The vertical structure of aerosol and its relationship to boundary-layer thermodynamics over the rural UK

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SUMMARY

The vertical structure of aerosol, covering sizes from 0.05–4 μm radius, was examined under conditions of subsidence during winter and summer over the rural UK. Under well-mixed boundary layer conditions, dry accumulation mode aerosol was found to be well mixed with height. During the winter campaigns, nocturnal cooling resulted in the development of a stable surface layer, typically 100 m in depth, within which the surface emitted pollutants became trapped leading to concentrations significantly greater than that observed in the mixed boundary layer above it. Under stable boundary-layer conditions, the aerosol and water vapour vertical profiles exhibited strong negative gradients with height and were indicative of suppressed turbulence associated with stable boundary-layer conditions. During summer, the boundary layer was normally decoupled and possessed two cloud layers: cumuli forming just below, and penetrating the surface-layer inversion; and stratocumulus occupying the region under the boundary-layer capping inversion. Aerosol profiles under decoupled conditions exhibited considerable variability with peak concentrations being observed in the vicinity of cloud edges. Average aerosol concentrations in the main boundary-layer ranged from 209–651 cm\(^{-3}\) and 0.89–4.3 μm\(^{-3}\) cm\(^{-3}\) for dry number and volume respectively, whilst concentrations and volumetric loadings of 239–2430 cm\(^{-3}\) and 1.1–13.5 μm\(^{-3}\) cm\(^{-3}\) were encountered in surface layers. The majority of the aerosol number and mass concentrations were almost exclusively derived from the size range 0.05–0.2 μm radius with mode radii often occurring at 0.1 μm or larger. By comparison, free tropospheric aerosol possessed typically an order of magnitude lower concentrations and mass with an associated mode radius of 0.05–0.06 μm or less.

KEYWORDS: Airborne observations Boundary-layer decoupling Pollution Stratocumulus Sulphate aerosols Subsidence inversions

1. INTRODUCTION

In recent years, much attention has been directed towards understanding the effects of aerosols on a variety of processes in the earth’s atmosphere. Aerosols play an integral role in limiting visibility; they serve as nuclei for the formation of fog and cloud droplets; they affect the earth’s radiative budget, and thus climate, both directly and indirectly; and they inhibit the propagation of electromagnetic radiation. It has long been known that aerosols directly interfere with the incoming solar radiation flux (Angstrom 1929; Bergeron 1928) and that any perturbation of aerosol in the troposphere is likely to cause a concomitant perturbation in the earth’s planetary albedo and consequently climate. Estimates of the direct radiative forcing of anthropogenic aerosol and sulphate aerosol (Charlson et al. 1991) have illustrated that this effect is opposite in sign and comparable in magnitude to the radiative forcing from greenhouse gases such as CO\(_2\), with a typical forcing of −1.2 W m\(^{-2}\) in the northern hemisphere and between −1 and −2 W m\(^{-2}\) globally averaged (Charlson et al. 1992). The anthropogenic SO\(_2\) emissions reported in Charlson et al. (1992) suggest that the radiative forcing possesses an increasing trend of −0.2 W m\(^{-2}\) per decade (Harshvardhan 1993). More recent estimates of the effect of anthropogenic sulphate and soot aerosol radiative forcing are more conservative with a global mean forcing of −0.34 W m\(^{-2}\) resulting from sulphate aerosol and +0.06 to +0.24 W m\(^{-2}\) for soot aerosol depending on whether it is externally mixed (lower estimate) or internally mixed (upper estimate) with sulphate aerosol (Haywood and Shine 1994).

Twomey (1974) also suggested that aerosols will provide an indirect radiative forcing through modification of cloud albedo resulting from variations in cloud nuclei source strength. Based on estimates of the anthropogenic contribution of aerosol sulphate in

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eastern North America, Leaitch et al. (1992) reported that anthropogenic sulphate had increased the supply of cloud condensation nuclei (CCN) and thus augmented cloud-droplet number concentrations in that region and estimated the present-day climate forcing to be between $-2 \ W\ m^{-2}$ and $-3 \ W\ m^{-2}$. Modelling studies by Jones et al. (1994) suggest a global mean indirect radiative forcing of $-1.3 \ W\ m^{-2}$ by anthropogenic sulphate aerosol, although more recent simulations suggest a forcing between $-0.3$ to $-1.3 \ W\ m^{-2}$ (Jones and Slingo 1996). A similar range of forcing ($-0.3$ to $-1.5 \ W\ m^{-2}$) was found by Boucher and Lohmann (1995) in a sensitivity study of two general circulation models, however, they argued that these results possessed an associated uncertainty greater than the reported range of forcing.

In order to estimate the effect of aerosols on climate, accurate knowledge of the spatial and vertical distribution of aerosols is necessary. Although there is an abundance of data on aerosol concentrations, size and composition available from surface based platforms, information on the vertical structure is more sparse (Patterson et al. 1980; Kleinman and Daum 1991; Kilby and Smith 1990). The vertical structure of aerosol is required to validate extrapolation of surface layer measurements to the full atmospheric boundary layer. A general assumption of a well-mixed boundary layer within which the aerosol is also well mixed may not always be the case as illustrated in some studies (Frisbie and Hudson 1993; Heintzenberg et al. 1991; Leaitch and Isaac 1991). We present results from two airborne campaigns dedicated to examining the vertical structure of atmospheric aerosol and its dependence on boundary-layer thermodynamics over the rural north-west England.

2. Experimental

The UMIST research aircraft is a Cessna 184 light aircraft adapted for specialized atmospheric research. With modifications to accommodate instrumentation, logging computers, and control systems, it has a payload of 200 kg and two crew members—a pilot and an aircraft scientist. The aircraft has an operating ceiling of 4000 metres amsl and a typical cruising speed of 55 m s$^{-1}$. The Cessna meteorological and navigational instrumentation comprises much of the standard equipment found in research aircraft. For this study, the aircraft facility was modified to investigate the vertical structure and volatility of terrestrial boundary-layer and the lower free-troposphere aerosol.

(a) Ambient particle sampling techniques

The concentration and size of particles with radii between 0.05 $\mu$m and 4 $\mu$m were measured with a wing-mounted Particle Measuring System (PMS) FSSP and a PMS ASASP-X operating inside the aircraft. The FSSP was configured to sample particles over a range which covered the sizes 0.5 $\mu$m to 4 $\mu$m in 15 sub-channels whilst the ASASP-X sizes over the range 0.05 $\mu$m to 1.5 $\mu$m radius with 32 channel resolution. The manufacturers calibration was used and the ASASP-X was regularly calibrated using latex spheres. Optical particle counters are subject to sizing errors resulting from refractive-index variations associated with variations in aerosol chemical composition and thus, may result in a distortion of the size distribution. Calculation of the ASASP-X scattering response for soot carbon and ammonium sulphate aerosol indicates that, for sizes from 0.05–0.2 $\mu$m, the ASASP-X oversizes soot carbon by up to 20% (Jennings et al. 1994). Thus, size-distribution distortions may result if a significant amount of soot carbon is present, however, as particles in this size range are generally composed of a mixture of soot carbon and ammonium sulphate (Smith and O’Dowd 1995), with ammonium sulphate
normally dominating the concentrations, the error in the size distribution derived from
the ASASP-X is likely to be considerably less than 20%.

The ASASP-X had a dual configuration which allowed sampling of ambient air as
near to the intake as possible and also of thermally conditioned air from an array of
‘volatility’ heater tubes held at selective temperatures to infer aerosol composition. The
results of the volatility analysis are discussed in an associated manuscript (in preparation).
The air intake comprised a 2.5 cm diameter forward facing nozzle protruding 20 cm from
the left wing, aligned parallel to the direction of motion. The nozzle was attached to an air
duct, also of 2.5 cm diameter, which channelled the aerosol into the fuselage for analysis.
The air within the duct traversed two right-angled bends before being extracted from the
duct by a 0.31 cm diameter nozzle for direct sampling by the ASASP-X. After the first
sample extraction, the duct then fed a volatility box, comprising four heated tubes, each of
0.5 cm internal diameter, before providing thermally conditioned flow to the ASASP-X.
By insertion of a flow control plate with a specific orifice diameter in the exhaust duct,
overall flow in the sampling intake system could be controlled. The 0.31 cm diameter
nozzle for direct sampling by the ASASP-X faced into the direction of air flow for a length
of approximately 1 cm. Thereafter, a right-angle bend served to divert to-be-sampled air
out of the duct and into the ASASP-X via a no-loss selector valve which selected either
ambient or volatility flows. Initial tests to quantify any humidity effects on the aerosol
were performed by comparing aerosol sampled from one heater tube set to 40 °C extracted
directly from the intake duct. The test confirmed that the aerosol sampled directly was
‘dried’ by the intake duct and, possibly the optical chamber in the particle counter.

(b) Particle concentration enhancement and loss

Incorrect sampling techniques can lead to a distortion of the ambient size distribution
and, to ensure valid results, particle concentrations must be sampled in an isokinetic or near-
isokinetic manner. Such considerations are especially important in aircraft experimental
work as the sampling platform is moving at high velocities. In order to determine the
flow rate through the intake duct system, one flight was dedicated to differential pressure
measurements within the duct at varying aircraft speeds. These pressure measurements
were determined by inserting a pitot-static tube in the take-off for the ASASP-X which
fed into an airspeed indicator. Thus, the aircraft velocity and the air velocity within the
duct were recorded at different air speeds and were found to be linearly related. Typical
air speeds during research flights were 100 knots or 53 m s⁻¹. For this typical research air
speed, the air velocity in the intake duct was 9.5 m s⁻¹ (flow rate = 1484 cm³ s⁻¹, transit
time = 0.21 s) leading to non-isokinetic sampling. The non-isokinetic effect on sampling
efficiency for the aerosol particles is described by the ratio of ambient concentration C₀ to
sampled concentration C, following Davies (1968):

\[ \frac{C}{C₀} = 1 + \left[ \left( \frac{U₀}{U} \right) - 1 \right] \left[ 1 - \left( \frac{1}{1 + 2\Psi} \right) \right] \]

where \( U₀ \) is the aircraft velocity, \( U \) is the air velocity in the duct intake and \( \Psi \) is the inertial
impaction parameter (or Stokes number) and

\[ \Psi = \frac{\rho_p 2r_p^2 U₀}{9 \mu r_d} \]

where \( r_p \) is the particle radius, \( \rho_p \) is the particle density, \( \mu \) is the air viscosity and \( r_d \) is the
duct radius.
This non-isokinetic sampling enhanced large-particle concentrations at the duct intake, with an 18\% increase for particles with radii = 1.5 \mu m decreasing to 0.09\% for 0.1 \mu m radius. However, this effect was advantageous as, further down the duct, this enhancement was partially balanced by particle loss around the two right-angled bends. Particle loss in curvilinear motion occurs primarily when the inertia of the larger particles is too great for them to follow the flow stream lines around a bend.

The condition of equilibrium between centrifugal and frictional forces leads to the following expression for radial velocity:

\[
v_r = \frac{2\rho_p v^2 r_p^2}{9\mu R}
\]

where \( v \) denotes the gas velocity and \( R \) is the radius of curvature of the air flow. In the time of passage \( t \) the droplet migrates through a distance \( s' \):

\[
s' = v_r t = \frac{v_t \propto R}{v}
\]

so that the ratio of settling height \( s' \) to the channel width yields the separation efficiency \( \eta_f \):

\[
\eta_f = \frac{s'}{s} = \Psi \alpha
\]

where \( \alpha = 1.6 \) for a 90\° angle.

From the above equations, losses for a 90\° bend and air flow of 9.5 m s\(^{-1}\) were calculated and the maximum loss of 2\% was found to occur for the 1.5 \mu m radius particles. This loss was repeated for another two right-angle bends as the air travelled through the duct. The sample nozzle extracting aerosol from the duct to the ASASP-X was configured for near-isokinetic sampling with maximum particle gain reaching less than 1\% for the largest particles, but the right-angle bend induced a loss of 4\% for these large particles. The total accumulated size distribution distortion (plotted in Fig. 1) resulted in a concentration enhancement of 11\% for the largest particles (1.5 \mu m), 5\% for 1 \mu m radius particles and 0.1\% for 0.1 \mu m particles. This concentration increase was considered acceptable as there were also some minor gravitational losses for the larger particles in the duct, although simple calculations indicated that they were considerably less than the losses for the right-angle bends.

3. Results

Results are presented from winter and summer experiments which investigated aerosol properties and structure in the atmospheric boundary layer and lower free troposphere over the semi-rural north-west of England. These observations were obtained in the vicinity of Carlisle (54.33\°N 2.75\°W) during a period of persistent anticyclonic conditions which lasted for about two weeks during the winter case and 1 week during the summer case. Cases are presented which demonstrate the variation encountered during each field study and which could be considered representative of winter and summer aerosol loadings in stable- and weakly-convective boundary layers.

(a) Winter campaign

Meteorological Summary. The synoptic meteorological situation was characterized by a persistent high pressure system which deepened and tracked eastward over the duration
of the project. At the start of the project, the centre of the high pressure region was situated a few tens of kilometres to the west of the sampling area with a maximum pressure of 1034 mb. By 6 December 1991, the centre had moved over to the east coast of the UK and had strengthened to 1042 mb. The centre of this high pressure system had moved over Poland by 10 December 1991, with the surface pressure at the centre reducing slightly to 1040 mb. Generally, the local weather was characterized by freezing or near-freezing temperatures and light variable winds at the start of the project whilst further into the project, as the high pressure centre moved away, winds increased moderately and wind direction was usually in the SE–SW sector.

(i) Case I: 6 December 1991. Thermodynamic profiles for this case study, shown in Fig. 2, present a classic well-mixed boundary layer, capped by a strong inversion. Within this boundary layer, the air was buoyantly neutral with the air temperature following the dry adiabatic lapse rate and a corresponding increase in relative humidity with altitude throughout the layer. A shallow stratocumulus cloud-layer occupied the region just below the inversion, as indicated from the FSSP observations in Fig. 2. Above this strong inversion, the air was very dry, with the dew-point temperature declining rapidly from −5 °C to −35 °C and potential temperature, Θ, increasing from 5 °C to 17 °C. The equivalent potential temperature Θ_e indicated that the conditions in the boundary layer were well mixed.

During this study, the FSSP and ASASP-X size ranges employed were 0.5 μm ≤ r ≤ 4 μm and 0.05 μm ≤ r ≤ 1.5 μm, respectively. The concentration of accumulation-mode aerosol, measured by the ASASP-X, remained constant with height (N ≈ 500 cm⁻³) up
Figure 2. Thermodynamic and aerosol vertical profiles for Cases 1, 2 and 3 during the winter campaign. $N_x$, $V_x$ and $N_F$, $V_F$ are number concentration and volume measured by the ASASP-X and the FSSP respectively.
to cloud base confirming the well-mixed nature of the boundary layer. We should note again that the size distribution measured by the ASASP-X is a ‘dry’ size distribution and thus is not subject to variation with relative humidity. By contrast, particles measured by the FSSP are wet, and thus concentration is expected to increase as particles from the accumulation mode grow in the FSSP size range as relative humidity increases. This effect is seen in Fig. 2 where FSSP concentrations increase from less than 0.2 cm$^{-3}$ at the surface to 10–20 cm$^{-3}$ below cloud base and reaching nearly 300 cm$^{-3}$ in cloud. The FSSP number and volumetric loadings displayed a positive correlation with relative humidity but only became significant when compared with the ASASP-X values near and within the cloud layer. Air-mass back trajectories for this case also suggest a Polar/Arctic origin but with sufficient transit over the UK to incorporate a high aerosol loading into the air parcel.

The solid squares on the aerosol profile plots in Fig. 2 represent altitudes at which number concentration and volumetric size distributions were extracted for display in Fig. 3. The break in the size distribution in each case shown in Fig. 3 corresponds to the transition between ASASP-X and FSSP sizes. Number and volumetric particulate distributions for this case exhibit little variation within the well-mixed boundary layer with a slight shift in distribution towards larger sizes occurring between the 320 m and 870 m levels. Above the main inversion, a substantial change in the particle concentration and spectral shape may be noted, with the concentration reducing by more than an order of magnitude, and the mode radius shifting from 0.15 μm in the mixed layer to about 0.05 μm in the free troposphere. It may be noted that there is good correspondence between observations from the two instruments. Thus, with the ASASP-X intake configured for near-isokinetic sampling, the
distortion of the particle size distribution resulting from sampling artefacts was found to be negligible.

(ii) Case 2: 4 December 1991. Vertical profiles of the boundary-layer thermodynamic properties for case 2 (Fig. 2) reveal an interesting structure which comprised four layers within the lowest 1.5 km above ground level. The low-level structure consisted of a temperature inversion at about 100 m (1030 mb) above ground level, capping a cold, moist surface layer. This low-level layer resulted from strong nocturnal cooling and persisted throughout the day. Under this near-surface inversion, the relative humidity was approximately 90%, the air temperature, $T_{Air}$, was slightly above freezing at $\approx 1 \, ^\circ C$ with $\Theta \approx -3 \, ^\circ C$. Above this surface layer, a warmer drier layer extending to 1000 m was present with the relative humidity decreasing to 70% just above the surface inversion and $\Theta$ increasing to 6 C. This layer was thermodynamically neutral with the temperature profile following the dry adiabatic lapse rate. Immediately above the main inversion at 1000 m, there was a shallow warm ($\Theta = 15 \, ^\circ C$) layer about 200 m thick, within which the dew-point temperature fell to $-13 \, ^\circ C$, and which was itself capped by a much more stable, warm ($\Theta = 20 \, ^\circ C$) layer. $\Theta_{E}$ displays an almost identical profile to that of $\Theta$, also indicating a very stable surface layer under the well-mixed main boundary layer. Air-mass back trajectories at surface level and 850 mb for this case indicate both air parcels circulating around the centre of the high over the UK and Ireland for a period of about four days.

Aerosol concentration and volumetric vertical profiles for this case are also presented in Fig. 2. Peak aerosol concentration loadings are evident in the near-surface layer, with a concentration, $N$, between 2000 and 3000 cm$^{-3}$. Within the main boundary layer (100–1000 m), ASASP-X particle concentrations were generally well-mixed with values of $N$ around 500 cm$^{-3}$ which remained constant with altitude. Above the main inversion at 1000 m, concentrations fell initially to 20–30 cm$^{-3}$, before increasing to 80–90 cm$^{-3}$ in the shallow moist layer at 1200 m, and then declined again to around 20 cm$^{-3}$ at 1300 m. FSSP data correlated well with relative humidity changes, presumably due to particle growth from the sub-micron region to the super-micron region. Particles sampled by the FSSP ($r \geq 1 \, \mu m$) contributed little to the overall particulate volume or number concentration, possessing values more than an order of magnitude lower than the 'dry' ASASP-X particles with radii $r \leq 1.5 \, \mu m$.

In Fig. 3, particle size spectra over the range 0.05 $\mu m \leq r \leq 4 \, \mu m$ for Case 2 are presented for the altitudes indicated by the solid squares in Fig. 2. Peak concentrations were found within the surface layer, whilst the minimum concentrations occurred above the main inversion (1000 m). Within the mixed layer, there is no discernible difference between spectra sampled at 420 m and 720 m, illustrating that the aerosol in this layer is well mixed. A most interesting feature of these spectra is that the particle concentrations observed near the surface were almost an order of magnitude higher, for sizes with $r \leq 0.08 \, \mu m$, than those observed in the main boundary layer.

(iii) Case 3: 9 December 1991. Vertical profiles of the meteorological parameters, shown in Fig. 2, illustrate a stable and dry boundary-layer structure. A three-layer structure is seen here, with a stable surface layer 200 m in depth and surface air temperatures slightly below freezing. Within this surface layer, the relative humidity profile had a very sharp negative gradient (falling from 85% at the surface to 55% at the surface inversion), which is characteristic of a surface vapour flux being prevented from mixing vertically within this stable layer. $\Theta$ increased from $-2 \, ^\circ C$ at the surface to $5 \, ^\circ C$ at the surface layer inversion, above which $\Theta$ increases within the main boundary layer to $16 \, ^\circ C$ at what appears to be the main inversion at 800 m. $\Theta$ and $\Theta_{E}$ profiles suggest that boundary layer conditions were stable on this day thus suppressing vertical mixing.
Aerosol profiles reveal a slight negative gradient within the stable surface-layer but decrease more rapidly with altitude in the main boundary-layer. For this situation, both humidity and potential-temperature profiles would provide better information on the aerosol structure than equivalent potential temperature. Average particle concentration in the surface layer was $N = 2530$ cm$^{-3}$, with a dry volumetric loading of 13.8 $\mu$m$^3$ cm$^{-3}$. Above the surface inversion, concentration and particle volume fell sharply from about 1000 cm$^{-3}$ and 7 $\mu$m$^3$ cm$^{-3}$ to 20 cm$^{-3}$ and 0.2 $\mu$m$^3$ cm$^{-3}$, respectively, averaging 206 cm$^{-3}$ and 1.25 $\mu$m$^3$ cm$^{-3}$. Size distributions from this flight are shown in Fig. 3. Four spectra were taken, one from each of the layers described above, and number and volume concentrations were found to decrease significantly with altitude, as seen in the vertical profiles. The spectral shape, however, remained remarkably constant with altitude within the surface and main boundary-layers, with a near flat spectrum from 0.05 $\mu$m to 0.15 $\mu$m, but which declined rapidly for larger particle sizes. Above the main inversion, a notable change in both number concentration and distribution may be noted, with the spectral shape acquiring characteristics more like a log–normal distribution. The FSSP spectra do not agree well with the ASASP-X data and have much lower concentrations in the overlapping ranges. On take-off during the original flight on this day, the Cessna engine became engulfed in flames after technical problems, resulting in high concentrations of both smoke and fire extinguisher aerosol in the vicinity of the FSSP which almost certainly resulted in contamination of the probe optics. Due to the shortage of daylight hours at these latitudes during this time of year, once the Cessna was considered airworthy again, the flight was continued at the expense of FSSP maintenance and recalibration.

(c) Summer Campaign

Meteorological Summary. The synoptic meteorology throughout the summer field project was characterized by two high pressure systems straddling the UK with a low pressure system over Iceland. The extended occluded front associated with the Icelandic cyclone was just to the north of Carlisle in a SW–NE orientation. At the start of the project (24 June), the front was nearest Carlisle with associated winds of 5–8 m s$^{-1}$ in a NW direction. On 25 June, the front retreated northwards and winds backed to a WSW direction with surface winds at about 5 m s$^{-1}$. This pattern continued on 26 June with the front moving further north and the winds backing to a more south-westerly direction but remaining at about 5 m s$^{-1}$. The resulting daytime boundary-layer comprised a weakly convective surface layer with small cumuli topped by a slightly-stable mixed layer with stratocumulus cloud cover. Cloud cover in both layers increased inland as the ground level rose toward the Pennine Fells.

(d) Summer Case Studies

(i) Case 4: 24 June 1992. Meteorological observations indicated two levels of cloud with small cumuli between 500 m and 1200 m providing cover ranging from about 2–7/8th and stratocumulus between 1200 and 1500 m giving 5–8/8th coverage. Thermodynamic profiles are illustrated in Fig. 4 and show a boundary layer about 1500 m in depth, with a relatively strong capping inversion. Examination of the temperature profiles within the boundary layer indicates the presence of a convective surface layer with a weak inversion at approximately 600 m. The surface layer was slightly convective with a surface vapour flux, as reflected in the equivalent potential-temperature profile. Above the surface layer the $\Theta_e$ profile indicates that it was well-mixed. The thermodynamical structure is typical of a stratocumulus-covered well-mixed boundary layer which transitioned into a decoupled surface layer and a coupled sub-cloud and cloud mixed-layer after cloud-top radiative
Figure 4. Thermodynamic and aerosol vertical profiles for Cases 4, 5 and 6 during the summer campaign. $N_x$, $V_x$ and $N_F$, $V_F$ are number concentration and volume measured by the ASASP-X and the FSSP respectively.
cooling (Nicholls and Leighton 1986). The lack of mixing throughout the boundary layer results in the build up of moisture in the surface layer which can penetrate the surface inversion after forming cumuli. These cumuli can then transport moisture and aerosol into the coupled sub-cloud and cloud layer. Aerosol concentration and volumetric profiles are illustrated in Fig. 4 and show vertical variations of about 50%. A peak in the particle concentration of 320 cm\(^{-3}\) occurred above the interface between the convective surface layer and the sub-cloud layer in the vicinity of cumulus edges, although the average concentrations were similar in both layers (\(N = 239 \text{ cm}^{-3}\), \(V = 1.3 \mu \text{m}^{-3} \text{ cm}^{-3}\) in the surface layer and \(N = 251 \text{ cm}^{-3}\), \(V = 1.43 \mu \text{m}^{-3} \text{ cm}^{-3}\) in the sub-cloud layer). The variation in aerosol concentration within the boundary layer is thought to result from cloud-aerosol interactions where gas-phase aerosol precursors, such as SO\(_2\), are converted to aerosol mass in the aqueous phase thus modifying the size of existing aerosol (Hoppel et al. 1986). Above the boundary-layer inversion, particle concentrations fell off rapidly to approximately 35 cm\(^{-3}\) and, thereafter, remained constant with height, indicating little mixing across this strong inversion.

Size distributions, taken in the surface layer (at 75 m), around a cumulus-cloud edge (at 700 m) and just under stratocumulus-cloud base (at 1030 m), are shown in Fig. 5. In the vicinity of the edge of cumulus, the aerosol concentration was observed to be significantly higher (30%) than that observed in the surface layer, particularly at sizes smaller than 0.1 \(\mu\)m radius. What appears to be happening here is that aerosol smaller than 0.05 \(\mu\)m radius are activated in the relatively high supersaturations associated with cumulus clouds and in the cloud medium, they increase in size through heterogeneous reactions leaving
larger nuclei on evaporation. When the spectra near the cumulus edge, and just under stratocumulus cloud base, are compared the number concentration is similar, however, particles in the 0.05–0.08 μm range have grown into sizes around 0.1–0.15 μm. Kerminen and Wexler (1995) have shown that neither condensation or coagulation processes can occur over sufficiently short time-scales to be able to explain the growth of particles from sizes of 0.05 μm to 0.1 μm, or, the formation of the accumulation mode centred around 0.1 μm. Their growth calculations indicated that the only growth mechanism capable of explaining the frequently observed accumulation mode is aqueous-phase production of sulphate aerosol in cloud or fog droplets through heterogeneous oxidation of dissolved SO₂.

It appears that the supersaturation reached in the stratocumulus cloud is high enough only to activate and chemically process nuclei larger than 0.05 μm, resulting in a size distribution narrower in width but higher in amplitude. Since the smallest activated aerosol particles lead to the lowest solute concentration in cloud droplets, these nuclei are capable of acquiring more relative mass than larger nuclei and consequently they grow more rapidly than the larger particles, thus leading to a narrowing of the size distribution.

(ii) Cases 5 and 6: 25 and 26 June 1992. These two cases are combined since the observed thermodynamic and aerosol structure were very similar. The cloud structure was similar to the previous case and variations along the inland traverse made it difficult to discern any notable differences. The thermodynamic profiles in Fig. 4 indicate the presence of a convective surface layer up to 800 m on 25 June and 900 m on 26 June, each topped by slightly stable mixed layers. The presence of a stratocumulus-cloud layers, as previously mentioned, can lead to a decoupling of the mixed layer into a surface layer and a coupled sub-cloud and cloud-mixed layer. With this decoupling, the vapour flux from the surface to the top of the boundary layer is inhibited, possibly leading to the break-up of the stratocumulus-cloud layer. It should be noted that, generally, all soundings were conducted through breaks in the cloud, thus cloud layers are not very evident from the relative humidity profiles, however, on a couple of occasions cumuli were penetrated. Particle profiles on both days show similar distinctive aerosol concentration layers (Fig. 5), although for case 6, the boundary layer extends to a greater altitude when compared with case 5. For case 5, aerosol concentrations were relatively constant with height in both the surface layer and the decoupled sub-cloud layer, averaging around N = 500 cm⁻³ and N ≈ 184 cm⁻³, respectively. By comparison, for case 6, the surface-layer aerosol profile exhibited a more negative gradient with altitude and there was a build-up of aerosol just below the surface-layer inversion. Surface aerosol concentrations were about 550 cm⁻³, falling off to approximately 300 cm⁻³ at 900 m. The enhanced aerosol concentrations within the surface layer in case 6 relative to that of the previous day, might have been a consequence of the backing of the wind to a more southerly direction as the cold front straddling Scotland retreated further north, thus allowing the air mass a longer transit time over the UK and Ireland to accumulate pollutants. On this occasion, the aerosol concentration declined with altitude within the stratocumulus- and cumulus-cloud layers, due to a considerable mixing between the surface layer and the sub-cloud–cloud layer and between the sub-cloud–cloud layer and the free troposphere (O’Dowd and Smith, to be submitted). Average concentrations within this layer were about 169 cm⁻³.

Four concentration and volumetric distributions obtained within the boundary layer and free troposphere are shown in Fig. 5 for both cases. In the lower surface layer, the highest concentrations were observed at the smallest sizes, indicating new particle formation in the nucleation mode and subsequent condensation growth. Above the surface layer inversion, the distributions more closely resemble a log-normal curve and concentrations are
substantially lower than within the surface layer. On 26 June, despite the meteorological conditions being similar to the previous day, the shape of the particle spectra extracted from the surface layer observations were somewhat different with no indication of a nucleation mode, although the spectra were otherwise similar.

4. Discussion

The average aerosol-number and mass concentrations for all cases during the campaigns are tabulated in Table 1. Average boundary-layer concentrations varied from 157 cm$^{-3}$ to 2530 cm$^{-3}$ with the volumetric loadings ranging from 0.89 $\mu$m$^{-3}$ cm$^{-3}$ to 13.84 $\mu$m$^{-3}$ cm$^{-3}$. By comparison, free tropospheric concentrations and volumetric loadings were typically an order of magnitude lower and range from 22 to 60 cm$^{-3}$ and 0.11 to 0.27 $\mu$m$^{-3}$ cm$^{-3}$, respectively, for number and mass concentration. Mostly, the highest loadings were observed near the surface, particularly when nocturnal cooling resulted in a stable surface-layer within which pollutants emitted from the surface were trapped, however, peak concentrations were also observed in the vicinity of cloud edges, suggesting that evaporating nuclei in clouds possess greater mass when compared to their initial mass. Actual ambient aerosol size, and consequently volume, will be significantly higher than the values presented here since these particles are subject to growth with changes in ambient relative humidity.

Size distributions indicate that, by far, most of the aerosol mass resides within the 0.05–0.2 $\mu$m radius range. Aerosol larger than 0.2 $\mu$m contribute typically 1–2 orders of magnitude lower mass than sizes smaller than 0.2 $\mu$m. Boundary-layer number concentrations possess a peak concentration at sizes around 0.1 $\mu$m radius with a local lower minimum located around 0.05–0.07 $\mu$m, although, near the surface, some cases appear bi-modal with a secondary peak around 0.05–0.06 $\mu$m radius, reflecting the input from local sources. By comparison, free tropospheric aerosol consistently possessed a monomodal peak at sizes of 0.06 $\mu$m and less. In an associated manuscript, we illustrate further that cloud-free boundary layer aerosol and cloud-free free tropospheric aerosol possess similar spectral characteristics with a mode radius centred around 0.05–0.06 $\mu$m radius, and, by comparison, spectral characteristics of aerosol in the cloudy boundary layer always possess a much larger mode radius at around 0.1 $\mu$m. Growth-law calculations (Kerminen and Wexler 1995) suggest that the only process capable of producing the observed accumulation-mode peak at 0.1 $\mu$m is chemical processing of aerosol by clouds.
The difference between boundary-layer aerosol and free-tropospheric aerosol is thus attributed to cloud processing of boundary-layer aerosol which provides an effective mechanism for aqueous phase gas-to-particle conversion in cloud droplets. The variability of the aerosol throughout the boundary layer suggests that extrapolation of ground-based concentrations of aerosol to higher levels, and even to cloud base, will not be accurate unless the boundary layer is thermodynamically well mixed.

5. Conclusions

During the two anticyclonic periods examined, evidence was found for substantial variations in boundary-layer properties and the structure of the lower free troposphere, with concomitant variations in the vertical aerosol-number-concentrations and volumetric loadings. The conditions described in this study cover a neutral, well-mixed, boundary layer, a boundary layer containing well-mixed layers with a stable surface-layer, a stable multi-layered boundary layer and a stratocumulus-induced "decoupled" boundary layer. The strong nocturnal cooling associated with winter-time anticyclonic conditions was responsible for the multi-layered structure. Such cooling can give rise to very stable conditions within near-surface layers, typically of 100–250 m in height, which frequently contain low-level haze or fog due to the high humidities induced by nocturnal radiative cooling and day-time vapour fluxes. Since particulate and gaseous emissions from the surface can become trapped in such layers, the observed particle concentrations are often much higher than within the main boundary layer, and their relatively recent origin results in much greater concentrations of very fine particles (r < 0.08 μm). Typical concentrations observed ranged from about 1500 to 5000 cm⁻³ and could show either a positive or negative gradient with height.

In neutral boundary-layer situations where the dry aerosol is well-mixed throughout the layer, actual wet-particle volumetric loadings are dependent of relative-humidity variations with altitude. Under more stable conditions, atmospheric turbulence (and hence aerosol vertical transport) could be suppressed and particle concentrations were found to decline with altitude. Typical concentrations and volumetric loadings within the main boundary layer were around 500 cm⁻³ and 4 μm⁻³, respectively, for inversion heights ranging from 1100 to 1500 m. Stable potential-temperature profiles were generally associated with negative gradients in the aerosol and relative-humidity profiles. In these cases, the correlations between aerosol concentration and relative humidity were not thought to be due to particle-size dependence on relative humidity, but to the lack of vertical mixing of both precursor gases and water vapour within these layers, accompanied by decreased particle production associated with much drier air at the higher altitudes.

Decoupled mixed boundary-layer characteristics were observed under stratocumulus cloud more often in summer than in winter. This decoupling resulted in convective surface layers under initially well-mixed cloud and sub-cloud layers, which developed into stable layers through mixing with the free troposphere and thus resulted in the break-up of the cloud decks. During the summer-time decoupled conditions, the surface layer contained small cumuli which penetrated into the upper boundary-layer and in the vicinity of the small cumuli, enhanced particle concentrations were observed. The presence of clouds is thought to be a very important factor in the modification of aerosol concentration and volume characteristics and, when these clouds evaporate, the residual nuclei can possess greater size when compared with the initial nucleating aerosol.

In the free troposphere above the boundary-layer inversion, particle concentrations were much reduced, being typically 20 cm⁻³, but varied between 8 and 80 cm⁻³ depending upon structure and moisture content. Also, the size distributions observed in the free
troposphere were distinctly different from those recorded within the lower boundary-
layer, with the upper distributions exhibiting mode radii of around 0.05 μm in all cases,
compared with the generally observed mode radius of about 0.1 μm in the boundary layer.
The difference between boundary-layer aerosol size distributions and free-tropospheric size distributions is attributed to enhanced growth of aerosol through cloud processing.

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