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# Effect of Climate Change between 1984 and 2007 on Precipitation Chemistry at a Site in Northeastern USA

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Climate change predictions for the northeastern US call for an increase in tropical storms and a decrease in extra tropical cyclones including continental storms. We ran 24-h back trajectories for each precipitation event that occurred at the Cary Institute of Ecosystem Studies in southeastern New York, USA from 1984 to 2007 and analyzed precipitation chemistry as well as air mass position 24 h prior to the onset of each precipitation event. The results showed an increase in marine precipitation and a slight but statistically insignificant decrease in continental precipitation during the 1984–2007 period. The chemistry of precipitation from the two directions was quite different: marine storms were higher in  $\text{Na}^+$  and  $\text{Cl}^-$  but lower in solutes associated with acid precipitation ( $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ ). Annual mean concentrations of acid precipitation solutes declined for storms from both directions during the period. We used a simple mixing model based on the current rates of increase and decrease of marine and continental precipitation respectively to show that chemical changes in precipitation resulting from the shift in storm tracks are small compared to chemical changes due to emissions reductions.

## Introduction

Climate change predictions for the northeastern United States call for the following: increased precipitation, an increase in the frequency of nor'easters (large, coastal storms that usually occur during winter), an increase in the proportion of winter precipitation that falls as rain, possibly an increase in tropical storms, more flooding events in winter months, and more frequent and longer growing season droughts (1). A northward shift in the jet stream has already occurred, resulting in a decrease in extra tropical cyclone (ETC) frequency at mid latitudes (30–60 deg N) with a corresponding increase in ETC activity at high latitudes (60–90 deg N) since 1959 (2). These changes in the jet stream have already and perhaps will continue to reduce the number of midlatitude storms and increase the number of subtropical storms (3). A decrease in the frequency of extra tropical cyclones and/or an increase in the frequency of tropical or marine storms could result in a subsequent increase in the proportion of precipitation in the northeastern US from marine sources. To date, precipitation events in the northeastern US have been reported to

come predominantly from the west and carry acidic solutes from the area of the country with the highest density of coal burning power plants (4, 5). Acid precipitation has declined in the northeastern US largely due to the decline in emissions of acid precipitation precursors from power plants, which are controlled by the US Clean Air Act (6–9). A decrease in the amount of precipitation from the continental US and an increase in precipitation of marine origin could alter precipitation chemistry by decreasing acid precipitation solutes more than expected from controls on precursor emissions alone.

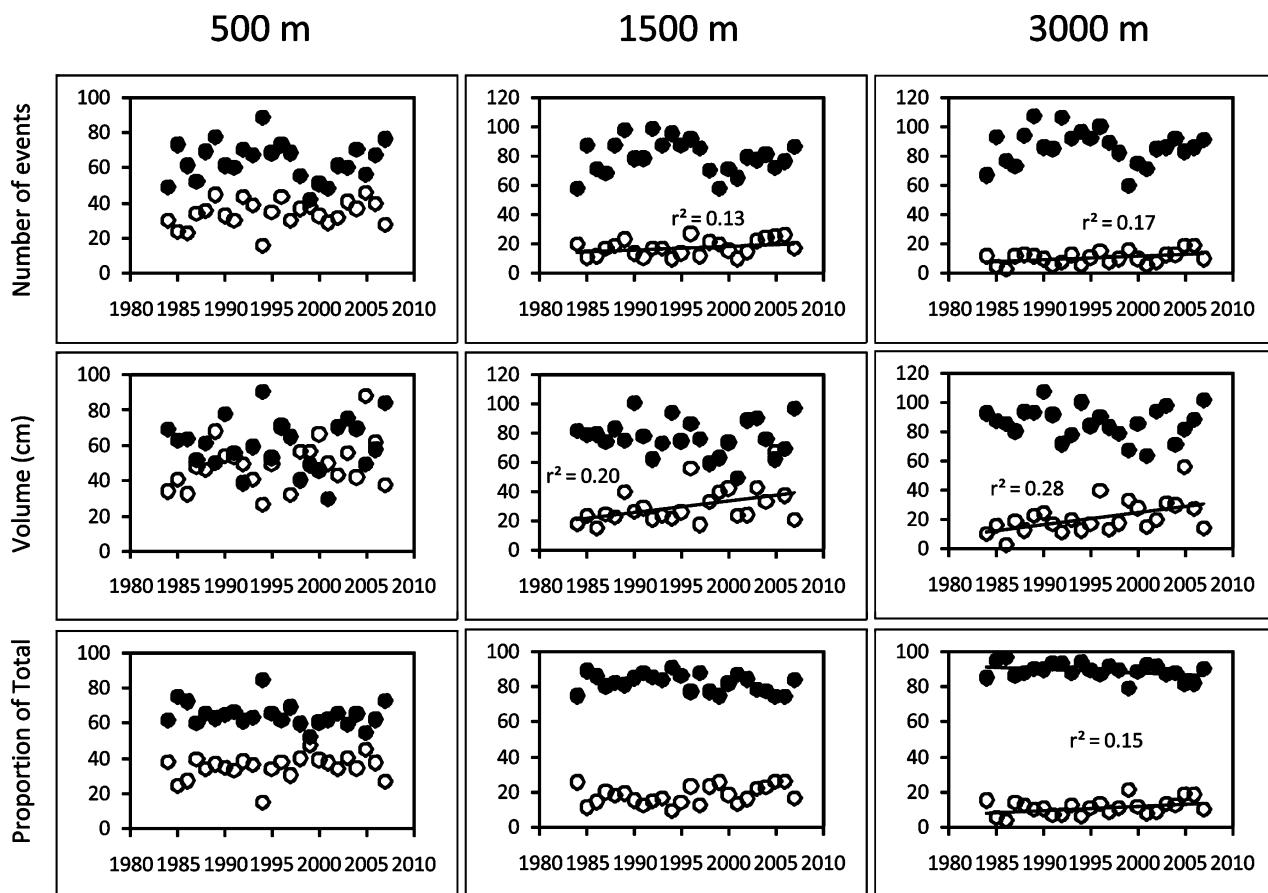
We used the National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (10) to estimate backward trajectories of air masses preceding the onset of each precipitation event that occurred at the Cary Institute of Ecosystem Studies in Millbrook, New York between 1984 and 2007. The position of air masses 24 h prior to the onset of each event together with the chemistry of each event allowed us to examine temporal changes in storm direction as well as the effect of such changes on precipitation chemistry during this period. This paper addresses three questions relative to our precipitation monitoring site: 1. Has there been a change in the direction from which air masses originate prior to precipitation events? 2. Has precipitation chemistry changed as a result of a change in the origin of air masses prior to precipitation events? 3. What impact might these changes have on future trends in acid precipitation?

## Materials and Methods

**Precipitation Sampling.** Precipitation samples were collected from 1984 to 2007 using the wet side of an Aerochem Metrics automatic wet–dry collector. The collector uses a moisture sensor that causes a motor to remove a cover from a clean bucket when it senses precipitated moisture. Samples were collected on an event basis; an event was defined as continuous precipitation that had not been interrupted by more than a 6-h hiatus. At the end of an event, the sample was collected, weighed, and transferred to a labeled clean sample bottle. When the sample was frozen, it was allowed to sit at room temperature until the entire sample was melted before analysis. Samples were analyzed at the Rachel L. Carson Analytical Facility of the Cary Institute of Ecosystem Studies for pH,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{PO}_4^{3-}$ ,  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{K}^+$ . Nitrate,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$  were measured with a Dionex DX500 ion chromatograph (AS4A and AG4A columns, micromembrane suppressor), and  $\text{NH}_4^+$  was measured by continuous flow analysis (Alpkem Flow Solution III, phenate method). If the quantity of sample was insufficient for all of these analyses, as many of the analyses as possible were completed with preference given to pH,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  in that order. In 1999 we changed the sample-handling protocol as follows. A 60-mL aliquot of sample was preserved with approximately 0.08 mL of chloroform and refrigerated. This aliquot was analyzed for  $\text{PO}_4^{3-}$ ,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$ . Comparisons of  $\text{PO}_4^{3-}$ ,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  were made for one year between samples treated with chloroform and samples left untreated. There was no significant difference between treated and untreated samples for  $\text{NH}_4^+$  and  $\text{NO}_3^-$ . Precipitation volume was determined using the wet–dry collector (1984–1987) and a Belfort Instrument Co. universal recording rain gauge (1988–2007).

**Estimation of Event Direction.** Using the NOAA ARL HYSPLIT model (10), we calculated 24-h back trajectories for air masses at 500, 1500, and 3000 m above sea level. We

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**FIGURE 1.** Change over time (1984–2007) in air mass origin at 500, 1500, and 3000 m in the number of events, total volume, and proportion of total number of events that were of marine origin (open symbols) or of continental origin (closed symbols) at the Cary Institute of Ecosystem Studies, Millbrook, NY, USA. Data are based on 24-h back trajectories from onset of precipitation events. Regression lines shown were significant at  $p < 0.10$ .

used the National Weather Service's National Centers for Environmental Prediction (NCEP) the National Center for Atmospheric Research (NCAR) Reanalysis Project data set archived by ARL for meteorological data. The precipitation event location is the Cary Institute of Ecosystem Studies in Millbrook, New York, USA (latitude 41.786 and longitude  $-73.741$ ). We analyzed a total of 2371 events occurring from 1984–2007. Events were considered of marine origin if the air mass was to the east of the Cary Institute or over the Atlantic Ocean 24 h prior to the onset of the event. We considered all other events continental. The 24-h back trajectory end points provided objective, midrange data for analysis of air mass position prior to precipitation onset; analysis of 6-, 12-, and 18-h back trajectory end points yielded the same results.

**Mixing Model.** To help put these changes in context, we created a simple, spreadsheet-based mixing model of  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  concentration using an equation (eq 1) where VWM is volume-weighted mean, and the subscripts M and C refer to events that were of marine origin and continental origin, respectively. We estimated future increases in the volume of precipitation of marine origin as 1 cm year $^{-1}$  and decreases of continental precipitation as 0.19 cm year $^{-1}$ , which were the average rates of increase and decrease, respectively, during the 1984–2007 period. VWM concentrations were kept constant at 2007 levels.

$$\begin{aligned} \text{MixedVWM} &= (\text{VWM}^* \text{volume})_M + (\text{VWM}^* \text{volume})_C \\ &= \text{Total Flux} / \text{Total Volume} \end{aligned} \quad (1)$$

Relationships among variables and trends over time were determined using SAS Version 9.1.3 Service Pack 4 (SAS Institute 2007).

## Results

**Storm Direction.** Overall, there were more events that had air masses that were continental 24 h prior to precipitation onset than marine (Figure 1). For air mass elevations of 500, 1500, and 3000 m above sea level, respectively, there were a total of 1522, 1902, and 2069 events that had air masses that were continental 24 h prior to onset of precipitation. This represented 65%, 82%, and 89% of the total number of events for air mass elevations of 500, 1500, and 3000 m, respectively. The lower proportion of continental air masses at lower air mass elevations may reflect the fact that lower elevation air masses are more subject to shifts in direction because of orographic and other effects than higher elevation air masses, which are more heavily influenced by prevailing westerly winds. Vertical shear between 500 and 1500 m was evident for 20% of the back trajectories and between 1500 and 3000 m for 10% of the back trajectories. Overall the volume of precipitation delivered during continental events was higher than from marine events (Figure 1). However, for nine of the 24 years there was a greater volume of marine precipitation for events that had air masses that were 500 m at the onset of precipitation (Figure 1). Seven of those nine years occurred in the latter half of the study period. There was one year, 2005, that also had a greater volume of marine precipitation for events that had air masses that were 1500 m at the onset of precipitation (Figure 1). In 2005, there were an extraordinary number of tropical storms in the Atlantic basin (28 named storms), which may explain the high volume of marine precipitation in that year. The absolute number of events and total volume of events that were of marine origin significantly increased ( $r^2 = 0.13$ – $0.28$ ,  $p < 0.10$ ) over the 24-year period, 1984 to 2007, for air masses that were at

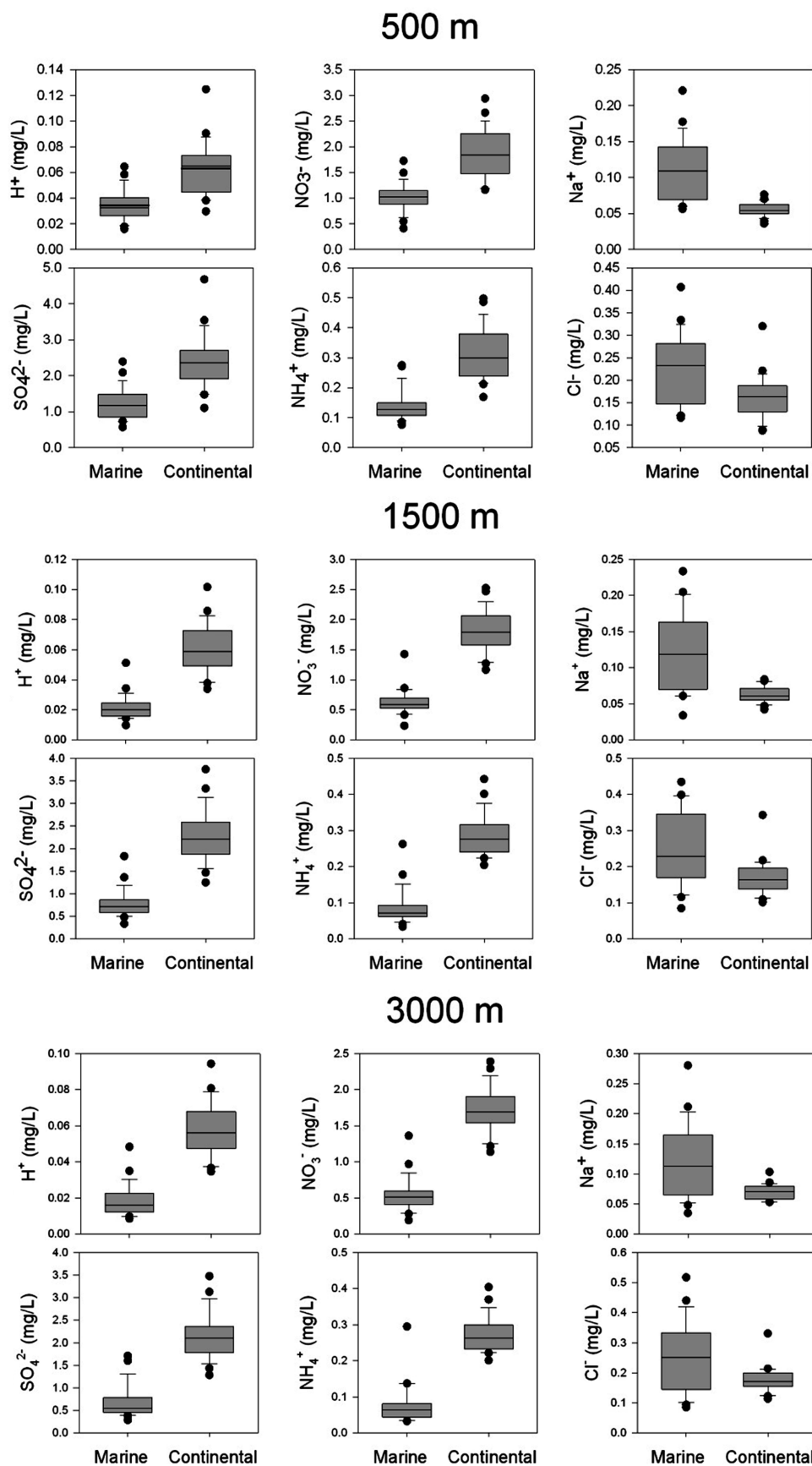


FIGURE 2. Box and whisker plots showing median, 25th and 75th percentiles, 10th and 90th percentiles, 5th and 95th percentiles, and absolute minimum and maximum annual volume-weighted mean concentration (mg/L) of events that were of marine or continental origin during the 24-year period from 1984 to 2007 for air masses at 500, 1500, and 3000 m at the Cary Institute of Ecosystem Studies, Millbrook, NY, USA.

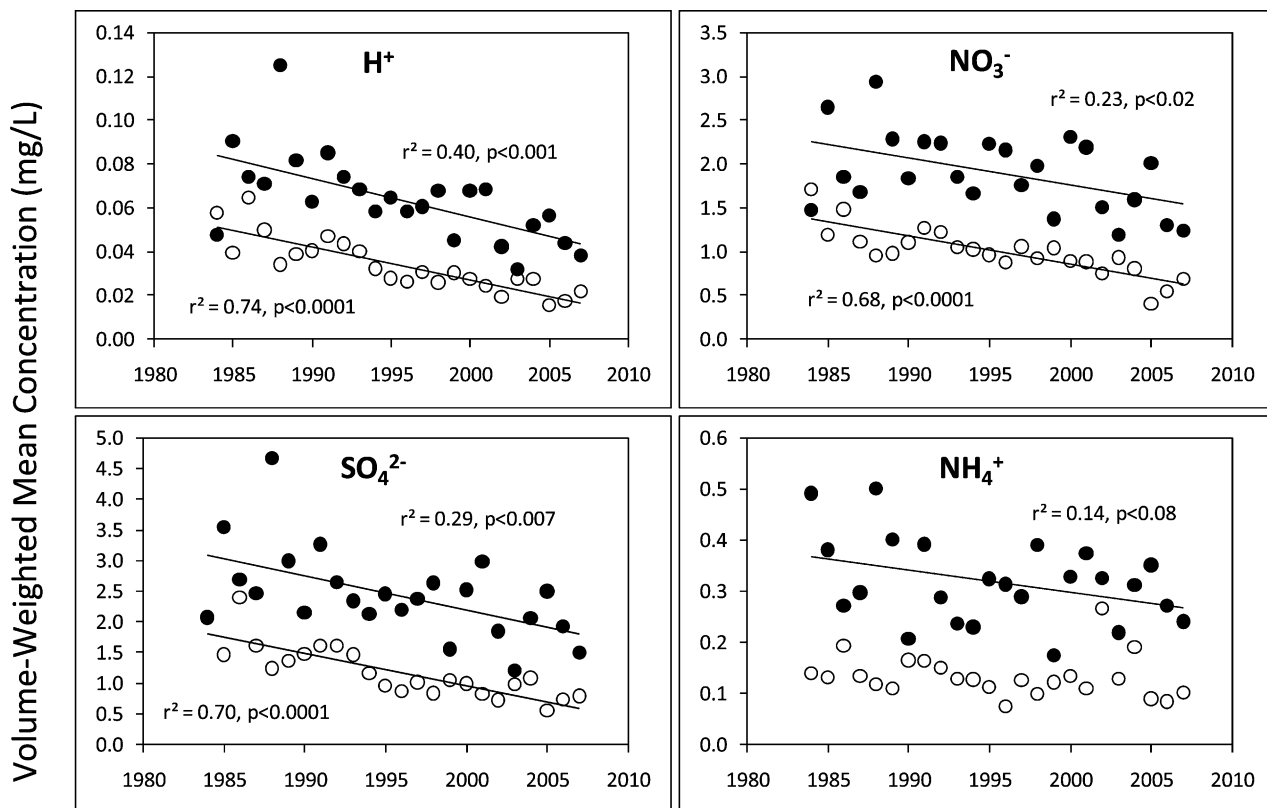


FIGURE 3. Annual volume-weighted mean concentrations of  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  of events that were of marine origin (open symbols) or of continental origin (closed symbols) at the Cary Institute of Ecosystem Studies, Millbrook, NY, USA. Events were considered of marine origin if any of the back trajectories (air mass elevations 500, 1500, or 3000 m) was over the Atlantic Ocean 24 h prior to the onset of the precipitation; all other events were considered continental.

TABLE 1. Slopes, Standard Errors of Slopes, and  $r^2$  Values for Linear Regressions of the Change over Time in Annual Volume-Weighted Mean Concentrations (mg/L) of the Acid Precipitation Solutes  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  for Regressions That Include All Events and Regressions That Exclude Events That Were of Marine Origin at the Cary Institute of Ecosystem Studies, Millbrook, NY, USA<sup>a</sup>

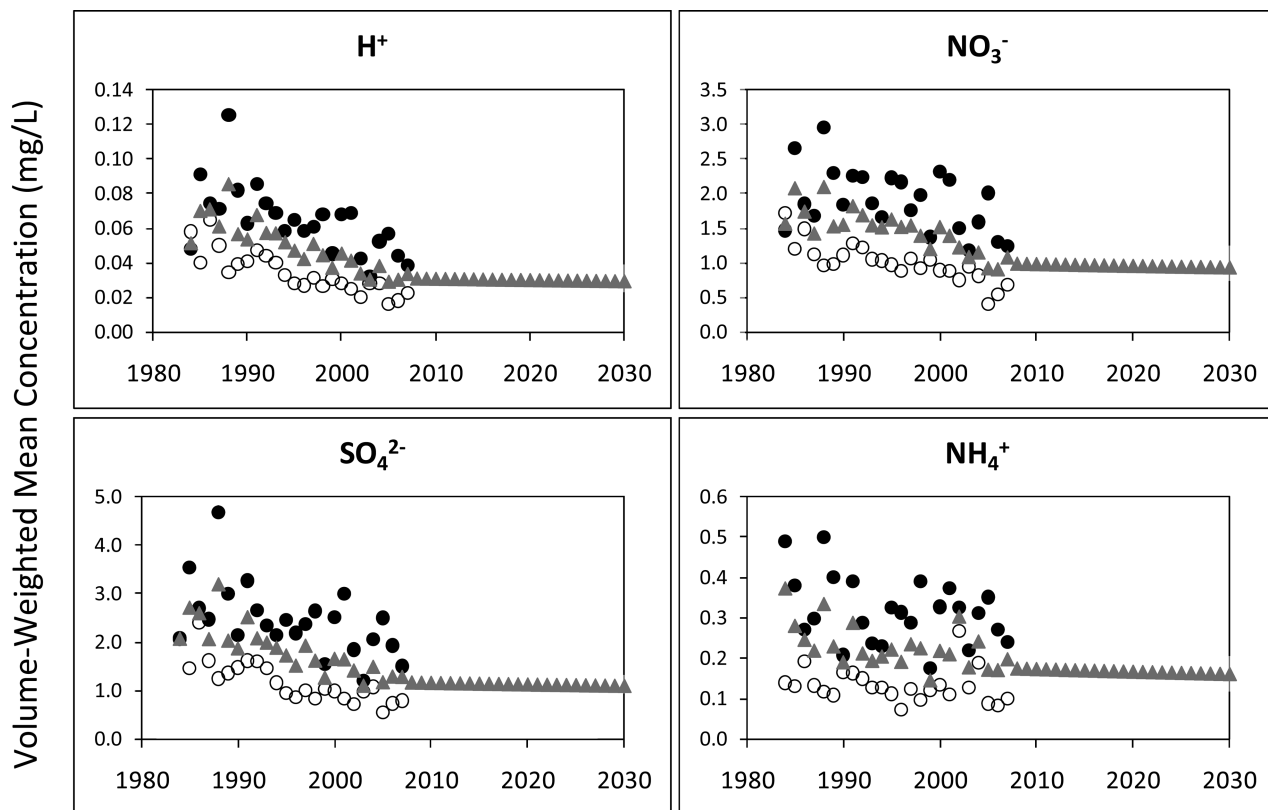
	all events			without marine events		
	slope	slope SE	$r^2$	slope	slope SE	$r^2$
$H^+$	-0.00178	0.00022	0.75	-0.00177	0.00046	0.40
$SO_4^{2-}$	-0.061	0.008	0.70	-0.056	0.018	0.30
$NO_3^-$	-0.036	0.005	0.69	-0.030	0.012	0.23
$NH_4^+$	-0.004	0.001	0.29	-0.004	0.002	0.14

<sup>a</sup> Differences between slopes were not significant.

1500 and 3000 m at the onset of precipitation (Figure 1). There was a slight but not statistically significant decrease in the number and the total volume of events that were of continental origin over the same period (Figure 1). With respect to the total number of events, the proportion of marine-origin events increased and the proportion of continental-origin events decreased (Figure 1). In sum, the volume and number of precipitation events that originated east of our site and were marine in origin increased, but there was not a significant decrease in the number or volume of precipitation events from the continental west or southwest during the study.

**Precipitation Chemistry.** For the acid precipitation solutes  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$ , volume-weighted mean (VWM) concentrations for events that were of continental origin were higher than for those of marine origin (Figure 2). In contrast, and as expected, VWM concentrations of  $Na^+$  and  $Cl^-$  were higher for events that were of marine origin than for events that were of continental origin (Figure 2). The  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  VWM concentrations declined during the 1984–2007

period for events that were of continental origin ( $r^2 = 0.40, 0.29, 0.23,$  and  $0.14,$  respectively), and  $H^+$ ,  $SO_4^{2-}$ , and  $NO_3^-$  declined for events that were of marine origin during that same period ( $r^2 = 0.74, 0.70,$  and  $0.68,$  respectively) (Figure 3). To determine whether the increase in marine events has affected the decreasing trend in acid precipitation solutes, we compared the slopes of linear regressions of annual VWM concentrations that excluded marine events with regression slopes that included marine events. A greater decreasing slope in mean concentrations when marine events were included would indicate that the increase in marine storms has made an important contribution to the decline in acid precipitation. The slopes of the change over time in the VWM concentration of  $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  were not significantly different when events that were of marine origin were included in the estimate than when events that were of marine origin were excluded (Table 1). Thus, these data suggest that the increase in events that are of marine origin has not caused a significantly steeper decline in concentration of acid precipitation solutes than the decline due to reduced emissions of acid precipitation precursors.



**FIGURE 4.** Results of the mixing model for annual  $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  volume-weighted mean concentration (mg/L) for events that were of marine origin (open symbols), of continental origin (closed symbols), or a mixture of events (triangles) at the Cary Institute of Ecosystem Studies, Millbrook, NY, USA. Projection of mixed precipitation beyond 2007 assumes an annual increase in marine precipitation of 1 cm and an annual decrease in continental precipitation of 0.19 cm with no further decreases from 2007 levels of  $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  concentrations. Thus this projection illustrates the case in which air mass trajectories continue to change at the rates observed over the last 24 years, but no further reduction in emissions of acid rain precursors occurs.

**Model Predictions.** Results of the mixing model indicate that, even with an increase in precipitation from marine sources of  $1 \text{ cm year}^{-1}$  and with a decrease in continental precipitation of  $0.19 \text{ cm year}^{-1}$  after 2007, the change in  $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  VWM concentration would not decline substantially (Figure 4). This result suggests that the decline in solutes contributing to acid precipitation between 1984 and 2007 was not due to an increase in precipitation from marine storms.

## Discussion

**Climate Change and Shifts in Air Mass Origin.** Many researchers have predicted that climate change will increase the effects of air pollution (11–17), while others have predicted that the effect of climate change on pollution will be insignificant compared to emission changes (18, 19). Changes in precipitation volume due to climate change could change sulfur and nitrogen deposition by either increasing or decreasing deposition, depending on the direction of change in precipitation volume (20, 21). Climate change predictions specifically for the northeastern US call for an increase in storms of tropical origin (1) and a decrease in midlatitude storms (2, 3). In this study, we focused on the change in the direction from which storms have originated over time and the effect that such a shift has had on precipitation chemistry. We asked if there had been an increase in marine storms in the northeastern US, and if so, whether that had an effect on the chemistry of precipitation in this region. Our results show a small increase in the number and volume of marine precipitation events between 1984 and 2007 and a slight, but not statistically significant, decrease in the number and volume of western, continental precipitation events. There was, however, important year-to-year

variability, with some years, e.g. 2005, in which there were an extraordinary number and volume of tropical events. The data from 2005 suggested that an increasing trend in marine precipitation had occurred, yet 2006 and 2007 data did not follow the same pattern. It is important not to allow short-term data from a few unusual years to influence our view of the long-term trends (22). Overall, the predominant direction of storm events was westerly and continental, which is consistent with past studies (4, 5, 23). Thus, while our analysis corroborated the prediction of increased marine precipitation as a result of climate change, we did not see a significant decrease in westerly storms. While the jet stream has shifted poleward (2, 3), our results suggest that this shift has not resulted in a significant decrease in continental storms at our site. It will be important to monitor trends in marine, subtropical precipitation as climate changes in the future.

**Acid Precipitation.** Storm events that were of marine origin had lower concentrations of acid precipitation solutes than events that were of continental origin. This finding is consistent with earlier studies of precipitation from marine and continental sources (4, 5), which reflect the predominant source of emissions of the acid precipitation precursors,  $\text{SO}_2$  and  $\text{NO}_x$ , from the Midwestern US. Acid precipitation solutes decreased in marine storms as well as continental storms during the 24-year period from 1984–2007, suggesting that marine air contained acid precipitation precursors that were originally from the continental US, as would be expected (24). Some back trajectories showed that air masses can be blown offshore and then back onshore, thus carrying pollutants from the continent to sea and then back to the continent (25). In addition, shipping emissions, which have been increasing over time, have recently been identified as

important contributors to acid precipitation in Europe and elsewhere (26, 27) and could be affecting the precipitation in the northeastern US.

**Clean Air Legislation.** The increasing volume and number of marine precipitation events has not caused a significantly greater decline in acid precipitation solutes than would have occurred had marine precipitation volume remained unchanged during 1984–2007 at the Cary Institute of Ecosystem Studies. Thus the increase in marine storms, while diluting the precipitation with lower concentrations of acidic solutes, has not had as comparable an effect on precipitation chemistry as controls on emissions of acid precipitation precursors. Model predictions suggest that, if the volume of marine precipitation continues to increase at the current rate of approximately 1 cm year<sup>-1</sup> and emissions of acid precipitation precursors do not decrease, precipitation pH would be 4.52 in 2030, only 0.04 pH units less than the 2007 mean precipitation pH measured at the Cary Institute of Ecosystem Studies. Thus, at this point in time, only minor changes in chemistry can be expected from shifting air mass origins alone, and further progress on reducing acid precipitation will require deeper cuts in pollutant emissions.

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