



The impact of long-term regional air mass patterns on nutrient precipitation chemistry and nutrient deposition within a United States grassland ecosystem

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Abstract

Changes in the frequency of precipitation as a result of a changing climate, as well as anthropogenic induced deposition of nitrogen (N), both have the potential to alter grassland productivity and diversity. Central U.S. weather patterns are dominated by three major air mass trajectories including regional sources from the Gulf of Mexico (marine tropical, Mt), the Pacific Northwest (mild pacific, mP), and the Desert Southwest (continental tropical, Ct). In this work, the Hybrid Single Particle Lagrangian Integrated Trajectory model was used to determine trends in the proportion of precipitation events from these air mass sources from 1983 to 2006 relative to Konza Prairie Biological Station (KPBS), KS. The annual volume-weighted mean (VWM) concentrations and wet deposition of a variety of precipitation dissolved solutes were linked to source regions north or south of KPBS. The proportion of precipitation events from Mt significantly increased, while the proportion of events from Ct and mP decreased significantly over the study period. The annual VWM concentrations of most solutes were typically higher from precipitation sourced to the north of KPBS. However, wet deposition of four ecologically relevant solutes (NH_4^+ , NO_3^- , H^+ , and SO_4^{2-}) was higher from events from the southern region, likely due to higher precipitation amounts. The proportion of reduced N increased significantly over the study period but was not affected by source region despite the higher use of fertilizers for agriculture in the northern source region. Given the location of this site relative to three dominant air mass paths, future shifts in these patterns will likely impact wet nutrient deposition.

Keywords Precipitation chemistry · Wet deposition · Konza prairie · NADP · HYSPLIT

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1 Introduction

Precipitation is a primary driver of ecological activity within the central United States (Epstein et al. 2002, Fay et al. 2011). The importance of precipitation for grassland ecosystems is exhibited by the change in plant species composition across the east-west gradient from the base of the Rocky Mountain chain to eastern Texas through Illinois (Luauenroth et al. 1999). Annual precipitation in eastern Colorado and western Kansas can be up to 10 times less than their eastern grassland counterparts. This drives the general transition from shortgrass steppe in the west to tallgrass prairie in the east (Luauenroth et al. 1999). Thus, precipitation gradients are highly correlated with plant community structure, function, and productivity (Heisler-White et al. 2009). In central U.S. grasslands and much of the United States, a changing climate will likely result in less frequent, but more intense precipitation events (Kunkel et al. 2008). The effects of changing precipitation regimes in U.S. grasslands could include altered annual productivity of grasslands (Fay et al. 2003) and agricultural fields (Lal et al. 2011), changes in plant community structure (Knapp et al. 2008), and a possible shift in the boundaries of grassland and forest ecosystems (Clark et al. 2001). Human inputs of nutrients, specifically nitrogen (N), by both wet and dry deposition has likewise altered grasslands (Vitousek et al. 1997). Short-term experiments (Zavaleta et al. 2003) and investigations across existing deposition gradients (Stevens et al. 2004) have shown that excess N deposition results in decreased grassland diversity. Anticipated changes in precipitation regimes and nutrient deposition will likely interact to affect grassland ecosystems in the central U.S., but the extent of this alteration is unclear.

While predicting how the interactions of anthropogenic ecosystem alterations will affect North American grasslands in the future is complex, the use of existing long-term data sets can increase our understanding of historical trends in regional weather patterns and nutrient deposition. One tool, the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model allows for the determination of backward trajectories of precipitation causing air masses relative to a specific location from historical data. The HYSPLIT model has been used in previous studies to determine the origin of precipitation causing air masses relative to a point of interest (Cohen et al. 2002, Kelly et al. 2009). For example, Kelly et al. (2009) used this model to determine annual changes in the proportion of marine and continental air masses relative to a research site on the U.S. east coast. When precipitation chemistry data exist, they can be easily correlated with model outputs to determine trends in precipitation chemistry from different source regions.

Historical analyses of weather patterns suggest that the central U.S. is dominated by three major air mass trajectories, each with a different regional source (Bryson 1966). These sources include the Gulf of Mexico (marine tropical, Mt), the Pacific Northwest and Canada (mild pacific, mP), and the Desert Southwest (continental tropical, Ct). Atmospheric conditions often favor these paths as they move through the central U.S. and eastward. These data have been used to delineate zones where a specific weather pattern is more dominant than others (i.e. more than 50% of air masses come from a single source; see Fig. 13 in Bryson 1966 and Fig. 1 in this text). Between each of these zones is a ‘buffer area’ where there is no dominant air mass (i.e. there are equal chances of an event originating from any of the adjacent zones). For example, between the Mt and mP zones, this buffer stretches from west/central Colorado through central Kansas and eastward into Pennsylvania (Fig. 1). The locations within these buffer regions will be the most susceptible to changes in regional weather patterns.

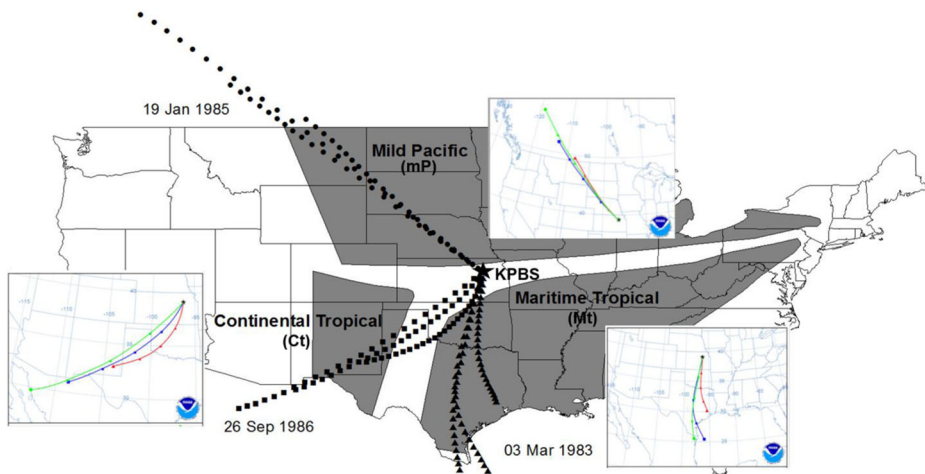


Fig. 1 Delineation of the three most common air mass source regions (gray; derived from Bryson et al. 1966) with example backward trajectory HYSPLIT model outputs from various dates relative to KPBS (39.08 N, -96.35 W). In this study, backward trajectories track the movement of an air parcel beginning 24 h from midnight of the date of a precipitation event greater than 1 mm at KPBS. On each date, the different colored trajectories represent three vertical levels above ground level (red = 500 m, blue = 1500 m, and green = 3000 m)

The goal of this study was to determine historical trends in the total number and proportion of precipitation events from multiple source regions relative to Konza Prairie Biological Station (KPBS), a central U.S. grassland research site located within the buffer area between the Mt and mP zones. Weekly precipitation chemistry and precipitation event sources were also compared to determine if source location is important for the deposition of a variety of ecologically relevant solutes. Information from this study can be used to determine the extent of changing air mass patterns and precipitation chemistry and the possible effect of interactive changes on central U.S. grassland ecosystems.

2 Methods

2.1 Study area

Konza Prairie Biological Station is in the Flint Hills ecoregion and is characterized by rolling hills, with cherty limestone and shale outcrops. The Flint Hills retains the largest extent of intact native tallgrass prairie in the Great Plains (Omernik 1987). The KPBS is a 35-km² nature reserve and Long-Term Ecological Research (LTER) site owned by The Nature Conservancy and managed by the Kansas State University Division of Biology. Watershed-level treatments include prescribed burning (with burn frequencies of 1, 2, 4, and 20 years in sub-watersheds), and grazing by the American bison (*Bison bison*; 1/3 of total area openly grazed by bison).

2.2 Precipitation collection and chemistry

Daily precipitation events and volumes were used from the KPBS Long-Term Ecological Research (LTER) database. Daily precipitation volumes were sampled during the length of the dataset using either a Belfort weighing rain gauge (1983–2000) or an Ott Pluvio² rain gauge

(2000-present). Weekly precipitation volumes and chemistry were taken from the Konza Prairie (site ID: KS31) National Atmospheric Deposition Program (NADP) database. Briefly, weekly precipitation samples were collected from an Aerochem Metrics Wet/Dry collector and directly sent on ice to the Central Analytical Lab, Champaign, IL to be analyzed for conductivity, pH, and the concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , Cl^- , Na^+ , K^+ , Ca^+ , and Mg^+ . More information regarding the NADP program can be found online (<https://nadp.slh.wisc.edu/>). Annual volume-weighted mean (VWM) concentrations and deposition were calculated using the following equations.

$$\text{annual VWM} = \frac{\sum \left(\text{weekly solute concentration} \left(\frac{\text{mg}}{\text{L}} \right) \times \text{weekly precipitation} (\text{L}) \right)}{\sum (\text{weekly precipitation} (\text{L}))} \quad (1)$$

$$\text{nutrient deposition} = \text{weekly solute concentration} \left(\frac{\text{mg}}{\text{L}} \right) \times \text{precipitation} (\text{mm}) \quad (2)$$

2.3 Precipitation event trajectories

The online version of the NOAA Air Research Lab HYSPLIT model (<http://ready.arl.noaa.gov/HYSPLIT.php>; Stein et al. 2015) was used to determine the regional source apportionment via a 24-h backward trajectory of precipitation causing air parcels (precipitation events; defined as greater than 1 mm) across various vertical heights from 1983 to 2006 relative to KPBS (39.08 N, -96.35 W). The National Center for Atmospheric Research (NCAR) Reanalysis Project dataset, which has a 210 km grid spacing (Kalnay et al. 1996), was used as the model input (Kelly et al. 2009). Source regions were classified based on the observations of Bryson 1966 (see above and Fig. 1 for a delineation of source regions and example model outputs). Daily precipitation event sources were determined from the location of an air parcel 24 h prior to midnight on the day of a precipitation event greater than 1 mm occurring at KPBS. Each event included the trajectory source classifications at 500, 1500, and 3000 m above ground level (AGL). These levels were used to characterize vertical variability. Linear regression was used to determine statistical differences in the annual total number and proportion of events from each source at each vertical level.

2.4 Matching trajectories with precipitation chemistry

In order to align precipitation event source with wet deposition and nutrient concentrations, daily precipitation event sources were scaled to each week of available NADP data. Precipitation chemistry comparisons across event sources were made between events that originated north or south of KPBS (i.e. events originating from the Mt and Ct zones were combined into a south category). This was done because of the relatively few events that originated from the Ct zone. If multiple events were recorded from different zones within a week, that weekly measurement was removed from this part of the analysis. Similarly, only events that had a consensus source location for all three vertical levels from north or south were included in the dataset. For events from the south, a week of data was included if two of the three vertical heights were the same source (i.e. Mt at two levels and Ct at the other, or vice versa). If the event was from the north, all three vertical levels had to be from the mP source. There were a

total of 624 events with at least one vertical level from the north, and of these 352 (56%) were removed from this analyses because of disagreement at one vertical level. Another 70% were removed because of multiple events from different sources in the same week for a total of 82 events included in this analysis from the north. There were a total of 964 events with no event originating from the north source region (e.g. a combination of Mt, and Ct only), and of this 69% were removed because of multiple events from different sources in the same week, for a total of 303 events from the south. The annual average number of events that qualified for this dataset differed between the sources (13 from the south and 4 from the north). Furthermore, approximately 30% of the annual VWM measurements from the north included 2 or fewer events. In 2001 there were no events where all three vertical levels originated from the north. Note that due to the removal of these data the annual VWM do not represent the true average over a year, but the average of annual events for this study. I tested the potential for biased results due to differences in sample size by randomly selecting 4 data points from each year from events from the southern source region (to match the average from the north). Subsequent analyses found similar statistical results (Supplementary Information). Therefore, I elected to use the full dataset available.

Box plots were used to qualitatively compare the variation of annual VWM precipitation chemistry for each solute and source across the time period of this study. Analysis of Covariance (ANCOVA) was used to quantitatively compare differences in the deposition between north and south regions of four ecologically relevant solutes (NO_3^- , NH_4^+ , H^+ , and SO_4^{2-}), with Direction (North or South) as the main factor and Year as a covariate to account for annual variation. I also compared the annual average proportion of reduced N ($\text{NH}_4^+/\text{NH}_4^+ + \text{NO}_3^-$) between regions and over time. A significant Direction effect suggests a statistical difference in the intercept of the regression between the two directions after accounting for annual variability. A significant Year effect suggests that solutes are statistically different across the 23-year time period. If Year was not a significant factor in the ANCOVA model, the model was simplified to an Analyses of Variance (ANOVA) by removing the Year covariate. For both analyses data was square root transformed if the data failed the Shapiro-Wilk normality test. For all ANCOVA's there was not a significant interaction between Year and Direction. After checking that the interaction was not significant, the term was removed from the model. I tested the validity of the model by removing the Direction term and comparing this new model (Year only), with the full model (Direction +Year). For all comparisons, these models were significantly different, suggesting that Direction explained a significant portion of variance and should remain in the model.

3 Results

3.1 Directional trends in precipitation events

A total of 1588 trajectory models were run for all precipitation events greater than 1 mm across the 23-year period. The total number of precipitation events varied annually but did not change significantly across the length of the time period (between 27 and 40; mean = 33). Total annual precipitation also varied but was not statistically different across the 23-year study period (between 419 and 1191 mm; mean = 786 mm). Total annual precipitation did not change from events originating from any of the three directions during the 23-year study period (means, Ct = 67 mm, Mt = 364 mm, mP = 46 mm). For every year of the dataset, more events

originated from Mt locations at the 500 and 1500 m heights than any other location. At the 3000 m height, events from the Ct location were marginally dominant; however, data from this level were highly variable with no significant trends for any source location (Fig. 2). The number of events from Mt did not change significantly over time at the 500 or 1500 m levels; however, there were significant decreases in precipitation events originating from mP and Ct at the same levels (Fig. 2, Table 1).

The proportion of precipitation events from Mt was annually higher at the 500 and 1500 m levels. Data from the 3000-m level were again highly variable, with no significant differences or dominant trends. The proportion of precipitation events from Mt increased significantly over time at the 500 and 1500 m level, while the proportion of events from Ct and mP decreased significantly (Fig. 2). The declining frequency of precipitation events originating at the Ct and mP was similar at the 1500-m level (-0.0046 and -0.0036 , respectively).

3.2 Precipitation chemistry and deposition

The distribution of annual VWM concentrations was generally higher in the north compared to the south for most solutes (Fig. 3). The exceptions to this trend included Cl, and Na (Fig. 3), which were the only solutes that had similar distributions of concentrations from the north and south.

An ANCOVA was used to quantitatively determine differences between events originating north and south of KPBS of deposition of NO_3^- , NH_4^+ , H^+ , and SO_4^{2-} . Annual NO_3^- deposition changed significantly across the study period; however, events originating from the south were approximately twice as high as deposition from the north ($p < 0.001$; Fig. 4 a). Annual NH_4^+ deposition increased during the study period from both directions ($p = 0.01$). Ammonium deposition from the south nearly doubled over the study period, while NH_4^+ deposition from the north increased marginally (Fig. 4 b). Ammonium deposition from the southern region was significantly higher than from the northern region ($p < 0.001$; Fig. 4 c). The deposition of H^+ ions decreased during the study period from both directions ($p < 0.004$), with H^+ deposition decreasing by half from the south and decreasing marginally from the

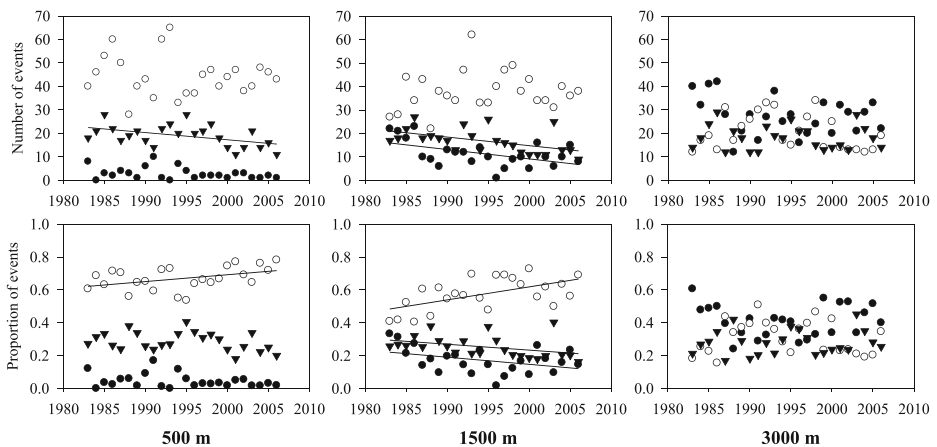


Fig. 2 Annual change in number and proportion of precipitation events across air mass origins (blank circles = Mt, triangles = mP, filled circles = Ct) at three different elevations above ground level from 1983 to 2006 relative to KPBS. Data with trend lines are statistically significant ($p < 0.1$, Table 1)

Table 1 Summary of significant linear relationships between the number and proportion of precipitation events from different sources across the 23 yr. period at different vertical levels above ground level. Significance was determined with a $p < 0.1$

Number of events	Location	r^2	Slope
500 m	mP	0.22	-0.3065
1500 m	Ct	0.25	-0.3987
1500 m	mP	0.26	-0.3670
Proportion of events	Location	r^2	Slope
500 m	Mt	0.18	0.0042
1500 m	Ct	0.15	-0.0043
1500 m	mP	0.15	-0.0036
1500 m	Mt	0.33	0.0079

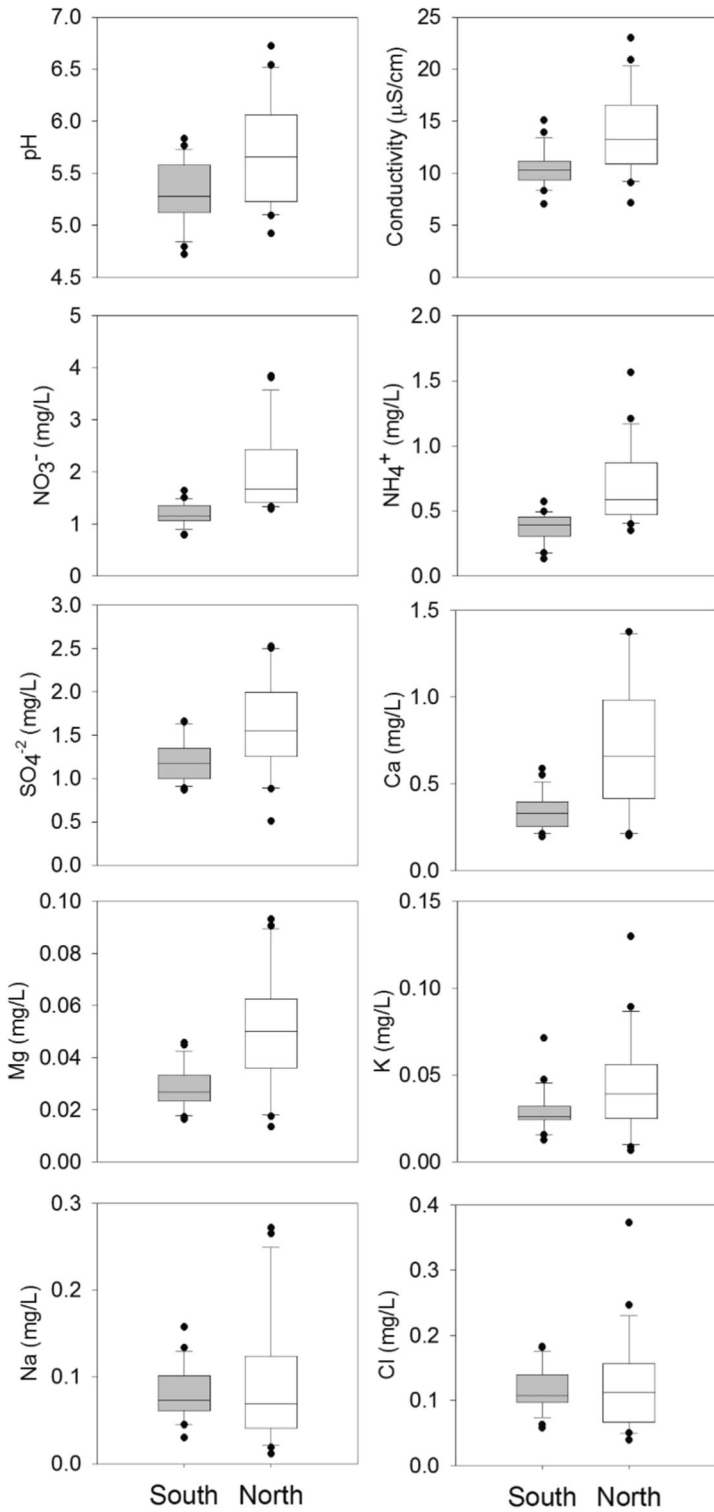
north. Hydrogen ion deposition from the south was significantly higher than from the north ($p < 0.001$; Fig. 4 c). The SO_4^{-2} deposition did not change statistically over the study period, but SO_4^{-2} deposition from the south was approximately 3-fold higher than the north ($p < 0.001$; Fig. 4 d). The proportion of reduced N increased significantly over the study period by approximately 5% ($p < 0.001$) and was not statistically different between source regions (Fig. 5).

4 Discussion

4.1 Implications of proportional shifts in air mass origin on nutrient deposition

Regional shifts in weather patterns are likely to affect the structure, function, and productivity of grassland ecosystems. Changes in weather patterns are especially important in the central U.S. grasslands, which are susceptible to weather pattern shifts because of their location between zones of major air mass dominance. At the 1500-m level, the proportion of events from each source location was less than 50% at the beginning of the study period, which is similar to historical analyses of air mass trajectory dominance for this region (Bryson 1966). Furthermore, the proportion of events from each source changed significantly at the 1500-m level during the study period. The annual number of events from the Mt location at the 1500-m level did not change significantly; however, decreases in the number of events from mP and Ct resulted in the proportion of events from the Mt location to increase from ~40 to ~60%. Therefore, the dominant zone of Mt air mass patterns has shifted northward to include at least portions of northeast Kansas.

The shift towards a higher proportion of events from the southern region may have implications for nutrient deposition for the Great Plains. While annual VWM concentrations were typically higher from the north, overall deposition rates were higher from the south due to substantially higher precipitation amounts from events originating in the southern source region. For both nitrogen species (NO_3^- and NH_4^+) this meant that events from the south have ~2-fold higher deposition than events from the northern region. Total nitrogen deposition on KPBS was relatively constant across the study period, with deposition rates varying between 4 and 7 $\text{kg ha}^{-1} \text{ yr}^{-1}$ (NADP 2007, site KS31). The critical load for N deposition (i.e. the deposition where detrimental ecological effects become evident) in the tallgrass prairie component of the U.S. Great Plains is estimated to be between 5 and 15 $\text{kg ha}^{-1} \text{ yr}^{-1}$ (Pardo



◀ **Fig. 3** Box plots of the annual volume-weighted mean (VWM) pH, conductivity, and concentrations of molecules and elements from precipitation events originating south (Ct and Mt; solid boxes) or north (mP source; open boxes) of KPBS across the 23-year study. Bars within boxes indicated median values. Upper and lower boundaries of boxes represent 25th and 75th quartiles, respectively. Error bars represent 10th and 90th percentiles, and solid circles represent those values outside the 10th and 90th percentiles

et al. 2011). A recent analysis of ecophysical, plant community and ecosystem metrics from KPBS during the time period of this study found little evidence for detrimental effects of excess N on ecosystem structure and function (McLauchlan et al. 2014). However, if the proportion of events continues to move towards the dominance of southern originating wet deposition, this could mean higher wet nutrient deposition rates than previously observed, possibly exceeding critical loads in the future.

4.2 Land-use did not affect ammonium deposition

Ammonium deposition increased over the study period, as has been observed for many parts of the U.S. (Li et al. 2016, Du et al. 2014). Ammonium deposition was significantly larger when

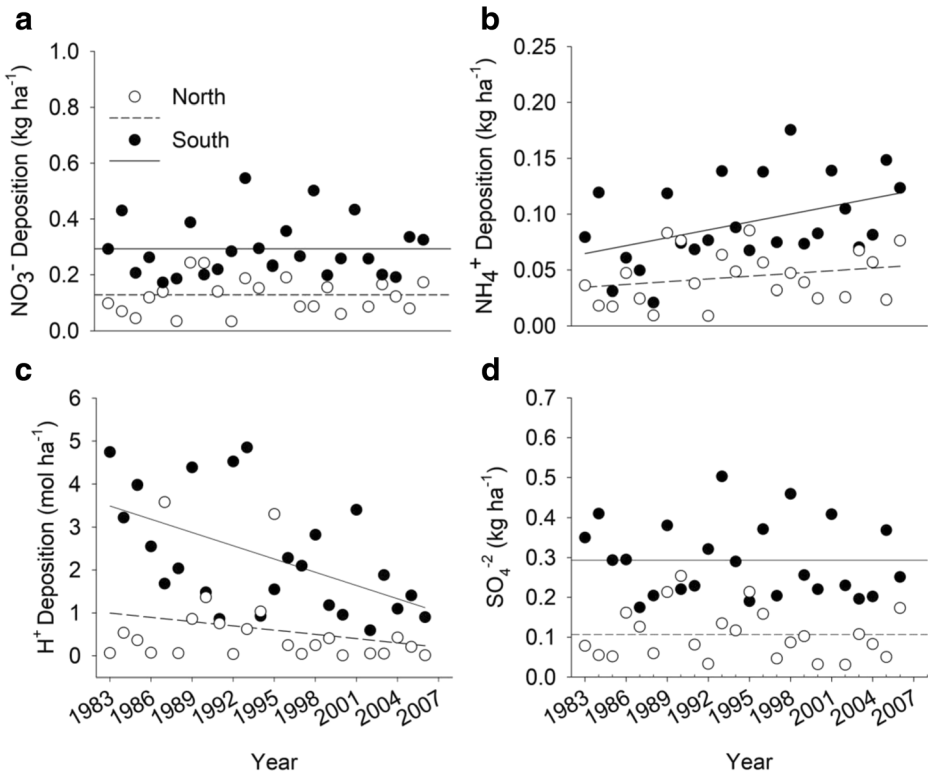


Fig. 4 Average annual deposition of ecologically relevant nutrients (**a** = nitrate, **b** = ammonium, **d** = sulfate) and hydrogen ions (**c**) across the 23-year period of this study and source region. The slopes and intercepts from each direction were tested using ANCOVA. Open circles represent average deposition from precipitation events originating from the north of KPBS, filled-in circles represent average deposition from the south of KPBS. Data with trend lines are statistically significant ($p < 0.05$). Nitrate and sulfate deposition did not change significantly over time but were different between source regions. Therefore, the line represents the mean from each source across the study period

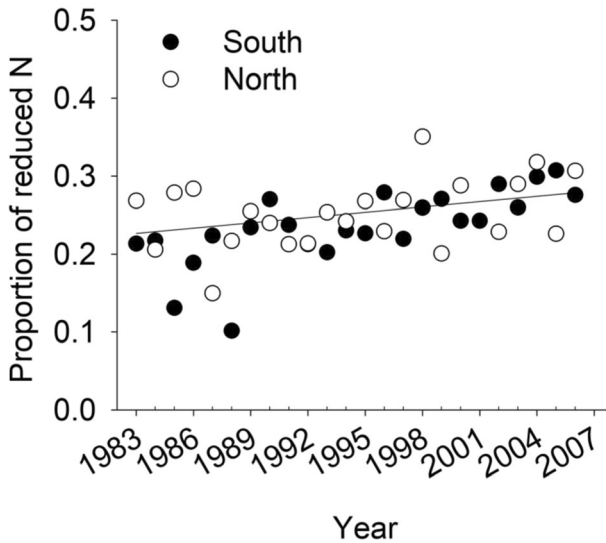


Fig. 5 The proportion reduced N between source regions over time. The proportion increased significantly over time but was not different between source regions

precipitation originated from the south, but as mentioned above this is likely due to higher precipitation amounts, as the proportion of reduced N was not significantly different between source regions. This is somewhat surprising given the drastic differences in the amount of agriculture, the primary emitter of reduced N, between the two regions. Total cropland (including cropland used for crops, cropland used only for pasture, and idle cropland) in the Northern Plains (e.g. The Dakota's, Nebraska, and Kansas; northern trajectory relative to KPBS) is 50.3% of the total area compared to 22.2% of the total area of the Southern Plains (e.g. Oklahoma and Texas; southern trajectory relative to KPBS) based on data from 2007 (Nickerson et al. 2011). In 2005, the total acres of corn treated with N fertilizers in the Northern Plains was 74,000 km² compared to 8500 km² in the Southern Plains (Ribaud et al. 2011). A meta-analysis of N loss from croplands suggests that approximately 8% of the N applied to cropland in industrialized countries is volatilized as ammonia (NH₃-N; Bouwman et al. 2002). Overall, wet ammonium deposition increased over the study period like national trends and was not affected by the source region of the precipitation. Nonetheless, increasing wet deposition of reduced N could result in the eutrophication of both terrestrial and aquatic ecosystems in the Great Plains.

4.3 Unexplained decreases in H⁺ deposition from the southern source region

The decrease in H⁺ deposition over time followed national trends, but deposition originating in the south decreased much faster than events originating from the north. In many regions of the U.S., decreases in H⁺ deposition is often associated with decreases in SO₄⁻² as it is the dominant strong acid anion present in precipitation (Rice et al. 2014, Vet et al. 2014) and has been steadily decreasing nationwide since the passage of the Clean Air Act (Baumgardner et al. 2002). However, SO₄⁻² deposition, along with NO₃⁻ deposition (another strong acid anion), was relatively constant from both source regions during the study, and likely do not explain the dramatic decrease in H⁺ deposition from the southern source region. It is beyond

the scope of this study to determine the exact explanation of changes in precipitation pH that may lead to changes in H^+ deposition, but possible explanations other than changes in SO_4^{2-} and NO_3^- could include changes in organic acids (Galloway et al. 1976, Vet et al. 2014), and Brønsted acids associated with dissolved Al and Fe (Galloway et al. 1976).

5 Conclusion

In this study, I used a long-term dataset (NADP) and an atmospheric model (HYSPLIT) to analyze patterns of wet deposition between two distinct source regions relative to a tallgrass prairie research station. These trends reveal that the proportion of precipitation events from the southern region increased significantly over the study period. Wet deposition of ecologically relevant solutes was higher from events from the southern region, likely because of higher precipitation amounts. Combined, more events from the southern region with higher wet deposition (particularly reduced N) may result in the eutrophication of terrestrial and aquatic ecosystems in this endangered ecosystem in the future.

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