Increasing Ca^{2+} deposition in the western US: the role of mineral aerosols

Abstract

Considerable research has focused on the role of industrial emissions in controlling the acidity of precipitation; however, much less research has focused on the role of mineral aerosols emitted from soils. According to data published by the National Atmospheric Deposition Network (NADP), over the past 17 years Ca^{2+} deposition has increased over large regions of the U.S. A trend analysis to determine regions of significant change in Ca^{2+} deposition revealed statistically significant increases in three broad regions within the western half of the country: the inter-mountain west, the midwest, and the northwest. We evaluated potential changes in sources of calcium to the atmosphere including soil erosion, industrial emissions, forest fires, and sea-salt aerosols to determine the cause of rising atmospheric calcium deposition. Based on our evaluation, the most parsimonious explanation for increased Ca^{2+} deposition is an increase in mineral aerosol emissions from within the western US. This explanation is corroborated by independent evidence showing increases in the frequency of dust storms and low-visibility days across regions of the western U.S. Furthermore, our analysis indicates that the increase in mineral aerosol emissions is most likely due to (1) increased aridity and wind transport and (2) increased area and intensity of upwind human activities. Changes in atmospheric dust concentrations can have important ecological implications through the contribution of acid neutralizing capacity to both precipitation and regions of deposition. Thus increased dust emissions have the potential to ameliorate the detrimental effects of acid precipitation on terrestrial ecosystems, though dust may exacerbate the impacts of air quality on human health.

Keywords
Dust, Precipitation, Alkalinity, Land-use, Wind, NADP, Calcium

1.0 Introduction

Our trend analysis in precipitation chemistry show increases in Ca^{2+} concentration across much of the US (NADP, 1994-2009). This new upturn is a reversal to the decline in Ca^{2+} deposition noted in the 1980s in the U.S. as well as Europe (Hedin et al., 1994; Hedin and Likens, 1996, Nilles 2001, Lynch 1995). Though the exact causes of the declines were speculative, some suggested that the decrease was due to industrial regulation of emissions (Hedin et al., 1994; Hedin and Likens, 1996; Lee and Pacyna, 1999), decreasing soil aridity in these regions (Sequiera, 1993), or a switch in monitoring methods from bulk to wet only precipitation monitoring (Sequiera, 1993). The aim of this paper is to determine the cause of the recent increase in atmospheric Ca^{2+} deposition over much of the U.S.

Many calcium-bearing minerals, in particular calcite ($CaCO_3$), affect precipitation chemistry through their acid-neutralizing capacities, (Sequiera, 1982; Young et al., 1988; Sequiera, 1993; Rogora et al., 2004), and can subsequently increase the buffering capacity of waters in depositional regions. The acidity of precipitation is a major environmental concern because of the wide variety of ecological impacts of low pH precipitation (Schindler, 1988). The decline in base-cation deposition in the late 1980s raised concern because acid emissions were increasing while buffering capacity was decreasing (Hedin et al., 1994).

In addition to the acid-neutralizing capacity of calcium bearing minerals, calcium is an essential nutrient for plants. In areas where chronic acid deposition has occurred, calcium and other base-cations have been leached from the soils (Likens et al., 1996). Increases in calcium deposition can reduce precipitation acidity, and but replenish Ca^{2+} that has been lost from soils due to decades of chronic acid deposition.

Calcium enters the atmosphere through a variety of mechanisms including sea-salt aerosols, forest fires, industrial emissions, and the wind erosion of soils and subsequent dissolution of calcium bearing minerals (Meszaros, 1966; Clayton, 1976; Sequiera, 1993; Lee and Pacyna, 1999). In the US, there are no published continental-scale budgets for Ca^{2+} in total suspended particulate emissions. In Europe, however, two-thirds of Ca^{2+} deposition is attributed to wind-blown soil, whereas most of the residual is associated with emissions of fly-ash from small power plants that do not yet have emission-abatement technology (Lee and Pacyna, 1999). Other potential industrial sources of Ca^{2+} appear to be negligible (Lee and Pacyna, 1999). In the U.S., major sources include mineral aerosols from soils and industrial emissions, as well as minor contributions from sea-salt aerosols and forest fires (Sequiera, 1993).

In the U.S., natural and industrial sources of airborne calcium will be tied spatially to their source regions and temporally to climate and human factors. Sea-salt aerosols can increase Ca^{2+} deposition but are limited to coastal areas. Dust associated Ca^{2+} deposition will be more prevalent closer to arid regions and the extensive loess deposits in the U.S., and will vary with climate and land-use (Reheis, 2006; Reheis and Urban, 2011). Most fires in the western U.S. occur during the summer months, with considerable inter-annual variability in frequency and aerial coverage (Westerling et al., 2003). Deposition of Ca^{2+} from fires would thus be strongly tied to fire locations and proximity, wind patterns, and the acreage burned. Similarly, industrial emissions would be greatest closest to large

concentrations of cement and power plants, and temporal trends in these emissions would parallel demand trends for energy and materials and the implementation of pollution-control systems.

In this paper, data from the National Atmospheric Deposition Program are used to determine regional trends in cation and anion concentration, pH, and alkalinity of precipitation for the continental US. These trends are compared with Ca^{2+} emission mechanisms related to human and climatic factors.

2.0 Methods

NADP preciptation chemistry data

Changes in Ca^{2+} concentration and overall precipitation chemistry in the U.S. were evaluated by individual site trend analyses of precipitation alkalinity, pH, and the ionic composition of wet deposition from 175 sites from the NADP network from 1994 to 2010. Data are analyzed from 1994 onwards because in 1994 NADP changed analytic procedures due to prior contamination of samples during shipment (NADP 1994). The NADP collection program involves an array of sites across the U.S. that were selected to be regionally representative of atmospheric precipitation chemistry (NADP, 2011). The NADP sites use the Aerochem Metrics 301 precipitation collector and the Belfort B5-780 recording rain gage. The precipitation collector is a wet-only collector and remains closed to the atmosphere until a sensor is triggered by precipitation. This set up excludes dry deposition. Though a dry bucket is collected by the NADP, this fraction not analyzed. All samples are sent to the Central Analytical Laboratory (CAL) of the Illinois State Water Survey. Quality assurance protocols are required for sample collection, transport, processing, chemical analysis, data validation, and verification prior to the transfer of data to the NADP program office.

The dissolution of calcium bearing carbonate minerals can have a direct effect on precipitation pH and alkalinity. Alkalinity, the ability for water to neutralize strong acids, is not measured by titration in the NADP network; however, we calculated alkalinity as the difference between strong base-cations and strong acid-anions (Schlesinger, 1997). Thus, the fraction of alkalinity change in rainfall attributable to any particular ion (ΔX_o) was calculated according to $|\Delta X_o|/\Delta Alk$.

$$\Delta Alk = \Delta Ca^{2+} + \Delta Na^+ + \Delta K^+ + \Delta NH_4^+ + \Delta Mg^{2+} - \Delta NO_3^{2-} - \Delta SO_4^{2-} - \Delta Cl.$$

Calcium can also be deposited in dry forms but at present, there is no nationwide network of dry deposition samplers that can be used in a manner similar to that used for the NADP analysis described above. The IMPROVE network of aerosol sampling sites does measure dry deposition but these measurements do not include the particles larger than 10 micrometers in diameter which can dominate the mass flux of particulates in arid environments (Lawrence and Neff, 2009). As a result, we do not attempt to quantify dry deposition of Ca here but note that the estimates based on wet deposition alone will be a conservative estimate of total Ca loading to ecosystems in the U.S.

2.1 Trends in Ca deposition

We examined trends at two levels: at individual stations and at larger regional scales. For each NADP station with more than 10 years of data we analyzed trends using the Mann-Kendall trend test.

The Mann-Kendall test is a non-parametric test for a monotonic trend (Sen, 1968). It is preferred over regression for trend analysis because it is less sensitive to outliers. Because the trend data from individual stations is short, and some stations may be close to unique disturbances that are not reflective of larger regional patterns, sites were grouped into US EPA level III ecoregions. Although the EPA regional separation is somewhat arbitrary, it provides an independent and unbiased approach for site selection for regional trend analysis. We note that it would be possible to show stronger regional trends by a more subjective clustering of NADP sample sites. In this analysis all sites are included in a given region, regardless of the trend, to obtain regional statistics. We constructed composite Ca^{2+} trends by compiling all sites within a region. Annual observations at each site (O) were normalized to their respective variance ((O -μ)/σ) before being averaged by year for a regional trend. Confidence limits were constructed based on a bootstrap re-sampling of sites with replacement in each ecoregion, a total of 1000 bootstrap realizations were conducted. The significance of the regional composite trend was also tested using the Mann-Kendall trend statistic. To correct for sea-salt-aerosol contribution, we analyzed the data for chlorine concentrations. Coastal regions and regions near the Great Salt Lake had higher Cl^- deposition rates. We corrected the calcium deposition rates by using a sea-spray Ca:Cl ratio of 0.02 (Gorham, 1957).

2.2 Analysis of mechanisms underlying trends in Ca deposition

2.2.1 Industrial sources

To evaluate coal-fired power plant emissions we examined production trends from coal fired power plants in the regions of interest. Data are obtained from the Emissions & Generation Resource Integrated Database (eGRID); available data years span from 1996-2009 (EPA, 2012). We evaluated the annual coal generation of MWh for each region (by state). Neither calcium nor particulate matter emissions are directly measured; instead we use annual energy production from coal as a proxy for potential trends in emissions. Though the amount of Ca oxides present in fly ash is well known, we cannot calculate an emission rate without knowledge of fly ash or particulate emissions through time. However, since regulations on emissions over the last few decades have resulted in a 95-100% reduction in solid-particle emissions (EPA 1993,1995), the amount of coal used serves as a conservative estimate because it defines the potential for fly-ash emissions without considering improvements in abatement technology, which should be leading to declines in overall Ca oxide emission.

The cement industry also does not report particulate emissions. However, according to a 1993 EPA report to Congress, air pollution control devices in the cement industry capture 98-100% of cement kiln dust (EPA, 1993). Therefore we can again assume that the regional trends in cement production are representative of potential particulate emissions, albeit conservative given the high efficiency the air pollution control devices (EPA, 1993). We calculate annual production regionally by grouping plants within our regions from the USGS cement production database (USGS, 2012).

2.2.2 Forest Fires

We examined trends in fire frequency and acreage burned using data from the National Interagency Fire Center (NIFC, 2012), available by year and by state from 2002 to 2012. Data for states that fall within our aggregate EPA ecoregions were grouped to evaluate trends over this time period.

2.2.3 Mineral sources

We evaluated trends in soil erosion through a variety of mechanisms. We associate dust emissions to factors such as seasonality, climate, and the influence of human activities. If Ca^{2+} deposition is strongly influenced by mineral aerosol concentrations, then we would expect a strong correspondence between changes in the factors that produce wind erosion and the observed changes in Ca^{2+} deposition.

Mineral aerosols are produced by a combination of factors including the erosive force of wind, and the surface properties that act to resist these forces. Wind speeds control the erosion potential of surface materials and subsequent transport, while soil properties and the presence of surface protectors (e.g. rocks, vegetation) determine the erodability of the soil (Goudie and Middleton, 2006). Climate, and specifically drought, can increase soil erodibility by reducing the amount of soil moisture and decreasing the amount of surface-stabilizing vegetation (Field et al., 2009). Human land uses can also increase the erodibility of soils by diminishing soil stability and decreasing or removing vegetative cover (Neff et al., 2005).

To examine possible trends in the climate factors that influenced dust generation over the period of the NADP Ca^{2+} deposition records, we compared regional Ca^{2+} deposition rates against a statistical model that combines regional palmer drought severity index (PDSI) and the number of days of wind speeds greater than 25.7 m s^{-1} to determine erosion or dust potential (NOAA, 2012). This wind speed threshold is used by the National Oceanic and Atmospheric Administration (NOAA) to classify large wind-storms (NOAA, 2012). Though wind erosion of soils can be initiated at much lower wind speeds (Goudie and Middleton, 2006), at the regional scale of this analysis it is difficult to create aggregate wind statistics that would be useful in analysis of Ca deposition trends. Instead, we use the NOAA windstorm statistics to quantify the number of strong wind events that could be responsible for regional scale wind erosion of soils. In the American west, PDSI and wind statistics were generated from the suspected source regions of the Mojave Desert and the Colorado Plateau (Sequiera, 1993; Reheis, 2006). In the northwestern region, PDSI and wind were determined for the Columbia Plateau in Washington, a well-known source of agricultural dust (Stetler and Saxton, 1996; Claiborn, et al., 1998; Saxton et al., 2000). The potential source region for the midwestern area was less easy to define spatially because loess deposits and agricultural areas are large in areal extent. To determine the source region for the midwest we used a spatial analysis of PDSI and compared this to the dominant midwestern Ca^{2+} trend. This analysis revealed high correlations to the Southern High Plains in northern Texas and Oklahoma. This region corresponds to the Dust Bowl region of the 1930s and contains extensive loess deposits and agricultural areas that continue to produce dust storms (Lee et al., 2012). Wind storm and PDSI statistics are show in Table 1. The dust generation models were constructed by first normalizing each variable $((O-\mu)/\sigma)$, then dust generation was related to climatic factors such that Dust generation = Days with windspeeds > 25.7 m s^{-1} (x) + (-PDSI)(y), where PDSI is included as a negative predictor variable because the drought conditions required to promote soil erosion correspond to negative PDSI values. Least-squares regressions were then performed using both factors (Wind Speed and PDSI) together and independently for all three dust-producing regions.

To independently corroborate that increases in Ca^{2+} deposition is due to increased dust storm activity, we compiled dust-storm activity in the regions of interest using incidences of reported dust events from the NOAA storm database and also low-visibility events from NOAA weather stations

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near desert and agricultural regions (NOAA, 2012b). Incidence of low visibility can arise from a variety of factors including, fog, smog, rain, and suspended particles (Spence, 1931; Pitchford et al., 1981). In semi-arid and arid regions fog, smog, and rain are far less likely than dust-storm activity. We define a low-visibility event as a time period during which visibility drops below 6000 and then 3000 meters; the event does not end until visibility increases above 6000 meters. These considerations ensure that (1) visibility changes within the same event are not counted multiple times, (2) changes in the frequency of data reporting does not affect our analysis, and (3) short non-substantive events are not counted.

3.0 Results

3.1 Deposition trends

On a site-by-site basis, trend analysis indicated that 116 of 175 sites across the U.S. have received increases in Ca^{2+} deposition over the last 17 years (Figure 1, Supplementary Table 1). Application of the non-parametric Mann-Kendall statistic to evaluate trend significance reveals that 26 of the site-level trends are significant at ($p \leq 0.05$). This result is 3x higher than expected by chance alone, implying that these temporal trends are not random. Furthermore, these sites tend to cluster within prominent areas in the inter-mountain west, northwest, and midwest (Figure 1), suggesting that these observed trends are not spatially random. To test this hypothesis, we used Moran's Index to determine whether or not the data is statistically spatially clustered or random. Moran's index was 0.451, the Z score was 5.12, and the p value was <0.000 indicating that the deposition data is indeed spatially clustered. Seven of eight EPA Ecoregion groups showed increases in Ca^{2+} concentration, and all grouped regions showed increases in pH and alkalinity (Table 2).

3.2 Regional patterns in potential sources

Increases in calcium deposition are seen across much of the nation, with three prominent hot spots in the Western US (Figure 1); trends of Ca^{2+} deposition in the inter-mountain west, northwest, and midwestern regions were significant at $p < 0.05$ (Table 3, Figure 2). Western states, particularly Colorado, Utah, and Wyoming, have seen the greatest increases in calcium deposition, with roughly double the Ca^{2+} deposition rate (average 230 mg m^{-2} yr^{-1}in the last 5 years) than that found in the eastern states. On average, these western states have seen a ~168% increase in calcium deposition over the last 17 years. The northwestern region has seen a 54 % increase, and the midwestern region a ~24% increase in Ca^{2+} deposition.

The industrial sources of calcium, namely coal-fired power plants and cement manufacturing plants, are dominantly located in the eastern portion of the country (EPA, 1993, 2012). There are 173 coal burning power stations in the mid-western (108) western (55) and northwestern states (10). In stark contrast there are 456 stations in the remaining eastern states accounting for 70% of the coal power production (EPA, 2012). A similar pattern is observed for cement production plants. Due to proprietary data reason, we cannot divide the exact number of plants by state; however the breakdown available is still informative. There are 4 active plants in Washington and Oregon, 13 in Colorado, Wyoming, Arizona, New Mexico, Idaho, Montana, Nevada, and Utah, 13, in Kansas, Iowa, Nebraska and South Dakota, and 58 in the eastern states that equate to 6x the production of both the northwestern and inter-mountain west states combined (USGS, 2012). Based on these spatial factors,

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the locations of the industrial sources of calcium (eastern US) are not spatially consistent with the regions of observed high Ca^{2+} deposition (western and midwestern US). Instead, the regions with the largest and statistically significant increase in the dissolved fraction of Ca^{2+} encompass or border the most prominent mineral aerosol source regions in the country: the southwestern and western deserts and the agricultural regions of the Southern Great Plains and Columbia River Basin.

3.3 Temporal patterns, peak years and seasonality of potential sources

Temporal patterns in the potential for fly-ash emission for coal-fired power plants in the regions of interest are inconsistent with the Ca^{2+} deposition trends observed. In the inter-mountain west region, coal MWh production statistics show an increase from 1998 to 2000 followed by a slow decline through the last decade (Figure 3). In the midwest, production slightly increased to 2004 where it stabilized until a decline in 2009, and in the northwestern region, production increased from 1999 to 2004 before declining through 2009. Even without considering the increases in emissions-abatement technology, the trends in coal production (and thus potential fly-ash emissions) do not match the trends and strong interannual variability observed in the Ca^{2+} deposition record for the inter-mountain west, midwest, and northwest. Similarly with the cement production statistics, here in all regions we see a slight increase from 2003-2007, followed by a decline through 2010 in all regions.

The recent trends in regional fire activity do not match the trends and interannual variability in Ca^{2+} deposition. Wild land fire acreage burned does not show a strong directional increase over the 1994-2010 period in the northwest or inter-mountain west, but shows a small increase to 2009 in the midwest. Further, the largest fire years do not correspond with the highest levels of Ca^{2+} deposition in any of the three regions. For example in the northwest, the period of peak burned acreage in 2006 and 2007 was a period of relatively low Ca^{2+} deposition as compared to adjacent years. (Figure 2, Figure 3). Further, studies of post-fire Ca^{2+} deposition rates have measured an increase in wet + dry deposition by only 29 mg m^{-2} yr^{-1} in areas immediately downwind of wildfires (Clayton 1976, Lewis 1974), this rate does not seem sufficient to raise the annual wet deposition rate by an average of 400 mg m^{-2} yr^{-1} as we have seen in some areas of the west (Supplementary Table 1). These observations indicate that although forest fires can emit calcium to the atmosphere, observed trends in forest fires are insufficient to explain the magnitude of observed changes in Ca^{2+} deposition.

Emissions of Ca^{2+} from the wind erosion of soils in the inter-mountain west should show strong spring and late winter peaks because dust emission events more commonly occur during these periods due to high winds (Reheis and Kihl, 1995, Reheis and Urban, 2011, Hahnenberger and Nicoll, 2012; Steenburgh et al., 2012).Whereas, dust emissions from the northwest and midwest tend to be smaller during winter largely because surfaces may be frozen or snow covered, however exposed fallow fields in winter may still be subject to erosion. The observed seasonal variations in Ca^{2+} deposition are consistent with soil-erosion sources. Seasonal analyses of the NADP Ca^{2+} deposition data show that the recent trend in the inter-mountain west states is largely related to increased late winter and spring dust deposition, with spring deposition dominating in magnitude for the time period analyzed (1994-2010)(Table 3). In contrast, in the midwest and northwestern regions, the seasons with the highest deposition rates are spring and summer, though both regions indicate that increased deposition rates are occurring in the winter and spring moths (Table 3).

3.4 Correlations of Ca deposition with climate factors

Calcium deposition was best explained using a combination of wind and drought variables across the three regions. Regression results, the squared correlation coefficient (r^2) and the *p*-values, are presented in Table 4. The number of high-wind events increased across all three regions during the time period investigated in this study. However, the PDSI as a measure of aridity only indicated increase drought frequency/intensity in the midwest and northwest. Despite some years of drought (2002-2003), the PDSI in the inter-mountain west region did not exhibit systematic drying over the last 17 years, suggesting that drought cannot explain recent increases in soil susceptibility to erosion in this area. In both the midwest and the northwest the correlation was improved by including both the number of wind events >25.7 m s^{-1} and PDSI in the correlation, though in the northwest wind was not a significant predictor of Ca^{2+} deposition on its own (Table 4). In the inter-mountain west, including the PDSI only marginally improved the correlation and decreased the significance. The PDSI and wind least-squares regressions correlated to the observed Ca^{2+} deposition rates in the midwest with an r^2 of 0.62 and in the northwest and inter-mountain west 0.41 and 0.55 (Figure 4).

Other factors that would be expected to align with an increase in soil aerosols include the incidences of reported dust storms and increased low-visibility events. We used observations from NOAA to test our working hypothesis that increases in Ca^{2+} deposition were due to increases in regional dust emissions. According to the NOAA storm database (NOAA, 2012), incidences of reported dust storms have increased in recent years (Figure 5). Because reports of dust storm events may not be a true measure of mineral aerosol emissions, we also evaluated low-visibility events from sites near our three regions of interest. As visibility measurements are taken every hour, these data represent a continuous measurement of potential dust-storm activity. In all three regions we see large increases in the number of low-visibility events that match general trends in Ca^{2+} deposition for our three regions (Figure 6). These independent observations support our working hypothesis that the observed increases in Ca^{2+} deposition in the Western US are caused by an increase in low-frequency, high-intensity dust storms as well as by higher frequency, lower intensity emissions that altogether suggest an increase in mineral aerosol loading to the atmosphere resulting in diminished visibility.

3.5 Precipitation alkalinity

Based on charge-balance equations, we calculated the fraction of precipitation alkalinity changes attributable to increases in Ca^{2+} concentration over the last 17 years, and compared this fraction to the fraction of alkalinity change attributable to declines in acid anions specifically NO_3^- and SO_4^{2-} (see Table 5 and Supplementary Figures 3-4). In all three areas with significant increases, Ca^{2+} contributes a large fraction to the overall change in alkalinity. In the inter-mountain west region, the ~168 % increase in wet calcium deposition over the last 17 years, provides an additional 442 μeq m^{-2} yr^{-1} acid-neutralizing capacity, with increase in Ca^{2+} accounting for 52% of the regional alkalinity change and as much as ~70% of the alkalinity increase at individual sites (Supplementary Table 1, Supplementary Figure 2).

4.0 Discussion

Of the potential sources we considered (industrial, forest fires, sea-salt, and soil dust); soil-dust emissions are the only source that is consistent with the spatial distribution, temporal trends, and inter-

annual variability of the observed Ca^{2+} in precipitation. Further, the NADP data indicate a nationwide increase in precipitation pH and alkalinity even in areas where acid emissions have not decreased substantially or have, in some cases increased (NADP, 1994-2009). This inconsistency between acid emissions and precipitation alkalinity strongly suggests that the pH and alkalinity of precipitation in parts of the western U.S. are increasing and the reason for this increase is most likely increased mineral-aerosol emissions. This interpretation is in line with Young's observation that SO_4^{2-} in western precipitation is more closely correlated with calcium than H^+ concentrations (Young et al., 1988). Furthermore, calcium isotope data suggest that atmospheric Ca^{2+} in California is primarily derived from a calcium carbonate source (Schmitt and Stille, 2005), despite the proximity to industrial, forest fire, and sea-salt sources.

The highest rates of Ca^{2+} deposition in this study are found near the southwestern deserts, specifically the Mojave Desert and the arid regions in the Four-Corners area that commonly emit dust under the influence of strong southwesterly winds, especially during late winter and spring (Figure 1)(Painter et al., 2007). Dust emissions in western U.S. deserts have been closely linked to anthropogenic factors in site level studies (Belnap and Gillette, 1998; Belnap et al., 2009; Munson et al., 2011). These areas are sensitive to human disturbance because soil crusts are destabilized through foot, vehicle, and livestock traffic, as well as through industrial and agricultural activities that greatly increase the likelihood of wind erosion (Belnap and Gillette, 1998). In the last 17 years, the population in the Mojave Desert has increased by 22% and on the Colorado Plateau by 29%, and recreational use of off-road vehicles has increased three to four fold (Cordell et al., 2008). In addition, the number of wells drilled for natural gas extraction quadrupled from 2000 to 2009 in the Colorado Plateau (COGCC, 2011; Utah.gov, 2011). Moreover, many natural factors contribute to western desert dust emissions, including interacting conditions of drought and wet periods and responses of vegetation dynamics (e.g., Urban et al., 2009) and dry lakes (Reynolds et al., 2007). Collectively, these changes provide an illustration of a region that is undergoing rapid and widespread changes that have the potential to cause changes in wind erosion of soil. The relative importance of each of these factors to dust generation remains uncertain and is an important area for future work.

Although much of the increase in Ca^{2+} deposition can be explained by climatic factors, such as transport and aridity, a large proportion of the variance remains unexplained. An unaccounted for variable in our analysis is the disturbance due to human land-use change, which can significantly increase soil mobility. Although it is very challenging to quantify the impact of human land-use change on soil properties, in the midwest Lee et al., (2012) show that modern dust emissions are strongly tied to human modifications where the cultivation of land has caused otherwise stable surfaces to erode. Agricultural practices and extensive loess deposits in both the midwest and northwest are known to contribute to windblown dust (Claiborn, 1998; Lee et al., 2012). Crop type affects erosion potential through soil exposure and the modification of soil properties, as well as through the duration of fallow periods (Lee et al., 2012). Active farm operations, such as plowing, also contribute to mineral-dust formation (Carroll, 1997). It has been estimated that soil erosion during wind storms in the northwest can reach 100 Gg day^{-1}, which is approximately 1% of the total global dust emission rate (Claiborn, 1998).

Our analysis identifies processes that are potentially controlling the chemistry of precipitation over vast areas of the western U.S. that may not be captured effectively by current monitoring networks. In the U.S. there is a spatially distributed network for monitoring of atmospheric particles

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less than 10 micrometers in diameter PM_{10}. Such monitoring does not appear to show increases in dust deposition that we report here and instead record downward trends in $PM_{2.5}$ and PM_{10} in western regions (Tong et al., 2012). A similar pattern is observed in most U.S. urban locations where the concentration of these particles has fallen over the past decade as the result of air quality regulations (Hand, 2011). Despite these trends in sub 10 micrometer particle deposition, atmospheric mineral aerosols – desert dust – often include a large fraction of particles larger than 10 micrometers particularly in and around desert source regions (Lawrence and Neff, 2009). At present, these particles are not routinely measured in U.S. monitoring networks and measurements of total particle concentrations in areas of Southeastern Utah and western Colorado, particles larger than PM_{10} dominate total atmosphere particle loads (Neff et al., *in review*). These particles are not routinely measured in monitoring networks and may explain the lack of correspondence between this study and other studies of aerosol concentrations. It is also important to note that the bulk of the desert dust in Utah/Colorado region is derived from local rather than far travel (Asian) sources (Neff et al., 2008; Lawrence et al., 2010).

4.1 Implications of increased dust

An increase in the concentration of atmospheric aerosols can have numerous effects on the chemistry of the atmosphere as well as on the biogeochemistry of downwind ecosystems. Dust-associated base-cations can add buffering capacity to ecosystems, increase surface-water pH, and replenish base-cations that have been lost due to years of chronic acid deposition (Likens et al., 1996; Rhoades, 2010). It is worth noting that the NADP was not set up to capture dust chemistry signals. The network measures wet-only deposition and there are no sites in some prominent dust emission and deposition regions, e.g southern Nevada. Thus our analysis likely represents a conservative estimate on the contribution of base-cations in many regions, especially areas where dry deposition is high.

In addition to neutralizing airborne acids in areas affected by acid rain, the emission and deposition of dust has the potential to impact source and sink ecosystems. The potential benefits of increasing alkalinity in precipitation are counterbalanced by the negative implications of soil erosion in source regions. Wind erosion preferentially removes the fine fraction of soils, which contains most of the soil's nutrients, cation-exchange capacity, and water holding capacity. As a result erosion can impoverish source region soils while enriching downwind areas (Pye and Vitousek, 1985; Reynolds et al., 2006; Ballantyne et al., 2010; Lawrence et al., 2011). Dust may also contain a variety of associated contaminants (e.g. pesticides, fertilizers, heavy metals, and industrial compounds) and nutrients that can have negative effects, especially in areas that previously received very little atmospheric input. For example, in the Sierra Nevada lakes of Spain, responses to elevated dust inputs were more pronounced in the more oligotrophic lakes (Morales-Baquero et al., 2006). Dust can also act as vector for the transport of pathogens that can affect human and ecosystem health (Griffin et al., 2001). For example, there have been large increases in the incidence of Valley Fever (*Coccidioidomycosis*) in southwestern states (CDC, 2013) from the mid-1990s to present. This disease is caused by the inhalation of a soil born fungus known as *Coccidiodes*.

4.2 Potential future dust scenarios

Long-term increases in dust generation are possible in response to projected changes in climate and land-use. We have demonstrated that dust emissions are likely increasing across regions of the

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western U.S. and that the frequency of high wind events appears to be playing a role in the erodibility and transport of soils as dust. We also suggest that land-use change may be playing a role in increasing dust emission, but more work is needed to make this link more definitive. Most model predictions suggest that continued warming will result in an increase in drought frequency over much of the southwestern U.S. (Seager, 2007). This condition would lead to drier soils related to the enhanced evapotranspiration that accompanies warmer temperatures (Seager et al., 2007, Cayan et al., 2010). Therefore increased drought frequency and greater soil aridity have the potential to increase dust emissions in the future, which can further contribute to regional aridity (Cook et al., 2009). Although our restricted analysis shows no discernible trend in drought over the southwestern region investigated here, our analysis does suggest that the southwestern region may be very susceptible to soil erosion when drought conditions are combined with an increasing frequency of high wind events.

In summary, dust emissions have increased over the last 17 years over large regions of the western US due to an interaction between wind speeds, drought cycles, and potentially changes in human land uses. This pattern suggests that dust may be an underappreciated contributor to the atmospheric particulate load in some U.S. regions near deserts or large agricultural areas. Changes in dust deposition have a clear impact on ecological systems and may also have broad implications for air quality, climate processes and human health. These results highlight the need to expand the measurements and locations of the atmospheric deposition network while focusing on the factors that influence dust emission, transport, and deposition

Tables

Table 1. The number of wind events and the PDSI statistics used in the dust generation statistical model. Note that a negative PDSI indicates drier conditions.

	Number of Wind Events > 27.5 m s-1			Regional PDSI		
Year	Colorado Plateau and Mojave Desert	Columbia Basin (east WA)	Southern Great Plains	Colorado Plateau and Mojave Desert	Columbia Basin (east WA)	Southern Great Plains
1994	5	0	1	-1.96	-2.17	-0.02
1995	20	3	0	0.19	2.52	0.62
1996	52	6	4	-3.16	3.63	-0.69
1997	25	11	5	-1.26	4.32	2.60
1998	31	4	2	1.14	0.44	0.31
1999	45	12	3	-1.51	-0.75	0.50
2000	41	10	2	-3.27	-1.14	-0.89
2001	36	12	6	-1.78	-2.71	1.27
2002	40	21	7	-4.88	-2.25	0.38
2003	25	13	5	-3.66	-1.24	-0.54
2004	19	10	7	-1.74	-1.77	1.71
2005	30	12	9	3.29	-2.42	1.14
2006	34	25	13	-2.20	0.84	-1.98
2007	59	13	3	-2.96	-1.16	1.94
2008	62	13	19	-0.95	-1.86	-0.51
2009	72	16	18	-2.07	-0.82	-0.51

Table 2. Summary of regional changes in precipitation chemistry analysis (See Supplementary Table 1 for individual site data). Bold indicates a significant result.

Region	% change in Ca^{2+} deposition (range)	Mann-K p-value on regional Ca^{2+} trends	Sites with increased pH (p <0.05)	Sites with increased Alkalinity (p <0.05)
Inter-Mnt West (n=22)	168 (-17 to 364)	**0.0003**	22 (20)	22 (20)
Northwest (n=9)	54 (-19 to 152)	**0.0045**	9 (7)	8 (6)
Southwest (n=14)	13 (-75 to 91)	0.7731	14 (13)	14 (11)
Westcoast (n=11)	44 (-15 to 133)	0.1275	11 (8)	10 (5)
Midwest (n=15)	24 (-39 to 66)	**0.0235**	15 (14)	15 (12)
Northern (n=17)	5 (-33 to 53)	0.3434	17 (17)	17 (16)
Northeast and East (n=45)	8 (-42 to 50)	0.4338	45 (45)	45 (44)
South and Southeast (n=37)	-4 (-51 to 56)	0.7108	37 (33)	33 (24)

Table 3. Statistics for the seasonal analysis of trends for the three significant regions, the inter-mountain west, the northwest, and the midwest.

		Fall	Spring	Summer	Winter
Inter-Mnt West (n=22)	Avg Dep Rate (mg m^{-2} yr^{-1})	34	79	33	24
	Inc Dep Rate (%)	61	1,076	28	155
	Number significant (Mann-Kendall)	4	7	4	1
Northwest (n=9)	Avg Dep Rate (mg m^{-2} yr^{-1})	9	22	16	6
	Inc Dep Rate (%)	20	797	-28	350
	Number significant (Mann-Kendall)	0	5	1	3
Midwest (n=15)	Avg Dep Rate (mg m^{-2} yr^{-1})	38	83	84	23
	Inc Dep Rate (%)	46	41	39	99
	Number significant (Mann-Kendall)	1	0	2	5

Table 4. Dust generation model statistics. Statistical model results are analyzed in comparison to Ca^{2+} deposition trends for each region. Both wind and PDSI data are used to create the trends in Figure 4, the statistics for these trends are shown in bold. We include the Akaike's Information Criterion (AIC), a relative goodness of fit measurement where lower numbers indicate an improved model.

	Wind > 25.7 m s^{-1}			PDSI			Wind > 25.7 m s^{-1} + PDSI		
	r^2	p-value	AIC	r^2	p-value	AIC	r^2	p-value	AIC
Inter-Mnt West	0.530	<0.001	18.10	0.030	0.500	29.56	**0.550**	**<0.01**	**19.4**
Midwest	0.370	0.012	16.29	0.490	<0.01	12.87	**0.620**	**<0.01**	**10.1**
Northwest	0.120	0.188	20.11	0.350	<0.05	15.17	**0.410**	**<0.05**	**15.8**

Table 5. Summary of regional changes in calculated alkalinity.

Region	Fraction of Alkalinity change due to Ca^{2+} (range)	Fraction of Alkalinity change due to NO$_3^-$ (range)	Fraction of Alkalinity change due to SO$_4^{2-}$ (range)
Inter-Mnt West (n=22)	0.52 (0.02 to 0.79)	0.11 (-0.08 to 0.26)	0.18 (-0.05 to 0.46)
Northwest (n=9)	0.22 (-0.04 to 0.61)	0.13 (0.03 to 0.37)	0.04 (-0.35 to 0.38)
Southwest (n=14)	0.18 (-0.18 to 0.52)	0.14 (-0.11 to 0.32)	0.21 (-0.08 to 0.57)
Westcoast (n=11)	0.12 (0.07 to 0.37)	0.13 (-0.01 to 0.29)	0.08 (-0.14 to 0.67)
Midwest (n=15)	0.07 (0.19 to 0.41)	0.29 (0.18 to 0.42)	0.40 (0.08 to 0.63)
Northern (n=17)	0 (0.24 to 0.26)	0.31 (0.12 to 0.46)	0.30 (0.02 to 0.54)
Northeast and East (n=45)	-0.01 (-0.07 to 0.12)	0.32 (0.15 to 0.41)	0.49 (0.22 to 0.63)
South and Southeast (n=37)	-0.04 (-0.45 to 0.15)	0.20 (-0.03 to 0.31)	0.40 (0.15 to 0.59)

Figure Captions

Figure 1. Changes in Ca²⁺ deposition across the U.S. plotted by the magnitude of the increase in mg m⁻², and by percent increase from 1994 values. Black dots represent sites where Ca²⁺ depositon decreased.
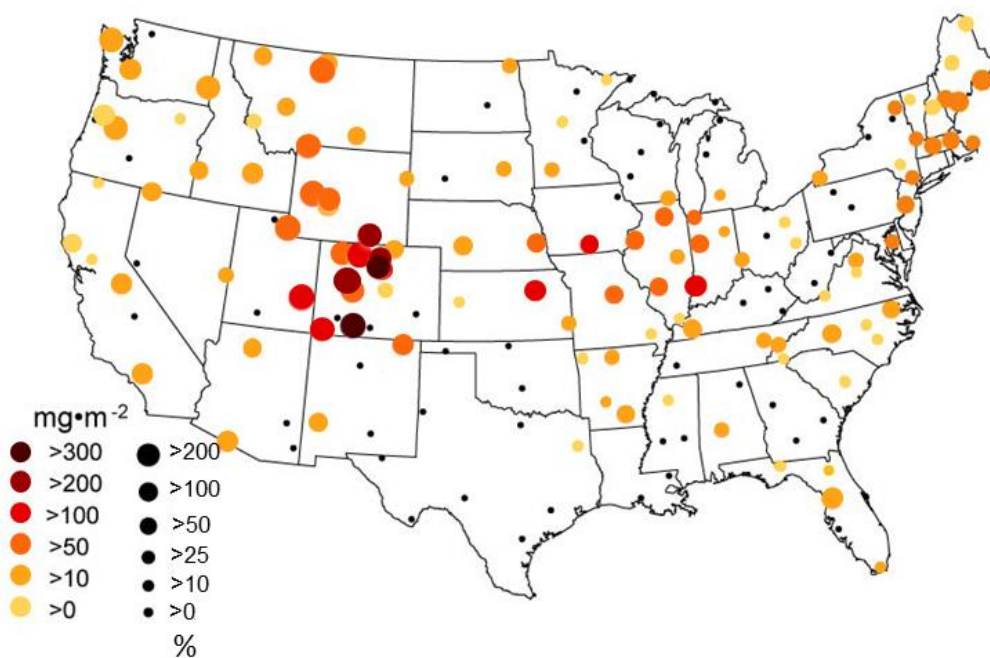
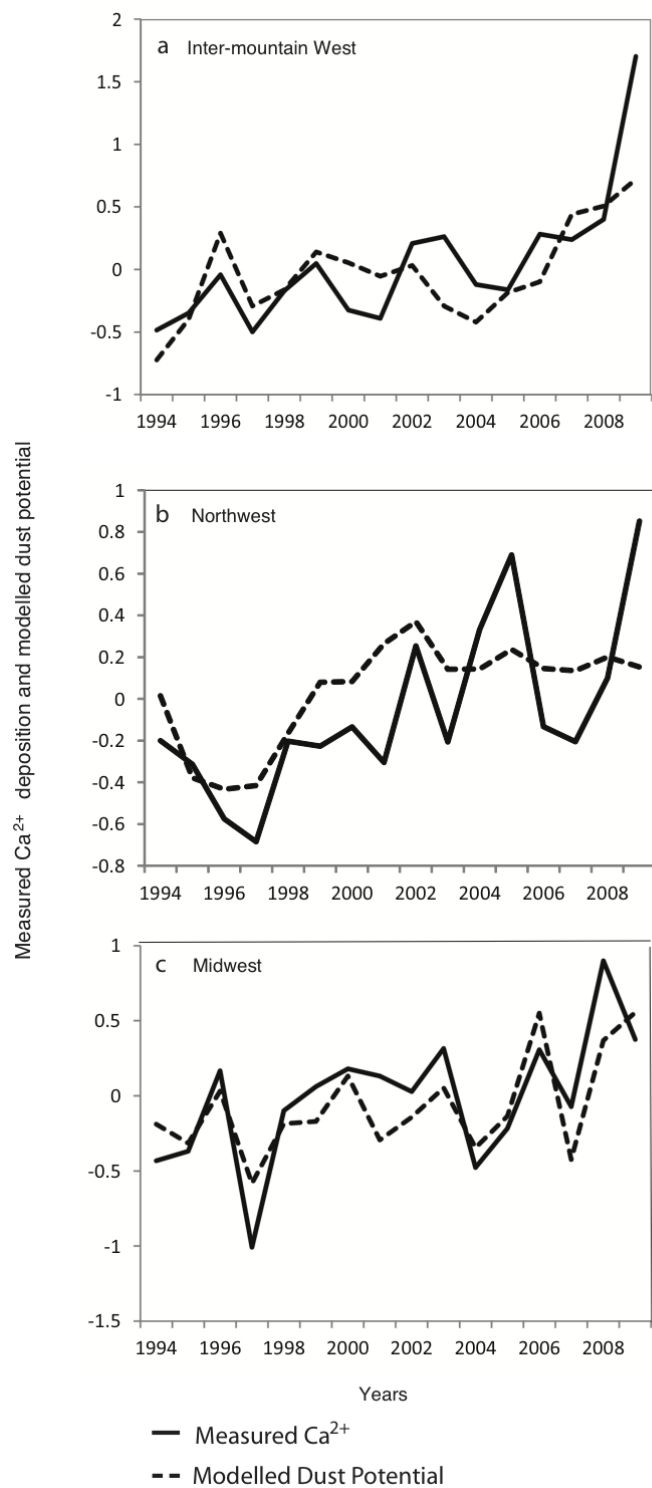Figure 2. Composite Ca²⁺ deposition trend a) Inter-mountain west Region, b)Northwestern region, c) Midwestern region. Note units in the Y-axis have been normalized. Shaded area represents the 95% bootstrap confidence intervals of the composite trends and the dashed line represents the significant trend as identified by Mann-Kendall trend analysis (see Methods).
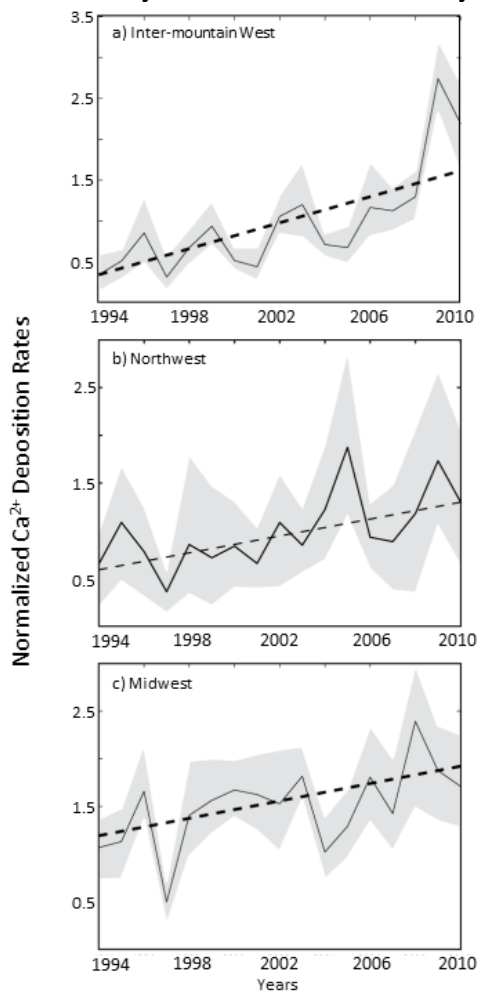
Figure 3. Recent trends in a) coal fired power plant production (MWh), b) trend in cement production by region
   c) trends in forest fire in acreage burned by region
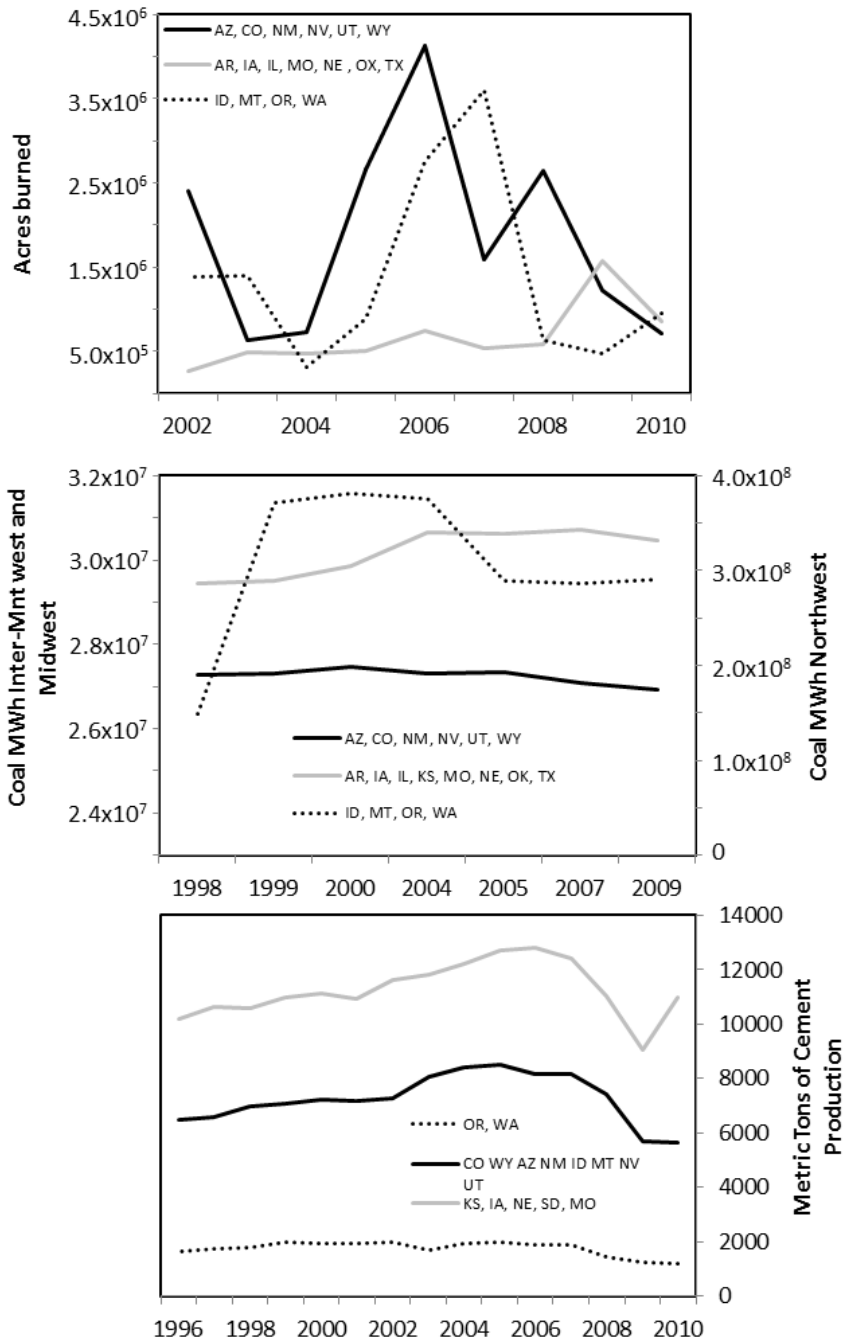
Figure 4. Composite Ca^{2+} deposition trends compared to modelled dust generation potential. a) Inter-mountian west region, r^2=0.55, b) Northwestern region, r^2=0.41, c) Midwestern region, r^2=0.62. Note units in the Y-axis have been normalized.

Figure 5. Composite trends of Ca^{2+} deposition rates in a) the Inter-Mountain West b) the Northwestern, and c) the Midwestern regions of the Continental US. Shaded area represents the 95% bootstrap confidence intervals of the composite trends and the dashed line represents the significant trend as identified by Mann-Kendall trend analysis (see Methods). Bar plots indicate the number of NOAA reported dust-storm events for each region
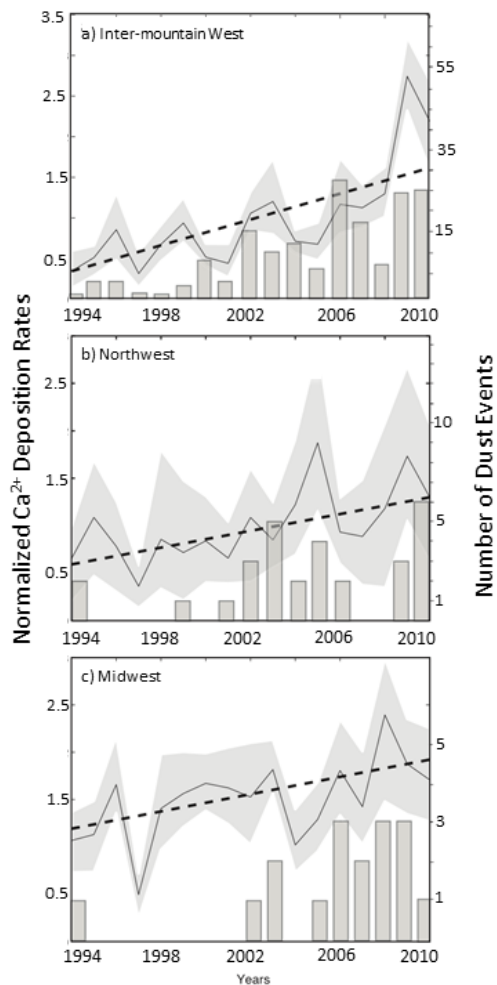
Figure 6. Low visibility events by year for sites near our three regions of significant increases in Ca^{2+} deposition. Data was obtained from the National Climate Data Center (NOAA, 2012b). Events are defined as a time period during which visibility drops below 6000 and then 3000 meters; the event does not end until visibility increases above 6000 meters, see main text for details.
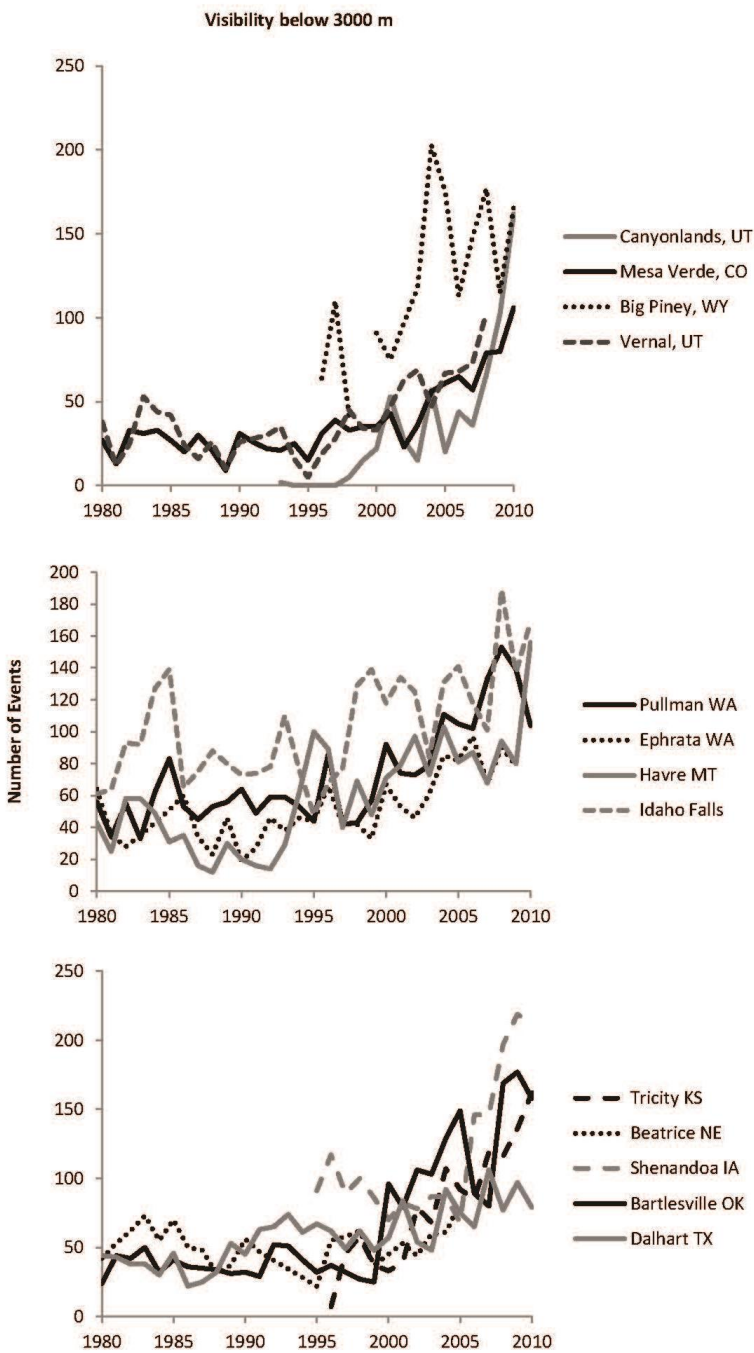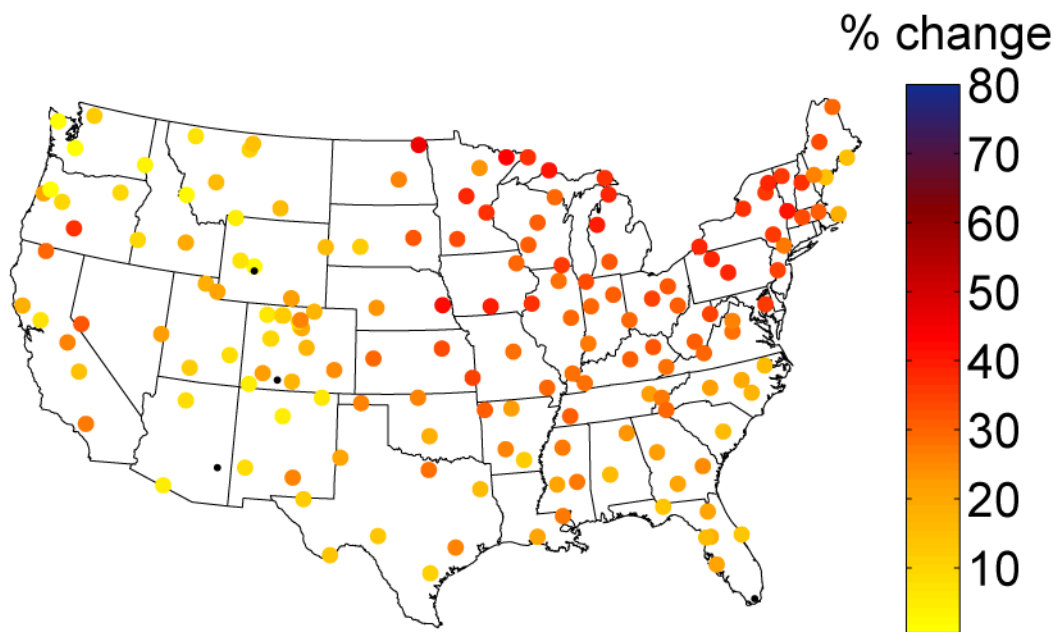
Supplementary Figure 1. Change in alkalinity of precipitation from 1994-2010 reported as μeq.L^{-1}
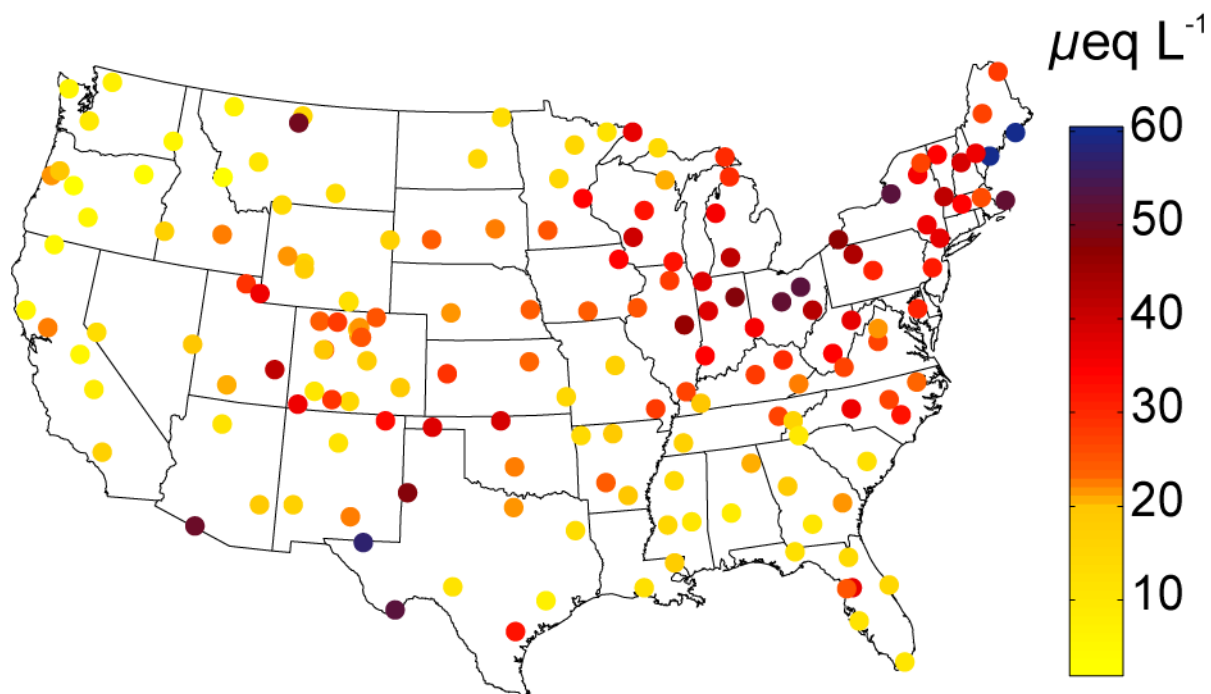
Supplementary Figure 2. Change in alkalinity from 1994-2010 due to increases Ca^{2+} concentration. Black dots represent areas where Ca^{2+} decreased over the time period.
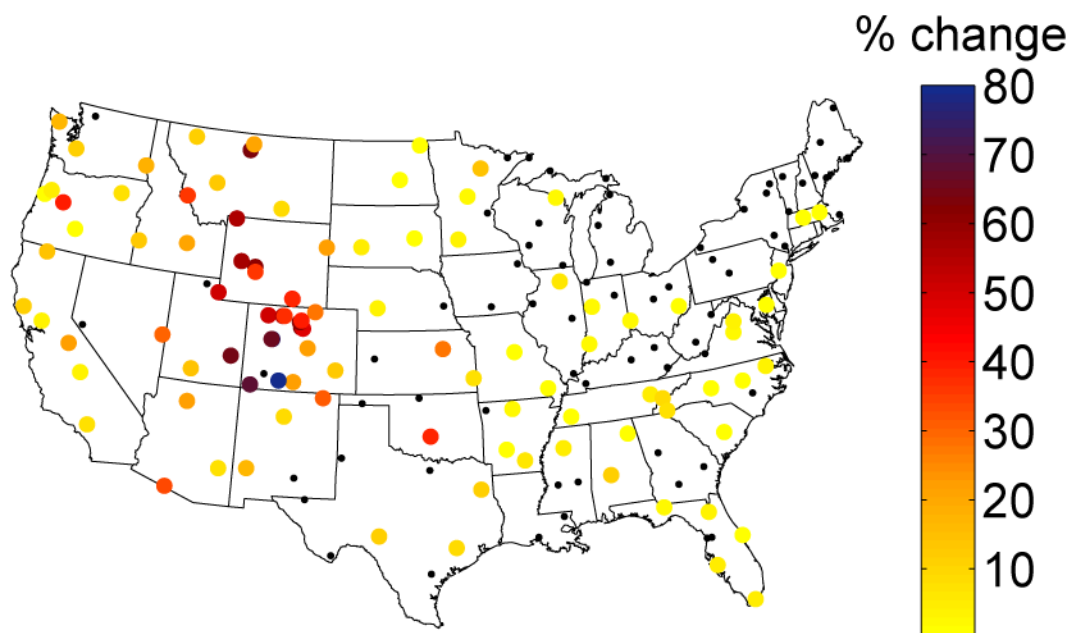
Supplementary Figure 3. Change in alkalinity from 1994-2010 due to decreases in NO_3^- concentrations. Black dots represent areas where NO_3^- deposition increased over this time period.

Supplementary Figure .4 Change in alkalinity from 1994-2010 due to reductions in SO_4^{-2} concentration. Black dots represent areas where SO_4^{-2} concentrations increased over this time period.
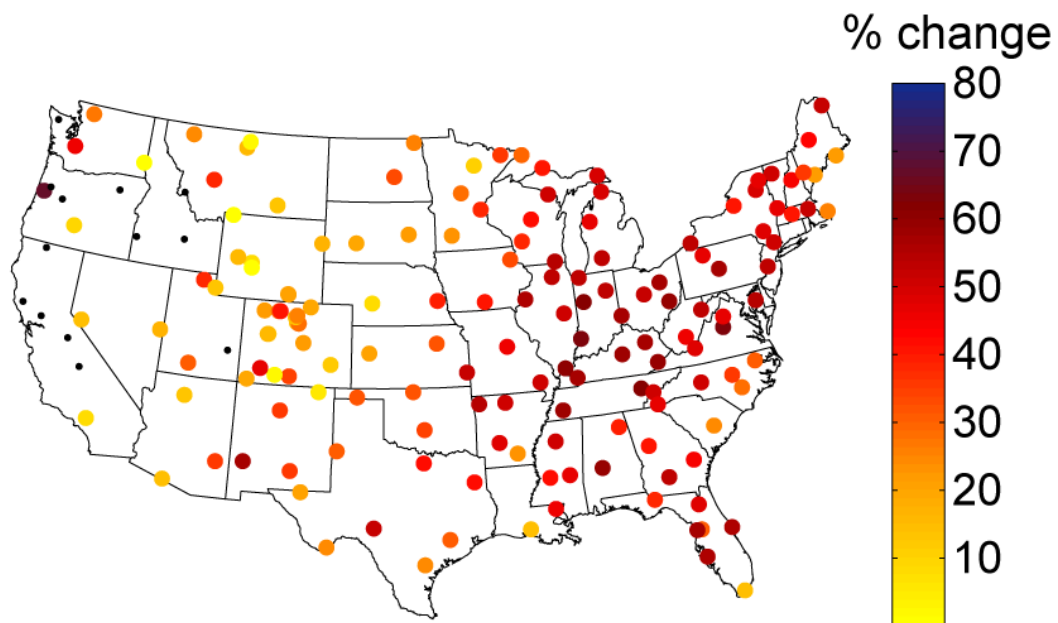
Acknowledgments

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