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Aerosol characteristics at Idaho Hill during the OH Photochemistry Experiment

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Abstract. This paper supports other measurements at Idaho Hill by describing measurements of the aerosol surface area, volume, and size distribution. The aerosol size distributions at the Idaho Hill site showed nuclei and accumulation modes similar to other clean continental sites. However, there were fewer large aerosols. Principal components analysis verifies that the modes present in the size distribution also had distinct time behavior. The coarse aerosol mode had two components with distinctly different time behavior: aerosols larger than 2 μm displayed a great deal of short-term variability not present in smaller aerosols. Accumulation mode aerosols had separate correlations with fresh and aged pollution, whereas smaller aerosols were correlated only with fresh pollution. Aerosols larger than 15 nm showed no diurnal variation in downslope conditions and an afternoon maximum in upslope conditions. Aerosol surface areas were too small to directly affect OH chemistry even if OH or HO_2 had fast surface losses. Aerosol volumes indicate that aerosol nitrate is not a likely candidate to balance the NO_y shortfall.

Introduction

Aerosol size distributions were measured during September 1993 as part of the Tropospheric OH Photochemistry Experiment at Idaho Hill, Colorado, for several reasons: to constrain calculations of the possible influence of heterogeneous chemistry on OH and HO_2 chemistry [*Cantrell at al.*, this issue], to constrain the NO_y budget [*Williams et al.*, this issue], and to investigate new particle formation at a remote continental site [*Marti et al.*, this issue; *Weber et al.*, in press]. Aerosol size distributions also correlate with the chemical composition of single particles, as measured with a novel mass spectrometer technique [*Murphy and Thomson*, this issue (a),]. Aerosols also provide additional tracers of the history of air parcels at the site and add context to measurements made at nearby Niwot Ridge, a site with an extensive history of gas phase chemistry measurements.

Idaho Hill is just east of the continental divide at an altitude of 3070 m (10,100 feet), approximately 25 km west of Boulder, Colorado. Meteorology at the site is described in more detail elsewhere in this issue [*Olson et al.*, this issue]. There are no major pollution sources for hundreds of kilometers to the west of the site, so downslope (west) winds bring clean continental air with little recent pollution. Upslope (east) winds at the site often bring polluted air from the Denver metropolitan area.

A variety of instruments characterized the size and chemical composition of aerosol particles at Idaho Hill. For size distributions from 15 nm to 9 μm diameter, the University of Minnesota operated a differential mobility analyzer and the NOAA Aeronomy Laboratory operated two optical particle counters. Measurements were also made by UM of the concentration of ultrafine aerosols, nominally 3 to 4 nm [*Marti et al.*, this issue]. Wetted wall denuder measurements of gas phase nitric acid and aerosol nitrate were made at the site [*Buhr et al.*, 1995], as well as filter measurements of sulfate, nitrate, ammonium, and other species. A new instrument, particle analysis by laser mass spectroscopy (PALMS), measured the chemical composition of single aerosol particles [*Murphy and Thomson*, this issue (a,b)]. This paper focuses on the results from the aerosol sizing instruments for particles larger than 15 nm.

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Sampling Techniques and Sizing Instruments

The NOAA Aeronomy Laboratory instruments collected aerosols from a 15.2 cm ID polyvinyl chloride (PVC) tube with an inlet 7.1 m off the ground. A cap over the inlet extended a few centimeters below the lip of the tube in order to exclude droplets larger than about 100 μm by gravitational settling. Flow in the PVC tube was about 900 L min^{-1}, or 0.9 m s^{-1}. There was one 90° bend before the air entered the trailer containing the optical particle counters and the PALMS instrument. Flows to the particle counters were sampled isokinetically from the large PVC tube, then passed through about 2 m of ~1 cm diameter copper tubing to the counters. To reduce losses, both counters shared the same line up to about 10 cm from the counters. Because the trailer was typically 5° to 10°C warmer than the outside, the air in the sample lines warmed and the relative humidity at the counters was usually less than 40%. Therefore all but very hygroscopic aerosols were measured dry. Transmission calculations for inertial deposition [*Liu and Agarwal*, 1974] and diffusion loss show that the combined losses for the PVC and copper tube sections should be small for particles less than 5 μm diameter. Particles smaller than 10 μm are calculated to make the turn from ambi-

ent wind into the inlet except in winds above about 13 m s^{-1} (30 mph). Particles larger than about 15 μm are lost by inertial deposition in the PVC tube and by gravitational settling in the copper tubing.

Two commercial optical particle counters were operated by the Aeronomy Laboratory. One was a PMS LasAir 1001 counter with a size range of 0.1 to 2 μm diameter and a He-Ne laser light source. The other was a Climet 208A counter with a size range of 0.35 to 9 μm and a white light source. The Climet analog output was fed into a custom all-digital pulse height analyzer board, which digitized the signal to 10 bits at 17 MHz and digitally selected peak heights. The PMS LasAir data were collected in seven size bins using its internal data collection system. Data were collected in 1-min intervals.

All optical particle counter data were corrected for dead time. Dead time corrections for the Climet were typically 5-10% and occasionally up to a factor of 2. This dead time correction is very reliable because it is due to a fixed readout time on the pulse height analyzer board rather than the Climet counter itself. In addition, the lower threshold of the Climet pulse height analysis was shifted between a low and high value every minute throughout the campaign. This checked the dead time correction by ensuring that the number of big particles didn not change when the much more numerous small particles were counted or not counted. A 0.72 ms dead time was used for the PMS LasAir dead time based on comparisons to the Climet and comparisons with and without a diluter on the PMS inlet. A diluter (Hiac/Royko) was added on September 9 to the PMS LasAir input to reduce dead time corrections. The dilution ratio, calibrated on site with both ambient aerosols and latex spheres, was found to be 9.6::1 for a nominal 10::1 diluter. After the diluter was installed, dead time corrections for the PMS LasAir were usually much less than 10%. The PMS and Climet counters tracked well in their size overlap range over large count rate fluctuations including the largest dead time corrections.

Both counters were calibrated with latex spheres (Duke Scientific) before deployment to Idaho Hill and on site. The Climet was also calibrated with a vibrating orifice generator before deployment. On the basis these calibrations, the manufacturer's calibrations were multiplied by 1.05 for the PMS LasAir and 0.93 for the Climet. The accuracy of the size calibrations is estimated to be ±10% plus index of refraction effects. Calibration with latex spheres can cause optical particle counters to underestimate the surface area of ambient aerosols by the order of 40% [*Hering and McMurry*, 1991]. The majority of the surface area was from aerosols measured by the differential mobility analyzer (DMA) system discussed below, so the use of latex spheres for size calibration does not affect calculations of possible heterogeneous reactions. The manual gain adjust to an internal light standard required by the Climet was performed every few days on site. Airflows were calibrated against a displacement flowmeter after the counters were set up at Idaho Hill. The combined calibration and stability of the flows is estimated to be ± 15%. The DMA and the optical particle counters tracked trends in surface areas extremely well in the size range covered by both, although the surface areas derived from the optical particle counters were about 25% larger than those from the DMA.

Optical particle counter data were nearly continuous from September 1 to September 30 except for the power outage after the September 13 snowstorm. Data have not been screened for the occasional traffic to the sampling site. Surface areas and volumes were calculated from the optical particle counter data by applying a spline fit to the log of the cumulative size distribution, then integrating the fit.

Fine particle size distributions were measured by the University of Minnesota with a differential mobility analyzer (DMA). The size range was usually 0.015 to 0.5 μm diameter, although the upper size limit was set to 0.26 μm on September 6 to 9 and September 25 to 27. The DMA was in a trailer located about 10 m from the optical counter instruments. Air was sampled from a downward facing inlet at a rate of about 36 L min-1 from a 2.5 cm copper tube at a height of about 5 m above ground level. Sample air was extracted from the centerline with a tube approximately 0.62 cm diameter and 50 cm long and transported to a neutralizer where an equilibrium charge was established on the particles. Sample flow rates were controlled by critical orifices. The flow rates are estimated to be accurate to within a few percent.

The DMA was operated in a scanning mode with a TSI 3760 CPC as the particle detector. A more complete description is given by *Marti et al.* [this issue]. The measurements were inverted using the method of *Hagen and Alofs* [1983] which accounted for particles carrying up to +10 elementary charges based on the charging theory of *Fuchs* [1963]. The inversion also corrected for aerosol transport losses [*Gormley and Kennedy*, 1949], losses in the DMA [*Reineking and Porstendorfer*, 1986], and the TSI 3760 CPC counting efficiency, as measured at the University of Minnesota for an ambient pressure of 700 mbar. A typical sampling interval for measurement of one size distribution was 2.5 min. Surface areas were calculated by integrating over the aerosol number distribution measured with the DMA.

Results and Discussion

Aerosol Size Distributions

The average aerosol mass distribution at Idaho Hill (Figure 1) showed three distinct modes, as is typical for a continental site. The break between the nuclei and the accumulation modes is not so clear in the average as it is on individual days, since the breakpoint varied between 40 and 150 nm for different days. The nuclei or Aitken mode was especially sharp on September 14, 15, 20, 22, 27, and 29. Figure 1 also shows size distributions for clean continental sites taken from figures in *Whitby* [1978] and *Seinfeld* [1986].

Although the Idaho Hill site is comparable to other clean continental sites for aerosols < 0.2 μm diameter, the Idaho Hill data show over a factor of 10 fewer coarse mode aerosols. Both the Seinfeld desert measurements and the Whitby measurements in the agricultural Midwest probably had more windblown dust than this site. The dip in the coarse mode near 2 μm hints at two components in the coarse mode. The presence of more than one coarse aerosol mode is confirmed in the principal components analysis below. The paucity of coarse mode aerosols is probably due to a lack of significant local sources of coarse aerosols at the Idaho Hill site. It is difficult to see how the difference between Idaho Hill and other clean continental sites could be an instrumental effect. Although it is possible that aerosols larger than 5 μm were lost in the inlet, it is difficult to find a mechanism for losing aerosols smaller than 3 μm in the inlet. Also, the drop away from other clean continental data in Figure 2 is evident at the upper end of the mobility analyzer data. The mobility analyzer used a different inlet and independent calibrations from the optical counters.

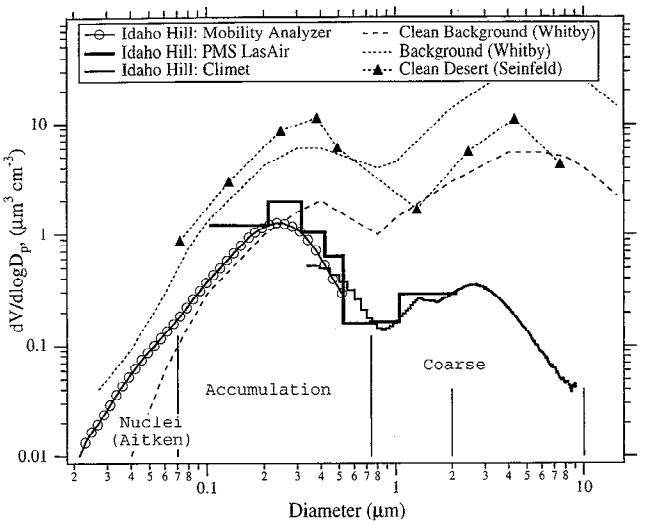

Figure 1. Average aerosol size distribution at Idaho Hill in September 1993. The vertical axis is the volume per unit logarithm of size. This emphasizes large diameter aerosols; by number, most of the aerosols are in the nuclei mode. Circles are data from the University of Minnesota mobility analyzer. Solid lines are data from optical counters operated by the Aeronomy Laboratory. Also shown are size distributions for clean continental background [*Whitby*, 1978], (dashed line) and clean desert background [*Seinfeld*, 1986], (dashed with triangles).

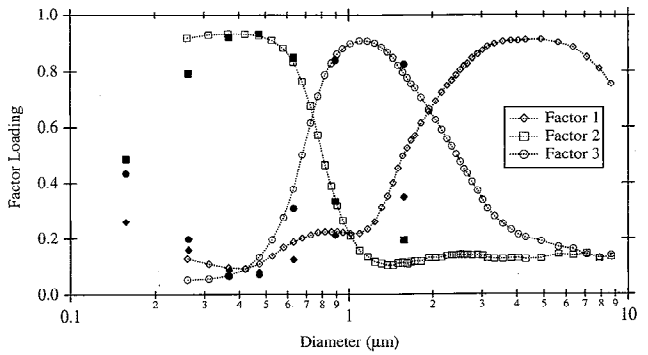

Figure 2. Factor loadings for a principal component analysis of the optical particle counter data. Open symbols are size bins from the Climet; solid symbols are size bins from the PMS LasAir. Three factors explained 86% of the variance. In this case, a factor loading is the correlation coefficient between a single size bin and each of the three linear combinations of size bins that explain most of the time history of the aerosol loadings. Note the close correspondence between these factors, derived from the time series of the data, with the modes shown in Figure 1, which contains no information about the time series.

The time series of the various aerosol modes were uncorrelated with each other. Correlations between aerosols in the 0.1 to 9 μm size range were also examined using principal components analysis (PCA) on the optical particle counter data (Figure 2). Data for each 15 min interval were collected into seven size bins for the PMS LasAir and 52 size bins for the Climet. PCA was performed using the Statistica package. The results clearly indicate three components to the aerosols measured by the optical counters. The largest three factors explain 86% of the variance; a fourth factor would only explain an additional 1% of the variance.

The accumulation mode is constrained to the same size range looking at either Figure 1 or Figure 2, since the boundary at 0.7 to 0.8 μm in Figure 2 between the accumulation mode and the smaller of the coarse aerosol modes corresponds almost exactly to the minimum in the size distribution (Figure 1). These two ways of defining the modes are completely independent. Figure 1 contains no information about the time series of aerosols of various sizes. Conversely, since each size bin is independently normalized in the PCA process, Figure 2 contains no information about the average number of aerosols of a given size.

The PCA confirms that the coarse mode is actually made up of two components, as suggested by the slight dip in Figure 1 just below 2 μm. A similar split of the coarse mode into two components has been seen in PCA of aerosol size distributions from aircraft data in the lower troposphere (M. Buhr, private communication, 1994). Short-term variability accounts for much of the difference between the two coarse mode components. The concentration of aerosols larger than 2 μm diameter was much more variable on minute timescales than would be expected from counting statistics alone. Aerosols smaller than 2 μm were not affected. The most dramatic variations were frequent "spikes" lasting 1 min or less when the concentration of aerosols larger than 2 μm increased two to tenfold. These spikes occurred in both polluted and nonpolluted air and in all wind speeds and directions. They occurred at all times of day, including times when there was nobody at the site, although they were more common during the afternoon than at night. Spikes were not significantly more common on weekends when there was more traffic on nearby four-wheel-drive roads. One possible explanation for the spikes is suspended particulates from the ground nearby, with the afternoon maximum due to decreased stability between the ground and the sampling inlet at 7 m. Figure 3 shows an example of how the size distribution changed during a spike in coarse aerosols. Below 2 μm the size distribution matches undisturbed conditions within counting statistics, but aerosols larger than 3 μm are strongly enhanced during the "spike."

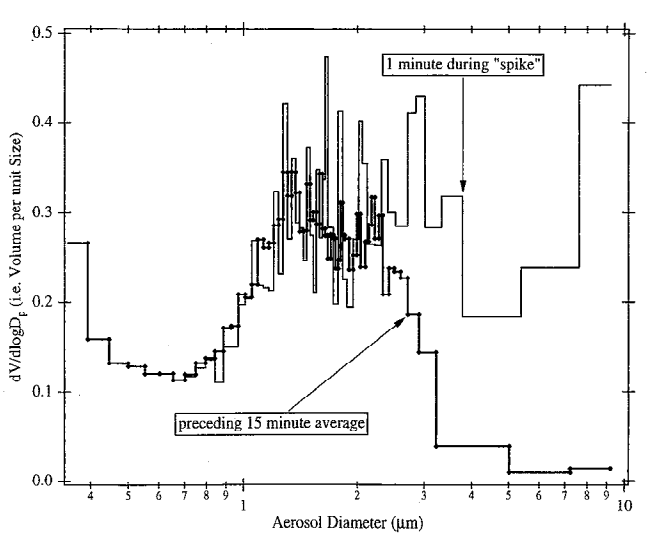

Figure 3. Aerosol size distributions for a short spike of enhanced coarse aerosols compared to an undisturbed period. See Figure 4 for the context of these distributions.

Correlations Between Aerosols and Gas Phase Species

Aerosol number, surface, and volume were sporadically correlated with various gas phase species. As an example, Figure 4 shows data on September 20. This day had downslope conditions in the morning switching to upslope near noon. All aerosol modes, along with NO_2, show a step change in less than 2 min when the wind changes from downslope to upslope. However, the correlation over the entire day is poor for two reasons: the aerosol volumes and NO_2 are uncorrelated during the morning, and the details of the afternoon rise are different. On this day, aerosol volume and NO_2 were more correlated during the upslope, but the reverse is true on some other days.

The pattern shown in Figure 4 is typical in the sense that aerosols were correlated with gas phase tracers at some times but not at other times. One explanation for the sporadic correlations between aerosols and gas phase species is that aerosols can represent either fresh pollution or the end result of photochemical processing of polluted air masses. For example, aerosols may correlate with species such as NO_2 in fresh emissions, then lose that correlation as the pollution ages. Support for this view is given by the correlation between carbon monoxide and aerosols (Figure 5). CO was correlated with accumulation mode aerosols, as expected, since both are from pollution sources. There were more accumulation mode aerosols for a given amount of CO in downslope conditions, probably because high-CO air in west winds has had much more time for secondary chemistry to add to the accumulation mode. In contrast to the accumulation mode aerosols shown in Figure 5, CO and the number of aerosols less than 0.1 μm were positively correlated only in upslope conditions. Ultrafine aerosols were correlated with gas phase H_2SO_4 [*Marti et al.*, this issue; *Weber et al.*, in press].

Overall correlations between aerosols larger than 0.1 μm and either anthropogenic or biogenic hydrocarbons were poor, even though hydrocarbons may represent an important source of aerosols. The poor correlations are understandable since small amounts of hydrocarbons may represent fairly clean air with few aerosols. But small amounts of hydrocarbons could also represent polluted air that has experienced considerable photochemical processing producing considerable amounts of aerosols. Some of the strongest correlations between aerosols and any of the gas phase species measured at Idaho Hill were between accumulation mode aerosol surface or volume and aged nitrogen species such as HNO_3 or (NO_y-NO_x). Ultrafine aerosols were strongly correlated with gas phase H_2SO_4 [*Marti et al.*, this issue].

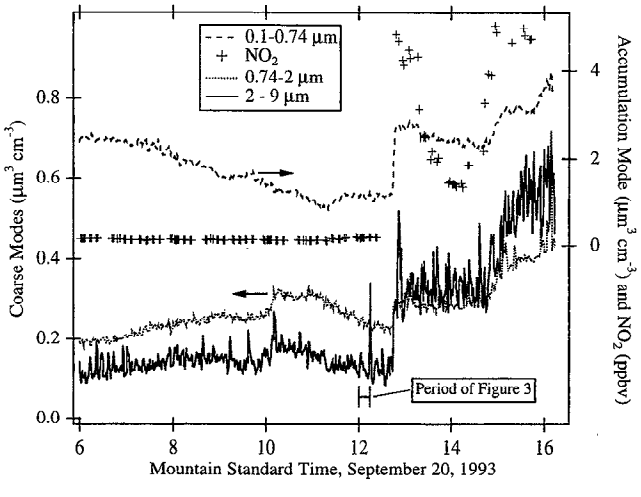

Figure 4. Time series of selected data on September 20, 1993, at Idaho Hill. There was change from west winds to upslope flow just after noon. Both the accumulation mode volume and NO_2 show a step change in less than 2 min when the wind changes. However, they are quite uncorrelated with each other or with the coarse mode aerosols during the morning. The variability of the aerosols larger than 2 μm exceeds that due to counting statistics, indicating atmospheric changes on very short length scales.

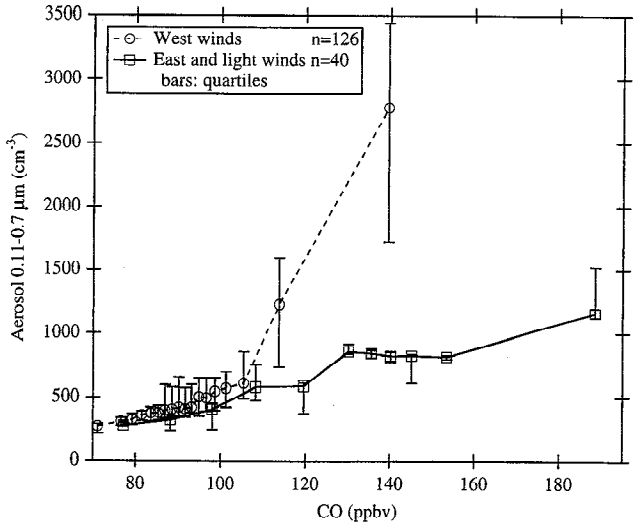

Figure 5. Accumulation mode aerosol volume as a function of carbon monoxide, separated by wind conditions. Each point is the median of either 40 or 126 three-minute averages. Correlations between CO and either coarse or nuclei mode aerosols were much weaker than the correlation shown.

Diurnal Variation

Aerosols showed distinctly different diurnal behavior depending on the size range and wind direction. Figure 6a shows the average aerosol surface area as a function of time of day. There is an afternoon and evening maximum in upslope conditions but not in downslope conditions. The average surface areas from 2300 to 0900 MST are the same for upslope and downslope conditions. This is very different than the behavior of some gas phase species such as benzene or NO_2, which are greater in upslope conditions at all times of day or night. The small peak in the downslope data just before midnight is due to a single event on September 27. The aerosol number peaks earlier in the day than the surface area or volume (Figure 6b). This is consistent with growth of aerosols through the afternoon due to daytime photochemistry.

Larger aerosols had a stronger diurnal variation in upslope winds. For the accumulation mode, the average afternoon volume (1200 to 1800 MST) was 2.0 times the average volume between 0000 and 0600 MST am. For the mode between 0.7 and 2 μm, the afternoon/nighttime ratio was 2.8; for coarse aerosols > 2 μm the ratio was 3.5. This increase in diurnal variation could be due to faster nighttime deposition of coarse aerosols compared to the accumulation mode. Except for ultrafine aerosols [*Marti et al.*, this issue], none of the size ranges showed much diurnal variation in downslope winds. In

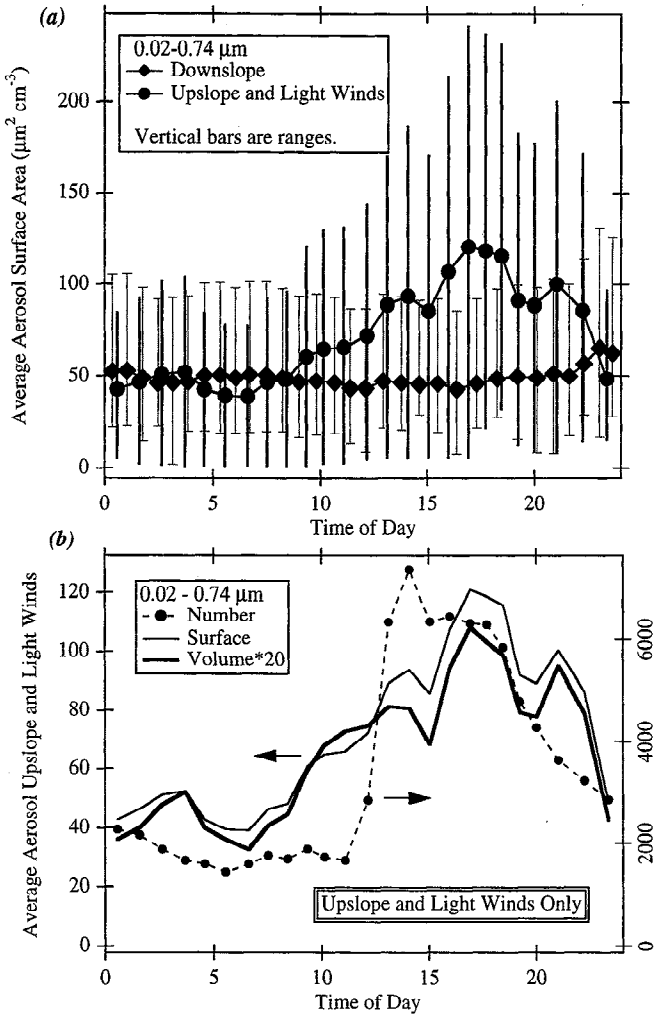

Figure 6. (a) Diurnal variation of aerosol surface area at Idaho Hill averaged from September 9 to 29. Surface areas are for aerosols 0.02 to 0.74 µm diameter and are computed from the mobility analyzer data supplemented by a small correction from optical counter data for the larger sizes. (b) Diurnal variation of aerosol number, surface, and volume at Idaho Hill for upslope and light wind conditions only. The number peaks before the surface and volume. For both plots, downslope conditions are defined as winds with a component from the west exceeding 1.8 m s^{-1}.

contrast to the size ranges discussed in this paper, the ultrafine diurnal variation was especially large in downslope winds.

NO_y Budget

Since nitrate was found primarily in aerosols larger than 0.7 µm [*Murphy and Thomson*, this issue *a*], the lack of many coarse mode aerosols at Idaho Hill means that aerosols probably contributed little NO_y (reactive odd nitrogen). In order for aerosol NO_y to balance the observed shortfall in NO_y of up to 600 parts per trillion by volume (pptv) [*Williams et al.*, this issue], four conditions would need to be met. First, the fraction of aerosols containing odd nitrogen would need to be near the upper limit set by the mass spectrometer data. Second, aerosol volumes would need to be approximately a factor of 2 larger than the nominal values from the optical counter data. Third, the aerosol NO_y would need to be in a chemical form not measured by the filter nitrate measurements made at the site. Finally, since aerosol volume was not well correlated with the NO_y shortfall, the aerosol composition would need to be rich in NO_y only during certain time periods. Together, these conditions make it unlikely that aerosol NO_y was a major contributor to the NO_y budget at Idaho Hill.

Possible Loss of OH and HO_2 to Aerosol Surfaces

Since the Idaho Hill measurements were part of the OH Photochemistry Experiment, it is of interest to assess the possible contribution of heterogeneous chemistry on OH. The aerosol surface areas were insufficient for direct effects on OH or HO_2. From Figure 6 the aerosol surface areas were almost always between 12 and 150 μm^2 cm^{-3}. Including transitional flow correction [*Wagner*, 1982], these surface areas imply a lifetime with respect to loss to aerosol surfaces of between 10 min and 2 hours if the reaction coefficient γ were 0.1. These time constants are long compared to photochemical time constants for OH and HO_2 [*McKeen et al.*, this issue], so heterogeneous chemistry was not directly important to OH at Idaho Hill even if there exist fast radical losses to surfaces. The only exception might be during the occasional episodes of fog at the site. Loss to aerosols could possibly be important for longer-lived species such as formaldehyde or nitric acid.

Conclusions

The size distribution of aerosols between 15 nm and 9 µm diameter was similar at Idaho Hill to other clean continental sites except that there were fewer large aerosols than other continental sites. Principal components analysis shows that the coarse mode has two components: a mode that varies slowly and is mostly below 2 µm diameter and a mode that varies rapidly and is mostly larger than 2 µm. This new use of PCA to examine aerosol size distributions gave very clean principal components. Aerosols were only sporadically correlated with gas phase species. Accumulation mode aerosols increased more rapidly with carbon monoxide in downslope flow than in upslope flow. This is consistent with aged pollution as the major source of accumulation mode aerosols. Aerosol surface areas were too small to directly affect OH chemistry even if OH or HO_2 had fast surface losses.

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