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Atmospheric concentrations and deposition of reactive nitrogen in Grand Teton National Park

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[1] The Grand Teton Reactive Nitrogen Deposition Study (GrandTReNDS) was conducted to provide a more complete look at atmospheric concentrations and deposition fluxes of various reactive nitrogen species in and around Grand Teton National Park (GTNP). Daily measurements of wet deposition, $PM_{2.5}$ composition, and gaseous ammonia and nitric acid concentrations were made at three locations. Weekly measurements of gaseous ammonia were made at eight additional sites. Ammonia concentrations were higher at the western sites; the study average ammonia concentration west of GTNP was 35 nmol m^{-3} and on the east side of the park it was 18 nmol m^{-3}. Concentrations of other measured reactive nitrogen species were lower than NH_3 and fairly similar at all sites, with averages of approximately 9, 1, and 3 nmol m^{-3} for ammonium, nitrate, and nitric acid, respectively. Wet deposition of ammonium and dry deposition of ammonia were the largest reactive nitrogen deposition pathways, together accounting for 56% and 62% of the nitrogen deposition on the east and west sides of GTNP, respectively. Nitrogen deposition of measured species totaled 2.38 kg N ha^{-1} west of GTNP at Driggs, ID (6 April to 21 September 2011), 0.85 kg N ha^{-1} west of GTNP at a high-elevation site (28 July to 21 September 2011) and 1.23 kg $N \cdot ha^{-1}$ at a location on the east side of GTNP (15 May to 21 September 2011). These measurements highlight the significant inputs of reactive nitrogen to regional ecosystems over the few months studied, the importance of including NH_3 dry deposition in nitrogen deposition budgets, and the need to conduct further research to capture sources and the annual cycle of deposition.

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1. Introduction

[2] Limited information exists concerning current levels of nitrogen deposition in Grand Teton National Park (GTNP). Several national network monitoring sites are located in the broader region surrounding the park, but due to complex topography, varying spatial emissions, and the short atmospheric lifetime of pollutants such as ammonia, it is unknown if these sites are representative of reactive nitrogen deposition in GTNP. Several intensive studies have previously taken place further south in Rocky Mountain National Park. These studies suggest we should be concerned about levels of nitrogen deposition in the Rocky Mountains both from an analysis of ecological communities and atmospheric measurements [*Rueth and Baron*, 2002; *Burns*, 2003; *Bergstrom and Jansson*, 2006; *Elser et al.*, 2009; *Beem et al.*, 2010; *Benedict et al.*, 2012, 2013]. This study was designed to characterize levels of nitrogen deposition further north in the Rocky Mountains in and near GTNP.

[3] Rates of reactive nitrogen deposition globally, and specifically in the Rocky Mountain region, have risen due to increased use of fertilizers and burning of fossil fuels [*Burns*, 2003; *Galloway et al.*, 2008; *Sutton et al.*, 2008]. The increase of nitrogen inputs from these sources has large and long-lived effects on both ecosystem productivity and nitrogen processing [*Schimel et al.*, 1997]. There is a growing weight of evidence that nitrogen deposition is adversely impacting ecosystems in the western United States [*Craig and Friedland*, 1991; *Williams et al.*, 1996; *Williams and Tonnessen*, 2000; *Baron et al.*, 2000; *Kochy and Wilson*, 2001; *Fenn et al.*, 2003]. Many natural plant communities are nitrogen limited, and altering the nitrogen content may damage the way the community functions, degrade water quality, and lead to forest decline [*Rattray and Sievering*, 2001]. National Parks are of particular interest since they

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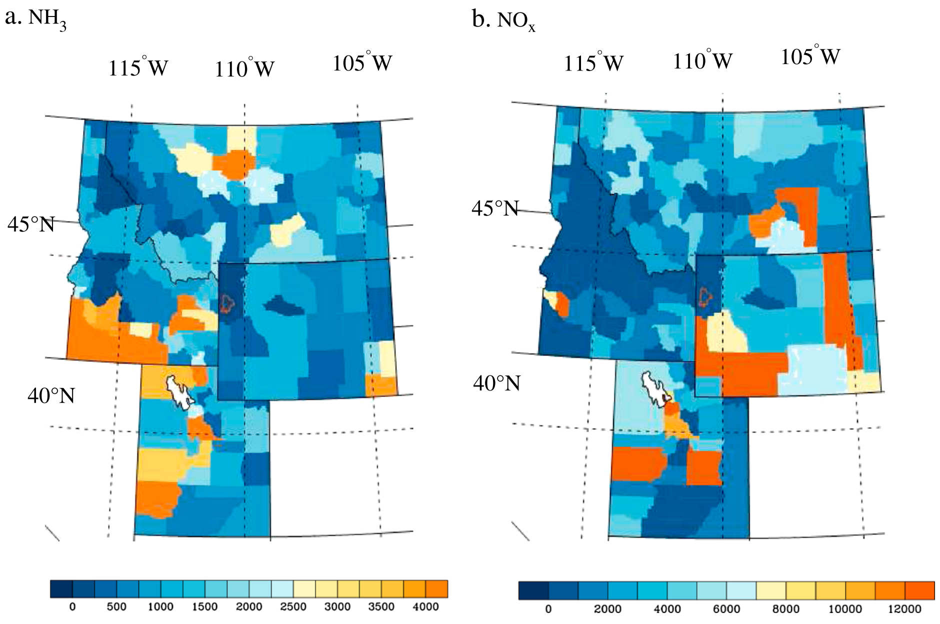

Figure 1. Annual emissions (in tons) by county for (a) NH_3 and (b) NO_x from the National Emissions Inventory, 2008 [*National Emissions Inventory*, 2008]. Grand Teton National Park is outlined in red for reference.

are protected from significant changes by U.S. federal law, including the Clean Air Act, the Wilderness Act, and the NPS Organic Act of 1969. Previous research in Rocky Mountain National Park (RMNP) has shown that increases in atmospheric deposition of nitrogen correlate with changes in biological species [*Baron*, 2006].

[4] Research to define the critical load, the level at which negative changes to an ecosystem begin to occur, and current rates of nitrogen deposition has been particularly important for policy makers to understand the impacts of nitrogen deposition and begin to take steps to reduce reactive nitrogen emissions and further protect sensitive ecosystems. Research in the Greater Yellowstone Ecosystem (includes GTNP) indicates changes in diatom assemblages from nitrogen enrichment that began around 1980 and corresponded to an estimated 1.4 kg N $ha^{-1}yr^{-1}$ wet deposition of nitrate and ammonium [*Saros et al.*, 2010]. Further south, in RMNP, changes in diatom species took place earlier, between 1950 and 1964 when wet inorganic nitrogen deposition was approximately 1.5 kg N $ha^{-1}yr^{-1}$ [*Baron*, 2006]. Increasing nitrogen deposition is a concern in both areas, because the ecosystems are very sensitive to excess nitrogen. The high-elevation ecosystems in RMNP and GTNP have several common characteristics including low buffering capacity from rocky soils and short growing seasons which contribute to their sensitivity. Research has found that 53% of lakes in RMNP and GTNP are very sensitive to acid deposition and have high rates of inorganic nitrogen and sulfate deposition [*Nanus et al.*, 2009]. Specifically, lakes above 2790 m in GTNP are predicted to have the greatest sensitivity to acidification from atmospheric deposition compared to lakes at lower elevations [*Nanus et al.*, 2005]. While some research has specifically targeted GTNP [*Clow et al.*, 2003], there is still an incomplete picture of the current level and trends of nitrogen deposition in the region

[5] Nitrogen deposition in GTNP has mainly been examined as an extension of research in Yellowstone National Park. However, GTNP is closer to several important source regions suggesting nitrogen deposition there could be very different from Yellowstone. Reactive nitrogen arises from a diverse array of sources, and GTNP is surrounded by sources of reactive nitrogen species that could be transported to and deposited in the park. These include large ammonia emissions from agricultural activities in the Snake River Valley and northern Utah to the west of GTNP (Figure 1a); emissions of oxidized nitrogen from oil and gas extraction and power plants in Wyoming and beyond (Figure 1b); and emissions of various nitrogen species from wild and prescribed fires. In addition, nitrogen emissions occur in the neighboring communities, more distant urban, industrial, and transportation sources, and natural processes within the park. High NH_3 emissions in the Snake River Valley and along the Idaho/Wyoming border are likely to impact GTNP since prevailing winds in the western U.S. are generally westerly. However, in the summer, circulation patterns associated with the North American monsoon can dominate surface winds (Figures S1 and S2 in the supporting information) and transport from regions farther south in Utah may also be important.

[6] Current data on nitrogen deposition trends in the region are provided by the National Acid Deposition Program/National Trends Network (NADP/NTN); however, there is no NADP site located in GTNP. The closest site is in Gypsum Creek, Wyoming, approximately 70 km southeast of GTNP. To examine nitrogen deposition in the region, the four NADP sites closest to GTNP are plotted on a map of the region along with seasonal deposition data (Figure 2). Two of the sites (Yellowstone and Pinedale) are colocated with Clean Air Status and Trends Network (CASTNet) sites which quantify dry deposition of ammonium (NH_4^+), nitrate (NO_3^-), and nitric acid (HNO_3). Deposition peaks in summer at all sites except Craters of the Moon, where deposition is largest in spring. On average dry deposition of NH_4^+, NO_3^-, and HNO_3 together contribute 30% and 20% of total

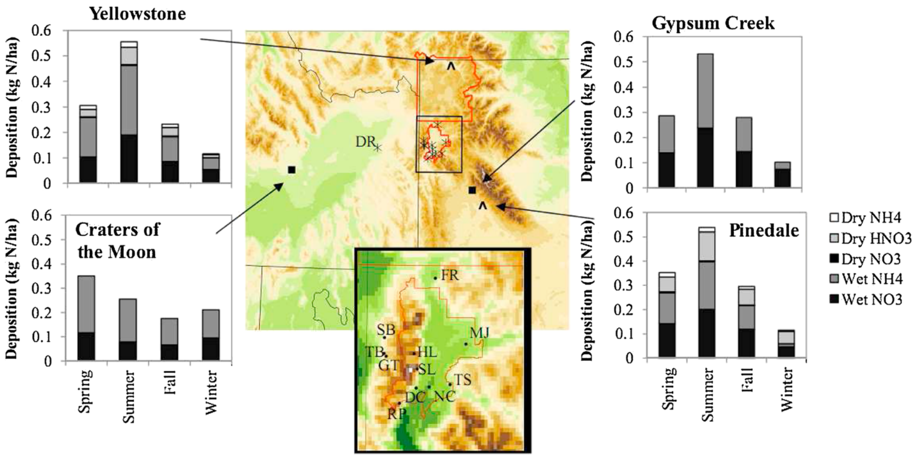

Figure 2. Site map showing topography in the region, national network sites in the region, and average (2005–2010) historical deposition data from those sites by season. National park boundaries are outlined in red and the black box shows the location of the inset map where sites for Grand TReNDS are labeled by abbreviation (see Table 1).

quantified nitrogen deposition based on the NADP and CASTNet measurements at Pinedale and Yellowstone, respectively. Average wet inorganic nitrogen deposition at Gypsum Creek was 1.2 kg N ha^{-1}yr^{-1} and at Pinedale was 0.94 kg N ha^{-1}yr^{-1}. Although these values are similar, given measurement uncertainties, it is important to make measurements at higher spatial resolution than currently available to assess deposition levels in GTNP. The complex topography in the region results in precipitation differences with altitude that can strongly impact deposition fluxes. These observations also do not include wet and dry organic nitrogen deposition or dry ammonia deposition. Studies in Rocky Mountain NP showed that wet organic nitrogen deposition and dry ammonia deposition accounted for about a third of the total measured nitrogen deposition [*Beem et al.*, 2010; *Benedict et al.*, 2012].

[7] In 2011, the Grand Teton Reactive Nitrogen Deposition Study (GrandTReNDS) was conducted to examine gas, aerosol, and precipitation chemistry in an effort to better understand the composition, levels, and spatial and temporal variability of the reactive nitrogen deposited in GTNP. In addition, understanding the sources influencing the reactive nitrogen deposition was a primary goal. The study focused on seasons with historically high regional deposition, spring and summer. Results discussed here include temporal and spatial variability in atmospheric nitrogen compound concentrations at several sites located in and around GTNP and construction of a reactive nitrogen deposition budget for the region for spring and summer 2011, including wet deposition of organic and inorganic nitrogen and dry deposition of ammonia, nitric acid, and fine particle organic and inorganic nitrogen. The findings are examined in the context of the critical load for nitrogen deposition in the region and priorities outlined for future research and monitoring efforts.

2. Methods

2.1. Site Information

[8] In 2011, measurements were made at a network of 12 sites in and around Grand Teton National Park (Figure 2). A list of sites including coordinates, elevations, and abbreviations is given in Table 1. Sampling began 6 April 2011 at Driggs, Idaho (DR), and sites were added at Grand Targhee Resort (TB) and the NOAA Climate Center (NC) at Moose Junction, Wyoming, shortly thereafter. The deployment of additional sampling sites was delayed due to high snowfall during the 2010–2011 winter season and resulting limited site access; sampling start dates are listed in Table 1. A core measurement site was located at 2722 m elevation atop a

Table 1. Site Information for 2011 Grand Teton National Park Study Including Coordinates, Elevation, and Measurements of Interest Ordered From West to East

		Latitude	Longitude	Elevation (m)	Start of Sampling	Passive Site	24 h Gas and Particle	Precipitation
Driggs	(DR)	43.7404	−111.8703	1947	6 April	X	X	X
Lower Grand Targhee	(TB)	43.7891	−110.9558	2454	21 April		X	
South Badger	(SB)	43.8504	−110.9543	2166	21 July	X		
Upper Grand Targhee	(GT)	43.7782	−110.9438	2722	28 April	X	X[a]	X
Rendezvous Peak	(RP)	43.5969	−110.8703	3176	21 July	X		
Holly Lake	(HL)	43.789	−110.7939	2826	24 July	X		
Death Canyon	(DC)	43.6566	−110.7818	2088	21July	X		
Surprise Lake	(SL)	43.7291	−110.7768	2922	24 July	X		
NOAA Climate Station	(NC)	43.6614	−110.712	1978	15 May	X	X	X
Flagg Ranch	(FR)	44.0827	−110.6828	2086	5 July	X		
Tetons Science School	(TS)	43.6709	−110.5996	2131	21 July	X		
Moran Junction	(MJ)	43.8276	−110.5156	2062	5 July	X		

[a] Precipitation sampling began 24 July 2011, and 24 h gas and particle sampling began 13 July 2011.

peak at Grand Targhee Resort (Upper Grand Targhee; GT). This site is just upwind (approximately 7 km to park boundary) of GTNP and was selected to better represent high-elevation inputs on the west side of the park (power and equipment access are not possible within the western part of the park itself). The Driggs site (1947 m elevation) is located in the Teton Valley, which is adjacent to the northeast end of the Snake River Valley and is closer to major agricultural NH_3 sources. The NOAA Climate Center is a lower elevation site (1978 m) on the east side of the Teton Range closer to NO_x sources in western Wyoming. On 28 April 2011, a passive ammonia sampler was deployed at Upper Grand Targhee site and was accessed by snowshoe every 2 weeks. Due to the high snowfall, however, the majority of measurements did not start until much later, when the access road to the site was clear of snow on 24 July 2011.

[9] At the three most comprehensive measurement sites (Driggs, Upper Grand Targhee, and NOAA Climate Center), daily URG annular denuder/filter-pack (URG Corporation, Chapel Hill, NC) samples and daily precipitation samples were collected. Additional measurements at the core site (Upper Grand Targhee) included high-volume particle samplers, high time resolution NO_x, NO_y, and NH_3 measurements, and particle size distribution measurements. High time resolution PM_1 composition was also measured using an Aerodyne high resolution aerosol mass spectrometer (AMS) for a portion of the study. Of these additional measurements, only analysis from the weekly high-volume particle samplers will be discussed here. (A. J. Prenni et al., Gas-phase reactive nitrogen near Grand Teton National Park: Impacts of transport, anthropogenic emissions, and biomass burning, submitted to *Atmospheric Environment*, 2013 discuss the gas-phase observations in details; other measurements will be discussed in forthcoming papers). High-volume samplers were also located at the NOAA Climate Center. Meteorological stations were located at Driggs, Upper Grand Targhee, and the NOAA Climate Center. Results presented here focus mainly on the 24 h gas and particle chemistry and precipitation measurements along with spatial gradients in ammonia concentrations measured with passive samplers across the full sampling network.

2.2. Sample Collection

2.2.1. URG

[10] Twenty-four hour samples were collected using URG annular denuder/filter-pack samplers from 8:00 A.M. to 8:00 A.M. MDT with a nominal volumetric flow of 10 L min^{-1}. Air was first drawn through a Teflon-coated cyclone ($D_{50}=2.5$ μm) followed by a denuder coated with a sodium carbonate solution for collection of nitric acid and sulfur dioxide [*Lee et al.*, 2008] and then a denuder coated with phosphorous acid solution to collect ammonia. Air then passed through a nylon filter (PALL Nylasorb 1 μm pore size, 37 mm) to collect particulate matter. Finally, the flow traveled through a backup denuder coated with phosphorous acid to capture any volatilized particulate ammonium. We have previously demonstrated [*Yu et al.*, 2005] that volatilized nitrate is retained by the nylon filter. The actual sample volume was measured using a downstream dry gas meter with an appropriate correction for the measured system pressure drop.

[11] Blanks were collected once a week to determine the minimum detection levels (MDL) which are as follows: 0.49, 0.40, 0.56, and 0.46 nmol m^{-3}, for Cl^-, NO_2^-, NO_3^-, and SO_4^{2-} and 0.42, 0.36, 0.04, 0.26, and 1.2 nmol m^{-3} for Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+} for the $PM_{2.5}$ filter samples. For the gas-phase measurements, the MDLs are 4.6, 0.48, and 0.08 nmol m^{-3} for NH_3, HNO_3, and SO_2. Sample concentration precision values (expressed as relative standard deviation (RSD)) were calculated from replicate measurements collected once a week. The $PM_{2.5}$ ion precision values were all less than 10% RSD, while the RSDs for NH_3, HNO_3, and SO_2 were 7%, 11%, and 21%. Sample pairs with concentrations less than the MDL were excluded from the precision calculation.

2.2.2. High-Volume Filter Sampling

[12] A Thermo Fisher Scientific TSP (total suspended particulate) High-Volume sampler with a $PM_{2.5}$ impactor plate was operated to collect daily and weekly samples at a flow rate of 850 L min^{-1}. Samples were collected on Whatman Quartz Microfibre Filters (20.3 × 25.4 cm) which were prebaked at 550°C for 12 h to remove background carbon and nitrogen. For total nitrogen analysis, 10 punches (4.9 cm^2/each punch) were extracted in 20 mL of deionized (DI) water by sonicating for 1.25 h at 35°C. The extracts were allowed to cool for 2.5 h and then were filtered (Whatman Puradisc 0.2 μm). Ten milliliters of extract were diluted with 10 mL of deionized (DI) water to get sufficient sample for analysis.

2.2.3. Precipitation Samplers

[13] Precipitation samples were collected using automated precipitation collectors (Yankee Environmental Systems TPC-3000 and NCON Systems ADS/NTN Atmospheric Precipitation Sampler). Sampling buckets were cleaned with DI water, shaken to remove water, covered with Al foil, and allowed to dry. The bucket was weighed prior to installation in the field and upon returning to the lab to determine the total sample volume. Blanks were taken periodically by pipetting 30 mL of high-purity deionized water into the bucket. The pH was measured after returning to the lab using a combination pH electrode calibrated with pH 7 and 4 buffers. The remaining sample was frozen until analysis. Sampling buckets were changed daily at Driggs and Upper Grand Targhee, and every other day at NOAA Climate Center because of the distance from the base of operations (Driggs). The MDLs of the nitrogen species are as follows: 0.026 mg N L^{-1} and 0.014 mg N L^{-1} for NH_4^+ and NO_3^- and 0.09 mg N L^{-1} for organic nitrogen. The RSDs for NH_4^+, NO_3^-, and organic nitrogen replicate analyses were 6%, 9%, and 7%.

2.2.4. Passive Samplers

[14] Radiello radial passive samplers were used to determine the ammonia spatial distribution across the sampling region, including at remote locations where electric power was unavailable. The ammonia passive sampler includes a blue microporous diffusive body (RAD 1201), an adsorbing cartridge impregnated with phosphoric acid (RAD 168) and a vertical adapter (RAD 122) (Radiello, Padova, Italy). The passive samplers were installed in an inverted bucket, a shelter to keep direct sunlight, precipitation, and wind from impacting the sampling cartridge [*Day et al.*, 2012]. Passive samples were deployed at 11 sites across GTNP; at Driggs, NOAA Climate Center, and Upper Grand Targhee; they were collocated with URG samplers. Passive ammonia samples were typically deployed for a 2 week duration to ensure sufficient ammonia mass collection. From the GrandTReNDS field blanks,

Table 2. Average Atmospheric Concentrations (nmol m^{-3}) at Standard Pressure From 13 July to 21 September 2011 for the Sites From This Study and From 13 July to 21 September for Studies Previously Conducted in Rocky Mountain National Park

	DR	TB	GT	NC	RMNP 2009	RMNP 2010
Na^+ (p)	0.8	0.8	0.7	0.7	0.4	0.5
NH_4^+ (p)	9.5	9.1	9.1	8.7	10.9	8.9
K^+(p)	0.6	0.5	0.6	0.5	0.3	0.3
Mg^{2+} (p)	1.0	0.3	0.3	0.4	0.2	0.1
Ca^{2+} (p)	5.5	1.6	1.5	2.2	0.9	0.5
Cl^- (p)	0.5	0.4	0.4	0.5	0.3	0.6
NO_3^- (p)	1.3	1.2	1.2	1.2	2.0	1.3
SO_4^{2-} (p)	3.6	3.3	3.0	3.4	4.1	2.6
NH_3 (g)	35.0	22.0	22.3	17.7	14.8	10.2
HNO_3 (g)	2.9	2.5	4.0	2.8	2.8	2.5
SO_2 (g)	1.4	1.4	1.6	1.3	1.9	2.1

the MDL was calculated to be 6.5 nmol m^{-3} for a 2 week sample. Details regarding the calculation of atmospheric concentrations and the comparison to URG samplers can be found in the supporting information (Figure S3).

2.3. Sample Analysis

[15] Aqueous denuder extracts, filter extracts, and precipitation samples were analyzed for both cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) and anions (Cl^-, NO_3^-, NO_2^-, and SO_4^{2-}) by ion chromatography. Denuder extracts were analyzed either for ammonium (phosphorous acid-coated denuders) or for sulfate and nitrate (carbonate-coated denuders). Cartridge extracts from the passive samples were only analyzed for ammonium. Cations were separated with 20 mM methanesulfonic acid eluent on a Dionex CS12A column followed by a CSRS ULTRA II suppressor and a Dionex CD-20 conductivity detector. Anions were separated with a 1.8 mM sodium carbonate/1.7 mM sodium bicarbonate eluent on a Dionex AS14A column followed by an ASRS ULTRA II suppressor and a Dionex CD-20 conductivity detector. Each ion chromatograph was calibrated daily using standards prepared from analytical grade salts.

[16] Total nitrogen in the precipitation samples was measured using a Shimadzu TOC V_{CSH} + TNM-1, a total organic carbon analyzer with a total nitrogen module. This instrument performs high-temperature oxidation, converting all reactive

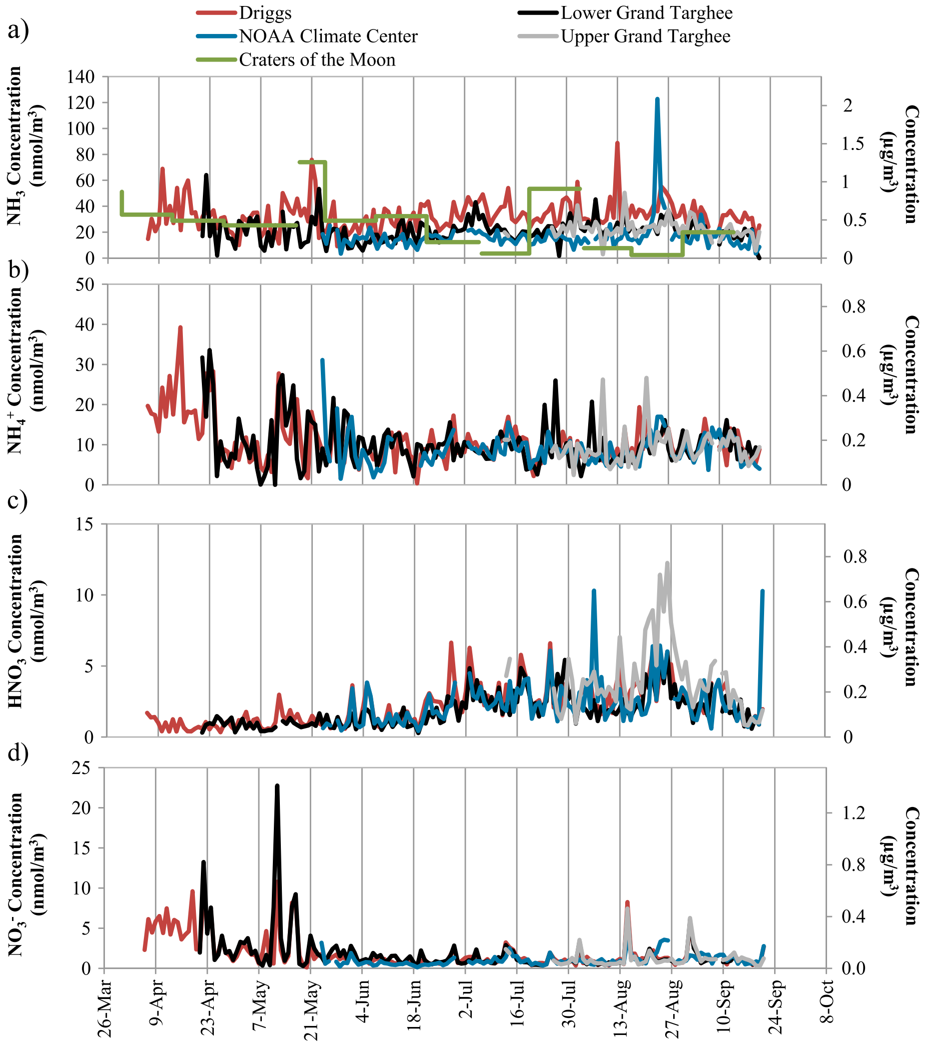

Figure 3. Twenty-four hour concentrations (adjusted to standard pressure) of (a) NH_3, (b) HNO_3, (c) NH_4^+, and (d) NO_3^- at all of the sampling sites. Two week ammonia concentrations from the AMoN network site located in Craters of the Moon National Park are also included in Figure 3a.

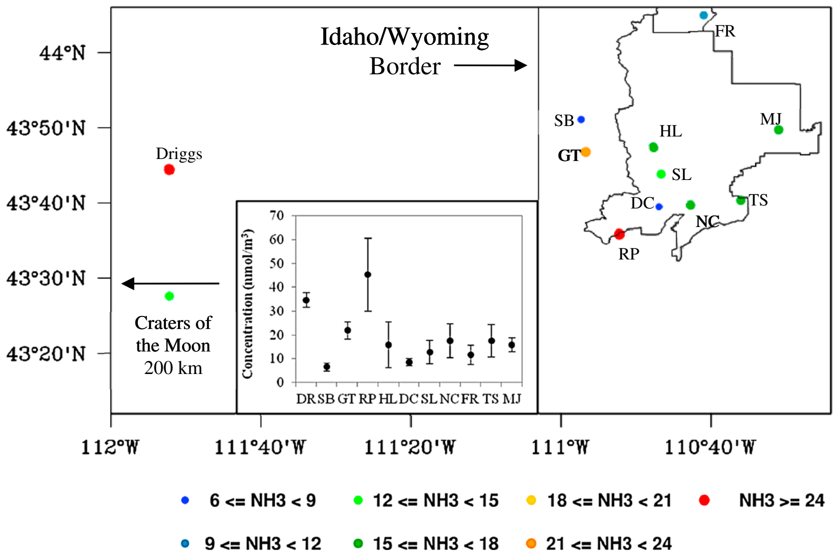

Figure 4. Spatial distribution of NH_3 (nmol m^{-3}) from July to September 2011 from biweekly passive samplers. Concentrations are adjusted to standard pressure. Data from the AMoN Craters of the Moon sites are from weekly passive samplers. This site is plotted below Driggs even though it is ~200 km west of Driggs. The color of each circle represents the average concentration at each sampling site. In the inset, sites are plotted according to longitude, west to east, and the error bars represent the standard deviation of concentrations during the study period at each site.

nitrogen in the sample to NO at 720°C and then detects NO by chemiluminescence. Measurement of organic nitrogen by high-temperature oxidation has been used previously to determine organic nitrogen concentrations in precipitation and aerosol samples [*Rastogi et al.*, 2011; *Keene et al.*, 2002; *Kieber et al.*, 2005; *Gioda et al.*, 2008; *Lin et al.*, 2010]. A stock solution of 1000 mg N L^{-1} was made using sodium nitrate and calibration and check standards were diluted to the appropriate levels from 0.5 mg N L^{-1} to 32 mg N L^{-1} prior to each analysis. Concentrations were determined from three replicate injections that were automatically performed by the instrument. Due to the replicate injections, a minimum of 15 mL is needed for analysis. Water-soluble organic nitrogen was determined from the difference of total nitrogen and inorganic nitrogen (NH_4^++NO_3^-) as measured by ion chromatography. Nitrite (NO_2^-) was less than 1% of total nitrogen measured in all samples and was not detected in the majority of samples. It is excluded from the discussion here.

3. Atmospheric Concentrations

3.1. Particle Composition

[17] Inorganic particle composition was measured at four of the GrandTReNDS sampling sites. Averages for the period when all sites were operating (13 July to 21 September 2011) are shown in Table 2. The average molar concentrations of NH_4^+ were largest, typically followed by SO_4^{2-}, Ca^{2+}, and NO_3^-. The remaining measured inorganic ion species concentrations all averaged less than 1 nmol m^{-3}. Timelines of NO_3^- and NH_4^+ concentrations are shown in Figure 3 (along with the gas-phase nitrogen species NH_3 and HNO_3). Concentrations of both $PM_{2.5}$ NH_4^+ and NO_3^- were low throughout the sampling period and similar at the four GrandTReNDS sites suggesting relatively uniform concentrations over the region, even from the west to the east side of the Teton Range. There was a slight decrease in concentrations of both NO_3^- and NH_4^+ at the end of May at Driggs and Lower Grand Targhee. An increase in HNO_3 and a simultaneous decrease in NO_3^- concentrations were evident during June, suggesting a shift in the gas-particle partitioning of ammonium nitrate, as expected at warmer temperatures, toward the gas phase.

[18] Daily or seasonal variations in the concentrations of the other inorganic species are not shown. For several species (SO_4^{2-}, Mg^{2+}, Ca^{2+}), there were changes in concentration from day to day, but they tended to be centered about the average with no clear episodes or trends, especially at NOAA Climate Center and Upper Grand Targhee. Only at Driggs did we observe a period of higher SO_4^{2-} concentrations in April which decreased into May (not shown). Elevated concentrations of Mg^{2+} and Ca^{2+} at Driggs indicated summer dust episodes beginning in July, where concentrations of both Mg^{2+} and Ca^{2+} would increase for several days and then decrease, repeating this pattern through the end of August. Concentrations of $PM_{2.5}$ Mg^{2+} peaked at 5.7 nmol m^{-3} and Ca^{2+} peaked at 28.6 nmol m^{-3} between 17 and 25 August.

[19] Increases in K^+ concentrations, a known biomass burning tracer, were also observed on several occasions throughout the study, indicating the influence of smoke in the region [*Andreae*, 1983]. At the NOAA Climate Center site, K^+ reached 6.2 nmol m^{-3}, and concentrations remained slightly higher in September compared to the first part of the study. Increases in K^+ concentrations were also observed at Upper Grand Targhee from 15 to 19 August peaking at 7.2 nmol m^{-3} and from 31 August to 4 September peaking at 5.4 nmol m^{-3}. At Driggs, more episodes of high K^+ were observed, but many of these episodes had overall smaller K^+ concentrations. These episodes occurred on 2 July (4.6 nmol m^{-3}), 4 July (3.4 nmol m^{-3}), 15–19 August

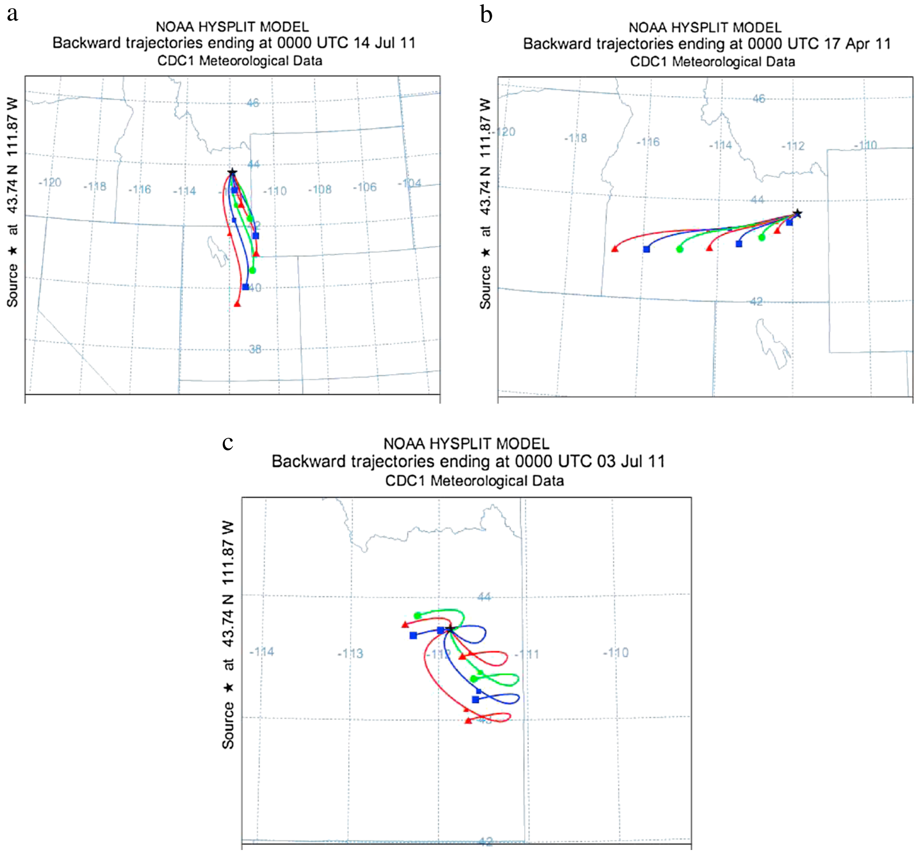

Figure 5. Example HYSPLIT back trajectories from days when high NH_3 concentrations (the 10% highest observed values) were observed at DR. Characteristic transport patterns during these periods were (a) transport from the south, (b) transport along the Snake River Valley, and (c) stagnation.

(9.2 nmol m^{-3}), and 29 August to 4 September (4.3 nmol m^{-3}). Several of these episodes overlap at the three sites indicating a regional smoke influence crossing the Teton Mountain Range.

[20] Average concentrations from measurements made in RMNP over a similar time period in 2009 [*Benedict et al.*, 2012] and 2010 are also included in Table 2. RMNP NH_4^+ and NO_3^- concentrations in 2009 were higher than GrandTreNDS observations, while RMNP 2010 concentrations were similar.

3.2. Ammonia

[21] The data from the passive ammonia sampling network provide higher spatial resolution and cover a larger area to better reveal how average NH_3 concentrations vary across the region. In Figure 4, we can see that there is a general decrease in ammonia concentrations from Driggs to the east. The observations of ammonia concentrations are consistent with higher emissions to the west of GTNP and westerly winds. Ammonia concentrations at the Craters of the Moon AMoN site (located north of the Snake River Valley approximately 1–2 km above the valley floor) averaged about half of the concentrations measured at Driggs. Biweekly average concentrations from the NADP Ammonia Monitoring Network (AMoN) site at Craters of the Moon, located approximately 200 km west of Driggs and north of the Snake River Valley, were similar to those measured at Driggs and our other sampling sites during the first part of the study. From mid-June through September, however, concentrations at the AMoN site were almost always lower than those measured at the GrandTReNDS sites.

[22] Two passive ammonia sites do not follow the general trend of ammonia decreasing eastward from Driggs: RP and SB. RP (Rendezvous Point), which had the highest average ammonia concentration (45 nmol·m^{-3}), is located at the top of Jackson Hole Ski Area, near the top of the tram which operates in the summer to carry hikers and tourists to the top of the mountain. A building at the top has restrooms and sells food and souvenirs. These activities likely elevated local ammonia concentrations, a hypothesis supported by the low ammonia concentrations measured at Surprise Lake (SL), which is at a similar elevation to RP. South Badger (SB), which had the lowest average ammonia concentration at 8.8 nmol m^{-3}, is located on the western slope of the Tetons in the national forest. It is located in a small isolated valley. Limited exchange of regional air into the valley and rapid ammonia deposition are hypothesized to reduce average ammonia concentrations locally.

[23] Daily NH_3 concentrations at the URG sites were fairly stable with only 13 days where concentrations increased to 50% above the average (Figure 3a). Average ammonia concentrations during the common measurement period from 13 July to 21 September 2011 were 34.0 nmol m^{-3} at Driggs, 21.1 nmol m^{-3} at Lower Grand Targhee, 22.2 nmol m^{-3} at Upper Grand Targhee, and 17.7 nmol m^{-3} at NOAA Climate Center. Ammonia concentrations at Driggs, Upper Grand Targhee, and NOAA Climate Center were all statistically

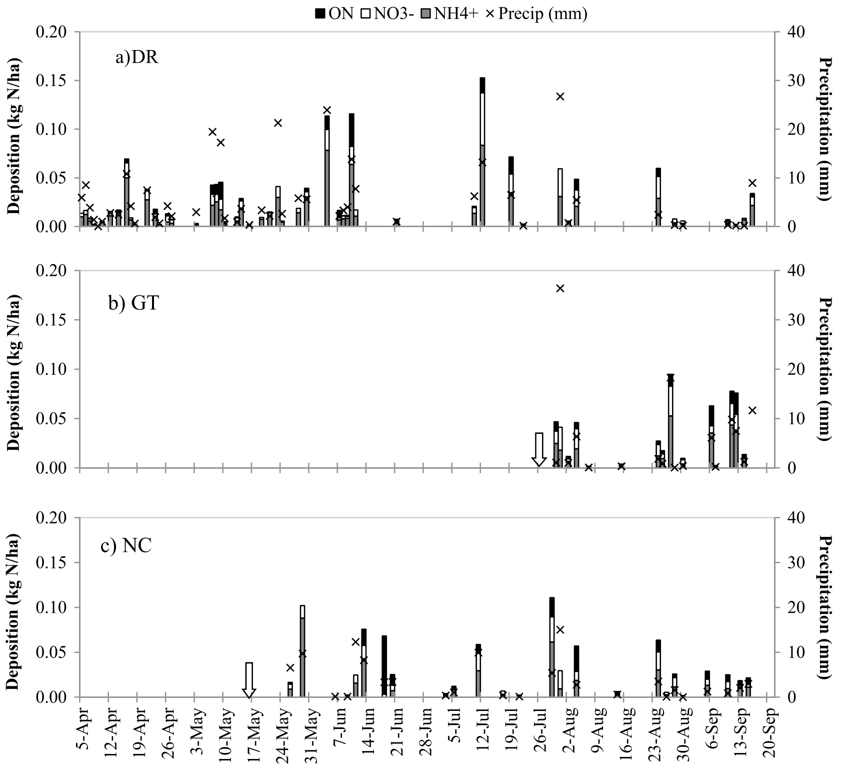

Figure 6. Timelines for wet nitrogen deposition (kg N ha^{-1}) throughout the study period at (a) Driggs (DR), (b) Upper Grand Targhee (GT), and (c) NOAA Climate Center (NC). Precipitation amount in millimeters is also plotted as determined from the volume of precipitation collected in the bucket. The arrows indicate when sampling began at GT (Figure 6b) and NC (Figure 6c).

different at the 95% confidence level. There was no significant difference between Upper and Lower Grand Targhee ammonia concentrations, not a surprising result given the close proximity of these two sites, although they are 270 m apart in elevation. Many of the periods of higher NH_3 concentrations occurred in August at the four sites. While the average ammonia concentrations were lowest at NOAA Climate Center, the maximum daily NH_3 concentration (122.7 nmol m^{-3}) during the study was measured there, indicating periods of higher concentration that occur and may be important for understanding conditions when pollutants are transported into the park.

[24] Back trajectories from the study period were examined to see if days with higher NH_3 concentrations had different transport patterns compared to other days during the study. Twenty-four hour back trajectories were run using NOAA HYSPLIT, the Hybrid Single Particle Lagrangian Integrated Trajectory Model (http://ready.arl.noaa.gov/HYSPLIT.php). The model was run every 3 h using NCEP/NCAR reanalysis with trajectories beginning at Driggs at 50 m above ground level. Days with NH_3 concentrations in the highest 10% of all values measured at Driggs were considered "high" NH_3 days. There are several common scenarios for the high NH_3 concentration days observed at Driggs: stagnation, transport from Utah, and parcel movement along the Snake River Valley (Figure 5). On other sampling days with lower ammonia concentrations, trajectories were more likely to arrive from a more northerly direction, from the southeast, or even from the west but not in the heart of the Snake River Valley.

3.3. Nitric Acid

[25] Nitric acid concentrations were similar across the GrandTReNDS sites, with averages for the 13 July to 21 September 2011 common measurement period of 2.9 nmol m^{-3} at Driggs, 2.5 nmol m^{-3} at Lower Grand Targhee, 4.0 nmol m^{-3} at Upper Grand Targhee, and 2.8 nmol m^{-3} at NOAA Climate Center. The highest measured daily HNO_3 concentration was measured at Upper Grand Targhee (12.2 nmol m^{-3}). The longer time records, available at Driggs and Lower Grand Targhee, reveal low nitric acid concentrations in spring, increasing concentrations during summer, and then a drop in concentrations moving into September (Figure 3b).

[26] During the latter part of August, nitric acid concentrations were more than 50% higher at Upper Grand Targhee compared to any other site (Figure 3b). From 19 August to 2 September, concentrations were at least two times higher than the rest of the sampling period. During these periods, high HNO_3 concentrations were not observed at Driggs, the NOAA Climate Center, or the lower elevation Grand Targhee base site, suggesting enriched nitric acid transport aloft. Back trajectories from these time periods indicate that flow was mainly from the east or southeast and likely was lofted over the mountains (Figure S4). On several occasions during GrandTReNDS, air masses showed signs of smoke (elevated K^+) at Upper Grand Targhee; however, the high concentrations of K^+ and HNO_3 did not coincide in August, suggesting a nonbiomass burning source of HNO_3 aloft.

[27] Average nitric acid concentrations were similar to RMNP observations at Driggs, Lower Grand Targhee, and the NOAA Climate Center, while at Upper Grand Targhee,

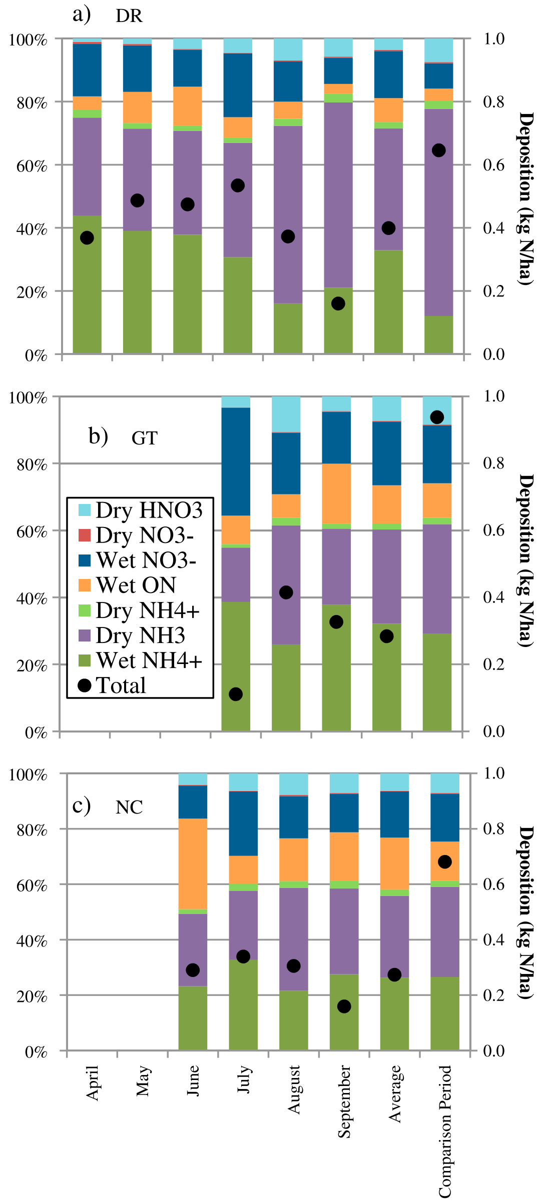

Figure 7. The monthly contributions of each nitrogen deposition pathway to total nitrogen deposition at (a) DR, (b) GT, and (c) NC. The study average for each site and the overlapping comparison period are also included.

they were much higher (approximately 40%). In general, the similar concentrations of HNO_3, NO_3^-, and $NH_4^+$and higher concentration of NH_3 in GTNP compared to RMNP may indicate important differences in the types of sources and reactive nitrogen deposition pathways impacting these two national park sites in the Rocky Mountain region.

4. Nitrogen Deposition

4.1. Precipitation Chemistry and Wet Deposition

[28] Daily precipitation samples were collected at Driggs, Upper Grand Targhee, and NOAA Climate Center. Across all sites, the precipitation pH ranged from a minimum of 4.76 (Upper Grand Targhee, 4 August 2011) to a maximum of 6.88 (Driggs, 14 May 2011) with volume-weighted average pH values of 5.42, 5.12, and 5.19, at Driggs, Upper Grand Targhee, and NOAA Climate Center, respectively. The range of pH reflects the combined influence of basic species and acidic species that contribute to precipitation chemistry. The average pH, which is somewhat lower than expected from equilibrium with atmospheric CO_2, is also influenced by scavenging of aerosol particles and other soluble gases, including ammonia and nitric acid. Higher pH values at Driggs are consistent with higher ammonia concentrations measured there.

[29] In June and July, precipitation at Driggs (~54 mm each month) was greater than at NOAA Climate Center (~30 mm), consistent with their locations upwind and downwind of the Teton Range, respectively. Wet nitrogen deposition was also greater at Driggs. In August and September, monthly precipitation was less than 10 mm at both Driggs and NOAA Climate Center, while at the higher elevation site, Upper Grand Targhee, 29 mm of precipitation fell in August and 36 mm fell in September. The highest average volume-weighted ion concentrations were of ammonium, calcium, nitrate, and sulfate at all three sites. At both Driggs and NOAA Climate Center, precipitation ion concentrations (μN) decreased in the order $NH_4^+ > NO_3^- > Ca^{2+} > SO_4^{2-}$, while at Upper Grand Targhee, the order was $NH_4^+ > Ca^{2+} > NO_3^- > SO_4^{2-}$. Nitrogen species were always more abundant than sulfur, on a molar basis, in the precipitation samples.

[30] In Figure 6, daily wet nitrogen deposition and precipitation amounts are plotted for each of the three sites in order to illustrate the frequency and amount of deposition per event. At Driggs, daily precipitation events occurred during the beginning of the study and began to decrease in frequency in June. Throughout the summer, at all three sites, there was little precipitation, leading to low amounts of wet nitrogen deposition. There were a few large precipitation events; for instance, on 31 July precipitation occurred at all three sites with between 20 and 38 mm of rain. The bars in Figure 6 represent the amount of nitrogen deposition by each pathway for the event. In general, ammonium was the largest contributor to wet deposition at each site, while the relative contributions of nitrate and organic nitrogen vary. Occasionally, organic nitrogen contributed more than ammonium.

4.2. Nitrogen Deposition Budget for GTNP

[31] As discussed above, wet deposition was determined from the amount of collected precipitation and the aqueous concentration for each sample. Dry deposition was estimated as a deposition velocity multiplied by the atmospheric concentration. Previously, in constructing deposition budgets at RMNP, deposition velocities were calculated based on observations from a colocated CASTNet station [*Beem et al.*, 2010]; however, the closest CASTNet monitoring site for GrandTReNDS measurements is in Pinedale, WY, 110 km SE of the NOAA Climate Center sampling site, and the next closest CASTNet site is located 140 km north in Yellowstone National Park. CASTNet calculates dry deposition with the NOAA Multilayer Deposition Velocity Model (MLM) described by *Meyers et al.* [1998] using meteorological and site conditions, including vegetative cover. To estimate representative average monthly deposition velocities for GTNP, the weekly HNO_3 deposition velocities from 2000–2009 for Pinedale and Yellowstone were averaged. Even though these sites are 225 km apart, 476 m different in altitude, and have

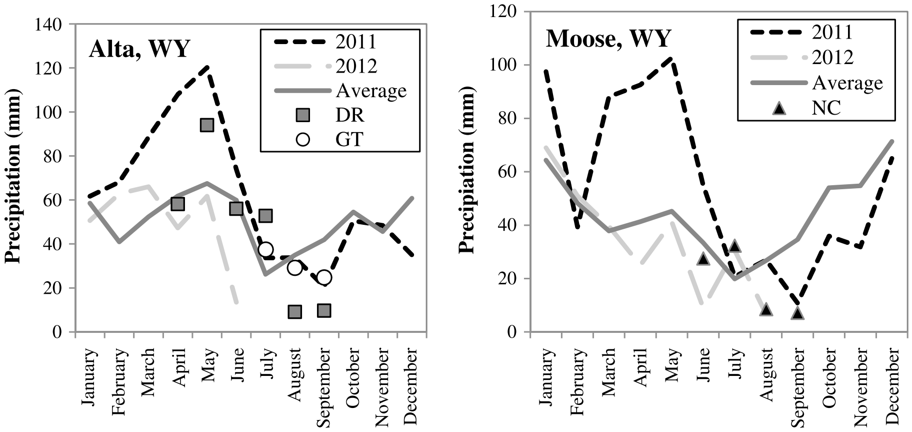

Figure 8. Monthly precipitation from the National Climatic Data Center for 2011, 2012 and the average 2000–2012 for Alta, WY located between DR and GT, and Moose, WY near our NC sampling site. The precipitation totals from each of the sampling sites, based on the bucket volume, are also plotted.

different vegetation, the deposition velocities were similar. Such agreement suggests that a regional deposition velocity may be used to estimate dry deposition in GTNP (Figure S5). The MLM does not calculate deposition velocities for NH_3. To estimate deposition of NH_3, 70% of the nitric acid deposition velocity was used following our earlier approach published for RMNP [*Beem et al.*, 2010; *Benedict et al.*, 2013].

[32] In Figure 7, the percent contributions of each reactive nitrogen deposition pathway are shown for April to September at Driggs, Upper Grand Targhee, and NOAA Climate Center. Dry deposition of NH_3 and wet deposition of NH_4^+, NO_3^-, and ON together accounted for more than 90% of total quantified nitrogen deposition at the three intensive GrandTReNDS sites. At Driggs, to the west of GTNP, dry deposition of NH_3 was largest (39%) followed by wet deposition of NH_4^+ (33%), wet deposition of NO_3^- (15%), and wet deposition of organic nitrogen (8%). At NOAA Climate Center, on the eastern side of GTNP, dry deposition of NH_3 was also the largest deposition pathway (29%), followed by wet deposition of NH_4^+ (26%), wet deposition of organic nitrogen (18%), and wet deposition of NO_3^- (17%). The relative importance of the deposition pathway at Upper Grand Targhee was slightly different; wet deposition of NH_4^+ was most important (32%) followed by dry deposition of NH_3 (28%), wet deposition of NO_3^- (19%) and wet deposition of organic nitrogen (11%). Low atmospheric nitric acid concentrations resulted in its low contributions to nitrogen deposition at all three sites. $PM_{2.5}$ nitrate and ammonium deposition contributions were low because of low fine particle deposition velocities. The similarity in deposition amount for ammonium wet deposition and ammonia dry deposition contrasts with results from other studies and mountain locations where wet deposition typically is greater than dry deposition [*Burns*, 2003; *Beem et al.*, 2010].

[33] The relative contributions of reduced, oxidized, and organic nitrogen deposition changed across the three sites. At Driggs, deposition of reduced nitrogen (wet NH_4^+, dry NH_3, and dry NH_4^+) contributed on average 73% to total quantified nitrogen deposition, while at Upper Grand Targhee and NOAA Climate Center, reduced nitrogen contributed 62% and 58%, respectively. This trend is consistent with the location of the largest ammonia sources west of the study region. While large NO_x sources are located to the east of the study region, the spatial trend in oxidized nitrogen deposition contributions was not as clear as for reduced nitrogen. The oxidized nitrogen deposition contribution was substantially higher at NOAA Climate Center (23%) than at Driggs (11%), but the Upper Grand Targhee contribution (27%) slightly exceeded the value at the NOAA Climate Center. The contribution of wet organic nitrogen deposition to total quantified nitrogen deposition increased from west to east, contributing 8%, 11%, and 18% at Driggs, Upper GrandTarghee, and NOAA Climate Center. Given the dominance of reduced nitrogen to the total nitrogen deposition budget, of course, spatial trends in relative contributions of the other forms of nitrogen partly reflect the spatial trend in reduced nitrogen deposition. NH_3 sources dominate the spatial trend of nitrogen deposition and atmospheric concentrations even on the east side of the Teton Range.

[34] Monthly/seasonal changes in the relative importance of the deposition pathways corresponded to changes in precipitation amount. The example at Driggs is particularly striking (Figure 7a). At Driggs, a larger fraction of wet deposition (~40%) was observed in early spring when there was more precipitation, while dry deposition of ammonia was more important later in the study period, corresponding to a decrease in precipitation (Figure 8). The absolute amount of precipitation in 2011 was higher than the historical average during the wet period, indicating that dry deposition of NH_3 may typically be a more important contributor also to spring nitrogen deposition than that observed during GrandTReNDS.

[35] Precipitation scavenges both gases and particles from the atmosphere leading to reduced atmospheric concentrations. The GrandTReNDS observations of NH_3 concentrations and precipitation in 2011 combined with subsequent observations from 2012 suggest that we may be seeing this interaction. Passive ammonia sampling has continued at Driggs since the end of GrandTReNDS to more fully assess the full seasonal cycle of ammonia concentrations and interannual variability. In the spring and summer of 2012, NH_3 concentrations were almost twice the level measured in 2011 (Figure 9). Precipitation during 2012 was lower than in 2011 and was more similar to the 10 year average (Figure 8). While more years of observations are needed to better understand the seasonal trends in

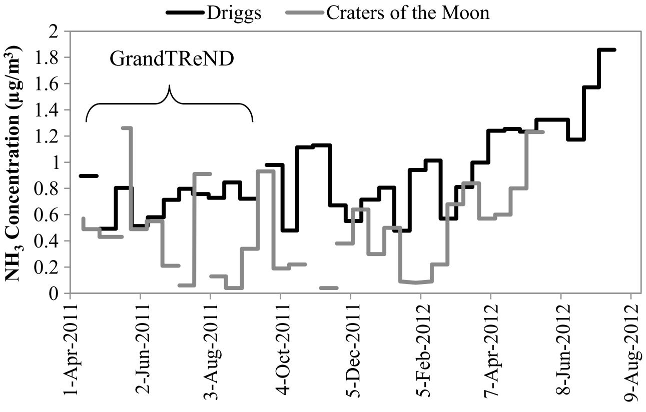

Figure 9. Ammonia concentrations measured in Driggs, ID using passive samplers from April 2011 to July 2012 along with ammonia concentrations measured at the Craters of the Moon AMoN site.

ammonia concentration in this region, the 2012 data suggest that dry deposition may typically be more important than what the GrandTReNDS results indicate.

[36] Dry deposition of particulate organic nitrogen (PON) was also determined using concentrations of water-soluble PM$_{2.5}$ organic nitrogen measured from weekly high-volume filters at Upper Grand Targhee. Weekly dry deposition of PON ranged from 0.004 to 0.01 kg N/ha accounting for approximately 0.8% of total measured nitrogen deposition at this site. In comparison, dry deposition of NH_4^+ was 1.8% of the total measured nitrogen deposition at the site and dry deposition of NO_3^- accounted for 0.2%. Contributions from water insoluble PON might increase the importance of PON somewhat, but low deposition velocities for fine particles will limit the relative size of this nitrogen deposition pathway. Dry deposition of organic nitrogen gases was not included in these deposition budgets. However, the high wet deposited organic nitrogen and low concentrations of soluble particulate organic nitrogen suggest that dry deposition of organic nitrogen gases could be an important nitrogen deposition pathway.

[37] Most important for understanding the impacts on the ecosystems and the relationship between deposition totals and critical load is the deposition flux. In Table 3, the flux of nitrogen per week was calculated from the deposition totals for each site. The standard errors, propagated from the analytical precision and uncertainties in each measurement are included for the weekly deposition rates and the study totals. Calculated errors were similar between sites; the error given for each individual pathway is the maximum standard error for that pathway across all sites. Included in this table is dry deposition of particulate organic nitrogen (PON), which was only sampled at Upper Grand Targhee, as well as deposition fluxes from RMNP from 2009 and 2010. Data from RMNP are included to provide a comparative measure of deposition since RMNP has been more extensively studied. While included in Table 3, Driggs is excluded in the following discussion since it is not representative of GTNP due to its proximity to ammonia source regions. Total measured nitrogen deposition rates were similar in GTNP and at the RMNP site. The deposition pathways had different relative contributions reflecting lower particulate and nitric acid concentrations and higher ammonia concentrations near GTNP. Upper Grand Targhee had the highest total measured nitrogen deposition for the period when all sites were operating. Upper Grand Targhee is just upwind of GTNP, suggesting that high-elevation lakes on the western side of GTNP may have similarly high deposition rates. Dry deposition rates tended to be similar at GTNP and RMNP for all pathways except dry deposition of NH_3 which tended to be larger at GTNP. The weekly deposition averages were similar at Driggs, NOAA Climate Center, and RMNP; only Upper Grand Targhee was different, likely reflecting the shorter operational period, the influence of the high nitric acid event on deposition, and higher precipitation amounts at the site in August and September compared to the other sites.

[38] To obtain a more complete annual nitrogen deposition budget estimate for GTNP, data from Rocky Mountain Regional Snowpack Chemistry Monitoring Study (http://co.water.usgs.gov/projects/RM_snowpack/index.html) from the 2010–2011 winter sampling were used. Deposition amounts from several sampling sites in the region, Garnet Canyon, Rendezvous Mountain, and Lewis Lake Divide, were averaged together to estimate winter inorganic nitrogen deposition. Average inorganic nitrogen deposition from these three sites, which includes both wet deposition and dry deposition accumulation in the winter snowpack, was 1.25 kg N ha^{-1}. Adding this amount to the NOAA Climate Center deposition budget from GrandTReNDS, we can better represent a full year of GTNP nitrogen deposition; though this will miss

Table 3. Total Deposition Flux Per Week by Pathway for Each Site Where Wet Deposition Samples Were Collected (kg N/ha/week)[a]

		DR	GT	NC	RMNP 2009	RMNP 2010
Dry NH_3 (±0.009)		0.032	0.024	0.017	0.014	0.0022
Wet NH_4^+ (±0.004)		0.012	0.027	0.018	0.023	0.026
Wet ON (±0.001)		0.003	0.0096	0.0095	0.009	0.0038
Wet NO_3^- (±0.003)		0.009	0.016	0.012	0.013	0.023
Dry HNO_3 (±0.002)		0.004	0.0061	0.0037	0.0041	0.0035
Dry NH_4^+ (±0.005)		0.0014	0.0015	0.0013	0.0018	0.0013
Dry PON (±0.0002)			0.0007		0.0008	0.0005
Dry NO_3^- (±0.00007)		0.0002	0.0002	0.0002	0.0003	0.0002
Total N Deposition per week		0.061	0.085	0.061	0.065	0.061
	6 April to 21Sept.	2.38 ± 0.2			2.34 ± 0.18	2.12 ± 0.16
Total N Deposition	15 May to 21 Sept.	1.77 ± 0.16		1.23 ± 0.1	1.68 ± 0.12	1.48 ± 0.11
	24 July to 21 Sept.	0.62 ± 0.08	0.90 ± 0.07	0.62 ± 0.06	0.65 ± 0.06	0.61 ± 0.05

[a] Deposition flux at RMNP was calculated from both the 2009 and 2010 data sets for the same dates of operation as the three GTNP sites (24 July to 21 September). Uncertainties are presented as ±1 standard deviation based on propagation of measurement errors.

nitrogen deposition that occurred in the fall before snowfall began and in the spring after the snowpack sampling and before measurements started. In addition, the organic nitrogen in the snow pack was not measured. Given these caveats, the annual GTNP nitrogen deposition at the NOAA Climate Center totals approximately 2.5 kg N ha^{-1} from October 2010 to September 2011. Adding the winter snowpack data to the Upper Grand Targhee budget yields total nitrogen deposition of 2.2 kg N ha^{-1}, but a larger portion of the year is missing measurements. These lower bound estimates substantially exceed the critical load established for GTNP of 1.4 kg N ha^{-1}, although that load is defined based solely on wet deposition of inorganic nitrogen. If we consider just the wet deposition at Upper Grand Targhee plus the winter snowpack data (which includes some dry nitrogen input), we estimate wet inorganic nitrogen deposition for the year of 1.7 kg N ha^{-1}. This value does not include wet inputs during the spring and fall which were not captured in either the GrandTReNDS or snowpack sampling data sets.

5. Conclusions

[39] The observations presented and discussed here provide new key information about the atmospheric concentrations of nitrogen species in and around GTNP and their contributions to reactive nitrogen deposition. During GrandTReNDS, ammonia was the most abundant of the measured reactive atmospheric nitrogen species measured, followed by nitric acid. Ammonia concentrations generally decreased from west of GTNP moving eastward. Ammonia concentrations were typically higher at GTNP than that observed previously at RMNP to the south, likely reflecting the location of large ammonia sources upwind of GTNP. Concentrations of the other nitrogen species were generally similar between GTNP and RMNP, except for somewhat elevated nitric acid concentrations at the high-elevation Grand Targhee site.

[40] Reduced nitrogen was the most important form of reactive nitrogen in both atmospheric and precipitation samples. Deposition of reduced nitrogen (wet + dry) contributed the most to total measured deposition at all three GrandTReNDS deposition sites, both west and east of the Teton Mountain Range. The dominance of reduced nitrogen was somewhat surprising on the eastern side of the park which is closer to NO_x sources in western Wyoming and farther from agricultural ammonia sources in Idaho. The importance of reduced nitrogen in the region indicates that we need to be concerned about continued increases in NH_3 emissions near GTNP and likely across the country.

[41] These measurements highlight the need for increased monitoring of ammonia which has traditionally been left out of national ambient air quality and deposition monitoring networks. The recent additions of AMoN NH_3 and IMPROVE NH_x ($NH_3 + NH_4^+$) measurements help to address one missing piece of the nitrogen deposition budgets from national monitoring sites. More measurements and research into ammonia deposition are also needed. The importance of wet organic nitrogen deposition in GrandTReNDS also points to the need to better constrain this often overlooked deposition pathway. Wet deposition of organic nitrogen is not addressed easily because of the difficulties in making high-quality measurements including issues concerning sample stability and contamination [*Cornell and Jickells*, 1999; *Hill et al.*, 2005; *Walker et al.*, 2012]. Additionally, dry deposition of gas-phase organic nitrogen was not included and could potentially be an important deposition pathway.

[42] Elevated nitric acid concentrations and dry nitric acid deposition were observed at the high-elevation Upper Grand Targhee measurement site just outside the GTNP western boundary. Since nitric acid is a secondary pollutant, it is likely that the high concentrations observed were the result of long-range transport and not local emissions from the ski resort. Further investigation of this phenomenon is important, since long-range transport of reactive nitrogen pollutants at altitude could be an important—and likely overlooked—contribution to nitrogen deposition to other remote, high altitude, sensitive ecosystems as well.

[43] Additional work remains to be done to examine this rich data set in more detail, along with an examination of regional transport patterns and source regions in this area. A combined analysis of these pieces of information (e.g., as done for RMNP by *Gebhart et al.* [2011]), is expected to provide considerable insight into the contributions of different source types and regions to reactive nitrogen deposition in GTNP. The limited transport analysis for high concentration periods conducted here, along with observed spatial gradients in ammonia concentrations, indicate that high emissions of ammonia in the Snake River Valley west of GTNP are likely important contributors to reduced nitrogen deposition in GTNP. The transport analysis also suggests that regions of northern Utah may also be important contributors to elevated ammonia concentrations and deposition in GTNP and the surrounding region.

[44] It is important to note that while the critical loads for Rocky Mountain and Grand Teton National Park are set at 1.5 and 1.4 kg N $ha^{-1} yr^{-1}$, respectively [*Baron*, 2006; *Saros et al.*, 2010], many areas experience much higher levels of nitrogen deposition and higher critical loads have been determined in other locations. For instance, the median critical load for all of Western Europe is 14 kg N $ha^{-1} yr^{-1}$ [*Reinds et al.*, 2008], and a recent study examines the deposition gradient across Europe, where deposition ranges from 2–44 kg N $ha^{-1} yr^{-1}$ [*Stevens et al.*, 2010]. A critical load is dependent on a host of factors including soil, vegetation, climate, and land use history which results in the low critical loads for remote mountainous areas in the U.S. In the United States, critical loads have been set at the point where there is scientific evidence for negative ecosystem effects and protecting natural ecosystems is the first priority, which has not always been the case in other countries. Critical loads across the U.S. vary from 1–39 kg N $ha^{-1} yr^{-1}$ depending on location and the affected vegetation [*Pardo et al.*, 2011].

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