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Abstract: The current study uses case studies of model-predicted regional precipitation and wet ion deposition over 5-year periods to estimate errors in corresponding regional values derived from the means of site-specific values within regions of interest located in the eastern US. The mean of model-predicted site-specific values for sites within each region was found generally to overestimate the corresponding model-predicted regional wet ion deposition. On an annual basis across four regions in the eastern US, these overestimates of regional wet ion deposition were typically between 5 and 25% and may be more exaggerated for individual seasons. Corresponding overestimates of regional precipitation were typically <5%, but may be more exaggerated for individual seasons. Period-to-period relative changes determined from the mean of site-based model-predicted wet deposition for the current regional ensembles of sites generally estimated larger beneficial effects of pollutant emissions reductions in comparison to changes based on model-predicted regional wet deposition. On an annual basis site-based relative changes were generally biased low compared to regional relative changes: differences were typically <7%, but

they may also be more exaggerated for individual seasons. Spatial heterogeneities of the wet ion deposition fields with respect to the sparse monitoring site locations prevented the monitoring sites considered in the current study from providing regionally representative results. Monitoring site locations considered in the current study over-represent the geographical areas subject to both high emissions and high wet ion deposition and under-represent the geographical areas subject to low emissions and low wet deposition. Since the current case studies consider only those eastern US site locations that have supported concurrent wet and dry deposition monitoring, similar errors may be expected for dry and total deposition using results from the same monitoring site locations. Current case study results illustrate the approximate range of potential errors and suggest caution when inferring regional acid deposition from a network of sparse monitoring sites.

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Short abstract/summary ☒

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Ms. Ref. No.: ATMENV-D-08-01198R1
Title: Errors in Representing Regional Acid Deposition with Spatially Sparse Monitoring: Case Studies of the Eastern US Using Model Predictions
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Authors' Responses to Referees' Comments
NOTE: Reviewer #2's comments were found within the Editor's (H.B. Singh) 02-26-09 emailed letter, and our responses are given below. Reviewer #3's comments were not located in the Editor's 02-26-09 letter, but were found on the AE website when we started to resubmit our manuscript with revisions per Reviewer #2. We are confused about whether to respond to Reviewer #3's comments, but to be safe we have chosen to provide revisions and responses to Reviewer #3's comments following those for Reviewer #2, below.

Referees' comments:

Reviewer #2: The manuscript is definitely improved. A few minor revisions are still in order.

The authors are still using the means of sites within the region to represent regional deposition, whereas an estimate of deposition from the site-specific model using distance-weighted values for regional deposition would be more appropriate. They should at least mention that other forms of estimating deposition (rather than just taking the mean of the sites in a region) from the 34 sites might yield estimates closer to their regional model estimate.
Whether interpolation of the site data (e.g. by some form of distance weighting) would give an estimate of regional deposition that is less biased than the arithmetic average of the site values when compared to the regional deposition for the entire model output is uncertain. As interpolation (including arithmetic averaging) goes, the interpolated value generally falls between the minimum and maximum values of the data used to make the interpolation, and the interpolated values are weighted averages of the underlying data. Whether the average of all interpolated values is different from the arithmetic average of the underlying data depends on the system of weights that is employed in the interpolation.
Our paper attempts to see if the data derived from CASTNET locations (using the most likely first choice approach, arithmetic mean) can be used in a "regionally representative" sense. The paper does not address questions of network design (e.g. determining locations of sites that <u>would be</u> "regionally representative" in a defined sense) or the "sphere of influence" for particular sites or collections of sites. Although beyond the scope of our effort, these would be questions where interpolation in a distance-weighted sense would prove useful.
For our manuscript revision in response to the reviewer's comment, see revised lines 127-132.

In both the abstract and the conclusions the authors should explain a major reason why they get a positive bias for the two time periods with the site-specific model. Section 3.4 does a nice job of explaining this (CASTNet sites are located in areas near high emissions and deposition within a region, areas of low emissions and deposition in a region are under-represented at the CASTNet sites).
For our manuscript revision in response to the reviewer's comment, see revised lines 26-28 and 372-374.

Table 2 shows a bias for changes during the two time periods, but in general these biases (relative differences) are very low. Annual biases are <5% in most cases. This is important in terms of assessing the impact of air pollutant legislation. The authors need to stress that even though there are biases in the two approaches in terms of deposition, there is very little bias in the relative changes between the two time periods. This is an important result and should be in the abstract.
For our manuscript revision in response to the reviewer's comment, see revised lines 19-24 and 363-369.

Reviewer #3: I am glad that the authors found some of my earlier comments useful. I do believe the paper has been improved with the revisions. However, my basic reaction to the paper is still lukewarm. What came through to me was the focus on wet deposition, not the question of the utility of sparse networks for regional assessments. I believe this emphasis should be reversed. I take this opportunity to reiterate the "Focus" comments from my first review.

Related to my earlier "Analysis" comments, I view the addition of the CDFs (Figure 4) as a major improvement, but the authors have chosen to retain their analytical focus on the results of the mean comparisons. I find this puzzling in that the CDFs were much more informative to me than the means (and the authors have thoughtfully included the means on the CDFs). For example, note the striking fact that (at least for sulfate, nitrate, and apparently hydrogen) the site numbers are higher than the modeled grid numbers across all percentile levels. The text indicates that the corresponding plots for ammonium and precipitation did not show this same degree of separation; this suggests that it would be useful to present these CDFs as well. Another question that occurred to me was: "What do the CDFs look like for the latter time period?" Given the paper's comparison of the two periods on the basis of the means, I'd think the CDFs would be of at least as much interest along this line.

The authors also did not accept my suggestion (at least for this paper) of utilizing the NADP network, as opposed to only CASTNET sites. They attempted to justify this by pointing out that they wanted to extend the application of their results to total deposition. While I did not have a problem with their general discussion of the relationship between wet, dry, and total deposition, the fact remains that the paper analyzed only wet deposition. The arguments about extending the results to total deposition apply equally as well to an NADP based analysis as they do to a CASTNET only one.

Having said all this, I imagine the authors are responding along the lines of "Wait a minute -- it's our paper." And this is a valid point. The choice of emphasis and analytical approach are (within reason and journal guidelines) the prerogatives of the authors.

I did not find any major mistakes, though I do have some general comments and specific suggestions that are indicated below. I do believe the paper is suitable for publication in *Atmospheric Environment*. My comments above simply reflect my view that the paper could have been made stronger and more interesting. I hope that the authors will be able to utilize some of my suggestions in their future efforts.

General comments: The de-emphasizing of the model to monitoring comparison (Figures 3a and 3b) as a justification for the modeled value to modeled value comparison did eliminate the "glaring contradiction" I mentioned in my first review. However, I still feel as before that Figures 3a and 3b and the text justification that accompanies them is not needed. I still suggest that Figures 2 and the citation to the Grimm and Lynch publication can be used to emphasize that an "apples to apples" comparison is being conducted using a published, peer-reviewed model. For this paper, it is not Figures 3a and b but the "apples to apples" approach that needs to be sold.

Since Arkansas was included as part of the southern region, I suggest putting it on the maps in Figures 1 and 2.

With one exception, I strongly suggest that the phrase "acid deposition" be replaced throughout the paper with "atmospheric deposition." The one exception is on page 2 where the paper explicitly mentions the acid-base balance. Other than this, "atmospheric deposition" is the more appropriate term.

Specific comments:

Title: I suggest changing the title to "Errors in representing regional wet deposition with spatially sparse monitoring: Case studies of the eastern US using model predictions and CASTNET sites."
We disagree and consider our title to represent of our work more accurately than the title suggested above.

p.1, line 16: Change to "regional mean wet ion …."
We feel insertion of the term "mean" is unnecessary (and potentially misleading).

p. 2, lines 38-39: Change "Acid deposition" to "Deposition."
Using "Deposition" in this context lacks specificity and could refer to deposition of any constituent unrelated to acid deposition (e.g., Hg). We want to convey the concept of acid deposition to the reader to tie in with the remainder of the sentence, paragraph, and manuscript.

p. 3, line 60: Add a comma after "species."
OK, see revised line 65.

p. 4, line 77: Change "pollutants" to "pollutant."
OK, see revised line 82.

p. 4, lines 85-87: Rephrase "It is unlikely … sparse coverage …" to "This paper seeks to ascertain how well the sparse coverage…." Delete ", that currently available monitoring results."
The suggested change would alter our intended meaning as well as disrupt the flow within the Introduction section. This change would diminish the strength of our statement of need for this work and would preempt our goal statement (revised lines 105-109).

p.4, line 94: Delete "by in similar ways."
"by" has been omitted; see revised line 99.

p. 5, line 119: Replace the semicolon with a comma.
OK, see revised line 124.

p. 6, line 121: Replace the comma with a semicolon.
This passage has been rephrased, eliminating the need for the suggested revision; see revised lines 125-127.

p. 6, line 136: Replace "the cited studies." with "Sickles and Shadwick (2007a and b)."
OK, see revised line 146.

p. 7, line 153: I suggest adding a data source reference for PADMN, similar to those provided earlier for NADP and CASTNET. General comment on the discussion of the model used: The text on the bottom of page 6 and top of page 7 suggest to me that the model employed for this paper was the model found in the Grimm and Lynch (2004) article. I took the rather detailed discussion on the rest of page 7 to be a description of what was done in that model. However, page 9 states (line 209) that the 2004 Grimm and Lynch model was modified for this current paper. Are the modifications to the Barnes algorithm described on page 7 the modifications that generated the model used here? If so, what other modifications were done (if any)? If my initial reading of page 7 is correct, what was done to modify the 2004 version of the model. The discussion on pages 6 and 7 should be rephrased to clarify what changes were made to the Grimm and Lynch (2004) model before it was applied in this paper.
The most recently published version of the model employed in the current study is Grimm and Lynch (2004); however, models evolve, and this model has evolved since it was submitted for publication in 2002. The essential change in the current incarnation of the 2004 version of the Grimm and Lynch model accommodates the inclusion of more comprehensive radar-based precipitation (which were not available in the earlier version), as described in the text.

p. 8, lines 180 and 181, and p. 9, line 189: What was done with boundary cells that crossed over either a regional or circular boundary?
Cells were included when their centroids were contained within their corresponding regions of interest; see revised lines 191 and 198.

p. 8, lines 182 and 183: Delete "and are termed … comparisons." I'd simply call these baseline values. The authors should not use the word "true" in referring to any of the values; all comparisons are being done with modeled values (or summary numbers calculated from modeled values). Using the word "true" is misleading in the context of the paper; many readers may equate it with "monitored." This creates an impediment to the reader being able to recognize the advantages of the model-to-model (apples to apples) comparisons that are actually being done.
OK, see revised line 192.

p. 9, line 193: Define explicitly how the relative standard deviations are calculated.
The relative standard deviation is the standard deviation of an ensemble of data relative to their mean, expressed as a percentage. Since it is a commonly used index of variability, we did not include it in our original text. The text has been revised to include its definition (see revised lines 203-204).

p. 9, lines 192-204: Somewhere in this discussion of spatial variability, it is worth noting that the values mentioned are underestimates because they do not account for spatial correlation.
OK, see revised lines 204-205.

p. 9, line 210 and p.10, line 211: I took the 17% to refer to the quarterly time frame and 10% to refer to the annual. But then this is followed by three variables: precipitation, sulfate, and nitrate. Did all three have the same estimation errors within quarters or years? This should be clarified.
This is accurate as stated; see Grimm and Lynch (2004) for more details on the bias by species.

p. 10, lines 212-214: Are 10% and 17% mentioned in the previous sentence averages from this cross-validation exercise?
Yes; see Grimm and Lynch (2004) for more details.

p. 11, line 252: Delete "and substantial." I believe this whole paragraph calls for a bit more circumspection. Some of the biases reported in Table 1 would be characterized as "substantial" but many would not; personally, I don't feel that, generally speaking, substantial biases were found, particularly in light of what's reported in Figures 3a and b.
Biases are tabulated to permit each reader to develop an opinion. The word "substantial" is an opinion-based adjective; we have inserted "sometimes" before "substantial," see revised line 263.

p. 11, line 254: Rephrase "… studies generally overestimates the true regional wet ..." to "… studies may overestimate the regional wet …." As with the immediately preceding comment, I think this rephrasing better reflects what has been found. Again, don't use the word "true."
Again, biases are tabulated to permit each reader to develop an opinion. In 38 of the 40 annual cases the biases were positive; since overestimates occurred 95% of the time, the modifier "generally" seems appropriate. The word "true" has been omitted.

p. 11, lines 255-256: The annual numbers reported in Table 1 for the ions range from -2% to 27%. Seven of these are 20% or above (with five of these due to hydrogen), seventeen are between 10% and 17%, and eight are less than 10%; the median is 15%. A better summary sentence for these two lines is "Annually across the four regions in the eastern US, this bias is typically between 10% and 17%, with a range of -2% to 27%. Biases may be larger for individual seasons."

Again, biases are tabulated to permit each reader to develop an opinion. Notice that for the annual numbers, 73% of the time (29 of 40 cases) biases fall between 5 and 25%. In contrast, in 17 of 40 cases (only 43% of the time), biases fall between 10 and 17%. Thus, our statement that "…bias is typically between 5 and 25%…" seems to be at least as accurate as that suggested above.

p. 12, line 259: Change "… season, and the …" to "… season, but the …."
OK, see revised line 270.

p. 12, line 260: Before the last sentence of the paragraph, insert the sentence on the CAA of Section 3.3 (p. 13, lines 288-291) here. **This suggested change would eliminate the background discussion necessary as a foundation later in the development of section 3.3. Instead, we have inserted revised lines 137-139.**

p. 12, line 277: Change "… CASTNET site locations." to " CASTNET site locations alone."
"alone" is unnecessary.

p. 13, lines 280-282: As I mentioned above, based on these comments, I strongly suggest adding Figures 4c and d for the ammonium and precipitation CDFs. I also strongly suggest adding Figures 5a-d that would correspond directly to Figures 4a-d, but for the later time period. Of course, additional text discussing these figures will be needed as well.
This would add unnecessary figures and text to a manuscript that is currently within the journal guidelines without adding substantially to the intended message of our paper.

p. 13, lines 288-291: As noted above, move the CAA sentence to p. 12.
This suggested change would eliminate the background discussion necessary as a foundation in the development of section 3.3, see above.

p. 13, lines 300-301: Given the definition of relative change presented here, it would be worth noting that this quantity would be expected to be less than zero. This would help with the interpretation of Table 2 and the understanding of the discussion of it (p. 14, lines 310-319).
Although we had no expectation, we feel that this observation is noted in revised lines 328-330.

p. 313, line 313: Change "… overestimated the …" to "… estimated a larger …."
OK, see revised lines 322-323.

pp.14-15: I suggest moving the entire Section 3.4 to the Conclusions.
We feel that it is inappropriate to include material in the "Conclusions" that has not been developed/discussed within the text. Conclusions drawn from section 3.4 are presented in the "Conclusions."

p. 15, line 348: Delete the word "true."
OK, see revised line 359.

p. 18: The caption for Figure 3 needs to explain the lines on the figures.
OK, see the revised list of figure captions.

Errors in Representing Regional Acid Deposition with Spatially Sparse Monitoring: Case Studies of the Eastern US Using Model Predictions

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Abstract. The current study uses case studies of model-predicted regional precipitation and wet ion deposition over 5-year periods to estimate errors in corresponding regional values derived from the means of site-specific values within regions of interest located in the eastern US. The mean of model-predicted site-specific values for sites within each region was found generally to overestimate the corresponding model-predicted regional wet ion deposition. On an annual basis across four regions in the eastern US, these overestimates of regional wet ion deposition were typically between 5 and 25% and may be more exaggerated for individual seasons. Corresponding overestimates of regional precipitation were typically <5%, but may be more exaggerated for individual seasons. Period-to-period relative changes determined from the mean of site-based model-predicted wet deposition for the current regional ensembles of sites generally estimated larger beneficial effects of pollutant emissions reductions in comparison to changes based on model-predicted regional wet deposition. On an annual basis site-based relative changes were generally biased low compared to regional relative changes: differences were typically <7%, but they may also be more exaggerated for individual seasons. Spatial heterogeneities of the wet ion deposition fields with respect to the sparse monitoring site locations prevented the monitoring sites considered in the current study from providing regionally representative results. Monitoring site locations considered in the current study over-represent the geographical areas subject to both high emissions and high wet ion deposition and under-represent the geographical areas subject to low emissions and low wet deposition. Since the current case studies consider only those eastern US site locations that have supported concurrent wet and dry deposition monitoring, similar errors may be expected for dry and total deposition using results from the same monitoring site locations. Current case study results illustrate the approximate range of potential errors and suggest caution when inferring regional acid deposition from a network of sparse monitoring sites.

Keywords: Spatial representation, Deposition, Sulfate, Nitrate, Ammonium, Hydrogen Ion, Precipitation

1. Introduction

Chemical species contained in air pollutant emissions are frequently transformed through chemical and physical processes in the atmosphere before they are deposited to the surface of the Earth. Acid deposition occurs when chemical species that can alter the acid-base balance of ecosystems are transferred from the atmosphere to the surface. Acid deposition is also spatially and temporally variable, stressing both landscapes and ecosystems, and can occur by dry deposition of gases and aerosols and by wet deposition from clouds, fogs and precipitation. The effects of acid deposition are diverse, ranging from eutrophication of coastal waters to acidification of lakes, streams, and forest soils with attendant reductions in species diversity (Dennis et al., 2007).

Acid deposition monitoring networks have been established to meet various goals (e.g., determining trends, spatial patterns, and site-specific behaviour). The Clean Air Status and Trends Network (CASTNET) estimates dry deposition using air quality and meteorological data monitored at more than 90 sites (53 east of the Mississippi River) by the US Environmental Protection Agency (US EPA) and the National Park Service (NPS). An archive of these data and estimates of dry deposition is maintained by the US EPA (http://www.epa.gov/castnet/). For the period between 1990 and 2004, several recent studies indicate that the number of dry deposition sites in the eastern US with relatively complete data records range between 30 and 40 (Holland et al., 1999 and 2004; Baumgardner et al., 2002; Mueller, 2003; Sickles and Shadwick, 2007a and b). Wet deposition is monitored in the United States (US) at more than 250 National Atmospheric Deposition Program / National Trends Network (NADP/NTN) sites (127 east of the Mississippi River). An archive of these and related data is maintained by NADP (http://nadp.sws.uiuc.edu/).

For a given chemical species, the sum of dry deposition and wet deposition of that species is termed total deposition. Although dry and wet deposition monitoring sites are frequently collocated, there is a relatively small number of paired CASTNET dry and NADP wet deposition monitoring sites with a sufficiently complete record to permit long-term examination of observed total deposition at these paired sites. Since the dry deposition network has fewer sites (see above), the number of paired sites available to yield observed total deposition is usually limited by the existing dry deposition monitoring sites.

Deposition of a relevant chemical species (e.g., sulfur or nitrogen), represents the amount of that chemical species deposited to an area, or a region, over a period of time and is often expressed in units of kg/ha y. Acid deposition is frequently considered to be a regional stressor of landscapes and ecosystems, where the region may range in size from a small watershed or forest to a multistate area. Although the dry and wet deposition monitoring results, noted earlier, are site-specific, they are sometimes aggregated across sites in an attempt to represent the regions where the sites are located (e.g., see US EPA, 2009). Large regions often have large heterogeneities in their spatial distribution of land cover (e.g., crops to forests), terrain (e.g., flat to montane), pollutant species (e.g., SO_2, NO_2, and NH_3), pollution sources (e.g., agricultural, mobile, and industrial), and pollutant emissions density. These varied features influence the magnitude and spatial distribution of both dry and wet deposition and may be responsible for sizeable differences reported for nearby monitors (Brook, et al., 1997; Reid et al., 2001; Gego et al., 2005). In those cases where quantification of total deposition to a large (e.g., multistate) region is desired, it is tempting to use the mean (or some other measure of central tendency) of the total deposition values monitored within a region of interest as an index of its regional value (e.g., see US EPA, 2009). It is unlikely, given the previously noted sparse coverage provided by available dry deposition monitoring sites, that currently available monitoring results can be used to provide accurate representations of regional total deposition. It is also unclear if observed changes in regional total deposition aggregated from such a sparse network are regionally representative.

The NADP was established approximately a decade prior to CASTNET. Wet deposition monitoring results from NADP provided guidance in the design of CASNET for the monitoring of dry deposition (Holland et al., 1994). At any given location both dry and wet deposition of common species are influenced in similar ways by common sources and meteorological patterns. As a result, site-specific dry deposition may be estimated to be very roughly proportional to collocated wet deposition (Sullivan et al., 2008). While wet deposition generally exceeds dry deposition at sites in the eastern US, the proportion varies depending on species, site location, and season (Sickles and Shadwick, 2007a). Multi-year finely spatially resolved model estimates of dry deposition are currently not available; however, such estimates are available for wet deposition. The goal of this paper is to use case studies of model-predicted regional wet deposition to estimate errors in regional wet deposition and in temporal changes of wet deposition derived from the means of model-estimated values obtained for the specific sites where only total deposition has been monitored in the eastern US. Although strictly applicable only to the interpretation of wet deposition, the results are given to provide a basis for the inferential interpretation of dry and total deposition monitoring results.

2. Approach

2.1. Description

The goal of the current study is to compare 5-year averages of model-predicted (defined in section 2.3), finely spatially resolved (i.e., nominally 330-m) regional precipitation and wet sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+) and hydrogen (H^+) ion deposition for four regions in the eastern US with the corresponding means of values obtained from the same model-predicted precipitation and wet ion deposition gridded surfaces but at specific monitoring site locations. Comparisons of period-average annual values and of period-average seasonal values are made. The term seasonal refers to precipitation and wet ion deposition values associated with climatic seasons (i.e., winter = December + January + February), while the term annual refers to the summation of the corresponding seasonal values. Model-predicted site-based wet deposition is considered at the specific locations where collocated monitoring of both dry and wet deposition (i.e., total) has occurred over the past 15 to 20 years. The mean of model-predicted site-specific values was adopted in the current study to estimate site-based regional wet deposition for subsequent comparison with regional model predictions of wet deposition. It is unclear how site-based regional wet deposition estimates based on more complex methods (e.g., distance-weighting) would compare to corresponding regional model predictions of wet deposition.

2.2. Monitoring data

Wet, dry and total deposition, derived from monitoring data collected at or near 34 CASTNET sites located in the eastern US, have been recently examined for the 5-year periods, 1990-1994 and 2000-2004 (Sickles and Shadwick, 2007a and b). Between 1990-1994 and 2000-2004, reductions of emissions densities and corresponding atmospheric concentration and deposition of oxidized sulphur and nitrogen species were reported. In these studies the eastern US was divided into four geographical regions, with 10 sites in the northeast, 10 sites in the midwest, 14 sites associated with the south, and 34 sites in the east, represented by the combination of the previous three regions. These regions and site locations (except for a southern site, located in Arkansas) are illustrated on maps of model-predicted annual wet SO_4^{2-} and NO_3^- deposition for 1990-1994 in Figs.1a and b.

For more information about the sites (e.g., terrain type, elevation, latitude, longitude), see Sickles and Shadwick (2007a and b).

2.3. Model-predicted precipitation and wet ion deposition

Model estimates of average annual and seasonal precipitation and wet ion deposition were made for the eastern US for the two 5-year periods, 1990-1994 and 2000-2004. The model employed is a moving neighborhood, weighted least squares regression algorithm that uses precipitation and wet ion concentration measurements along with elevation, slope and topographic aspect input derived from 3-arc-second US Geological Survey Digital Elevation Model (USGS DEM) output (Grimm and Lynch, 2004). Precipitation, measured daily at approximately 4400 National Oceanic and Atmospheric Administration (NOAA) sites in the eastern US, was used for years prior to 2001. Precipitation for subsequent years was derived from NOAA's radar-based Quantitative Precipitation Estimate data set after bias-correction according to NOAA's Global Historical Climatology Network measurements. A modified, three-pass Barnes (1964) objective analysis algorithm was applied to measurements of quarterly volume-weighted wet ion concentrations summarized from weekly precipitation samples collected at NADP/NTN sites and at Pennsylvania Atmospheric Deposition Monitoring Network (PADM) sites to estimate ion concentration in precipitation across the eastern US. The Barnes algorithm was modified by adjusting the weighting parameter for each 0.5-degree geographic sub-domain to minimize the root mean square error (RMSE) of first-pass estimates at the six nearest NADP/NTN and PADM sampling locations. The algorithm applied to the concentration data does not directly account for elevation, slope aspect, or "rain shadow" effects. However, the localized terrain regression algorithm used to estimate quarterly precipitation does directly account for both elevation and slope-aspect effects and, consequently, also accounts for local "rain shadows." These concentration and precipitation estimates were then combined in the model, accounting for location, elevation, and slope to produce annual and seasonal deposition estimates at 12-arc-second (i.e., nominally 330-m) resolution for each year.

In the current study, each year's model output for the corresponding periods, 1990-1994 and 2000-2004, was combined using Arc/Info to produce period-average annual and seasonal gridded surfaces of precipitation and wet ion deposition for the eastern US (e.g., see Fig. 1). Period-average model estimates were also subsequently used to create corresponding gridded surfaces of relative change between 1990-1994 and 2000-2004 of average annual and seasonal precipitation and wet ion deposition, where relative change = 100 [value(2000-2004) – value(1990-1994)] / value(1990-1994). Example maps of the 1990-1994 to 2000-2004 relative change in wet SO_4^{2-} and NO_3^- deposition for the four study regions are shown in Figs. 2a and b.

Regional coverages for each of the four study regions (east, northeast, midwest, and south) were used to obtain corresponding period-average maps of annual and seasonal precipitation and wet ion deposition. Example maps of model-predicted annual wet SO_4^{2-} and NO_3^- deposition for 1990-1994 for the four study regions are shown in Figs.1a and b. For the time period of interest, regional estimates of annual and seasonal precipitation and wet ion deposition were determined by averaging across all grid cells with centroids contained within each region of interest. These regional estimates are considered to be baseline values in subsequent comparisons.

Site-specific coverages for 2-km diameter circles surrounding each of the 34 monitoring locations (latitude and longitude) were also determined. Using the period-average gridded surfaces of model-predicted seasonal precipitation and wet ion deposition, estimates of site-specific annual and seasonal precipitation and wet ion deposition were determined by averaging across the 25 to 30 grid cells with centroids contained within each 2-km site circle of interest for each of the two periods.

In the current study, one measure of temporal (i.e., year-to-year) variability is based on the relative standard deviations (%) for the period means of model-predicted annual average precipitation and wet ion deposition for each region, where relative standard deviation = 100[standard deviation/mean]. Note that covariance between cells induced in the interpolation by data has not been considered in this calculation. These values were generally <10%, ranging regionally between 3% for precipitation during 1990-1994 in the east to 11% for wet NH_4^+ deposition during 2000-2004 in the northeast. Using period means of annual average precipitation and ion deposition for each grid cell, spatial (i.e., grid cell-to-grid cell) variability was also determined for each study region. As suggested in Figs. 1a and b, spatial variability within regions may be considerable. Based on the relative standard deviations determined by averaging across all grid cells within each region of interest, these values were generally <30%, but ranged between 13% for precipitation during 1990-1994 in the midwest to 56% for H^+ during 2000-2004 in the midwest. As expected, the spatial variability within the 2-km site circles was small, with mean relative standard deviations <2%.

3. Results and Discussion

3.1. Model applicability

Using an earlier version of the current model, Grimm and Lynch (2004) reported mean quarterly and annual estimation errors of 17 and 10%, respectively, for modeled estimates of precipitation and wet SO_4^{2-} and NO_3^- deposition to the eastern US. These estimation errors were calculated by individually withholding observations at each NADP/NTN site location and estimating the value at the withheld location using the remaining observations.

The following comparisons were performed to provide additional documentation of the fidelity of the model for subsequent application in the current study (see sections 3.2 and 3.3). For the two 5-year periods under consideration in the current study, 19 NADP sites (among the 34 paired CASTNET-NADP sites noted above in section 2.2), were both located within 2 km of their paired CASTNET sites and had sufficiently complete data records to permit analysis. Five-year seasonal monitoring values from these 19 NADP sites determined in earlier studies (Sickles and Shadwick, 2007a and b) were identified for subsequent comparison with model estimates. This yielded 152 (i.e., 19 sites x 4 seasons x 2 periods) 5-year average seasonal monitoring values of each variable (i.e., precipitation and wet ion deposition) for subsequent comparison with modeled results. Model-predicted site-specific seasonal precipitation and wet ion deposition for the 19 site locations under consideration were identified (see section 2.3) and paired with the corresponding previously identified monitoring values. Scatter plots were prepared for each variable, and examples for wet SO_4^{2-} and NO_3^- deposition are shown in Figs. 3a and b. Both the square of the correlation coefficient and the slope of an unweighted linear regression forced through the origin were determined for each variable. The corresponding r^2 and slope values are: precipitation, 0.72 and 1.03; wet SO_4^{2-} deposition, 0.89 and 1.00; wet NO_3^- deposition, 0.86 and 1.01; wet NH_4^+ deposition, 0.87 and 0.96; and wet H^+ deposition, 0.90 and 0.98. These results suggest that the Grimm and Lynch (2004) model provides estimates of precipitation and wet ion deposition that are sufficient for the main purpose of this paper (i.e., investigation of errors in representing regional acid deposition with sparse monitoring).

3.2. Comparison of modeled regional versus mean of modeled site-specific precipitation and wet ion deposition

Regional estimates of annual and seasonal precipitation and wet ion deposition were determined by averaging for each period and season across all grid cells within each region (see section 2.3). Site-specific estimates of annual and seasonal precipitation and wet ion deposition were also determined as described in section 2.3. The means of these model-predicted site-specific values across all sites within each region were determined for each period and season. These two sets of values were compared using relative difference (%) with the regional value as the standard. The resulting relative difference between model-predicted regional value and mean of model-predicted site-specific values for all sites within that region are summarized in Table 1 for each region, period, season, and variable.

These findings generally show positive and sometimes substantial biases for wet ion deposition. This indicates that the use of the mean of site-based values as proxies for the regional values in the current case studies generally overestimates the baseline regional wet deposition. Annually across the four regions in the eastern US, this bias is typically between 5 and 25% and may be more exaggerated for individual seasons. Although precipitation is generally biased in the same direction, annually in the eastern US the magnitude of the bias for precipitation is typically <5% and may also be more exaggerated for individual seasons. The order varies with region and season, but the magnitude of the bias is roughly ordered $H^+ > SO_4^{2-} > NO_3^- > NH_4^+ >$ precipitation. The magnitude of the bias also appears to be larger for the more polluted time interval, 1990-1994, than for the less polluted 2000-2004.

Cumulative distribution functions (e.g., Rohatgi, 1976) were also prepared to compare distributions of modeled gridded values with distributions of samples drawn from the modeled gridded values at CASTNET site-specific grid locations. Example cumulative distribution functions for wet SO_4^{2-} deposition and wet NO_3^- deposition are shown in Figs. 4a and b for the annual period 1990-1994 for the eastern US and the 34 CASTNET site locations in the east. Solid lines represent the cumulative distribution functions for modeled gridded values, and individual points represent the cumulative distribution functions for the corresponding CASTNET site locations. In both illustrations the cumulative distribution functions for the CASTNET site locations lie largely to the right of the cumulative distribution functions for modeled gridded values, and both the mean and median values for the CASTNET site locations exceed those for the corresponding modeled gridded distributions. This suggests that higher values from the modeled gridded distributions are over represented (and that the lower values are under represented) by corresponding distributions from CASTNET site locations. Other species, periods, seasons and regions are not shown due to space limitations. However, comparisons of paired cumulative distribution functions for wet H^+ deposition are similar to those shown in Figs. 4a and b for SO_4^{2-} and NO_3^-. In contrast, comparisons for wet NH_4^+ deposition and precipitation show smaller differences between their respective paired cumulative distribution functions.

3.3. Comparison of 1990-1994 to 2000-2004 relative changes in regional precipitation and wet ion deposition inferred from modeled regional versus mean of modeled site-based values

It is often important to track changes in ecological stressors over time to evaluate the impacts of legislatively mandated changes in pollutant emissions. The Clean Air Act Amendments and other legislation established controls that resulted in reductions of SO_2 and NO_x emissions between 1990 and 2002 in the eastern US of 39% and 22%, respectively (Sickles and Shadwick, 2007b). Between the two periods, 1990-1994 and 2000-2004, relative changes in regional wet ion deposition are expected to be associated with some of the cited changes in pollutant emissions. Errors associated with quantifying

1990-1994 to 2000-2004 relative changes in regional precipitation and wet ion deposition are examined in this section.

Regional estimates of annual and seasonal precipitation and wet ion deposition were determined by averaging across all grid cells within each region for 1990-1994 and for 2000-2004 (see section 2.3). These values were used to compute the regional 1990-1994 to 2000-2004 relative changes (%) for each region and season, where relative change = 100 [value(2000-2004) – value(1990-1994)] / value(1990-1994). Site-specific estimates of annual and seasonal precipitation and wet ion deposition were also determined as described in section 2.3. The means of these model-predicted site-specific values across all sites within each region were determined for 1990-1994 and for 2000-2004. These values were used to compute site-based 1990-1994 to 2000-2004 relative changes for each region and season (as defined above). Differences between these two estimates of 1990-1994 to 2000-2004 relative change are summarized for each region and season in Table 2 for each variable.

Period-to-period relative changes determined from the mean of site-based modeled deposition for the current regional ensembles of sites generally estimated larger beneficial effects of pollutant emissions reductions in comparison to changes based on modeled regional estimates. Site-based relative changes were generally biased low compared to regional relative changes, with differences typically <7% on an annual basis. The magnitude of the bias varies with precipitation, ion, and season and may be more exaggerated for other regions and seasons (e.g., midwest in summer; northeast in winter;

and south in fall). The direction of this bias in period-to-period change is consistent with the earlier finding of a larger bias in regional site-based wet ion deposition estimates for the more polluted than for the less polluted time intervals.

3.4. Site representativeness

Over half of the electric generating units targeted by the Clean Air Act Amendments for SO_2 and NO_x emissions reductions are located in a six-state source region along the Ohio River (IL, IN, KY, OH, PA and WV). Fifteen of the 34 monitoring sites considered in the current study are located in this region. Examples of model-predicted annual wet SO_4^{2-} and NO_3^- deposition for 1990-1994 for the four study regions are shown in Figs.1a and b. Examination of Fig.1 reveals that during 1990-1994 approximately 28 of the 34 sites (i.e., >80%) are located in areas of high modeled wet SO_4^{2-} deposition (median ≈ 18.5 kg/ha y) and approximately 25 of the 34 sites (i.e., >70%) are located in areas of high modeled wet NO_3^- deposition (median ≈ 12.1 kg/ha y). It appears that the 34 monitoring site locations considered in the current study over-represent the geographical areas subject to both high emissions and high wet ion deposition and under-represent the geographical areas subject to low emissions and low wet deposition. Maps of period-average model-predicted wet ion deposition for each ion during each period are spatially heterogeneous with respect to the monitoring site locations (not shown due to space limitations). In the current study, ensembles of sparse monitoring site locations were used to represent large spatially heterogeneous model-predicted regional precipitation and deposition fields. Thus, sparse monitoring in nonrepresentative locations of spatially heterogeneous variable fields yielded the biases shown in sections 3.2 and 3.3 for each of the variables.

4. Conclusions

The current case studies use 5-year averages of model-predicted, finely spatially resolved (i.e., nominally 330-m) regional precipitation and wet SO_4^{2-}, NO_3^-, NH_4^+ and H^+ ion deposition for four regions in the eastern US. The mean of model-predicted site-specific values of wet ion deposition for sites within each region was found generally to overestimate the corresponding model-predicted regional wet ion deposition. On an annual basis across four regions in the eastern US, these overestimates of regional wet ion deposition were typically between 5 and 25% and may be more exaggerated for individual seasons. Corresponding overestimates of regional precipitation were typically <5%, but may be more exaggerated for individual seasons. Period-to-period relative changes determined from the mean of site-based modeled wet deposition for the current regional ensembles of sites generally estimated larger beneficial effects of pollutant emissions reductions in comparison to changes based on modeled regional estimates. On an annual basis site-based relative changes were generally biased low compared to regional relative changes: differences were typically <7%, but they may also be more exaggerated for individual seasons. Spatial heterogeneities of the model-predicted wet ion deposition fields with respect to the sparse monitoring site locations prevented the monitoring sites considered in the current study from providing regionally representative results. Monitoring site locations considered in the current study over-represent the geographical areas subject to both high emissions and high wet ion deposition and under-represent the geographical areas subject to low emissions and low wet deposition. Since the current case studies consider only those eastern US site locations that have supported concurrent wet and dry deposition monitoring, similar errors may be expected for dry and total deposition using results from the same monitoring site locations. Current case study results illustrate the approximate range of potential errors and suggest caution when inferring regional acid deposition from a network of sparse monitoring sites.

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Fig. 1. Maps of model-predicted annual wet ion deposition (kg/ha y) for 1990-1994 for the northeast, midwest, south, and eastern regions of the US with CASTNET monitoring site locations identified: (a) sulfate; (b) nitrate.

Fig. 2. Maps of relative change (%) from 1990-1994 to 2000-2004 of model-predicted annual wet ion deposition for the northeast, midwest, south, and eastern regions of the US with CASTNET monitoring site locations identified: (a) sulfate; (b) nitrate.

Fig. 3. Paired matched 5-year average seasonal wet ion deposition (kg/ha y) from CASTNET/NADP monitoring results and from model estimates for 1990-1994 and 2000-2004: (a) sulfate; (b) nitrate. Lines indicate the relative difference (%) from perfect agreement (i.e., the 1:1 line).

Fig. 4. Cumulative distribution functions of annual wet ion deposition (kg/ha y) for 1990-1994 for modeled gridded values for the eastern US (solid lines) and the 34 eastern CASTNET site locations (individual points): (a) sulfate; (b) nitrate. Horizontal lines represent the 25^{th}, 50^{th} (median), and 75^{th} percentiles, and M represents mean values.

Table 1
Click here to download Table: Errors-Case Studies Table1.xls

Table 1. Relative difference (%) between model-estimated regional value and mean of model-estimated site-specific values for all sites within that region for specific periods and seasons

Region	Period	Season	100 (Mean Modeled Site-Specific Value - Modeled Regional Value) / Modeled Regional Value				
			PPTN	SO4	NO3	NH4	H+
East	1990-1994	Annual	2	21	16	10	25
East	2000-2004	Annual	0	15	10	7	20
East	1990-1994	W	5	17	20	7	23
East	2000-2004	W	3	10	12	4	17
East	1990-1994	Sp	6	21	21	9	30
East	2000-2004	Sp	3	16	13	6	25
East	1990-1994	Su	-2	23	12	9	25
East	2000-2004	Su	-4	18	6	8	20
East	1990-1994	F	3	19	14	16	22
East	2000-2004	F	0	12	10	6	18
MW	1990-1994	Annual	3	15	6	1	23
MW	2000-2004	Annual	4	14	5	-2	27
MW	1990-1994	W	14	24	9	8	25
MW	2000-2004	W	14	25	8	4	28
MW	1990-1994	Sp	7	15	8	-1	24
MW	2000-2004	Sp	0	9	0	-6	23
MW	1990-1994	Su	-2	15	7	-2	24
MW	2000-2004	Su	2	17	7	2	30
MW	1990-1994	F	-1	10	1	7	18
MW	2000-2004	F	5	12	7	-2	27
NE	1990-1994	Annual	4	17	12	16	15
NE	2000-2004	Annual	2	17	8	15	13
NE	1990-1994	W	8	20	10	20	15
NE	2000-2004	W	-2	8	-3	6	4
NE	1990-1994	Sp	4	16	13	18	13
NE	2000-2004	Sp	1	16	8	18	11
NE	1990-1994	Su	7	21	17	18	20
NE	2000-2004	Su	7	22	14	17	21
NE	1990-1994	F	0	11	5	9	9
NE	2000-2004	F	0	12	7	12	11
SO	1990-1994	Annual	6	21	19	13	26
SO	2000-2004	Annual	1	12	10	9	16
SO	1990-1994	W	9	17	26	8	25
SO	2000-2004	W	7	9	21	8	20
SO	1990-1994	Sp	11	26	30	14	39
SO	2000-2004	Sp	8	18	19	12	28
SO	1990-1994	Su	-3	20	6	7	20
SO	2000-2004	Su	-7	12	0	7	11
SO	1990-1994	F	6	23	19	25	24
SO	2000-2004	F	0	7	5	7	10

Region: East=MW+NE+SO, MW=midwest, NE=northeast, SO=south;
Season: Annual=W+Sp+Su+F, W=winter (December+January+February), Sp=spring, Su=summer, F=fall.

Table 2
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Table 2. Differences (%) between model-estimated regional relative 1990-1994 to 2000-2004 change and model-estimated site-based relative change

Region	Season	Difference in Estimates of 1990-1994 to 2000-2004 Change (%)				
		Site-Based 1990-1994 to 2000-2004 Change - Regional 1990-1994 to 2000-2004 Change				
		PPTN	SO4	NO3	NH4	H+
East	Annual	-2	-4	-5	-3	-3
East	W	-1	-5	-6	-3	-3
East	Sp	-3	-4	-7	-3	-3
East	Su	-1	-3	-5	-1	-3
East	F	-3	-5	-3	-8	-3
MW	Annual	1	0	-1	-3	2
MW	W	0	1	-1	-4	1
MW	Sp	-7	-5	-8	-7	0
MW	Su	4	1	0	4	2
MW	F	6	1	5	-8	5
NE	Annual	-3	0	-3	-1	-1
NE	W	-8	-7	-9	-12	-6
NE	Sp	-2	0	-4	0	-2
NE	Su	0	1	-2	-1	0
NE	F	0	0	1	2	1
SO	Annual	-4	-7	-7	-4	-6
SO	W	-2	-5	-3	0	-3
SO	Sp	-3	-6	-7	-3	-6
SO	Su	-4	-6	-6	-1	-6
SO	F	-7	-11	-11	-16	-9

See Table 1 for definition of abbreviations.

Figure 1a
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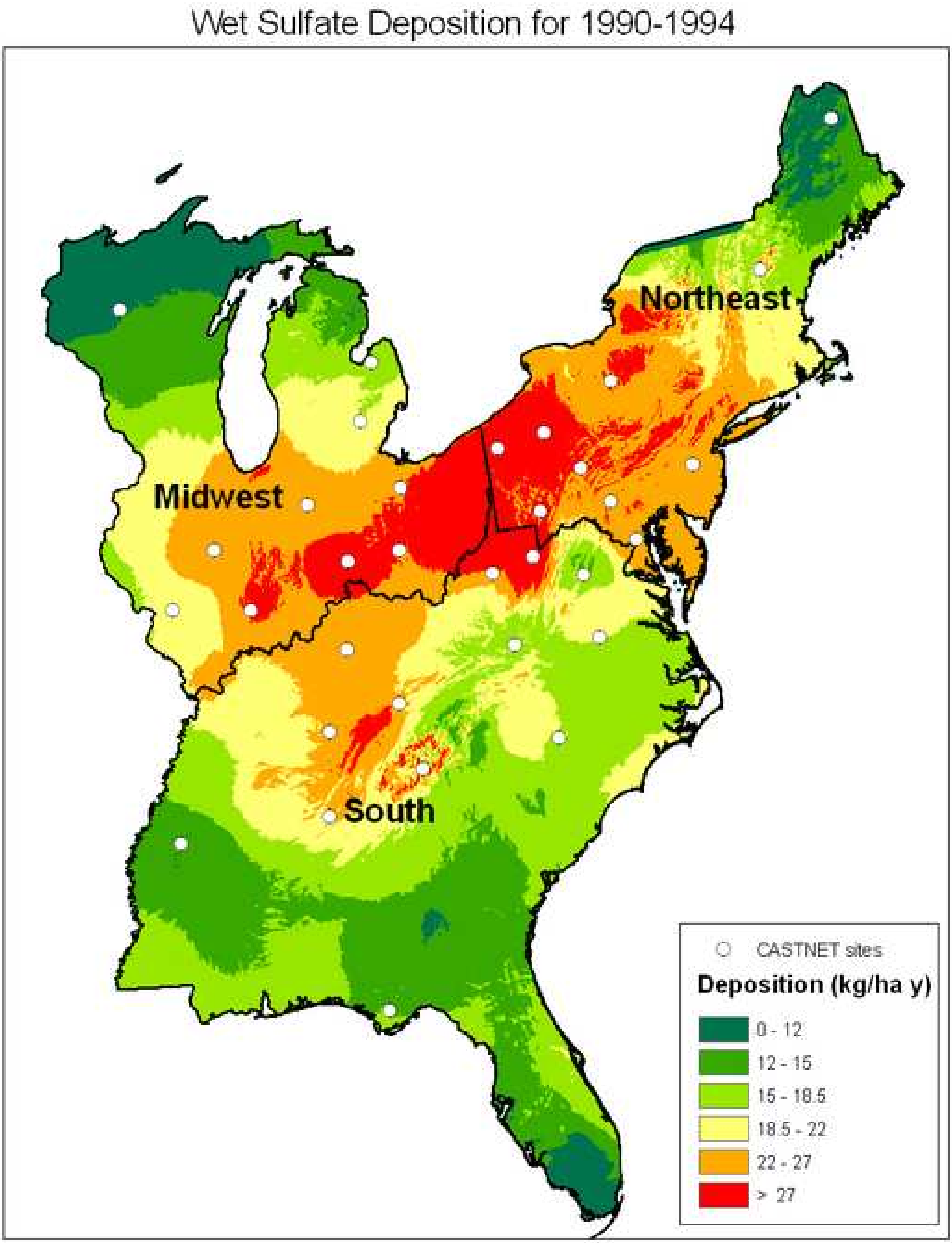

Figure 1b
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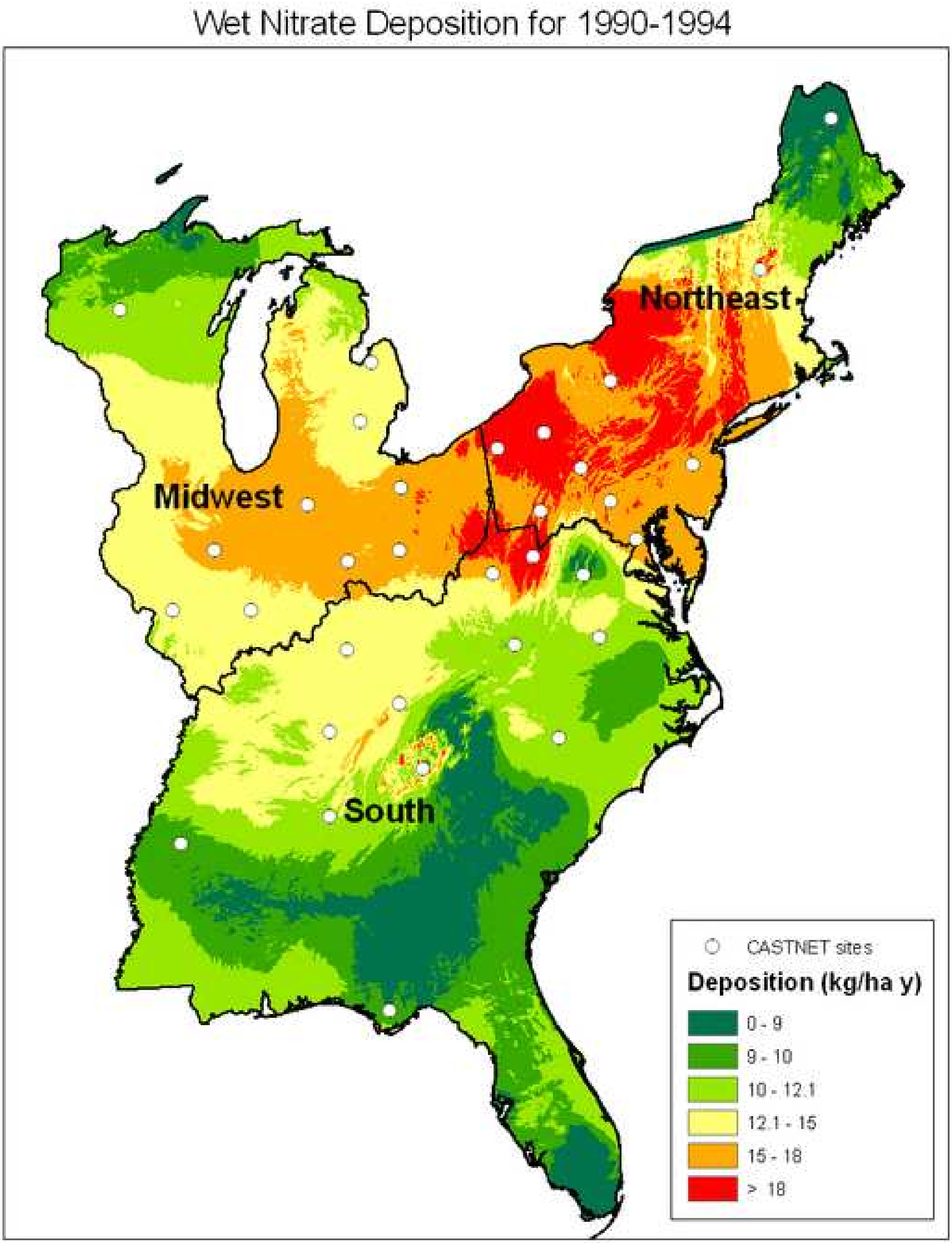

Figure 2a
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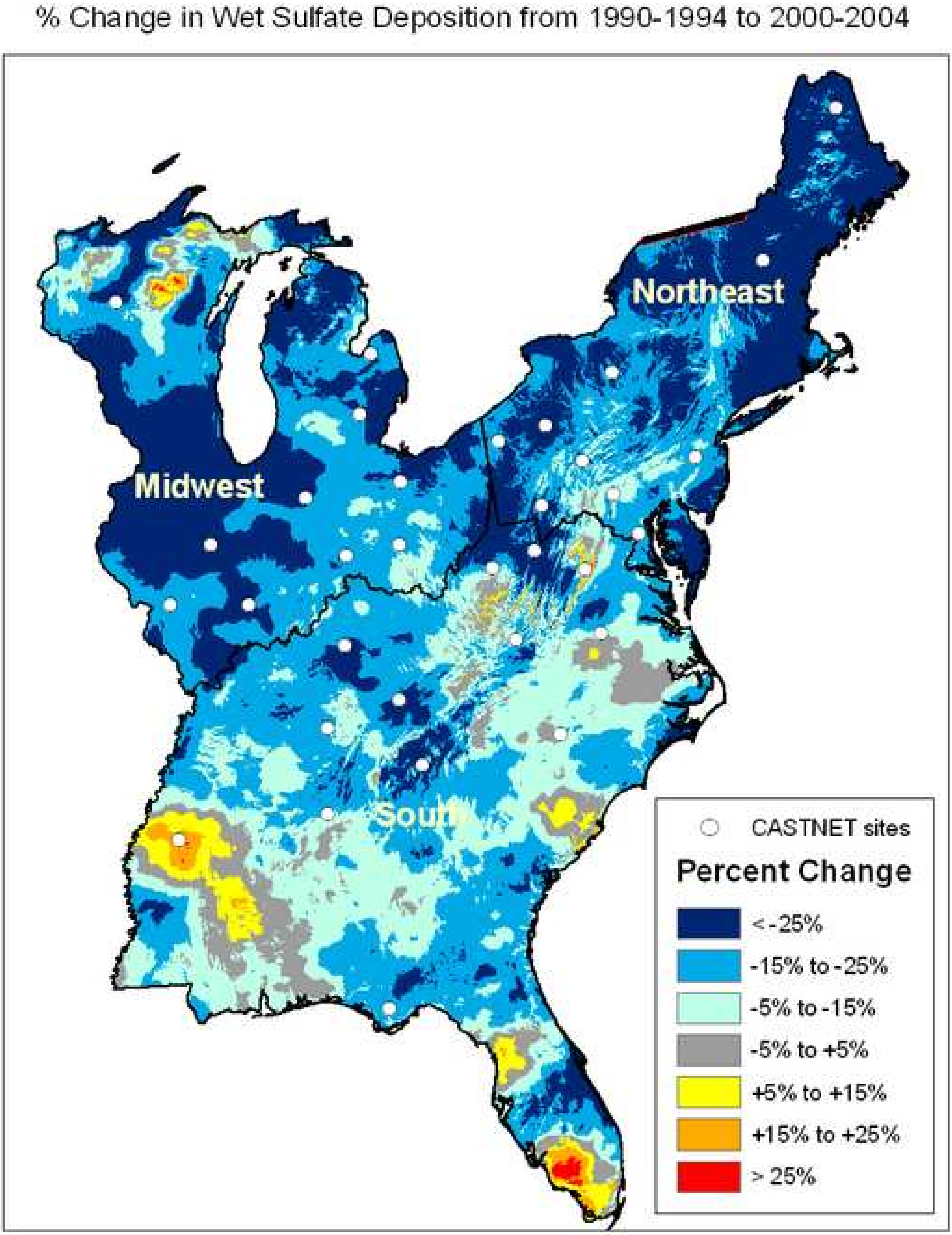

Figure 2b

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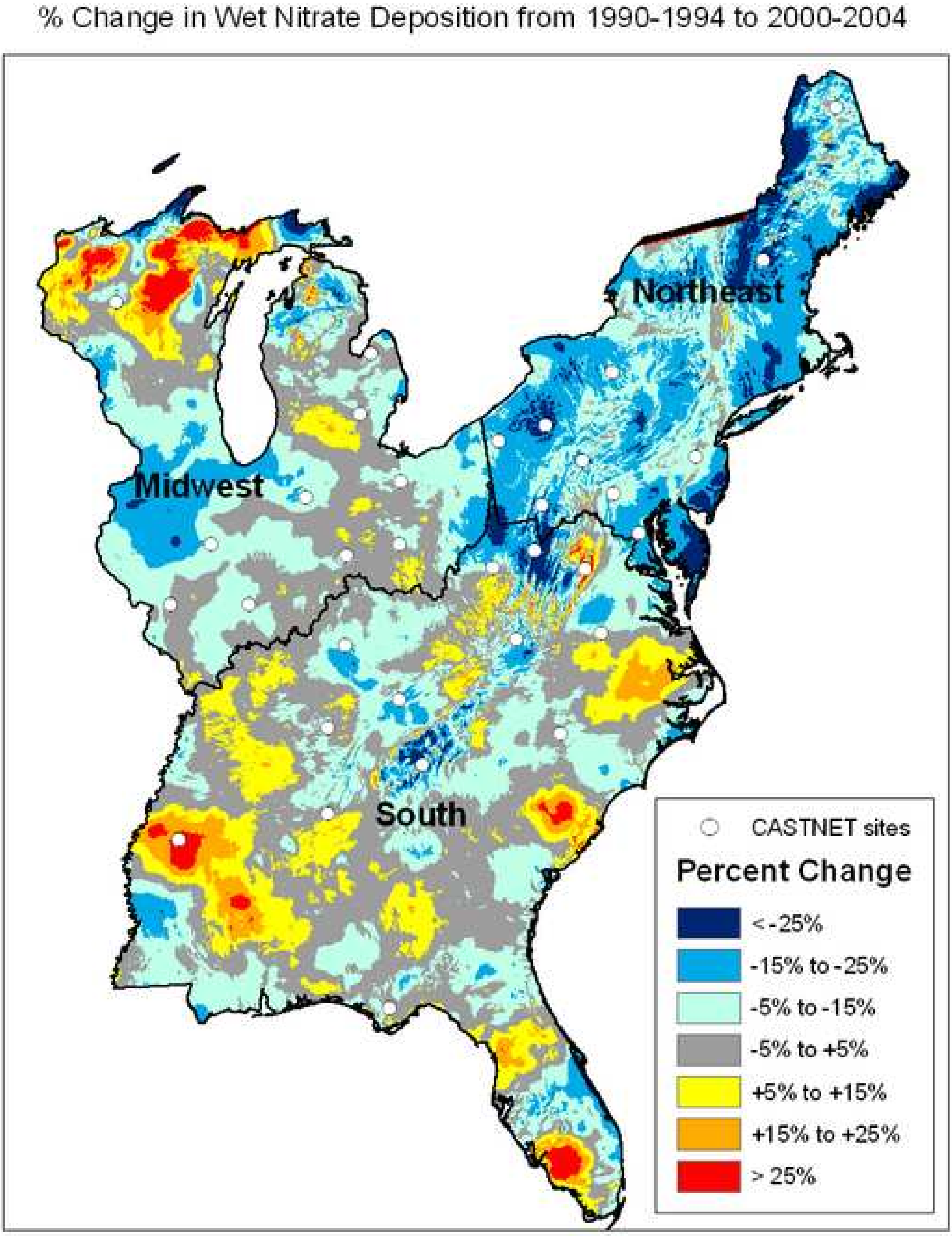
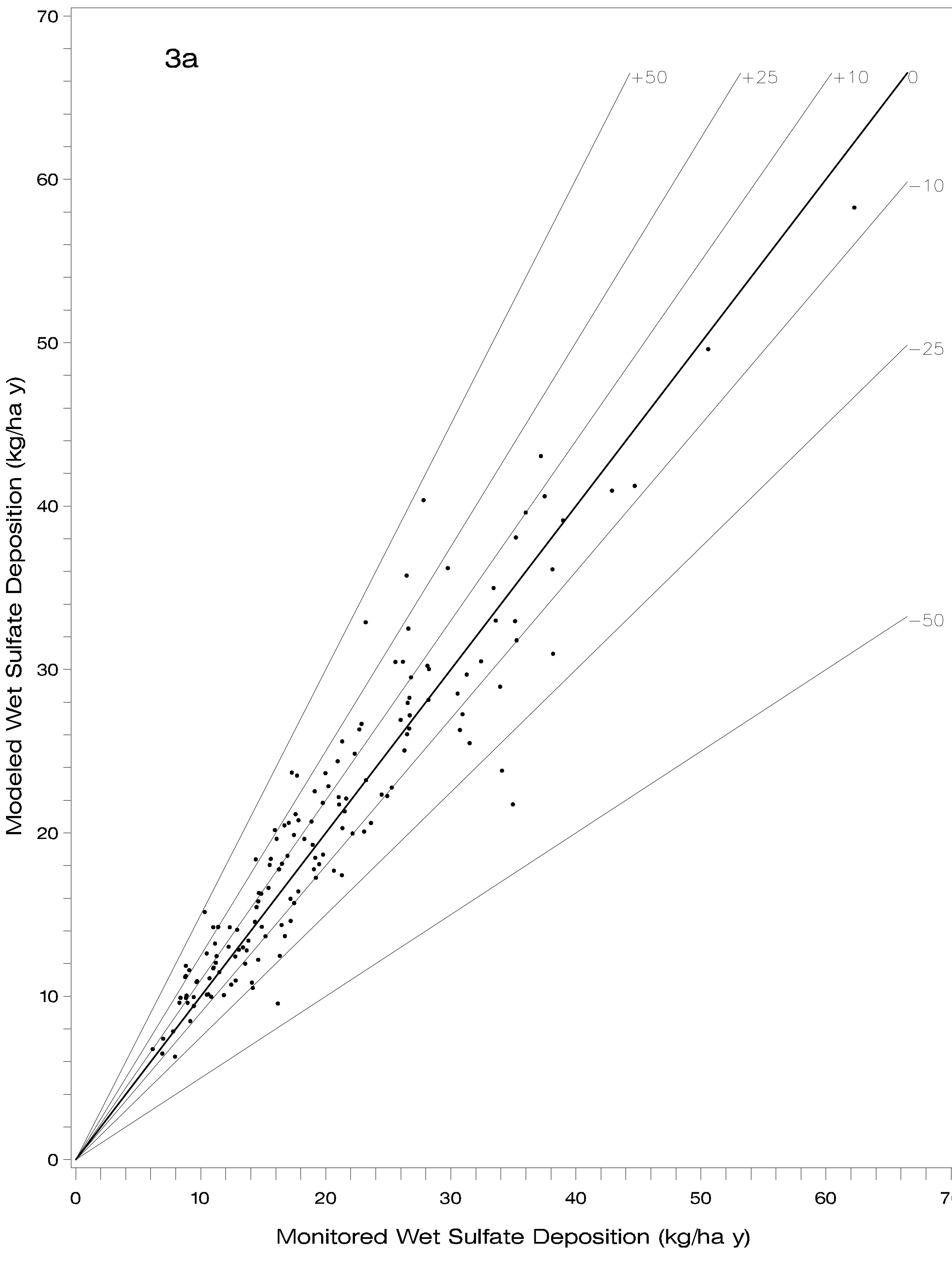


Figure 3a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−10
−25
−50
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50
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30
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10
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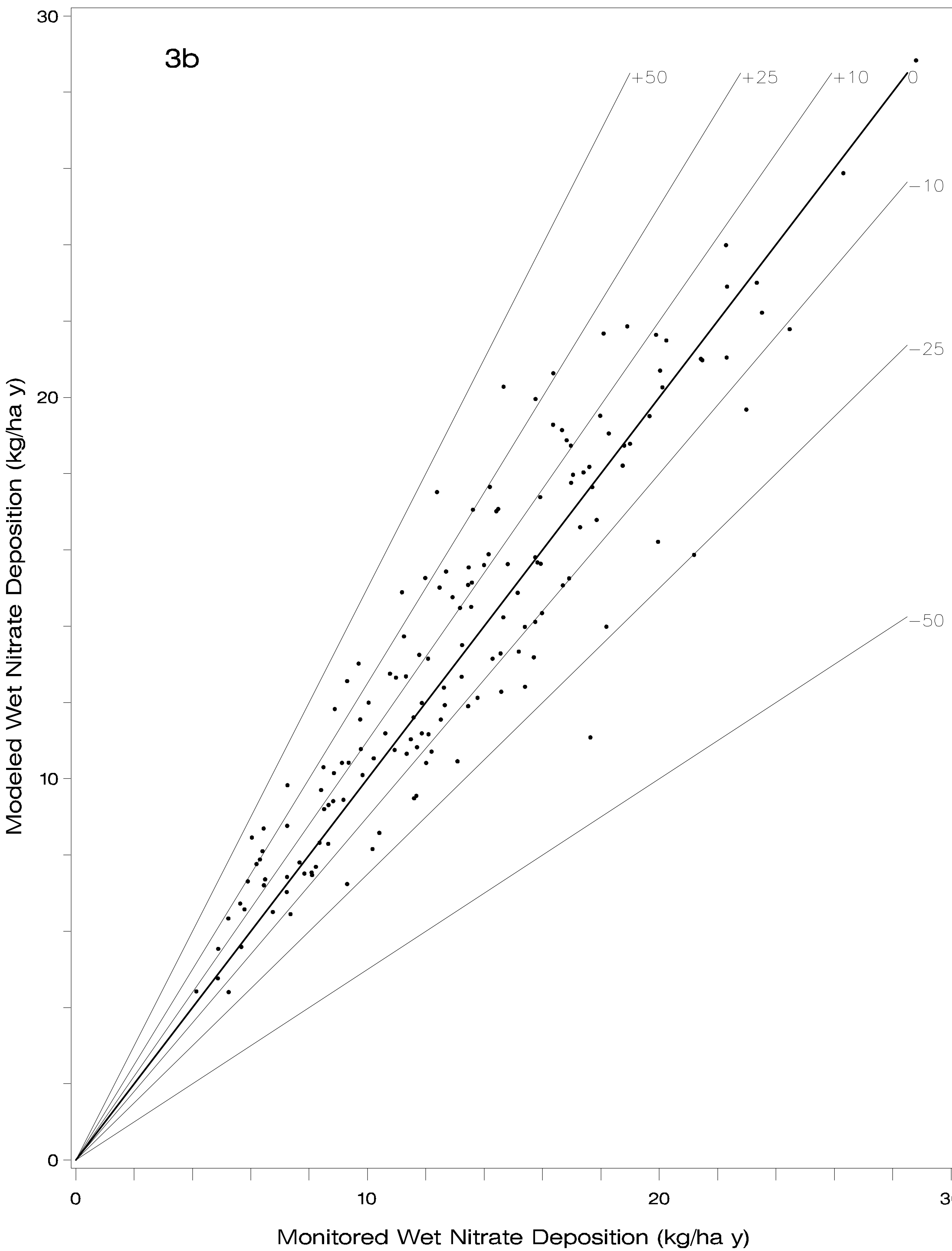
40
50
60
70
Modeled Wet Sulfate Deposition (kg/ha y)
Monitored Wet Sulfate Deposition (kg/ha y)

Figure 3b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Modeled Wet Nitrate Deposition (kg/ha y)
Monitored Wet Nitrate Deposition (kg/ha y)

Figure 4a

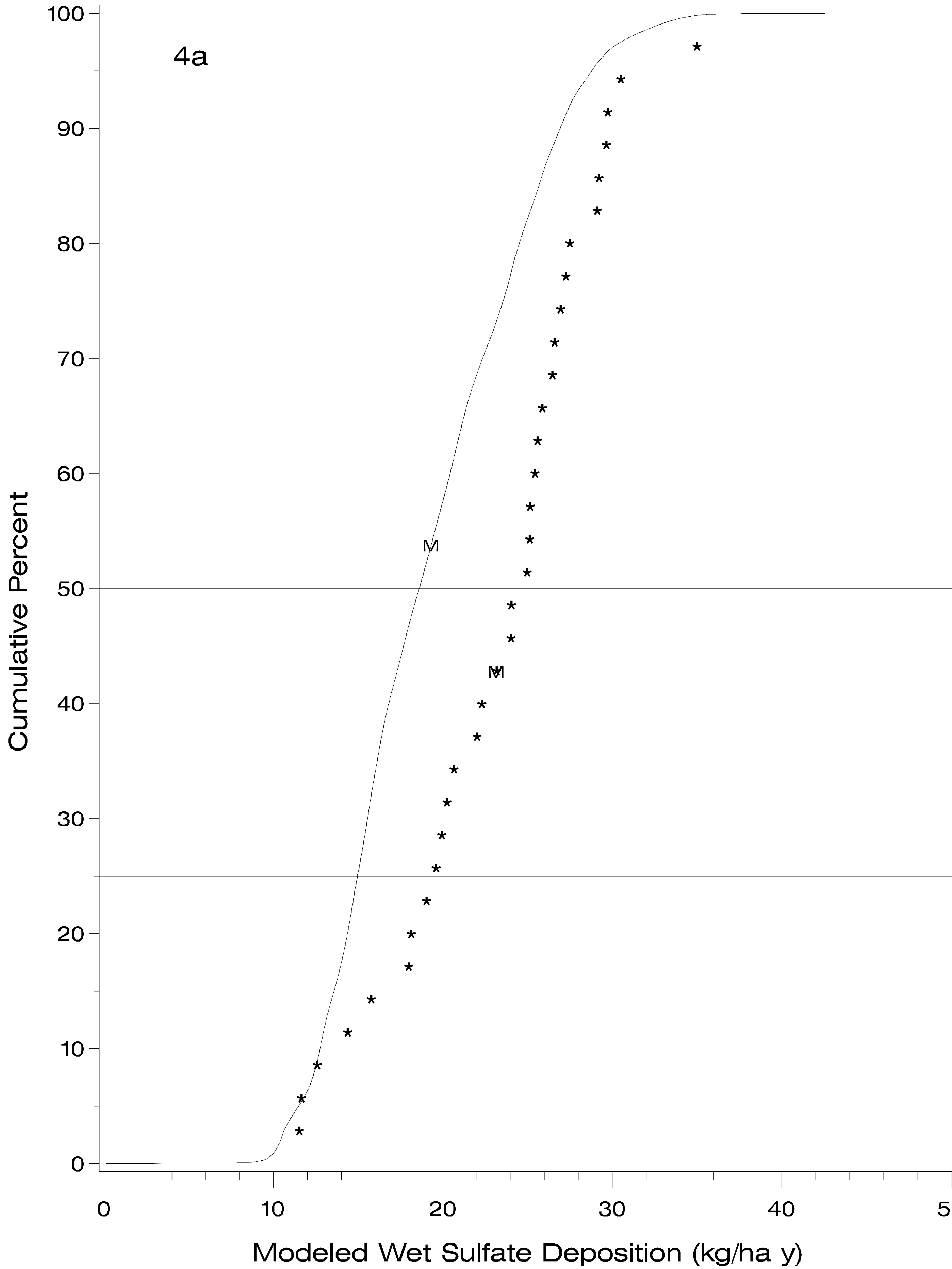
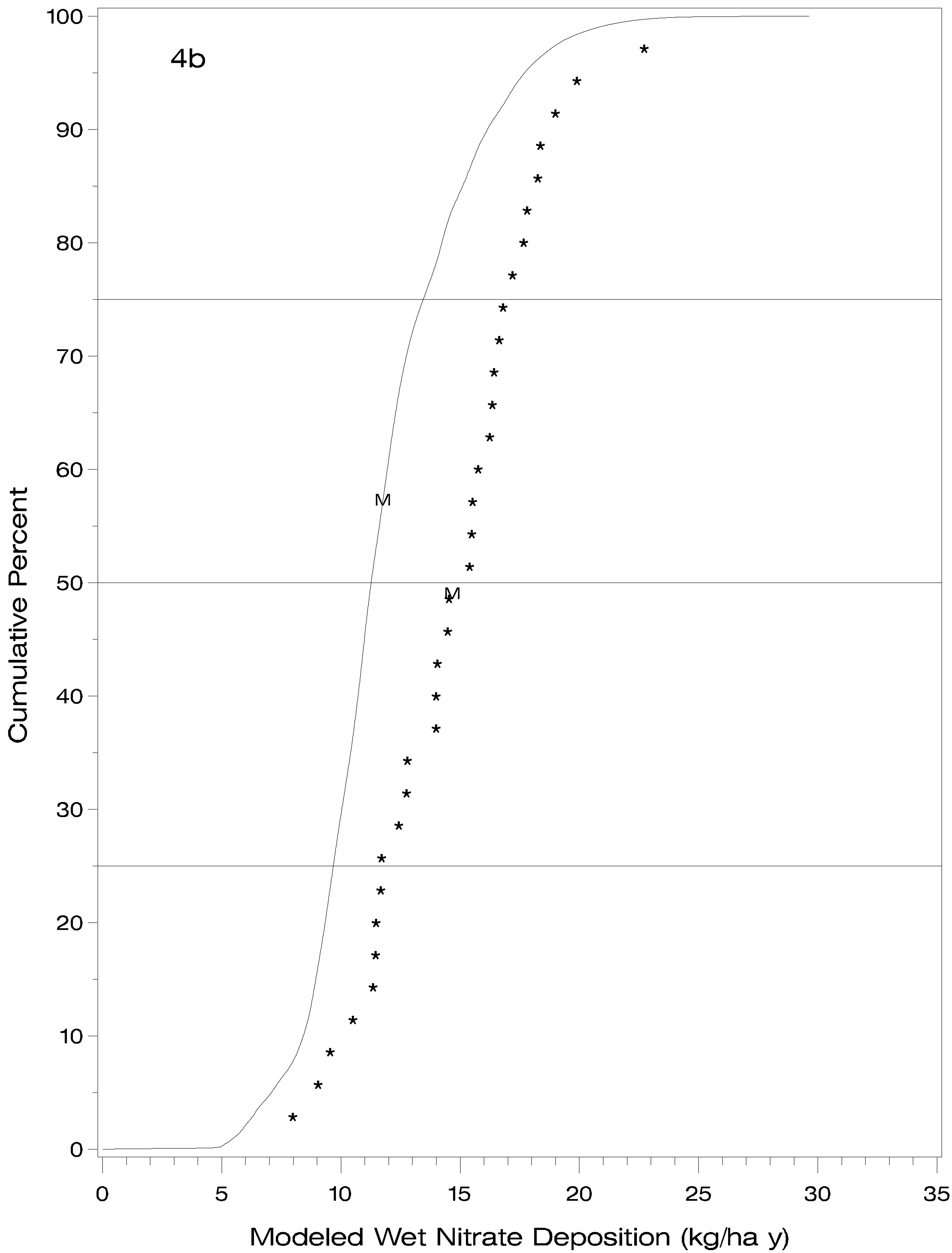

4a
Cumulative Percent
0
10
20
30
40
50
60
70
80
90
100
M
M
0
10
20
30
40
50
Modeled Wet Sulfate Deposition (kg/ha y)

Figure 4b



4b
Cumulative Percent
Modeled Wet Nitrate Deposition (kg/ha y)
M
M
0
10
20
30
40
50
60
70
80
90
100
0
5
10
15
20
25
30
35