The effect of mixing height on maritime aerosol concentrations over the North Atlantic Ocean

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SUMMARY

An empirical investigation of the effect of the stability of the atmosphere on near-surface maritime aerosol concentrations is presented. The data base consists of an extensive series of aerosol experiments conducted on the western coast of the Outer Hebrides.

In the absence of any well-defined capping inversion over the Atlantic, stability is characterized in terms of convective and mechanical mixing heights, derived from radiosonde ascent data and from friction velocity, respectively. After wind-speed-production effects have been removed from the data, a simple relationship of the form

\[ \text{concentration} = A + B/(\text{mixing height}) \quad (A, B \text{ constants}) \]

was found to adequately describe most data subsets. A discussion of the errors involved in such a model is given.

1. INTRODUCTION

The concentrations, radius distribution, and profile of the sea-salt aerosol particles in the atmosphere are important for several reasons. The particles attenuate electromagnetic radiation; they are implicated in cloud formation and the development of precipitation; they may play a significant role in the sea–air moisture flux under high wind conditions (Smith et al. 1983); and they may carry pollution and bacteria from the sea to the air (Blanchard and Syzdek 1972).

In order to examine the effects of various meteorological parameters on the concentrations of aerosol particles in the size range 0.1 ≤ r ≤ 23.5 μm, an extensive series of aerosol experiments was conducted at Ardvichlar Point, on the island of South Uist, off the north-west coast of Scotland. These data were analysed primarily for wind speed effects, but it appeared reasonable to postulate that aerosol concentration at South Uist could also be dependent on atmospheric stability. The relationship between aerosol concentration and wind speed was shown to be considerably stronger in more stable tropical maritime air masses than in less stable polar maritime air masses and it was hypothesized that this was owing to the greater particle concentration build-up possible when there was a lesser volume of atmosphere available for occupancy by these particles (Exton et al. 1985).

The phenomenon of the dilution of concentrations of surface-produced aerosol particles with increasing turbulence has been reported over land; Choularton et al. (1982) found that diurnal variations of the height of the mixing layer (as gauged by Richardson’s number) have a profound effect on the concentration of all particles at a remote, high-level sampling site. For particles of radius less than about 3 μm, concentrations were enhanced in stable conditions owing to the constrained depth over which mixing could occur; for particles larger than this, concentrations were depleted under stable conditions, as gravitational settling could then remove them. Also, the concentration in the atmosphere of radon (a naturally-produced radioactive gas given off by the earth at a fairly constant rate) has been shown to be inversely related to the amount of convective activity (Larson and Hoppel 1973), wind speed and turbulence (Li 1974), and mixing layer height, as measured by acoustic radar (Hsu et al. 1980).
These results, however, were not in agreement with other work published upon the subject. Goroch et al. (1980) considered a simple model in which gravitational settling was balanced by upward turbulent transport. They looked at theoretical aerosol concentrations at different heights under various conditions of wind speed, humidity and air and sea temperatures. They found that for wind speeds of greater than 10 m s⁻¹, stability was unimportant in determining aerosol concentrations, and that, for low wind speeds, at a height of 20 m, the amount of aerosol present was increased by up to an order of magnitude under unstable, as opposed to neutral, conditions, for aerosol of sizes 1–10 μm radius.

Davidson and Schütz (1983) used these theoretical considerations to investigate the effects of stability in JASIN-78 data. Stability was characterized by z/L, calculated from Barker and Baxter's (1975) expression. A theoretical neutral stability concentration was calculated from a published equation for each wind speed and used to normalize the measured particle concentrations for wind speed and humidity. For 1 μm particles, there were more present in unstable conditions than in neutral conditions, and more present in neutral conditions than in stable conditions—the opposite of the putative South Uist findings.

The stability of the atmosphere may have a significant effect on marine aerosol particle concentrations, but more detailed work is required to discover the exact nature of the relationship. This paper is an attempt to derive an empirical formulation of the relationship between concentrations of particles of various sizes and stability, for all available maritime South Uist data sets.

2. THE MODEL

The ejection of sea water droplets in the size range sub-micrometre to several millimetres radius from the sea surface by the bubble-bursting mechanism is well documented (Blanchard 1963; Day 1964; MacIntyre 1972; Cipriano et al. 1983). The number and size of the droplets produced depend upon many factors, e.g. wind speed history, fetch, water temperature, air/sea temperature difference, salinity, water contaminants, but primarily upon the number and size distribution of the entrained air bubbles, and thus, indirectly on the wind speed (Monahan and O'Muircheartaigh 1980; Exton et al. 1985). The particles thus generated will initially inhabit the lowest metre of the atmosphere (Wu 1979a) where the sea-salt concentration is very high, sometimes greater than 1000 μg m⁻³ (Monahan 1968). From here, some particles, especially the larger ones, will fall back into the sea, and others are transported upwards by turbulence. At some height, H, the particles will cease to move upward, either because the convective plume is no longer buoyant at that altitude, or because the buoyancy forces are equalled by gravitational ones. H may then be defined as the height of the mixing layer.

This idea of a mixing layer was illustrated by data collected by an instrumented aircraft operating over the North Sea off the coast of Lincolnshire, during a field project at Donna Nook, Lincolnshire. Profiles of Aitken nuclei, aerosol particles and temperature were collected at about midday on 4 September 1981. Figure 1(a) shows radius/concentration spectra from size range 3 (1.5 ≤ r ≤ 10 μm) of the Axial Scattering Spectrometer Probe (ASSP), obtained during level runs at various altitudes, with the relevant size range from UMIST's ground-based Forward Scattering Spectrometer Probe (FSSP) included, and Fig. 1(b) shows the total concentration in the same size band and the Aitken count plotted against altitude. On this day, a strong anticyclone had given rise to a pronounced capping inversion at about 850 mb (1400 m), with a dry layer above. Considering the two parts of Fig. 1, it is reasonable to conclude that there was a region
below the inversion which was well-mixed (although the concentration fell off slightly as the inversion was approached, especially for the larger particles). Above the inversion the spectra are similar to each other, with particle concentrations some 1–2 orders of magnitude less than below the inversion, and a marked absence of large particles. The Aitken count is also depleted above the inversion, which is consistent with its suspected anthropogenic origins, it being the stubble-burning season.
These facts lead to the assumption that, under equilibrium conditions, all particles from Aitken size to 8 µm radius are reasonably evenly distributed up to the top of the mixing layer. Therefore, it follows that the concentration of aerosol at any height within the mixed layer should be inversely related to the depth of the mixing layer, all other factors remaining constant.

When there is a strong capping inversion, with unstable conditions underneath, $H$ is readily identifiable as the inversion height, as in the above example. However, there is rarely a well-marked capping inversion above the North Atlantic, although such inversions are frequently used to define the height of the mixing layer in the analysis of Pacific Ocean data. There are two mechanisms which will produce turbulent activity, and thus affect the height of the mixing layer: one is convective plume activity and the other is the wind shear near ground level. Some quantification of these two factors is required. In order to avoid the complexities of a full physical model of the marine atmospheric boundary layer, involving turbulent diffusion, gravitational settling, entrainment across the upper boundary layer, turbulent deposition, etc., the characterization of which would require many approximations and assumptions, it was decided to try a simple representation.

A model of diurnal mixing height variation was given by Benkley and Schulman (1979). It consists of a convective regime building up after radiation sunrise, and dying away after radiation sunset. The hours between radiation sunset and radiation sunrise are filled by a nocturnal mixing height estimate, modelled as a nocturnal surface inversion. For daylight hours when there is little convective activity, the nocturnal model was used. Although this diurnal variation of mixing height, driven by radiation, is clearly inapplicable here, it was decided to employ the basic philosophy of this model. This required a convective regime, with mixing heights calculated from radiosonde ascent data, and a separate, competing, mechanical regime, with mixing heights calculated from wind speed data.

Any convective activity will be driven by the difference between sea-surface temperature and air temperature. The sea-surface temperature shows little diurnal variation, and the temperature of the air immediately above the sea surface is more influenced by prevailing air mass than by solar radiation. When the sea is warmer than the air above it, convective activity is likely, and when the air temperature is higher than the sea-surface temperature, stability should result. Examination of sea-surface and air temperatures for the North Atlantic suggests that, during winter months, the situation is almost always unstable, and frequently very unstable, and during the summer months it is usually slightly unstable, with periods of neutral stability. This is illustrated in Figs. 2(a) and (b), for sections of data from summer and winter in 1980.

![Figure 2. Temporal variations in air and sea-surface temperatures. (a) For August 1980.](image-url)
A convective mixing layer height, $H_c$, was defined as that height to which a parcel of air at sea-surface temperature and site humidity could rise under its own buoyancy according to the Stornoway radiosonde ascent. In fact, $H_c$ is likely to exceed any real mixing layer height, as entrainment of environmental air into the convective plume will lessen its buoyancy, but it will serve as a comparative estimate of the amount of convective activity present.

Stornoway radiosonde ascent data were available from the Meteorological Office for 0001 and 1200 GMT on most relevant days, and sea-surface temperature was taken from the isotherms, averaged over five days' data, issued by the Meteorological Office.

The turbulence generated by the wind velocity may be modelled by a diagnostic formula based on similarity theory, relating mixing height directly to the friction velocity, $U_*$ (e.g. Yu 1978; Venkatram 1980). Although very simple, a diagnostic model of the type

$$H_m = C U_* / f$$

where $C =$ dimensionless constant, $f =$ Coriolis parameter and $H_m =$ height of turbulent layer, explained nearly 50% of the variance of the depth of nocturnal (i.e. non-convective) turbulence, when tested against an estimate of the actual depth of turbulence computed from profiles of the Richardson number (Mahrt et al. 1982). This type of model was adopted for the present purposes. The value for $C$ suggested in the literature varies considerably: Benkley and Schulman suggest 0.185, Gathman (1982) uses 0.23, and Mahrt et al. concluded that a value of 0.06 gave the best results.

A mechanical mixing layer height, $H_m$, was defined from Eq. (1), using a roughness length of 2 mm for the sea surface (McIntosh and Thom 1981), and following Benkley and Schulman both in using a value of 0.185 for $C$ and in setting von Kármán’s constant at 0.35, to give

$$H_m \sim 95U(2) \text{ (m)}$$

$$H_m \sim 75U(10) \text{ (m).}$$
The derivation of Eqs. (2) and (3) from Eq. (1) is strictly valid only under conditions of neutral stability. Varying the input value of the constant parameters alters the constant in Eqs. (2) and (3); for example, using a small roughness length of 0.4 mm, and the minimum value of the constant $C$ of 0.06, reduces the value of the constant in Eq. (3) to about 20. Given the other errors in this type of analysis, this was deemed acceptable. Equation (2) or (3) was used according to whether the wind speed data available were 2 m or 10 m data. The wind speed value used was the average over the hour preceding the time for which the mixing layer height was required.

3. **Validation of Technique**

The techniques of calculating $H_c$ and $H_m$ were tested at a remote fell-top field station, Great Dun Fell (GDF), in Cumbria, where an acoustic sounder and an instrumented glider were available to provide measurements to compare with the estimates made (Latham et al. 1983). The instrumented glider provided on-site temperature profiles, and direct observation of cloud and mixing status. The acoustic sounder was oriented vertically, and emitted 200 ms pulses at a frequency of 1678 Hz. The backscattered signal from the sounder is dependent on temperature, wind speed, and, to a lesser extent, humidity. The sounder had a vertical limit of 900 m, and could not be operated at wind speeds above 10 m s$^{-1}$, or during rain, owing to excessive noise on the return signal. Periods of convective activity show up clearly as holes in the tracer signal (Gardiner 1982). Meteorological Office sonde data, similar to Stornoway information, were obtained for the stations nearest GDF. As there are three of these, none very close, the choice of the most suitable ascent was made on the basis of the daily air mass trajectory.

The convective mixing depth was estimated using site dry-bulb temperature, dewpoint, and pressure, this last being estimated as 100 mb less than that recorded at a site in the valley bottom. The mechanical mixing depth was estimated from

\[ H_m = 100U(10) \]

which was obtained from Eq. (1), using a roughness length of 2 cm, calculated for the site in a previous study (Blyth et al. 1980).

For periods when the sounder suggested that there was strong convective activity, mixing depth estimates were plotted against cloud top as reported by the glider (when it was able to fly) and against inversion height as measured by the sounder (when it was within range of the sounder). The sounder data were used only if the inversion itself could be seen (Kaimal et al. 1982). The results are shown in Fig. 3. The estimated heights are usually somewhat lower than the measured ones, with a maximum discrepancy of about 30%.

When the sounder indicated no convective activity, the values of mixing height obtained from the sounder were compared with the estimate made with Eq. (4). This was done on the assumption that what the sounder was 'seeing' was a marked temperature variation between the mixed layer below and the unmixed layer above. The results are shown in Fig. 4.

Although data were limited, it was concluded that these methods of obtaining heights of the mixing layer were acceptable, and should provide a reasonable first estimate.
4. THE DATA

The data from the South Uist experiments were used. These are described more fully in the Exton et al. (1985) paper, but basically consisted of aerosol concentration measurements in the size band $0.1 \leq r \leq 23.5 \, \mu m$ using various Particle Measuring Systems (PMS) aerosol spectrometers at heights of 2 and 10 m above the ground. Wind speed and direction, and dry- and wet-bulb temperatures were also measured and recorded. However, no direct measurements of atmospheric turbulence were made. The structure of the upper air was known only from radiosonde ascents taken at Stornoway, which is situated on the eastern side of the Isle of Lewis, some 40 km wide at that point. Low hills on Lewis ($\sim 300 \, m$) further distort the flow of air from the North Atlantic, and modify the lower part of the atmosphere from the open ocean state.
Details of such problems as site suitability, the non-isotropic, non-isokinetic sampling and multi-valued response of PMS probes, together with questions of data selection and treatment, are dealt with in the Exton et al. paper. In all, some 85 days of data were collected in maritime air from the North Atlantic, during several 2- to 4-week periods, at various times of the year, between 1980 and 1983.

In order to look at the effect of mixing height on particle concentration, the effect of wind speed on production rate had to be removed from the data. A relationship of the form

\[ n = c U^d \]

where \( n = \) particle concentration (cm\(^{-3}\)), \( U = \) wind speed (m s\(^{-1}\)) and \( c, d \) are constants, was assumed for the normalizing process, following Monahan et al. (1983). The wide range of values for the dependence of aerosol concentration on wind speed both within the various South Uist data sets, and in the published data, are given in Exton et al., and the reasons for the discrepancies discussed. The value of \( d \) chosen was 1.5 for particles larger than 0.25 \( \mu \)m radius, and 1.0 for smaller particles, which values were shown to be good averages of all South Uist maritime data.

All aerosol data were then corrected to the same wind speed. The value of 6 m s\(^{-1}\) was chosen as the wind speed value to which all concentrations were corrected, and the wind-speed-corrected aerosol concentration, \( n_{wsc} \), then became

\[ n_{wsc} = n(6/U)^d. \]

The value of 6 m s\(^{-1}\) was originally chosen for use in Eq. (6) as it is below the value at which aerosol particle concentrations appeared to begin to increase rapidly with wind speed in the South Uist data. However, the actual value is unimportant, provided that all concentrations are corrected to the same value of wind speed. The concentrations used were the integrated values for the hour preceding the time of the mixing layer height estimate.

5. RESULTS

The values of \( H_c \) and \( H_m \) obtained were compared; the unstable conditions prevalent at South Uist meant that \( H_c \) usually had the greater value.

Initially, the wind-speed-corrected concentrations were correlated with both \( 1/H_m \) and \( 1/H_c \) individually. However, it is worth noting that there are problems with correlating the wind-speed-corrected concentrations with the wind-speed-driven mixing height, \( H_m \), for prolonged periods during which \( H_m \) is the dominant mixing height. As both the concentration and the mixing height are wind speed dependent, the effect of increasing the mixing height would show up merely as an overall decrease in the wind-speed–particle-concentration relationship, i.e. the empirically derived value of the term \( d \) in Eq. (6) would be reduced by unity, and no mixing height effects would be apparent. It is therefore impossible to use this type of analysis for data periods during which the mechanically driven mixing height frequently exceeds the convectively driven one. This is not the case for the South Uist data set.

Plots of \( n_{wsc} \) versus \( 1/H_m \) were largely scatter plots, showing that there was no systematic relationship between the two variables in the data sets studied, except for the data from February and March 1981, when correlations were persistently negative and, for larger particles, significant. This was thought to be spurious; examination of the air mass trajectories for this period shows fairly extensive recent passages over land, even for air masses classified as maritime. This recent land crossing means that the production
equation (5) no longer holds, more especially for larger particles with their shorter residence times. Concentrations collected at higher wind speeds were being over-corrected, leading to low particle concentrations for high values of \( H_m \). Accordingly, data from February/March 1981 were not used in the further analysis.

Plots of \( n_{\text{wsc}} \) versus \( 1/H \) showed reasonable correlation except for values of \( H \) of <500 m. This was due to two factors; first, as discussed in the section on the data base, these are the heights for which Stornoway data are least appropriate and secondly, it is when the convective mixing layer height is low that the mechanical mixing layer height becomes dominant. Subsequently, \( H \) was taken to be the higher of \( H_c \) or \( H_m \), and a linear regression routine using a simple least-squares fitting procedure was used to obtain a relationship of the form

\[
n_{\text{wsc}} = A + B/H.
\] (7)

Generally, this led to an increase in the correlation coefficients compared with using \( H_c \) alone; for example, for CSASP (see below) range 0, the correlation coefficient improved from 0.61 to 0.77 for December 1980, and from -0.09 to 0.64 for August 1980, when stability was very variable.

All available valid data sets were analysed; in addition data were combined into larger sets where possible. The 1980 data were all collected at 10 m by a Classical Scattering Aerosol Spectrometer Probe (CSASP), which measures particles in the size range 0.25-16 \( \mu \)m radius, and an Active Scattering Aerosol Spectrometer Probe (ASASP), which measures particles in the size range 0.08-1.5 \( \mu \)m radius, and the 1982 and 1983 data were collected at 2 m by an FSSP, which measures particles in the size range 0.25-23.5 \( \mu \)m radius, and an ASASP. It is not reasonable to combine 2 m data with 10 m data, and it proved impossible to combine 1982 and 1983 data as the FSSP was overhauled between projects, and was counting approximately twice as high on one project as the other. This is not uncommon with this type of probe. Although the CSASP and FSSP cover approximately the same size band, they do have somewhat different responses, so that not all of the discrepancy in their results is due to their not being positioned at the same height.

Examples of plots of adjusted concentration versus inverse mixing layer height, are shown in Figs. 5(a)-(c), along with the lines of best fit to the data, corresponding to the relationship in Eq. (7). A full set of the empirical relationships is given in Table 1, together with the significance level of the correlations.

The only large particle data for which the results are not significant are those which were collected in April 1980. Also the relationships are stronger in 10 m than in 2 m data, probably owing to the larger number of other influences on aerosol particle concentration at 2 m. Tides are an important influence, but salutation, pollen release, seaweed emissions and fragments, sand flies and general disturbance by other users of the site environs, may occasionally cause data contamination. For the 1982 data, the correlations for large particles are noticeably lower than for 1983, possibly owing to the stronger tidal perturbations encountered in 1982, as described in detail in Latham et al. (1984).

There is a tendency for the larger particles to have more significant correlations; this is reasonable considering that bubble bursting at the sea surface is the major source of these particles, whereas the smaller ones are also created by gas-to-particle conversion, and can be advected in from remote sources.

The column marked \( B/A \) in Table 1 shows the dependence of particle concentration upon mixing height with the concentrations normalized for background, or infinite mixing height concentration. When these values are compared for the December 1980 and for
1983 data (these are the only two data sets for which all probe ranges are available), it may be noted that, in general, the larger the particle, the stronger the dependency on mixing height. This is initially somewhat surprising (and contrary to the land-based findings of Choularton et al. (1982)) as gravitational sedimentation might be expected to

![Graph](image)

Figure 5. The relationship between mixing height and wind-speed-corrected particle concentration. (a) All 1980 CSASP size range 0 (1 ≤ r ≤ 16 μm) maritime data. (b) All 1982 FSSP size range 2 (0.5 ≤ r ≤ 8 μm) maritime data. (c) All 1983 FSSP size range 2 maritime data.
### Table 1: Mixing Height Relationships of the Form Given in Eq. (7) for Maritime Data

<table>
<thead>
<tr>
<th></th>
<th>ASASP size band 1 (0.22 ≤ r ≤ 0.5 μm)</th>
<th>ASASP size band 2 (0.1275 ≤ r ≤ 0.3025 μm)</th>
<th>ASASP size band 3 (0.08 ≤ r ≤ 0.15 μm)</th>
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<tr>
<td></td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
</tr>
<tr>
<td>Dec 80</td>
<td>22 4.60 20.97 4.56 0.46 5</td>
<td>23 34.91 96.85 2.77 0.33</td>
<td>23 40.25 103.71 2.58 0.36 10</td>
</tr>
<tr>
<td>1982</td>
<td>18 5.22 16.15 5.02 0.45 10</td>
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<tr>
<td>1983</td>
<td>21 6.26 2.79 0.45 0.43 10</td>
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<tr>
<td>CSASP size band 0 (1 ≤ r ≤ 16 μm)</td>
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<td></td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
</tr>
<tr>
<td>Apr 80</td>
<td>18 1.10 0.69 0.63 0.32 1</td>
<td>23 3.75 17.09 4.56 0.74 1</td>
<td></td>
</tr>
<tr>
<td>Aug 80</td>
<td>15 0.08 2.25 28.13 0.64 1</td>
<td></td>
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</tr>
<tr>
<td>Dec 80</td>
<td>23 0.18 2.43 13.58 0.77 1</td>
<td></td>
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<tr>
<td>Cum 80</td>
<td>56 0.61 1.47 2.41 0.54 1</td>
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<td></td>
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<td>FSSP size band 0 (1 ≤ r ≤ 23.5 μm)</td>
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<tr>
<td></td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
</tr>
<tr>
<td>1982</td>
<td>28 0.85 0.71 0.84 0.39 2</td>
<td>28 0.80 0.82 1.03 0.42 2</td>
<td></td>
</tr>
<tr>
<td>1983</td>
<td>24 0.19 0.61 3.21 0.82 0.1</td>
<td>24 0.28 0.56 2.00 0.69 0.1</td>
<td></td>
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<tr>
<td>FSSP size band 2 (0.5 ≤ r ≤ 8 μm)</td>
<td></td>
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<tr>
<td></td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
<td>n A B B/A cc si</td>
</tr>
<tr>
<td>1982</td>
<td>28 2.25 3.32 1.48 0.57 0.2</td>
<td>28 3.12 4.29 1.38 0.56 0.2</td>
<td></td>
</tr>
<tr>
<td>1983</td>
<td>24 0.98 1.85 1.89 0.79 0.1</td>
<td>24 2.07 2.57 1.24 0.56 1</td>
<td></td>
</tr>
</tbody>
</table>
be the dominant force on particles larger than about 10 μm radius, leading to a depletion rather than an accretion of these particles in more stable conditions. However, the channel counts for the smallest sized particles dominate the concentration for all probe size ranges, so that the increasing strength of the relationship between particle concentration and mixing layer height may be valid only for particles up to about 7 μm radius.

6. DISCUSSION

It is not difficult to understand the discrepancy between the model of Goroch et al. (1980) and the present results. If the type of model suggested by Yu (1978) is correct (Eqs. (2) and (3)), then, even with the low values of the input parameters, only very light wind speeds are necessary to produce mixing depths in excess of the height of the probe position, so that the concentrations are always measured within the mixing layer. Within this layer, depletion rather than enhancement is likely with increasing instability, as found at South Uist. Later models of aerosol concentrations and altitude profiles tend to incorporate a term of this type (e.g. Gathman 1982), except when the atmosphere is extremely stable.

The results found by Davidson and Schütz (1983) are more difficult to reconcile with the present work, especially as the data were obtained in a similar environment to the studies reported here. The data set which they used for analysis is comparatively short and may be overly influenced by other factors, e.g. frontal systems. Their analysis is also dependent on several algorithms formulated by other workers, which may not be appropriate for their experimental conditions. In particular the algorithm for stability, characterized by z/L (where z is the measurement height and L is the Monin-Obukhov parameter) may not be applicable at wind speeds greater than 10 m s⁻¹.

It must also be considered whether equilibrium conditions are likely to be reached in the time available. As the major changes in mixing layer height over the North Atlantic are caused by the arrival of a different air mass, conditions may have as long as several days to come into equilibrium, which must be adequate.

When examining the variation of aerosol concentration with mixing layer height, another point needs to be considered. Wu (1979b) proposed that the extent of white-capping, and hence the amount of aerosol produced, is a function of atmospheric stability. His empirical formulae suggest that the number of particles emitted by the sea should be about twice as high in stable (dT < -0.4 degC) as in unstable (dT > +0.6 degC) conditions (dT is the difference between the sea-surface and the air temperatures). However, as illustrated in Fig. 2, conditions at South Uist are almost always unstable, so that this factor is not directly applicable here. Monahan and O'Muircheartaigh (1986), however, state that, for a given value of the wind speed, whitecapping increases as dT becomes more positive, i.e. as the atmosphere becomes increasingly unstable. This would operate to increase particle concentration as the mixing height increased, and, if present in the South Uist data, is clearly being swamped by the dilution effect. Attempts to correlate wind-speed-corrected particle concentrations with dT rather than H resulted in the same sort of inverse relationships, but with far lower correlation coefficients in every case. While increasing instability may well result in a lower particle production rate as well as an increased particle concentration dilution effect, the latter is thought to be the more important factor in these circumstances.

There are, of course, many over-simplifications and errors built into this technique of estimating H; the use of Stornoway ascents, and the simple derivation of Hₙ are obvious examples. Another involves the use of site humidity for obtaining the value of
Figure 6. Wind-speed-corrected particle concentrations for FSSP size range 1 (1 ≤ r ≤ 16 μm) for all 1983 maritime data. (a) Using site air temperature instead of sea-surface temperature. The correlation coefficient is −0.15. (b) Using the dry ascent only from sea-surface temperature. The correlation coefficient is −0.15. (c) Using the height of the lowest significant inversion. The correlation coefficient is 0.2.
$H_e$. The relative humidity immediately above the sea surface is about 98%, and then falls dramatically in the first few metres. Eriksson (1959) suggested a sharp fall across the first millimetre, a less rapid fall across the next metre, and a gradual fall up to about 8 m, where the environmental level is reached. The humidity devices employed in these experiments were positioned about 3 m above the high water mark, so it is likely that site humidity is an under-estimate of the humidity in the lowest metre of the boundary layer. The humidity on site varied from 60% to 100%, with the great majority of the data collected at humidities close to the mean value of 85%. Also, the derivation of Eqs. (2) and (3) from Eq. (1) is strictly valid only under conditions of neutral stability, but this is not a major source of error in the South Uist data analysis, as conditions were usually close to neutral on the few occasions when the mechanical mixing height became dominant.

Not all the scatter around the straight-line relationship is due to errors in the method, however; there are other factors affecting particle concentration which are not dealt with here, e.g. detailed air mass history, changes in humidity, precipitation, tide movements (especially for the 1982, and to a lesser extent, the 1983 data), and aerosol loss or modification within cumulus clouds.

Other methods of characterizing $H$ were attempted, including: using site temperature instead of sea-surface temperature; using only a dry ascent from sea-surface temperature (i.e. ignoring buoyancy due to latent heat of condensation); and using as $H$ the height of the lowest significant inversion (where one could be detected). Results of these alternative analyses are given in Figs. 6(a) to (c). As can be seen, poorer correlations and weaker relationships resulted in every case.

In the absence of a well-defined capping inversion, as is often the case over the North Atlantic, it is necessary to find an alternative method of characterizing the mixing layer height. The method presented here, using only sea-surface temperature, temperature and humidity profiles, and wind speed, seems to give a workable estimate of the effect of atmospheric stability on marine aerosol concentrations within the mixed layer.

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