, especially for the cold months.
2. Dry and wet depositions of the road salt aerosol in the November to April period, at the IL19 site, were about 103 and 14  $\text{mg}/\text{m}^2$ , respectively. Thus it is absolutely necessary to include dry deposition sampling when studying the deposition of road salt aerosols. For the May to October period of warm months, dry and wet depositions of sodium were about 24  $\text{mg}/\text{m}^2$ .
3. Eight-week dry deposition values of the road salt aerosol at IL19 are well correlated ( $R^2 = .75$ ) with the road salt applied during the sampling periods within about 20 kilometers of the sampling site.
4. For sodium at the IL19 site, the wet-side concentration, not the wet-side deposition, is indicative of the road salt emission temporal pattern.

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## **Appendix C**

# **Atmospheric Deposition of Deicing Salt Applied to Highways in the Southwest Chicago Area**

**Number :** 05-A-1350-AWMA

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## **ABSTRACT**

Last year, at the 2004 AWMA Indianapolis meeting, we presented a paper on sodium and chloride wet and dry deposition at a suburban site at Argonne National Laboratory, about 40 kilometers southwest of downtown Chicago<sup>1</sup>. The present paper continues that discussion by describing results for five sites located about three kilometers west of the Argonne site but still having I-55 as a potentially dominant salt source. The deposition samples are obtained with a wet/dry deposition sampler, which automatically exposes to the atmosphere one 14 liter plastic bucket during precipitation periods and the other 14 liter bucket during non-precipitating periods. The monthly data record for dry deposition at the five Lemont sites extends from 1997 to 2004 while the data record for the IL19 Argonne site was 1980-1998. Air quality measurements show that most of the sodium aerosol mass is in particles greater than 10 microns in diameter. Thus one can expect that the simple bucket measurement of dry deposition is a valid sampling approach for the large sodium particles. The daily application rate of road deicing salt for state roads in the area is known and is found to have a high correlation with salt dry deposited. The winter dry depositions of sodium at the five Lemont sites are presented. Site 5 has very high salt deposition values, due to a nearby street. Site 3 values are unusually low, perhaps due to trees filtering out large salt particles. The IL19 Argonne site values are in reasonable agreement with the Lemont site values.

## **INTRODUCTION**

This report describes results from a project to characterize the atmospheric emission and deposition of road deicing salt, composed mainly of sodium chloride (NaCl). The focus of the effort is to determine the airborne concentration and deposition

of road salt aerosol near the location of the proposed roadway, FAP 340, which would join Interstate 55 (I-55)/Tollway 355 (I-355) and Interstate 80 (I-80) southwest of Chicago (Figure 1). Five monitoring sites were positioned to both characterize the road salt deposition prior to the construction of the proposed roadway and to measure the increase in deposition once the new roadway is built. Results for the 1997-2004 period are summarized from measurements at the five monitoring sites established during this project.

This paper also reviews results from 18 years of sodium deposition measurements at a suburban site about 40 km southwest of downtown Chicago (site IL19, which is part of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN, described at <http://nadp.sws.uiuc.edu>)). The sodium deposition record at IL19 is compared to the deicing salt applied to state roads and expressways in the area.

A simple conceptual model for road salt in the atmosphere is as follows. The tires of vehicles traveling on salted roads inject salt particles into the air. The salt particles can be either liquid or solid. The emission process will be referred to as wet emission when the tires are thrusting salt droplets into the air from a wet/slushy road and will be referred to as dry emission when the tires are thrusting solid salt particles into the air from a dry road. In either case, turbulence caused by the moving vehicles will rapidly mix the emitted particles into a vertical column behind the vehicles and the wind will transport the particles away from the road. The particles will be deposited onto the ground by wet deposition and by dry deposition. The wet deposition process refers to precipitation (i.e. raindrops and snowflakes) falling to the ground. The precipitation contains a background level of salt (from in-cloud scavenging of ambient aerosols) that will be increased as the precipitation falls through a deicing salt plume (i.e. below-cloud scavenging of particles). Dry deposition of deicing salt particles refers to the gravitational and inertial impaction processes that result in the salt droplets and solid salt particles being removed from the atmosphere, during periods when precipitation is not occurring. Impaction processes result in deicing salt being deposited onto vegetation and other obstacles while gravitational settling results in deicing salt being deposited into sampling vessels or onto the ground. Large, traffic-generated salt drops, referred to as splash drops, fall out close to the road. Arbitrarily the strip of ground from the edge of the road out to 15 meters will be referred to as the splash deposition zone. Smaller drops, referred to as droplets or spray, are deposited in the spray and solid particle deposition zone, which begins 15 meters from the road.

Many different deicing substances are applied to roads but due to its relatively low cost, sodium chloride is the dominant deicing material used in the USA<sup>2</sup>. For this paper the term salt or deicing salt refers to sodium chloride. The amount of sodium chloride used in the United States as a highway deicing salt rose steadily from about one million tons per year in the early 1950's to the fluctuating level of 8-11 million tons per year from 1968 to 1992<sup>3</sup> and then to the fluctuating level of 15-20 million tons per year from 1993 to 2001.

The effects of salt and other highway deicers on the environment have been the subject of hundreds of research papers and reports in the last 50 years. Studies of the impacts on motor vehicles, highway infrastructure, human health, soils, surface water, groundwater, vegetation and biological communities (in soils and surface waters) have been reviewed.<sup>4,5,6</sup> The field studies, where impacts of deicing salt have been examined,

have generally focused on distances within a couple hundred meters of the road emission source. In these impact studies salt deposition rates were sometimes quantified, using bulk collectors (containers continuously exposed to the atmosphere to collect precipitation (wet deposition) and dry deposition of salt aerosols) or by washing off salt deposits that had accumulated on vegetative surfaces or by measuring salt levels in soil samples at various distances from the road sources. Occasionally road deicing salt has been identified as one of many emission sources that influence ambient air quality and ambient precipitation quality. Gélinas and Schmit<sup>7</sup> carried out two years of bulk sampling at sites in Montreal (2 sites) and in a small watershed about 30 kilometers away from the city (14 sites). Site location and seasonality of the data led the authors to identify road salt as one of the sources influencing their bulk deposition data. Dasch and Cadle<sup>8, 9</sup> collected dry deposition and wet deposition samples for two years at an urban site in the Detroit area and at a rural site north of Detroit. Here the patterns in sodium deposition were attributed to road salt emissions. Two bulk sampling studies have been done in the Chicago area to quantify the airborne transport and deposition of road salt. Kelsey and Hootman<sup>10</sup> measured bulk deposition at the Morton Arboretum for two winters, at distances from 15 meters to 1018 meters from nearby expressways. A winter 1999/2000 study near South Elgin, IL included measurements of bulk deposition for one winter at 19 sites at distances from 8 to 285 meters from the road.<sup>11</sup> Williams and Stensland<sup>12, 13</sup> gave preliminary reports of the air quality and deposition study investigating road salt levels at five sites 40 kilometers southwest of downtown Chicago, near Lemont, IL. This last study, completing its field measurements in April 2004, was the first to include standard ambient aerosol sampling equipment to determine airborne concentrations and size distributions for road deicing salt ambient aerosols.

## **METHODS**

### **Description of Baseline Monitoring Sites**

The proposed tollway (FAP 340) is a 12-mile road segment that would extend southward from the intersection of I-55 and I-355, joining I-80 at New Lenox (Figure 1). A bridge approximately two kilometers long would span the Des Plaines River, the Sanitary and Ship Canal, and the I&M Canal. Figure 2 is an aerial view of the area showing the approximate position of the proposed roadway with respect to the five sampling sites established in 1996.

The primary strategy behind the locating of the sampling sites was to provide locations for determining the impact of the new road. Essentially the sites are positioned in an area that would be affected by salt deposition from the new roadway, and the project goal was to contrast measurements of the salt deposition at the sites before and after construction of the roadway. As shown in Figure 2, all 5 sites are close to the proposed roadway. Sites 1 and 2 are in the river valley, in an area that would be near the proposed bridge, at two different distances toward the east. Future measurements from dry deposition and aerosol samplers should show larger values adjacent to the bridge compared to lower values at a farther distance away. Both the magnitude and rate of

falloff with distance from the source are important parameters for characterizing the impact of the new road. Sites 3 and 4 are located, similarly to sites 1 and 2, at different distances east of the proposed roadway. These sites are outside the river valley in an at-grade position with respect to the new roadway. I-55, north of the five sites, is considered to be a major source of road salt in this area, before FAP 340 is built

Additional considerations for site locations were the practical aspect of acquiring access to the land. Sites 1 and 2 are along a portion of the historic I&M Canal on land managed by the City of Lemont. Site 4 is in the DuPage County Forest Preserve, and Sites 3 and 5 are on properties of Citizens Utilities Corporation. Several aspects of the sites raise questions for the interpretation of the data presented here. The salt deposition at site 5 should be influenced by I-55, although the effect of streets, buildings and other obstructions shown in Figure 3 are complicating factors that potentially affect the salt deposition pattern. A considerable portion of the warehouse construction between site 5 and I-55 has been done since the beginning of the study and there has been significant new construction since the photograph in Figure 3 was taken. There is no direct pathway between I-55 and site 5 that is unobstructed by warehouse buildings. International Parkway is within 30 m (100 ft.) of site 5, and although this roadway is limited to slowly moving vehicles, the strength of the salt source is potentially important. Joliet Road, which lies between site 5 and I-55, is also a potentially strong salt aerosol source for site 5. It is a divided highway that joins I-55 and carries heavy rush hour traffic.

Sites 3 and 4, while less obstructed from I-55 emissions by the buildings, are located in wooded areas and are obstructed by trees. Figure 4 provides an even more detailed aerial photograph of sites 3 and 4. Site 3 is clearly in the middle of woods and the trees could filter a salt aerosol originating from any direction before arriving at site 3. Site 4 is obstructed by the same woods for winds from the south and east. Two open fields separated by a line of trees lie to the north and east of site 4. There are no obvious local sources to sites 3 and 4 such as International Parkway or Joliet road that may affect salt deposition at site 5. Their seclusion in wooded areas should serve to decrease the salt deposition in comparison to the other sites because of distance from local sources and the potential filtering effect of the trees.

Sites 1 and 2 are isolated from I-55 by larger distances, and they have the additional complicating effect of the intervening river valley. The Village of Lemont lies immediately to the south of the sampling sites. As shown in Figure 5 New Avenue runs in an east and west direction passing both sites at a distance of approximately 0.5 km to the south, and is a potential source of salt aerosol. The bridge crossing the canal area at Lemont Road is located about 1.5 km to the east of site 2. The elevated source of the bridge on Lemont Road may be of some importance for salt emission. Like Joliet Road, Lemont Road connects with I-55 and has a considerable rush-hour traffic load with speeds of about 45 mph. Traffic rates are about 145,000 vpd on I-55 and 15,000 on Lemont Road. Traffic flow on Joliet road is not known but appears to be similar to that on Lemont Road. The I-55 traffic speed is of course greater than that on Joliet Road or Lemont Road so to maintain safe driving conditions, more deicing salt per lane-kilometer must be applied on I-55.

The NADP/NTN site operated by Argonne National Laboratory (ANL), known as the IL19 site (cf. Figure 2), is located about 40 km southwest of Navy Pier, which is on the edge of Lake Michigan near downtown Chicago. The deposition sampler is in the

southwest portion of the ANL property, in a grass-covered area, and is far enough away from obstacles, such as buildings or trees, to meet the NADP siting criteria (also refer to photos at <http://nadp.sws.uiuc.edu/sites/siteinfo.asp?net=NTN&id=IL19>).<sup>14</sup> The site is 1.9 km southeast/downwind of the I-55 salt source when the wind is perpendicular to the road, and 0.8 km downwind of Lemont Road salt source with a west wind. The I-55 expressway is hypothesized to be the dominant sources of sodium deposition measured at IL19 in the cold season months, with smaller deposition components from Lemont Road and from a more general suburban background level source.

## Overview of Sampling Procedures

The main instruments used for sampling the airborne road salt include dichotomous aerosol samplers (dichots) and high-volume aerosol samplers (hivols). The dichot samplers collect particles in the two diameter (D) ranges of  $D < 2.5 \mu\text{m}$  and  $2.5 \mu\text{m} < D < 10 \mu\text{m}$ . The hivol collects all “suspended” particles with approximate  $D < 30 \mu\text{m}$ . Table 1 summarizes sampling terminology used in this paper. The airborne sodium particles are deposited on filter substrates in the instruments: quartz for the hivol and Teflon for the dichot. In the laboratory the filters are extracted in deionized water and then sodium is measured in the solutions by ion chromatography.

Dry and wet deposition of sodium was measured at the five Lemont sites using automatic wet/dry precipitation samplers ( Aerochem Metrics Model 301). A picture of the sampler at site 2 is on the web: <http://www.sws.uiuc.edu/atmos/deposition/deposit2.asp?sz=bgr&pn=2>. The sampler is often referred to as “the Aerochem” or “the Aerochem sampler”. The Aerochem sampler is used at IL19 and at all the other sites in the NADP/NTN (NADP/NTN is described at <http://nadp.sws.uiuc.edu> ). The Aerochem holds two 14-liter plastic buckets, with a lid that is automatically positioned over the wet-side bucket during periods without precipitation and over the dry-side bucket during precipitation events. A sensor for rain or snow particles activates a motor that moves the lid between the two bucket positions. The sample buckets are cleaned in the laboratory with deionized water and stored in plastic bags for transport to and from the field sites. The rain and snow captured in the wet-side buckets is filtered in the laboratory. For the dry-side buckets, 250 milliliters of deionized water are added to the bucket, and after a minimum of 24 hours for soaking, the dry-side sample is handled like a wet-side sample. The Aerochem sensor works very well for rain events but for snow events, especially during cold temperatures when snowflakes are small, it is common for some of the snow to end up in the dry-side bucket because the sensor has not detected the snowflakes and so not moved the lid to cover the dry-side bucket.

Samples from the Aerochem samplers at Lemont sites 1-5 were analyzed for sodium by ion chromatography, with a detection limit of about 0.1 milligram/liter (mg/L). Sodium in samples from the Argonne site IL19, and all other NADP/NTN sites, is analyzed by flame atomic absorption spectrophotometry, with a detection limit of about .003 mg/L.

Average monthly air concentrations for the super-coarse PM (>10), coarse PM (2.5-10), and fine PM (<2.5) size fractions, averaged over all sites, are shown in Table 2. The values are broken into two classes represented by sampling periods when at least one

snow event occurred (snow periods) and periods when no snow events occurred (non-snow periods). The hivol data are blank corrected and the occasional slightly negative values that occur after the blank subtraction are set to zero. The data in Table 2 show that the sodium bearing aerosols in the vicinity of five Lemont sampling sites are large sized. During sampling periods that included some snowfall, about 4% of the sodium mass is in particles with diameters from 0-2.5  $\mu\text{m}$ , 18% in particles from 2.5-10  $\mu\text{m}$ , and 78% in particles greater than 10  $\mu\text{m}$ . During snow periods, the super-coarse particle Na concentrations are over two and one-half times larger than for the non-snow periods. The sodium in coarse particles is over 8 times higher during snow periods compared to non-snow periods. Figured as the percent of particle mass composed of NaCl, the salt percent mass is 2.3% for the super-coarse particles during non-snow months and increases to 9.5% during months with snow. The NaCl mass percent for the coarse particles is less than 0.7 for non-snow periods and 7.3% for snow periods.

A comparison of the mean airborne sodium concentrations in Table 2 shows that during snow periods there about 4 times more sodium mass in the super-coarse compared to the coarse particles. Thus about 20 % of the salt mass is in particles less than 10  $\mu\text{m}$  in diameter. Particles of this size are able to remain suspended in the atmosphere for longer periods of time than super-coarser particles, and can be transported for very long distances. If emitted from a roadway, 10  $\mu\text{m}$  diameter particles typically would not deposit within the first few km. So, there is clearly a component of the salt aerosol that deposits in a regional rather than a local spatial scale. This suggests that Chicago or the Chicago Metropolitan Area could be significant sources of salt aerosol at the Lemont sampling sites under the right conditions.

## **Issues in Dry Deposition Sampling with Buckets**

In general the measurement of dry deposition of gases and particles is a difficult and complicated process. Exposed buckets, such as those in the dry-side of the Aerochem samplers, would not be expected to measure adequately the dry deposition of gases or aerosols composed of small particles (e.g. sulfate), but would be expected to work much better for aerosols composed of large particles. The aerosol sampling in this project, with hivol and dichot samplers, has shown that the sodium and calcium in the ambient aerosol is predominantly in particles greater than 10 microns aerodynamic diameter and thus large enough that buckets are reasonable to use for dry deposition.

In early 1980's a debate was occurring within NADP/NTN as to whether the dry-side bucket data provided credible and useful data for the various analytes. At that time the primary interest was in determining the deposition of sulfur and nitrogen species. To determine the true dry deposition of something like sulfur it was argued that one needed to measure separately the deposition of gaseous species (i.e. sulfur dioxide), fine sulfate particles ( $D < 2.5 \mu\text{m}$ ), course sulfate particles ( $2.5 < D < 10 \mu\text{m}$ ) and super-coarse particles ( $D > 10 \mu\text{m}$ ), and that one should not assume that sulfate ion measured in dry-side bucket extractions was likely to represent or be well correlated with the sum of these four components. For crustal aerosols like calcium, which have a mass median diameter greater than 5  $\mu\text{m}$ , it was noted that the data from dry-side buckets was more representative.

Dry deposition can be calculated by multiplying the measured aerosol concentration by the dry deposition velocity. With the data in Table 2 showing that most of the aerosol mass is in particles sizes greater than 10  $\mu\text{m}$ , one can assume that the dry deposition velocity for the road salt aerosols is equal, to a good approximation, to the terminal settling velocity. To implement this dry deposition measurement approach effectively one would need to know the aerosol concentration as a function of particle size, where the size classes should probably not be more than 3-4  $\mu\text{m}$  in width. Accurate sampling of the road salt particle size distribution is difficult because the large momentum of the particles means it is difficult to keep them from impacting onto surfaces of the sampler other than the actual collection media. The sampling difficulty for large particles was dramatically demonstrated in a “shootout” type of equipment intercomparison study by May et al.<sup>15</sup> in which 17 devices were compared in collecting 20, 30, and 50  $\mu\text{m}$  test aerosols during about 15 tests under field conditions. The intercomparison study demonstrated that large sampling biases can occur when attempting to sample large particles. The greatest bias occurred with a modified Anderson cascade impactor that only collected 4%, 7% and 12% of the 50, 30, and 20  $\mu\text{m}$  test aerosols, respectively. Gélinas et al.<sup>16</sup> used the bulk method to determine total deposition but commented, “Dry and bulk deposition fluxes are more reliably determined using airborne concentrations along with a dry deposition velocity model.” We agree in principal with this statement but are not aware of a study for large sized aerosols like road salt aerosols that demonstrates the practicality of the statement. Considering the challenges in properly sampling large particles and the fact that multiple sites are usually necessary, it appears that resources will seldom be available to determine deposition over long periods of time without resorting to something like bulk or dry-side/wet-side sampling with surrogate surfaces.

In the winter 1999/2000 field study in South Elgin, IL<sup>17,10</sup>, biweekly bulk sampling was done to look for road salt deposition decreasing with distance from a highway with traffic flow of about 12,000 vpd. In one part of the experiment seven plastic sampling buckets were deployed at seven sites from 13 to 136 meters from the road. During each of seven two-week sampling periods when deicing salt was used on the highway, the deposition of sodium decreased with distance from the highway as was expected. The consistency of the data qualitatively supports the bulk sampling approach for wintertime road salt aerosols.

A concern with the dry-side samples has been that they are quite frequently contaminated by bird droppings and thus might not be useful for scientific studies. Hicks (1986)<sup>18</sup> referred to the problem as a severe limitation but did not provide support for the conclusion. Feely et al. 1985<sup>19</sup> collected six dry-side samples daily for 23 summer days. Comparison of the visible contamination recorded for each sample and the ion concentrations in the extraction solutions led Feely et al.<sup>18</sup> to conclude that bird droppings were the only visible contaminant that seemed to significantly affect the chemical composition. Gatz<sup>20, 21</sup> examined the contamination comments recorded for the samples in the NADP/NTN dry-side data set and found that the deposition calculations for the ions sodium and calcium were not strongly affected by inclusion of samples that had contamination comments, including comments identifying bird dropping contamination. However, for other ions like potassium, ammonium, and phosphate, the summary statistics were strongly influenced by inclusion of samples with contamination comments

indicating the presence of bird droppings in the dry-side sample bucket. The contamination comments in the database resulted from observations by laboratory personnel, and to some extent by field personnel, of the visible state of the dry-side bucket sample. If one were going to attempt to examine patterns of dry deposition for ions such as potassium, ammonium or phosphate, through the use of such dry-side bucket data, one would certainly need to have a means to screen out those samples affected by bird contamination.

We thus conclude that for the dry-side samples, sodium and calcium are not among the ions significantly influenced by the bird contamination. However, to further guard against the influence of contaminants in the buckets, medians were used, instead of means, to summarize the dry-side data.

### **Road Salt Applied Records**

Records of the amount of road salt applied daily, by the state, in the Chicago area in the 1987-2002 period were obtained from the Illinois Department of Transportation (IDOT). The records are for the IDOT District 1, the Chicago area, composed of Cook County and the 5 counties that share boundaries with Cook County. Twenty-two winter operations teams, distributed across the area, apply most of the deicing salt for IDOT in District 1. IDOT salts all the major highways and expressways in the Chicago area. Other road salting occurs in the area, including salting of city streets by the city of Chicago and by other municipalities in the area. By comparing the salting activity by one team and with the salting activity of nearby teams it was observed that the salting patterns are well correlated. That is, when a snow event occurs, all the IDOT teams and municipal teams will be simultaneously salting streets and roads in their jurisdiction.

## **RESULTS and DISCUSSION**

### **Dry Deposition of Sodium at Argonne Site IL19**

A total of 93 dry-side Aerochem samples were collected at the IL19 site from 1980 until early 1998 and were analyzed for a suite of ions including sodium.<sup>1</sup> The mid-value of a sampling interval was used to assign the samples to months. The number of dry-side samples by month was 8,8,7,9,7,7,7,8,6,8,5, and 13 for January through December, respectively. Since wet-side samples were weekly in duration, the number of wet-side samples is approximately 8 times the number of dry-side samples. Wet-side deposition data were collected at IL19 from 1980 to 2004. Median dry deposition values at IL19 for sodium are shown in Figure 6. Dry deposition is being operationally defined as that amount collected with the Aerochem dry-side sampling protocol. For each month the median and the +/- 95% confidence intervals<sup>22</sup> are shown.

Wet deposition is calculated as the product of the weighted average ion concentration for a month times the precipitation amount for the month, where the later is determined with the Belfort precipitation gage at the NADP/NTN sites<sup>23</sup> and not from the volume of water in the Aerochem buckets. The sodium wet deposition values have a maximum in April and a minimum in September, and are relatively uniform throughout the year when compared to the dry deposition pattern shown in Figure 6 where the maximum values are about 7-8 times the minimum values. The wet and dry deposition

data are combined to calculate the percent that dry deposition of sodium is of total deposition (wet plus dry), with the result being shown in Figure 7. For December-March, the months with the largest dry deposition, the percent dry deposition values are greater than 70%; for Jan-Feb, with the really large dry deposition values for sodium, the percent dry deposition is about 85%. In the warm months May to October, when road salting is not occurring, the average wet and dry deposition values of sodium are both about 4 mg/m<sup>2</sup>/month. Soil dust and sea salt aerosols transported into the Chicago area are likely sources for this warm period deposition of sodium, with industrial emissions and demolition and construction activities also being possible sources. We assume these warm period background values are the same during the November to April cold months, when road salt aerosols are present. Subtracting these background levels from the cold month values, and summing over the six cold months gives an estimate for total salt aerosol deposition due to road salt of about 103 mg/m<sup>2</sup>/6 winter months for dry deposition and 14 mg/m<sup>2</sup>/6 winter months for wet deposition. For the six warm months, May – October, the dry and wet deposition values of sodium are both about 24 mg/m<sup>2</sup>/6-months. The total winter, sodium, dry-side deposition is (103+24)/6 = 21.2 mg/m<sup>2</sup>/month. Figure 9 summarizes the IL19 values.

The snowfall data for 1980-1998 for the closest National Weather Service Cooperative Observer Site, near Midway Airport, were examined. The largest precipitation values are in the warm months and the largest snowfall values are in January and February. It is reasonable to hypothesize that the quantity of deicing salt applied to roads in the area will have a temporal pattern similar to that for monthly snowfall. Thus the road salt emissions should be the largest in January and February, the two snowiest months, and the dry deposition should also be the largest in January and February, as is observed in Figure 6.

IDOT Team 135 applies the salt in the vicinity of site IL19 (on I-55 and within about 20 km of site IL19). The Team 135 daily salt applied values were summed into time periods corresponding to the exposure periods (about 8 weeks) of the IL19 dry-side bucket samples. A scatter plot of Team 135 salt applied versus the corresponding sodium deposition determined from the dry-side buckets is shown in Figure 8. Dry-side samples that included exposure only during the months of May through October, when no deicing salt was applied, are not included. From Figure 8 it is seen that the dry deposition of wintertime sodium particles at the IL19 site is well predicted by the road deicing salt applied in the Team 135 area, with a linear fit giving  $R^2 = 0.748$ .

The estimated dry-side plus wet-side deposition of road deicing sodium at the suburban site IL19 site for the six cold months is 117 mg/m<sup>2</sup>/6-cold months. This deposition value is relatively small compared to values at sites very close to the ocean. The NADP/NTN year 2000 wet-side deposition values for sodium for the Cape Cod Massachusetts site (MA01) and the Everglades Florida site (FL11) are 184 and 55 mg/m<sup>2</sup>/month, respectively. These coastal wet deposition values, multiplied by 6, are much greater than the IL19 value, even without adding on the dry deposition component. For coastal sites sea spray is the dominant source of sodium being deposited.

### **Dry Deposition of Sodium at Lemont Sites 1-5**

The dry-side bucket measurements of sodium dry deposition are shown in Table 3 for the 30 sampling periods that occurred in winter. Table 3 gives the midpoints of the

sample periods and the duration of the periods. Winter samples were defined to be those with sample midpoints falling in the months of November to April. Summer samples were defined to be those with midpoints in June – September. Even for samples with durations of 6-8 weeks, this definition of summer will assure that road salt does not influence the summer samples.

Working hypothesis #1 has been that I-55 was the dominant source of winter sodium for each of the five sites. Therefore, based on distance from this source, one expects the dry deposition of sodium for winter periods to have the pattern: site 5 > site 4 > site 3 > site 1 and site 2. Sites 1 and 2 are about the same distance from I-55 so we expected them to have similar sodium levels.

Dry-side deposition data are shown on the left side of Table 3. Careful data screening, including thorough reviews of notes recorded by the field and laboratory technicians, resulted in data gaps as shown on the table. The sites were usually only visited during sample change trips because the sites were a 2.5 hour drive from the offices/shops/labs of the project staff in Champaign, IL. Equipment malfunctions at a site sometimes led to a few consecutive samples being lost in the data screening process. Being closer to the sites to allow for more frequent checks on the equipment would have reduced the number of such lost samples.

On the right side of Table 3 is a tabulation of site-to-site comparisons, which constitute non-parametric statistical tests, also called binomial tests. It is concluded that site 5 has much higher dry deposition values than the other four sites. This must be due to the considerable traffic on the adjacent local street, International Parkway. This new street and the many new large storage buildings now filling the space from site 5 all the way north to I-55 were not present when the site 5 location was selected in 1996. Of the five Lemont sites, only site 5 will be on the west side of the new tollway, and was intended to be similar to site 4 except on the other side of the new road. The growth of traffic activity near site 5 will prevent the site 4 versus site 5 comparisons. However, with the baseline measurements for site 5, it will still be useful to look for salt deposition increases due to traffic on the new toll road. The other strong conclusion from Table 3 is that the dry deposition at site 1 is larger than the dry deposition at site 3. If I-55 were in fact the current dominant salt source for sites 4, 3, 2, and 1, then site 1 should not have higher deposition than site 3. Since sites 1-4 are quite far from I-55, it might be that I-55 becomes unrecognizable, being just one more component of the urban/regional salt plume, so that sites 1-4 would have similar dry deposition values. Another point to consider is that site 3, being surrounded on 3 sides by many trees, might have lower concentrations of airborne salt particles because the trees filter out the large salt particles before they get to site 3. Our opinion is that filtering by the trees is important to explain lower values at site 3, but that site 1 (and site 2 to a lesser degree) have elevated values due to local sources in Lemont, just to the south of the sites, that combine with the other sources, including I-55. The density of trees around site 4 is less than for site 3, so any filtering effect by the trees would be less than at site 3. Although not statistically significant, the binomial test data suggest the following additional tendencies: site 1 > site 2 ( $p=11.9\%$ ); site 4 > site 2 ( $p=13.2\%$ ) and site 4 > site 3 ( $p<10\%$ ).

It is useful to have quantitative values for salt deposition at the five sites and these are given in Tables 4 and 5. For Table 4 all the data in Table 3 are used, which results in different numbers of samples at the sites, varying from  $N=21$  to  $N=29$ . To avoid the

influence of a few very large values, we suggest using the median values. The site 5 median is greater than the medians for the other five sites; the site 1 median is greater than the site 3 median; the site 1 median is greater than the site 2 median, the site 4 median is greater than the site 2 median, and the site 4 median is greater than the site 3 median. These five median comparisons all agree with the statistical tendencies from the binomial tests discussed in the previous paragraph.

For Table 5 the Table 3 data are reorganized to provide equal N values for site groups. The last two columns on the right contain the 22 winter events where both sites 4 and 5 have dry-side deposition values. Site 5 is a factor of 2.8 larger than site 4 when medians are compared and a factor of 2.3 larger if means are compared. In the third through sixth columns from the right in Table 5, the data for the 14 cases where sites 1,2,3,and 4 all had dry-side deposition values are shown. The four median values show the same patterns discussed in the paragraph above, i.e. the site 1 median is larger than the site 3 median, etc. The biggest difference in median values, in comparing Tables 4 and 5, is for site 3.

In summary, using the median values on Tables 4 and 5, site 5 is about 2.5 larger than site 4 and the winter dry-side deposition values for sites 4,3,2, and 1 are 18.9, 11.5, 16.3, and 21.9 mg/m<sup>2</sup>/month, respectively. The value for the IL19 Argonne site is 21.2 mg/m<sup>2</sup>/month, in reasonable agreement with the Lemont site 4 value considering that the Argonne dry-side data record is for 1980-1998 while that for the Lemont sites is 1997-2004.

## SUMMARY AND CONCLUSIONS

In this study air concentration and deposition of airborne road deicing salt (i.e. sodium) has been measured at five sampling sites near Lemont, IL. The primary objective was to determine winter baseline deposition levels that can be compared with the levels after a new tollway (connecting I-355 and I-80) is constructed. Six years of winter dry and wet deposition data were collected at the five Lemont sites, which were quite far (about 1.5 to 4.5 km) from a present major road source, I-55. A nearby NADP site, IL19, at Argonne National Laboratory, has a longer record (1980-1998 for wet and dry deposition) and a superior wet deposition dataset (due to the use of a laboratory method with a lower detection limit for sodium) so it was used to supplement the Lemont data. The IL19 site is about the same distance from I-55 as Lemont sites 3 and 4, and is about 6 kilometers to the northeast of these sites.

Conclusions are:

1. Sodium bearing aerosols in the vicinity of five Lemont sampling sites are large sized. During sampling periods that included at least one snow event, about 4% of the mass is in particles with diameters from 0-2.5  $\mu\text{m}$ , 18% in particles from 2.5-10  $\mu\text{m}$ , and 78% in particles greater than 10  $\mu\text{m}$ . With this relatively large sized sodium aerosol, the dry-side "bucket" sampling technique is a reasonable approach for measuring dry deposition. Contamination of the dry-side samples by bird droppings is not a significant issue for sodium, especially for the cold months when bird activity is low. The large airborne salt

particles were sampled with the standard high volume air sampler, which likely under-collects the largest particles due to inlet problems, so the 78% value is biased low.

2. Average dry-side and wet-side deposition of the road salt aerosol in the November to April period, at the IL19 Argonne site, was about 127 and 38 mg/ m<sup>2</sup>/6-months, respectively, or about 21.2 and 6.3 mg/m<sup>2</sup>/ month, respectively (salt as sodium). Thus it is absolutely necessary to include dry deposition sampling as well as wet deposition when studying the total deposition of road salt aerosols. For the May to October period of warm months, dry and wet deposition of sodium were each about 24 mg/m<sup>2</sup>/6-months. If one assumes the summer salt values, which are from non-road salt sources, are present at the same levels in the winter months, then one can subtract them from the winter values to estimate the road salt contribution. With this assumption one gets an average dry deposition value of road salt sodium for the six winter months of about 103 mg/m<sup>2</sup>/6 months or an average winter month value of about 17.2 mg/m<sup>2</sup>/month. Although relatively large, it is much smaller than deposition within a few kilometers of an ocean coastline.

3. Eight-week dry deposition values of the winter road salt aerosol at IL19 are well correlated ( $R^2 = .75$ ) with the road salt applied (within about 20 kilometers of the sampling site) during the sampling periods.

4. Dry deposition data for 30 winter sampling periods for the five Lemont sites are shown on Tables 3-5. On the right side of Table 3 is a tabulation of site-to-site comparisons, which constitute non-parametric statistical tests, also called binomial tests. It is concluded that site 5 had much higher dry deposition values than the other four sites, by a factor of about 2.5. The other strong conclusion from Table 3 is that the dry deposition at site 1 is larger than the dry deposition at site 3. If I-55 were in fact the current dominant salt source for sites 4,3, 2, and 1, then site 1 should not have higher deposition than site 3. Since sites 1-4 are quite far from I-55, it might be that I-55 becomes unrecognizable, being just one more component of the urban/regional salt plume, so that sites 1-4 have similar dry deposition values. Another point to consider is that site 3, being surrounded on 3 sides by many trees, might have lower concentrations of airborne salt particles because the trees filter out the large salt particles before they get to site 3. Our opinion is that filtering by the trees is important to explain lower values at site 3, but that site 1 (and site 2 to a lesser degree) have elevated values due to local sources in Lemont, just to the south of the sites, that combine with the other sources, including I-55. Summary statistics for the Lemont sites are shown on Table 4, with median, winter, dry-side sodium deposition values for sites 1-5 being 21.9, 16.3, 11.5, 18.9, and 34.0 mg/m<sup>2</sup>/month, respectfully. The site 4 median value of 18.9 agrees reasonably well with the Argonne IL19 value of 21.2.

5. It is fairly common to hear or read the opinion that large particles, which would include road salt particles, are not really an air quality issue since they are too large to travel any significant distance. Our study shows that some fraction of the airborne road salt particles travels large distances, i.e. several kilometers. Our opinion is that in the Chicago area an urban/regional salt plume will develop and exist, for many hours to a

few days, each time weather conditions result in substantial salt being applied to provide safer driving conditions.

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