

Appendix D: Compilation of Available Information on Atmospheric Deposition Rates Compiled by NC Collaboratory Third Party Reviewers

Review of Atmospheric Nitrogen Deposition Data and Trends for the Falls Lake and Jordan Lake basins

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Introduction

Total nitrogen deposition includes wet and dry nitrogen (N), both made up of oxidized and reduced portions. Wet deposition is the result of precipitation events (rain and snow) that removes particles and gases from the atmosphere. Dry deposition is the transfer of gases and particles to the landscape when there is no precipitation (Baumgardner et al., 2002). Oxidized N is primarily produced from the burning of fossil fuels, whereas reduced N is primarily emitted from agricultural and livestock systems. Reduced N includes both Ammonia (NH_3) and particulate Ammonium (NH_4^+). Oxidized N mainly comprises nitrogen oxide (NO), nitrogen dioxide (NO_2), and nitric acid (HNO_3) (Paerl et al., 2002). However, more comprehensive definitions of oxidized N include HNO_3 , NO_x , Dinitrogen pentoxide (N_2O_5), Nitrous acid (HONO), organic nitrates, and Peroxyacyl nitrates (PAN) (Schwede and Lear, 2014).

N deposition Databases

Total Deposition Science Committee (TDEP)

EPA has developed a hybrid approach to mapping total deposition that combines measured and modeled values (Schwede and Lear, 2014). This Total Deposition Science Committee (TDEP) was formed within the National Atmospheric Deposition Program (NADP) in 2011. Wet deposition values are obtained from combining NADP/National Trends Network (NADP/NTN) measured values of precipitation chemistry with precipitation estimates from the Parameter-elevation Regression on Independent Slopes Model (PRISM). Dry deposition values are obtained by combining air concentration data with modeled deposition velocities. Air concentration data are from the Clean Air Status and Trends Network (CASTNET), the NADP/Ammonia Monitoring Network (NADP/AMoN), and the Southeastern Aerosol Research and Characterization (SEARCH) network, while deposition velocities are estimated from the Community Multiscale Air Quality (CMAQ) model. These point values for deposition are merged spatially with modeled dry deposition values from the CMAQ model. CMAQ predicts hourly concentration and deposition values using a numerical air quality model (Byun and Schere, 2006).

TDEP maps are available for dry, wet, and total deposition. Dry N deposition is available for total N, reduced N, oxidized N, total Nitrate, and Ammonium, while wet N deposition is available for inorganic N, Nitrate (NO_3^-), and Ammonium. Finally, total deposition is available for total N, reduced N, and oxidized N. The total Nitrate (in the dry deposition) refers to HNO_3 and particulate NO_3 (TDEP). Based on an [NADP brochure](#), inorganic N deposition contains Ammonium and Nitrate depositions. However, comparing maps of inorganic N, Ammonium, and Nitrate wet depositions shows that wet Nitrate depositions are higher than the other two (perhaps due to a nitrate mapping error). TDEP maps can be found in <https://www3.epa.gov/castnet/mapcharts.html>, but the underlying data are not available for online download. A semi-quantitative exploration of these maps shows that in our study area, total N deposition is likely dominated by oxidized N, especially in urban areas (Figure 1). Total deposition of oxidized N in urbanized areas is approximately 2 times higher than rural areas, whereas reduced N is higher in the livestock regions east of our study area (Figure 1). The dry N deposition is also dominated by the oxidized portion, particularly in urban areas (Figure 2). Dry N deposition has decreased from 2000

to 2019 (Figure 3) mainly due to the decrease in the oxidized portion. The dry N deposition does not appear to be correlated with precipitation. The wet inorganic N deposition has decreased gradually from 2000 (dry year) to 2019 (wet year) (Figure 4). This is true even though (comparing more maps from 2000-2019) wet inorganic N deposition is generally higher in wet years.

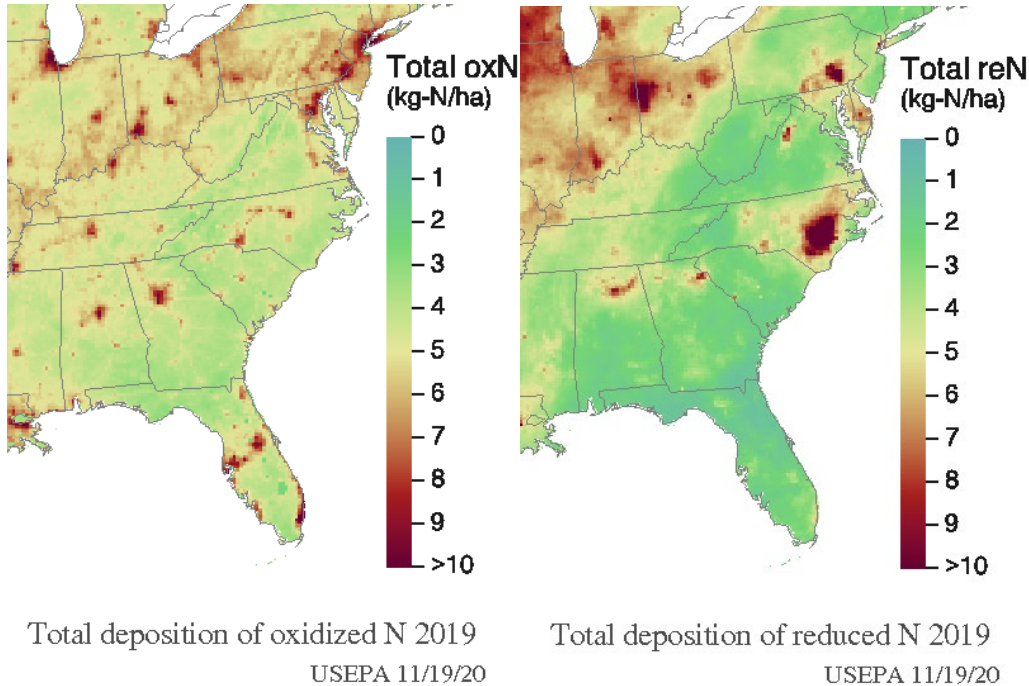


Figure 1. Total deposition of oxidized (left) and reduced (right) N in 2019 (<https://www3.epa.gov/castnet/mapcharts.html>).

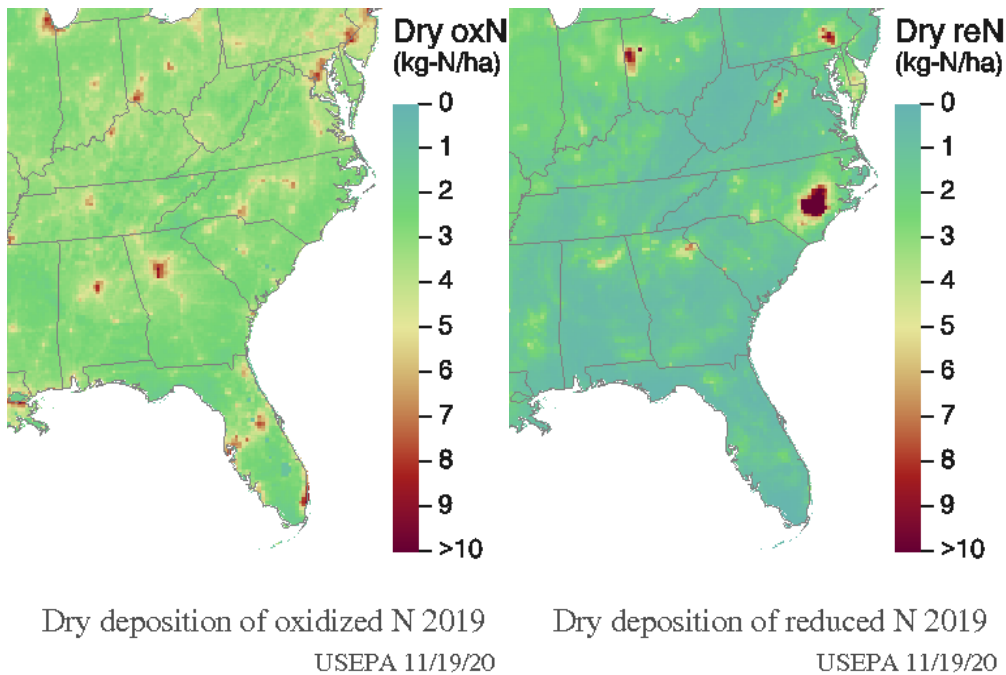


Figure 2. Dry deposition of oxidized (left) and reduced (right) N in 2019 (<https://www3.epa.gov/castnet/mapcharts.html>).

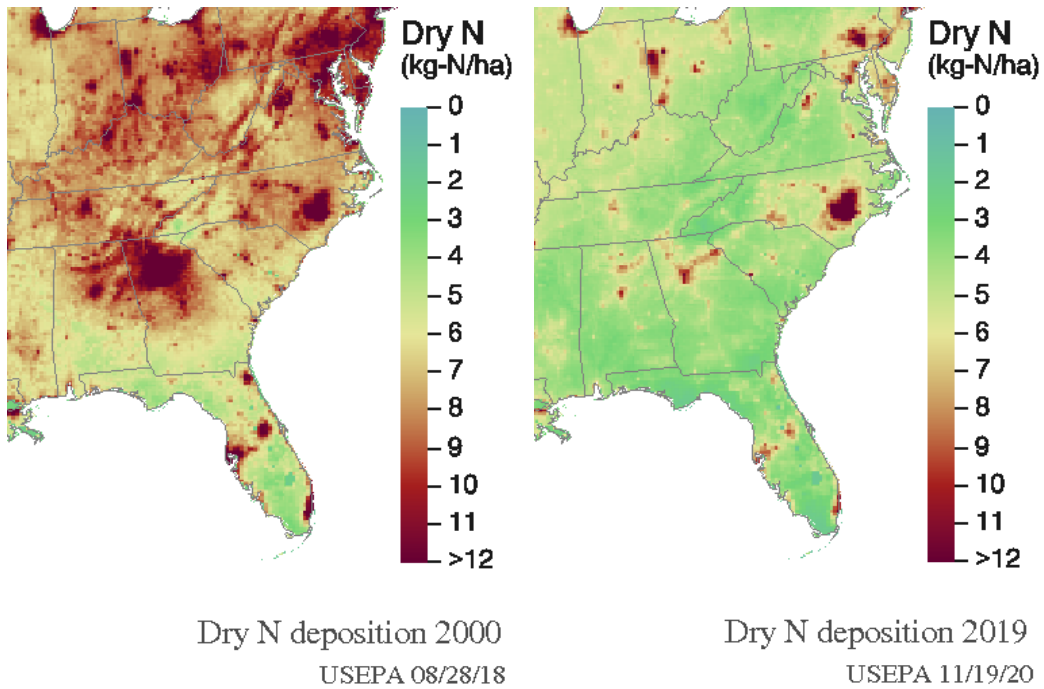


Figure 3. Dry N deposition in 2000 (left) and 2019 (right) (<https://www3.epa.gov/castnet/mapcharts.html>).

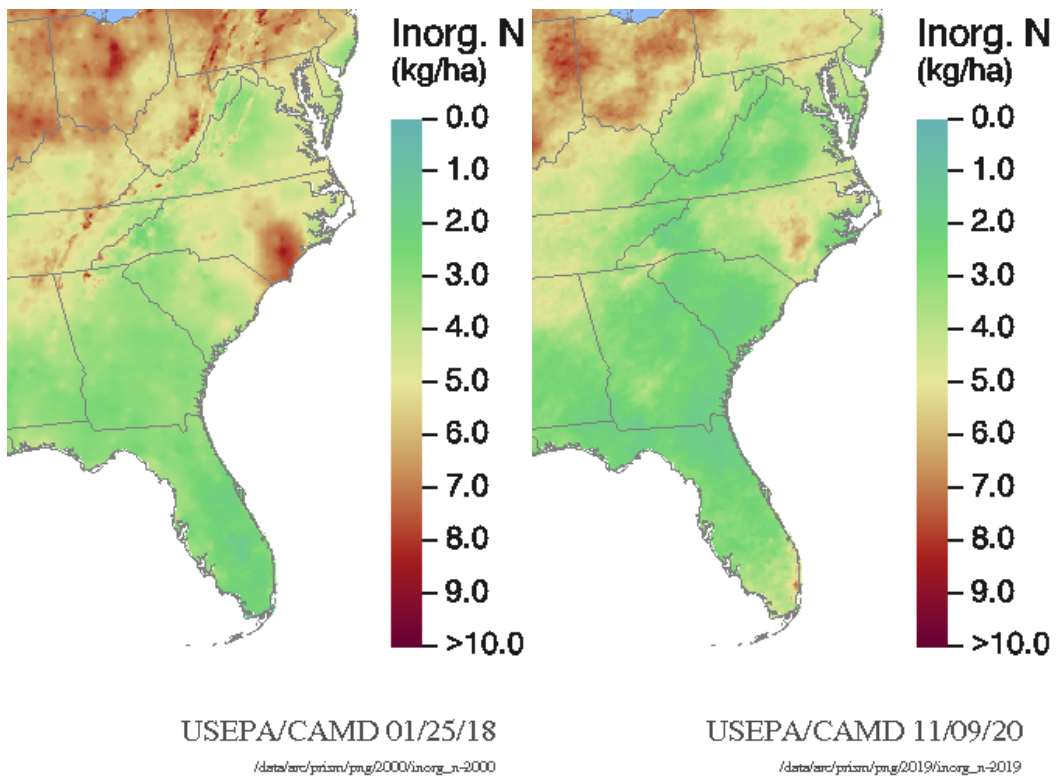


Figure 4. Wet deposition of inorganic N in 2000 (left) and 2019 (right) (<https://www3.epa.gov/castnet/mapcharts.html>).

The NADP National Trends Network (NTN)

The NADP NTN (<http://nadp.slh.wisc.edu/ntn/>) has a long-term record of precipitation chemistry and wet deposition at over 250 stations in the U.S. It collects weekly precipitation samples and measures a number of analytes including SO_4 , NO_3 , and NH_4 (Schwede and Lear, 2014). The NTN sites located in the Piedmont region are (Figure 5, Table 1). NTN provides weekly concentrations, seasonal and annual wet depositions (Figure 6).

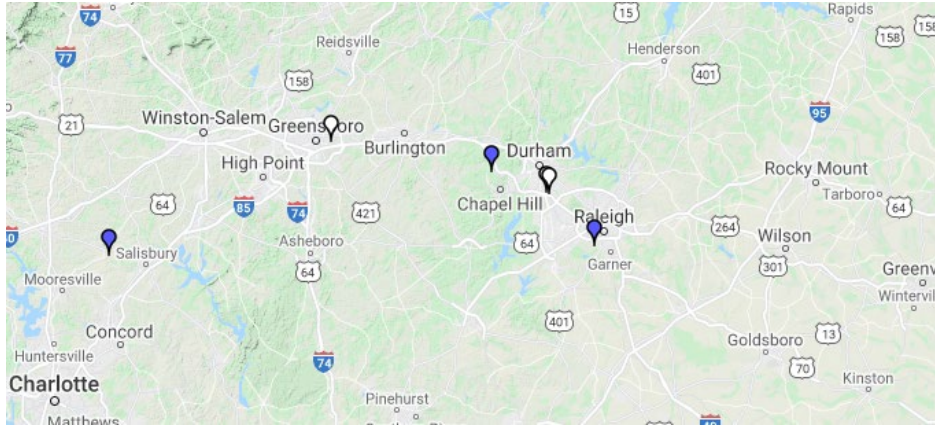


Figure 5. Location of the NTN/NADP stations.

Table 1. NTN station description.

Site ID	Location	Nearest city	Data availability
NC 30	Duke Forest	Durham	2020
NC 17	Greensboro	Greensboro	2015-2019
NC 11	Research Triangle Institute	Durham	1980-1982
NC 33	Research Triangle Park	Durham	1980-1983
NC 34	Piedmont Research station	Salisbury	1978-2019
NC 41	Finley Farm	Raleigh	1978-2019



Figure 6. NTN annual wet N deposition (upper) and precipitation (lower).

The median NTN (wet) deposition for NH_4 , NO_3 , and total N for these stations are 2.4, 2.2, and 4.6 kg/ha/y, respectively. The higher deposition values mostly correspond with higher precipitation years. However, the correlation between annual deposition and precipitation varies in each form of N, with the lowest correlation with NO_3 (Table 2). On average, the total N wet deposition in wet years is about 5.5 kg/ha/y, while in dry years it is about 3.8 kg/ha/y. The total N and NH_4 wet deposition values are generally consistent with the TDEP inorganic N and NH_4 wet deposition; however, NO_3 wet deposition values from NTN measurements are lower than the TDEP values.

Table 2. Correlation of precipitation with each N form of annual deposition from NTN measurements. The values are shown for the two stations that had the longest record.

NTN site\variable	NH_4	NO_3	Total
NC34	0.59	0.35	0.61
NC41	0.73	0.30	0.77

The monitored values can also be summarized by season to explore the seasonal variability of N deposition (Figure 7). The boxplot of each N form deposition shows higher deposition in spring and summer, which is likely related to higher precipitation in these seasons, especially in late summer. These results also indicate substantial year-to-year variability in seasonal wet N deposition.

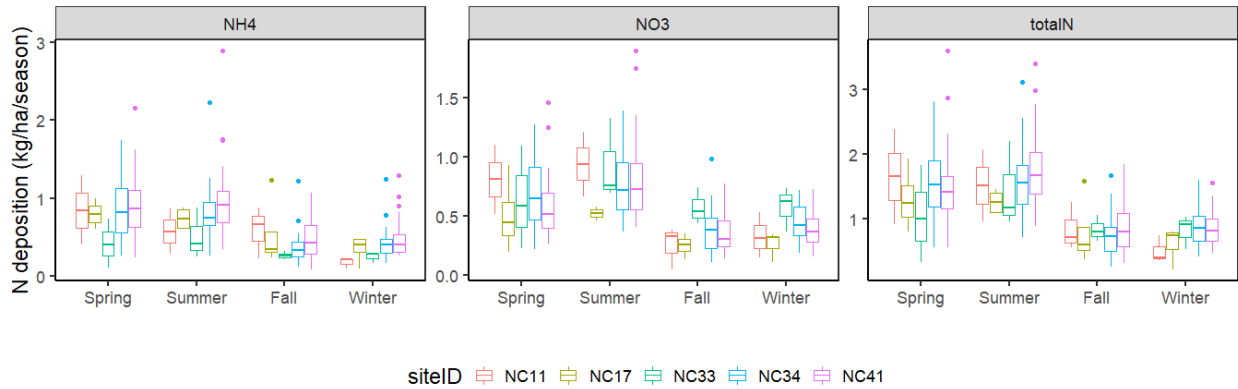


Figure 7. Seasonal wet N deposition from NTN measurements.

Clean AirStatus and Trends Network (CASTNET)

CASTNET (www.epa.gov/castnet) measures ambient concentrations of sulfur and nitrogen species as well as rural O₃ concentrations. The concentrations are used to calculate dry deposition fluxes. It complements NTN, with nearly all sites co-located with or near an NTN site. The CASTNET monitoring network has 2 stations in the Piedmont region located in Duke Forest and Research Triangle Park. These two sites are close to NC30 and NC33, however, they provide a longer and more recent record than NTN measurements (from 2000-2017). The CASTNET website provides dry, wet, and total N annual deposition (Figure 8). Similar to TDEP maps (Figures 3), Figure 8 shows that dry N deposition has decreased over time. Wet N deposition has more variability due to its positive correlation with precipitation (Table 3). The median total, wet, and dry deposition values are 12.2, 4.9, and 7.3 kg/ha, respectively. The median wet deposition from CASTNET measurements (4.9 kg/ha) is close to the NTN measurement (4.6 kg/ha). On average, dry deposition makes up 60% of total N deposition in these two stations. This is generally consistent with the information from the TDEP maps.

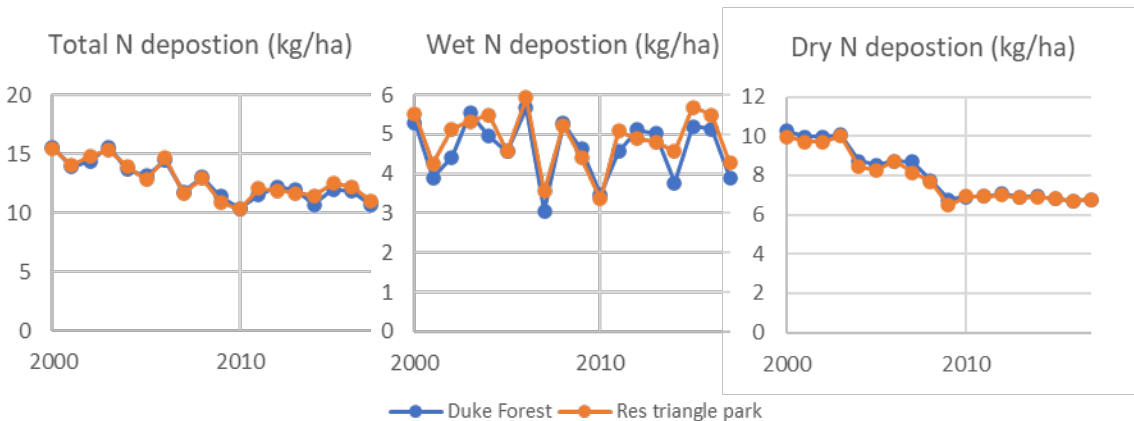


Figure 8. Total, wet, and dry N deposition from CASTNET.

Table 3. Correlation of precipitation with annual wet and dry N deposition from CASTNET measurements.

CASTNET site\variable	Wet N deposition	Dry N deposition
Duke Forest	0.71	-0.21
Research Triangle Park	0.62	-0.04

Baumgardner et al. (2002) collected data as part of the CASTNET deposition monitoring conducted during the 1990s. Dry deposition contributed approximately 35% of total nitrogen deposition in North Carolina, rather than the 60% observed in the more recent CASTNET data for our study area (above). This discrepancy may be due to using older measurements (1990s) as well as using different monitoring sites.

Community Multiscale Air Quality (CMAQ) model

CMAQ is an advanced regional air quality model developed by EPA to simulate the fate, transport, and deposition of air pollutants under varying atmospheric conditions (Byun and Schere, 2006). The data provides hourly estimates and aggregated daily datasets from 2002-2014 (<https://www.epa.gov/cmaq/cmaq-output>). The output shapefile is also available through their [FTP server](#). There are some databases that computed aggregated annual CMAQ estimates.

EnviroAtlas national map is an interactive map that portrays annual nitrogen deposition (kg/ha/y) within each 12-digit hydrological unit code (HUC) watershed for 2011 (which is a fairly normal precipitation year for the study area). This map provides information from the CMAQ model (<https://enviroatlas.epa.gov/enviroatlas/interactivemap/>). The map provides annual wet, dry, and total deposition for reduced and oxidized N deposition (Figures 9-12). Comparing the CASTNET values in 2011 with this interactive map shows that the values are similar (less than 10% difference).

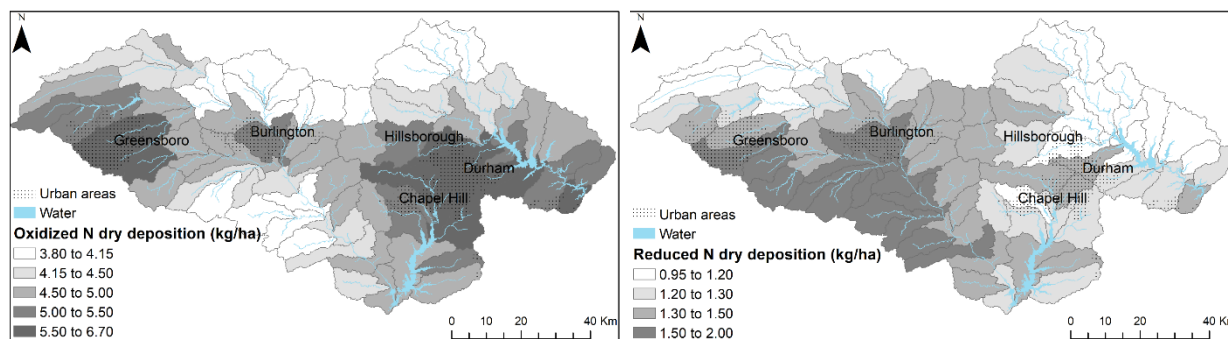


Figure 9. Dry annual oxidized (left) and reduced (right) N deposition (from EnviroAtlas national map).

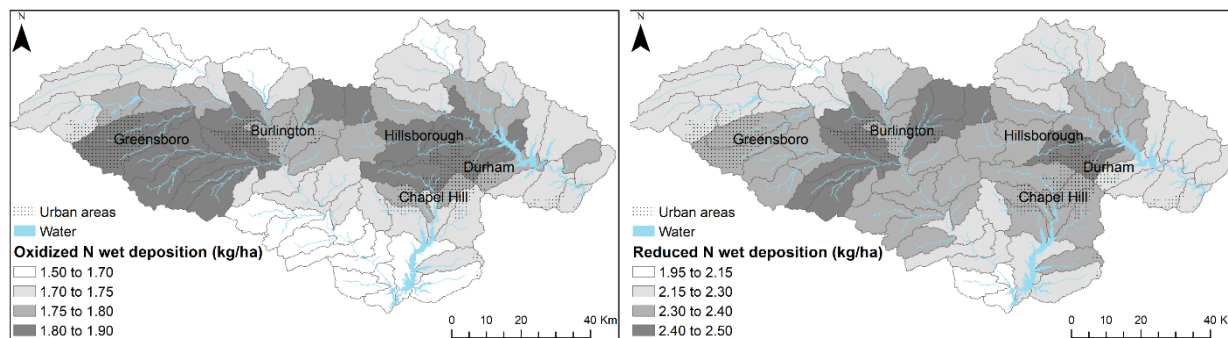


Figure 10. Wet annual oxidized (left) and reduced (right) N deposition (from EnviroAtlas national map).

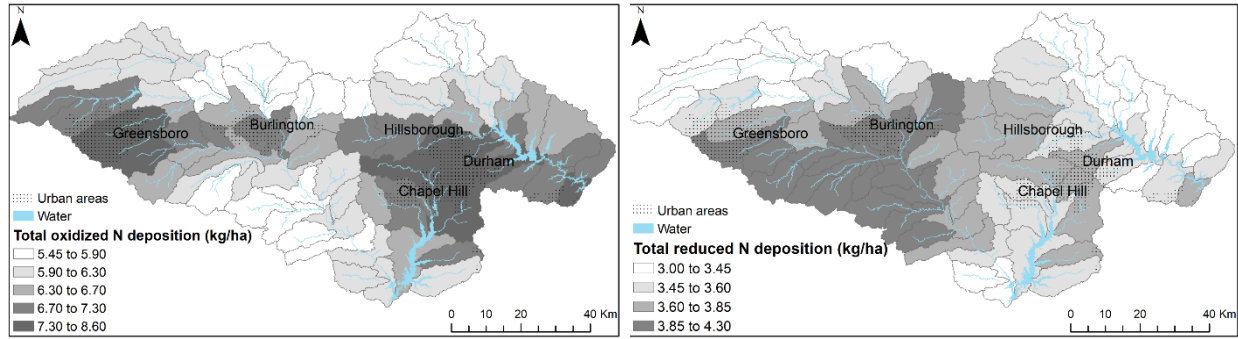


Figure 11. Total annual oxidized (left) and reduced (right) N deposition (from EnviroAtlas national map).

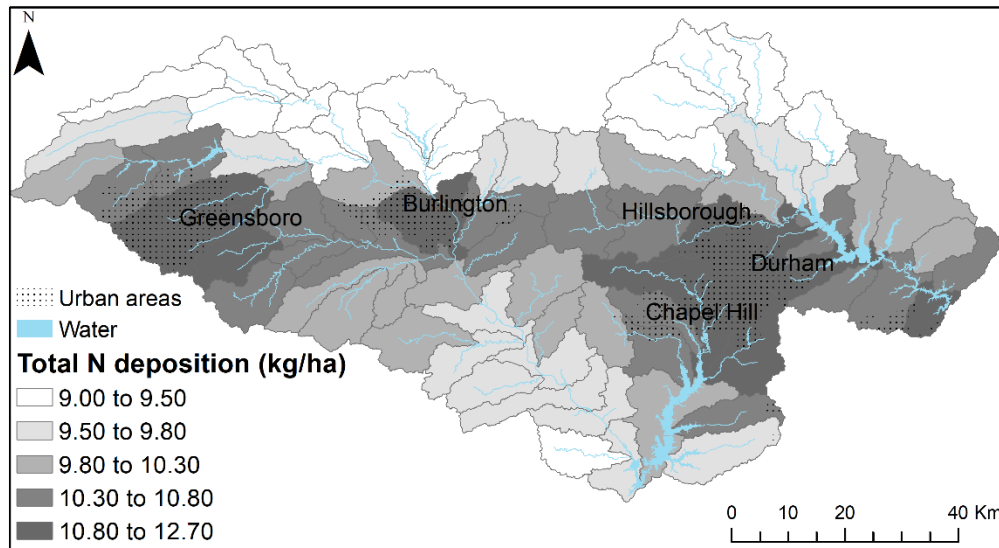


Figure 12. Total N deposition (from EnviroAtlas national map).

The oxidized N deposition values are higher in urban areas, especially in Greensboro and Durham (Figures 9-11, left). However, the reduced wet and dry N deposition does not show a strong spatial pattern related to urbanization (Figures 9-11, right). The median N deposition for each form is provided in Table 4. N dry deposition and total N deposition are both dominated by the oxidized form. This complies with the findings of the TDEP maps (Figures 1-2). Unlike dry and total N deposition, wet deposition is dominated by the reduced form. The wet Ammonium deposition maps from TDEP also shows its dominance in the inorganic N deposition. However, since TDEP does not provide wet Ammonia deposition maps, comparing wet deposition from the EnviroAtlas map and TDEP maps is not straightforward. Finally, dry deposition (median= 5.89 kg/ha) is the dominant form of total deposition. The dry deposition makes up about 60% of total deposition, which complies with the CASTNET measurements.

Table 4. Median deposition values (kg/ha/y) for each N form in Figures 9-12.

Deposition type	Oxidized	Reduced	Total
Wet	1.74	2.32	4.06
Dry	4.61	1.29	5.89
Total	6.41	3.59	10.05

The USGS SPARROW model for North Carolina includes N deposition as an input variable (Gurley et al., 2019). The model input is a 3-year average (2010, 2011, 2012) of total deposition (Figure 13) derived from a national database (Wieczorek et al., 2018). Wieczorek et al. (2018) created national databases of information (including annual N deposition from the CMAQ model) linked to the National Hydrography Dataset-Plus (NHD+) catchments (Moore and Dewald, 2016). These estimates are aggregated based on NHD+ catchments (not shown in Figure 13). Instead, Figure 13 shows the HUC12 boundaries for visualization purposes. The deposition maps show higher N deposition in urbanized areas. The median total N depositions in 2010, 2011, and 2012 are 9.65, 10.22, and 10.52 kg/ha, respectively. Comparing Figure 13 with the EnviroAtlas map (Figure 12) shows that the total deposition values are close.

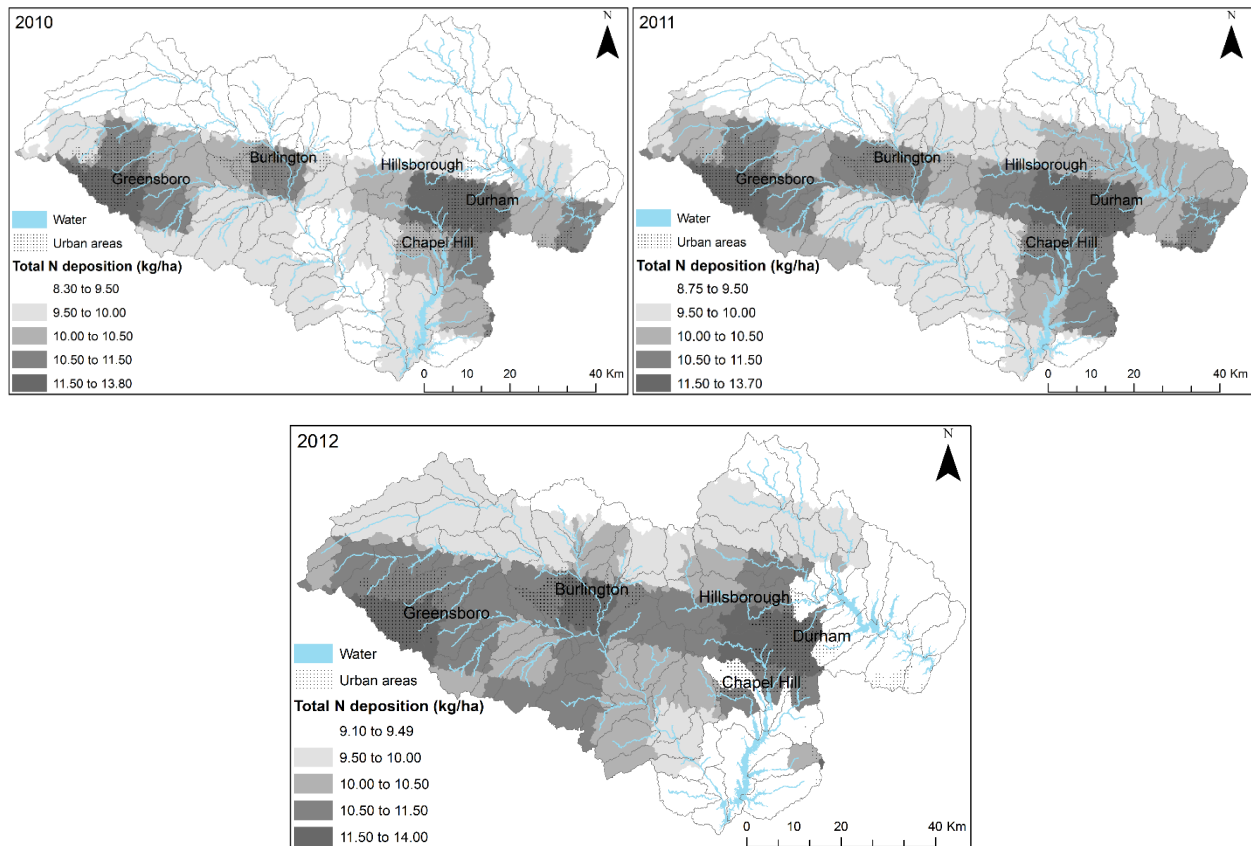


Figure 13. Total (wet + dry) N deposition in 2010 (left), 2011 (right), and 2012 (lower) from (Wieczorek et al., 2018).

Summary

- The median total annual N deposition for the study area is 12 kg/ha/y based on CASTNET data (2000-2017).
- Spatially distributed estimates of total N deposition for 2010-2012 range from 8-14 kg/ha/y (Wieczorek et al., 2018).
- Total N deposition is highest in summer, followed by spring, mainly due to higher precipitation in our study area during these seasons (NTN).

- Total annual N deposition is positively correlated with annual precipitation. This is primarily due to precipitation increasing wet deposition (NTN and CASTNET).
- Dry N deposition makes up about 60% of total deposition (EnviroAtlas map and CASTNET).
- Total N deposition is higher in urban areas, primarily due to higher dry oxidized deposition (TDEP and EnviroAtlas map).
- Oxidized N accounts for 40% of wet deposition, 80% of dry deposition, and 65% of total deposition (EnviroAtlas map).

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Review of Atmospheric Phosphorus Deposition Data for the Falls Lake and Jordan Lake basins

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Introduction

Historically, atmospheric phosphorus (P) deposition has been considered a minor contributor to overall P loading, relative to other sources like fertilizers. However, some studies suggest that it can be important for particular lakes and urban areas (Jassby et al., 1994; Newman, 1995; Anderson and Downing, 2006; Tipping et al., 2014). Primary sources of atmospheric P deposition are marine aerosols, dust from agricultural activities and deserts, biomass burning, soil erosion, and coal and oil combustion (Ahn and James, 2001; Tipping et al., 2014). Atmospheric P transport is different from nitrogen (N) transport in that it does not have a stable gaseous phase in the Earth's atmosphere and is mainly restricted to aerosols (Mahowald et al., 2008). Also, unlike N, P deposition data is not generally available through the National Atmospheric Deposition Program (NADP). Thus, atmospheric P deposition data is relatively scarce, and it must often be estimated from small-scale measurement studies or models (Ahn and James, 2001; Sabo et al., 2021)

Atmospheric P deposition on a global and regional scale

Several studies have explored P deposition on a global scale. Graham and Duce (1979) compiled atmospheric total P (TP) samples from 1973-1975, globally. Based on their study, TP deposition in North America ranged from 0.07-1.5 kg/ha/yr, with an average of 0.3 kg/h/yr. The higher values were often (but not always) associated with urban areas. More recent large-scale studies indicate similar levels of TP deposition (Gibson et al., 1995; Tsukuda et al., 2006; Mahowald et al., 2008). For example, Tsukuda et al. (2006) reported a range and average P deposition of 0.07-1.07 kg/ha/yr and 0.4 kg/ha/yr, respectively, in North America. These data-synthesis studies provide a foundation for our understanding of P atmospheric deposition. However, most sampling occurred over limited time intervals and may underestimate P deposition in some cases (Benitez-Nelson, 2000). Mahowald et al. (2008) estimated TP deposition on a global scale using the Model of Atmospheric Transport and Chemistry (MATCH; Rasch et al., 1997). The model estimated deposition for central NC to be 0.01-0.05 kg/ha/yr in 2000, which is relatively low. The other global studies did not report deposition values for central North Carolina.

Sabo et al. (2021) developed estimates of TP deposition for USGS HUC-8 watersheds across the conterminous United States (CONUS) from 2002-2012 using atmospheric models (Wang et al., 2015). The average P deposition in 2012 was 0.08 kg/ha/yr for the CONUS and ranged from 0-0.19 kg/h/yr. No clear pattern was found between the P deposition values and the urbanized areas. In central NC, estimated P deposition was between 0.06-0.1 kg/ha/yr in 2012 (Figure 1), somewhat higher than in 2002. More precisely, the database associated with this study indicates that P deposition was 0.09 and 0.08 kg/ha/yr in the Jordan Lake and Falls Lake basins in 2012, respectively.

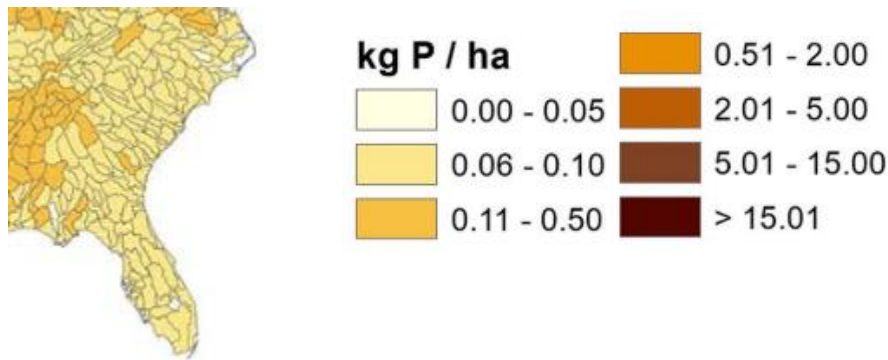


Figure 1. Atmospheric TP deposition rates for HUC-8 subbasins in the southeast US in 2012 (Sabo et al., 2021).

In addition to global and national studies on P deposition, regional studies may provide more insights on spatial and seasonal patterns. In this regard, Delumyea and Petel (1978) determined wet and dry P deposition in Lake Huron from April-October 1975 by collecting samples from 11 stations. They measured available P, indicating the sum of the water- and acid-soluble fractions of P. The dry and wet available P deposition values were 0.06 and 0.08 kg/ha/yr. The total available P deposition was determined 0.24 kg/ha/yr (notably different from wet+dry deposition, which was attributed to sample contamination). The total available P deposition was highest in June and September, and this was attributed to agricultural activity near Lake Huron.

Willey and Kiefer (1993) collected P deposition samples from 1988-1991 on the campus of UNC Wilmington. Wet and dry P deposition was small compared to N. The N:P molar ratio of atmospheric deposition was 51 (mass ratio of 23), which is notably higher than the Redfield molar ratio of 16 (Chapra, 2008). The wet and dry P deposition values were reported as <0.19 and <0.01 kg/ha/yr, respectively, indicating higher contributions of wet deposition. Total P deposition (<0.2 kg/ha/yr) is generally consistent with the deposition values derived from the CONUS model for central NC (Sabo et al., 2021).

Ahn and James (2001) estimated the total weekly P deposition in South Florida using wet and dry samples collected from April 1992 to October 1996 at 13 sites. The average P deposition was 0.41 kg/ha/yr, and the standard deviation was 0.14 kg/ha/yr across sites. The average ratio of total dry to wet deposition was 2.8, which unlike Willey and Kiefer (1993) indicates that the dry portion was dominant. Individual site means ranged from 0.11 kg/ha/yr at a remote station in a marsh area to 0.77 kg/ha/yr in pastureland. Comparing monthly values indicated that the mean P values were lowest in January (0.31 kg/ha/yr) and highest in October (0.54 kg/ha/yr). The average P deposition during the wet seasons (June–October) was about 26% larger than that of the dry season, mainly due to higher precipitation. No long-term trend was found in the data.

Anderson and Downing (2006) characterized wet and dry deposition to wet and dry surfaces in Iowa, using samples collected by 12 automated samplers at six sites across the region in 2003. The annual total deposition was 0.3 kg/ha/yr of P, 7.7 kg/ha/yr of N, and 6.1 kg/ha/yr of Si. The N:P ratio of total atmospheric deposition for this study (as atoms) was 60 (mass ratio of 27). Dry and wet P deposition values were 0.25 and 0.05 kg/ha/yr, respectively, indicating P deposition was dominated by the dry portion, similar to the findings of Ahn and James (2001). The dry deposition was greatest in spring and declined through the summer. Both dry and wet P deposition declined in winter. The seasonal variation

in P deposition may be related to tillage and fertilization schedules in agricultural areas, since they are usually tilled and fertilized prior to planting in spring.

Maccoux et al. (2016) characterized the nutrient sources to Lake Erie from 2003-2013, with measured atmospheric deposition as one of the nutrient sources. The estimated wet+dry P deposition was roughly 0.18 kg/h/yr, which is on the higher end of the range estimated by Sabo et al. (2021). The atmospheric P deposition had the highest standard errors among the source categories, which can be related to the variability in the measurements of TP concentrations and precipitation depth.

Summary

- Estimates of atmospheric TP deposition vary widely across different studies. In the U.S., estimates typically range from 0.05 to 0.5 kg/ha/yr.
- In the most recent study (Sabo et al., 2021), atmospheric TP deposition for the Falls Lake Basin was approximately at 0.08 kg/ha/yr in 2012, but with notable uncertainty.
- The N:P deposition molar ratio was 51 in Wilmington, NC (Willey and Kiefer, 1993) and 60 in Iowa (Anderson and Downing, 2006), indicating an N:P mass ratio of about 25. If we assume 12 kg/ha/yr of TN deposition in our study area (see previous memo on N deposition) and applying the ratio of 25, this yields 0.5 kg/ha/yr of P deposition, which is relatively high compared to other estimates.
- In three studies, the dry TP deposition was dominant (Ahn and James, 2001; Anderson and Downing, 2006), while another study indicated the wet portion was dominant (Willey and Kiefer, 1993).
- Temporal variability in P deposition may be driven by precipitation and agricultural activities (Delumyea and Petel, 1978; Anderson and Downing, 2006).
- Higher P deposition is often found in agricultural areas (Delumyea and Petel, 1978; Anderson and Downing, 2006) but trends with urbanization are less clear across studies (Graham and Duce, 1979; Sabo et al., 2021).

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