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Airshed Domains for Modeling Atmospheric Deposition of Oxidized and Reduced Nitrogen to the Neuse/Pamlico System of North Carolina

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ABSTRACT

Atmospheric deposition is important to nutrient loadings to coastal estuaries. Atmospheric emissions of nitrogen travel hundreds of kilometers as they are removed via atmospheric deposition. Long-range transport from outside the Neuse/Pamlico system in North Carolina is an important contributor to the total (wet + dry) deposition of nitrogen to the watershed, estuary and Sound. We need to delimit the extent of long-range transport that significantly contributes to deposition and thus loadings. Since airsheds do not have natural boundaries, in contrast to watersheds, an approach to define principal airsheds has been developed using simulations of the Extended Regional Acid Deposition Model. Principal airsheds for the deposition of oxidized nitrogen (nitrates/nitric acid), and reduced nitrogen (ammonium/ammonia) are defined and characterized. The principal airshed for oxidized nitrogen is 665,600 km^2 and for reduced nitrogen it is 406,400 km^2, 25 and 15 times larger than the drainage area, respectively. Nitrogen oxide emissions from within the oxidized nitrogen principal airshed are estimated to explain 63% of the oxidized nitrogen deposition to the Neuse/Pamlico system. Ammonia emissions from within the reduced nitrogen principal airshed are estimated to explain 60% of the reduced nitrogen deposition to the system for 1996 emissions. North Carolina emissions contribute a large share of the reduced nitrogen deposition, but a small share of the oxidized nitrogen deposition, estimated to be 45% and 20%, respectively. Thus, a large regional atmospheric perspective is necessary for multi-media modeling involving nutrient deposition to coastal estuaries.

Key Words: Airsheds, atmospheric deposition, estuaries, nutrient deposition, eutrophication.

INTRODUCTION

Nitrogen is a major factor controlling primary production in estuarine waters (Ryther and Dunstan 1971, Nixon 1986). The fluxes of nitrate from rivers have been significantly increasing. For example, nitrate concentrations in the Mississippi River have more than doubled since 1965 (Turner and Rabalais 1991, Justic´ *et al.* 1995). Nutrient flux to coastal systems is central to productivity; yet, the flux has increased to the extent that degradation of the marine environment is now a widespread occurrence and is creating extensive concern (Vitousek *et al*. 1997*)*. Evidence suggests a long-term increase in the frequency of phytoplankton blooms, including the Chesapeake Bay (Officer *et al.* 1984) and the Neuse River and Pamlico Estuaries in North Carolina (Copeland and Gray 1991, Paerl *et al.* 1998).

Atmospheric deposition of nitrogen is estimated to contribute an important fraction of the flux of new nitrogen into coastal estuaries (Valigura *et al.* 1996, Valigura *et al.* 2000). For many estuaries the fraction coming from atmospheric deposition falls within the range of 20 to 30%. This large percentage is judged to be significant, and atmospheric loading contributions are starting to be considered when addressing the problems of coastal eutrophication (e.g., Whitall and Paerl, 2001).

Atmospheric deposition of nitrogen contributes to the nutrient load via two pathways. The first pathway is direct deposition to the water surface. The second is deposition to the watershed and then terrestrial release of a fraction into streams and rivers to be carried, with in stream attenuation, to the estuary. Typically, the major pathway is the second, indirect one (Valigura *et al.* 2000). Thus, total (wet plus dry) atmospheric deposition to the entire watershed drainage area is important.

Total deposition of nitrogen comes in two key inorganic forms: (1) oxidized-nitrogen (wet nitrate and dry particulate nitrate and gaseous nitric acid), and (2) reduced-nitrogen (wet ammonium and dry particulate ammonium and gaseous ammonia). While total oxidized-N deposition can be estimated from monitoring measurements, total reduced-N deposition cannot be because ammonia gas is not monitored routinely. Our modeling analysis suggests that oxidized-N accounts for approximately 65% and reduced-N approximately 35% of the total inorganic nitrogen deposition for many U.S. East Coast estuaries. Oxidized nitrogen stems mainly from combustion of fossil fuels and the release of nitrogen oxides. Reduced nitrogen stems mainly from agricultural releases of ammonia (fertilizer application and animals). However, the relative importance of reduced-N deposition is expected to increase in the future because major reductions in nitrogen oxides emissions are being mandated as part of the ozone control program and ammonia emissions are expected to increase.

From a coastal biogeochemical perspective and to better assess the budget of nutrients originating from the atmosphere, we would like to know from where and from how far away are the emissions that are most responsible for the atmospheric deposition of nutrients. Since oxidized-N deposition is a component of acidic deposition (better known as acid rain) one expects long-range transport to be a factor in oxidized-N deposition. On the other hand, there is a range of views about how far reduced-N travels as it deposits.

Sorting out nutrient transport and lifetimes in the atmosphere and airsheds requires a regional air quality model. We have used one such model to estimate the range of transport of oxidized-nitrogen emissions for the eastern U.S. and have used this information to develop airsheds of oxidized-nitrogen deposition for several estuaries (Dennis 1997, Valigura *et al.* 2000). In this paper our work is expanded to include the range of transport of reduced-nitrogen and development of the airshed of reduced-nitrogen for the Neuse River-Pamlico Sound system.

In the next section, we briefly describe the Extended RADM, the regional air quality model modified for this study, provide a sense of its performance, and summarize the approach used to develop annual averages. We next assess the range of influence and lifetimes of oxidized- and

reduced-nitrogen that provide a basis for developing airsheds. We then explain the concept of a principal airshed and characterize the principal oxidized- and reduced-nitrogen airsheds developed for the Neuse River/Pamlico Sound system.

MODEL USED

To address the fate of inorganic nitrogen in a consistent modeling framework, the Regional Acid Deposition Model (RADM) (Chang *et al.* 1987) was enhanced. The RADM is an Eulerian (fixed-grid) model that was developed under the National Acidic Precipitation Assessment Program (NAPAP) as a state-of-the-science model to address regional and urban gas-photochemistry, aqueous chemistry, cloud processes, transport, and wet and dry deposition (Chang *et al.* 1990). RADM was developed with grids that are 80-km on a side. We enhanced RADM by adding a new module adapted from the Regional Particulate Model (Binkowski and Shankar, 1995) and modifying others to represent the physical and chemical pathways governing the fate of emitted NH_3, particularly the sulfate-nitrate-ammonium-water aerosol composition based on equilibrium thermodynamics. The module to represent aerosol equilibrium is based on the work of Saxena *et al.* (1986) that was further modified by Binkowski and Shankar (1995). No distinction of aerosol size was retained because the inorganic particles are commonly in the fine aerosol size fraction; typically, < 1 micron. Aqueous chemistry and wet and dry deposition modules were enhanced to include particulate nitrate and ammonium. The enhanced model is termed the Extended RADM.

Because RADM predicts hourly photochemistry and is, hence, very computationally intensive, an aggregation technique developed during NAPAP (Dennis *et al.* 1990) is used to create annual estimates of nutrient deposition. Meteorological cases of 5-day duration are grouped by wind-flow pattern through cluster analysis and sampled proportionate to their frequency of occurrence (Brook *et al.* 1995a and 1995b). A total of thirty cases constitute the aggregation sample. The aggregation technique produces a climatological average of transport and deposition.

Comparisons of model predicted, climatological annual averages with National Acid Deposition Program (NADP) and Clean Air Status and Trends Network (CASTNet) measurements averaged for several years are summarized below.

Wet deposition:	$SO_4^{=}$:	Ext.RADM = 1.20 x NADP, R^2 = 0.73, (n=124)
	NH_4^{+}:	Ext.RADM = 1.17 x NADP, R^2 = 0.36, (n=121)
	NO_3^{-}:	Ext.RADM = 1.16 x NADP, R^2 = 0.64, (n=124)
Ambient concentrations:	HNO_3:	Ext.RADM = 1.37 x CASTNet, R^2 = 0.36, (n=44)
	$HNO_3 + NO_3^{-}$:	Ext RADM = 1.41 x CASTNet, R^2 = 0.61, (n=44)
	NO_3^{-}:	Ext.RADM = 1.28 x CASTNet, R^2 = 0.63, (n=44)
	$NO_3^{-}/(HNO_3 + NO_3^{-})$:	Ext.RADM = 1.04 x CASTNet, R^2 = 0.47, (n=43)
	NH_4^{+}:	Ext.RADM = 1.25 x CASTNet, R^2 = 0.60, (n=41)

It is difficult for a grid model operating from first principle descriptions of the processes, combined with comparing its grid predictions with point measurements, to achieve correlations greater than 0.7 (R^2 = 0.49). Five of the eight correlations look quite good, one acceptable, and two, wet ammonia and ambient HNO_3, not as good. More recent modeling studies of ammonia indicate the large uncertainty in ammonia emissions is significantly affecting model performance (Gilliland, 2001). Thus, we believe the lower NH_3 correlation is not so much an issue with the model, but rather

is an issue of a poor ammonia emissions input. For HNO_3, during the warm half of the year when the spatial signal is more pronounced, due to more active photochemistry, the R^2 is 0.52, more comparable to the wet nitrate deposition. For ambient NO_3^-, during the cold half of the year when its spatial signal is more pronounced the R^2 is 0.71. The R^2 of 0.61 for total nitrate ($HNO_3 + NO_3^-$), where there is less concern about measurement bias, is quite reasonable and the partitioning of total nitrate is basically unbiased with an acceptable R^2 of 0.47. Thus, we believe the Extended RADM to a sufficient degree is capturing the spatial patterns in annual wet deposition, ambient concentrations and partitioning of key species.

Also, the strong trend in wet NH_4^+ measured at the Clinton, NC site (NADP site NC35) has been corroborated with the Extended RADM. Wet deposition of NH_4^+ at this site increased by approximately 55% for the period 1989-1992 compared to the period 1995-1997. Emissions in the 80-km grid cell overlying site 35 were doubled to account for the increase in agriculturally-related NH_3 emissions. Wet deposition of NH_4^+ in Extended RADM increased by 44% in this grid cell, but increased only small amounts in cells containing other NADP sites. This is consistent with Walker et al., 2000 who found no statistically significant trends at NADP sites other than NC35, providing further credibility for the model predictions.

The Extended RADM is imperfect; nonetheless, its calculations and spatial predictions are in adequate agreement with measurements. Hence, insights developed from the Extended RADM should be reasonable.

DEFINING AIRSHED DOMAINS

From a biogeochemical perspective, we would like to know from where do the atmospheric emissions emanate that deposit and affect the watershed of interest. We do this through the concept of an airshed. However, an airshed for air pollutants is a fuzzy entity. There are no clear boundaries preventing the flow of chemicals in the atmosphere as there are for the flow of surface waters in watersheds. The absolute influence from a source continuously diminishes with distance to extremely small levels. As we reach smaller levels of deposition we also reach a point of diminishing returns in terms of the efficiency with which emissions from that source deposit on a receptor watershed and the overall proportion of the budget being accounted for.

Thus, we have developed the concept of a Principal Airshed. Its size and shape is related to the distances pollutants transport and deposit as a function of climatology. The principal airshed boundary is defined from the perspective of the climatological deposition range of influence of the sources surrounding the region of interest. A source subregion's climatological range of influence is normalized by its emission rate, hence, is independent of the emission rate. Thus, the principal airshed boundary is established in terms of the climatological range of deposition from a source, not in terms of percent contribution to the deposition due to the current configuration of emissions. The climatological range of deposition is quantified by the normalized range of influence. To compute the normalized range of influence, first, the climatological pattern of deposition from a source subregion is defined via the model; second, the deposition is summed to define the total deposition, across the modeling domain, from that source. Then we go back to pattern of deposition and start at the point of maximum deposition and work down the pattern along contours of constant deposition, adding up the deposition as we go. When we reach a pre-established percentage of the original total deposition, we mark this contour of equal deposition and continue the process. Typically we

demarcate the contours at 25%, 50%, 65%, and 80%. The normalized range of influence for oxidized-N and reduced-N is shown in Figure 1a and 1b, respectively, for a source subregion associated with the NC/TN/GA border area. We have created these same types of contours for 100 source subregions across the eastern U.S. and Canada with typical sizes of subregions being 160 x 160 km and 160 x 240 km.

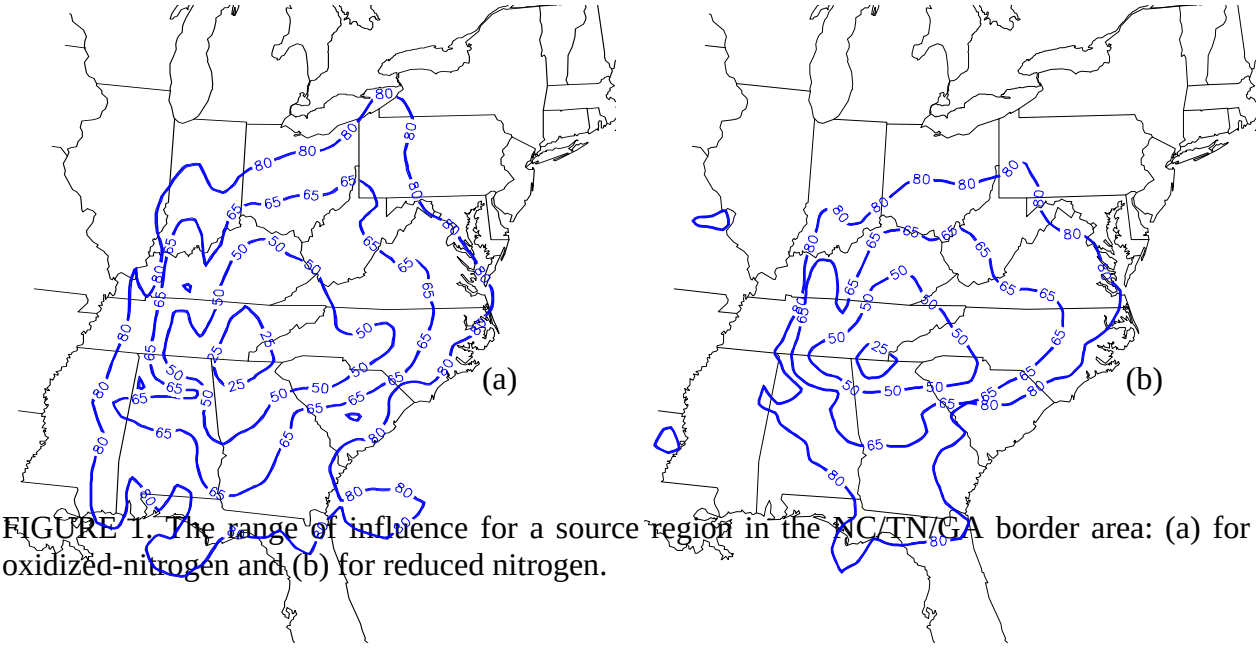

FIGURE 1. The range of influence for a source region in the NC/TN/GA border area: (a) for oxidized-nitrogen and (b) for reduced nitrogen.

The effective influence distance of a source's deposition can be defined similar to that as defining a chemical lifetime, but focusing on what has been lost rather than what is left. In this case we quantify the distance at which the accumulated deposition encompasses 1-(1/e) of the total, or roughly 65%. This 1-(1/e)-distance becomes the range of influence of the source region being studied. The normalized range of influence from NO_X emission sources in the eastern U.S. is the order of 400-600 km for the 65% contour in the prevailing directions of transport (see also Dennis, 1997). The distance is shorter in the direction opposite the prevailing wind. The influence of prevailing climatology on the pattern of the range of influence is evident in Figure 1. The normalized range of influence from NH_3 emission sources in the eastern U.S. is roughly 3/4^{ths} that for oxidized nitrogen, that is, the order of 300-450 km. These ranges are consistent with a residence time of approximately 1 to 1½ days for oxidized-N and up to 1 day for reduced nitrogen (Schwarz 1989). The major determinate of the lifetime and range is cleansing of the atmosphere through take-up of gases and particles into cloud water and subsequent wet deposition of the oxidized and reduced nitrogen through precipitation. However dry deposition of oxidized nitrogen is as influential as wet deposition for that form of nutrient because oxidized nitrogen is mostly in the form of nitric acid which dry deposits extremely rapidly (at the highest feasible rate).

Operationally, we have found that a good distance of demarcation for setting the principal

airshed boundary is the 65% contour of the normalized range of influence. Two examples are shown in Figure 2 in relation to the airshed boundary. Figure 2a shows a source region that is “in” the principal airshed and Figure 2b shows a source region that falls just “outside” the principal airshed boundary. The boundary of the principal airshed shown in Figure 2 is the line of separation between more-efficient (the 65% contour encompasses a part or all of the watershed of interest) and less-efficient (the 65% contour is barely inside, just touching or is outside the watershed perimeter) emission sources.

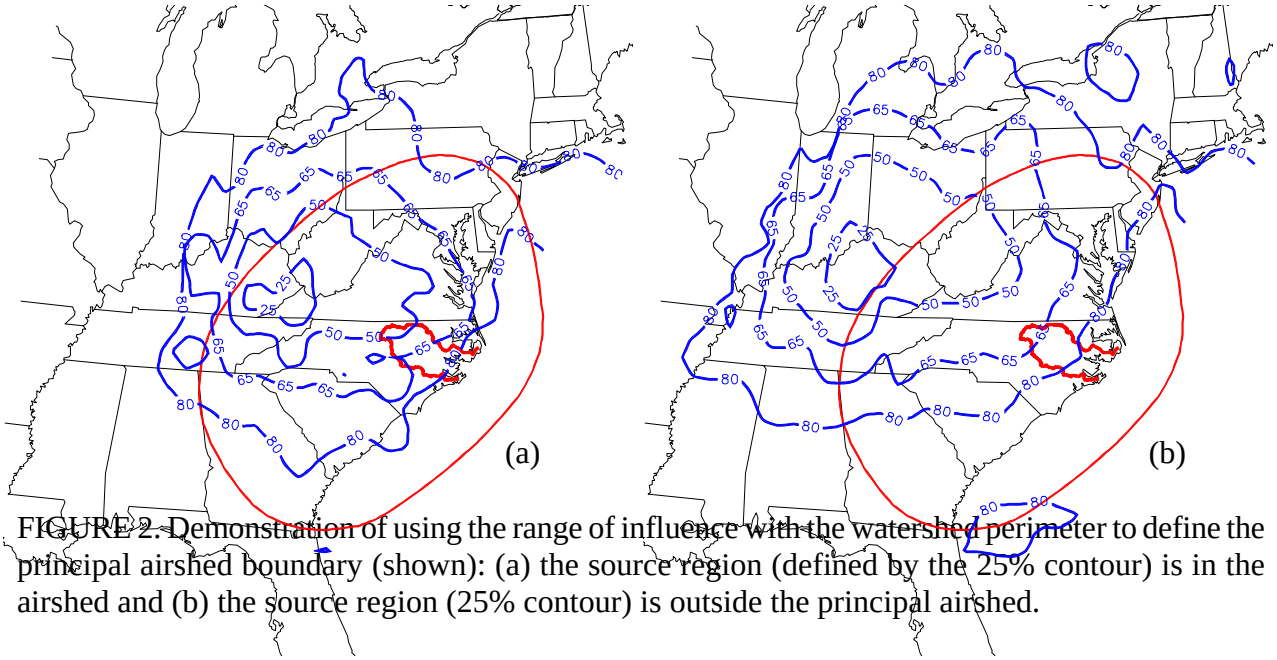

FIGURE 2. Demonstration of using the range of influence with the watershed perimeter to define the principal airshed boundary (shown): (a) the source region (defined by the 25% contour) is in the airshed and (b) the source region (25% contour) is outside the principal airshed.

This approach results in a consistent, physical-chemical-meteorological definition of the airshed that can be applied anywhere, even where there are few emissions, and allows a consistent cross-comparison of airshed attributes for different airsheds. It is a concept of “were there emissions there” they would be sufficiently efficient in depositing to the watershed that they would be our first focus of attention. The principal airshed is the area to “look to” first and it is expected that a majority of the deposition to the watershed will be explained by the emissions emanating from the airshed.

RESULTS

Principal airsheds for oxidized-N and reduced-N have been defined for the Neuse River/Pamlico Sound system, using the procedures described above. These are shown in Figure 3. The characteristics of the airsheds are given in Table 1.

TABLE 1. Characteristics of the Neuse River/Pamlico Sound system principal airsheds

	Airshed Size (km^2)	Size Factor Over Watershed Area	Percent Deposition Explained	Airshed Emissions as % of E. North American	Percent Deposition Explained from NC Emissions
Oxidized Nitrogen:	665,600	25			
1990's NO_X emissions			63%	18 %	20%
Reduced Nitrogen:	406,400	15			
1985 NH_3 Emissions			50%	5.7 %	30%
1990 NH_3 Emissions			55%		
1996 NH_3 Emissions			60%	6.8%	45%

Both airsheds are large compared to the watershed. The factor of 25 for the oxidized-N airshed relative to the watershed area is very close to the median ratio for 20 estuaries that have been characterized in a similar manner (Valigura, *et al*. 2000; Paerl, *et al*. 2002). The percent of the oxidized-N deposition explained by NO_X emissions for many estuaries ranges from 60-70%. Thus, 63% is at the lower end of typical values, indicating long range transport is important for the Neuse/Pamlico system. Furthermore, emissions of NO_X from North Carolina explain only a small fraction (20%) of the oxidized-N deposition. Hence, transport of oxidized-N from outside the state is very important. The percent explained of reduced-N deposition from NH_3 emissions is surprisingly low, 55% for 1990 emissions. The percent explained increases from 50% to 60% with the large growth of confined animal operation emissions in eastern NC. NC NH_3 emissions went from explaining sixty percent of the 50% contribution from the principal airshed for 1985 emissions to explaining seventy-five percent of the 60% contribution for 1996 emissions. Even for reduced-N,

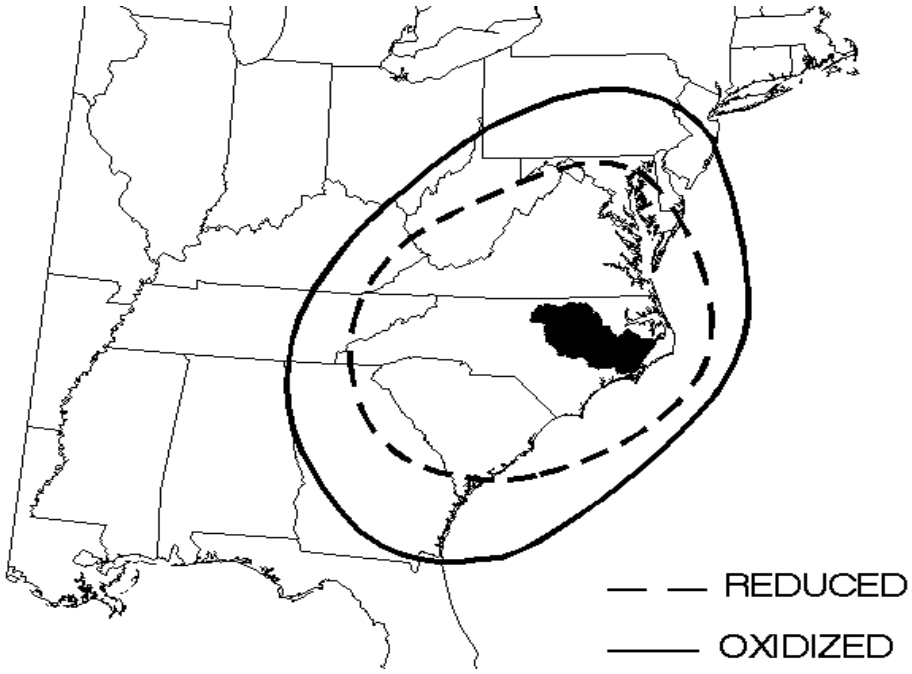

FIGURE 3. Principal airsheds of oxidized- and reduced-nitrogen for the Neuse/Pamlico system

long-range transport from outside NC is important, accounting for a majority of the reduced-N deposition to the Neuse/Pamlico system.

DISCUSSION AND CONCLUSIONS

The analysis with the Extended RADM indicates that airsheds are very large compared to watersheds, covering several states in geographic extent. Based on evaluation studies (e.g., Cohn and Dennis 1994) and the overprediction bias of 16-20% in the wet deposition comparisons on page 3, most likely the size of the oxidized-nitrogen principal airshed is underestimated in these studies. To date, most evaluation interpretations suggest that the range of influence is biased short. Sensitivity tests show that the grid size of the model, on the other hand, does not appear to make a difference in the range of influence for oxidized-nitrogen at the 50% and 65% contour distances.

It is not clear if the size of the reduced-nitrogen principal airshed is over- or under-estimated. Evaluation interpretations point in both directions, suggesting the range of influence is potentially biased short as well as potentially biased long. For example, the ammonia wet deposition bias suggests the range is biased short, as for oxidized-N, while the large grid size for ammonia hot spots is expected to result in a range of influence that is biased long. Errors potentially offset each other. The grid-size effect on the range of influence for reduced-nitrogen is the subject for further study.

Less than two-thirds of the nutrient deposition of both oxidized-N and reduced-N to the Neuse River/Pamlico Sound system is explained by emissions emanating from the principal airsheds. Given our understanding of the model, this result for oxidized-N percent contribution is

probably a valid perspective. The percent explained for reduced-N deposition is smaller than expected. The large grid size may be a contributor to this for high emissions areas like the coastal plains of North Carolina. Yet, the model reproduces many features of the local trends in reduced-N wet deposition. Fowler *et al.* (1998) studied a single livestock source with measurements and a model and concluded from the measurements that the local gradient of ambient NH_3 was extremely steep and that local deposition of NH_3 within 300 m of the source is a small fraction, 3% (and in some circumstances possibly up to 10%), of the local emission source. Asman (1998) estimated that 19%, $\pm$ a factor of 2, of the local emissions were dry-deposited within 2 km of the source when an NH_3 compensation point is assumed. The Extended RADM local grid-cell deposition calculations are consistent with this work, being closer to the results of Fowler *et al.* 1998. Our conclusion is that the reduced-N budget for North Carolina produced by the model is of the right order. As noted, the effect of grid size is an area for further investigation. Thus, for the reduced nitrogen system we learned that long-range transport is much more important than suspected. Long-range transport is definitely important to North Carolina estuaries for oxidized-N deposition.

Even though the principal airsheds are large, they are still too small for the scope of multi-media modeling involving nutrients because 30-40% of atmospheric deposition originates outside these airsheds. If the oxidized-N principal airshed is doubled in size, then the percent of oxidized-N deposition explained by the NO_X emissions increases to 80%. If the reduced-N principal airshed is doubled and then tripled in size, the percent of reduced-N deposition explained by the NH_3 emissions increases to 71% and 75%, respectively. The size of the tripled reduced-N principal airshed happens to be roughly the same as the doubled oxidized-N principle airshed and covers all or portions of 18 states east of the Mississippi River. To capture the long-range transport forming the background concentration affecting the east coast estuaries, we typically model the eastern third of the lower 48 states, including southeastern Canada. Thus, to provide a proper chemical context of nutrients, together with the hydrology, the atmospheric regionality is the order of at least one-third to one-half of the continental U.S., comparable in size to the drainage area of the Mississippi River Basin.

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