**Atmospheric
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Discussions**

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Interactive comment on "Observations of sulfur dioxide uptake and new particle formation in a midlatitude cumulus cloud" *by* J. R. Peter et al.

J. R. Peter et al.

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We would like to thank the referee for their constructive comments. Below are our responses.

2. Comments

2.1

We are aware of the work of Husain and the Se tracer technique. We made a reference to the technique (pg. 7478, line 23), although a later reference (Husain, 2004) than the reference suggested by the referee (Husain, 1991).

2.2

The reviewer commented that we should mention ternary nucleation. We have added

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the following sentence (pg. 7474, line 12),

Observations have, however, found that observed H_2SO_4 concentrations are insufficient for binary nucleation to occur, but are sufficient for ternary nucleation of the H_2SO_4—NH_3—H_2O system (e.g. Weber, 1999).

2.3

We have replaced "size" (page 7475, lines 8,9) with "radius" to make it explicit that radius is the determinant of particle size rather than diameter. The remainder of the paper, including all size distributions, utilises radius as the measure of particle size.

2.4

There are three points to be addressed in reply:

(1) The data acquisition rate: the 64 Hz data acquisition rate referred to is the aircraft data acquisition rate. Analog output, from the ASASP and 43S, was obtained at 64 Hz and then averaged to 1 Hz. We have made this explicit in the text.

(2) The temporal resolution required to obtain the measured values: the SO2 concentrations measured were obtained in three regions; sub-cloud, entrained and detrained air. The sub-cloud and entrained region concentration measurements are an average of 1 minute of data and the detrained concentration was found from an average of 3 minutes and 50 seconds of measurements. These measurements are thus within the required temporal resolution (1 minute) suggested by the referee. (3) How do we ensure that we are looking at detrained air from the cumulus?

The flight path of the aircraft was specifically designed to examine air detrained from the cumulus. This involved flying parallel to the wind such that the aircraft was flown both upwind and downwind of the cloud band. This flight pattern increases the likelihood of sampling detrained air from a dynamical perspective.

The mixing diagram (Fig. 1) is proof that we are sampling detrained air. The thermo-

dynamic measurements of clear air sampled downwind of the cumulus band lie to the right of the sounding and fall on a straight line between the cloud base sample and the PSEA. This provides us with two pieces of information: (1) air sampled downwind of the cumulus band is warmer and has a greater water vapour content than air at the corresponding altitude in clear air upwind of the cumulus band; (2) since we are plotting conserved thermodynamic properties that mix linearly, or nearly linearly in the case of θ_q, and the resulting samples fall on a straight line, the mixing is a result of combination of air from two distinct air regions; cloud base and the PSEA.

The only manner in which air in the cloud layer can acquire a component of subcloud air is if it has been transported vertically through the cloud. The thermodynamics of the parcels thus indicate that we are sampling cloud detrained air.

Although not shown (and it is not a necessity for the paper) clear air sampled on the upwind side of the cloud band was also examined to ascertain its source. The samples in this case lay almost exactly on the sounding line indicating that air was being detrained from the cloud mainly on the downwind side of the cloud band. This is dynamically consistent with observations indicating that updrafts occur most frequently on the upwind side of the cloud band, while downdrafts and detrainment occurs mainly on the downwind edge of cloud (Kitchen, 1981; Perry and Hobbs, 1994; Kollias, 2000; Lu, 2003).

To address the above three points we have added the following text pg. 7478.

The subcloud and entrained region concentrations are averages of 1 minute of data, and the detrained concentration was evaluated from 3 minutes and 50 seconds of data. These acquisition times are within the response time (1 minute) of the TSI-43S SO_2 analyzer. The time taken for data acquisition in the detrainment region may seem to indicate a rather poor spatial resolution for examining detrainment from the cloud; the airspeed of the King Air was about 100 m s^{-1} resulting in a horizontal distance of around 23 km being covered during sampling of the detrained air. However, satellite

images revealed that the cloud band had held a consistent structure for at least two hours before the observational period (Peter, 2006). The wind speed at the flight level was about $10 \, \mathrm{m \, s^{-1}}$ suggesting that air detrained from the cumulus may have travelled several hundred kilometres downwind of the cumulus. Quantitative verification that detrained air was sampled downwind of the cumulus is given in Fig. 1; the clear air samples downwind of the cumulus contain a fraction of subcloud air, which can only be achieved by the process of convection. The SO_2 concentration in the detrained region cited in Table 1 corresponds exactly to the same times of the clear air samples in Fig. 1.

2.5

This point addresses one of the major issues raised by the reviewer, namely the error associated with the F-value. There are two issues which we would like to address.

(1) The reviewer suggests estimating the uncertainty in the regression mixing line and the departures of the in-cloud points from the mixing line. We have calculated the R-square value for the line of best fit which passes through the cloud base sample, the clear-air samples and the primary source of entrained air, and obtained 0.98. This is an exceedingly high value and indicates that there are no substantial departures of the clear-air samples from the line of best fit. This is evident visually, however, quoting the R-square value quantifies this departure. We conclude that the error obtained in estimating the PSEA from the line of best fit which passes through the cloud base sample and the clear-air samples is minimal.

(2) The second source of error in obtaining the F-value may come from measurement errors. For the purposes of the mixing diagram, the sources of error will be in measurements of temperature, water vapour mixing ratio and pressure. We address these now in succession.

- Temperature. The error in temperature for the Rosemount sensor, which was

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used to measure the temperature of the clear-air samples is $\pm 0.3\ ^\circ$C.

- The wet-bulb temperature, from which the dew point temperature is calculated also has an error of $\pm 0.3\ ^\circ$C., in clear air.

- Pressure. pressure was measured with a Rosemount pressure transducer. The error associate with this sensor is ± 0.35 hPa.

- In total these errors represent a maximum error in θ_q of 0.8°C and in Q_{tot} of 0.2 g kg^{-1}.

The maximum error that could be induced by these changes, corresponds (at most) to modifying θ_q of the PSEA to be in the range 305–309 K. This range corresponds to the extremities of the horizontal section of the sounding where the PSEA is marked.

The propagation of the above errors results in the F-fraction now being in the range 0.61–0.67. For the analysis, we used a value F=0.64, but we now quote F=0.64 $\pm$0.03.

It should be noted that the errors contributing to the F-fraction are minor when compared with concentration measurements of SO$_2$ or aerosol particles, which have been accounted for. When these errors are propagated through to the ratio calculation (Figure 4), the error bars are of the same order of size as the thickness of the lines.

Additionally, when the error in the F-fraction is propagated in evaluating the predicted concentration of SO$_2$, the error (± 0.21) remains unchanged (to two significant figures).

We conclude that the error incorporated by measurement errors and evaluation of the F-fraction is insignificant. As such, our conclusion that particle nucleation may be occurring within cloud is valid.

2.6

The error quoted in the text (± 0.17) was a typographical error. The correct error (as pointed out by the referee) is ± 0.21, which is the error which was quoted in Table 1.

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2.7

The sentence regarding provision of rate constants (page 7479, line 5) has been re-moved.

The rate constants for aqueous-phase oxidation of SO_2, by either O_3 H_2O_2, are well quantified (Seinfeld and Pandis, 1998). By deducing the amount of SO_2 scavenged within cloud (as we have done) coupled with knowledge of the concentrations of ox-idising species O_3 and H_2O_2 (which we don't have) would enable the time a parcel has spent in cloud to be evaluated. We agree that the there are many errors that may be propagated when attempting such a calculation, however, there are currently no methods to determine in-cloud parcel residence time. An attempt at such a calculation is needed to constrain cloud microphysical models. The feasibility of making such a calculation is high. We have demonstrated the feasibility of determining the amount of SO_2 scavenged within cloud and, if we had measurements of O_3 and H_2O_2, would have conducted this calculation ourselves.

2.8

Entrainment is probably the most likely explanation for the decrease in cloud droplet number concentration (CDNC), and this is in fact evident from the mixing line of the in-cloud points on the conserved variable diagram (Figure 1.). The in-cloud points correspond to 1 Hz samples of cloudy air at the same altitude at which the clear air samples were obtained. It can be seen that there are only a few cloudy samples that come close to being adiabatic (i.e. having the same total water content as the cloud base sample). This is very typical of observations within cumulus clouds as has been shown in several studies (Warner, 1967; Jensen, 1985). To examine how the aerosol size distribution may have been modified by production of sulfate in cloud droplets, we distributed the mass of SO_2 lost among the cloud droplets. We came to the conclusion that distributing the available mass among the cloud droplet size distribution resulted in a larger fractional increase in radius of the smallest aerosol. This conclusion will not

change, if we consider the mass to be partitioned among the number of cloud droplets measured at cloud base or at the flight level (800 m above cloud base). We settled on a value that was the average of these values as sulfate production will occur continuously throughout the extent of the cloud until in-cloud parcels have detrained.

We have used the assumption that the clear air samples measured on the downwind side of the cumulus band were detrained at their level of neutral buoyancy, which observations suggest is the most realistic description of the process of detrainment in cumulus clouds. This means that the parcels that started their vertical ascent at cloud base and were then detrained at the flight level at which we measured the clear air samples. These parcels presumably encountered cloudy air that had CDNC concentrations ranging from the cloud base number to the number measured at the flight level.

The second point mentioned here is that the conserved variable analysis will be invalid if there is appreciable precipitation, which we do acknowledge. However, there was no precipitation that reached the surface. The precipitation was measured as appreciable concentrations of 2D-C sized droplets in the cloud anvil. It was shown in a previous paper (Peter, 2006) that there were appreciable downdrafts on the downwind side of the cumulus band which helped sustain the general circulation occurring within the cumulus cloud. The mixing diagram includes the liquid water content measured by the 2D-C, however, the position of the in-cloud samples was unaffected if the 2D-C measured LWC content was ignored. We are therefore confident that, although precipitation-sized droplets may have contributed to downdrafts and the maintenance of the convection, precipitation did not effect the conserved variable analysis.

Figure 2 has been modified to show that there were downdrafts on the downwind side of the cumulus band, not precipitation as was indicated previously, to avoid confusion. **2.9**

The monotonic increase in the ratio of the predicted to observed concentration is due to the highly size-dependent nature of the Brownian diffusional scavenging of the CN

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by cloud droplets. This fact is encompassed in Eq. (9) and Eq. (10).

The other issue raised was how the series of curves in Fig. 4 was arrived at without size distribution information for the CN number size distribution. This issue was also raised by Anonymous Referee #1 in their point 1, so we regard it crucial to explain this in more detail.

In Section 3.3 (page 7481 lines 25–28) we state that the ratio of the predicted-to-observed concentration was calculated from the differential concentration. Thus, the differential concentration calculated for the TSI-3010, which counts particle with a radius greater than 6 nm, was calculated as the measured concentration minus the ASASP concentration. The ASASP measures particles with a radius larger than 66 nm. The concentration difference between the two instruments is therefore the differential concentration dN in the size range 6–66 nm. Similarly, the difference between the TSI-3025 and TSI-3010 gives the differential concentration of particles in the size range 1.25–6 nm. Derived concentrations, such as dN/dr and dN/dlogr can then also be calculated. We thus have two size bins, in the size range 1.25–66 nm, which are represented by the horizontal lines (which look like error bars) in Fig. 4. The nominal radius in each size bin is also represented, by a circle for the TSI-3025 and a square for the TSI-3010.

To arrive at the series of dashed curves, which incorporate the loss expected due to Brownian diffusion of the CN to cloud droplets we assumed that the initial aerosol size distribution, $n_a(r_a, 0)$ in Eq. (9) was monomodal and the number concentration was specified as the differential concentration of the particular differential size bin it resided. Thus is r_a was in the radius range $1.25 \leq r_p \leq 6.0$ nm, then the concentration $n_a(r_a, 0)$ was set at the differential concentration predicted by mixing (Eq. (3)) for the TSI-3025 probe. Similarly, if r_a was in the radius range 6 nm$\leq r_p \leq$66 nm, then $n_a(r_a, 0)$ was set to the value of the differential concentration predicted by mixing for the TSI-3010 probe.

In effect, this provides the maximum and minimum number of CN that could be scav-

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enged by Brownian diffusion to cloud droplets. If all particles are of the size specified by the minimum radius of the size bin (1.25 and 6 nm for the TSI-3025 and TSI-3010 respectively) then solution of Eqs. (9) and (10) yields the maximum number of particles that could be scavenged because they have the largest diffusion coefficient. Conversely, if all the particles are considered to be of the size specified by the maximum radius of the size bin it is in (6 nm and 66 nm for the TSI-3025 and TSI-3010 respectively) then the solution is the minimum number of particles that could be scavenged by Brownian diffusion. Therefore, the ratio $CN_{predicted}/CN_{observed}$ will be a minimum at the minimum radius of the size bin, and a maximum at the maximum radius of the size bin. Because the diffusion coefficient is a strong function of particle size, we see that $CN_{predicted}$ is nearly zero at small radii and asymptotes the value predicted by mixing as the particle radius increases. However, over the entire size range of the first size bin (TSI-3025), the predicted to observed ratio is consistently below unity, implying that there must be a source of new particles to balance their diffusive loss to cloud droplets.

The following text has been added,

"Since we only have two size "bins," provided by the differential concentration of the CN counters and the ASASP, it was necessary to make assumptions about the form of the aerosol number size distribution $n_a(r_a, t)$. We assumed the aerosol size distribution to be monomodal and its number concentration specified by whether it was in the size range of the first size bin (determined by the differential concentration of the TSI-3025 and TSI-3010) or the second size bin (determined by the differential concentration of the TSI-3010 and the ASASP). In effect, this specifies the maximum and minimum particles that could be left in a size bin. If all particles are considered to be of the size specified by the left hand size of the size bin (i.e. their smallest possible value) then they will have the largest possible diffusion coefficient and be subject to the largest possible scavenging. Conversely, if all particles are the size specified by the upper limit of the size bin, then the particles will have the smallest diffusion coefficient and be subject to the least amount of scavenging."

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The need for vertical error bars has been discussed in the points above.

2.10

We have modified the figure to utilise the full page width, rather than a column width. An appropriate size will need to be chosen for the figure during production, and we will consult with copy editors to address this.

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