The effect of sulphur chemistry on the scattering properties of particles

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SUMMARY

A model has been developed of the interaction of aerosol with shallow layer clouds. The model includes aqueous phase chemical processes, particularly the oxidation of sulphur dioxide (SO_2) occurring in droplets formed on cloud condensation nuclei of known chemical composition. The cloud microphysics as a function of height above cloud base is predicted along with changes to the aerosol size distribution and hygroscopic properties after a single cycle through cloud. The predictions of the model are compared with observations from a hill cap cloud experiment for a single cloud pass. A stratocumulus cloud model then examines the changes in the cloud microphysics and aerosol population arising as the modified aerosol distribution undergoes a further passage through cloud. The cycling process is repeated a further nine times. We show that the cloud processing of the aerosol results in a strongly bimodal aerosol size distribution which can significantly affect the direct radiative forcing of the aerosol. The processing is strongly oxidant-limited and forced entrainment of hydrogen peroxide through cloud top is required to maintain the oxidation. A strong surface source of ammonia substantially enhances the observed modification. The processing affects the cloud condensation nucleus spectrum, initially increasing the number of droplets activated. However, multiple cycling has a tendency to stabilize the droplet number.

1. INTRODUCTION

Atmospheric aerosols affect the Earth's climate through their influence on the radiation budget. This occurs both directly through the scatter of solar radiation by aerosol particles and indirectly as they act as cloud condensation nuclei (CCN). In this latter role, the aerosols control the number of droplets which activate within a given cloud system. This in turn controls the droplet effective radius for a given liquid water content, and hence the albedo of the cloud (see Slingo (1990) for a discussion of the sensitivity of the radiative properties of cloud to changes in the droplet effective radius). The natural aerosol has been substantially perturbed by anthropogenic aerosols, for example by sulphates formed from SO_2 emissions. The global mean radiative forcing due to the direct effect of anthropogenic aerosols has been calculated to be of a similar magnitude, but opposite sign, to the radiative forcing of anthropogenic greenhouse gases (Charlson *et al.* 1991). The indirect effect is poorly quantified but may be at least as important as the direct effect. Despite their importance, the effects of aerosol particles are still very poorly represented in global climate models. This is partly a result of a lack of globally distributed data, but it is also due to the absence of a clear understanding of the chemical processes linking gaseous precursor emissions (e.g. sulphur compounds and oxides of nitrogen) to the size distribution and hygroscopic properties of aerosols, aerosol optical depth and cloud reflectivity.

It has been demonstrated by observational studies of the processing of aerosols by cloud, for example Hoppel

et al. (1994), that a bimodal aerosol size distribution is produced. Modelling studies (e.g. Bower & Choularton 1993) have also shown that cloud processing can produce a bimodal aerosol size distribution and can enhance the number of CCN available for subsequent cloud formation.

In this paper, a model of the stratocumulus cloud-capped boundary layer is presented, and the evolution of the atmospheric aerosol as it undergoes multiple cycles of mixing through the depth of the boundary layer is described. This process affects both the direct and indirect radiative forcing of the aerosol particles. First, the chemical processing of those particles which are nucleation-scavenged by the cloud droplets in the stratocumulus causes the accumulation mode particles to grow larger. Secondly, the increased soluble mass of sulphate aerosol may increase the number of CCN, and hence modify the cloud microphysics.

2. THE MODEL

This is based on the cloud chemistry model of Bower & Choularton (1993). In outline, the initial aerosol spectrum is based on an external mixture of ammonium sulphate aerosol (small sizes) and sea salt (large sizes). When these data are applied to specific case studies, the size partitioning of aerosol chemistry is obtained from cascade impactor measurements. Each aerosol category is also given an insoluble fraction, determined from the measurements made by a hygroscopic Tandem Differential Mobility Analyzer (TDMA) instrument when running in experimental studies.

(a) Dynamics

A dynamic framework for the stratocumulus cloud is prescribed which consists of eddies with a cycle equal to the boundary layer depth. The vertical velocity at cloud base is prescribed and set to 0.2 m s^{-1} for the results presented below. Entrainment was sometimes allowed to occur at cloud top, at a prescribed rate, introducing heat, moisture, aerosol and trace gases (particularly hydrogen peroxide). The latter was considered to be particularly important, since the S(IV) oxidation was often strongly oxidant-limited at the average droplet acidities calculated.

(b) Microphysics and chemistry

As an air parcel ascends towards cloud base, the growth of each individual class of aerosol particles is calculated. As the supersaturation rises and activation takes place, the take-up of trace gases by each individual category is explicitly calculated. Out-gassing of ammonia (NH_3) (and other species) is allowed to occur. The assumption of an equilibrium between the gas and liquid phases is not made. The out-gassing of the trace gases NH_3 and HNO_3 may be important for the growth of Aitken mode interstitial aerosol particles. Within the droplets, the oxidation of S(IV) to sulphate is computed, hydrogen peroxide (H_2O_2) and ozone (O_3) being the dominant oxidants. The parcel of air and droplets proceed upwards to the cloud top where entrainment of air from the free troposphere above the capping inversion can occur. This includes a flux of heat, moisture, aerosol particles of specified number, size distribution and chemical composition and trace gases (especially the oxidant, H_2O_2) and the detrainment of drops. The parcel then descends through the cloud base. As it does so, the droplets evaporate and modified aerosol particles are generated. Out-gassing also occurs. This parcel then recycles and undergoes another similar cloud pass. This process is repeated up to nine times.

3. RESULTS OF MODELLING STUDIES

Table 1 summarizes the initial physical conditions put into the model for each of the model runs presented. Variations in the chemical conditions for the three cases discussed are summarized in table 2. Typical results from case 1 are shown in figures 1 and 2. Figure 1

Table 1. *The initial physical conditions input to the model for case studies 1–3*

physical parameter	input value
boundary layer depth	600 m
cloud base altitude	300 m
relative humidity at cloud base	99 %
cloud base temperature	12.5 °C
cloud base pressure	900 mbar
tropospheric air; temperature	cloud top T + 2.5 °C
tropospheric air; relative humidity	85 %
vertical wind velocity	0.2 m s^{-1}

shows the changes in the aerosol size distribution after successive passages through cloud. This case uses the initial chemical conditions that are regarded as the standard input for this study. The characteristic bimodal size distribution occurs as particles which are just large enough to activate become larger due to the addition of sulphate mass. However, the oxidation process is very strongly oxidant-limited. With these input conditions, oxidation by ozone was very inefficient. In order to maintain the modification in successive cycles, entrainment of H_2O_2 into the cloud had to be simulated. This was achieved by restoring the concentration of H_2O_2 in the region close to cloud top to 0.3 ppbv ($\frac{1}{3}$ the input concentration at cloud base). This essentially regulated the S(IV) conversion rate. Aircraft data (Gallagher *et al.* 1991) all support the suggestion that the free troposphere is a major source of H_2O_2 .

Ozone is present in abundance, but for this to be an effective oxidant, a surface source of ammonia is required to raise the cloud water pH (which in these examples was less than 3.5). Such a source will be found over many continental and some oceanic regions. In addition, there is now evidence (Wells *et al.* 1996)

Table 2. *The input gas phase concentrations for model case studies 1–3*

species concentration (ppbv)	case 1	case 2	case 3
SO_2	10	10	10
NH_3	0.1	0.1†	2.0†
H_2O_2	1.0	1.0	1.0
H_2O_2 troposphere	0.0*	2.0	2.0
O_3	30.0	30.0	30.0
O_3 troposphere	30.0	30.0	30.0

* Simulated entrainment only (i.e. reset to 0.3 ppbv if below this concentration at cloud top).

† Reset to initial cloud base concentration at cloud base for each successive cycle.

Figure 1. The aerosol size distribution predicted by the model after one, five and ten cycles through the stratocumulus cloud in case study 1, compared to the unprocessed (input) aerosol distribution. Entrainment of hydrogen peroxide at cloud top was simulated by resetting the gas phase concentration to 0.3 ppbv (if below this value).

Figure 2. The CCN activity spectrum after one, five and ten passes through stratocumulus cloud in case study 1.

Figure 3. The peak cloud supersaturation attained in successive cycles through the processing stratocumulus cloud in case study 1.

that ammonia can be transported long distances in dry aerosol as ammonium. When cloud forms, this can then be repartitioned between cloud droplets.

Figure 2 shows the effect of the cloud processing on the CCN activity spectrum. It can be seen that a significant change occurs in the number of CCN available for subsequent activation following each cycle. However, as indicated in figure 3, the peak supersaturation attained within successive cloud passes (with the same vertical velocity) declines. This occurs because those droplets which activate in the first cloud pass produce on evaporation larger CCN with a reduced critical supersaturation. These particles then gather water at an increased rate in subsequent activation cycles. This process tends to stabilize the droplet number concentration (at least when the updraught is fixed), and it becomes very insensitive to the gas phase concentration of primary pollutants and the subsequent injection of secondary aerosol. Only a substantial injection of primary aerosol which is very efficient as CCN can alter the cloud droplet number concentration.

In the absence of entrainment the effect of cloud processing is limited to the first cycle through cloud.

Thereafter, little modification is observed. However, a similar result is obtained even when entrainment is more realistically simulated, i.e. with 2 ppbv of H_2O_2 in the free troposphere and using a prescribed entrainment rate capable of reducing the cloud top liquid water content by 10%, as in case 2 (illustrated in figure 4a) discussed below.

It is interesting to note that with the fitting of flue gas desulphurization units to some power stations in the UK, the SO_2 concentrations have dropped substantially, by much more than the emissions, but the sulphate aerosol loadings have remained almost unchanged (Review Group on Acid Rain 1996, to be published). This is exactly what this model predicts; the sulphate production is controlled by oxidant availability. It is strongly suggested that reducing SO_2 emissions has not reduced indirect or direct radiative forcing of aerosol in polluted regions near to sources of SO_2 .

(a) The effect of ammonia on the aerosol size distribution

Over the UK and Europe, the airstream being advected from a source of SO_2 will pass over agricultural land, which can be a strong source of ammonia. In these regions gas phase ammonia concentrations are maintained at levels of 1 or 2 ppbv, typically during the warmer months of the year (Bower *et al.* 1995). Model runs were carried out to investigate the effects of varying the availability of ammonia on the cloud processing. In these runs full entrainment of O_3 and H_2O_2 (at 30 ppbv and 2 ppbv in free troposphere), heat and moisture, and detrainment of cloud drops (but no entrainment of fresh aerosol) was enabled, and the concentration of ammonia at cloud base in successive cycles was reset to the initial input value of 0.1, 1 or 2 ppbv. Other trace gas inputs were as in case 1.

Figures 4a and 4b show the resulting aerosol size modification in successive passes for case studies 2 and 3 with inputs of 0.1 and 2.0 ppbv of NH_3 respectively. Figures 5a and 5b show the corresponding bulk sulphate and ammonium loadings following processing for the same cases. It can be seen that in the high ammonia case (case 3), the amount of sulphate produced increases in successive cycles, and that the size modification generated is similar to that of case 1 (where a forced entrainment of H_2O_2 was simulated). This is because the ammonia maintains a high cloud water pH and so enables ozone to be an effective oxidant. As in case 1, the bimodality of the aerosol size distribution continues to increase markedly as extra sulphate mass is added in successive cloud passes, increasing the direct radiative effect of the aerosol. However, in the absence of significant NH_3 levels (case 2), the extra oxidant introduced (by the entrainment of H_2O_2 through cloud top) is not able to maintain a significant additional modification after the first cycle (in contrast to case 1), even though the entrainment rate is sufficient to reduce cloud top liquid water content by up to 10%. Although drops are also lost through detrainment, the overall effect of processing

Figure 4. (a) The aerosol size distribution predicted by the model after one, five and ten cycles through the stratocumulus cloud in case study 2, compared to the unprocessed (input) aerosol distribution. Gas phase inputs are as in case 1 (including 0.1 ppbv NH_3) but with full entrainment of heat, moisture and trace gases (2 ppbv H_2O_2 , 30 ppbv O_3 present in tropospheric air). (b) As figure 4a but for case 3, with NH_3 set to 2 ppbv initially and at cloud base in successive cycles.

Figure 5. (a) Bulk aerosol loadings of sulphate and ammonium after successive cycles through the stratocumulus cloud of case 2 (corresponding to figure 4a). (b) As figure 5a except for case 3 (corresponding to figure 4b).

on the cloud microphysics is negligible, for the reasons explained above (in case 1).

4. COMPARISON OF MODEL PREDICTIONS WITH FIELD STUDIES

(a) Field experiments at Great Dun Fell

Two major field projects have been performed, one in the autumn of 1991 and the second in the spring of 1993. Both used the Great Dun Fell cap cloud as a natural flow-through reactor to study the physical and chemical processes relevant to a number of scientific objectives. Those relevant to this work include investigations into the sulphur and nitrogen chemistry of the system and the evolution of the aerosol size distribution as air passed through the cloud covering the hill. In the second of these experiments the suite of measurements was supplemented by contributions made by several other groups who joined the experiment forming the third EUROTRAC GCE field campaign (Choularton *et al.* 1997).

Great Dun Fell forms part of a long ridge running

north-west to south-east. The prevailing winds in the area are from either the south-west or the north-east, hence the site is ideal for a flow-through reactor study as air is forced to rise over the ridge. The ridge is frequently enveloped by a capping cloud. The 1993 experiment was performed at five major sites. These were as follows:

- Site 1: Wharley Croft (elevation 206 m)
- Site 2: Fell Gate (elevation 430 m)
- Site 3: Mine Road (elevation 670 m)
- Site 4: Great Dun Fell summit (elevation 847 m)
- Site 5: Moor House (elevation 550 m).

Sites 1 and 2 are generally below cloud base on the south-western side of the ridge. Site 3 is generally close to or above cloud base, site 4 is at the hill summit and site 5 is close to cloud base on the north-eastern side of the ridge.

The main measurements made at each of the sites are summarized in table 3. Contributions to these measurements were made by all GCE member groups and other British groups (Choularton *et al.* 1996).

Table 3. *The measurements made at Great Dun Fell during the GCE field campaign in 1993*

gas phase species	site	aerosol measurements	site	cloud measurements	site
NO_x NO_y	1 3 4 5	aerosol physics: size distribution DMPS	2 4(i/s) 5	cloud microphysics: drop size distribution FSSP/PVM	3 4
SO_2	1 3 4 5	size distribution ASAPX	1 5	liquid water content PVM	3 4
O_3	1 3 4 5	CN ultrafine 3025	1 5	counterflow virtual impactor CVI	4
NH_3	1 3 4 5	hygroscopic growth TDMA RHP	2	droplet-aerosol analyser DAA	
HNO_3	1 2 3 4 5	LAMMA	2 4 5	cloud chemistry: cloud collector (active)	3 4
NMHC	1			cloud collector (passive)	4
PAN	5	4 stage impactors ($\times 2$)	1 5	2-stage collector (active)	4
HNO_2	2	8 stage impactors ($\times 2$)	4	continuous chemistry: pH, NO_2^- , S(IV), H_2O_2	3 4
H_2O_2 (g)	2 3 4	bulk aerosol (R & M)		bulk chemistry:	
Organic acids (Formic/Acetic+)	2 4	bulk interstitial aerosol	3	cloud droplet size chemistry	4
HCHO	4	aethelometer	1 4	major ions	3 4
Other:		hi-vol impactor organics	2 4	transition metals	4
Automatic Weather Stations	all sites	other: UVB/radiation	2 4		
SODAR	1	JNO ₂ /radiation	2 5		

Figure 6. Aerosol size distribution measured upstream and downstream of Great Dun Fell by an optical particle counting probe (hour averaged spectrum 18.00–19.00, 22 April 1993). The wind was blowing from the south-west, i.e. from Wharley Croft to Moor House. The figure shows the growth in mid-sized aerosol due to sulphate production in the cloud.

(b) Results from the Great Dun Fell measurements

(i) Aerosol processing by cloud

A sample of the results showing some aerosol data, measured by UMIST, illustrates some of the processes operating during both field experiments. Figure 6 shows the aerosol size distribution measured at sites 1 and 5 by PMS (Particle Measuring Systems) ASAPX optical scattering probes. The data are from the first case study on the 22 April 1993. On this occasion the wind was from the south-west and site 1 (Wharley Croft) is upwind and site 5 (Moor House) downwind. It is evident that a loss of particles from the smallest size ranges has occurred. In the middle size ranges a growth of particles has occurred. These particles in the middle size ranges are mostly incorporated into the cloud droplets by nucleation-scavenging. The growth was due to the addition of sulphate by aqueous phase cloud chemistry, measured by impactors at sites 1 and

Figure 7. Aerosol size distributions measured by the DMPS system upwind, within the hill cap cloud, and downwind of Great Dun Fell (07.00–08.00, 10 May 1993).

5. The changes observed during both experiments were consistent with the results of our model described above.

Figure 7 shows data obtained from the three DMPS instruments operated at sites upwind of the cap cloud, within the cap cloud and downwind of the hill cap cloud. These data were gathered by Dr Wiedensohler of Lund University (now at IFT Leipzig). This diagram illustrates features which are found in much of the data set, when the airflow between the three sites is connected and aqueous phase oxidation of S(IV) to sulphate is occurring in the cloud water (Bower *et al.* 1997). At the largest sizes the aerosol particles are activated to form cloud droplets. The DMPS at the summit is only measuring interstitial particles. At the downwind site, the cloud droplets evaporate, returning the CCN to the aerosol phase, with some growth due to sulphate production. Careful analysis of the full data set suggests that in conditions where sulphate production by aqueous phase oxidation is occurring, some growth of the interstitial aerosol also occurs, although

this is much less than for the particles which are nucleation-scavenged. At the smallest sizes in the 'new nucleation mode' it is sometimes observed that new particles are produced on leaving the cloud (see figure 7). The number of particles produced is very variable, and is sometimes much larger than shown. On other occasions a loss of these small particles is observed as the air flows across the hill and through the cap cloud. This is probably due to the Brownian capture of small particles (which are in the interstitial air within the cloud) by the cloud droplets.

The mechanism of the formation of the small particles is of interest. The measurements of ammonia concentrations made by our collaborators from the Institute of Terrestrial Ecology are probably relevant. They show that ammonia does out-gas from evaporating cloud droplets as the air stream leaves the cloud. The aerosol and cloud water chemistry measurements show that some droplets form on large sea salt aerosol whilst the majority form on ammonium sulphate aerosol. Sulphate production within those droplets forming on sea salt aerosol is predicted to result in the out-gassing of HCl as these droplets evaporate. Preliminary modelling results suggest that the nucleation of ammonium chloride aerosol may explain the new particle production which is observed.

(ii) *Cloud microphysics*

In these experiments detailed measurements of the size distribution of the aerosol entering the cloud were available, along with measurements of the hygroscopic properties of the particles (from the TDMA). The predicted droplet size distributions from our model, using these data as input (along with a description of the airflow over the hill) were compared to the observed size distributions. Generally, reasonable agreement was obtained, although the model tended to overestimate the number of activated droplets observed at the hill top, typically by 20%. On a few occasions, a larger discrepancy was observed, but usually the differences were less than at Kleiner Feldberg (site of the second EUROTAC GCE field experiment in Central Germany).

The possibility exists that some component of the aerosol droplet chemistry affects the properties of the particles during the activation process, most probably the condensation coefficient and/or the droplet surface tension. The most likely candidate components for this are soluble organic compounds (Schulman *et al.* 1996).

A marked relationship, found during both Great Dun Fell experiments, exists between sulphate aerosol loading and droplet number concentration, an example of which is shown in figure 8. However, in continental air masses, even with low sulphate loadings, the droplet number seems to be independent of the sulphate loading and points lie along the plateau region of the curve. Other species, such as nitrate, then control the droplet number.

(c) *Flights with the C-130 aircraft*

During the spring of 1994, the UK Meteorological Office C-130 aircraft made a series of passes through

Figure 8. Relationship between aerosol sulphate loading upstream of Great Dun Fell and the cloud droplet number at the hill summit (hourly averages, April–May 1993).

Figure 9. Time history of droplet effective radius as the C-130 aircraft flew straight and level in cloud above Great Dun Fell across the ridge at an altitude of 1100 m above mean sea level on 30 April 1994. The vertical line marks the time where the aircraft crosses the hill summit.

the clouds at Great Dun Fell, both in stratocumulus clouds upwind and downwind of the hill and through the hill cap cloud itself. The objective of this experiment was to investigate whether the modification of the CCN activity spectrum predicted above was actually observed in the microphysics of the stratocumulus clouds upwind and downwind of Great Dun Fell. Vertical profiles through the stratocumulus were also made upwind and downwind of the hill. At the same time, measurements of the cloud microphysics and cloud chemistry were made on the ground at the fell's summit. The main features of the results from this study were: (i) aqueous phase oxidation of S(IV) to sulphate was occurring in the hill cap cloud; and (ii) the droplet effective radius, for a given liquid water content, was approximately 2 µm smaller in the stratocumulus clouds downwind of the hill than in the stratocumulus upwind of the hill. Figure 9 shows a transect across the hill through cloud showing the marked drop-in-droplet effective radius which occurs

downwind of the hill summit. This change was due to the activation of droplets in the hill cap cloud producing additional CCN, which became available for activation in the stratocumulus downwind. These observations are in broad agreement with the predictions of our cloud processing model discussed above.

5. CONCLUSIONS

1. When sulphate production occurs due to the oxidation of S(IV) by hydrogen peroxide and ozone, there is a marked evolution of the chemical properties of the aerosol particles on which the droplets form.

2. A significant modification of the aerosol size distribution and hygroscopic properties was modelled and observed.

3. The microphysics of the processing stratocumulus cloud are very insensitive to the trace gases entering the system. However, the CCN produced will be available for activation in less vigorous clouds forming elsewhere as their critical supersaturation has been considerably reduced. Generally, sulphate production reduces the critical supersaturation of processed aerosol particles. On some occasions, however, out-gassing of ammonia (with other gases) can cause an increase in the critical supersaturation of smaller CCN.

4. All these processes are strongly oxidant-limited unless a strong source of surface ammonia is present. In the absence of ammonia, entrainment of hydrogen peroxide from the free troposphere is rate-limiting for the S(IV) oxidation process. If ammonia is present in concentrations of 1 ppbv or greater, then this has a marked effect, producing a large amount of particulate ammonium sulphate and a strongly bimodal aerosol size distribution, enhancing the direct radiative effect of the aerosol. The effect on the cloud microphysics of the processing cloud is small.

5. New particle production is observed as the air stream exits from cloud. This is associated with sulphate production in the droplets and is probably due to the out-gassing of ammonia and hydrochloric acid from different droplets.

6. The growth of interstitial haze particles is also observed.

The funding for this project was provided by the UK Natural Environment Research Council, the Commission of the European Union and the Hadley Centre of the UK Meteorological Office.

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Discussion

K. P. SHINE (*Department of Meteorology, University of Reading, UK*). Professor Choularton presented evidence of the change in effective radius due to cloud processing of aerosols upwind and downwind of Great Dun Fell. Were coincident radiation measurements made from the C-130 and, if so, did they show a coincident increase in albedo consistent with the decrease in droplet-effective radius?

T. W. CHOULARTON. Measurements were made of the cloud albedo by the C-130 immediately upwind and immediately downwind of the range of hills. The droplet effective radius was smaller downwind with more activated droplets for a given liquid water content. The cloud downwind of the hills had been more strongly affected by entrainment of dry air from the free troposphere than the upwind cloud. Hence the liquid water path downwind was reduced. The effect of this was that the cloud albedo downwind of the hill was reduced despite the reduction in droplet effective radius.

K. P. SHINE. Since cloud processing of aerosols is oxidant-limited, would the processing have been different in a pre-industrial world when oxidant levels were probably lower? Specifically, how much of the sulphate aerosol burden is dependent on oxidant availability rather than on the SO₂ emissions?

T. W. CHOULARTON. The oxidant levels, particularly ozone, were lower in the pre-industrial period. The sulphur dioxide concentrations were, however, also much lower. If ozone is the dominant oxidant then the concentration of ammonia, the only basic trace gas, is important. It is not clear how much this has changed but it is probably higher now. Overall, the increase in sulphur dioxide since pre-industrial times almost certainly dominates the increase of oxidant availability so the chemical transformation of S(IV) to S(VI) has become more oxidant-limited. The effect of all this is that close to SO₂ sources the sulphate burden will be controlled by the availability of oxidant. This dominance decreases with increasing distance from the source regions. Over remote oceans, the sulphate burden will be controlled by the availability of S(IV) with hydrogen peroxide (in the aqueous phase) and OH (in the gas phase) being the dominant oxidants.

P. BORRELL (*EUROTRAC, Garmisch-Partenkirchen, Germany*). The excellent work, in a polluted atmosphere, with its remarkable agreement between experiment and modelling, was done with Professor Choulaton's own model, presumably designed for this purpose. In the previous paper (Ayers) we saw a similarly impressive agreement for remote marine atmospheres. Are there substantial differences in the models or assumptions used, or are they simply the same model applied with the same assumption to different air masses?

G. P. AYERS (*CSIRO Division of Atmospheric Research, Australia*). Although I am not familiar with the details of Professor Choulaton's work, my impression is that the cloud chemistry models used in his work and our work are of a similar type, based on measured sub-cloud aerosol size distributions, relatively standard droplet growth laws, gas exchange and droplet chemistry followed explicitly as a function of droplet size across the droplet size spectrum.