Air parcel thermodynamic properties and aerosol particle concentrations in the maritime atmospheric boundary layer

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SUMMARY

In some circumstances, the distribution of particles in the maritime atmosphere may be simply related to the atmospheric profile. For data obtained in the Outer Hebrides in 1982 by the Meteorological Office Research Flight aircraft, a strong correlation is found between particle concentrations over the radius range from 0.25 to 4 μm and the thermodynamic properties of air parcels at various levels within the marine atmospheric boundary layer. This approach offers a potential tool toward better understanding of particle distribution and provides a means for estimating particle concentrations at various levels on the basis of a standard meteorological sounding.

1. INTRODUCTION

In recent years, there has been considerable interest in the employment of conserved thermodynamic properties in unifying our understanding of cloud physical processes, as exemplified by the work of Betts (1973, 1982a, b and 1983) and Paluch (1979). Paluch’s seminal study of the usefulness of thermodynamic air parcel tracing in studying the properties of clouds has been clearly demonstrated in a number of investigations of mixing between cumulus clouds and their environments. Basically, when applied to the study of cumulus cloud properties, this technique employs two parameters which are conserved during adiabatic altitude changes and which mix in a nearly linear manner. These parameters are the cloud water specific humidity, Q, and a temperature parameter related to the specific entropy, named by Paluch the wet equivalent potential temperature, θ_w; clearly, both these parameters must take into account the contribution of the liquid phase when dealing with cloud properties. Utilizing this approach for studies of non-precipitating Colorado cumulus clouds, Paluch presented data from successive horizontal traverses, at various altitudes, on a graph of Q against θ_w. By plotting an environmental sounding (taken in the clear air outside the cloud) on the same graph, she demonstrated that the air within the cloud could only consist of a proportionate mixture of air from cloud base with that situated at the cloud top level.

The need for a better understanding of the processes governing the distribution of aerosol particles, produced at the sea surface, within the marine atmospheric boundary layer prompted the application of this elegant parcel-tracing technique to the North Atlantic marine aerosol studies being conducted at UMIST.

In the maritime environment typical of the North Atlantic, the relatively warm ocean surface is often overlaid by a cooler air mass. Thus, air parcels—warmed and moistened by contact with the sea surface—will rise and mix with air from higher levels, which contains reduced quantities of water vapour. For these maritime air masses, the aerosol particle concentration, for radii greater than about 0.25 μm, is dominated by particles released from whitecaps generated at the sea surface by the action of the wind. These particles, apart from those large enough to have appreciable fall velocities relative to the air motions, will be similarly transported within their air parcels and diluted by this environmental mixing.
In order to investigate these processes, information is required on particle concentrations within the size range of interest, together with measurements of the related atmospheric thermodynamic properties, thoughout the marine boundary layer. Such a sequence of measurements was obtained by the Meteorological Office Research Flight aircraft in collaboration with the UMIST field programme conducted on South Uist (one of the islands in the Outer Hebrides situated off the north-west coast of Scotland) during summer 1982. The aircraft operated on two occasions within this period, on 18 and 26 August, but relevant data are only available for the earlier date, and these data form the basis of this analysis.

2. METEOROLOGICAL SITUATION

The synoptic meteorological situation is illustrated by the surface weather chart for 12 GMT on 18 August (Fig. 1) and the associated satellite photograph (Fig. 2) taken at 1420 GMT. The situation was dominated by a low pressure area centred to the south-east of Iceland with a further low located over the North Sea. A cold front, situated over central and southern England at 09 GMT, had moved southwards with its associated rainfall, leaving the remainder of the British Isles affected by a showery north-westerly airstream. The 12 GMT sounding at Stornoway indicated wind directions at the surface and at 500 mb of 290° and 245°, respectively, with associated wind speeds of 4.5 m s⁻¹ and 8 m s⁻¹. Surface observations at Benbecula gave corresponding values of 270° and 8.5 m s⁻¹, respectively.

Standard trajectory analysis indicated that the air mass present over the site had travelled from a location to the south of Iceland during the previous 24 hours and, consequently, was of polar maritime type. Such air masses are usually found to be unstable and are generally associated with shower activity. Cloud base was estimated at about 600 m, which corresponded closely with observations made from the aircraft on the day.

Figure 1. Surface weather chart for 12 GMT on 18 August 1982. The arrow indicates the experiment site.
Figure 2. Satellite photograph (visible wavelength) for 1420 GMT on 18 August 1982. Photograph by courtesy of University of Dundee.
3. FIELD MEASUREMENTS

A comprehensive description of the field site at Ardvachar Point on the north-west tip of South Uist is incorporated in Exton et al. (1985), which also includes details of the ground-based instrumentation and its layout.

For the measurements on 18 August, the C-130 aircraft was equipped with two Particle Measuring Systems (PMS) probes, an ASSP covering the radius range from about 1 to 10 µm, and an FSSP for particle radii from 0.25 to 23 µm. Measurements of Aitken nucleus concentrations were also made. In addition, the aircraft was instrumented to provide information on various meteorological parameters including dry-bulb and dew-point temperatures, specific humidity, and sea surface temperature together with wind speed and direction.

Upon arrival at the field site, the aircraft made a spiral descent over the sea, close to Ardvachar Point, followed by a sequence of horizontal flights at a number of specific altitudes above the sea and, finally, a spiral ascent, again over the sea, at the end of the measurement period. Each constant-altitude flight consisted of an outward and return run terminating at the ground site and oriented along the prevailing wind direction, which was around 270° during the observation period.

Throughout this data analysis, particle concentrations measured on range 3 of the FSSP, covering a radius range from 0.25 to 4 µm, are utilized since these particles are sufficiently large to be overwhelmingly of sea surface origin within the prevailing air mass and yet are small enough to have quite low fall velocities. Also, they are generally sufficiently numerous to give statistically realistic concentrations and size spectra over relatively short horizontal displacements (or sample periods).

Although the ground-based UMIST instrumentation was not fully operational during this aircraft flight, measurements of aerosol concentration were being recorded from an FSSP device, together with local meteorological parameters. Thus, these data are available for comparison purposes.

4. DATA ANALYSIS

One difficulty in considering the variation of particle concentration with altitude concerns the substantial variability which frequently exists within any given data set. This variability is clearly apparent in the ground-based measurements made, over the years, at South Uist during periods when the ambient conditions were relatively constant. When dealing with observations made from an aircraft, these variations are even more noticeable since the aircraft may rapidly transit from one region to another during flight. An example of the variability of particle concentration along a typical horizontal trajectory is illustrated in Fig. 3, where changes by a factor of two or so are apparent between successive aerosol samples. Since each particle concentration value represents an integration over a 10-second period, corresponding to a horizontal displacement of approximately 1 km, the true variability is likely to considerably exceed this factor. These fluctuations in concentration are substantially greater than the statistical sampling errors, which are indicated in the figure by vertical bars. The corresponding variations, in the horizontal, of specific humidity, relative humidity and wet equivalent potential temperature (calculated as shown below) are included in this figure for comparison purposes.

A vertical atmospheric sounding, taken by the aircraft during the project, is shown in Fig. 4, which illustrates the variation of potential temperature and specific humidity with altitude. The potential temperature, $\theta$, was derived from the dry-bulb temperature, $T$, by the equation

$$\theta = T \left(\frac{1000}{(p - e)}\right)^r$$  \hspace{1cm} (1)
where \( p \) and \( e \) are atmospheric and water vapour pressures in millibars, respectively, and \( T \) is in K. The ratio of the gas constant to the molar heat capacity, \( x \), is given by the expression

\[
x = x_d (1 + 0.61Q)/(1 + 0.87Q)
\]

where \( x_d \) is the value for dry air.

Figure 3. Typical horizontal aircraft traverse at an altitude of 150 m showing the variability of: (a) particle concentration, \( N \), \((0.25 < r < 4.0 \mu m)\); (b) specific humidity, \( Q \); (c) relative humidity, \( RH \); and (d) wet equivalent potential temperature, \( q_e \). \( D \) is the distance from the measurement site at Arivachar Point.

Figure 4. Variation of specific humidity, \( Q \), and dry air potential temperature, \( \theta \), with altitude.
This sounding is fairly typical of a maritime atmospheric boundary layer and indicates a well-mixed layer up to about 1 km, at which height there is a well-defined inversion. Also, it may be noted that the specific humidity decreases rapidly above this capping inversion. Throughout this layer, the air is cooler than the sea surface and, therefore, atmospheric instability and the concomitant mixing into this region of air parcels, warmed at the sea surface, is to be expected.

In order to consider more fully the question of atmospheric stability, the contribution of water vapour must be taken into account. Following Paluch, values of wet equivalent potential temperature were calculated using the expression:

$$\theta_w = K \theta$$  \hspace{1cm} (3)

where

$$K = (1000/p_{at})^{-1.12r} \exp[rL/(T_c(c_{pd} + c_{w}r))]$$  \hspace{1cm} (4)

in which $r$ is the water vapour mixing ratio ($= Q/(1 - Q)$), $p_{at}$ and $T_c$ are the partial pressure of dry air and the temperature, respectively, at the condensation level. $L$ is the latent heat of vaporization and $c_{pd}$ and $c_{w}$ are the specific heats of dry air at constant pressure and liquid water, respectively.

These values of $\theta_w$ are plotted against altitude in Fig. 5, together with specific humidity: the substantial degree of correspondence between the two parameters, especially in the lower 1 km, is clearly visible. High values of $\theta_w$ near the surface may be noted and are due to the contribution of moisture to atmospheric stability at these levels, whilst the high values above about 3.5 km are a consequence of the high potential temperatures at these altitudes. Thus, the atmosphere is conditionally unstable up to a height of about 4 km, which corresponds approximately to observations of cloud top altitudes on the day in question.

![Figure 5. Variation of specific humidity, Q, and wet equivalent potential temperature, $\theta_w$, with altitude.](image-url)
The aerosol particle concentrations for the lower 2 km of the atmospheric boundary layer, measured during the aircraft ascent, are presented in Fig. 6 with the $\theta_q$ and $Q$ curves plotted alongside. It may be noted that there is a general similarity of shape between all three curves apart from two regions (marked X and Y), at about 730 m and 1100 m, where anomalously large concentrations are found. More detailed examination of the data for these regions indicates the presence of significant numbers of particles in the FSSP larger-size channels, suggesting that the higher counts are a consequence of condensational growth, a view supported by relative humidity measurements in excess of 80%. The associated descent profile, taken about 2 hours previously, shows a similar atmospheric structure but with only the lower anomalous region present at a lower altitude of about 600 m. Thus, the region X may form part of a continuous layer, whereas the region Y seems more localized. The close similarity between the $\theta_q$ and $Q$ curves shows that the variations in $\theta_q$, at these low altitudes, are dominated by changes in water vapour density rather than by variations in the temperature structure. Above the marine boundary layer inversion, at altitudes greater than 1200 m or so, the particle concentrations are very low, which confirms the view that the aerosol within this size range is predominantly of sea surface origin.

![Diagram showing particle concentration, temperature, and humidity variations](image)

Figure 6. Variation of particle concentration, $N$, with altitude, $H$, with $Q$ and $\theta_q$ curves included for comparison.

As mentioned previously, Paluch demonstrated that air parcels which consist of a mixture of air from two levels should fall along a straight line when plotted on a graph of $Q$ against $\theta_q$. Furthermore, this straight line would intersect the environmental sounding, when plotted on such a graph, at points corresponding to the two levels in question. When the ascent data from this project are plotted in this manner (Fig. 7), it may be noted that the points corresponding to altitudes from the surface to almost the 2 km level fall approximately on a straight line. Thus, air between these two levels may be regarded as a proportionate mixture of air from close to the sea surface with air from just above the marine inversion. Since the air above this inversion is essentially particle-free, we might anticipate that aerosol concentration would show a similar proportionate mixing within the mixed layer.
Figure 7. Variation of specific humidity, $Q$, with wet equivalent potential temperature, $\theta_e$, for the C-130 ascent.

Figure 8. Variation of particle concentration, $N$, with $\theta_e$ for aircraft (○) and ground-based (+) data. The regression line is derived from the 562 aircraft data points only (excluding those marked X and Y—see text) and has the form $N = 0.482\theta_e - 12.38$ (correlation coefficient is 0.82).

A graph of particle number concentration, $N$, against $\theta_e$ is presented in Fig. 8 and does, indeed, show a close correlation between these two parameters, thus strongly supporting the proposed hypothesis. In these specific circumstances, the higher particle concentrations are clearly correlated with air parcels containing more substantial quantities of air originating close to the sea surface. The points associated with the anomalously high particle concentrations are labelled X and Y, as in the ascent profile, and are
excluded from the correlation analysis. The simple least-squares regression line, fitted to
the aircraft data only, is shown in this figure and it may be noted that the values of
particle concentration and $\theta_q$ for the ground-based measurements obtained over the
concerning period (shown as crosses) are in good agreement with the projection of
this line. Thus, for these data, the variation of particle concentration throughout the
mixed layer is explicable simply in terms of the dilution of particle-laden air whose origin
is close to the sea surface with particle-free air from above the capping inversion.

Since the sea surface is a source of water vapour in addition to aerosol, a similarly
strong correlation between $N$ and $Q$ might be expected, especially with changes in $\theta_q$
closely following variations in $Q$ (as mentioned in connection with the ascent profile,
Fig. 6). This hypothesis is confirmed by a scatter plot of $N$ against $Q$, shown in Fig. 9,
although the correlation coefficient is somewhat reduced compared with the previous
figure. In contrast, however, similar analysis between $N$ and $\theta$, the dry air potential
temperature, indicates a much poorer—and inverse—correlation, refuting the intuitive
notion that $\theta$ would be equally appropriate in this analytical approach.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig9.png}
\caption{Variation of particle concentration, $N$, with specific humidity, $Q$, for the aircraft data only. The
regression line was derived as in Fig. 8 and is given by $N = 0.793Q - 2.884$ (correlation coefficient, 0.78).}
\end{figure}

5. Discussion

The analysis presented for the C-130 data gathered on 18 August 1982 demonstrates
that, for particles produced at the sea surface and generally present in low concentrations
above the marine atmospheric boundary layer, a simple model of air parcel mixing is
adequate to describe their concentrations within this mixed layer.

Since these aerosol particles are strongly hygroscopic, their equilibrium sizes will
depend upon the ambient relative humidity and, therefore, their concentrations should
be adjusted to take account of relative humidity changes throughout the mixed layer.
However, previous aerosol studies in the Outer Hebrides (Exton et al. 1985) suggest that
the influence of humidity upon particle concentrations, within this size range, will be
comparatively small for relative humidities below about 90%. With the exception of the
three points excluded from the correlation analysis, the mean relative humidity for all the aircraft observations was approximately 68% with a standard deviation of 4%. Thus, it seems unlikely that variations in relative humidity are a major contribution to the scatter in the data set, which is more probably due to differences in response characteristics of the measuring instruments and to the inherent limitations of this approach.

Also, strictly, the exchange of water vapour between the aerosol and the atmosphere should be included in the calculations of $\Theta_q$ in an analogous manner to cloud liquid water in the cumulus studies. However, this contribution to $\Theta_q$ should be insignificant compared with the other components in situations relevant to the analysis presented here.

Clearly, many more data are required under a variety of differing circumstances in order to investigate the general applicability of this approach. The effects of relative humidity upon particle concentration must also be incorporated if similar analysis is to be applied to air parcels approaching the saturation point. The technique will only be relevant for specific atmospheric conditions, although such conditions should occur fairly frequently in the North Atlantic environment.

The limited temporal resolution of the aircraft observations of atmospheric thermo-dynamic properties render it uncertain as to whether the air parcels are truly mixed or are simply the instrumental integration of successive plumes of air with differing properties and particle concentrations. It is anticipated that the UMIST aircraft, which flies more slowly than the C-130 and has been instrumented with fast-response humidity sensors in addition to PMS probes, should be capable of resolving this question and of providing a much more comprehensive data set during continuing measurements in the Outer Hebrides.

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