

Acid concentration of cloud mass in relation to Size Distribution and Origin in Steamboat Springs, Colorado

Jacob Goldberg

Department of Atmospheric and Oceanic Sciences-University of Wisconsin-Madison

April 2010

ABSTRACT

Seven experiments were conducted throughout the day on 1 April 2010, collecting size distribution, wind direction, and rime pH data at the Storm Peak Laboratory (SPL) in Steamboat Springs, Colorado. Analysis of 6 of the 7 experiments resulted in average pH, and Median Volume Diameter (MVD values of 4.24, 13.41 μm respectively. Number concentration and liquid water content (LWC) both show a general positive trend over the experimental period, indicating an increase in cloud density and total water. Back trajectories spanning 48, 120, and 240 hours behind the time of the Experiment imply that a significant portion of the sulfate aerosols were transported across the Pacific from East Asia and that local contributions during the event were predominantly from the northwest United States.

Introduction

Aerosols, especially acidic aerosols such as sulfates, can have a varied effect on numerous aspects of cloud physics including the dynamics of precipitation processes, size distribution of the cloud, and, in turn, an ecosystem's potential for biodiversity. The fine particulates present in the Colorado Rocky mountain region have a variety of origins, both local and global in nature. These particulates can travel thousands of miles and physically affect cloud microphysics and ecosystem processes in multiple regions at various distances from the initial source. For this reason, aerosol origin and dispersal is a global issue. For the purposes of clarification, local constitutes contributions from the Western United States and global focuses on trans-national particulate contributions.

From 31 March 2010 to 1 April 2010, a winter storm event occurred over Steamboat Springs, Colorado and specifically saw a substantial amount of time in which the Storm Peak Laboratory (SPL) was encapsulated by cloud. Multiple experiments were conducted in situ throughout the storm event throughout much of the day on

1 April 2010. The purposes of this research are to analyze the relationship between the particle-size distributions of orographically induced cloud measured at various times to the pH of collected cloud samples and correlate this with relative origin. It is proposed that decreasing (increasing) average particle diameter will have the effect of decreasing (increasing) the pH and that particle trajectories for this event will indicate a transnational influence and focus the local influence of particles from the Northwest United States. This paper will demonstrate two things: (1) pH is not directly linked to any one specific cloud physical characteristic, but that analysis of multiple size distribution characteristics leads to a greater understanding of pH variation and (2) there is a transnational contribution of particles stemming from East Asia and local contributions originate in the Northwest United States.

Background

In certain conditions, one affect that aerosols can have on the surrounding environment is that, depending upon the type and distribution of aerosol present in an air-mass, precipitation processes can be hindered or even completely inhibited. The

Rocky Mountain region is particularly susceptible to fluctuations in aerosol concentration because much of its precipitation is orographically induced. Much of the particulate mass can accumulate in the thin mountain boundary layer, where the orographic clouds are generated, and thereby alter the propensity for these clouds to precipitate.

Fine particulate aerosols, such as sulfates, have multiple effects on cloud microphysics that can lead to both precipitation suppression and increased acid deposition. One consequence of increased anthropogenic aerosols is their so-called “indirect effect”. The aerosols act as minute cloud condensation nuclei (CCN) which form a high density of tiny cloud droplets (Givati & Rosenfeld, 2004). Borys et al. (2003) found that the addition of only $1 \mu\text{g m}^{-3}$ of anthropogenic aerosol to the Colorado Rocky air-mass can hinder or even shut off the snow particle rime growth process and reduce the orographically induced snowfall rate by up to 50%.

Another effect of anthropogenic aerosols is their ability to interrupt ice nucleation in higher, colder regimes. Generally, insoluble particles, such as soot and dust, can act as highly efficient ice nuclei (IN) by contact freezing supercooled cloud droplets. The nucleability of IN is greatly reduced when anthropogenic aerosol gases such as SO_2 , NO_2 , and NH_3 are absorbed at active sites on the forming ice particle (Pruppacher and Klett, 1997). The concentration of acidic aerosols, depending upon their origin, can therefore act to greatly vary the snowfall efficiency according to their anthropogenic aerosol composition. Air-masses that originate in areas of higher pollution would then likely entrain more of those pollutants and thereby variably reduce their snowfall rate.

Fitzgerald (1974) investigated the effect that various ammonium sulfate aerosol

concentrations had on the droplet size distributions of adiabatically lifted, condensationally produced cloud droplets. In his paper, he found that the range of droplet size distribution produced by the mixed nuclei (soluble and insoluble) was no greater than that for pure salt nuclei. He notes too that work of various researchers, including Belyaev (1961), investigated the effect that turbulence has on droplet growth and that more research in this area is needed. What is less understood, and is a main component to the research here is the relation of aerosol, especially sulfate and its associated forms, to the size distribution of mechanically driven, condensationally formed cloud droplets. Keck (2007) experimentally verified that the median volume diameters (MVD) for stratified and convective clouds peak near $14 \mu\text{m}$ and $18 \mu\text{m}$ respectively and that most all MVDs fall between a $15 \mu\text{m}$ gap near this distribution due to minimal collision-coalescence of cloud particles below $30 \mu\text{m}$. Less research has been done on orographically induced clouds and an aim of this research is to ascertain whether or not the MVDs of clouds at SPL are within this range.

An additional effect of high acidic aerosol concentration is that this inevitably increases the likelihood of acid deposition by multiple schemes including low pH precipitation and acidic fogs. Cloud droplets and, therefore, precipitation normally contain a variable concentration of dissolved ionic species, such as carbonic acid, which make its pH slightly acidic. Atmospheric acids can come from multiple sources, but the most significant are anthropogenic in nature. Acidic fogs and wet deposition by precipitating orographic clouds can have a detrimental affect on various biota and the overall biodiversity of the region (EPA, 2009). A water pH of 5.5 to 6 is a relative threshold below which danger to local and regional biota is considered an environ

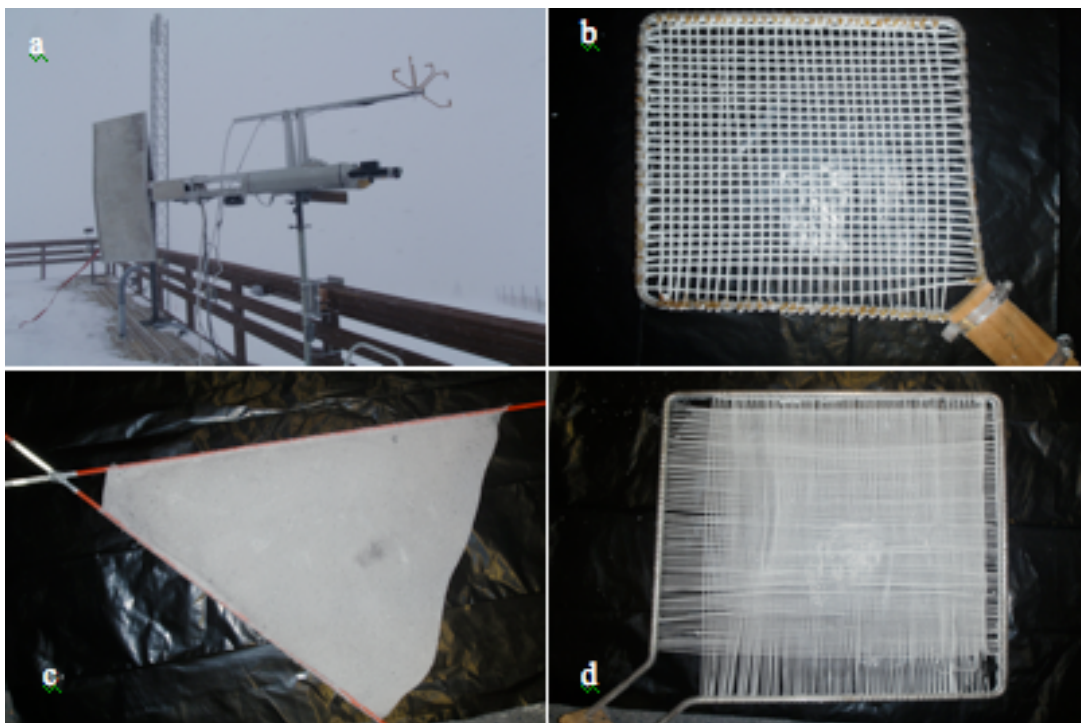


Figure 1 a) — This is the DMT SSP-100, which measures cloud droplets on the range of 3-57 μ m in diameter. b) – Nylon Cloud sieve (with collected rime) with large grid spacing. c) – Anodized aluminum cloud sieve (with collected rime) with small grid spacing.

mental threat (Doka et al, 2003). Trans-Pacific transport of sulfate pollution accounts for up to 30% of the background sulfate aerosol concentration in the United States (Park et al., 2004). Local pollution accounts for a considerable amount of the fine air mass in the western U.S., with 28% in the North West and 38% in the South West (Chin et al, 2007). Both of these sulfate aerosols occur in our atmosphere due to oxidation of SO₂ to H₂SO₄ through reactions with hydrogen peroxide, oxygen, and ozone. Currently, Colorado has 33 coal-fired power plants in operation and more are proposed to come online in 2010. Although there are laws in place to assure that the sulfate emission of these power plants is reduced, they still account for much of the fine air mass in the region, especially in Steamboat Springs.

Methods

The research here is aimed at determining a relationship between acid concentration and origin of clouds passing over the Storm Peak research area between March 31st and April 1st. The clouds are of interest in that their size distributions and pH yield information as to the presence of atmospheric pollutants such as sulfates, and the prevailing wind that carries them is indicative of the general direction of the pollutant origin. The size distribution data is gathered using a Droplet Measurement Technologies (DMT) Scattering Spectrometer Probe (SSP)-100, which measures droplet sizes on a range of 3-57 μ m. Data is constantly recorded while in cloud as the probe is allowed to swivel freely in the horizontal plane in order to face into the predominant wind direction (Figure1a).

Rime was collected utilizing three different grid sized cloud sieves made of two materials: nylon and anodized aluminum (*Figures 1b, 1c & 1d*). Under the assumption that the nylon sieve material is

completely non-conductive and non-reactive, making its contribution to pH and conductivity negligible, calibration of the aluminum sieve involved measuring the difference between averaged pH and conductivities and using that value as an adjustment to remove any adverse material contributions. The sieves were placed on the roof of the laboratory and allowed to build rime as the clouds passed through. The cloud collection devices were brought down after the sieve with the smallest grid spacing appeared to have rimed to the degree that air-flow was significantly restricted. Wind direction was collected from the SPL Mesonet data available from the SPL website. In addition, backward parcel trajectories to determine the relative origin of the airmass during the event were calculated using the NOAA Air Resources Laboratory (ARL) Hysplit model. Specifically, the meteorological data set used comes from The National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS).

Results

For reference in all of the data figures, only Experiments 2-7 are used for analysis. Using archived SPL Mesonet Data, wind direction over the course of the day at the lab is displayed in *Figure 2*.

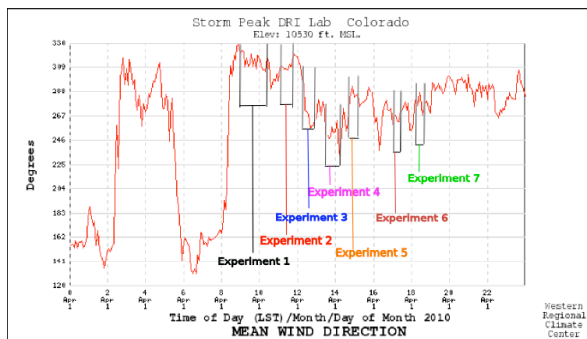


Figure 2 – This is a graph (mesonet data) of the mean wind direction (in degrees) over time at SPL on April 1, 2010. Note that the local standard time (Mountain Standard Time) is 7 hours behind UTC and Mountain Daylight Time (used for this paper) is 6 hours behind UTC or 1 hour ahead of MST.

Apparent from the graph are numerous wind shifts throughout the day, with a significant shift from southwest to northwest at approximately 9-3:30 a.m. MDT. This major wind shift signals the beginning of both the series of experiments conducted while in cloud and the genesis of a snow event over SPL. Also noted in this figure are the relevant wind directions during the various experimental runs, which, due to blocking by upstream topography, play a significant role in the development of orographic clouds at SPL.

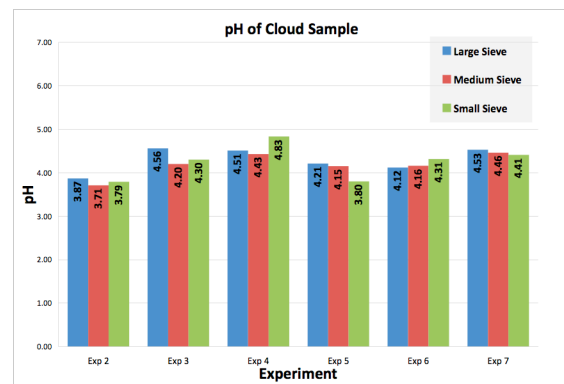


Figure 3 – This image displays the average pH values for each sieve (see legend) during each experiment.

Figure 3 illustrates the variation in pH of experiments 2-7. Although there are slight variations in pH values between the three sieves for each experiment, after calibration they are each well within one standard deviation of each other and would therefore be considered highly representative of the actual cloud pH. The maximum values reached between 4.50 and 4.83 with the lowest average pH at 3.79 for experiment 2. The overall average pH value for all the experiments is 4.24. *Figures 4 & 5* denote the averaged number concentration in ($\text{particles}/\text{cm}^3$) and averaged median volume diameter (MVD) in μm respectively. Higher average number concentrations indicate a relatively denser cloud.

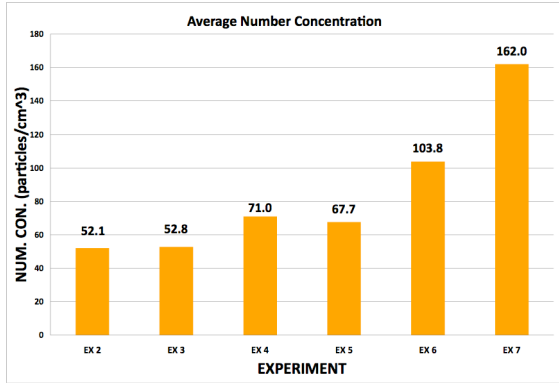


Figure 4 – This figure displays the average number concentration (as a proxy for cloud density) for each experiment in particles per cubic centimeter.

The cloud became denser over the experimental period, beginning with an average number concentration of 61 ($\text{particles}/\text{cm}^3$) for experiment 2 and ending with a value of 160 ($\text{particles}/\text{cm}^3$) for experiment 7. The density of cloud does not seem to correlate strongly with pH and is likely an independent cloud

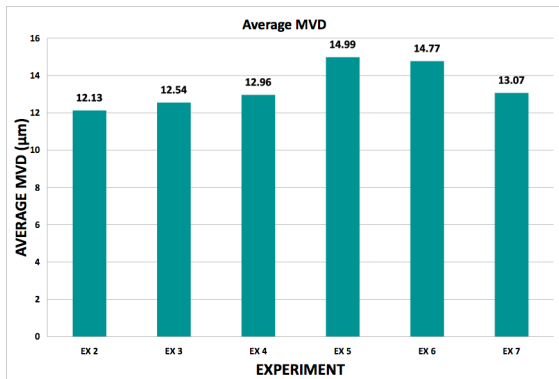


Figure 5 – This image displays the average median volume diameters (MVD) in μm for each experiment.

characteristic. The MVDs fluctuate slightly between experiments, with experiment 1 the lowest at 12.1 μm and Experiment 5 the highest at 14.99 μm , but all the experiments combined have an overall average of 13.41 μm (Figure 5). These values fall in line with experiments conducted by Keck (2007) and, in particular, fall nearer the range of stratiform cloud MVD distributions.

Upon comparison to corresponding pH values between the experiments, MVD and pH seem to have a slight correlation. The magnitude of change from adjacent experiments is within 1% for both pH and MVD and these changes between pH and MVD have a general positively correlation. That is, pH increases are usually denoted by an increase in MVD. In conjunction with Average Liquid Water Content (LWC) data (Figure 6), a more representative picture of the cloud characteristics is revealed. LWC values generally increased throughout the day. There was a dip to the lowest value at Experiment 4 with 0.0246 (g/m^3) and the highest value at Experiment 7 with 0.144 (g/m^3) (Figure 6).

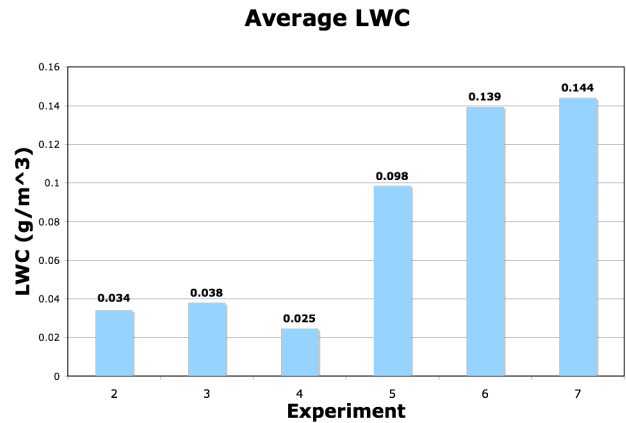


Figure 6– This figure shows the liquid water content (LWC) of each experiment in (g/m^3).

Figures 7 (a, b and c) are all back-trajectories calculated utilizing the ARL Hysplit Trajectory Model. These trajectories are calculated to 240 hours, 120 hours, and 48 hours before 1 April 2010 2300 UTC. Figure 7a displays a trans-national origin, specifically from the Sea of Japan, to the particles detected at the laboratory. Figure 7b focuses the trajectories on a smaller time and spatial scale, where it is discerned

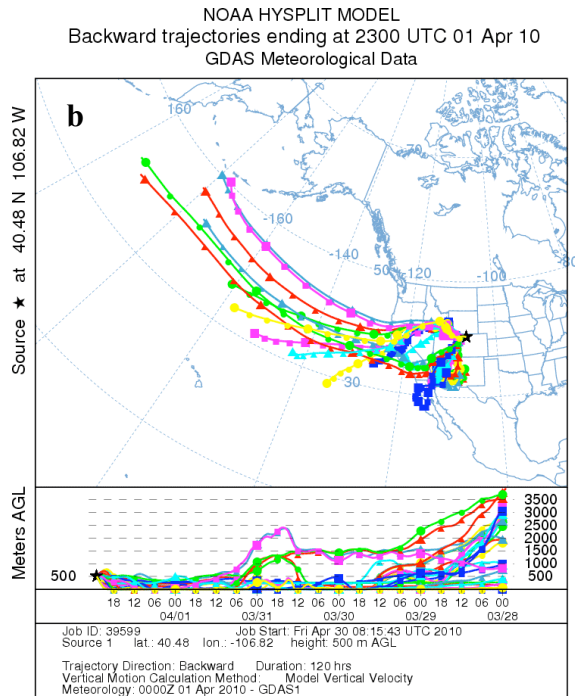
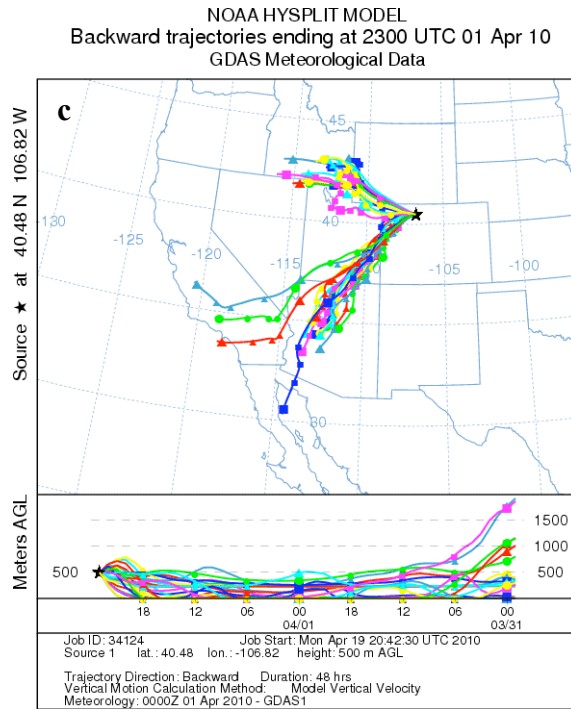
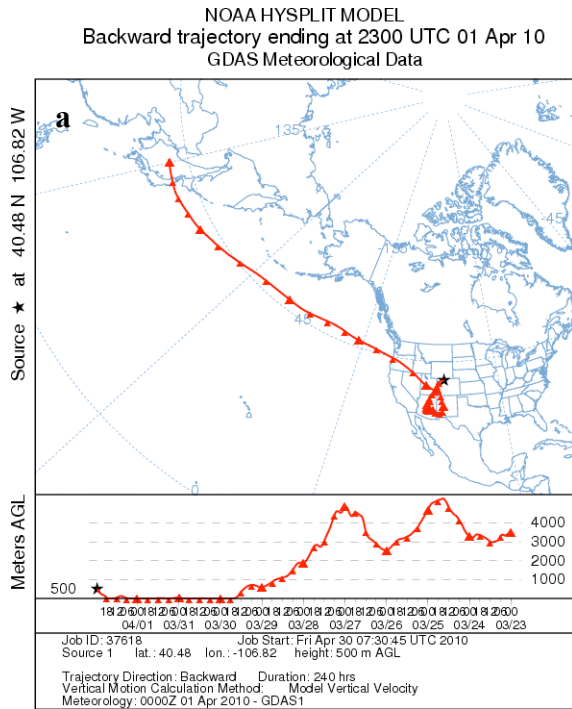


Figure 7 – a) Long-range Hysplit backward trajectory of 240 hours starting at Steamboat Springs, CO on 2300Z 1 April 2010. b) – Same as a) but for a medium range of 120 hours. c) – Same as a) but for short range (local) 48 hours [wind shift highly evident].

Figure 7c further focuses the trajectories on an even smaller time and spatial scale. Here it is evident that as the 700 hPa trough and associated low pressure system (*Figure 8*) and mid level winds passed over the area, the particle contribution shifted from southerly to a northerly influence. Under the experimental verification of sulfate aerosol trans-national transport by Park et al. (2004), it is asserted that a significant quantity of particles that were measured at the lab had an origin somewhere in East Asia. Upon investigation of the finer spatial and temporal scales of the Hysplit data, it is also asserted that a significant amount of the sulfate aerosol arriving at the lab may have been entrained more locally from specifically the northwest region of the United States, to the exclusion of southwestern U.S. contributions.

that particles arriving at the lab had two major sources.

The two major sources at 120 hours are out over the far north Pacific, likely in the polar jet stream, and, to a smaller extent, a southerly origin off the coast of California.

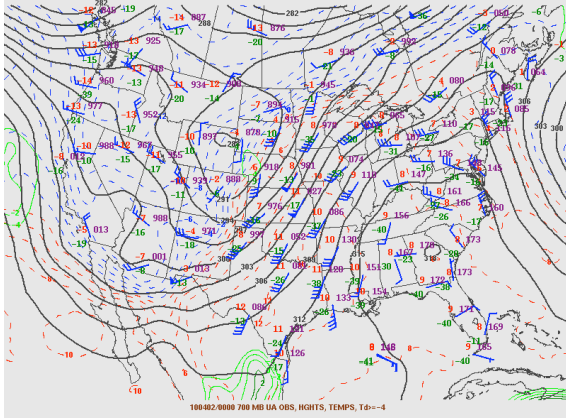


Figure 8– This is a 700 hPa upper air analysis at 0000Z 2 April 2010. This mid level is representative of altitudes on and around the Storm Peak Laboratory at Steamboat Springs.

Discussion

These experimental findings indicate that pH is independent of any singular cloud characteristic such as MVD, average number concentration, and LWC. However, the pH values are logical given a simultaneous examination of all the characteristics at once. According to Finstad et al. (1988), MVD values are highly representative indicators of drop size distribution and inherent collision efficiency of the cloud particles. This means that larger MVD values denote larger average droplet size and therefore the increased likelihood of collision and coalescence. Assuming all other things equal, a larger droplet with the same number of dissociated hydrogen ions as a smaller droplet would register as a higher pH being that the hydrogen ions would now be in a lower molar concentration due to the added water. It is thought that increased drop size would necessitate an increase in pH and it is interesting to note that this effect is realized for the most part, but it is not the only contributor to the cloud sample pH.

For instance, upon examination of *Figure 3* and *Figure 5*, it is apparent that while pH increased from Experiment 6 to Experiment 7, the associated MVD values

dropped by almost 2 μm . This can be explained by examining *Figure 4* and *Figure 6*, noting that both the number concentration and the LWC increased between Experiments 6 & 7 respectively. This indicates that there was a larger volume of water in a more dense cloud. This increase in LWC would necessitate a drop in acidity (increase pH) by overall dilution in the volume measured. It is interesting to note that this dilution by increased LWC overpowers the counter effect of decreasing the average droplet size between these two experiments, as evidenced by the drop in MVD, which would act to increase the acidity (lower pH) by increasing the ratio of dissolved acid to water.

Contrarily, between Experiments 4 & 5, there is an increase in both MVD and LWC, but a drop in average pH. This inconsistency leads to an overall inconclusive analysis as to the effects that MVD and LWC on pH and their relative magnitudes of effect compared to each other. While compared to every other experiment, the simultaneous comparison of all these values leads to what are relatively logical conclusions, the conflict between experiment 4 & 5 requires there to be additional investigation and experimental verification before any strict conclusions can be made.

However, in terms of pH data, at a value of 4.24, the average pH falls well under the threshold of danger to biodiversity. Snow, and therefore the resultant meltwater, would have a higher pH due the fact that snow scavenges less efficiently than rain and can form readily on small, non-hygroscopic particles, diluting the overall acid concentration. While meltwater concerns cannot be directly addressed by this research without supplemental data regarding snow pH (see Vanden Boogart, 2010 for snow pH discussion), the danger posed by acidic fog is still an issue as these low pH values could pose a definite risk to the both optimal plant and

insect growth regimes. This could then have a possible impact on the local biodiversity, both due to acidic fog deposition and possible meltwater concerns.

The back trajectories of the cloud samples measured indicate that at least a portion of the particles have a trans-national origin. Their low pH values denote significant levels of sulfate aerosol as defined in the context of this research. East Asian origin is a very reasonable estimation, as air mass flowing off the coast of China would likely be entraining large amounts of sulfate and other pollutant aerosol from the large-scale burning of coal that dominates Chinese energy production. On a more local scale, much of the dissolved acid detected in the cloud samples may owe its origin to entrainment of sulfate aerosols from coal plants upstream of SPL. While extent of sulfate contribution between local and trans-national sources cannot be completely determined for this event, it can be held, as evidenced by the back trajectories, that at least part of the low pH values are due to transport across the Pacific. While these finer scales were not investigated in the scope of this research, the direct effect of proximal upstream aerosol input could be modeled using the UW Non-Hydrostatic Modeling System (NMS) to ascertain the impact of local aerosol dispersion during this event.

Conclusion

The results of this series of experiments indicate that particle-size distribution and cloud pH rely on the simultaneous analysis of multiple cloud characteristics, specifically upon the interrelation between LWC, MVD, and number concentration. While correlations between individual physical characteristics and pH are statistically significant, they must be analyzed in a greater spectrum of cloud features in order to determine the dominant contributing fac-

tors. More research into separation and quantification as to the extent of influence each characteristic has on the resultant cloud pH is needed.

The overall averaged MVD value of 13.41 μm places the orographically generated cloud particles near the stratiform MVD regime. More experiments verifying or questioning this finding would be valuable to further classify MVD regimes for orographic clouds, which are currently an under-researched area.

In addition, it is highly likely that a portion of the sulfate aerosol for this event had a trans-national origin and this origin lead to the incidence of low pH. Upon investigation of finer spatial scales, it was determined that aerosol contributions from the northwestern U.S. dominated contributions from local sources (as defined by the scope of this research) over the southwestern U.S. for this particular orographic event.

References

- Borys, R. D., Lowenthal, D. H., Cohn, S. A., and Brown, W. O. J., 2003: Mountaintop and radar measurements of anthropogenic aerosol effects on snow growth and snowfall rate, *Geophys. Res. Lett.* (retrieved from scholar.google.com).
- Belyaev, V. I., 1961: Drop-size distribution in a cloud during the condensation stage of development. *Isv. Akad. Nauk SSSR, Ser. Geofiz.*, No. 11, 1209-1213 (retrieved from scholar.google.com).
- Chin, M., T. Diehl, P. Ginoux, and W. Malm, 2007: Intercontinental transport of pollution and dust aerosols: implications for regional air quality, *Atmos. Chem. Phys.* (retrieved from scholar.google.com).
- Doka et al., 2003 S.E. Doka, D.K. McNicol, M.L. Mallory, I. Wong, C.K. Minns and N.D. Yan, Assessing potential for recovery of biotic richness and indicator species due to changes in acidic deposition and lake pH in five areas of

southeastern Canada, *Environ Monit Assess* **88**
(retrieved from scholar.google.com).

EPA, 2009: Aquatic biodiversity: Air pollution and acid rain. Retrieved from EPA website: <http://www.epa.gov/bioiweb1/aquatic/airpoll.html>.

Finstad, K. J., E. P. Lozowski, and L. Makkonen, 1988: On the median volume diameter approximation for droplet collection efficiency. *J. Atmos. Sci.*, 45.

Fitzgerald, J.W., 1974 : Effect of aerosol composition on cloud droplet size distribution: A numerical study. *J. Atmos. Sci.*, **31**, 1358-1367.

Givati, A. & Rosenfeld, D., 2004: Quantifying precipitation suppression due to air pollution. *J. Appl.* (retrieved from scholar.google.com).

Jeck, R.J., 2007: Distance-scaled water concentrations versus mass-median drop size, temperature, and altitude in supercooled clouds. *J. Atmos. Sci.*, 65, 2087-2106.

Nakicenovic, N. et al., 2000: IPCC Special Report on Emissions Scenarios (Cambridge Univ. Press, Cambridge, UK (retrieved from scholar.google.com).

Park, R. J., Jacob, D. J., Field, B. D., Yantosca, R. M., and Chin, M., 2004: Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *J. Geophys.* (retrieved from scholar.google.com).

Pruppacher, H. R. and Klett, J. D., 1997: *Microphysics of Clouds and Precipitation*, Kluwer Acad., Norwell, Mass. (retrieved from scholar.google.com).