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Wang, Jian



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Observation of ambient aerosol particle growth due to in-cloud processes within boundary layers

Jian Wang,¹ Peter H. Daum,¹ Lawrence I. Kleinman,¹ Yin-Nan Lee,¹ Stephen E. Schwartz,¹ Stephen R. Springston,¹ Hafliði Jonsson,² David Covert,³ and Robert Elleman³

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[1] Aerosol microphysical and optical properties were measured on board the CIRPAS Twin Otter aircraft during 16 flights at the Southern Great Plain (SGP) site in northern central Oklahoma as part of the Aerosol Intensive Operation period in May 2003. Within well-mixed boundary layers on four cloudy days, vertical profiles measured on board the Twin Otter show that dry aerosol size, volume concentration, and scattering coefficients all increased with increasing altitude, whereas the total number concentration remained essentially constant. A one-dimensional model, which uses simultaneous meteorological measurements as inputs, shows that the observed increase in aerosol volume concentration with increasing altitude is consistent with in-cloud sulfate production at the top of the boundary layer. The sulfate production rate was sufficiently fast to overcome the homogenization resulting from turbulent mixing. In contrast, on cloud-free days, measurements on a second aircraft show nearly uniform aerosol volume concentrations within well-mixed boundary layers. The observed vertical gradients in aerosol volume concentration suggest that even within well-mixed boundary layers, surface measurements may not be representative of aerosols properties (e.g., loading and scattering coefficients, etc.) at elevated altitudes, especially when SO₂ concentration and cloud coverage are high.

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1. Introduction

[2] Atmospheric aerosols affect the global energy budget by scattering and absorbing sunlight (direct effects) and by changing the microphysical structure, lifetime, and coverage of clouds (indirect effects). Although it is widely accepted that anthropogenic aerosols could have significant effects on global climate, at present the magnitudes of these effects are poorly quantified. The Intergovernmental Panel on Climate Change (IPCC) estimates that the direct and indirect effects of aerosols remain the most uncertain components in the radiative forcing of climate change over the industrial era [IPCC, 2001]. One of the major chemical components in anthropogenic aerosols is sulfate. Sulfate is produced mainly through oxidation of gaseous SO₂, which can take place both in the gas phase or in liquid cloud droplets. Since 1981, a number of field studies have shown that the aqueous phase oxidation of SO₂ is an important pathway for sulfate

production [Hegg and Hobbs, 1981; Daum *et al.*, 1984; Chandler *et al.*, 1988a, 1988b; Gervat *et al.*, 1988; Seinfeld and Pandis, 1998]. It is generally agreed, based on modeling studies, that the liquid phase oxidation dominates the sulfate production on global scale [Hegg, 1985; Langner and Rodhe, 1991; Benkovitz *et al.*, 2004]. There have also been a number of studies that focused on the modification of aerosol properties through in-cloud oxidation of SO₂. Yuskiewicz *et al.* [1999] studied the effects of passage through an orographic wave-cloud on aerosol size distribution, and found substantial increases in aerosol mass concentration. Through Mie calculations based on the pre- and post-cloud size distributions, Yuskiewicz *et al.* [1999] also found that a modification of aerosol size distribution due to in-cloud sulfate production led to increases in aerosol mass scattering efficiency. Hegg *et al.* [2004] studied the aerosol mass scattering efficiency in the outflow region of marine stratocumulus clouds and found it to significantly exceed that within non-detraining air. The increase was attributed to in-cloud oxidation of SO₂ that adds sulfate to small particles and grows them to a larger, more scattering efficient size range.

[3] In this paper, we report an interesting phenomenon observed during the Aerosol Intensive Operation Period (AIOP), which was carried out in northern Central Oklahoma

¹Atmospheric Science Division, Brookhaven National Laboratory, Upton, New York, USA.

²Naval Postgraduate School, Monterey, California, USA.

³Department of Atmospheric Sciences, University of Washington, Seattle, Washington, USA.

during May 2003. Within well-mixed boundary layers (BLs) on cloudy days, vertical profiles measured on board a research aircraft show that dry particle size increased with increasing altitude while the total particle number concentration remained unchanged. The increasing particle size led to increases of both the dry aerosol volume concentration and light scattering coefficient. This increase in particle size is attributed to in-cloud sulfate production at the top of the BL. This interpretation is supported by measurements conducted on clear days, which show a nearly uniform aerosol scattering coefficient within well-mixed BLs. Simulations using a one-dimensional model also indicate that the observed vertical gradient in aerosol volume concentration is consistent with in-cloud sulfate production. This in-cloud sulfate production via aqueous oxidation of SO_2 was sufficiently fast to compete with turbulent mixing within the BL, and led to larger aerosol particle size, and higher volume concentration and scattering coefficient at higher altitudes. The observed inhomogeneity of aerosol properties suggests that even within well-mixed BLs, surface measurements may not be representative of the aerosol properties at elevated altitudes when cloud coverage is substantial.

2. Measurements

[4] From 5 to 31 May 2003, an Aerosol Intensive Operation Period (IOP) was conducted at the Southern Great Plains (SGP) Climate Research Facility [Ferrare *et al.*, 2006]. The SGP Climate Research Facility covers an area of $\sim 142,000 \text{ km}^2$ in North Central Oklahoma and South Central Kansas, and is the first and largest field measurement site established by the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) Program. The central facility at SGP is located at 36.60°N and 97.48°W , with an altitude of 319 m. Since 1996, continuous measurements have been carried out, which provide long time records of aerosol, cloud, radiation, and atmospheric properties at SGP. During the IOP, a suite of aerosol instruments were deployed on board a Twin Otter aircraft to characterize aerosol properties and their vertical profiles and horizontal variability at the SGP central facility site. The measurements included particle number concentrations, aerosol size distributions, and various aerosol optical properties. The Twin Otter aircraft, operated by the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) based at Marina, California [Bluth *et al.*, 1996], performed 16 research flights out of the Ponca City airport, Oklahoma (32 km east of the SGP central facility). Most of the flight patterns were centered on the central facility at the SGP site. Data from the second aircraft, a Cessna 172-N is also used in this study. As part of the In situ Aerosol Profiling (IAP) program, the Cessna has regularly (2–3 times per week) measured aerosol optical property profiles over the SGP site since 2000 [Andrews *et al.*, 2004]. The aerosol optical properties measured on Cessna include aerosol light scattering and absorption coefficients. During the IOP the Cessna flew 14 of its standard profile flights. Among all six cases examined in this study, the profiles were conducted at the SGP central facility, except for the ascent on 22 May 2003, which was carried out at 36.69°N , 96.63°W , 80 km east of the SGP central facility.

2.1. Measurements on Twin Otter

[5] The number concentration of particles larger than 10 nm in diameter was measured by a Condensation Particle Counter (Model 3010, TSI Inc., Minneapolis, Minnesota), and aerosol size distributions were measured by a Scanning Mobility Particle Sizers (SMPS) and a PCASP. A detailed description of the SMPS is given by Wang *et al.* [2003]. The major components of the SMPS are a cylindrical Differential Mobility Analyzer (Model 3081, TSI Inc., Minneapolis, Minnesota) and a condensation particle counter (Model 3010, TSI Inc., Minneapolis, Minnesota). Prior to measurements, the relative humidity (RH) of aerosol sample was reduced to below 25% inside a Nafion drier. Aerosol size distribution ranging from 20 nm to ~ 800 nm was measured every 73 seconds. The SMPS was calibrated during the IOP using polystyrene latex standards. Data from the SMPS were reduced using the data inversion procedure described by Collins *et al.* [2002].

[6] In addition to the SMPS, a Passive Cavity Aerosol Spectrometer Probe (PCASP, PMS Inc., Boulder, Colorado) with a SPP-200 data system (DMT Inc., Boulder, Colorado) was deployed on board the Twin Otter. The PCASP measured particles in the diameter range of $0.11\text{--}2.69 \mu\text{m}$ at a time resolution of 1 Hz. During the AIOP, the de-icing heater of the PCASP was turned on to dry aerosol samples. Strapp *et al.* [1992] showed that the de-icing heater effectively removes water associated with particles when the ambient RH is below 79%. At higher ambient RHs, the de-icing heater does not dry hydrated particles completely. The PCASP was calibrated using spheres of three different refractive indices (RI = 1.33, 1.42 and 1.58) using the methodology described by Liu *et al.* [1992]. The bin diameters used in this study were based on the RI = 1.58 calibration.

[7] Aerosol light scattering coefficients were determined using a 3-wavelength integrating nephelometer (model 3563, TSI, St. Paul, Minnesota). The nephelometer was calibrated against particle-free air and CO_2 before and at multiple times during the field deployment and was zeroed with particle-free air before each flight. The TSI nephelometer was operated at a flow rate of 30 l/m with its inlet heater operated at $\sim 35^\circ\text{C}$. This resulted in the RH inside the instrument being considerably lower than the ambient RH. The RH inside the TSI nephelometer ranged from near 0 to 35% depending on the ambient RH. Aerosol light absorption $\sigma_{\text{ap}}(\lambda)$ was measured using an improved version of the three-wavelength filter-based Particle Soot Absorption Photometer (PSAP, $\lambda = 467, 530, 660 \text{ nm}$, Radiance Research, Seattle, Washington) described by Virkkula *et al.* [2005]. The data reduction and correction scheme of Bond *et al.* [1999] was applied. Because $\sigma_{\text{ap}}(\lambda)$ was measured just downstream of the TSI nephelometer, it was measured under subambient RH (i.e., the same RH as inside the TSI nephelometer to minimize RH-dependent artifacts due to the filter substrate). The nephelometer and PSAP sampled aerosol from a shrouded intake whose inlet transmission efficiency was characterized in airborne and wind tunnel experiments by Hegg *et al.* [2005]. They found no appreciable loss in transmission efficiency for particles smaller than $\sim 3.5 \mu\text{m}$ diameter at the typical Twin Otter speed of 50 m/s. For larger particles, the efficiency decreases rapidly but levels off at an efficiency of slightly

higher than 0.6 for particles $5.5 \mu\text{m}$ through the limit of their measurements at $9 \mu\text{m}$. During the AIOP, a $1 \mu\text{m}$ impactor was installed upstream of the nephelometer and PSAP only during the first two hours of the flight on 25 May.

2.2. Measurements on Cessna

[8] The major instruments of the aerosol package on the Cessna included an integrating nephelometer (Model 3563, TSI Inc., Minneapolis, Minnesota) and a single wavelength Particle Soot Absorption Photometer (PSAP, Radiance Research, Seattle, Washington). Details of the aerosol package on the airplane can be found in *Andrews et al.* [2004]. Inside the airplane, the aerosol sample was gently heated, if necessary, to maintain a RH below 40%. Downstream of the heater, there was a $1 \mu\text{m}$ impactor upstream of the aerosol instruments (corresponding to a geometric size cut of approximately $0.79 \mu\text{m}$) to eliminate uncertainties due to particle losses and inlet transmission inefficiencies for larger particles. The uncertainties in the IAP nephelometer measurements depend primarily on the flight segment length (~ 10 min for the four highest levels, ~ 5 min for the five lowest levels) and on the amount of aerosol present [*Clarke et al.*, 2002]. For very clean ($\sigma_{\text{sp}} \sim 1 \text{ Mm}^{-1}$) upper flight levels the uncertainty in the scattering measurement is approximately 40%, whereas for flight levels with higher aerosol loadings ($\sigma_{\text{sp}} > 20 \text{ Mm}^{-1}$) the uncertainty is less than 10%.

3. Results and Discussion

[9] In this section, we first show vertical distributions of aerosol and meteorological properties measured within well-mixed BLs for four cases identified from Twin Otter flights. In addition, two cases of well-mixed BLs were found from Cessna measurements on clear days, and the vertical profiles are presented. A one-dimensional model is then developed to estimate the vertical gradient of aerosol volume concentration using measured meteorological parameters, which is then compared to the observations.

3.1. Case I of Well-Mixed BLs on Cloudy Days, 25 May 2003

[10] The Twin Otter took off at local time 13:53 (local daylight saving time, GMT - 5hrs is used throughout this paper) and flew a series of overpasses over the SGP central facility at various altitudes. In particular, four overpasses within the BL ranged from 950 m above ground level (AGL) at 15:07 to 180 m AGL at 15:32. During this period, the sky was covered with boundary layer stratus with cloud base height near 1000 m AGL. Data from a TSI total sky imager, which is a part of the routine measurements conducted at SGP, shows that cloud coverage decreased from 95% at 14:30 to 30% at 17:00. The average cloud coverage during the period of the four overpasses was $\sim 40\%$.

[11] Figure 1 presents aerosol and meteorological parameters measured on board the Twin Otter during multiple overpasses over the SGP site on 25 May. The uniform potential temperature profile shown in Figure 1b indicates the BL to be well-mixed. The uniform potential temperature profile is confirmed by the temperature measurements on board the radiosondes launched at 12:30 and 18:30. The existence of a well-mixed BL is also supported by the uniform profiles of water vapor mixing ratio and total particle concentration (Figure 1c). The total particle number concentration shown in Figure 1c was measured by a Condensation Particle Counter, and corrected to standard temperature and pressure (STP, 1013.25 hpa and 273.15 K, all aerosol extensive properties are reported at STP throughout this paper). On account of the decreasing temperature with altitude, the ambient RH increased with altitude and reached 100% at about 1000 m, consistent with the presence of clouds.

[12] The aerosol number concentration integrated from SMPS aerosol size distribution was uniform within the BL at about 2000 cm^{-3} (Figure 1d). This concentration agrees well with the direct measurements of the CPC. In contrast to the aerosol number concentration, the volume-average particle diameter (\bar{D}_{pv}) derived from SMPS size distribution increased with increasing altitude within the BL (Figure 1e). As a result, the corresponding volume concentration of particles smaller than $0.8 \mu\text{m}$ in diameter ($V_{0.8\mu\text{m}}$) increased by $1.3 \mu\text{m}^3/\text{cm}^3$ from $5.6 \mu\text{m}^3/\text{cm}^3$ at 180 m to $6.9 \mu\text{m}^3/\text{cm}^3$ at 950 m (Figure 1e). It is important to point out that the increases of particle size and aerosol volume concentration were due to an increase in particle dry mass and not due to water uptake. Despite the steady increase in ambient RH with altitude, the RH inside the SMPS was much lower since the aerosol sample was dried by a Nafion drier. In fact, the RH measured inside the SMPS actually decreased slightly with increasing altitude and was below 23% throughout the BL (Figure 1f). As the low RH inside the SMPS was expected to remove all water associated with aerosol particles, the measured size distributions were representative of dry aerosol. The aerosol volume concentration ($D_p < 0.8\mu\text{m}$) derived from PCASP size distribution also showed similar increases with increasing altitude (Figure 1g). Below 650 m, the ambient RH was lower than 73%, at which point the PCASP de-icing heater was expected to effectively dry aerosol particles prior to measurements [*Strapp et al.*, 1992]. The increasing dry volume concentration measured by the PCASP below 650 m supports the conclusion that the increase of aerosol volume concentration was due to the increase in aerosol dry mass rather than a RH effect. The number concentration integrated from the PCASP size distribution also increased with increasing altitude (Figure 1g).

[13] Vertical profiles of aerosol scattering (σ_{sp}) and absorption (σ_{ap}) coefficients are shown in Figure 1h. Similar

Figure 1. Vertical profiles of aerosol and meteorological measurements during Twin Otter multiple overpasses of SGP site on 25 May 2003. (a) Flight time and ambient temperature; (b) potential temperature and ambient RH; (c) water vapor mixing ratio and total particle number concentration measured by CPC; (d) particle number concentration derived from SMPS size distribution; (e) particle volume concentration and volume averaged particle diameter derived from SMPS size distribution; (f) RH inside nephelometer and SMPS; (g) volume concentration of particle smaller than $0.8 \mu\text{m}$ and number concentration derived from PCASP size distribution; (h) dry aerosol scattering and absorption coefficients corrected to STP.

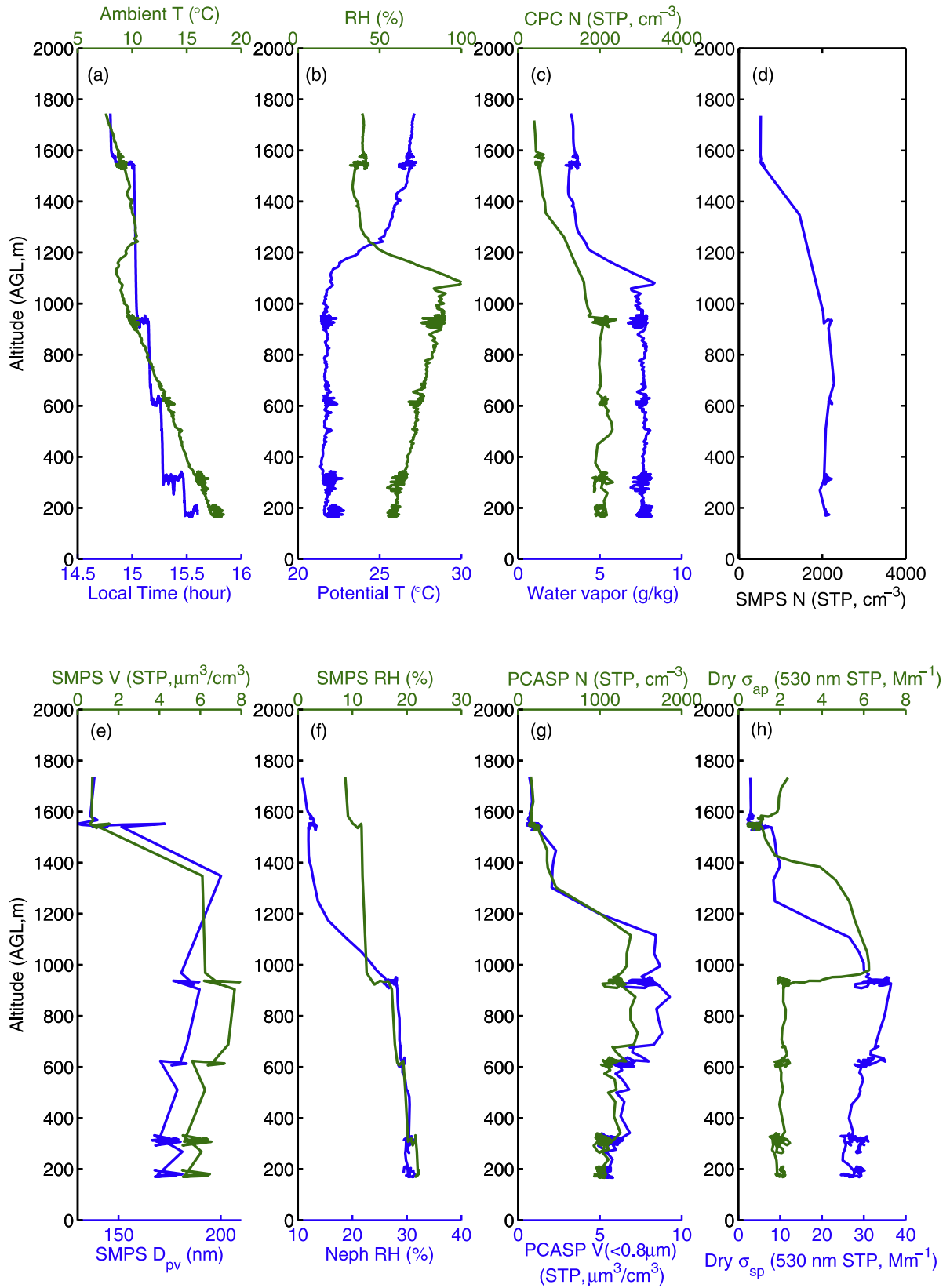


Figure 1

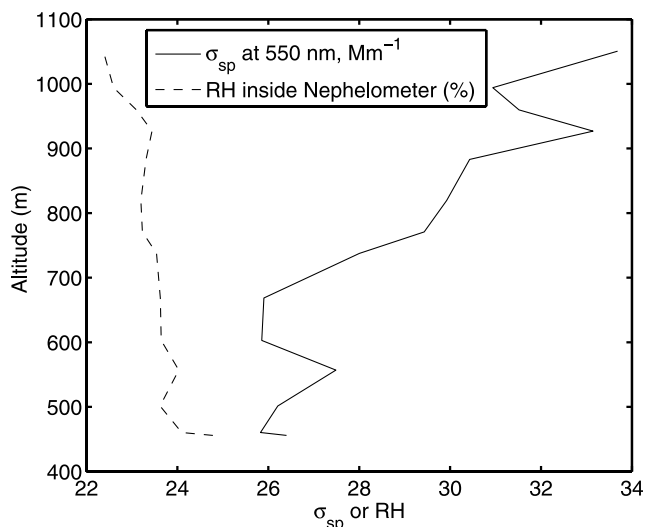


Figure 2. Submicrometer scattering coefficient and RH inside the nephelometer during an ascent of Cessna aircraft on 25 May 2003.

to the RH inside the SMPS, the RH inside the 3-wavelength nephelometer was very low ($<31\%$) and decreased slightly with altitude within the BL. As expected, the dry aerosol scattering coefficient increased with increasing altitude as a result of increasing particle diameter. In contrast, the aerosol absorption coefficient remained constant throughout the BL. During the overpasses, a $1\ \mu\text{m}$ impactor was installed upstream of the nephelometer. Based on nephelometer measurements, the aerosol dry scattering coefficient increased by $6.8\ \text{Mm}^{-1}$ from $26.2\ \text{Mm}^{-1}$ at $180\ \text{m}$ to $33.0\ \text{Mm}^{-1}$ at $950\ \text{m}$. Using the dry aerosol size distribution measured by the SMPS and assuming a particle refractive index of 1.53, we carried out Mie scattering calculation [Bohren and Huffman, 1983], and found the increase of submicron aerosol scattering coefficient from $180\ \text{m}$ to $950\ \text{m}$ to be $6.7\ \text{Mm}^{-1}$, in agreement with the increase derived from the nephelometer measurement.

[14] Since measurements at various altitudes were carried out at different times, it is possible that the observed differences in aerosol particle size, volume concentration, and scattering coefficient were due to the variations of aerosol properties over the sampling period rather than to increasing altitude. This possibility is addressed by examination of the data from the Cessna, which performed a quick ascent from $400\ \text{m}$ to $1100\ \text{m}$ near the SGP site, from 15:28 to 15:31. The measured aerosol light scattering coefficient and the RH inside the Cessna nephelometer are shown in Figure 2. The RH inside the Cessna nephelometer was below 25% within the BL, and decreased slightly with increasing altitude. Similar to the Twin Otter measurements, the aerosol scattering coefficient measured on board the Cessna also increased with increasing altitude. It is worth noting that during the overpasses of the Twin Otter, measurements started at high altitudes and concluded near the surface, whereas the time sequence of the measurements at different altitudes was opposite during the Cessna's ascent. The increasing aerosol scattering coefficient with increasing altitude observed during both the overpasses and the ascent

indicates the observed differences were due to an actual vertical gradient of aerosol properties rather than to any temporal variations.

[15] The observed increases of aerosol particle size and volume concentration with increasing altitude suggest either removal of aerosol volume near the surface or production at the top of the BL. For such processes to be manifested in a vertical gradient requires also that the removal or production process be sufficiently rapid to compete with turbulent mixing within the BL, which tends to homogenize the aerosol volume concentration. Since there was no precipitation observed on 25 May, and the dry deposition of submicron aerosol particles is negligible on the time scale of turbulent mixing [Seinfeld and Pandis, 1998], the observed vertical gradients of dry aerosol size and volume concentration are attributed to in-cloud sulfate production at the top of the BL. Since sulfate is not absorptive, this interpretation is supported by the observed uniform aerosol absorption coefficient within the BL (Figure 1h). In addition, the observed increase with altitude of total number concentration obtained from the PCASP size distribution, is also consistent with this interpretation. The PCASP has a lower detection limit of $\sim 110\ \text{nm}$ in particle diameter. In-cloud sulfate production grew some of the small particles, which were originally below the PCASP detection limit, to larger sizes that could be detected by the PCASP. As a result, the particle number concentration measured by the PCASP increased with increasing altitude within the BL.

3.2. Cases II, III, and IV of Well-Mixed BLs on Cloudy Days

[16] Three additional cases of well-mixed BLs are found on 22, 20, and 17 May, among the 16 Twin Otter flights. The well-mixed BLs are identified by uniform potential temperature, water vapor mixing ratio, and total particle number concentration within the BLs. On 22 May, the cloud base height was about $1000\ \text{m}$. The cloud coverage decreased from 45% at 11:40 to about 10% at 12:30. The Twin Otter performed an ascent over the SGP site from 12:14 to 12:21. On this day, the average cloud coverage during the ascent was 15% . Similarly on 20 May, the Twin Otter performed a spiral descent from 13:00 to 13:13. Cloud coverage was 100% at 11:00, and gradually decreased to 0% at 14:30. During the descent, the average cloud coverage was 30% . Vertical profiles of aerosol and meteorological properties measured on 22 and 20 May are shown in Figures 3 and 4, respectively. For both cases, the aerosol scattering coefficients and volume concentrations derived from PCASP size distributions increased with increasing altitude whereas the aerosol absorption coefficients remained constant within the BL, all of which is consistent with in-cloud sulfate production. The size distribution measured by the SMPS is not shown for these two cases due to the low time resolution of SMPS measurements and the short periods of the ascent and descent on the two days.

[17] On 17 May, the sky was overcast with stratus cloud. The cloud coverage was near 100% throughout the day, and the BL was well-mixed as indicated by the uniform potential temperature within the BL. Figure 5 shows measurements during a quick descent from 16:48 to 16:53. The cloud base height was at $\sim 730\ \text{m}$. Unlike the previous three cases, no appreciable increase with altitude was observed in

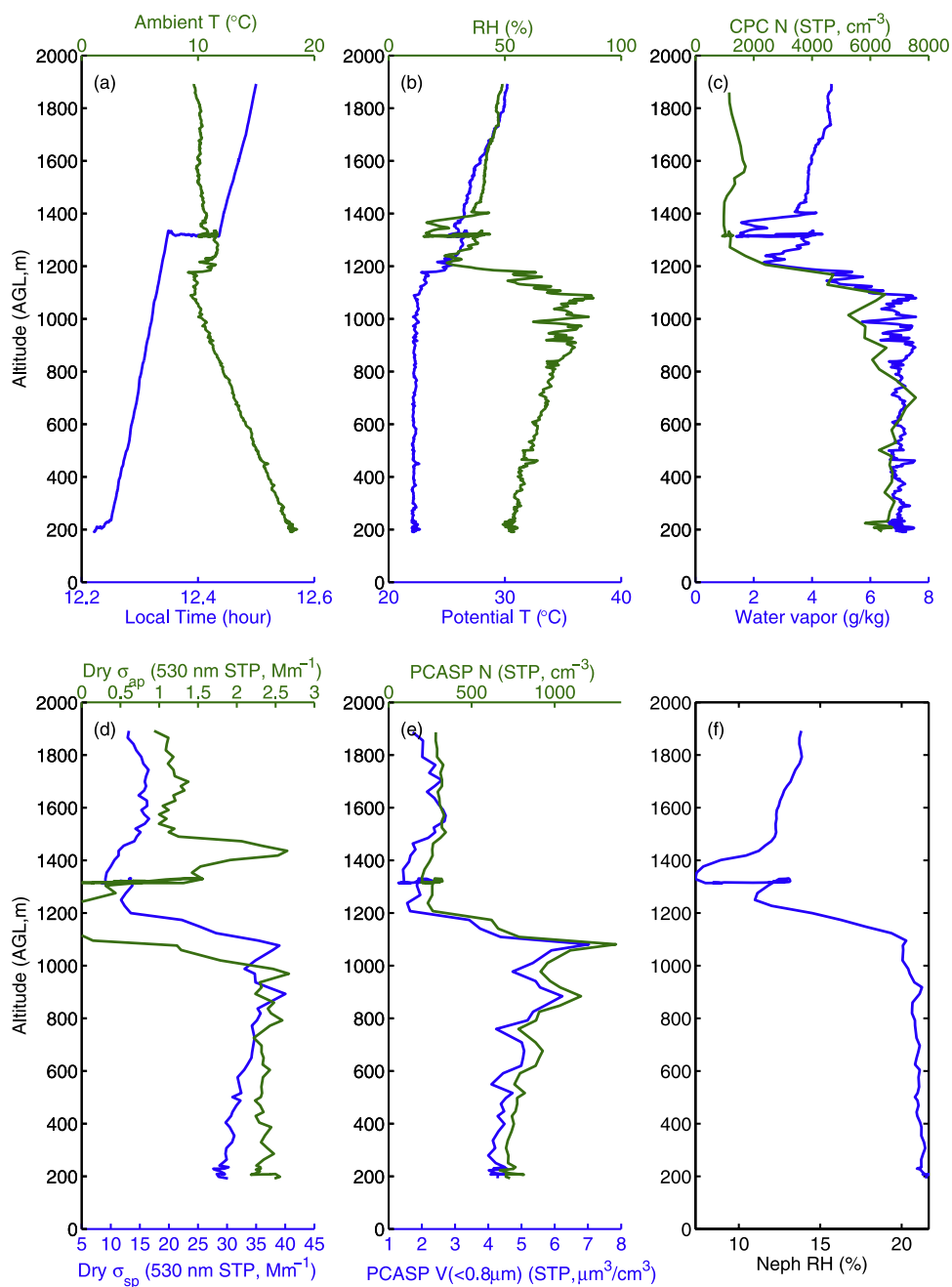


Figure 3. Vertical profiles of aerosol and meteorological measurements during a Twin Otter ascent on 22 May 2003. (a) Flight time and ambient temperature; (b) potential temperature and ambient RH; (c) water vapor mixing ratio and total particle number concentration measured by CPC; (d) dry aerosol scattering and absorption coefficients corrected to STP; (e) volume concentration of particle smaller than $0.8\ \mu\text{m}$ and number concentration derived from PCASP size distribution; (f) RH inside nephelometer.

either the dry aerosol scattering coefficient or the aerosol volume concentration ($V_{0.8\mu\text{m}}$) derived from the PCASP size distribution below the cloud. The increase of PCASP $V_{0.8\mu\text{m}}$ inside the cloud at an altitude of $\sim 750\ \text{m}$ was likely due to particle water uptake as the de-icing heater does not dry particles completely at high ambient RHs [Strapp *et al.*, 1992]. It is expected that the ambient concentration of SO_2 will decrease as in-cloud production of sulfate proceeds. As a result, when there is insufficient SO_2 available for in-

cloud sulfate production, turbulent mixing within the BL will then result in a constant aerosol volume concentration throughout the subcloud layer. Compared to the other three cases, the cloud coverage on 17 May was substantially higher, nearly 100% for more than 24 hours prior to the descent. One of the possible explanations for the absence of a vertical gradient in aerosol volume concentration is that the high cloud coverage over an extended time period could lead to the depletion of SO_2 or H_2O_2 as a result of in-cloud

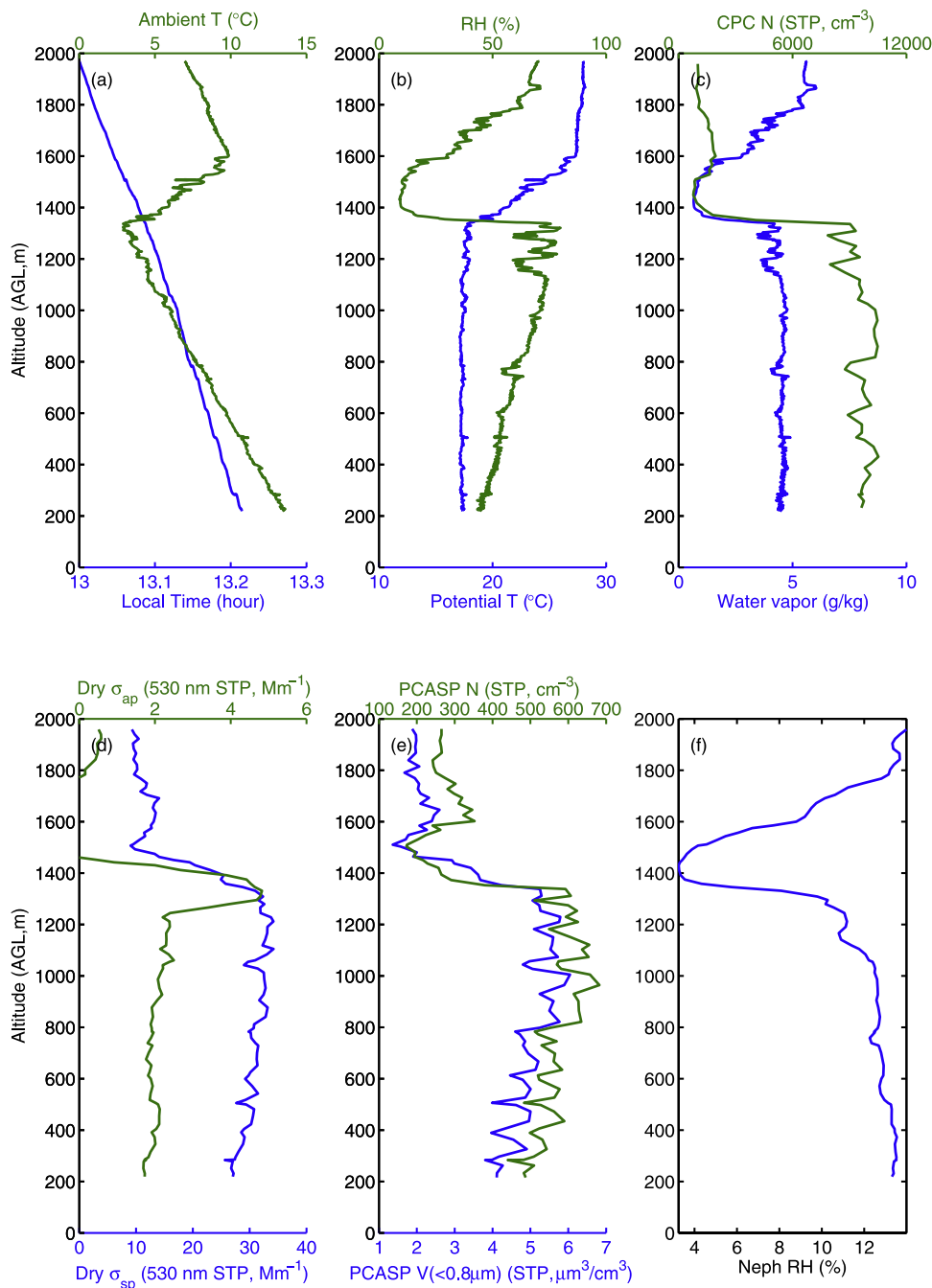


Figure 4. Vertical profiles of aerosol and meteorological measurements during a Twin Otter descent on 20 May 2003. (a) Flight time and ambient temperature; (b) potential temperature and ambient RH; (c) water vapor mixing ratio and total particle number concentration measured by CPC; (d) dry aerosol scattering and absorption coefficients corrected to STP; (e) volume concentration of particle smaller than $0.8 \mu\text{m}$ and number concentration derived from PCASP size distribution; (f) RH inside nephelometer.

reaction. The absence of a vertical gradient could also be due to an increase in vertical eddy diffusivity. Without SO_2 measurements, it is difficult to pinpoint the exact reason. The four Twin Otter cases, all of which were on cloudy days, are summarized in Table 1. To further test our explanation of in-cloud sulfate production, vertical profiles of aerosol properties measured on clear days are presented and discussed in the following section.

3.3. Cases of Well-Mixed BLs on Cloud-Free Days

[18] Two cases of well-mixed BLs on nearly cloud-free days were identified from potential temperature measurements on board the Cessna aircraft on 29 May and 5 May. In both cases, no appreciable increase in aerosol scattering coefficient with increasing altitude was observed. Figure 6 shows examples of the vertical profiles of the 29 May flight. The Cessna performed an ascent from 450 m to 1100 m

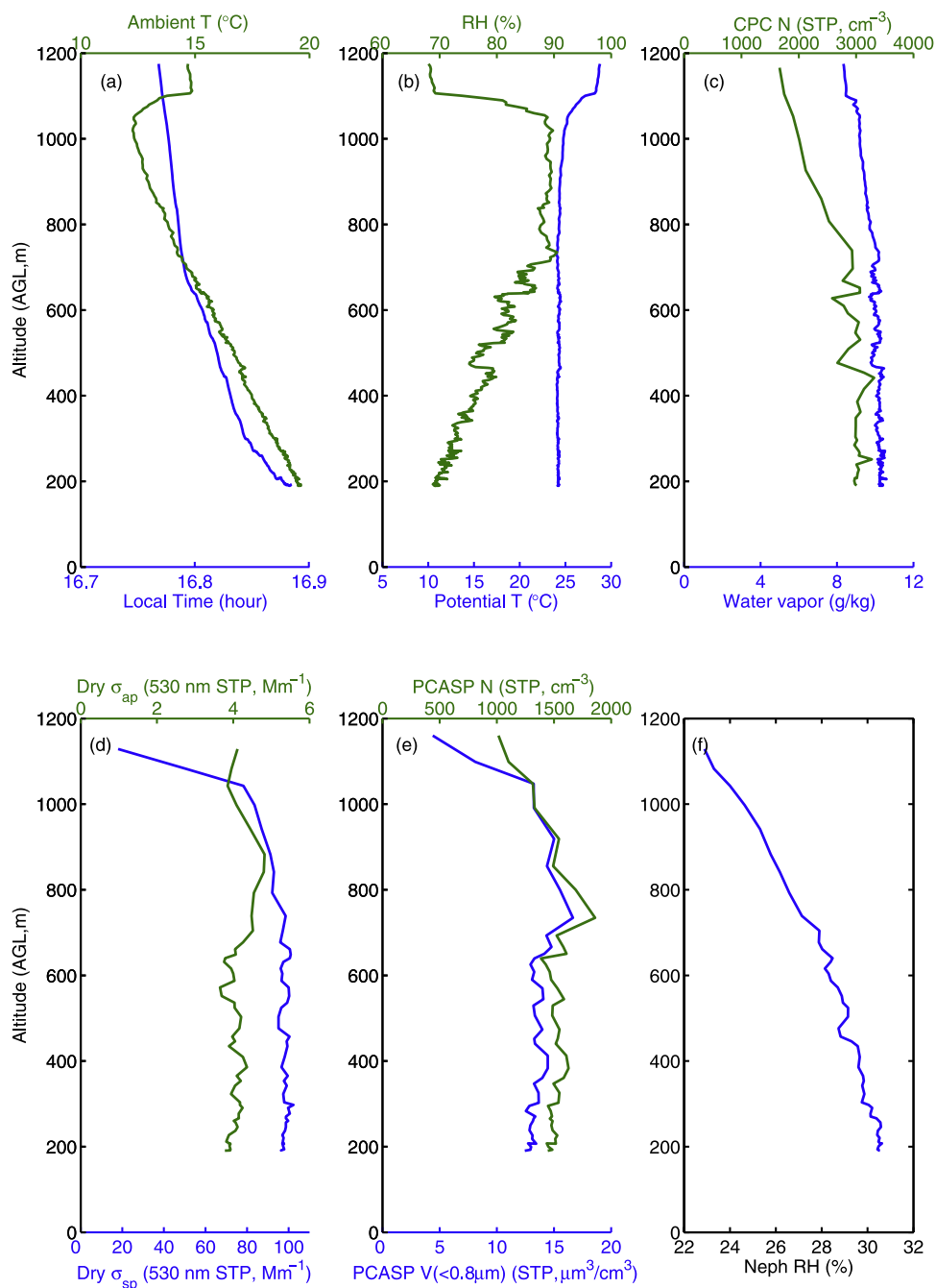


Figure 5. Vertical profiles of aerosol and meteorological measurements during a Twin Otter descent on 17 May 2003. (a) Flight time and ambient temperature; (b) potential temperature and ambient RH; (c) water vapor mixing ratio and total particle number concentration measured by CPC; (d) dry aerosol scattering and absorption coefficients corrected to STP; (e) volume concentration of particle smaller than $0.8 \mu\text{m}$ and number concentration derived from PCASP size distribution; (f) RH inside nephelometer.

around 11:00. The RH inside the nephelometer was below 20% during the ascent. In contrast to the three cases on cloudy days, the dry aerosol scattering coefficient remained nearly constant within the BL. The absence of a vertical gradient in dry aerosol scattering coefficient on these clear days lends strong support to our conclusion that the increases in aerosol volume concentrations and scattering coefficients observed on cloudy days were due to in-cloud sulfate production.

3.4. Estimation of the Vertical Gradient of Aerosol Volume Concentration Using a One-Dimensional Model

[19] To further examine our explanation that the observed vertical gradient in aerosol volume concentration was a result of in-cloud sulfate production, we developed a one-dimensional model to determine if in-cloud sulfate production was sufficiently fast to compete with turbulent mixing. The vertical gradient of aerosol volume concentration was estimated using the one-dimensional model and measured

Table 1. Summary of the Six Cases of Well-Mixed BLs From the Twin Otter and Cessna Measurements

Date	Platform	Period (Local Time)	σ_{sp}^a Mm ⁻¹	$\Delta\sigma_{sp}^b$ Mm ⁻¹	V_c^c $\mu\text{m}^3/\text{cm}^3$	ΔV_c^d $\mu\text{m}^3/\text{cm}^3$	Cloud Coverage
17 May	Twin Otter	16:48–16:53	97.9	1.8	13.1	0.74	~100%
20 May	Twin Otter	13:00–13:13	29.5	5.6	5.0	1.3	30%
22 May	Twin Otter	12:13–12:30	32.7	6.9	4.9	1.2	15%
25 May	Twin Otter	14:48–15:36	29.6	6.8	6.4	1.3	40%
5 May	Cessna	15:37–16:12	6.1	−0.2	N/A	N/A	0%
29 May	Cessna	11:00–11:06	27.4	−1.9	N/A	N/A	0%

^aAverage scattering coefficients measured by Twin Otter nephelometer (530 nm) or Cessna nephelometer (550 nm) within BLs.

^bIncreases of the measured scattering coefficient from near surface to the top of BL.

^cAverage volume concentration of particles smaller than 0.8 μm derived from SMPS size distribution (25 May) and PCASP size distributions 17, 20, and 22 May) within BLs.

^dIncreases in volume concentration of particles smaller than 0.8 μm from near surface to the top of BL.

meteorological parameters, and compared to the observed gradient for the first case on 25 May. Simulations were not carried out for the two other cases (20 and 22 May) when appreciable vertical gradients were observed, because cloud liquid water path measurements were not available on these days.

[20] Figure 7 shows a schematic of the one-dimensional model that describes the aerosol volume concentration within a BL 100% covered with clouds. The cloud base height and the cloud thickness are represented by l_b and l_c , respectively. Under the assumption that the BL is horizontally homogeneous, and the initial aerosol volume concentration is uniform, the aerosol volume concentration $c(z, t)$ ($\mu\text{m}^3/\text{cm}^3$) can be described as a function of altitude z and time t by the following equation:

$$\frac{\partial c}{\partial t} = K_{zz} \frac{\partial^2 c}{\partial z^2} \quad (1)$$

with initial condition $c(z, 0)$, where K_{zz} is the eddy diffusivity in the vertical direction. Since the dry deposition of submicron particles is negligible on the time scale considered here, the boundary conditions of equation (1) can be written as:

$$\begin{aligned} K_{zz} \frac{\partial c}{\partial z} \Big|_{z=l_b} &= F \\ K_{zz} \frac{\partial c}{\partial z} \Big|_{z=0} &= 0 \end{aligned} \quad (2)$$

where F is the flux of aerosol volume due to in-cloud sulfate production. For the above boundary conditions, the solution of equation (1) is [Carslaw and Jaeger, 1959]:

$$\begin{aligned} c(z, t) = c_0 + \frac{F_t}{l_b} + \frac{Fl_b}{K_{zz}} \left[\frac{3z^2 - l_b^2}{6l_b^2} \right] - \frac{2Fl_b}{\pi K_{zz}} \left[\sum_{n=1}^{+\infty} \frac{(-1)^n}{n^2} \right. \\ \left. \cdot \exp\left(\frac{-K_{zz}n^2\pi^2 t}{l_b^2}\right) \cos \frac{n\pi z}{l_b} \right] \end{aligned} \quad (3)$$

The last term of equation (3) decreases exponentially with increasing t . When $t > 2\frac{l_b^2}{\pi^2 K_{zz}}$ (22 minutes evaluated using parameters listed in Table 2), equation (3) can be approximated using the following asymptotic solution:

$$c(z, t) = c_0 + \frac{F_t}{l_b} + \frac{Fl_b}{K_{zz}} \left[\frac{3z^2 - l_b^2}{6l_b^2} \right] \quad (4)$$

Equation (4) shows that in-cloud sulfate production together with downward mixing of this sulfate through eddy diffusion results in increases of the aerosol volume concentration with both time and altitude.

[21] The aerosol volume flux at the cloud base, F , is estimated by the following approach. Considering an area S at the cloud base and assuming there is no air exchange (detrainment or entrainment) at the cloud top, we can estimate the average residence time of an air parcel within the cloud as:

$$t_r = \frac{V}{Q} = \frac{S \cdot l_c}{Q} \quad (5)$$

where V is the volume of cloud over the area S , l_c the cloud thickness, and Q the air exchange rate at the cloud base over the area of S . It is worth noting that the assumption of no air

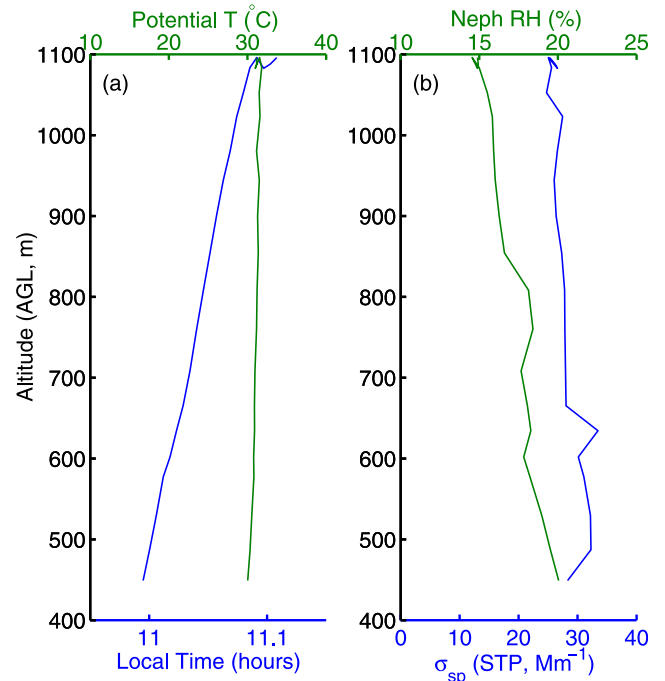


Figure 6. Vertical profiles of aerosol and meteorological measurements during an ascent of the Cessna aircraft on 29 May 2003. (a) Flight time and potential temperature; (b) dry scattering coefficient measured at 550 nm and RH inside the nephelometer.

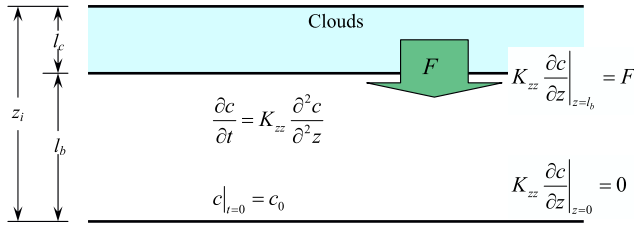


Figure 7. Schematic of a one-dimensional model that describes aerosol volume concentration within well-mixed BLs covered with clouds.

exchange at cloud top may not be accurate under certain circumstances. At the cloud base, the particle volume flux is:

$$F = \frac{(c'(l_b, t) - c(l_b, t))Q}{S} \quad (6)$$

where $c'(l_b, t)$ and $c(l_b, t)$ represent the aerosol volume concentration of air parcels exiting (detrainment) and entering (entrainment) clouds, respectively (Figure 8). Let R_c denote the sulfate volume production rate inside the cloud, $c'(l_b, t)$ can be written as:

$$c'(l_b, t) = c(l_b, t - t_r) + R_c t_r \quad (7)$$

Combining equations (6) and (7), we can write the aerosol volume flux as:

$$F = \frac{(c(l_b, t - t_r) + R_c t_r - c(l_b, t))Q}{S} \quad (8)$$

From equation (4), the following equation can be derived:

$$c(l_b, t - t_r) - c(l_b, t) = -\frac{F t_r}{l_b} \quad (9)$$

Inserting equation (9) into equation (8), and noting that $t_r = \frac{V}{Q} = \frac{S \cdot l_c}{Q}$, we have:

$$F = \frac{\left(R_c t_r - \frac{F t_r}{l_b}\right)Q}{S} = \frac{\left(R_c - \frac{F}{l_b}\right)S \cdot l_c Q}{S} = \left(R_c - \frac{F}{l_b}\right)l_c \quad (10)$$

Rearranging equation (10), we can write the particle volume flux as:

$$F = R_c \frac{l_c l_b}{l_c + l_b} \quad (11)$$

[22] Aqueous oxidation of SO_2 in liquid water clouds occurs by two main reactions, oxidation by hydrogen peroxide, H_2O_2 and by ozone, O_3 . The relative contributions of these two reactions depend on the concentrations of the two oxidants and on cloud water pH, and to lesser extent on temperature [Schwartz, 1988]. For typical concentrations of the two oxidants (a few ppb H_2O_2 ; a few tens of ppb O_3) the H_2O_2 reaction is dominant for pH below about 5. The strong decrease of the $\text{SO}_2 - \text{O}_3$ reaction rate with decreasing pH makes this reaction self-limiting as production of sulfuric acid lowers the pH and slows down further reaction; the $\text{SO}_2 - \text{H}_2\text{O}_2$ reaction rate is independent of pH. At the cloud temperature of 10°C , the $\text{SO}_2 - \text{H}_2\text{O}_2$ reaction rate R_{aq} (M hr^{-1}), which represents a lower limit for the total sulfate production rate, is given by [Schwartz, 1988]:

$$R_{\text{aq}} = 7.26 \times 10^{-4} p_{\text{SO}_2} p_{\text{H}_2\text{O}_2} \quad (12)$$

where p_{SO_2} and $p_{\text{H}_2\text{O}_2}$ are the mixing ratios of SO_2 and H_2O_2 with the unit of ppb. Equation (12) assumes that gas- and aqueous- phase mass-transport limitations are negligible, as is the case under typical conditions [Schwartz, 1988]. The in-cloud production rate of aerosol volume can be written as:

$$R_c = V_m R_{\text{aq}} \frac{L}{\rho_w} \quad (13)$$

where V_m is the sulfate molar volume, L the cloud liquid water content, and ρ_w the water density (1000g/L). Based on the assumption that the sulfate was completely neutralized by ammonium, the molar volume of ammonium sulfate ($74.6 \text{ cm}^3/\text{mole}$) is used in the following estimations. Inserting equation (12) into (13), we have:

$$R_c = 54.14 p_{\text{SO}_2} p_{\text{H}_2\text{O}_2} L \quad (14)$$

Table 2. Parameters Used as Inputs of the One-Dimensional Model for Estimation of the Aerosol Volume Concentration Increase From 180 to 950 m Altitude on 25 May 2003

Parameters	Values Used	Notes
p_{SO_2}	1 ppb	Assumed
$p_{\text{H}_2\text{O}_2}$	1 ppb	Assumed
l_b	1000 m	Active Remote Sensing of CLOUDs product that combines the data from millimeter cloud radars, laser ceilometers, microwave radiometers, and micropulse lidars to produce a time series of vertical distributions of cloud hydrometeors over the ARM sites.
l_c	500 m	Same as l_b
K_{zz}	150 m^2/s	Calculated as $0.1w \cdot (l_b + l_c)$, w is assumed as 1 m/s
L	0.13 g/m	Calculated as the liquid water path divided by the cloud thickness (l_c). The liquid water path was derived from Microwave Radiometer measurements, and was averaged over four sites during the time of the overpasses.

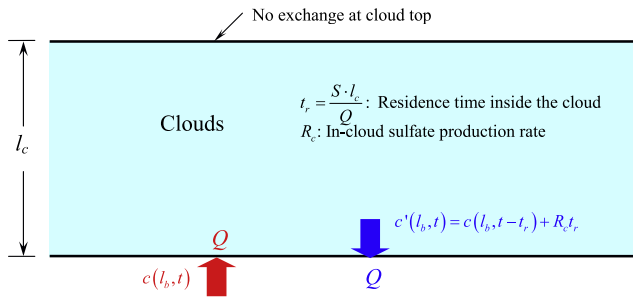


Figure 8. Derivation of aerosol volume flux as a result of in-cloud sulfate production.

where F is given in the unit of $\mu\text{m}^3 \text{cm}^{-3} \text{hr}^{-1}$, and the unit of L is g/m^3 . Combining equations (11) and (14), we can write the aerosol volume flux as:

$$F = 1.5 \times 10^4 p_{\text{SO}_2} p_{\text{H}_2\text{O}_2} L \frac{l_c l_b}{l_c + l_b} \quad (15)$$

where F has a unit of $\mu\text{m}^3 \text{cm}^{-2} \text{hr}^{-1}$, and the unit of l_b and l_c is m.

[23] Because the mixing ratio of H_2O_2 was not measured during the project, it is necessary to estimate this quantity from other measurements. Vertical profiles of H_2O_2 mixing ratio in Ohio in June 1987 were reported by *Kleinman and Daum* [1991]. The average H_2O_2 mixing ratio measured at 875 hpa (corresponding to the cloud height discussed here) was about 2 ppb. *Ray et al.* [1992] reported the H_2O_2 mixing ratio at 875 hpa averaged 1 ppb and 3 ppb at a central Arkansas site during spring and summer, respectively. Due to the lack of measurements, we took 1 ppb as an estimate of H_2O_2 mixing ratio in our calculations. We also assumed the mixing ratio of SO_2 to be 1ppb. The cloud base height and cloud thickness, which are given by Active Remote Sensing of CCloud (ARSCL) product, were 1000 and 500 m, respectively. The ARSCL product combines the data from millimeter cloud radars, laser ceilometers, microwave radiometers, and micropulse lidars to produce a time series of vertical distributions of cloud hydrometeors over the ARM site. The vertical eddy diffusivity K_{zz} was estimated using an empirical expression applicable to unstable conditions [*Lamb et al.*, 1975; *Lamb and Duran*, 1977]. The empirical expression shows that K_{zz} starts from 0 at the surface and first increases with increasing altitude. K_{zz} reaches its maximum value of $\sim 0.21w \cdot z_i$ at a height of $0.5z_i$, where w is the vertical velocity and z_i is the inversion base height. K_{zz} then gradually decreases to near zero as the altitude further increases to z_i . The average K_{zz} within the BL is estimated as:

$$K_{zz} = 0.1w z_1 = 0.1w(l_b + l_c) \quad (16)$$

Assuming the updraft velocity as 1 m/s on this cloudy day, we estimated K_{zz} using equation (16) as $150 \text{m}^2/\text{s}$. The cloud liquid water content is calculated by dividing the liquid water path by the cloud thickness (l_c). The liquid water path was determined by averaging Microwave Radiometer measurements at four locations near the SGP site during the aircraft overpasses. The calculated liquid water content

was $0.13 \text{g}/\text{m}^3$. All parameters discussed above are listed in Table 2. It is important to note that the one-dimensional model was based on 100% cloud coverage. Cloud coverage strongly influences the total liquid water volume that is available for aqueous sulfate production. As the liquid water content was derived from Microwave Radiometer measurements averaged over the period of overpasses at different sites, the cloud coverage is implicitly accounted for by the average or “effective” liquid water content, which was used to derive the aqueous phase oxidation rate.

[24] Using parameters listed in Table 2, we estimated the particle volume flux using equation (15) as:

$$F = 6.5 \times 10^5 \frac{\mu\text{m}^3}{\text{m}^2\text{sec}} \quad (17)$$

The difference in aerosol volume concentration between 950 m (z_2) observed at 15:07 (t_2) and 180 m (z_1) observed at 15:32 (t_1) can be estimated by combining equations (4) and (17):

$$\Delta c = -\frac{F}{l_b}(t_1 - t_2) + \frac{F}{2K_{zz}l_b} \cdot (z_2^2 - z_1^2) = 0.91 \mu\text{m}^3/\text{cm}^3 \quad (18)$$

[25] The magnitude of the estimated difference in volume concentration, $0.91 \mu\text{m}^3/\text{cm}^3$, is in qualitative agreement with the observation of $1.3 \mu\text{m}^3/\text{cm}^3$. Although this agreement suggests that in-cloud sulfate production may have been sufficiently fast to account for the observed vertical gradient of aerosol volume concentration, it must be stressed that there are many assumptions associated both with the one-dimensional model and some parameters used in our calculation were assumed or estimated due to lack of measurements. The one-dimensional model is expected to have limitations since it treats many processes through greatly simplified approaches. Furthermore, additional in-cloud reactions such as oxidation of SO_2 by O_3 , neglected in the present estimate, might increase the sulfate production rate. Consequently the agreement between the present estimate and the measured increase of aerosol volume concentration should be viewed as a consistency check, rather than a claim that the model accurately predicts the rate of in- cloud sulfate production.

3.5. Implication of In-Cloud Sulfate Production on the Inference of Aerosol Properties at Higher Altitudes From Ground-Measured Measurements

[26] The observed vertical gradients in dry aerosol particle size and volume concentration suggest that surface aerosol measurements may not be representative of aerosols at higher altitudes even within a well-mixed BL when cloud coverage is substantial. The vertical profiles measured at SGP show the difference in aerosol volume concentration between the cloud base and the surface was about $1 \mu\text{m}^3/\text{cm}^3$, and larger difference can be expected elsewhere. SO_2 concentrations at SGP are likely to be much lower than those in the Northeast and upper Midwest of the United States, where higher SO_2 concentrations could lead to larger vertical gradient of aerosol volume concentration. *Kleinman and Daum* [1991] reported SO_2 concentration in the vicinity of Columbus, Ohio reached as high as 21 ppb, and the average SO_2 concentration was over 5 ppb near the surface.

Assuming the concentrations of SO₂ and H₂O₂ as 5 ppb and 1 ppb, respectively, and using the same meteorological data listed in Table 2, we estimate the difference in aerosol volume concentration at the two altitudes (180 and 950 m) as:

$$\Delta c = \frac{F}{2K_{zz}l_b} (z_2^2 - z_1^2) = 9 \mu\text{m}^3/\text{cm}^3. \quad (19)$$

4. Conclusions

[27] Vertical profiles of aerosol properties were measured on board the Twin Otter aircraft during the ARM Aerosol Intensive Operation Period at SGP site in northern central Oklahoma in May 2003. Out of a total of 16 flights, well-mixed BLs were found on four cloudy days. The well-mixed BLs were identified by constant potential temperature, water vapor mixing ratio, and total particle concentration within the BL. In three of the four cases, aerosol size distributions measured from a SMPS and/or a PCASP show that dry particle size increased with increasing altitude, leading to increases in both dry aerosol volume concentration and light scattering coefficient. Given the presence of cloud coverage during the measurements, the observed vertical gradient in dry aerosol volume concentration is likely due to in-cloud sulfate production. Using a one-dimensional model, we simulated the aerosol volume concentration increase within the BL under the observed meteorological conditions. While there are many assumptions associated with the one-dimensional model and some required parameters are estimated or assumed, the simulation suggests that the increase in dry aerosol volume concentration with increasing altitude is consistent with in-cloud sulfate production through aqueous oxidation of SO₂. The explanation is also consistent with the constant aerosol absorption coefficients measured by a PSAP, since sulfate is not absorptive. In addition, two cases on cloud-free days identified from the Cessna flights (IAP program) showed that in contrast to the 3 cases on cloudy days, the dry aerosol scattering coefficient was uniform throughout the well-mixed BLs. The absence of a vertical gradient in aerosol volume concentration on clear days lends strong support to our explanation that the observed increases in particle size and aerosol volume concentration were due to in-cloud sulfate production. It is important to point out that the lack of trace gas measurements prevents more accurate predictions of the sulfate production rate and more rigorous checks on our interpretation. The conclusion drawn here should be verified in future field studies when simultaneous aerosol and trace gas measurements are available.

[28] The observed increase of dry aerosol volume concentration due to cloud processing is $\sim 1 \mu\text{m}^3/\text{cm}^3$ from the surface to the cloud base. In a location with higher SO₂ concentration, such as the Northeast and upper Midwest United States, the vertical gradient of dry aerosol volume concentration could be much larger. Using the one-dimensional model, we show that at an SO₂ concentration of 5 ppb, the difference in aerosol volume concentration between cloud base and surface could reach as high as $9 \mu\text{m}^3/\text{cm}^3$. This suggests that even within a well-mixed BL, surface measurements of aerosol properties, such as size

distribution and scattering coefficients, may not be representative of aerosols at higher altitudes, especially when the cloud coverage is substantial.

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- D. Covert and R. Elleman, Department of Atmospheric Sciences, University of Washington, Seattle, WA 98195, USA.
- P. H. Daum, L. I. Kleinman, Y.-N. Lee, S. E. Schwartz, S. R. Springston, and J. Wang, Atmospheric Science Division, Brookhaven National Laboratory, Upton, NY 11973, USA. (jian@bnl.gov)
- H. Jonsson, Naval Postgraduate School, Monterey, CA 93933, USA.