A study of aerosol properties and solar radiation during a straw-burning episode using aircraft and satellite measurements

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SUMMARY

Aircraft measurements of radiative fluxes over the North Sea during a heavy straw-burning pollution episode are presented and compared with results from a radiative transfer model.

The model uses aerosol properties derived from aircraft and satellite measurements, and acceptable agreement with the measured fluxes is obtained.

High levels of aerosol concentration are described, with correspondingly large radiative effects. Boundary layer heating rates of 3 K day$^{-1}$ are measured, and rates of 5–10 K day$^{-1}$ in regions of higher aerosol loading are inferred.

1. INTRODUCTION

In recent years there has been considerable interest in the effect of aerosol on solar radiation in the cloud-free atmosphere, much of it directed at influences on global and regional climate. Concern over possible global warming or cooling by anthropogenic aerosols has been heightened, especially by current work on the aftermath of a nuclear exchange as well as the effects of widespread deforestation. Study of the behaviour of radiation and aerosol tends to have been channelled into three areas, with limited interaction; measurement and collation of aerosol properties, modelling of the atmospheric radiation field, and measurement of the radiation field.

A large amount of information is available on the physical and optical properties of aerosol of different types and localities (Carlson and Benjamin 1980; Harris and Gerber 1982; Patterson et al. 1986; Kilsby and Smith 1987). However, owing to the extreme variability and range of concentration and optical properties of atmospheric aerosol, it is very difficult to model the radiation field reliably from climatological or standard aerosol properties.

Modelling of local and global climatic effects has been carried out using measured and standard aerosol properties (Braislaw and Dave 1973; Wendling et al. 1985; Blanchet et al. 1986). Much uncertainty remains however, and a range of aerosol single scattering albedos exists at which under certain conditions, a line dividing global cooling and warming may be drawn (Charlock and Sellers 1980).

The ultimate test of such modelling is comparison with measurements of the real radiation field; unfortunately these are difficult measurements to make with sufficient accuracy. Radiation profiles allowing flux divergences to be inferred have been measured (Roach 1961; De Luisi et al. 1976; Method and Carlson 1982; McArthur 1984; Kitchen and Squires 1984), but seldom with sufficient accuracy or detail to stand comparison with profiles modelled using simultaneous aerosol measurements.

This work presents results of an experimental and theoretical study of a highly polluted atmosphere. Aircraft and satellite measurements of properties of aerosol resulting from straw-burning are combined in a radiative transfer model to be compared with measured profiles of downward, downward diffuse and upward radiative fluxes. Comparisons are made for the total solar band (0.3–3 $\mu$m), and the visible (0.3–0.7 $\mu$m) and near infrared (0.7–3 $\mu$m) parts of the solar band and good agreement is obtained
throughout. It is found that although the aerosol has its origin in combustion, it is only a moderate absorber, in contrast with the very strong absorption assumed in some "nuclear winter" scenarios. These scenarios assume the production of absorbing aerosol from combustion of material in urban environments rather than combustion of vegetation. Combustion of vegetation undoubtedly forms a substantial part of any nuclear winter scenario, and additionally occurs with increasing frequency in slash and burn agriculture and deforestation world wide. In this paper, a severe pollution event is described, and boundary layer heating rates of 5–10 K day$^{-1}$ are calculated.

The comparison of satellite-derived aerosol optical depths with aircraft measurements is particularly important, since validation of the satellite technique is essential before it can be used to provide much needed global and temporal information on aerosol. This information can be used to correct satellite-measured surface parameters (e.g. vegetation indices and albedo) as well as being directly included in climate models.

2. THE EXPERIMENT

(a) Conditions

On 11 and 12 September 1985 the centre of an anticyclone was situated over Denmark and produced clear skies over the North Sea (Fig. 1). Following a fairly wet summer, a dry spell in early September gave farmers a good opportunity to burn straw and stubble. Straw-burning fires were widespread over the cereal-growing areas of England, and imagery from NOAA-9 Advanced Very High Resolution Radiometer (AVHRR) channel 3 (section 2(c)) shows some 750 separate fires (~1 km$^2$ resolution) each consisting of a blazing field (Fig. 2).

The south to south-westerly surface flow carried aerosol from the fires over the North Sea, trapped under two temperature inversions at 920 mb and 1000 mb (Fig. 3).

Figure 1. Surface synoptic chart for 1200 GMT on 12 September 1985. The aircraft measurement runs are shown around 56°N, 0°W. The location of Hemsby upper air station is marked by a cross.
Aerosol concentrations were particularly high near to the east coast of England, with large amounts of pollution also in the eastern North Sea, probably originating in the industrial regions of central Europe.

Since the current day's smoke plumes appear to have reached only some 70 km distance from the coast (section 3(b)) the aerosol in the operating area (Fig. 1) on 12 September must be assumed to be the product of earlier burning on 11 September. Comparison of the aircraft profiles with radiosonde ascents from Hemsby at 0000 GMT and 1200 GMT shows that the lowest inversion (at 1000 mb) is consistent with that produced by overnight cooling, sustained by the cool (~13°C) sea surface during its transport to the operating area. This process resulted in very high concentrations of smoke in the lowest layer, whereas over land and nearer the coast the aerosol was mixed throughout a much deeper boundary layer (and so was more dilute).

(b) Aircraft flight pattern and instrumentation

On 12 September the Hercules of the Meteorological Research Flight (MRF) performed a sortie around 56°N, 0°W (between 1050 and 1445 GMT)—the measurement runs of the flight pattern are shown in Fig. 1. In addition a series of air sampling runs were performed off the north east coast of England on 11 September. The Hercules is
equipped with meteorological and navigational equipment and other specialised instrumentation (Readings 1985), of which on this occasion the radiation and aerosol systems were used. The flight pattern consisted of a series of straight and level runs of 20km length. Runs were performed at 12 levels between 60 and 8000m with interconnecting profiles for measuring temperature, dewpoint and aerosol properties.

Solar radiation measurements were made with four Eppley PSP pyranometers arranged in pairs facing upwards and downwards. One of each pair was fitted with a clear glass dome (WG295 transmitting between 0.3 and 3 μm) and the other with a red dome (RG715 transmitting between 0.7 and 3 μm). This arrangement allows measurements to be made in three spectral bands: solar, near infrared and (by subtraction) visible. Total upward and downward fluxes were measured as well as the diffuse component of the downward flux. The measurement procedure is described in section 4.

The aerosol scattering coefficient at 0.48 μm was measured continuously using a MRI model 1550 integrating nephelometer. This was calibrated before flight using Freon 12, Freon 22 and filtered air according to the procedure proposed by Ruby and Waggoner (1981). Number concentration and size spectra in the radius range 0.25–3.75 μm were measured using a PMS Forward Scattering Spectrometer Probe (FSSP). Aerosol samples were taken on 0.4 μm pore size Nuclepore membrane filters over 20-minute periods. These were analysed firstly for mass loading (± 20 μg) and secondly for absorption coefficient at 8 wavelengths between 0.5 and 1 μm using the Integrating Sandwich technique (Clarke 1982; Foot and Kilsby 1989).
(c) Satellite data

NOAA-9 (AVHRR) data from a pass overhead the United Kingdom at 1346 GMT on 12 September 1988 have been used for two purposes. Firstly the imagery from the near infrared channel 3 (3.7 μm) shows the distribution and extent of straw-burning over England (Fig. 2). Channel 3 is very sensitive to the presence of small hot spots in the field of view due to the non-linearity of the Planck function (Saunders 1986) and has been proposed as a means of monitoring straw-burning (Muirhead and Cracknell 1985), and used to monitor tropical deforestation in South America (Malingreau et al. 1989).

Radiances from the visible channel 1 (0.63 μm) and infrared channel 2 (0.86 μm) are also used quantitatively to derive aerosol optical depths. The technique is described in detail in section 3(b).

3. AEROSOL PROPERTIES

(a) Aircraft measurements

The vertical distribution of aerosol is best shown by the total number concentration (0.25 < r < 3.75 μm) measured with the FSSP (Fig. 4). This shows some aerosol situated below a temperature inversion at 900 m, but the majority is concentrated below a lower inversion at 90 m. No significant amounts of aerosol were observed above 900 m.

![Figure 4. Profile of aerosol concentration (0.25 < r < 3.75 μm) measured with FSSP.](image)

The vertical distribution of aerosol scattering coefficient, measured with the integrating nephelometer during aircraft profiles and level runs, is shown in Fig. 5. This corresponds very well with the FSSP measurements, but there is some discrepancy between the profile measurements, taken during a 2.5 m s\(^{-1}\) descent, and the level runs which were performed in an ascending staircase. This could be due to time lags in the instrument sampling and in flushing the sample volume of the instrument, especially after filling with the very polluted air of the lowest run of the ascent. The best estimate of scattering coefficient is considered to be between these measurements, although an underestimate may occur near the surface in the very high concentrations.

The size distribution by volume measured by the FSSP averaged over 12 minutes in the dense haze is shown in Fig. 6, and displays a dip at 0.9 μm radius characteristic of
Figure 5. Profile of scattering coefficient at 0.48 μm measured with the integrating nephelometer. The continuous line is a descending profile, the circles (with error bars) are values from runs in an ascending 'staircase'.

Figure 6. Size spectrum of particle volume concentration per unit radius derived from 12 minutes of FSSP measurements below 180 m compared with the volume spectra for the three SRA aerosol models. The model concentrations have been scaled to give an extinction coefficient of $3.7 \times 10^{-4} \text{m}^{-1}$ at 0.55 μm to match the measured value.
previous FSSP measurements in a variety of conditions (Kilsby and Smith 1987). This dip is believed to be instrumental in origin. The instrument is incapable of detecting the very small \((r < 0.2 \mu m)\) smoke particles expected from straw-burning, and high concentrations of large particles suggest that at a distance of 200–300 km from the source, some coagulation may have occurred.

A number of filter samples were collected on the 2 days of the experiment; three off the north east coast on 11 September at 60 and 300 m, and one on 12 September between 60 and 300 m over a 25-minute period. The results of gravimetric and optical absorption analysis of these filters are given in Table 1. A spectral plot of absorption coefficient \((\beta_{abs})\) is given in Fig. 7 for the 12 September filter. This displays the inverse wavelength dependence characteristic of a grey absorber such as elemental carbon.

Combination with mass loading measurements allows the specific absorption and effective imaginary refractive index \((n_2)\) to be calculated (Gerber 1983). The specific absorption \((B_a)\) of \(\sim 2.2 \text{ m}^2\text{g}^{-1}\) is very high for normal atmospheric aerosols but is in line with accepted values for products of flaming combustion. Values in the literature range from 0.7 to 2.4 \text{ m}^2\text{g}^{-1}\) for burning pine needles (Patterson et al. 1986), through 2.0 \text{ m}^2\text{g}^{-1}\) for assumed nuclear winter smoke (NRC 1985) to 10–12 \text{ m}^2\text{g}^{-1}\) for carbon soots.

The single-scattering albedos \((\omega_0)\) at 0.55 \(\mu m\) have been calculated using the measured absorption coefficient and the scattering coefficient, measured at 0.48 \(\mu m\) scaled

<table>
<thead>
<tr>
<th>Date</th>
<th>Height (m)</th>
<th>Mass loading ((\mu g\cdot m^{-2}))</th>
<th>Absorption coeff. ((10^{-4}\text{m}^{-1}))</th>
<th>Specific absorption ((\text{m}^2\text{g}^{-1}))</th>
<th>(n_2)</th>
<th>(\omega_0)</th>
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<td>11 Sept 85</td>
<td>60</td>
<td>54</td>
<td>0.17 ± 0.03</td>
<td>3.2</td>
<td>0.018</td>
<td>0.87</td>
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<tr>
<td></td>
<td>300</td>
<td>85</td>
<td>0.19 ± 0.03</td>
<td>2.2</td>
<td>0.013</td>
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<tr>
<td></td>
<td>300</td>
<td>84</td>
<td>0.2 ± 0.03</td>
<td>2.4</td>
<td>0.014</td>
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<tr>
<td>12 Sept 85</td>
<td>60–300</td>
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<td>0.39 ± 0.06</td>
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<td>0.92</td>
</tr>
<tr>
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<td>±15%</td>
<td>±0.8</td>
<td>±0.005</td>
<td>±0.02</td>
</tr>
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</table>

Figure 7. Absorption coefficient measured on 12 September with the Integrating Sandwich, at heights between 60 and 300 m. Uncertainty in absorption coefficient is ±15%.
to 0.55 μm. The scaling factor of 1.16 was calculated from the wavelength dependence of the Standard Radiation Atmosphere (SRA) continental aerosol model (WMO 1986). The value of 0.92(±0.02) for 12 September is higher than those for 11 September but if a scattering coefficient for the profile is used rather than one for the level runs, a value of 0.90 is obtained. The relation between $n_2$ and $\omega_0$ is easily tested if assumptions of size distribution and $n_1$ (real part of the refractive index) are made. This is best illustrated by Fig. 8, in which the solid line is a mean relationship between $n_2$ and $\omega_0$ calculated with Mie theory for a large number of observed size distributions with a refractive index, $n_1$, of 1.5 (Gerber 1983). The line is a good approximation to a number of measurements, and the straw-burning values are not exceptional. This comparison gives confidence in the three separate constituent measurements; absorption coefficient, mass loading and scattering coefficient.

Estimates of the total aerosol extinction optical depth on 12 September at 0.48 μm may be made using the nephelometer data shown in Fig. 5. The best estimate is 0.25 ± 0.04 assuming a single scattering albedo of 0.92, but this technique assumes horizontal homogeneity of aerosol concentration. A comparison with other estimates using different methods may be made using Fig. 10, and this vertical integration of the scattering coefficient appears somewhat low.
(b) Satellite measurements

The derivation of aerosol optical depth, and its spatial distribution over the ocean from satellites is a well established technique (Griggs 1984; Durkee et al. 1986; Masuda et al. 1988), though not without difficulty. In principle any visible or near infrared satellite channel may be utilised, and AVHRR channels 1 and 2 have been used here. AVHRR data are readily available and these channels give a useful compromise in spectral and spatial resolution, as well as being situated at nearly ideal wavelengths (0.58–0.68 and 0.72–1.1 μm) for accuracy of retrieval.

The scheme used in this work was operated as part of the APOLLO processing software (Saunders and Pescod 1988) with cloud screening performed before retrieval, and is described in more detail elsewhere (Kilsby 1987). It is based on the principle of separating the measured radiances into three components.

\[
L_S = L_A + L_R + L_W
\]  

(1)

where \(L_S\) is the satellite measured radiance; \(L_A\) is the aerosol backscattered radiance; \(L_R\) is the Rayleigh scattered radiance; \(L_W\) is the radiance reflected from the ocean; \(L_R\) is calculated using the Rayleigh scattering phase function and optical depths averaged over the channel responses. \(L_W\) is calculated in two parts. A specularly reflected diffuse (sky) radiance component, is estimated using model results (Plass et al. 1976). The other component is the diffusely reflected "upwelling" radiance which depends on water quality and is taken as zero for channel 2. Measured reflectances from Morel (1980) have been used. \(L_W\) is small for clean water, but can match the aerosol component for very polluted or shallow coastal waters (Prangsma and Roozekrans 1989).

These components are all derived taking account of absorption due to ozone (channel 1) and water vapour (channel 2) calculated using LOWTRAN 5. It is assumed that the aerosol back-scattering takes place below the Rayleigh scattering and gaseous absorption—this is a good approximation for this case as most of the aerosol was in a shallow layer below 150 m. The aerosol radiance \(L_A\) is then considered to have an approximately linear dependence on optical depth, \(\delta\), as follows (Durkee et al. 1986; Liou 1980).

\[
L_A = \frac{F\omega_0 P(\theta) \delta}{4 \cos \phi}
\]  

(2)

where \(F\) is the effective solar irradiance; \(\omega_0\) is the single scattering albedo; \(\theta\) is the scattering angle; \(P(\theta)\) is the aerosol scattering phase function; \(\phi\) is the satellite zenith angle.

For optical depths over 1, this approximation may lead to an overestimate of up to 20%.

The two largest uncertainties of the scheme arise here. Firstly, since insufficient size distribution data (normally, no data) are available to calculate the phase function, a model must be used. The SRA continental model (WMO 1986) has been used here (and throughout the rest of this work) because amongst the commonly used models it most closely approximates the observed optical and physical properties of the straw-burning aerosol. For areas close to the source, a much smaller (mass mean radius <0.2 μm) smoke aerosol would be more appropriate. An uncertainty of ±25% was estimated by consideration of sensitivity to variation in the phase function. A range of plausible functions was used and the estimate covers the resulting retrievals for this geometry. The asymmetry parameter \((g)\) of the continental model is 0.633 at both AVHRR wavelengths.

Secondly the incoming effective solar irradiance depends on the calibration and
Figure 9. Optical depth map for Channel 1 at 0.63 μm on 12 September at 1346 GMT. Contours are labelled in units of optical depth multiplied by 10. The area in which the low level measurements were performed is denoted by a box at the top of the diagram.

Figure 10. Aerosol extinction optical depth plotted against wavelength, measured with three techniques. The vertical error bars are estimates of experimental uncertainty, the horizontal bars define the wavelength band for each measurement. The SRA continental model is plotted, scaled to an optical depth 0.4 at 0.55 μm.
response of the satellite channel. The values used in this work are those of Rao (1987). These two effects combine to produce an error of $\pm 25\%$ in the optical depths.

The scheme was run for the afternoon pass (1344 GMT) of NOAA-9. A contour map of the distribution of extinction optical depth at 0.63 $\mu$m for the western North Sea on 12 September has been produced from the scheme's false colour output imagery (Fig. 9). It can be seen that the highest optical depths ($> 1$) are close to the coast, downwind of the densest areas of straw-burning (Fig. 2) and this was verified observationally during the return flight. Optical depths further downwind in the experimental area are somewhat less and are shown in Fig. 10 for comparison with estimates derived by other means. This decrease in optical depth may be due to either a combination of dispersion and deposition or simple variation with time of burning during the previous day.

4. Radiative Fluxes

(a) Measurement

The total downward, diffuse downward and diffuse upward solar fluxes were measured directly in 2 wavelength bands (total solar and near infrared) as outlined in section 2(b). They were measured at 12 levels between 60 and 8000 m; no cloud affected the measurements throughout the experimental period.

The total downward fluxes were measured flying towards the sun. They have been corrected for instrument orientation, using the aircraft Internal Navigation System (INS) and measurements of the thermopile orientation relative to the aircraft frame of reference (Cluley 1978). Temperature and cosine error corrections were applied. The latter is based on laboratory measurements made at MRF (Foot et al. 1986). Finally the measurements were reduced to a single equivalent zenith angle of 51.5° (noon) using a modified cosine dependence derived from model simulations (section 4(b)). This enables inter-comparison of data both internally and externally with modelled fluxes (section 4(e)).

The diffuse downward fluxes were measured flying away from the sun, obscuring the solar disc from the instrument with a fixed pillar. A geometrical correction scheme using a realistic scattering phase function (Foot et al. 1985) was applied, producing positive corrections of up to 20%.

The diffuse upward fluxes were measured on both up- and down-sun legs, and a small correction for solar zenith angle was calculated using a dependence derived from model runs at a number of zenith angles.

The experimental error in measured solar fluxes is estimated to be about $\pm 5$ W m$^{-2}$.

(b) Modelling

The solar visible and near infrared fluxes were simulated using a delta-Eddington multiple scattering solar radiation scheme (Slingo and Schrecker 1982) with 24 spectral bands between 0.25 and 4 $\mu$m. The model atmosphere was specified by temperature, humidity and ozone density at 50 levels, based on aircraft measurements up to 8000 m and the AFGL mid latitude summer model above. The model was run at a solar zenith angle of 51.5° for comparison with the measured fluxes. A sea surface albedo of 0.09 below 0.7 $\mu$m and 0.045 above was specified, derived from the aircraft pyranometer measurements at the lowest level.

Aerosol was included in two layers, specified by extinction coefficient, single scattering albedo and asymmetry parameter for each of the 24 spectral bands.

Because the measured size distribution data from the FSSP extend only to 0.25 $\mu$m, and are inadequate for derivation of phase function and spectral properties, a fully
specified aerosol model is required. Three standard models are widely used and provide benchmarks for further comparisons—the Standard Radiation Atmosphere (SRA) continental, maritime and urban models (WMO 1986).

The SRA continental model was chosen as the most appropriate for several reasons. Firstly the single scattering albedo in the visible is closest to that observed—the maritime and urban models are respectively much less and much more absorbing.

Secondly, the size distribution is closest to that observed. A comparison of size spectra (Fig. 6) shows that the urban model lacks a large-particle mode whilst the maritime model has a preponderance of large particles. The continental model is similar to that of the FSSP, allowing for the tail-off in FSSP measurements above 2 μm radius due to the effect of short sampling time on counting statistics for the largest particles.

Thirdly the integrated volume concentration of the continental model is closest to that observed when scaled to give the same extinction coefficient. Assuming a density of 1 g cm⁻³, the filter sample equivalent concentration of 178 ± 20 μm³ cm⁻³ compares with 235, 400 and 75 for the continental, maritime and urban models respectively.

The wavelength dependence of extinction coefficient, single scattering albedo and asymmetry parameter of this model are shown in Fig. 11. The model was scaled to

![Graphs showing optical properties of the aerosol model. (a) Extinction coefficient, βₑₑₑ. (b) Single scattering albedo, ωₑ. (c) Asymmetry parameter, g.](image-url)

Figure 11. Optical properties of the aerosol model. (a) Extinction coefficient, βₑₑₑ. (b) Single scattering albedo, ωₑ. (c) Asymmetry parameter, g.
give an extinction coefficient at 0.55 μm of $10.0 \times 10^{-4} \text{m}^{-1}$ between 0 and 180 m, and $3.0 \times 10^{-4} \text{m}^{-1}$ between 180 and 890 m to represent the aerosol profile (Figs. 4 and 5) scaled to give an aerosol optical depth of 0-4, in line with the measurements (Fig. 10).

(c) Comparison and discussion of flux profiles

The model results and aircraft measurements are presented in Figs. 12-14. The total downward fluxes are the most fundamental quantity, and are difficult to measure, requiring a range of corrections for instrument attitude, as well as being vulnerable to high cloud contamination. Figure 12 shows a reduction of some 32% of the solar extraterrestrial flux at sea level. When split into visible and near infrared bands the different absorption and scattering mechanisms become apparent. The visible flux undergoes continuous reduction due to Rayleigh scattering throughout the atmosphere, and a sharp reduction due to aerosol scattering and absorption below 920 mb. The near-infrared flux is reduced mainly by water vapour absorption. All three wavelength bands are in good agreement throughout, and internal consistency justifies the separate correction procedures.

The diffuse downward fluxes are a sensitive measure of aerosol effects and the model representation. Figure 13 shows comparisons between modelled and measured fluxes for all three bands. There is a large Rayleigh component present in the visible band and a very large aerosol component at low level—in fact the visible direct-diffuse ratio at the lowest measurement level is only $\sim 1.5$. The absence of the near-infrared diffuse component at high level confirms the absence of any thin cirrus, which is otherwise difficult to eliminate from the results.

The measured diffuse fluxes appear to be 10–20 W m$^{-2}$ higher than the modelled values in the total and visible bands. Since this discrepancy exists at all levels, it can only partly be due to inadequacies in the aerosol model (for instance, low value of $g$). Most of the discrepancy is due to the different definitions of diffuse flux used in deriving the values. The measurements have been corrected so that they represent the total hemispheric downward flux scattered out of the direct solar beam. The modelled fluxes represent the fraction of the total downward flux considered to be outside the ‘direct’ flux or delta function implicit in the delta-Eddington approximation. This ‘direct’ flux includes scattered radiation travelling in very nearly the same direction as the incident beam. Therefore, the modelled fluxes will always be an underestimate of the true diffuse flux, particularly for aerosols with highly asymmetric phase functions. A discussion of the delta-Eddington approximation and its associated errors is given in Joseph et al. (1976), and comparison with other methods in King and Harshvardhan (1986).

The upward fluxes (Fig. 14) show good agreement and comprise a combination of sea surface reflectance, Rayleigh and aerosol backscatter.

(d) Aerosol optical depth

Extinction optical depths may be calculated for each waveband from the reduction in the direct beam through the boundary layer. The model was used to calculate the extinction optical depth without aerosol; this ‘clean atmosphere’ value was then subtracted from the calculated optical depths to derive aerosol optical depth. These aerosol optical depths have been plotted against wavelength (Fig. 10) for comparison with estimates from satellite and nephelometer measurements. Agreement is reasonable given the differences in time and position of aircraft and satellite data ($\pm 2$ hours and 200 km), and horizontal inhomogeneities. The nephelometer-derived optical depth is low, compared with that expected from extrapolation of the wavelength dependence illustrated by the other methods. An explanation may be that the calibration and previous experi-
Figure 12. Total downward fluxes. Points are aircraft measurements, continuous lines are model values. Uncertainties in measurements are ±5 W m⁻². (a) Solar (Total 0.3–3 μm); (b) Visible (0.3–0.7 μm); (c) Near infrared (0.7–3 μm).
Figure 13. Diffuse downward fluxes. Uncertainties in measurements are about 15%. (a) Solar (0.3-3μm); (b) Visible (0.3-0.7μm); (c) Near infrared (0.7-3μm).
ments were conducted at much lower scattering coefficients than those encountered here, and there may be inadequate calibration or sampling losses in these conditions. The nephelometer measurements were made in level runs at one height and spot in between profiles, and so can only be representative of relatively small areas. The satellite retrievals and pyranometer measurements are representative of much larger areas.

(e) Heating rates and flux divergence

The effect of aerosol in the atmosphere can be expressed in terms of solar heating rates calculated from flux divergences in the boundary layer. Table 2 illustrates the aerosol effects quantified in terms of heating rates split into visible and near infrared wavebands for a layer between 1007 and 841 mb (80 and 890 m) which contains most of the aerosol.

The measured total heating rate of 3·1 K day$^{-1}$ is seen to be split 3:2 between the

| TABLE 2. Boundary layer heating rates (K day$^{-1}$) for boundary layer from 80 m–890 m |
|---------------------------------|-----------------|-----------------|-----------------|
|                               | Solar           | Visible         | Near infra-red  |
| Measured total heating rates   | (a) 3·1         | 1·2             | 1·8             |
| Modelled total heating rates   |                 |                 |                 |
| SRA continental model          | (b) 2·7         | 1·3             | 1·5             |
| No aerosol                     | (c) 0·8         | 0·05            | 0·7             |
| $\omega_0 = 0·8$               | 3·5             | 1·9             | 1·6             |
| $\omega_0 = 0·7$               | 4·7             | 2·8             | 1·9             |
| Aerosol heating rate           |                 |                 |                 |
| Measured (a)–(c)               | 2·3             | 1·2             | 1·0             |
| Modelled (b)–(c)               | 2·0             | 1·2             | 0·8             |
near-infrared and visible bands, the extra absorption in the near infrared being due to water vapour effects. The modelled total heating rate is a little less, but the split between wavebands shows extra absorption in the near infrared.

In addition to the case with aerosol specified as measured, the model was run for cases with no aerosol, and with more absorbing aerosols (the continental model but with $\omega_0 = 0.8$ and 0.7). The case with no aerosol reveals that 0.73 K day$^{-1}$ of the total 3.1 K day$^{-1}$ heating rate is due to water vapour absorption in the near infrared. This allows inferred aerosol heating rates to be derived by subtracting the 'no aerosol' case from the total case. We can then see that more absorption from aerosol occurs in the visible band than in the near infrared.

The cases with more absorbing aerosol give a total solar heating rate of 3.5 and 4.7 K day$^{-1}$ for $\omega_0 = 0.8$ and 0.7 respectively. These extreme heating rates would only be possible if a combustion source were producing large amounts of elemental carbon (soot), but they do illustrate the dependence of heating rate on $\omega_0$.

The distribution of heating throughout the atmosphere is shown in Fig. 15 for the modelled solar band. At high level appreciable gaseous absorption occurs, with aerosol heating present only below 920 mb. The deduced boundary layer heating rate is superimposed for comparison, and it can be seen that this is exceeded by a factor of 3 for the layer of dense aerosol below the inversion at 1000 mb, which may lead to erosion of the inversion and destabilisation.

5. Conclusion

A comprehensive set of measurements and model results from a highly polluted atmosphere have been presented. The case represents one of the most severe aerosol
situations likely to be found in mid-latitudes, and as such affords an ideal opportunity for an assessment of aerosol effects on radiative transfer and a trial of measurement and modelling techniques.

Large aerosol heating rates of 2–3 K day\(^{-1}\) have been found in the boundary layer, and the aerosol has been shown to be moderately absorbing, with a single scattering albedo, \(\omega_0\), of around 0.90–0.92. Comparisons may be made with nuclear winter scenarios, where early estimates of \(\omega_0\) were considerably lower, implying large amounts of elemental carbon in the aerosol although these scenarios include a large proportion of combustion in urban environments. Recent estimates however have used measurements of aerosols derived from burning vegetation which are in line with this work. It would seem that instances of \(\omega_0 < 0.88\) are rather rare in nature and require an urban or anthropogenic source.

The use of the Integrating Sandwich technique for determining aerosol absorption parameters \((\beta_{\text{abs}}, n_2, \omega_0, B_0)\) is fairly reliable. However this technique and the integrating nephelometer must be used with care if large relative humidities are encountered since neither are true \textit{in situ} techniques. An essential addition to the aerosol instrumentation is a particle sizing probe capable of measuring in the 0.05–0.5 \(\mu\)m radius range, permitting the determination of phase function and wavelength dependence of extinction without resort to an aerosol model.

The use of the SRA continental model was fairly successful in two areas—the satellite retrieval of optical depth and the radiative transfer model. In the absence of sufficient size data, a model must be used and standardisation in this area is long overdue.

The radiative flux measurements are the central aim and result of this work, and the consistency of these measurements and their successful modelling are a satisfactory consequence of the considerable effort involved in the measurements and subsequent corrections. The use of a wavelength split at 0.7 \(\mu\)m has enabled the separation of aerosol and water vapour effects, whilst the measurement of the downward diffuse flux has allowed greater certainty in attributing extinction to aerosol and defining its altitudinal variation.

The technique of deriving optical depths using AVHRR, although limited to clear skies and ocean areas, is extremely useful for inferring radiative effects in the absence of other measurements. In this study, it may be assumed that some very large heating rates would have occurred near the coast where aerosol optical depths exceeded unity.

It is planned to perform similar studies in a variety of aerosol conditions using improved instrumentation and techniques (particularly a better aerosol sizing probe) to allow more accurate description, modelling and understanding of the atmospheric radiation field.

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