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# The chemical composition of fogs and intercepted clouds in the United States

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## Abstract

Over the past decade, the chemical compositions of fogs and intercepted clouds have been investigated at more than a dozen locations across the United States. Sampling sites have been located in the northeast, southeast, Rocky Mountain, and west coast regions of the US. They include both pristine and heavily polluted locations. Frontal/orographic clouds (warm and supercooled), intercepted coastal stratiform clouds, and radiation fogs have all been examined. Sample pH values range from below 3 to above 7. Major ions also exhibit a wide concentration range, with clouds at some locations exhibiting high sea salt concentrations, while composition at other locations is dominated by ammonium and sulfate or nitrate.

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## 1. Introduction

Clouds and fogs play an important role as processors of atmospheric aerosols and soluble gases. Of particular interest are the roles they play in new particle production (e.g., via aqueous oxidation of sulfur dioxide) and particle removal (e.g., via particle scavenging and removal by fog drop deposition or precipitation). Over the past decade, we have undertaken several studies of the drop size-dependence of fog and cloud drop composition

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and its influence on cloud and fog processing of aerosols (e.g., Collett et al., 1994; Bator and Collett, 1997; Hoag et al., 1999; Xu et al., 1999; Rattigan et al., 2001; Reilly et al., 2001). During each of the field campaigns, we also used more traditional cloud/fog collectors to collect “bulk” samples of fog/cloudwater. Here, we examine results of those bulk cloud and fog measurements and compare the compositions of fogs and clouds sampled at several locations in the United States.

Although bulk cloud/fog composition measurements have been reported at many locations across the US, few investigators have attempted to employ similar sampling approaches at a network of locations spanning the country. While the data presented here do not represent a complete picture of cloud and fog composition at the sites studied, nor provide complete geographic coverage across the US, they are unusual inasmuch as similar sampling approaches have been used at a large number of locations in several regions of the US. In this sense, the results provide an interesting comparison with findings published by Weathers et al. (1988), the only investigators to previously publish results from such a large US cloud-sampling network.

## 2. Experimental

Fogs and intercepted clouds were sampled at numerous US locations during the period 1992–1999. These included studies of radiation fogs, coastal stratiform clouds, and frontal and orographic clouds. Fig. 1 depicts the site locations. Site elevations range from near sea level to more than 3200 m. The list of sites is presented in Table 1, grouped into seven regions. Radiation fogs were sampled in several winter campaigns at numerous locations in the Central Valley of California. Several sampling sites were in the southern Sacramento



Fig. 1. Locations of fog and cloud sampling sites, 1992–1999.

Table 1  
Fog and cloud sampling sites

Region	Site	Study period	No. samples
Sacramento Valley, CA	Woodland, CA	Dec. 1992	10
	Davis, CA	Dec.–Jan. 1998/1999	53
	Walnut Grove, CA	Jan. 1996	3
San Joaquin Valley (SJV), CA	Bakersfield, CA	Jan. 1993, Jan. 1994, Dec.–Jan. 1995/1996	25
	Kern Wildlife Refuge, CA	Dec.–Jan. 1995/1996	20
	Fresno, CA	Dec.–Jan. 1995/1996	26
Oregon Coast	Angora Peak, OR	July 1993	7
Mt. Mitchell, NC	Mt. Mitchell, NC	August 1993	11
Colorado Rockies	Storm Peak Lab, Steamboat Springs, CO	January 1997	27
	Southern California Coast	Cabrillo National Monument, San Diego, CA	June 1993
	La Jolla Peak, CA	June–July 1993	18
	San Pedro Hill, Los Angeles, CA	June 1992	4
	Whiteface Mountain, NY	Whiteface Mountain, NY	Sept. 93, July 98

Valley with additional sites in the central and southern San Joaquin Valley. These events were generally associated with low wind speeds and region-wide stagnation resulting from an overlying subsidence inversion. Coastal stratiform clouds were sampled at three locations in southern California: Cabrillo National Monument in San Diego, San Pedro Hill in Los Angeles, and La Jolla Peak southeast of Oxnard. Additional samples of coastal stratiform clouds were collected at Angora Peak on the northern Oregon coast. Sampling of coastal clouds at all locations was conducted during spring and summer, when stratiform clouds frequently move onshore. Orographic clouds and orographically enhanced frontal clouds were sampled at three additional locations: Mt. Mitchell, NC, Whiteface Mountain, NY, and Storm Peak Laboratory (SPL) in Steamboat Springs, CO. Clouds were sampled in summer and/or fall at Mt. Mitchell and Whiteface Mountain and during winter at SPL.

Bulk fog and cloud samples were collected during these campaigns with one of two collector types. Supercooled clouds (at SPL) were sampled with a Caltech Heated Rod Cloudwater Collector (CHRCC, see Fig. 2). The rest of the samples were collected using version 2 of the Caltech Active Strand Cloudwater Collector (CASCC2, see Fig. 2). The CASCC2 and CHRCC are described by Demoz et al. (1996). Both are active collectors, using a fan to draw drop-laden air across a bank of cylinders which serve as inertial impaction surfaces. The collection surfaces are 508- $\mu\text{m}$  Teflon strands in the CASCC2. The theoretical lower 50% size cut for this sampler is 3.5  $\mu\text{m}$  (drop diameter). The CHRCC features 3.2-mm diameter stainless steel rods as collection surfaces and a theoretical lower size cut of 9  $\mu\text{m}$ . Supercooled cloud drops freeze upon impaction, forming a rime deposit. At the end of each sampling period, the fan is turned off and the collection rods are internally heated to melt the accumulated sample. Samples were collected at 1-h intervals during many of the campaigns, but shorter or longer sampling periods were used in some cases.



Fig. 2. The Caltech Heated Rod Cloudwater Collector (CHRCC) (left) and the Caltech Active Strand Cloudwater Collector version 2 (CASCC2) (right).

Collectors were cleaned prior to sampling by rinsing with high purity deionized water ( $> 18 \text{ M}\Omega \text{ cm}$ ); blanks were taken routinely to ensure the adequacy of collector cleaning procedures. Sample pH values were measured on-site, shortly after collection, using a pH meter and combination microelectrode calibrated with pH 4 and 7 standard buffers. Additional sample was refrigerated and major ions were analyzed later in our laboratory using standard techniques of ion chromatography. Replicate sample analyses typically show a precision of a few percent for the major ion concentrations reported below and a pH uncertainty of 0.1 pH unit or less.

### 3. Results

More than 250 samples of bulk fog and cloudwater were obtained from the 1992–1999 sampling campaigns. These samples were grouped by region as indicated in Table 1. Averages and standard deviations were computed for sample pH, major ion concentrations, and nitrate/sulfate ratios. These results are presented below. Additional information about the composition of some of the San Joaquin Valley fog samples was previously presented by Collett et al. (1999). In the results reported below, it should be noted that there is a slight composition bias introduced by the use of the CHRCC collector ( $D_{50} = 9 \mu\text{m}$ ) in the supercooled Rocky Mountain clouds vs. the CASCC2 collector ( $D_{50} = 3.5 \mu\text{m}$ ) used elsewhere. As shown by Xu et al. (1999), the smallest drops in these clouds typically have lower pH and higher nitrate, sulfate, and ammonium concentrations than are found in the larger drops. Since the total water mass is typically dominated by drops bigger than the CHRCC size cut, however, this bias is minor and does not affect the main conclusions reported below.

Fig. 3 depicts the average fog/cloud pH found for each region. The average pH values vary widely between regions. The highest pH values were observed in the San Joaquin Valley and Sacramento Valley fogs. The average fog pH measured in each of these regions exceeded 6.0. The most acidic regions were the southern California coastal sites (average cloud pH = 3.3) along with Mt. Mitchell (2.8) and Whiteface Mountain (3.5). Minimum cloud pH values recorded at these sites were 2.83 (southern California), 2.45 (Mt.

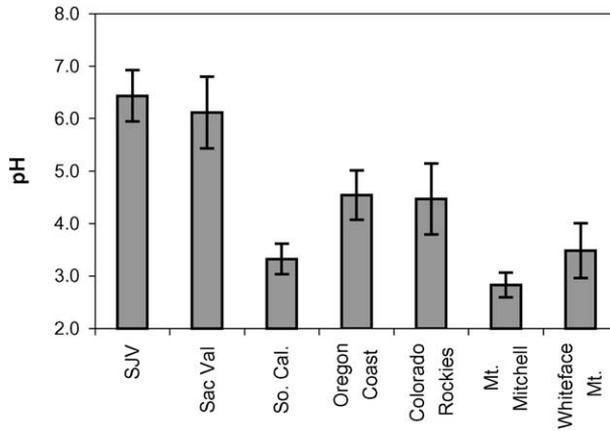


Fig. 3. Average cloud/fog pH values observed at each of the study sites/regions. The error bars indicate one standard deviation observed spread from the mean.

Mitchell), and 2.77 (Whiteface Mountain). Intermediate pH values were found in clouds at the Oregon coast and Rocky Mountain sites.

Concentrations of nitrate and sulfate found in fog and cloud samples are depicted in Fig. 4. Average concentrations of both species exhibit a large range from the cleanest to most polluted sites.

Cloud/fog nitrate is derived from both scavenging of aerosol nitrate and uptake of gaseous nitric acid. The mean nitrate concentrations range from 23  $\mu\text{N}$  (micronormal) at Angora Peak, OR and 58  $\mu\text{N}$  at Storm Peak, CO to nearly 1200  $\mu\text{N}$  at the Sacramento

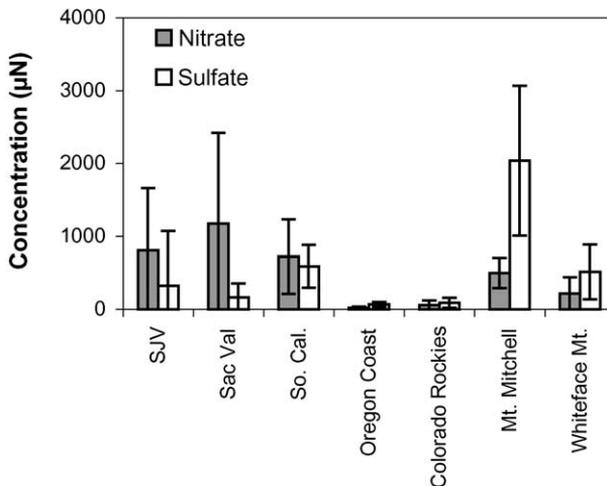


Fig. 4. Average cloud/fog nitrate and sulfate concentrations observed at each of the study sites/regions. The error bars indicate one standard deviation observed spread from the mean.

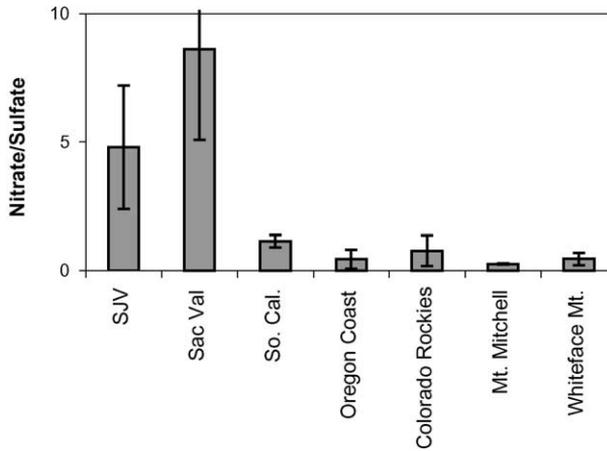


Fig. 5. Average nitrate/sulfate concentration ratios (equivalent/equivalent) observed in fog and cloud samples from each of the study regions. The error bars indicate one standard deviation observed spread from the mean.

Valley sites. Average nitrate concentrations were also high at the San Joaquin Valley (807  $\mu\text{N}$ ) and southern California (718  $\mu\text{N}$ ) coastal sites, while moderate average nitrate concentrations were observed at the two eastern US locations. Intersample variability in nitrate concentrations was high, relative to the mean value, in many of the regions.

Fog and cloud sulfate concentrations, which can derive from aerosol sulfate scavenging or in situ oxidation of dissolved sulfur dioxide, also vary considerably between regions. The lowest mean values were again observed at the Oregon coast (69  $\mu\text{N}$ ) and Rocky Mountain (87  $\mu\text{N}$ ) sites, with concentrations a bit higher in the Sacramento Valley (166

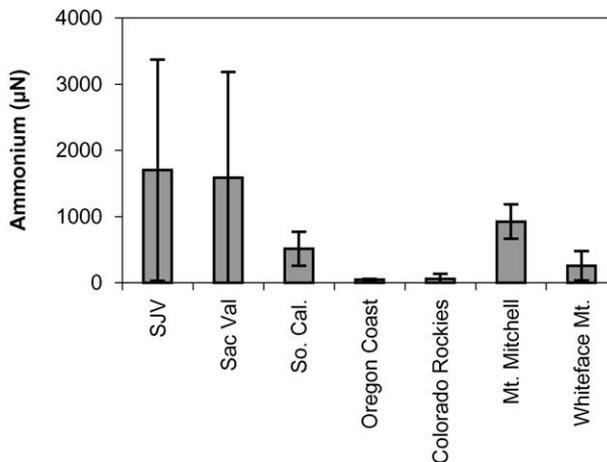


Fig. 6. Average cloud/fog ammonium concentrations observed at each of the study sites/regions. The error bars indicate one standard deviation observed spread from the mean.

$\mu\text{N}$ ) and San Joaquin Valley (324  $\mu\text{N}$ ) radiation fogs. The highest sulfate concentrations were measured at Mt. Mitchell (average of 2033  $\mu\text{N}$ ), with moderately high average concentrations also observed in clouds collected at the southern California coastal sites (586  $\mu\text{N}$ ) and at Whiteface Mountain (511  $\mu\text{N}$ ).

When nitrate and sulfate concentrations are compared, it becomes evident that fog/cloud composition is typically sulfate-dominated at some locations, but dominated by nitrate at other locations. Fig. 5 depicts the average nitrate/sulfate ratios for each region. The highest ratios are observed in the San Joaquin and Sacramento Valley radiation fogs, with average values of 4.8 and 8.6, respectively. The lowest ratio (0.25) is observed at Mt. Mitchell. The Oregon coast, Rocky Mountain, and Whiteface Mountain clouds exhibit average ratios between 0.4 and 0.8, while the southern California coastal clouds have an average nitrate/sulfate ratio of just over 1.

Fig. 6 illustrates average ammonium concentrations observed in each study region. Concentrations are again lowest at the Oregon coast and Rocky Mountain sites, both averaging near 50  $\mu\text{N}$ . The highest average ammonium concentrations were observed in the San Joaquin (1701  $\mu\text{N}$ ) and Sacramento (1588  $\mu\text{N}$ ) valley fogs, where agricultural and livestock activities emit large quantities of ammonia. High average ammonium concen-

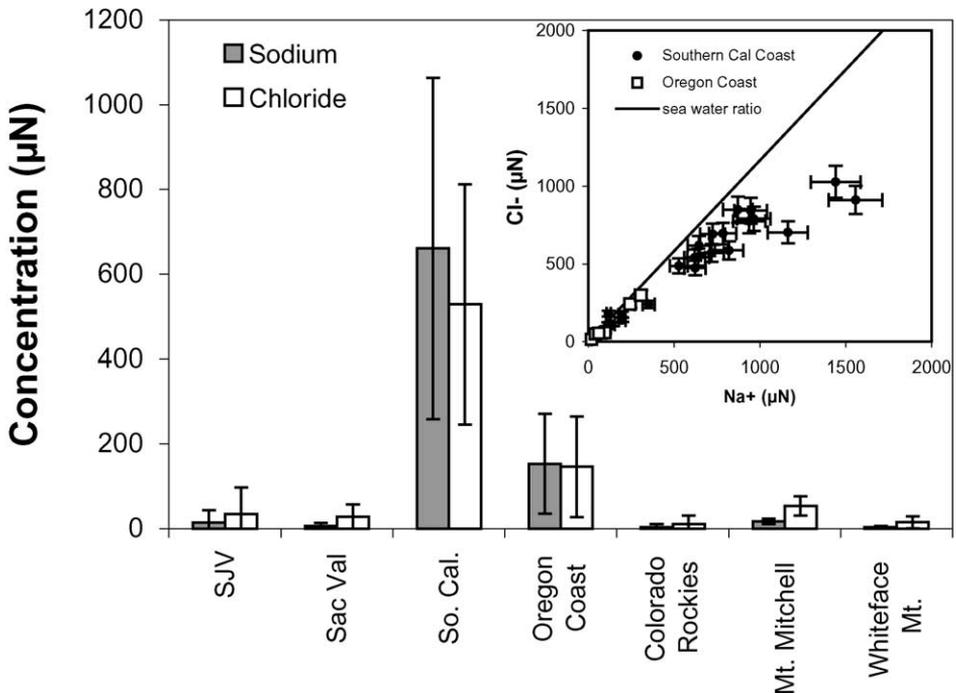


Fig. 7. Average cloud/fog  $\text{Na}^+$  and  $\text{Cl}^-$  concentrations. The error bars in the bar graph indicate one standard deviation observed spread from the mean. The inset figure compares the ratio of  $\text{Cl}^-$  to  $\text{Na}^+$  concentrations in cloud samples collected along the coasts of Oregon and southern California with the ratio found in standard sea water ( $\text{Cl}^- = 1.17 * \text{Na}^+$ ). Error bars on the plotted points indicate the approximate measurement uncertainty.

trations (924  $\mu\text{N}$ ) were also observed in Mt. Mitchell clouds, with modest average ammonium concentrations found in intercepted clouds from southern California (512  $\mu\text{N}$ ) and Whiteface Mountain (256  $\mu\text{N}$ ).

Average concentrations of sodium and chloride ions in fog and cloud samples from each region are depicted in Fig. 7. Concentrations of these species are highest in clouds sampled at the southern California and Oregon coastal sites. Sodium and chloride are the major ionic species measured in the pristine Oregon coast clouds and important contributors to the ionic strength of the polluted southern California clouds. The ratio of chloride to sodium in cloud samples from both locations is generally lower than the ratio typically found in seawater (1.17), with average  $\text{Cl}^-$  to  $\text{Na}^+$  ratios of 0.93 (Oregon coast) and 0.85 (southern California Coast).

#### 4. Discussion

The observed compositions of the clouds and fogs sampled at the different locations are not surprising. Measured pH values, for example, can be largely explained by the balance of acidic and alkaline inputs to the cloud/fog water. The most acidic samples were obtained in southern California, Mt. Mitchell, and Whiteface Mountain, all locations heavily influenced by acidic anthropogenic emissions. Clouds at all three locations have high concentrations of nitrate and/or sulfate, reflecting large inputs of sulfuric and/or nitric acids. Ammonium concentrations, representing neutralizing inputs from ammonia either to precursor aerosol particles or directly into the cloudwater, are much lower than the sum of nitrate and sulfate concentrations at these locations, resulting in only partial acid neutralization and acidic cloudwater. The situation is much different in central California radiation fogs. Here, nitrate concentrations are the highest observed in any of the regions, yet large concentrations of ammonia, emitted in part by agriculture and livestock operations, are more than sufficient to neutralize acidity from nitric and sulfuric acids, resulting in fogs with pH values typically much higher than found in unpolluted regions. The pH values in intercepted clouds sampled on the Oregon coast and in the Rocky Mountains both average 4.5, close to the value expected in the absence of strong anthropogenic inputs of acids or bases (naturally occurring acids can depress the background atmospheric pH value up to a pH unit below the value of 5.6 expected from equilibrium with atmospheric  $\text{CO}_2$ ).

Among the polluted sites, sulfate is the dominant anion in cloudwater at the two sites in the eastern US, nitrate strongly dominates anion concentrations in the California radiation fogs, and shows a slight enhancement over sulfate in intercepted clouds at southern California coastal locations. The greater contributions by sulfate in the eastern US are not surprising, given the large emissions of sulfur dioxide from coal-fired power plants in the Ohio River Valley and other locations frequently upwind of Mt. Mitchell and Whiteface Mountain.  $\text{NO}_x$  emissions are much larger than  $\text{SO}_2$  emissions in California (California Air Resources Board, 1999), so it is not surprising to see nitrate contributing much more than sulfate to fog composition in the Central Valley. If sulfate derived from sea salt is considered in southern California coastal clouds, the importance of anthropogenic contributions of nitrate vs. sulfate is also clearly evident.

Comparison of  $\text{Cl}^-$  to  $\text{Na}^+$  ratios in coastal cloud samples revealed a typical deficiency of  $\text{Cl}^-$ , relative to the ratio expected from sea water. This chloride deficiency was stronger in the southern California cloud samples than in clouds sampled on the Oregon coast. The observed chloride deficiency is a typical result of reactions by which nitric or sulfuric acids can displace chloride (as gaseous hydrochloric acid) from sea salt particles. It is not surprising that the chloride deficiency is stronger in the clouds from southern California, where larger acid concentrations are present.

Differences in fog and cloud composition observed between the study sites can result from many factors. While differences in regional emissions characteristics are important, they do not represent the whole story. Radiation fog formation in central California, for example, relies on air mass stagnation and strong nighttime cooling. Under these conditions, horizontal advection is slow and the zone of influence of emissions is relatively small, perhaps up to a few hundred kilometers for long-lived accumulation mode aerosol species. In cases where orographic clouds or orographically enhanced frontal clouds are collected at mountain sites, transport is typically much more vigorous and the collected cloudwater may include solute contributions from emissions sources located at much larger upwind distances. Differences in boundary layer depths between environments producing radiation fogs and those associated with frontal or orographic cloud formation can also result in different degrees of emission dilution by vertical mixing. Concentration levels of various solutes observed in fogs may also be higher than typically seen in clouds due to the lower liquid water contents (hence, less dilution) typically observed in fogs.

The cloud and fog compositions presented here are broadly consistent with those reported by other investigators for sites located in the same regions. Weathers et al. (1988), for example, report a sulfate concentration range in Whiteface Mountain cloudwater that is similar to, but slightly lower than, the range observed in this study. Anderson et al. (1999) report cloud compositions from three high elevation eastern US sites, including Whiteface Mountain. As in this study, sulfate was found to be the dominant anion and the clouds were on average quite acidic, with annual mean pH values typically between 3.5 and 3.8. Deininger and Saxena (1997) examined how cloud composition at Mt. Mitchell, NC varied with air mass origin. They used computed back trajectories to define three categories: highly polluted (trajectories arriving from  $290^\circ$  to  $65^\circ$ ), polluted marine (from  $65^\circ$  to  $240^\circ$ ) and polluted continental (from  $240^\circ$  to  $290^\circ$ ). Although differences were apparent between the compositions of clouds associated with transport from these three sectors, all showed the main features observed at Mt. Mitchell in the current study: a high sulfate/nitrate ratio and a mean pH below 3.5. The general cloud composition features observed at Whiteface Mountain and Mt. Mitchell in this study have also been reported by other investigators for a number of elevated sites in the eastern US (see e.g., Weathers et al., 1988; Saxena and Lin, 1990; Kim and Aneja, 1992; Mohnen and Vong, 1993) and Canada (Schemenauer et al., 1995). New England coastal fogs have also been found to be quite polluted and very acidic (Weathers et al., 1988; Klemm et al., 1994).

The compositions of fogs in California's Central Valley are broadly consistent with those reported more than a decade ago (e.g., Jacob et al., 1984, 1986), although sulfate concentrations in the current study are considerably lower, consistent with an intervening decline in regional sulfur dioxide emissions. The mean composition reported here for

Oregon coastal clouds is similar to the mean composition reported by [Vong et al. \(1997\)](#) for nonprecipitating clouds collected at the Cheeka Peak coastal site located farther north in Washington state. [Vong et al. \(1997\)](#) report slightly lower values of mean pH (4.2), mean nitrate/sulfate ratio (0.33), and mean ammonium concentration (15  $\mu\text{N}$ ) than found here (4.5, 0.44, and 46  $\mu\text{N}$ , respectively). Average sea salt contributions to the [Vong et al.](#) cloud samples were also somewhat higher than found here. Clouds sampled 100 km from the ocean at Marys Peak, OR by [Weathers et al. \(1988\)](#) had a mean pH of 4.6, close to the value observed here at the coastal Oregon site (4.5). The average composition of southern California coastal clouds reported here is similar to the findings of [Munger et al. \(1989\)](#) who reported a slightly higher mean nitrate/sulfate ratio (1.3 vs. 1.1) in coastal stratiform clouds sampled at San Pedro Hill, CA. Average nitrate and sulfate concentrations reported by [Munger et al.](#) were somewhat higher than reported here, while strong evidence of chloride loss from sea salt was observed in both sets of observations. The cloud pH values measured at Storm Peak Lab, in the Rocky Mountains of Colorado, are in the range of those typically observed by [Hindman and Campbell \(1994\)](#) at the same site in the early 1990s, but generally higher than the values they observed in the mid-to-late 1980s.

## 5. Summary

More than 250 fog and cloud samples were collected at several US locations during the period 1992–1999. Because the sampling campaigns at each location were of limited duration, the data obtained should not be considered to provide a completely representative picture of cloud and fog composition in the studied regions. Nevertheless, a comparison of the results suggests some interesting regional patterns. A wide range of acidities and ion concentration levels was observed across regions. The cleanest samples in the study came from sites on the Oregon coast and in the Rocky Mountains of northern Colorado. Much more polluted cloud and fog samples were collected at coastal sites in southern California, in the interior Central Valley of California, and at mountain locations in the northeastern and southeastern US. Among polluted sites, sulfate was observed to dominate anion concentrations in the eastern US, while nitrate was often much more important in the west. Sodium and chloride concentrations were highest in coastal clouds, where evidence of acid displacement of chloride was also evident. The results presented here are broadly consistent with those reported by several other investigators at similar locations.

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California Air Resources Board, the San Joaquin Valleywide Air Pollution Study Agency, or its Policy Committee, their employees or their members. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as actual or implied endorsement of such products.

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