#### **THESIS**

# TRANSPORT OF POLLUTANTS FROM EASTERN COLORADO INTO THE ROCKY MOUNTAINS VIA UPSLOPE WINDS

Submitted by

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In partial fulfillment of the requirements For the Degree of Master of Science Colorado State University Fort Collins, Colorado Fall 2013

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#### ABSTRACT

# TRANSPORT OF POLLUTANTS FROM EASTERN COLORADO INTO THE ROCKY MOUNTAINS VIA UPSLOPE WINDS

The confluence of mountain meteorology and major pollution sources come together to transport pollutants across the Front Range, especially nitrogen species (NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, orgN, NO<sub>3</sub><sup>-</sup>, and HNO<sub>3</sub>) from agricultural and urban regions, into the Rocky Mountains. The focus of this study was to examine the meteorological conditions in which atmospheric wet deposition of inorganic nitrogen in the Rocky Mountains was anomalously high. We analyzed 19 years (1994-2013) of precipitation and concentrations of wet inorganic nitrogen data from three National Atmospheric Deposition Program (NAPD) sites in the Rocky Mountains: Beaver Meadows (CO19), Loch Vale (CO98), and Niwot Ridge (CO02). Beaver Meadows (2477 m), Loch Vale (3159 m), and Niwot Ridge (3520 m) are all within 40 km but differ in elevation, resulting in different seasonal precipitation composition and totals. The North American Regional Reanalysis (NARR) was used to observe synoptic conditions that influenced two high wet deposition events from August 18-20, 2006 and July 6-8, 2012. Interestingly, anti-cyclones in southern Canada and high precipitable water values associated with monsoonal flow played significant roles in initiating convection that caused high values of wet deposition of inorganic nitrogen in the Rocky Mountains. The Advanced Research WRF model was then used to simulate the meteorology at a high spatial and temporal resolution for the two time periods to examine the contribution of cloud-scale convection to wet nitrogen deposition in the Rocky Mountains. A mesoscale mountain circulation caused by differential heating between mountains slopes and the

plains was the main driver of the slow westward transport towards the mountains while cloud-scale convection contributed greatly to the transport of nitrogen along the Colorado Front Range.

#### **ACKNOWLEDGEMENTS**

I give many thanks to Matt Bishop for software installation and computing help—this project could not have been completed without him. Thanks to Cameron Homeyer for WRF tracer code and Gus Alaka for help with post-processing of WRF output. National Centers for Environmental Prediction (NCEP) NARR was provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website at http://www.esrl.noaa.gov/psd/. Stage IV radar data was provided by NCAR/EOL under sponsorship of the National Science Foundation. http://data.eol.ucar.edu/.

I want to think the I-WATER program for their support during my time in graduate school.

I am extremely grateful for the guidance and mentoring from my advisor, Dr. A. Scott Denning. Additionally, I would like to thank my committee members, Drs. Russ Schumacher and Jay Ham, for great feedback throughout this project.

Lastly, I want to thank friends, mentors, and family for their continuous support during my time away from them.

This research was supported by the National Science Foundation under Grant No. 0966346 and the National Park Service under award P13AC00599, project number CSURM-273.

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#### 1. Introduction

Nitrogen (N) is a limiting nutrient for aquatic and terrestrial ecosystems in Colorado's Rocky Mountains (Morris and Lewis, 2006). From the continual addition of pollutants into the Rocky Mountains, such as transported N from Colorado's Front Range, alpine ecosystems are very susceptible to change (Vitousek et al., 1997). Generally, alpine watersheds in the Rocky Mountains are upwind of pollutant sources East of the Colorado Front Range, but the combination of diurnal mountain circulation, monsoonal moisture flow, and synoptic circulation patterns can bring together these two unrelated landscapes into biogeochemical contact. With 83% bare rock, ice, and boulder, 11% tundra, 5% forest, and 1% subalpine meadow (Arthur, 1992), the sparse vegetation in Loch Vale Watershed leaves alpine lakes vulnerable to accumulating pollutants from runoffs streams that were never taken up by coarse-textured soils (Swackhamer et al., 2004; Walthall, 1985). Hence, most deposited ions end up in runoff streams into mountain lakes shown in Figure 1.

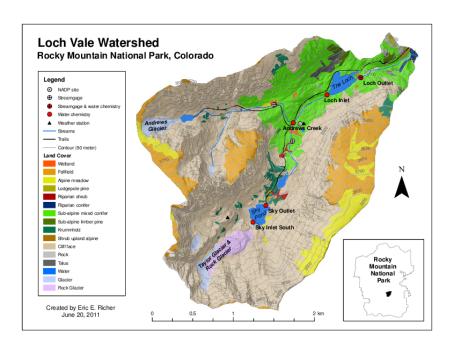


Figure 1. Figure of Loch Vale Watershed including height contours (m), alpine streams and lakes, and measurement sites

#### 1.1 Pollution effects

Concentrations of plant-available N have been increasing due to various anthropogenic activities (Nichols et al., 2001; Elser et al. 2009). Consequently, concern about pollutant transport into the Rocky Mountain National Park (RMNP) is on the rise. Pollutant gases along the Front Range such as nitrogen oxides ( $NO_x$ ) from engine combustion and industrial processes, sulfur dioxides ( $SO_2$ ) from coal combustion and gas processing, and ammonia ( $NH_3$ ) from agriculture and livestock can convert to particle phases through the following reactions found in Beem (2008), Jacob (1999), and Jacobson (2002):

 Gaseous NH<sub>3</sub> dissolves in water. NH<sub>3</sub> scavenges H<sup>+</sup> in rain/water to form ammonium (NH<sub>4</sub><sup>+</sup>) ions

$$NH_3$$
 (g)  $\Leftrightarrow NH_3$  (aq) (dissolved ammonia) (1)

$$NH_3 (aq) + H_2O(l) \Leftrightarrow NH_4^+ (s) + OH^-$$
 (2)

 Nitrate (NO<sub>3</sub>-) scavenges H<sup>+</sup> ions to form nitric acid (HNO<sub>3</sub>). HNO<sub>3</sub> combines with NH<sub>3</sub> to form ammonium nitrate

$$NO_{3}^{-}(g) + H_{2}O(1) \Leftrightarrow HNO_{3}(g) + OH^{-}$$
 (3)

$$HNO_3$$
 (g)+  $NH_3$  (g)  $\Leftrightarrow NH_4NO_3$  (s) (4)

• Sulfur dioxide ( $SO_2$ ) oxidizes in rain droplets to sulfate ( $SO_4^{2-}$ ), which scavenges H<sup>+</sup> ions to form sulfuric acid ( $H_2SO_4$ ).  $H_2SO_4$  combines with  $NH_3$  to form ammonium sulfate

$$SO_2 (g) \Leftrightarrow SO_2 \cdot H_2O$$
 (5)

$$SO_2 \bullet H_2O \Leftrightarrow HSO_{3^-} + H^+$$
 (6)

$$H_2O_2(g) \Leftrightarrow H_2O_2(aq)$$
 (7)

$$HSO_{3}^{-} + H_{2}O_{2}$$
 (aq) + H<sup>+</sup>  $\Leftrightarrow SO_{4}^{2-} + 2H^{+} + H_{2}O$  (8)

$$SO_4^{2-}(g) + 2H_2O^+(g) \Leftrightarrow H_2SO_4(g) + 2OH^-$$
 (9)

$$H_2SO_4(g) + 2NH_3(g) \Leftrightarrow (NH_4)_2SO_4(s)$$
 (10)

Mountain ecosystems are often assumed to be pristine environments and devoid of anthropogenic influence, which includes direct defacing of mountain surfaces or indirectly degrading land or water by atmospheric deposition of pollutants. N loading of Rocky Mountain aquatic and terrestrial ecosystems occurs when there is increased atmospheric deposition of N. Some effects of "cultural eutrophication" are reduced water transparency and oxygen depletion near the bottom of lakes (Elser et al., 2009). At a smaller scale, macronutrient ratios (nitrogen: phosphorous: potassium) become unbalanced when unnatural amounts of N enter an ecosystem, leading to loss in biodiversity (Gough et al. 2000; Stevens et al., 2004; Suding et al., 2005; Bobbink et al., 2010; Bowman et al., 2012). Species thriving in N-rich ecosystems may not be able to physically endure harsh winters in the mountains. Baron et al. (2009) found N increases at Loch Vale in RMNP is a result of N release from melting glaciers due to warmer summer and fall mean temperatures, exacerbating terrestrial and aquatic eutrophication.

The National Park Service, the U.S. Forest Service, Colorado Department of Public Health and Environment, and the U.S. Environmental Protection Agency teamed up to study trends and effects on air-quality issues facing RMNP, known as the Rocky Mountain National Park Initiative. Morris et al. (2013) concluded Loch Vale observed no trend in wet N deposition between 1984 and 2012 and Beaver Meadows saw an increase in wet N deposition between 1980 and 2012. No trend in nitrate was observed while ammonium showed an upward trend (Morris et al. 2013).

#### 1.2 Pollution sources

The Colorado Front Range is a major emitter of NO<sub>x</sub>, SO<sub>2</sub>, and NH<sub>3</sub>. The Rocky Mountain Atmospheric Nitrogen and Sulfur (RoMANS) study was conducted during spring and summer periods of 2006 with the objective of finding the origins and sources of N and sulfur that deposits in RMNP. With the exception of NO<sub>x</sub> emissions in northwest Colorado, the urban corridor between Colorado Springs and Fort Collins, CO is Colorado's biggest source region of NO<sub>2</sub> and SO<sub>2</sub> relative to other regions in the state (Benedict et al., 2013). From the RoMANS study, Malm et al. (2009) found about half of the wet-deposited N in RMNP originated west of the park while the other half came from the east—both cases having different meteorological transport mechanisms. This was interesting because sources of N for RMNP that are to the west are far away from the park (i.e. California, southern Nevada, Four Corners region (Malm et al., 2009), while sources to the east are relatively closer to the park (Colorado Front Range). However, because yearly-averaged winds are from the west, there is a persistent transport of N from western sources into RMNP. The other half of the wet-N deposition that originates east of RMNP come from regions of high N emission including the Colorado Front Range urban corridor, croplands, and concentrated animal feeding operations (CAFOs). Malm et al. (2009) pointed out that although transport from eastern Colorado into RMNP is infrequent, when upslope wind events occur, RMNP experiences its highest annual peaks of ambient N concentrations.

Weld County, Colorado has some of the largest concentrated animal feeding operations (CAFOs) in America with approximately 595,000 head of cattle (Lynn, 2012) in the county. From the CAFOs, notable emitted gases are methane (CH<sub>4</sub>), hydrogen sulfide (H<sub>2</sub>S), and NH<sub>3</sub> (Gómez-Moreno et al., 2010). Gaseous NH<sub>3</sub>, which is not plant-available, is

highly soluble in water. Gaseous NH<sub>3</sub> can convert to NH<sub>4</sub>+ from Reaction 2 (Jacob, 1999) through atmospheric processes and become a source of plant-available N. Inventory of all NH<sub>3</sub> emissions originating from the Colorado Front Range from National Park Service et al. (2010) are shown in Figure 2.

# 2002 Front Range NH3 Emissions

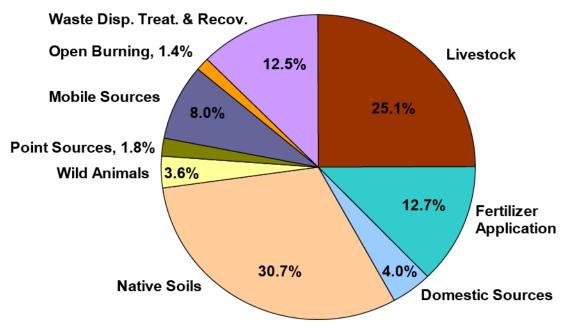


Figure 2. Estimates of Front Range  $NH_3$  emissions from 2002. Figure adapted from National Park Service et al. (2010).

#### 1.3 Transport mechanism

Though winds in Colorado are predominately from the west, thermally induced east winds during summer-time result from a mountain-valley circulation on the lee side of the Front Range in the event of little-to-no synoptic forcing (Markowski and Richardson, 2010). From daybreak, the mountain tops heat up quicker than air at the same altitude over the eastern-Colorado plains. Convection results from the higher temperatures on the mountain slopes, inducing a buoyancy-driven wind from the plains westward into the mountains, explained extensively by Wagner (1938), Ekhart (1948), Defant (1951), Thyer (1966), Toth

and Johnson (1985), Tripoli and Cotton (1989), and Wolyn and Mckee (1994). These upslope, or anabatic winds can travel 2-4 m/s (Defant, 1951). In the presence of moisture, atmospheric conditions become unstable, leading to convective precipitation over the mountain, emptying any pollutants entrained in the clouds (wet deposition) into the mountains (Baron and Denning, 1993; Benedict et al. 2013); with no moisture, advected pollutants from a mountain-valley circulation can still end up in the mountains through dry deposition—direct deposition of a gas or particle on a surface. Malm et al. (2009) found N deposition rates were about a factor of 2 higher in summer than spring, due to the difference meteorological processes that lead to precipitation, which was the motivation for this study (later emphasized quantitatively in Figure 6). In this study, we simulated the fine-scale meteorology of two summer (JJA) upslope events in which atmospheric deposition of N at three mountain sites was anomalously high.

#### 2. Methods

We analyzed 19 years (1994-2013) of precipitation (mm) and wet concentrations (mg/L) of inorganic N (NH<sub>4</sub>+ and NO<sub>3</sub>-) from three National Atmospheric Deposition Program National Trend Network (NAPD/NTN) sites: Beaver Meadows (CO19; 40.3639º N, 105.5810º W), Loch Vale (CO98; 40.2878º N, 105.6628º W), and Niwot Ridge (CO02; 40.0547º N, 105.5891º W). We then isolated two high wet-deposition summer events from the three mountain sites by choosing peaks from a time series of wet-N deposition at the Beaver Meadows site. The North American Regional Reanalysis (NARR) was used to observe synoptic conditions that led to or influenced the high-deposition events. The Weather Research and Forecasting (WRF) model was used to simulate the meteorology at a high time-and-space resolution for the progression of the upslope events that led to high deposition values of wet inorganic N in the Rocky Mountains.

2.1 National Atmospheric Deposition Program National Trends Network

The NADP/NTN monitors weekly precipitation chemistry for over 250 rural sites across the United States to study the effects of atmospheric deposition on ecosystems.

NADP/NTN sampling and analytical procedures are documented in Peden (1986).

Wet inorganic  $NH_4^+$  and  $NO_3^-$  concentrations (mg/L) and precipitation (mm) measured by NADP/NTN (National Atmospheric Deposition Program, 2013) were used to observe the seasonal and annual trends of wet inorganic N deposition at three different sites in the Rocky Mountains. Weekly wet-N deposition was calculated by multiplying the weekly precipitation, the concentrations of  $NH_4^+$  and  $NO_3^-$ , and the N contribution from each ion.

The sites, Beaver Meadows (2477 m), Loch Vale (3159 m), and Niwot Ridge Saddle (3520 m) were chosen as study sites to capture the precipitation differences for changing elevations. Beaver Meadows, located in RMNP, is surrounded by grassland meadows, sagebrush, lodgepole and ponderosa pine, and Douglas fir (Snyder, 2013). Loch Vale, also in RMNP, is near tree line and is surrounded by subalpine meadow, sub-alpine fir, Engelmann spruce, tundra, and bare rock/boulder/ice fields (Baron and Mast, 1992). Niwot Ridge Saddle, located about 40 km south of RMNP and 25 km west of Boulder, CO, USA, is above tree line and surrounded by alpine tundra (Niwot Ridge Long-Term Ecological Research Site, 2013). For all three sites, data from samples collected before January 11, 1994 were excluded due to changes in sample-handling procedures, which mostly affected free hydrogen ion concentrations (NADP/NTN, 2013).

The two sites in RMNP, Loch Vale (3159 m) and Beaver Meadows (2477 m), are located approximately 11 km apart but differ in height by 682 m resulting in different seasonal precipitation composition and totals discussed in Baron and Denning (1993). Baron and Mast (1992) showed deposition values at Loch Vale are closer to values at sites on the west side of the Continental Divide, whereas values at Beaver Meadows were better correlated with Estes Park data. This suggests N concentrations at high-elevation sites such as Niwot Ridge and Loch Vale come from synoptic-scale frontal disturbances with moisture from the Pacific Ocean carrying pollutants from the western United States. Conversely, deposition at Beaver Meadows is influenced by the transport of urban pollutants along the Colorado Front Range via large-scale cyclonic flow near the Texas Panhandle or mesoscale mountain-valley circulation, which both bring moisture from the Gulf of Mexico towards the mountains (Baron and Mast, 1992; Malm et al., 2009). A time series of wet deposition at

Beaver Meadows for the last decade (2003-2013) was used to select two high wet-N deposition events because it experiences most meteorological influence from the east.

#### 2.2 North American Regional Reanalysis

The NARR is a combined high-resolution model and assimilation dataset, providing 32-km meteorological data of North America 8 times per day since 1979 (Mesinger et al., 2006). With high temporal and spatial resolutions, the NARR is able to capture intricacies of small-scale phenomena such as diurnal heating and localized precipitation within large-scale systems. We used the NARR to observe the synoptic circulation and precipitation patterns leading to the high wet deposition events. Near the times of high wet deposition, monthly means of u-v winds and precipitable water were subtracted from the observed u-v winds and precipitable water to help diagnose future high-deposition scenarios based on anomalous weather patterns.

#### 2.3 Weather Research & Forecasting Model

The WRF model is a regional numerical model used for operational forecasting as well as atmospheric research. WRF is able to simulate real and idealized weather phenomena with scales ranging from meters to thousands of kilometers.

We used version 3.4.1 of the Advanced Research version of the WRF model (WRF-ARW) (Skamarock et al., 2008) to simulate and analyze details of two high wet deposition events at high spatial and temporal resolutions. Specifically, zonal wind direction and speed along with precipitation adjacent to the Colorado Front Range were of interest.

WRF was configured with one-way boundary conditions between four domains (shown in Figure 3) at a 3:1 parent-to-nest ratio. The coarse domain had grid spacing set to

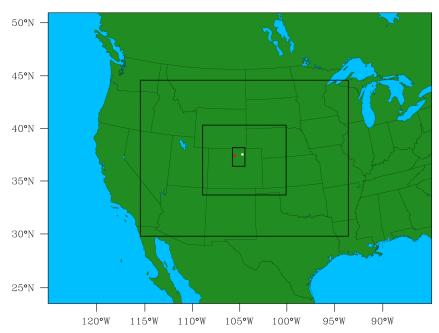


Figure 3. WPS domains with grid-spacings of 27 km, 9 km, 3 km, and 1 km for domains 1, 2, 3, and 4, respectively. Locations of the Kuner Feedlot in Weld County, Colorado and Rocky Mountain National Park are shown by the white and red dots, respectively.

27 km with a time resolution of 96 s. The vertical grid had 50 layers with a ceiling at 50 mb. WRF was initialized and nudged at the boundaries with data provided by the NCEP Operational Model Global Tropospheric Final Analyses (National Centers for Environmental Prediction, National Weather Service, NOAA, U.S. Department of Commerce, 2000). For each case, WRF was run for 48 hours with an output time interval of 10 minutes for each domain. We initialized WRF 6 hours prior to sunrise to avoid erroneous spin-up meteorological features during hours of interest (Weiss et al., 2008).

Model physics were based on and described in Nehrkorn et al. (2010), which analyzed high-resolution data of a time-inverted lagrangian transport model that used WRF output. Table 1 shows the physical schemes used in this study. The Mellor-Yamada-Janjic planetary boundary layer scheme that included a one-dimensional turbulent kinetic energy scheme was used for domains 3 and 4, where convection was not parameterized.

Table 1. Physics schemes used in WRF, which were outlined in Nehrkorn et al. (2010).

Physical parameter	Scheme	Domains applied
Microphysics	Lin et al. (1983)	All
LW Radiation	RRTMG (Iacono et al., 2008)	All
SW Radiation	New Goddard	All
	Yonsei University with Noah Land	
PBL	Surface Model and MM5 similarity	1, 2
	theory-based surface layer scheme	
	Mellor-Yamada-Janjic with Noah	
PBL	Land Surface Model and the Eta	3, 4
	similarity theory-based surface	
	layer scheme	
Cumulus Convection	Grell 3D (Grell and Dévényi, 2002)	1, 2

We included a passive tracer (tracer\_opt = 2 in the dynamics section of

namelist.input) discussed in Barth et al. (2012) in a 2x2 grid points of domain 4. The unitless passive tracer, released at every time step (dt =  $3.\overline{55}$  s) from the surface, represented emissions from Kuner Feedlot managed by JBS Five Rivers Cattle Feeding LLC in Weld County, CO, shown by the white dot in Figure 3. At each time step, the surface of the approximate location of the feedlot was set to a tracer value of 1. Winds advected and dispersed the tracer into the 3D space of the  $4^{th}$  domain. Tracer concentrations downwind of the feedlot were normalized to the release-concentration of 1.

#### 3. Results

#### 3.3 NADP

Monthly wet deposition of inorganic N and precipitation amounts from 1994-2013 from Beaver Meadows, Loch Vale, and Niwot Ridge are shown in Figure 4 (NADP/NTN, 2013). Beaver Meadows and Loch Vale both had bimodal precipitation distributions for the year, with a primary maximum in the spring and a secondary maximum in the summer.

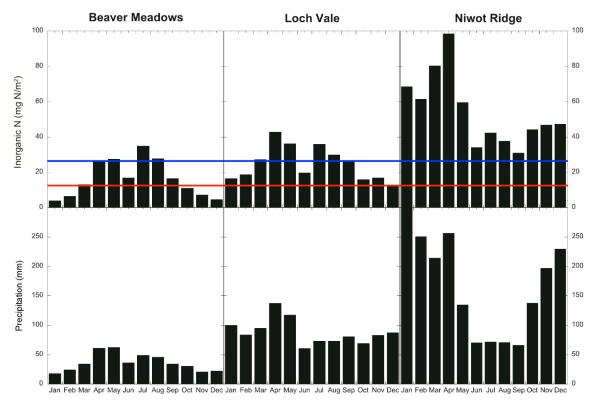


Figure 4. Inorganic N deposition and precipitation amounts for 3 NADP sites, Beaver Meadows (2477 m), Loch Vale (3159 m), and Niwot Ridge (3520 m), from 1994-2012 (NADP/NTN, 2013). The red line marks the ecosystem-response threshold for Loch Vale Watershed (12.5 mg N m $^{-2}$  mo $^{-1}$ ) in RMNP (Baron, 2006); the blue line marks the ecosystem-response threshold for alpine tundra (25.0 mg N m $^{-2}$  mo $^{-1}$ ; Bowman et al., 2012).

Niwot Ridge, the highest elevation site of the three, had most precipitation from winter and springtime winter storms. Interestingly, monthly distributions of wet deposition of inorganic N also show a bimodal distribution for the lower elevation sites; however, the primary maximum is in the summer while the secondary maximum occurs in the spring.

Warmer temperatures and longer hours of sunlight during summers yield more dynamic atmospheric reactions producing  $NO_3^-$  ions and gaseous  $NH_3$  (Baron et al., 1992; Hargreaves and Tucker, 2004). The red line in Figure 4 marks the ecosystem-response threshold for Loch Vale Watershed (12.5 mg N m<sup>-2</sup> mo<sup>-1</sup>) in RMNP (Baron, 2006); the blue line marks the ecosystem-response threshold for alpine tundra (25.0 mg N m<sup>-2</sup> mo<sup>-1</sup>; Bowman et al., 2012).

Figure 5 shows a decade (January 1, 2003 – January 1, 2013) of weekly wet deposition of N associated with  $NH_4^+$  and  $NO_3^-$  ions as well as the total wet N deposition from  $NH_4^+ + NO_3^-$  from Beaver Meadows, Loch Vale, and Niwot Ridge sites. The highest peaks in Figure 5 were springtime deposition events and, therefore, not taken into account for this study. Two anomalously high deposition events that occurred August 18-20, 2006 and July 6-8, 2012 (circled red peaks in Figure 5) were chosen as our study cases.

We then stratified summer weeks between 1994 and 2013 (n=187) at the lower elevation site (Beaver Meadows) by deposition values to show our chosen time periods were representative of other high wet inorganic N deposition events. The secondary frequency peak of high deposition of wet inorganic N in Figure 6, which included our chosen cases from 2006 and 2012, 32.56 and 28.37 mg N m<sup>-2</sup> wk<sup>-1</sup>, respectively, was the primary motivation for this study. The highest deposition case (farthest right column in Figure 6) was not considered because we were interested in high-deposition cases that were representative of other high-deposition cases.

#### *3.2 NARR*

Figures 7 and 8 are four-panel plots of meteorology at different height levels using the NARR for the August 2006 and July 2012 high wet deposition events, respectively. For

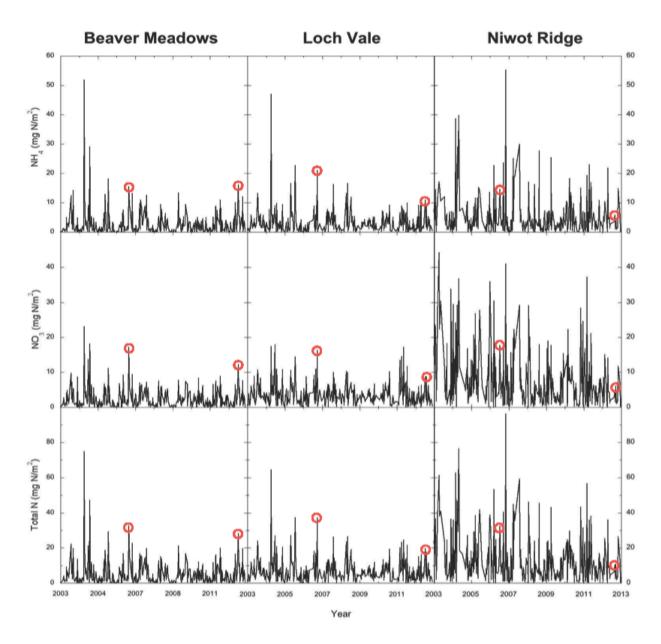


Figure 5. 10-year timeline (01/2003-01/2013) of wet inorganic N deposition for three NADP sites—Beaver Meadows, Loch Vale, and Niwot Ridge. Red-circled peaks are the deposition events of interest (August 2006 and July 2012) in this study (NADP/NTN, 2013).

both figures, the top left panel (a) is a CONUS 250-mb map with wind barbs (kts), contoured geopotential heights (m), and shaded isotachs (kts); the top right panel (b) is a CONUS 500-mb map with wind barbs (kts), contoured geopotential heights (m), and shaded relative vorticity  $(10^{-6} \, \text{s}^{-1})$ ; the bottom left panel (c) is a CONUS map of contoured

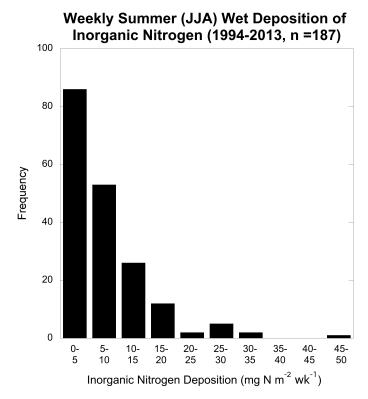


Figure 6. Frequency versus wet deposition of inorganic nitrogen for summer (JJA) weeks between 1994 and 2013 (n = 187) (NADP/NTN, 2013)

1000-500 mb heights (m) (red), contoured mean sea-level pressure (hPa) (MSLP; black), and shaded 700-mb relative humidity (%) (RH); the bottom right panel (d) is a map of Colorado and its surrounding area with 850-500 mb wind barbs (kts) removed from the August mean wind values (i.e. wind barb value = wind at specified time – average August wind value) and shaded precipitable water content values (mm).

From Figure 7a, an upper-level jet skirted northern Colorado around midnight. At 500 mb (Figure 7b), an anti-cyclone and a cyclone simultaneously developed over the Colorado-Wyoming-Nebraska triple point and southeastern Colorado. In figure 7c, a large-scale anti-cyclone in southern Canada is driving higher pressures from the northeast into the Colorado Rocky Mountains. The circulation pattern at low-mid levels, best shown in Figure 7d, was a recipe for enhanced pollutant transport from eastern Colorado into the

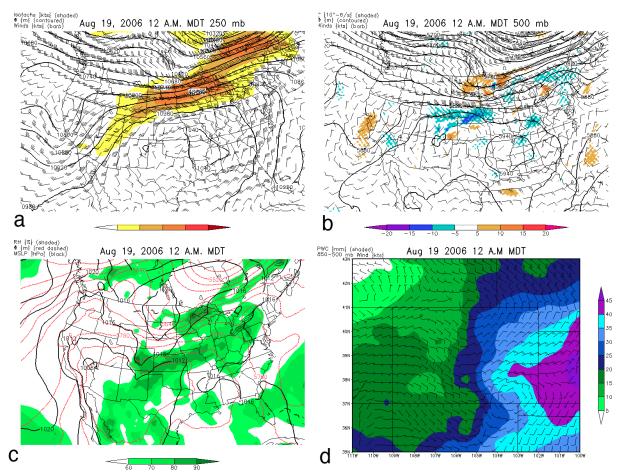


Figure 7. A 4-panel plot of the NARR at 12 A.M. MDT on August , 19 2006—top left: 250-mb map of wind barbs (kts), shaded isotachs (kts), and contoured heights (m); top right: 500-mb map of wind barbs (kts), shaded relative vorticity ( $10^{-6} \, \text{s}^{-1}$ ), and contoured heights (m); bottom left: shaded relative humidity (%) at 700 mb, contoured MSLP (hPa) (black), and contoured 1000-500 mb heights (m); bottom right: shaded precipitable water (mm) and winds (kts) subtracted from the entire month of August 2006.

Rocky Mountains. High RH values in the bottom left panel showed there was ample atmospheric moisture at low-mid levels for wet deposition into the mountains.

A broad high-pressure circulation was retrograding from the central U.S. to the Mountain-West states at the time of the still CONUS maps shown in Figure 8 (July 2012 event). The location of the broad upper-level ridge of high pressure, shown in Figures 8a and 8b, allowed for moist, southerly flow into Colorado. Similar to the August 2006 case, circulation patterns such as anomalous southerly winds from the location of the broad upper-level high pressure concurrently with a mid-level anti-cyclone located in

northeastern Colorado (shown in the Figure 8d), were once again responsible for the enhanced pollutant transport into the Rocky Mountains. And like the August 2006 high wet deposition event, a surface anti-cyclone in southern Canada drove winds from the northeast into the Colorado Rocky Mountains, shown by MSLP contours in Figure 8c.

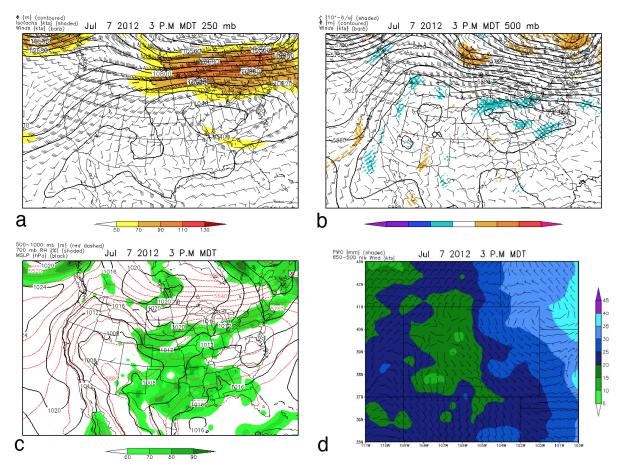


Figure 8. A 4-panel plot of the NARR at 3 P.M. MDT on July 7, 2012—top left: 250-mb map of wind barbs (kts), shaded isotachs (kts), and contoured heights (m); top right: 500-mb map of wind barbs (kts), shaded relative vorticity ( $10^{-6} \, s^{-1}$ ), and contoured heights (m); bottom left: shaded relative humidity (%) at 700 mb, contoured MSLP (hPa) (black), and contoured 1000-500 mb heights (m); bottom right: shaded precipitable water (mm) and winds (kts) subtracted from the entire month of July 2012.

### 3.3 WRF

Figures 9 and 10 show 48-hour time series of zonal wind from WRF output for Beaver Meadows, Loch Vale, and Niwot Ridge during the August 2006 and July 2012 high wet inorganic N deposition events, respectively. Because both simulations started at

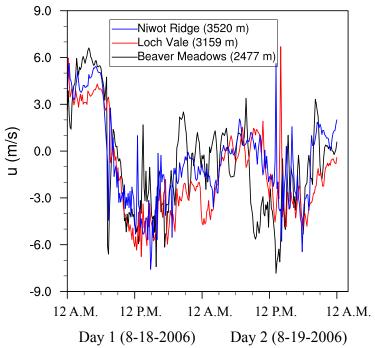


Figure 9. WRF output: 48-hour time series of zonal wind at 3 NADP sites starting August 18, 2006

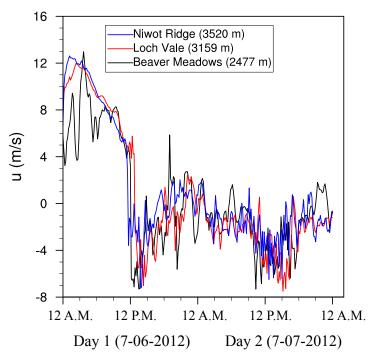


Figure 10. WRF output: 48-hour time series of zonal wind at 3 NADP sites starting July 06, 2012

midnight, hours 0-23 were considered "Day 1" while hours 24-47 were considered "Day 2".

For both time periods shown in Figures 9 and 10, a diurnal cycle of anabatic/katabatic winds is evident with a higher magnitude on Day 1. At all locations for 2006 and 2012, the peak east wind occurred around 2 P.M. local time on Day 1. Around 2 P.M. of Day 2 (hour 38), Niwot Ridge and Loch Vale experienced maximum west winds at the same time Beaver Meadows experienced maximum east winds. The wind speeds at all three locations were approximately the same. The opposite direction of concurrent maximum winds for the three locations on the afternoon of Day 2 in 2006 suggested a synoptic wind influence for the higher locations, Niwot Ridge and Loch Vale. On Day 2 of the 2012 event, winds were predominately from the east for all three locations for the entirety of the day, which suggested widespread upslope as a result of a more robust synoptic wind pattern.

Mountain-valley circulation alone did not lead to N transport for the August 2006 and July 2012 high wet inorganic N deposition events. Moisture associated with synoptic systems, shown in the lower panels of Figures 7 and 8, aided in the small-scale convective transport of pollutants from eastern Colorado westward into the Rocky Mountains.

Figures 11-16 show cross sections of wind streamlines, stippled cloud water concentrations (g/kg), and shaded base-10 logarithms of the unitless passive tracer concentrations at specific times within the two case studies (August 2006 case: Figures 11-13; July 2012 case: Figures 14-16). The cross section extends across the 1-km domain from Figure 3 and goes through RMNP and the approximate location of the Kuner Feedlot in Weld County, Colorado. The three red dots at the surface from high-to-low elevation are the approximate locations of Loch Vale, Beaver Meadows, and the Kuner Feedlot.

#### 3.3.1 Case 1: August 19, 2006

At 13:20 MDT (Figure 11), surface easterlies over the eastern plains extended from the surface to ~1 km above the surface and carried convected pollutants towards the mountains. Growing clouds reach the surface near the mountaintops and just below Beaver Meadows while a large area of tracer concentrations was transported up the mountainside while. However, before the deep concentrated tracer was transport by the upslope winds, cloud-scale convection was responsible for the vertical transport immediately west of the feedlot. Fast-forwarding 30 minutes to 13:50 MDT (Figure 12), the clouds near the mountaintops and near Beaver Meadows grew vertically and horizontally. The updraft associated with the cloud near Beaver Meadows entrained tracer well over 4 km ASL. The area of high tracer concentration just west of the Kuner feedlot grew much more in the vertical than the horizontal, indicating cloud-scale convection plays a large role in N

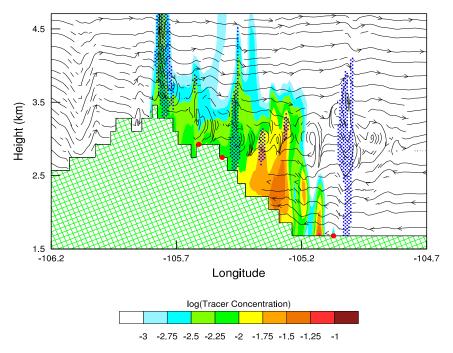


Figure 11. August 19, 2006 at 13:20 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

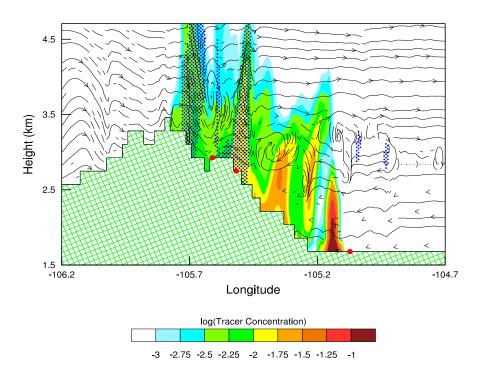


Figure 12. August 19, 2006 at 13:50 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

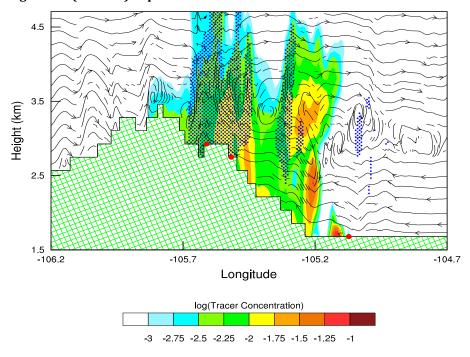


Figure 13. August 19, 2006 at 14:20 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

transport. By 14:20 MDT (Figure 12), the clouds that were once at the mountaintops and near Beaver Meadows grew and combined. The extensive cloud, reaching the ground at both Loch Vale and Beaver Meadows, was collocated with high tracer concentration (tracer concentration > 0.01).

#### 3.3.2 Case 2: July 7, 2012

At 14:00 MDT (Figure 14), surface easterlies on the eastern plains extend from the surface to ~1 km above the surface, similar to Case 1. From Figure 14, a large cloud extending down to Loch Vale and Beaver Meadows began to entrain a large volume of tracer, with some tracer already collocated with the cloud just above Beaver Meadows. Closer to the Kuner Feedlot, a large area of tracer indicated winds were blowing directly along the cross section from the feedlot towards RMNP. 30 minutes later, at 14:30 MDT (Figure 15), the tracer that was once near the feedlot started up the mountain before getting entrained into a convective cloud while the wind advected another plume into the cross section (shown by the small plume immediately west of the Kuner feedlot). Over the next 40 minutes to 15:10 MDT (Figure 16) the vertically-stretched volume of tracer between Beaver Meadows and the Kuner feedlot was advected by the upslope winds to the location of the cloud over Loch Vale and Beaver Meadows. Meanwhile, clouds moving towards the East entrained tracer, seen as the once-small plume in Figure 15 near the Kuner feedlot, well above 4.5 km. Interestingly, this rain event was largely responsible for extinguishing the High Park Fire West of Fort Collins, CO and subsequent flash flooding around the High Park Fire burn scar.

Realistically, for both cases, the tracer emitted from the feedlot would have been water-soluble ammonia and would either have been entrained into the cloud or scavenged

by falling rain droplets. Hence, any collocation of the tracer with a cloud extending to the surface would suggest wet deposition.

The Stage IV analysis of NEXRAD radar imagery, which includes manual quality control, was used to verify the location of cloud concentrations from WRF shown in Figures 11-16 were realistic. One should note replicating exact precipitation quantities to in order to quantify wet deposition was not the intention of the comparison of Stage IV analysis with WRF output. Shown in Figure 17 are regional 6-hour plots of precipitation from the 2006 (top panels) and 2012 (bottom panels) cases with Colorado as the main focus. Precipitation plots from WRF output were from domain 3 (3 km grid spacing). The location of RMNP can be seen by the black dot in north-central Colorado.

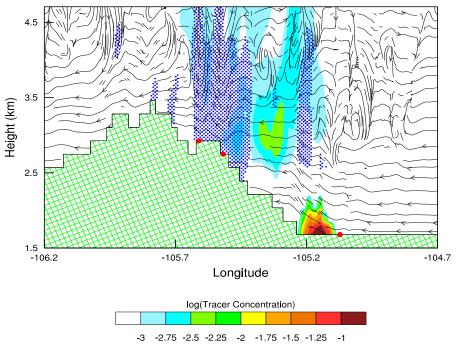


Figure 14. July 7, 2012 at 14:00 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

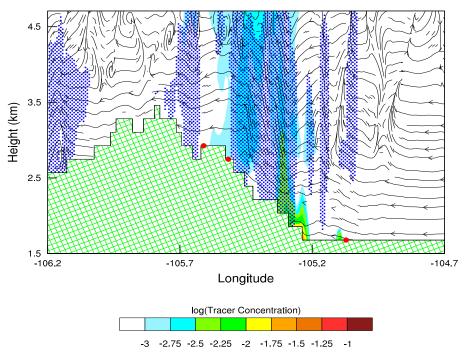


Figure 15. July 7, 2012 at 14:30 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

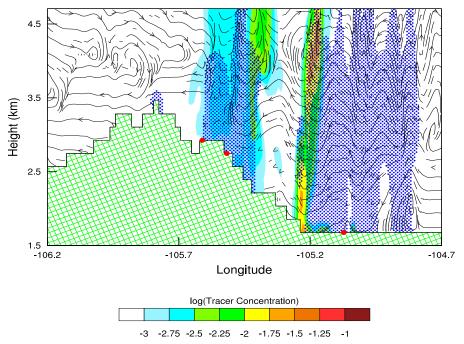


Figure 16. July 7, 2012 at 15:10 P.M. MDT. Cross-section across 1-km domain from Figure 3 going through the locations of Rocky Mountain National Park and Kuner Feedlot in Weld County, CO of wind streamlines, stippled clouds, and shaded logarithm (base 10) of passive tracer concentrations.

From the precipitation plots in Figure 11, RMNP received precipitation during the afternoon hours on August 19, 2006 and July 7, 2012. Radar imagery showing precipitation at RMNP enforces the claim of wet deposition provided by the collocation of tracer and clouds from WRF (Figures 9 and 10).

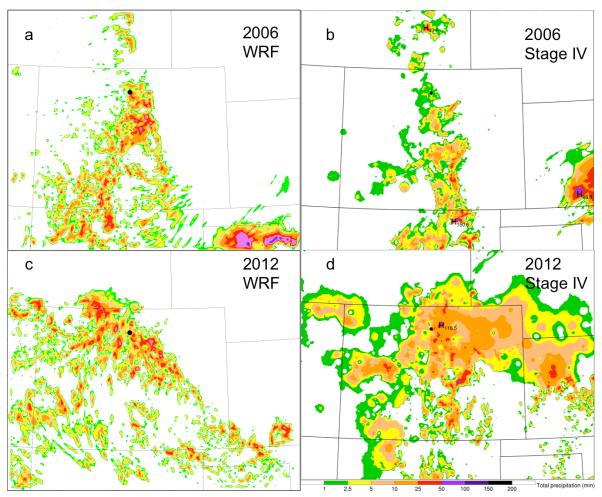


Figure 17. a. 6-hour precipitation from WRF output ending at 6 P.M. MDT on August 19, 2006; b. 6-hour precipitation from Stage IV analysis ending at 6 P.M. MDT on August 19, 2006; c. 6-hour precipitation from WRF output ending at 6 P.M. MDT on July 7, 2012; d. 6-hour precipitation from Stage IV analysis ending at 6 P.M. MDT on July 7, 2012. The black dot in north-central Colorado is the location of Rocky Mountain National Park.

#### 4. Discussion and Future Work

We isolated two recent anomalously high wet deposition events of inorganic N in the Rocky Mountains using data from NADP/NTN. From the NARR, we were able to identify circulations at low, mid, and upper levels as key players for bringing moisture into eastern Colorado. The NARR helped identify which meteorological patterns were possible high wet deposition days when viewing weather forecasts. We then simulated the meteorological set-up of the wet deposition events using WRF over northern Colorado with a 1-km grid and a passive tracer, illustrating NH<sub>3</sub> emissions from a feedlot in eastern Colorado.

Due to the consistency of the buoyancy-driven upslope winds and the presence of moisture with monsoonal flow from the south each year, high deposition during summer months were expected. However, unexpected mid-level synoptic circulation was found to be a key player for the penetration of higher precipitable water values, shown in Figures 7 and 8, into the Front Range of Colorado. Higher precipitable-water values led to enhanced convective activity during the case studies from 2006 and 2012. From Figures 11-16, we showed cloud-scale convective transport along played a very important role in N deposition in the Rocky Mountains in addition to the mesoscale mountain-valley circulation.

From this study, we will use WRF output as input to the Stochastic Time Inverted Lagrangian Transport (STILT) model mentioned in Nehrkorn et al. (2010) to geo-locate eastern Colorado feedlots that are responsible for high N emissions. Locating large sources of concentrated N will be one step further to reducing N loading in the Rocky Mountains. Bowman et al. (2012) mentioned land managers and air quality policy makers should identify any sign of ecological changes due to N deposition as an important prevention goal.

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