ACIDIC CLOUD EPISODES IN THE NORTHERN COLORADO ROCKIES: INADVERTENT WEATHER MODIFICATION?

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Abstract. Acidic cloud episodes were detected in January 1998 and January 2000 at Storm Peak Laboratory (SPL) in the northern Colorado Rockies. The episodes were characterized by increased concentrations of small droplets and condensation nucleus concentrations and reduced liquid water contents, snowfall rates and sub-cloud relative humidities. The trajectories of the air parcels arriving at SPL before, during and after the episodes were studied. The parcels arriving during the episodes encountered the least precipitation during their journey to SPL suggesting little cloud and precipitation scavenging of aerosol particles. Further, much of the difference in acidity can be explained by dilution of the cloud droplets. Thus, the episodes may be primarily a natural phenomenon.

1. INTRODUCTION

During the winter months, Storm Peak Laboratory (SPL) in the northern Colorado Rockies near Steamboat Springs is frequently enveloped by snowing supercooled clouds (Borys and Wetzel, 1997; Hindman, 2001). Almost every winter since the winter of 1990-91, during two-week periods in January, City College of New York (CCNY) researchers have been investigating the chemical and physical properties of the clouds. Occasionally cloud water pH values, that normally average around 5, decrease to 4 or less for measurable periods. These periods are the acidic cloud episodes. The cloud chemical and physical characteristics of the two episodes captured to date (January 1998 and 2000) are presented in this paper. The episodes were related to an absence of precipitation scavenging along the trajectories of the air parcels that reached SPL and not necessarily to a change in the chemical composition of the cloud droplet nuclei. Thus, the episodes may be primarily a natural phenomenon.

2. METHODS

Cloud and snow samples were collected and analyzed at SPL for their chemical properties following the methods detailed by Borys, et al. (2000). Cloud droplet spectra were measured using a spectrometer detailed by Borys, et al. Snowfall rates (water-equivalent) were determined following Borys, et al. (1988). PM_{10} measurements were made using a TSI DustTrak (www.tsi.com) in the upwind valley below SPL (only for the January 2000 episode). PM_{10} are the mass concentrations of particles ~ 0.1 to 10 μm in diameter, hence contain the larger cloud-forming nuclei. Condensation nucleus (CN) concentrations were made at SPL with a TSI Condensation Particle Counter (Keady, et al., 1986). CN are particles less than ~ 0.1μm in diameter, hence contain the smaller cloud-forming nuclei. In the laboratory at CCNY, cloud water samples, returned frozen from SPL, were analyzed by liquid ion chromatography for sulfate, nitrate and chloride mass concentrations.

3. CLOUD CHEMICAL CHARACTERISTICS

In January 1998, an acidic cloud episode, consisting of 3-hour, 12-hour and 18-hour cloud events, was identified in a eleven-day series of cloud events (Fig. 1). The episode occurred during the last three days of the cloud events. There was no post-episode because measurements ceased at 1100 MST on 23 January during the acidic cloud episode. The average cloud water pH during the episode was 4.08 and was 4.57 during the pre-episode. Additionally, average concentrations of the sulfate, nitrate and chloride ions in the cloud water were higher during the episode (Fig. 2): the concentrations were, respectively, 2.48, 4.48 and 0.79 mg/L during the episode and 2.35, 1.77 and 0.40 mg/L during the pre-episode. The corresponding snowfall pH values were 4.62 (episode) and 5.31 (pre-episode). The snow pH values are larger than the cloud values because the crystals are composed primarily of diffusion-grown ice with smaller amounts of accreted cloud droplets (Borys, et al., 1988). It is significant that the snow pH values were reduced during the episode reflecting the accretion of the more acidic cloud droplets

In January 2000, an acidic cloud episode occurred consisting of an 8-hour period with measurably reduced cloud water pH values imbedded

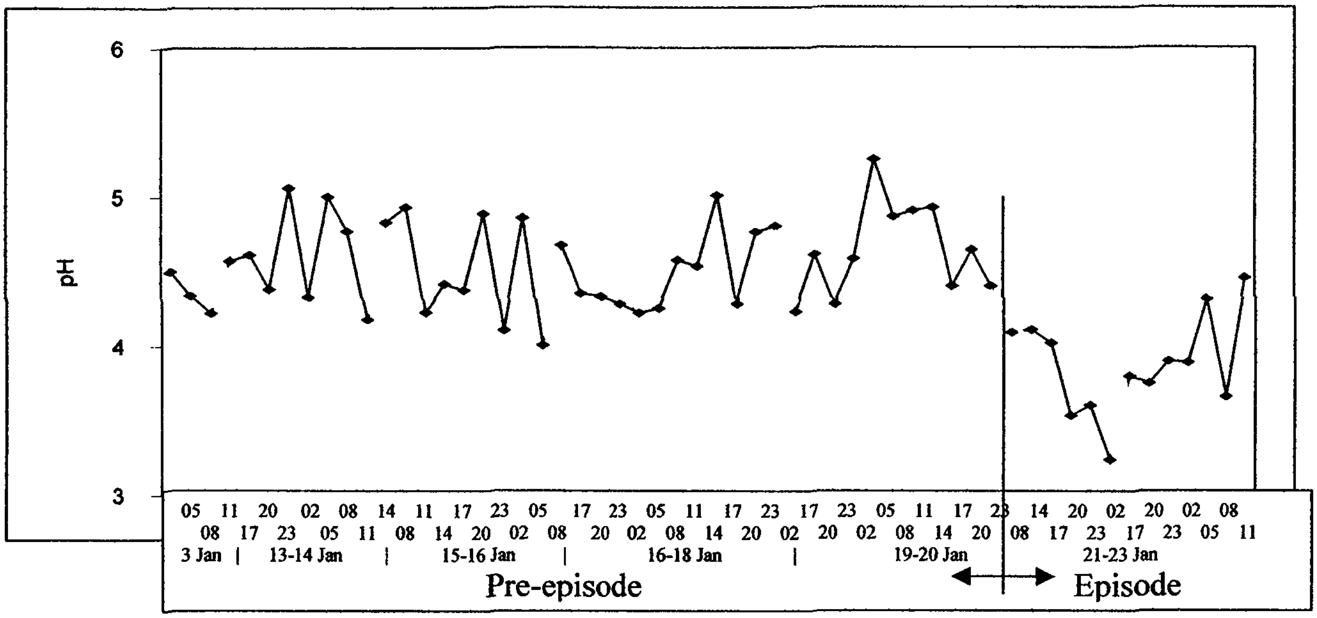

Figure 1: Cloud water pH at SPL during the January 1998 pre-episode cloud events (0500 MST, 3 Jan to 2300 MST, 20 Jan) and during the acidic cloud episode that began 0800 MST, 21 January.

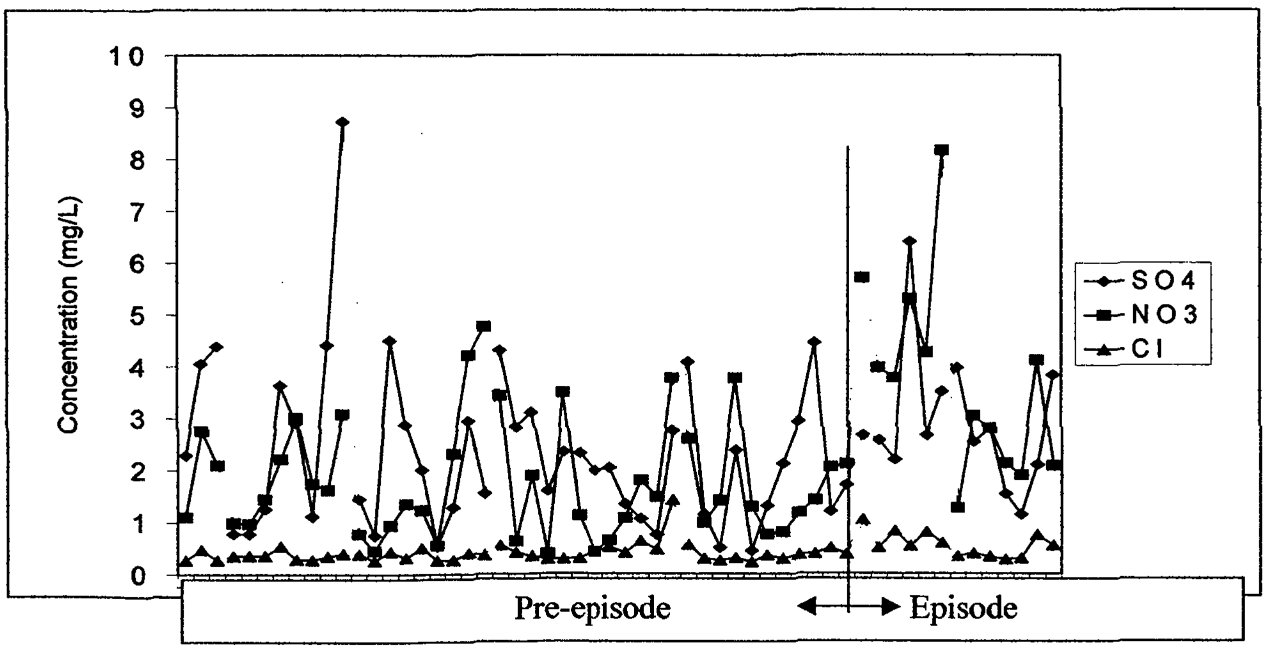

Figure 2. Cloud anion concentrations during the January 1998 pre-episode and acidic cloud episode (see Fig. 1 for dates and times).

in a three-day cloud event (Fig. 3). The anion concentrations were correspondingly enriched (Fig. 4). The episode pH averaged 4.12 while the pre-episode pH averaged 5.39 and the post-episode pH averaged 6.06. The corresponding snowfall pH values were 5.96 (pre-episode), 5.48 (episode) and it was not snowing during post-episode.

4. AEROSOL CONCENTRATIONS

The average CN concentrations measured at SPL during the January 1998 acidic cloud episode were larger than during the pre-episode (Fig. 5): the average concentration during the episode was 1597 cm^{-3} and was 688 cm^{-3} during the pre-episode. Variations in the concentrations during the pre-episode, though, make the difference between the average values not strongly significant.

The average CN concentrations measured at SPL during the January 2000 acidic cloud episode were larger than during the pre-episode (Fig. 6): the average concentration during the episode was 450 cm^{-3} and was 200 cm^{-3} during the pre-episode. The large increase of CN at the end of the post-episode occurred as the cloud base was lifting from below to above SPL; SPL was out of cloud shortly after 1300 MST ending the post-episode. These CN were associated with a new air mass arriving at SPL.

The PM_{10} measurements revealed a significant increase during the January 2000 acidic cloud episode (Fig. 7). This result is consistent with the increase in the CN measurements. There were no PM_{10} measurements during the January 1998 episode.

5. CLOUD PHYSICAL CHARACTERISTICS

It can be seen in Fig. 8 that there were more droplets of smaller size during the acidic cloud episode in January 1998. Accordingly, the cloud liquid water contents (LWC) were reduced: 0.11 gm^{-3} (pre-episode), 0.055 gm^{-3} (episode). Further, the snowfall rates at SPL were substantially lower during the episode: 1.03 mm/h (pre-episode), 0.18 mm/h (episode).

These characteristics were repeated during the January 2000 acidic cloud episode. As seen in Fig. 9, increased concentrations of small cloud droplets occurred during the episode. The corresponding LWC values were 0.22, 0.07 and 0.14 gm^{-3} during, the pre-episode, episode and post-episode, respectively. The corresponding snowfall rates were 1.39, 0.88 and no snow fell during the post-episode.

The variations in droplet measurements during the acidic cloud episodes are comparable to those reported for SPL by Hindman, et al. (1994). They reported the average concentration and mean diameter of the most acidic cloud water samples (pH= 3.4) were 329 cm^{-3} and 6.4 μm and 189 cm^{-3} and 8.0 μm for the least acidic samples (pH = 5.1).

6. AIR PARCEL TRAJECTORIES

Back-trajectories of air parcels arriving at SPL were determined by accessing NOAA's HYSPLIT program (Draxler and Hess, 1998) on the Internet.

6.1 January 1998

Back-trajectories determined for parcels arriving during the pre-episode and during acidic cloud episode were superimposed on a map-background (Fig. 10). The trajectories during the pre-episode varied widely in curvature suggesting the parcels encountered a both "ridges" and "troughs" (high and low pressure regions) during their journey to SPL. In contrast, the trajectories during the episode appear primarily anticyclonic in curvature indicating the parcels encountered primarily ridge conditions.

In troughs, air generally rises and in ridges, air generally sinks (Stull, 2000). Further, the average vertical excursion of the parcels can be determined from their pressure change because pressure decreases in rising air and vice versa. Accordingly, the average pressure change along the trajectories was calculated. It was found the air parcels rose 245 mb during the pre-episode and rose only 43 mb during the acidic cloud episode. These results are consistent with the ridges and troughs encountered by the pre-episode parcels and the primarily ridge conditions encountered by the episode parcels.

The behavior of the air parcels during the hour prior to arrival at SPL is illustrated in Figure 11. All the trajectories during the respective episodes were averaged to produce the figure. It can be seen the air parcels during the pre-episode were rising at both the 500 and 600 mb levels, conditions that would produce deep clouds and more intense precipitation rates. In contrast, air parcels during the acidic cloud episode were rising at the 600 mb level but were sinking at the 500 mb level, conditions that would produce shallow clouds and less intense precipitation. This result supports the smaller precipitation rates measured during the acidic cloud episode.

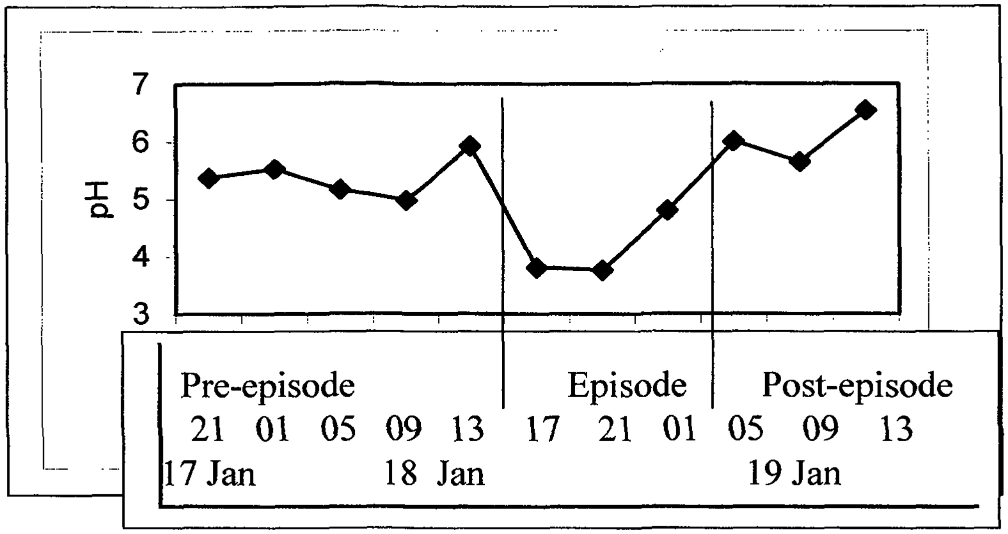

Figure 3. Cloud water pH at SPL during the January 2000 pre-episode, acidic cloud episode and post-episode. Times are MST.

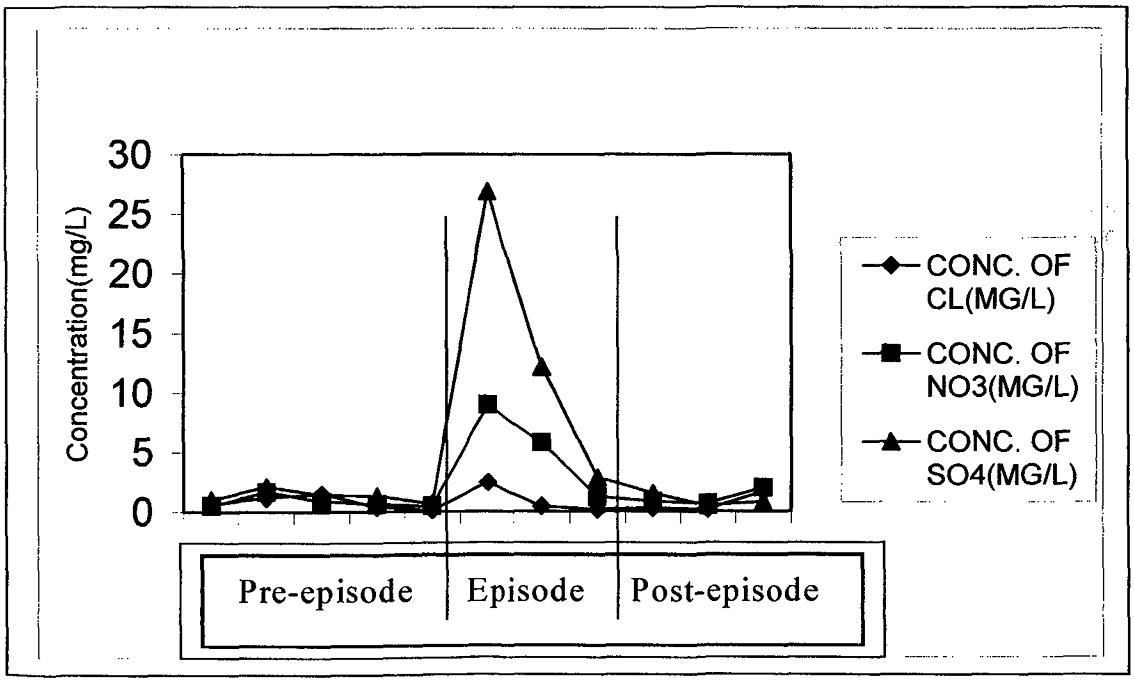

Figure 4. Cloud anion concentrations during the January 2000 pre-episode, acidic cloud episode and post-episode (see Fig. 3 for dates and times).

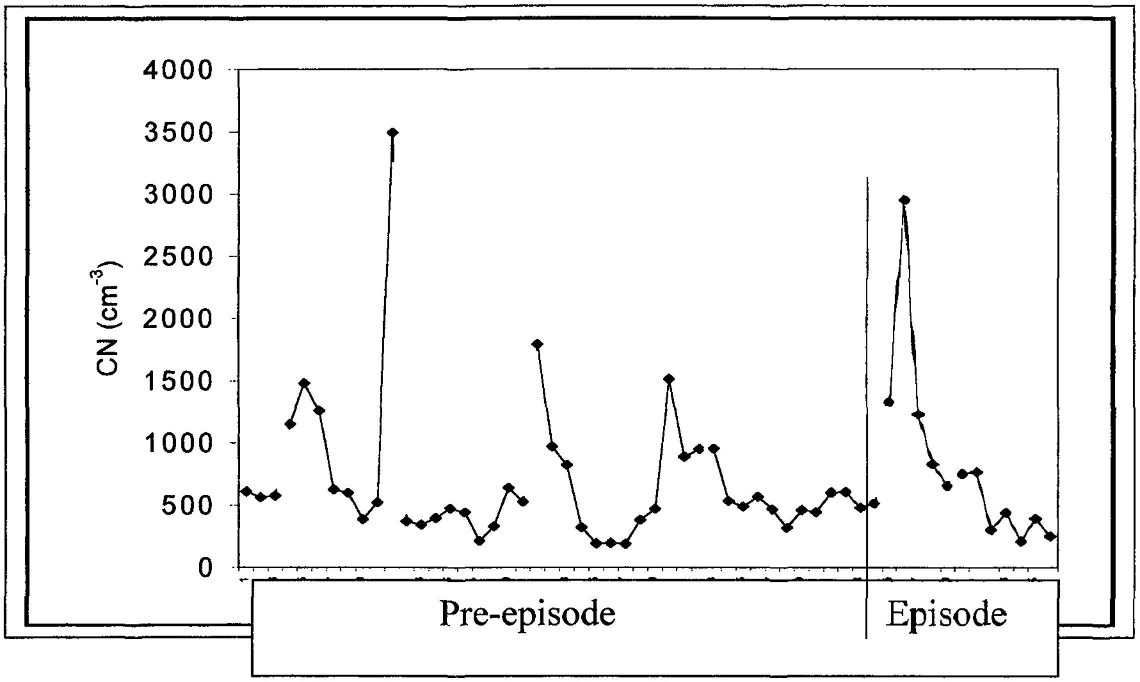

Figure 5. CN concentrations at SPL during the January 1998 pre-episode and acidic episode (see Fig. 1 for dates and times).

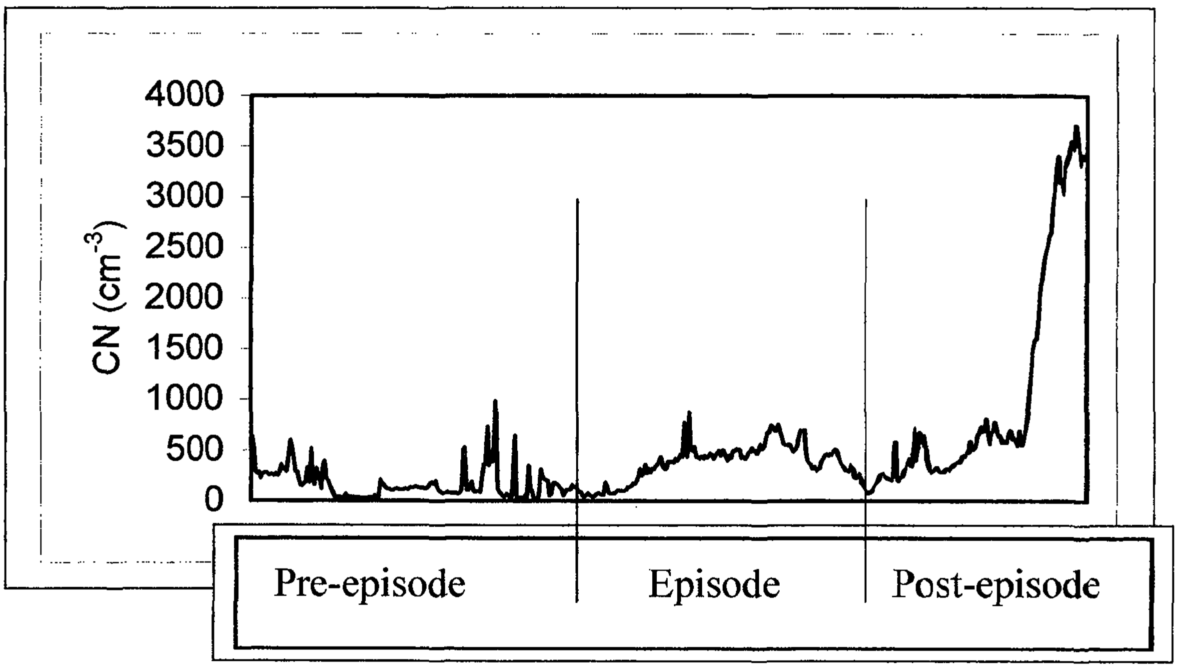

Figure 6. CN concentrations at SPL during the January 2000 pre-episode, acidic cloud episode and post-episode (see Fig. 3 for dates and times of the episodes).

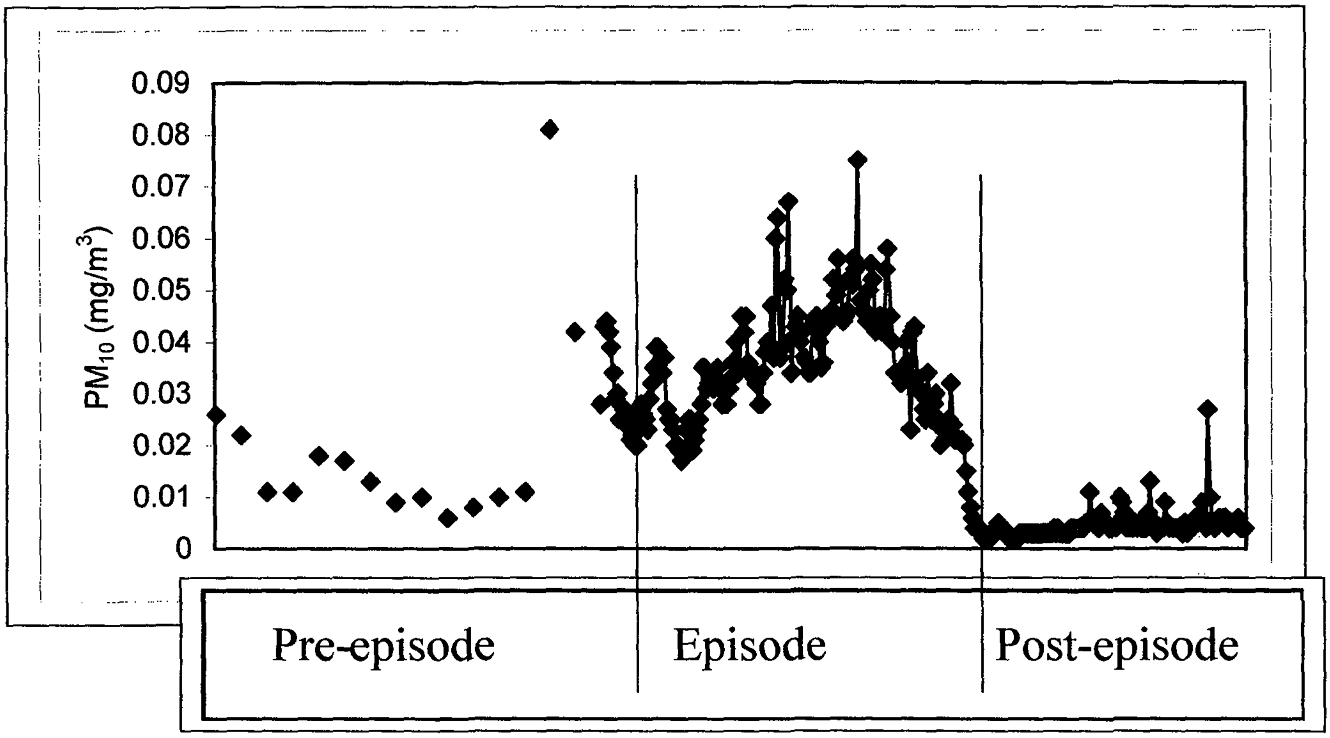

Figure 7. PM_{10} concentrations in the valley below SPL during the January 2000 pre-episode, acidic cloud episode and post-episode (see Fig. 3 for dates and times of the episodes).

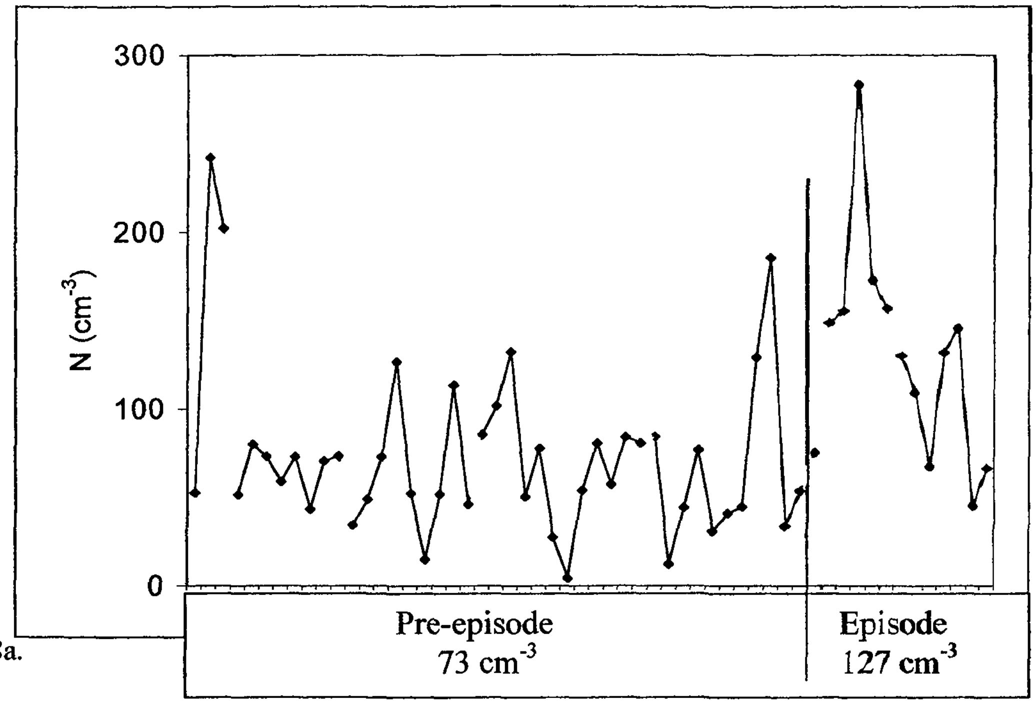

Fig. 8a.

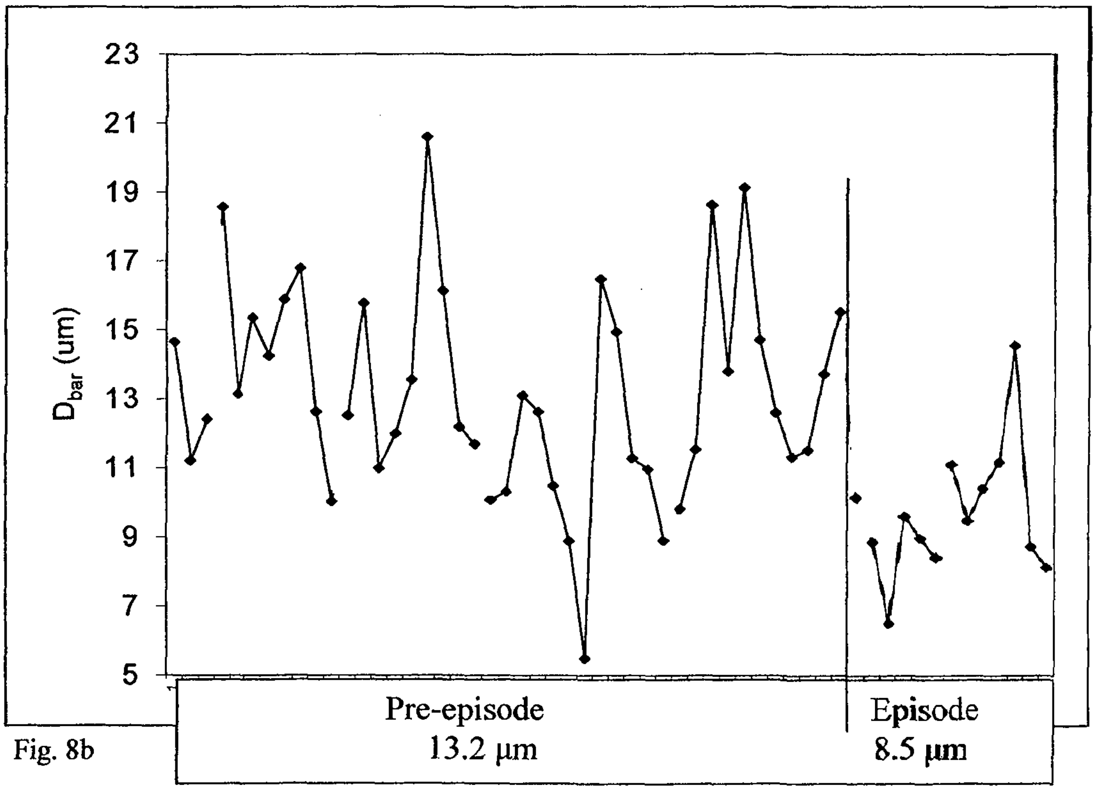

Fig. 8b

Figure 8. Cloud droplet concentrations (a) and average droplet diameters (b) during the January 1998 pre-episode and acidic cloud episode. Average values for the episodes are the inserted values. See Fig. 1 for dates and times.

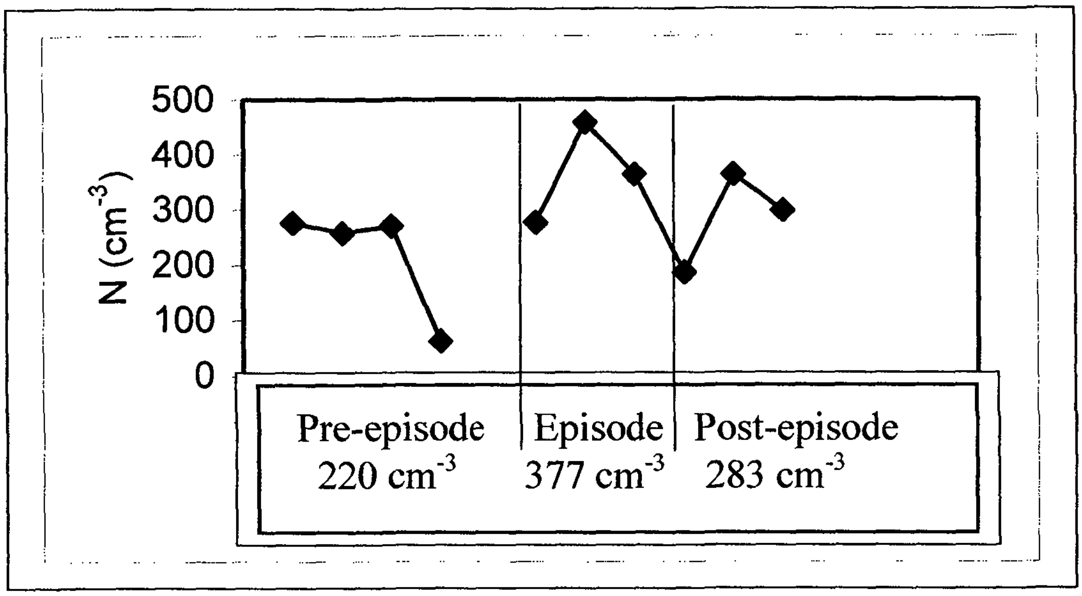

Fig. 9a

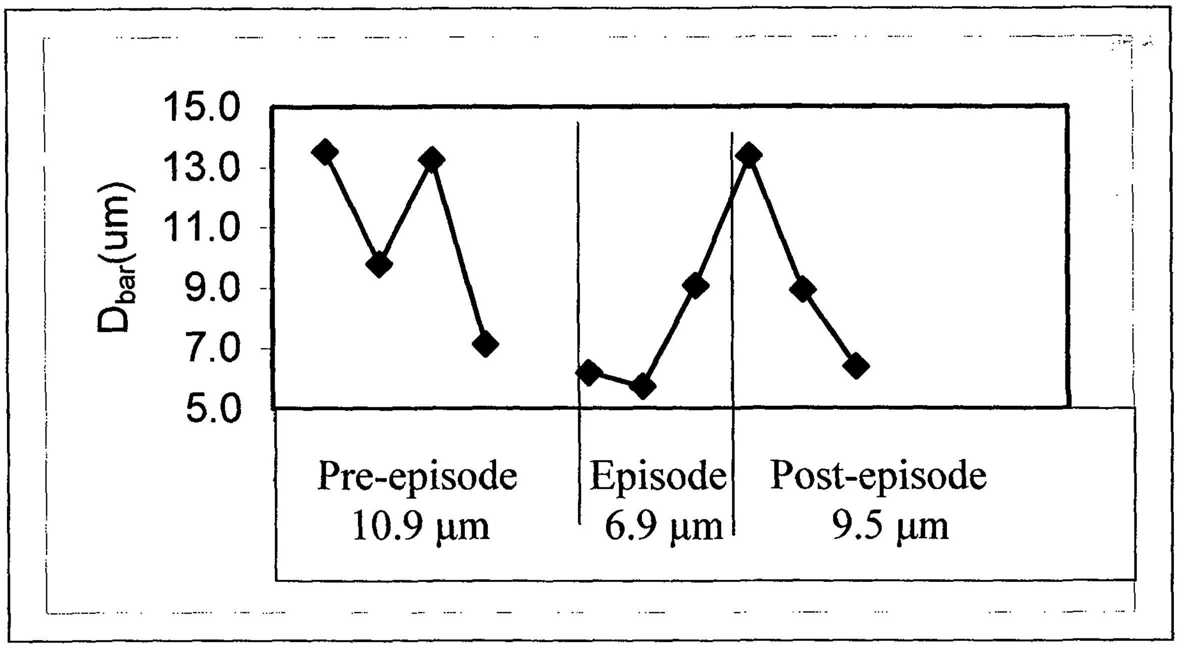

Fig. 9b

Figure 9. Cloud droplet concentrations (a) and average droplet diameters (b) during the January 2000 pre-episode, acidic cloud episode and post-episode. Average values for the episodes are the inserted values. A data point is missing in the pre-episode (13h) because the cloud spectrometer iced up (see Fig. 3 for dates and times of the episodes).

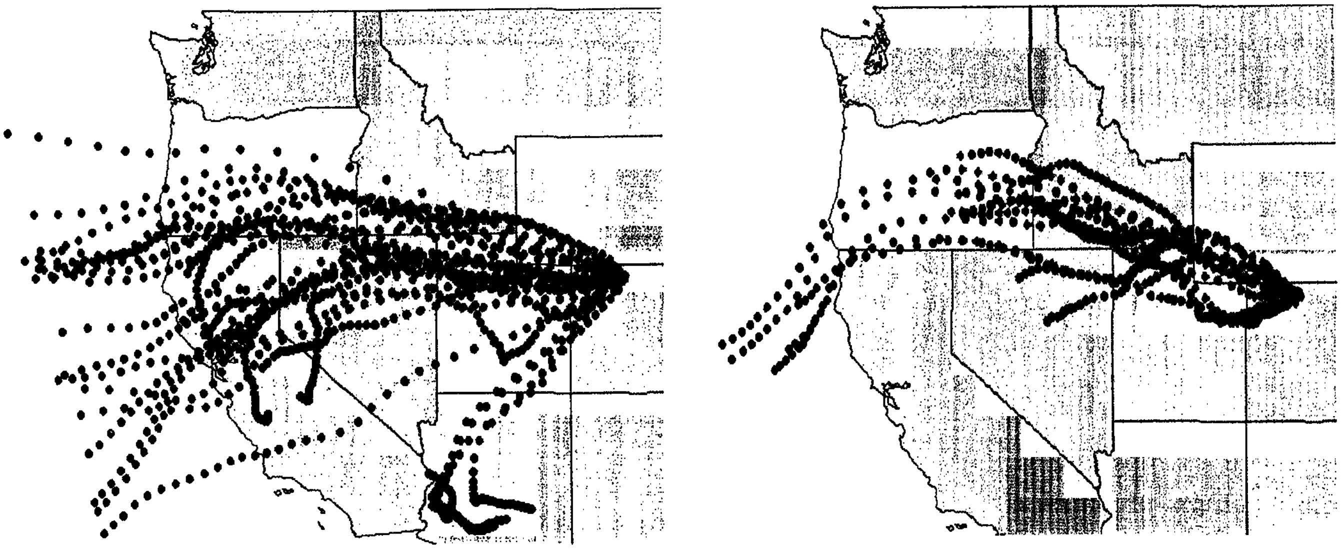

Figure 10. 48-hour air parcel trajectories during the January 1998 pre-episode (left) and acidic cloud episode (right). Each dot represents one hour of travel of the parcels. There was no post-episode period.

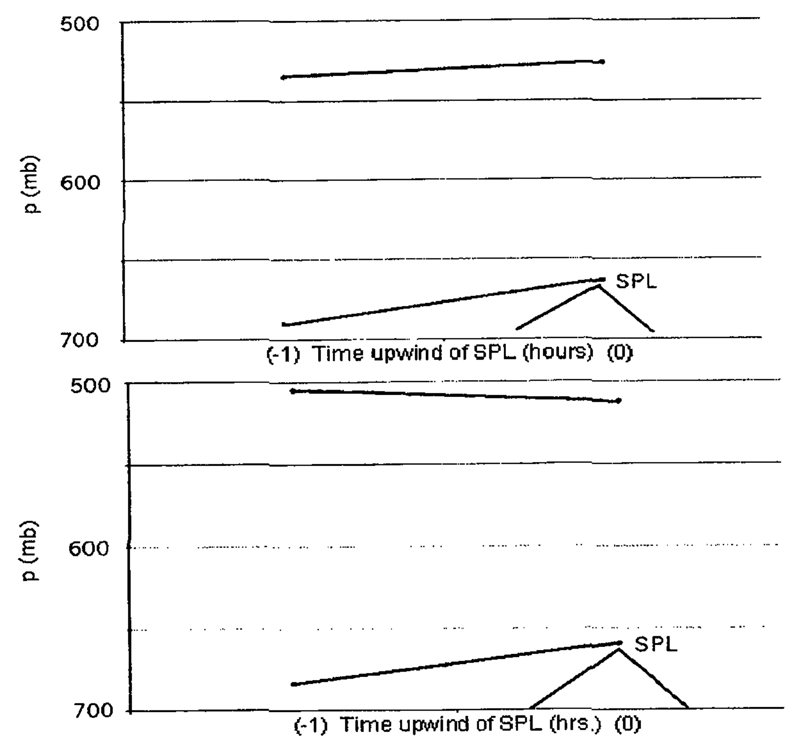

Figure 11. Average change in pressure of the air parcels during the last hour prior to arrival at SPL: (top) pre-episode, (bottom) acidic cloud episode.

6.2 January 2000

The horizontal back-trajectories of the air parcels arriving at SPL during the pre-episode, episode and post-episode are shown superimposed on a map-background in Figure 12. The path of the parcels during the pre-episode and acidic cloud episode periods were almost identical; the air originated from the Four-corners region. The southerly flow indicates low pressure was to the west of SPL and high pressure to the east. The vertical component of the trajectories (not shown) indicated the air arriving at SPL was forced to rise up the Park Range forming the reported clouds and precipitation. It can be seen in Figure 12 the post-episode period trajectories originated further west in response to the passage of the low pressure region.

7. TRAJECTORY-RADAR ANALYSES

The SPL snowfall measurements indicated substantially larger precipitation rates during the pre-episodes than during the acidic cloud episodes. Further, the trajectory analyses indicated the presence of predominantly low-pressure systems west of SPL during the pre-episodes. Thus, it was hypothesized that the air parcels arriving at SPL during the pre-episodes encountered greater precipitation, hence resulting in "cleaner" clouds with higher cloud water pH values.

To test this hypothesis, the amount of time air parcels encountered precipitation on their way to SPL was estimated. Hourly radar images from NOAA's National Climatic Data Center web site (www.ncdc.noaa.gov) were downloaded and superimposed on the corresponding horizontal back-trajectories, using the GIS program ArcView. A sample of trajectories representing the pre-episode, acidic cloud episode and the post-episode was selected for analysis. Figure 13 shows sample radar images with superimposed parcel trajectories during the pre-episode and acidic cloud episode for the January 1998 case.

The results of the trajectory-radar analyses supports the hypothesis. For the January 1998 case, the pre-episode parcels were in precipitation 35% of the time and the episode parcels 7% of the time. In fact, the parcel that arrived at SPL that produced the lowest pH value encountered no precipitation! For the January 2000 case, the pre-episode parcel was in precipitation 30% of the time, the episode parcel 18% of the time and the post-episode parcel 58% of the time. The post-episode parcel arrived at SPL at 0900 MST on 19 January. The abrupt increase in CN seen in Figure 6 began at 1000 MST on 19 January. Thus, one explanation for the abrupt CN increase may be the parcels arriving after 1000 MST traveled through little precipitation.

8. DISCUSSION

Both acidic cloud episodes were characterized by increased concentrations of small droplets, reduced liquid water contents, reduced precipitation rates and increased CN concentrations. Additionally, the air parcels arriving at SPL during both episodes encountered significantly less precipitation.

These results indicate the air parcels that traveled to SPL which produced the acidic cloud episodes had few aerosol particles removed by precipitation scavenging. The resulting increased CN and PM_{10} concentrations likely led to the increased concentrations of small droplets. Further, this shift in the droplet spectrum to smaller droplets most likely led to the reduction in precipitation rates during the episodes by a reduction in snow crystal riming. This reduction in riming has been recently measured at SPL by Borys, et al. (2000).

The reduced cloud LWC values during the acidic cloud episodes most likely were a result of reduced water vapor contents of the air parcels arriving at SPL. Relative humidity measurements made at the 2600 m elevation (SPL is at 3520 m), an elevation often just below cloud-base, revealed the humidity, indeed, reduced during the acidic cloud episodes. In January 1998, the pre-episode humidity was 94% and the episode humidity was 92% while in January 2000, the humidities were 95% (pre-episode), 88% (episode) and 98% (post-episode). The reduction in humidities of the episode parcels led to higher cloud bases and, therefore, less cloud depth below SPL producing smaller LWC values. The smaller LWC values and increased cloud droplet concentrations led to the increased acidity of the droplets. A similar finding was detected in non-precipitating stratiform clouds at Whiteface Mountain NY by Falconer and Falconer (1980) and at Mt. Brocken, Germany by Acker, et al. (2001).

The acidity of the aerosol particles in the source region of the parcels may not be a determining factor for the acidic cloud episodes. For example, the Four-corners region was the source for both the pre-episode and episode air parcels for the January 2000 case; the region contains a large coal-fired power plant (there are two such plants 36 and 90 km west of SPL, too). Therefore, it seems reasonable to assume the chemical composition of the aerosol particles were identical for both periods. Thus, the less numerous and larger cloud droplets during the pre-

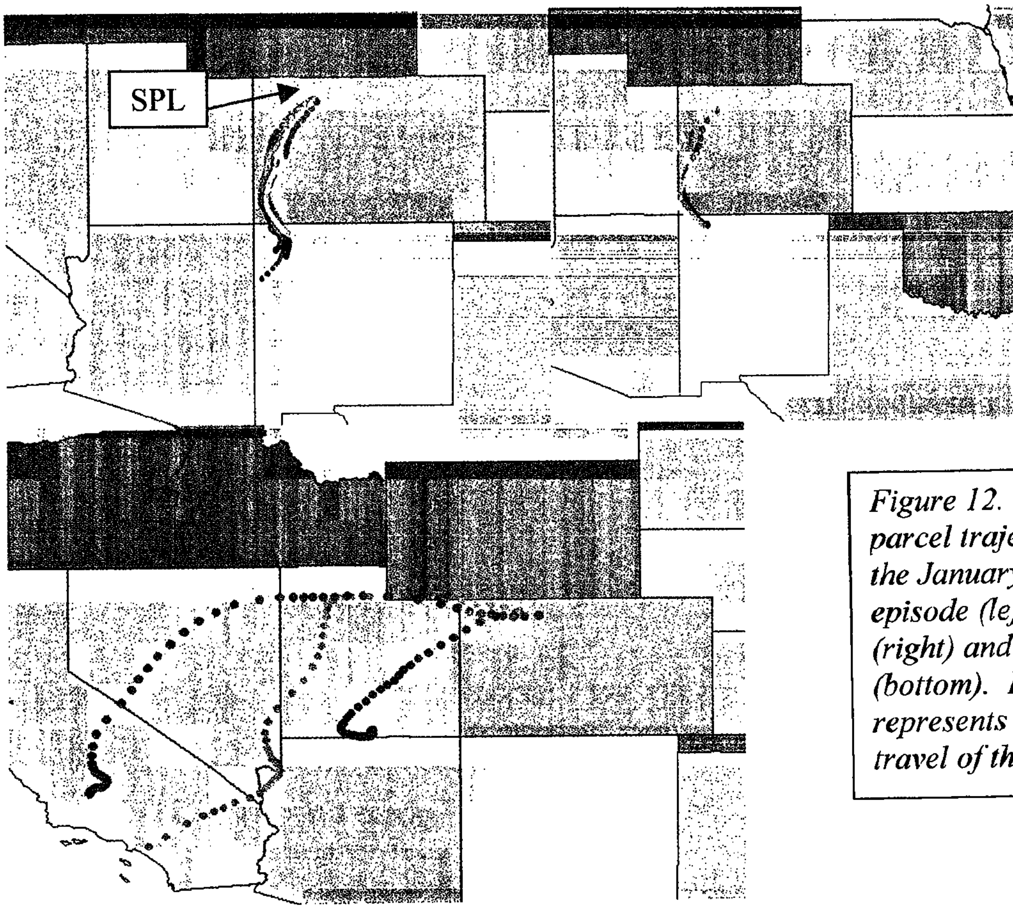

Figure 12. 48-hour air parcel trajectories during the January 2000 pre-episode (left), episode (right) and post-episode (bottom). Each dot represents one hour of travel of the parcels.

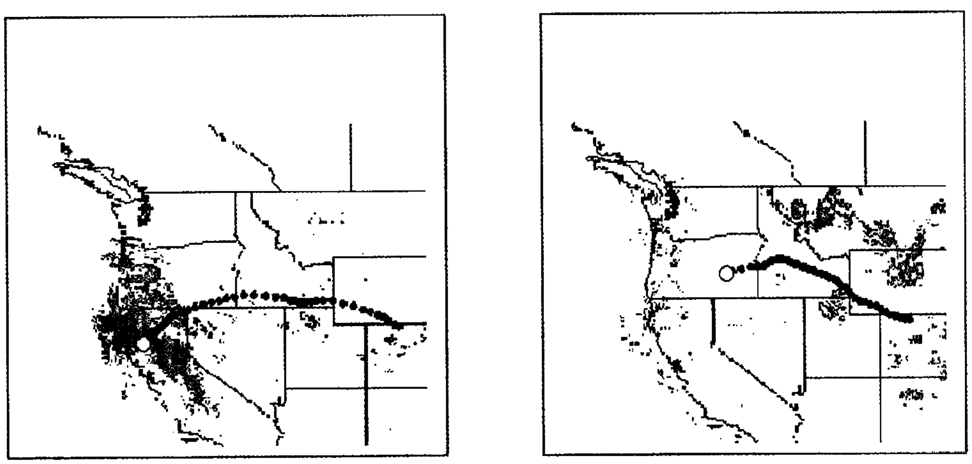

Figure 13. Sample radar images with superimposed back-trajectories for (left) the parcel that arrived at 0200 MST at SPL during the pre-episode on 14 January 1998 and for (right) the parcel that arrived at 0200 MST at SPL during the acidic cloud episode on 22 January 1998. The circle indicates the position of the air parcel at the time of the radar image.

episode may have been more chemically dilute that the smaller and more numerous cloud droplets during the episode. This dilution may explain much of the difference in average pH values between the pre-episode and episode.

To test this idea, the hydrogen ion concentrations $[H^+]$ from the pre-episode and episode periods for both January cases were calculated (pH = -log $[H^+]$). The $[H^+]$, then, were compared with the dilutions calculated from the average cloud droplet sizes (D_{bar}) and number-concentrations (N): Volume/drop = D_{bar}^3/N. For the January 1998 case the pre-episode $[H^+]$ was a factor of 3.1 less than the episode value and the corresponding dilution value was a factor of 6.5; the droplet dilution easily accounts for the difference in $[H^+]$. In contrast, for the January 2000 case, the pre-episode $[H^+]$ was a factor of 19 less than the episode value and the corresponding dilution value was a factor of 7; the droplet dilution accounts for a portion of the difference in $[H^+]$. Thus, dilution can explain much of the difference in average pH values between the pre-episode and episode keeping aerosol particle composition constant. This result is consistent with the findings of Falconer and Falconer (1980).

Alkaline cloud episodes, likewise, may occur at SPL. Borys (2001, personal communication) reported that the spring 2001 Asian dust episode documented by Prospero (2001) was detected in the aerosol measurements at SPL (there were no corresponding cloud measurements). These episodes occur most frequently between February through May as reported by Yienger, et al. (2000). The dust particles are expected to be alkaline and, thus, produce alkaline cloud episodes instead of acidic cloud episodes. Therefore, it would be instructive to determine the effects of Asian dust episodes on clouds and precipitation at SPL through a combination of remote (Rosenfeld, 2000) and in situ measurements.

9. CONCLUSIONS

The two acidic cloud episodes studied to date (January 1998 and 2000) occurred, respectively, for 30 and 38% of the duration of the parent clouds. The episodes appear to have been caused by air parcels which encountered little precipitation on their journey to SPL. As a result, the parcels contained increased aerosol particle concentrations and reduced relative humidities which altered the chemical and physical structure of the clouds enveloping SPL. It was shown that much of the difference in acidity between the pre-episodes and acidic cloud episodes could be explained by dilution of the cloud droplets and not necessarily a change in chemical composition of the aerosol particles from "natural" to "human-produced". Thus, the meteorological conditions upwind of SPL appear to be a major cause of the acidic cloud episodes. The acidic episodes, therefore, are not necessarily an indication of inadvertent weather modification.

10. ACKNOWLEDGEMENTS

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