Quart. J. R. Met. Soc. (1972), **98**, pp. 778-797

551.510.42 : 551.521.31

Aerosol and solar radiation in Britain

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(Manuscript received 14 February 1972; in revised form 12 June 1972)

SUMMARY

The irradiance of the solar beam was measured on cloudless days at Sutton Bonington in the English Midlands and at sites in north-west Scotland. Total and diffuse fluxes were also measured on some days. An attenuation coefficient for aerosol τ_a was defined by

$$S(\tau_a) = S(o) \exp (- \tau_a m)$$

relating the measured flux at normal incidence $S(\tau_a)$ to the flux calculated for a dust-free atmosphere when the air mass number is m. Changes of τ_a from day to day were related to changes of air mass origin; local sources of aerosol were relatively unimportant. In maritime air, τ_a ranged from 0·05 to 0·15, and in continental air, from 0·1 to 0·5. In a tropical maritime air mass, τ_a decreased from 0·13 at sea level to 0·07 at 1,340 m.

The fraction of (ultra-violet + visible) to total radiation was $(0{\cdot}54 - 0{\cdot}28\ \tau_a)$ and the ratio of diffuse to total radiation ($m < 2$) was $(0{\cdot}1 + 0{\cdot}7\ \tau_a)$. The ratio of total scattering to absorption by aerosol decreased from 4 at $m = 1{\cdot}1$ to 0·5 at $m = 2$.

Mean monthly values of τ_a at four Meteorological Office stations were calculated from records of solar radiation and hours of sunshine and corresponding values of total and diffuse flux were tabulated for ' isolated ', ' rural ' and ' urban ' sites.

1. INTRODUCTION

The presence of solid particles in the Earth's atmosphere has important consequences for the transmission of solar radiation and for the nature of the radiation régime at the ground. The absorption of solar energy by a layer of aerosol increases the radiative heating of the atmosphere and decreases the amount of energy available at the surface. Scattering by aerosol increases the amount of radiation which is reflected by the atmosphere into space and increases the downward flux of diffuse radiation at the Earth's surface. Attenuation also produces changes in the spectral composition of solar radiation which are significant biologically.

To estimate the amount of radiant flux which is absorbed and scattered by aerosol as distinct from other atmospheric constituents, measurements of direct and diffuse radiation at the ground may be compared with the fluxes predicted below a model atmosphere containing appropriate amounts of ozone, water vapour, and carbon dioxide (G. D. Robinson 1962, 1966). The height distribution of aerosol can be inferred by measuring solar radiation from aircraft (Roach 1961), and the presence of particles as high as 50 km has been demonstrated by measuring the scattering of light from searchlight beams (Elterman, Wexler and Chang 1969).

Concern about possible changes in global climate has stimulated new interest in the radiative effects of aerosol. Recent calculations by Rasool and Schneider (1971) imply that any future increases of aerosol content will decrease the mean surface temperature of the Earth and that the heat balance of the atmosphere may become increasingly sensitive to changes of aerosol content. To be able to detect the radiative effects of changing aerosol content, it is essential to establish baselines for the income of solar radiation in different parts of the world and to show how this income is related to aerosol load. This kind of exercise has been attempted at relatively few sites. Valko (1963) analysed turbidity measurements at Locarno-Monti in Switzerland and demonstrated marked daily and annual changes in the strength of the direct solar beam which he ascribed to differences in the composition of aerosol in different air masses. Flowers, McCormick and Kurfis (1969) reported measurements of turbidity from a network of stations in the USA equipped with sun photometers. These instruments record the irradiance of the direct beam at a wavelength of 0·55 μm but they are not accurate for measuring turbidity in air which has a small aerosol content. At Mauna Loa in the Pacific, the mean turbidity of the atmosphere increased rapidly between 1958 and 1967 (Peterson and Bryson 1968) but the trend has since reversed and is now interpreted as a prolonged effect of volcanic activity (Sawyer 1971; Dyer and Hicks 1968) rather than a sinister index of atmospheric pollution by man.

The state of radiation climatology in different parts of the world reveals the different priorities of national meteorological services. Research has been strongly supported in the USSR (Kondratyev 1969) but there has been less progress in the USA and Western Europe and little in Britain. Irrespective of geography, information about the radiative effects of aerosol is needed for atmospheric model building, for the interpretation of radiometric measurements from satellites, for estimating regional differences of surface water loss by evaporation, for ecological studies of spectral composition in relation to the growth and development of plants, and for architectural studies of the distribution of light and heat in buildings. With these diverse needs in mind, we report a series of measurements of solar radiation between 1967 and 1971 and analyse them in terms of aerosol distribution.

2. Theoretical background

The attenuation of monochromatic radiation in a clean atmosphere follows the Bouguer-Lambert Law except at very low solar angles irrelevant to this study. Beneath the atmosphere, the monochromatic irradiance on a plane perpendicular to the solar beam is given by the relation

$$S_{p\lambda} = S_{p\lambda}^* \exp\left[-\int_0^\infty a_\lambda \, \rho \, dh\right] \qquad . \qquad . \qquad . \qquad (1)$$

where $S_{p\lambda}^*$ is the irradiance per unit wavelength outside the atmosphere, a is a mass attenuation coefficient and ρ is the density in a pathlength dh. To relate $S_{p\lambda}$ more directly to solar angle, Eq. (1) may be rewritten in the form

$$S_{p\lambda} = S_{p\lambda}^* \exp\left(-\tau_\lambda \, m\right) \qquad . \qquad . \qquad . \qquad (2)$$

where $\tau_\lambda = \int_0^\infty a_\lambda \, \rho dh$ for a vertical beam and m is the air mass number (approximately sec z where z is solar zenith angle). The quantity τ_λ is the monochromatic optical thickness of the atmosphere.

The attenuation of solar radiation in the atmosphere takes place by gaseous absorption (g) molecular scattering (s) aerosol absorption (aa) and aerosol scattering (as). With appropriate suffices, τ_λ may be expressed as the sum of four components.

$$\tau_\lambda = \tau_{g\lambda} + \tau_{s\lambda} + \tau_{as\lambda} + \tau_{aa\lambda} \qquad . \qquad . \qquad . \qquad (3)$$

The coefficient τ_λ depends strongly on wavelength, because gaseous absorption is very selective and Rayleigh scattering is proportional to λ^{-4}.

For the whole solar spectrum, an integral optical thickness τ may be defined by the relation

$$\exp -(\tau \, m) = \int_0^\infty S_{p\lambda}^* \exp -(\tau_\lambda \, m) \, d\lambda \Big/ \int_0^\infty S_{p\lambda}^* \, d\lambda \quad . \qquad . \qquad (4)$$

or

$$\tau = -m^{-1} \ln \left\{\left[\int_0^\infty S_{p\lambda}^* \exp\left(-\tau_\lambda \, m\right) d\lambda\right] \Big/ \int_0^\infty S_{p\lambda}^* \, d\lambda\right\}$$

but because τ_λ is a function of wavelength, τ and other similar coefficients depend on air mass number and cannot provide a unique index of aerosol attenuation or composition. This behaviour is a major drawback in using τ, atmospheric transparency (Kondratyev 1969) or Linke's turbidity factor (Linke 1942) to study diurnal changes of turbidity.

To get a more consistent measure of attenuation by aerosol, Blackwell, Eldridge and Robinson (1954) compared measured fluxes of solar radiation with estimates for a clean moist atmosphere. In effect, they expressed $S_{p\lambda}$ as

$$\begin{aligned} S_{p\lambda} &= S_{p\lambda}{}^* \,[\exp -(\tau_{g\lambda} + \tau_{s\lambda})\, m]\, [\exp -(\tau_{aa\lambda} + \tau_{as\lambda})\, m] \\ &= S_{p\lambda}(0)\, [\exp -(\tau_{a\lambda}\, m)] \qquad (5) \end{aligned}$$

where $S_{p\lambda}(0)$ is the irradiance below an atmosphere free from aerosol and $\tau_{a\lambda}$ is a spectral attenuation coefficient for aerosol. Most of the wavelength dependence of $S_{p\lambda}$ is now contained in the term $S_{p\lambda}(0)$ and the integral coefficient derived from the expression corresponding to Eq. (4) is

$$\begin{aligned} \tau_a &= -m^{-1} \ln \left\{ \left[\int_0^\infty S_{p\lambda}(0) \exp -(\tau_{a\lambda}\, m)\, d\lambda \right] \Big/ \int_0^\infty S_{p\lambda}(0)\, d\lambda \right\} \qquad (6) \\ &= -m^{-1} \ln \left[S_p(\tau)/S_p(0) \right] \end{aligned}$$

where $S_p(\tau)$ and $S_p(0)$ are the measured and calculated values of irradiance over the whole spectrum measured normal to the solar beam.

For small values of τ_a, the fractional attenuation of radiation by aerosol is

$$[S_p(0) - S_p(\tau)]/S_p(0) = 1 - \exp(-\tau_a\, m) \simeq \tau_a\, m \qquad (7)$$

i.e. τ_a is approximately the fractional attenuation per unit air mass.

The calculation of $S_p(0)$ followed the method outlined by G. D. Robinson (1962) but it was based on an extra-terrestrial spectrum given by List (1966) equivalent to a Solar Constant of 1353 W m^{-2}. The absorption by ozone, assigned to the outer limit of a plane atmosphere, was calculated from the integral absorption coefficients of Fritz (1949) adopting the annual variation of ozone content given by N. Robinson (1966). Precipitable water was calculated from the appropriate radiosonde ascent (usually at Crawley) and the absorption of radiation by water vapour, carbon dioxide and oxygen was calculated from the absorptivities given by Yamomoto (1962). The reflection coefficient of the ground was assumed to be 0·25, an appropriate value for green vegetation, and the contribution of reflected radiation to the downward diffuse flux was allowed for by an approximation used by Robinson (1962).

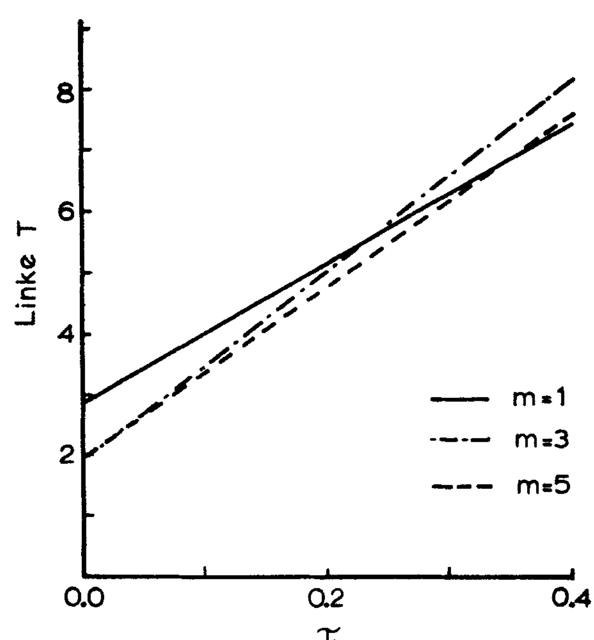


Figure 1. Dependence of the Linke turbidity factor on τ_a for three values of air mass.

To allow τ_a to be compared with other coefficients of turbidity, Fig. 1 shows the variation of the Linke turbidity factor T (Linke 1942) with τ_a for three air mass numbers, assuming 2 cm precipitable water. The Linke factor is defined by the relation

$$S_p(\tau) = S_p^* \exp -(\tau_s + \tau_a + \tau_w(um))\, m = S_p^* \exp(-T\,\tau_s\, m) \qquad (8)$$

so that

$$T = 1 + (\tau_a + \tau_w(um))/\tau_s$$

where τ_s and τ_w are the integral optical thicknesses for molecular scattering and water vapour absorption respectively, u is the optical path for water vapour and S_p^* is the irradiance outside the atmosphere. The factor T therefore represents the number of clean dry atmospheres which would produce the same attenuation as the actual atmosphere containing water vapour and aerosol. Because τ_w is a function of um, T is a function of air mass number as well as atmospheric water content. For example, if τ_a were constant at 0·05 over a whole day, Fig. 1 implies that the diurnal variation of T would exhibit a spurious maximum at noon and secondary maxima early and late in the day. Beneath a very turbid atmosphere, these spurious cycles would be reversed. N. Robinson (1966) drew attention to this possible explanation for changes of T recorded by Hinzpeter (1950).

3. Experimental background

Most of the measurements were made at Sutton Bonington (52·8° N, 1·3° W), a rural site within 15 mi of three large industrial conurbations: Nottingham (north-north-east) Derby (north-west) and Leicester (south-south-east). The nearest substantial source of man-made aerosol is the 2,000 MW coal-fired power station at Ratcliffe-on-Soar, about 2 mi north of the experimental site, but a careful note was made of the few occasions when this source was likely to affect radiation measurements. The radiation regime at the site is probably representative of much of England, the industrial midlands of Scotland and similar areas of western Europe between latitudes 50° and 55°N. The amount of industrial pollution reaching more remote areas is likely to be small and relatively strong insolation has been recorded at Aberporth on the Welsh coast (Monteith 1966), at Littlehampton on the south coast of England (Rees 1968) and over parts of Scandinavia (Black 1960).

Supplementary measurements were made at Strontian and on Ben Nevis (north-west Scotland) and in Northumberland, sites where the average aerosol content of the atmosphere was expected to be less than at Sutton Bonington. Several spot readings were made in Edinburgh during the clearance of a haar which descended on the Summer Meeting of the Royal Meteorological Society in August, 1970.

The direct component of solar radiation at normal incidence S_p was measured with a Linke-Feussner pyrheliometer. The filter ring at the head of the instrument contained a Schott RG 695 (formerly RG 8) filter transmitting radiation at wavelengths exceeding 0·7 μm. Irradiance in the ultra-violet and in the visible spectrum (0·30 to 0·70 μm) was found from the difference in output with and without the filter after correcting for absorption in the pass band and reflection. The time of each pair of readings was noted so that an accurate solar zenith angle could be calculated from the latitude of the site and from solar declination. The temperature of the thermopile was also recorded and the calibration of the instrument was adjusted using a temperature coefficient given by the manufacturers.

The output from the thermopile was measured with a Comark portable microvoltmeter and the sensitivity of the two scales used during the measurements (10 and 30 mV full scale) was checked frequently with a standard voltage source.

On a number of cloudless days, the irradiance of a horizontal surface S_t was measured with a conventional Kipp solarimeter. The diffuse component S_d was estimated by shading the thermopile and surrounding glass hemispheres with a small disc subtending the same angle as the aperture of the pyrheliometer (10°) and the direct component S_s ($= S_p \cos z$) was estimated from $S_t - S_d$. The solarimeter was calibrated regularly against a standard instrument in an integrating sphere at Kew Observatory.

Consistency between the calibrations of the solarimeter and the pyrheliometer was achieved by recording the direct component of radiation with both instruments simultaneously. Accepting the calibration of the solarimeter as correct, the sensitivity of the pyrheliometer determined by this method appeared almost constant during the period of the measurements and was always within $\pm$ 2 per cent of the manufacturer's calibration. The absolute error of the pyrheliometer readings is therefore estimated to be $\pm$ 2 per cent irrespective of solar angle. The corresponding error in solarimeter readings was probably about $\pm$ 2 per cent when the zenith angle was less than 70° increasing to between $\pm$ 5 per cent and $\pm$ 10 per cent when the sun was nearer the horizon.

4. Results

(*a*) *Case studies*

The turbidity of the atmosphere measured at any site depends partly on local weather which determines the input of aerosol from domestic and industrial sources and partly on the synoptic history of the prevailing air mass which determines the input of aerosol from much more distant sources and its distribution throughout the troposphere. Inter-diurnal changes of turbidity with changes of air mass are well documented (Kondratyev 1969; Joseph and Manes 1971) and there is some less conclusive evidence for an intra-diurnal cycle related to the vertical transport of dust and pollen by convection (Monteith 1962).

An examination of all the evidence from the present study revealed that on days when the synoptic situation was almost static, τ_a changed by less than $\pm$ 10 per cent during the day and showed no systematic diurnal cycle. Fig. 2 shows examples of this behaviour which was observed at all times of year and with many combinations of surface conditions and wind speeds. In contrast, on days when a distinct change of air mass could be inferred from a synoptic development, τ_a increased or decreased substantially, sometimes by a factor of 2 between sunrise and sunset.

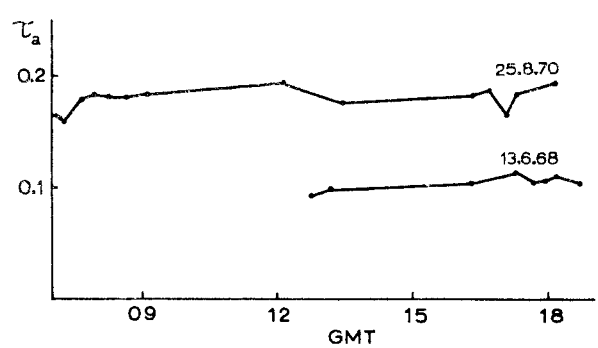


Figure 2. Diurnal variation of τ_a on 13 June 1968 and 25 August 1970.

Figs. 3 to 5 present the variation of τ_a over periods of several consecutive days and the corresponding synoptic situation over Europe and the North Altantic. Fig. 3 shows a sequence from 4 to 8 April 1969. On 4 April, relatively clean, maritime air was moving across Britain. By the 5th, air of continental origin began to intrude and τ_a increased initially. The slight decrease of τ_a during this day may be ascribed to small displacements of a rather poorly defined trajectory. On 6 April, the continental airstream became well established; τ_a increased and reached very large values on the 7th. On the 8th, the tropical continental air was gradually replaced by air of Polar continental origin and τ_a fell rapidly throughout the day.

Fig. 4 illustrates the contrasting behaviour of τ_a that sometimes occurred on consecutive days. On 7 June 1969, air of tropical maritime origin circulated in an anticyclone

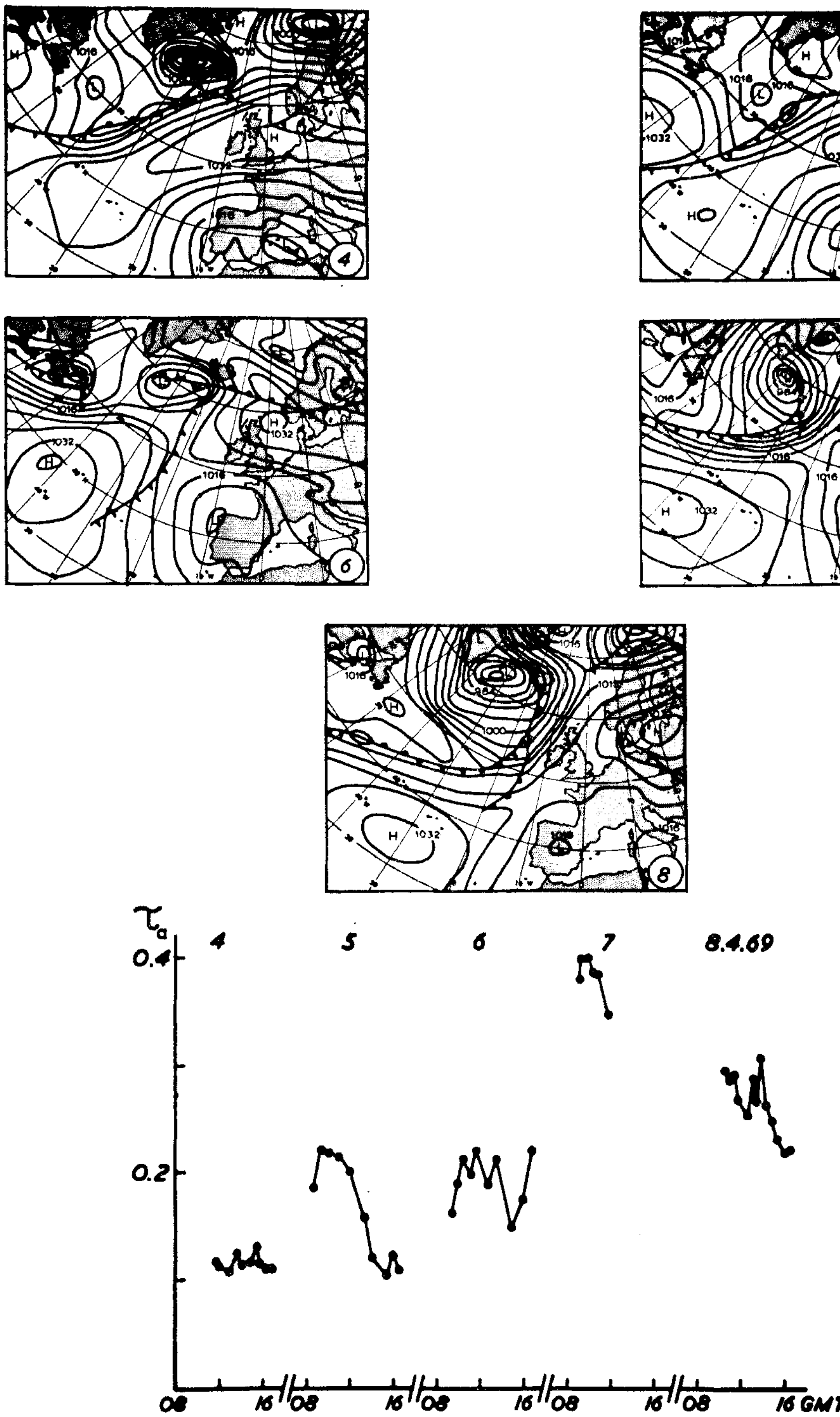
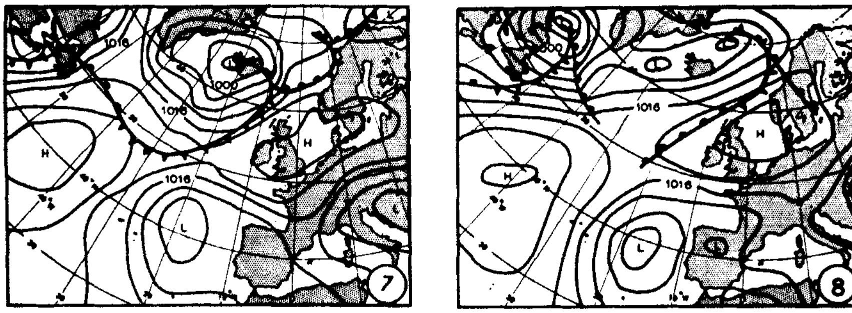


Figure 3. Diurnal variation of τ_a, from 4 to 8 April 1969.

over central England and the North Sea. Small values of τ_a are consistent with a record of exceptionally good visibility and the air became only slightly more turbid during the day. By 8 June, the atmosphere was much more turbid and large fluctuations of τ_a were



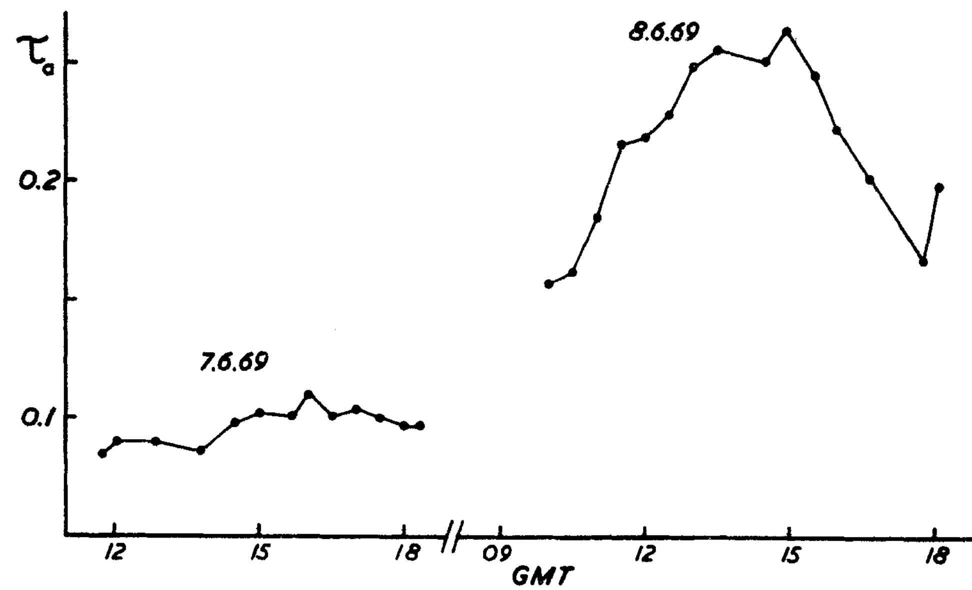
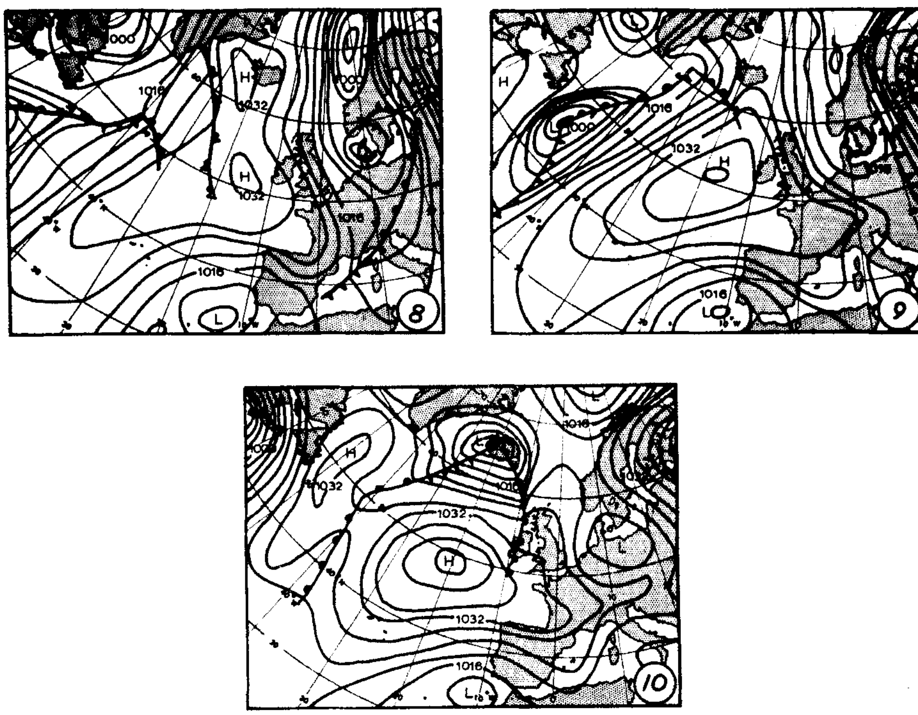


Figure 4. Diurnal variation of τ_a on 7 and 8 June 1969.

observed. Local surface conditions did not change from the previous day and there were no upwind sources of local man-made pollution to explain the erratic behaviour of τ_a. It appears that aerosol of continental origin, irregularly distributed as a result of trajectory changes, was probably responsible for these fluctuations.

As an exception to prove the rule, Fig. 5 presents a series of days in February 1969 which was the only period during which substantial changes of τ_a could not be explained in terms of the synoptic situation. On 8 February very cold air of Arctic maritime origin passed over the whole of industrial Lancashire and the Derby-Nottingham region, before reaching Sutton Bonington. Although there was no apparent change of trajectory, τ_a almost doubled during the day. As the anticyclone developed, the weather map indicated a possible incursion of tropical maritime air. Overnight, τ_a decreased substantially but it doubled again on the 9th. These increases of τ_a were probably caused by the accumulation of industrial and domestic smoke beneath an inversion which was recorded at about 1,000 m over Crawley. On 10 February, the increase of τ_a was repeated till noon but was followed by an equally rapid fall, probably following the advection of cleaner maritime air behind a warm front.

Many other observations support the contention that major changes in aerosol attenuation at a rural site such as Sutton Bonington are usually the result of changes in air mass type and that local sources of aerosol, either natural or man-made, are seldom responsible for comparable changes in the attenuation of solar radiation.



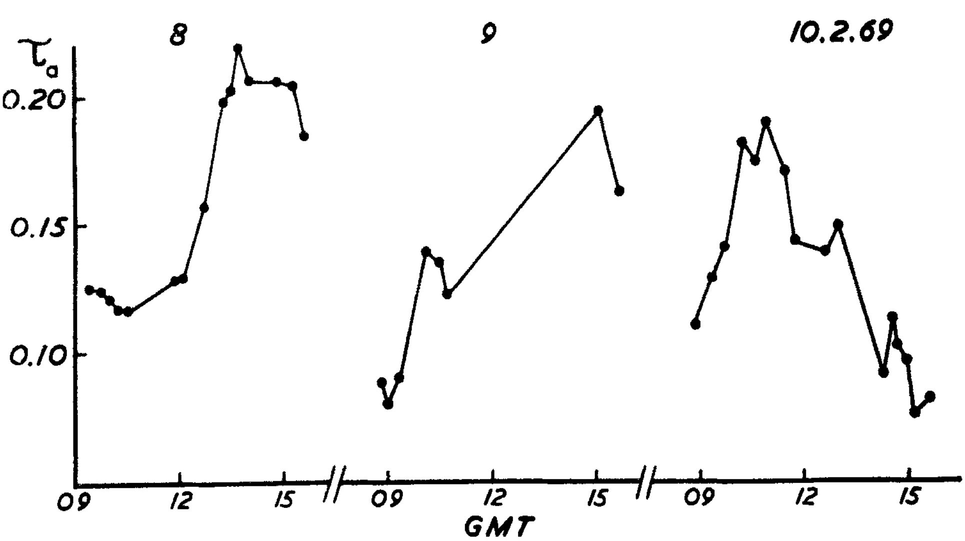


Figure 5. Diurnal variation of τ_a from 8 to 10 February 1969.

(*b*) *Air mass classification and* τ_a

Daily averages of τ_a were calculated for days when there were long clear periods. Although there was no pronounced annual change of $\bar{\tau}_a$, there was a tendency for larger values to be recorded more often in summer than in winter because of the greater frequency of continental air masses. This tendency is consistent with a previous tentative conclusion from the analysis of monthly radiation totals that the fractional loss of radiation attributable to aerosol is greater in summer than in winter (Monteith 1966).

Fig. 6 shows daily averages of τ_a classified by air mass type. Almost all the observations for Polar and Arctic continental air masses were made in summer when the two types were often indistinguishable and they were therefore left unseparated. The diagram shows that the minimum aerosol attenuation at Sutton Bonington is obtained in Arctic or Polar maritime air when $\bar{\tau}_a$ is about 0·1. The maximum attenuation occurs in tropical continental air when $\bar{\tau}_a$ is about 0·4 to 0·5.

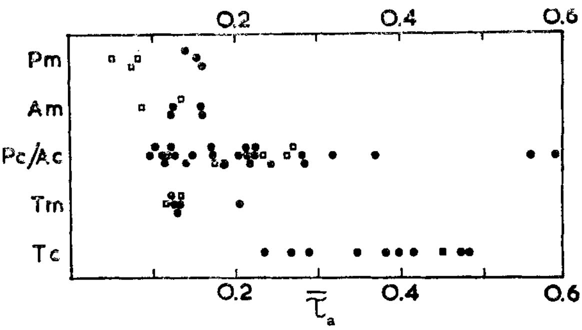


Figure 6. Daily averages of τ_a classified by air mass type
● Sutton Bonington, □ Strontian, ■ Edinburgh.

Fig. 6 also shows values of $\bar{\tau}_a$ at Strontian and a single point at Edinburgh. Comparison with the Sutton Bonington points suggests that $\bar{\tau}_a$ at stations remote from urban areas may be about 0·05 less on average than at ' rural ' sites close to cities or large industrial sources of pollution.

(c) *Change of τ_a with height*

On 20 August 1971, an almost cloudless day in parts of north-west Scotland, the irradiance of the solar beam was measured with the Linke-Feussner radiometer during an ascent of Ben Nevis (1340 m). The climb began from Fort William at 0930 GMT and readings were made at intervals of about 200 m until the summit was reached at 1330. A final reading was made at the base site in the late afternoon.

The synoptic situation during the week ending 21 August was dominated by the Azores anticyclone, and on the 20th a ridge of high pressure extended north-eastwards over the north of Scotland. The surface wind at Fort William was light and south-westerly. During the morning, a shallow layer of cumulus appeared over hills to the north-west of Ben Nevis but did not develop much higher than the top of a haze layer, visible at about 800 m. A shallow inversion between 400 and 700 m was recorded by the Stornoway radio-sonde ascent at 1100 GMT. During the afternoon, a few transitory patches of orographic cloud appeared over the summit of Ben Nevis.

The change of τ_a with height shown in Fig. 7 was calculated from Eq. (6), allowing for the decrease in atmospheric pressure and in precipitable water. The first three readings in the morning were rather erratic but the lower or the two base readings (τ_a = 0·127) was consistent with the reading after the descent (τ_a = 0·115). The average change of τ_a with height was about 0·004 per 100 m. No systematic change was detected in the relation between τ_a and height above and below the inversion layer which was very shallow compared with the depth of atmosphere in which the aerosol was dispersed.

Schonbachler and Valko (1965) reported rather larger changes of a turbidity coefficient with height in the Alps and their measurements appear consistent with figures given by Elterman (1968) for the distribution with height of the aerosol in a continental air mass. The Ben Nevis measurements reveal less turbidity at sea level and less decrease of turbidity with height as expected for a more dilute maritime aerosol distributed through a greater depth in the troposphere.

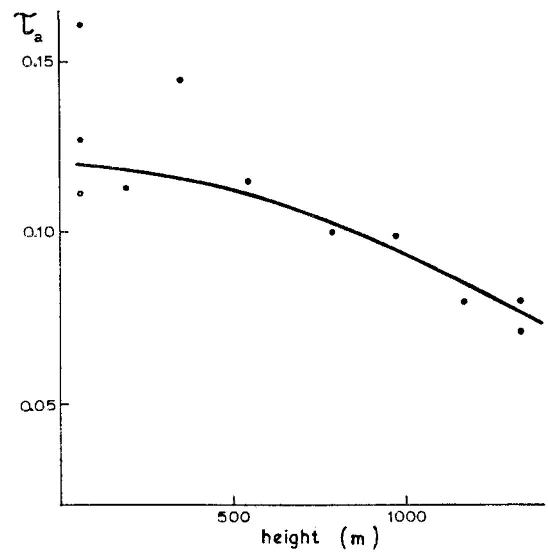


Figure 7. Variation of τ_a with height on Ben Nevis. ● during ascent, ○ after descent.

(*d*) *Spectral composition*

Below an atmosphere in which the attenuation of direct radiation takes place by Rayleigh scattering only, the fraction of radiation in the waveband 0·3 to 0·7 μm is $\epsilon_v = 0{\cdot}45$ for unit air mass number and decreases to 0·42 for air mass 2 (Linke 1942). The fraction is not sensitive to small changes in the spectrum adopted for extra-terrestrial radiation. In a real atmosphere, the main constituents which modify the value of ϵ_v are water vapour and aerosol. When the water vapour content of the atmosphere increases, the absorption of infra-red radiation increases and so does ϵ_v. Conversely, when the aerosol content increases, ultra-violet and visible radiation are likely to be scattered more than infra-red radiation so ϵ_v is expected to decrease. For arbitrary values of 2·0 cm precipitable water and 300 dust particles per cm^3 at ground level, Moon (1940) estimated that ϵ_v was 0·50 for $m = 1$ and 0·47 for $m = 2$. These figures agree with more rigorous calculations by Avaste, Moldau and Schifrin (1962) for an atmosphere of average turbidity i.e. 2·1 cm precipitable water and $\tau_\lambda = 0{\cdot}3$ at 0·55 μm. Analysis of these calculations indicates that when m is less than 2, aerosol content will usually be a more important determinant of ϵ_v than water vapour. At larger air mass numbers, increasing selective absorption and multiple scattering preclude the separation of aerosol effects from other types of attenuation.

The minimum air mass number in this series of measurements was 1·14 and since ϵ_v was expected to vary little between $m = 1{\cdot}14$ and 2·0, the analysis of measurements with the RG 695 filter was restricted to this range. Fig. 8 shows the dependence of ϵ_v on τ_a derived from these readings. If the dependence of τ_a on water vapour content is ignored, the best straight line through the points is

$$\epsilon_v = a - b\,\tau_a \qquad . \quad . \quad . \quad . \quad (9)$$

where $a = 0{\cdot}537 \pm 0{\cdot}004$ and $b = 0{\cdot}28 \pm 0{\cdot}01$. Taking a value of $\tau_a = 0{\cdot}2$ to represent the average amount of aerosol in the air masses concerned, the corresponding value of ϵ_v is 0·48.

Despite the scatter of points in Fig. 8, the observations contain some evidence for the dependence of ϵ_v on water vapour content. The calculations of Avaste *et al.* (1962) suggest that plots of ϵ_v against τ_a for specified amounts of precipitable water should consist of almost parallel straight lines. Some of our ϵ_v measurements support this conclusion, but there were very few days when τ_a was constant enough for a consistent analysis.

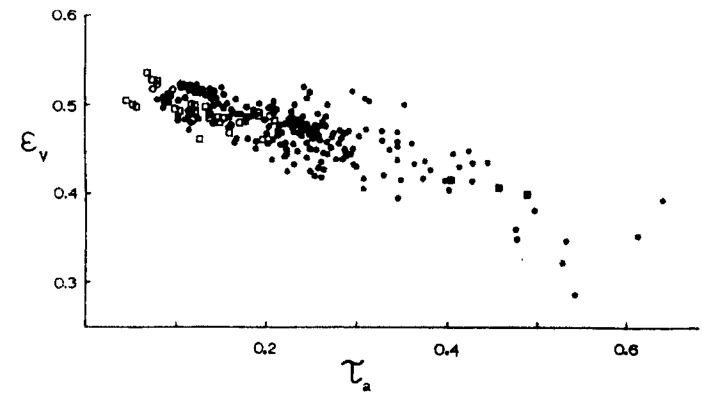


Figure 8. Variation with τ_a of ϵ_v, the fraction of direct radiation in the waveband 0·3 to 0·7 μm; zenith angle $z < 60°$. ● Sutton Bonington, □ Strontian, ■ Edinburgh.

The results do not support the conclusion drawn from a much smaller number of measurements at Kew that ϵ_v was independent of the aerosol content of the atmosphere (Blackwell *et al.* 1954).

(e) *Ratio of diffuse to total radiation*

When solar radiation is intercepted by aerosol particles in the size range 0·1 to 1 μm, theory predicts that most of the scattered radiation will travel forwards at a small angle to the direction of the incident radiation. The theory for multiple scattering is very complex, but for single scattering when the sun is more than 30° above the horizon, the ratio of downward to upward scatter by aerosol is probably between 5 and 10 (G. D. Robinson 1962). Because the scattered radiation is concentrated at small angles, the sky round the sun appears brighter than the rest of the hemisphere (in the absence of cloud). When diffuse radiation is measured with a solarimeter mounted under a shade ring, some of this scattered flux is not recorded, nor is it properly allowed for by a conventional shade ring correction based on the assumption that sky radiation is isotropic (Drummond 1955). Measurements of diffuse radiation with a shade ring therefore tend to underestimate the diffuse flux and this error increases with the aerosol content of the atmosphere.

To minimize the error, diffuse radiation was measured by shading a Kipp solarimeter with a disc occluding a solid angle of about 0·1 steradians. When the sky was cloudless and solar elevation exceeded 30°, the ratio S_d/S_t was expected to depend on the aerosol content of the atmosphere but not on solar angle (Dogniaux 1954). The ratio observed in these conditions was plotted against contemporary values of τ_a determined from measurements of S_p with the Linke-Feussner instrument. Fig. 9 shows that the points were well fitted by the straight line

$$(S_d/S_t) = c + d\,\tau_a \quad . \quad . \quad . \quad . \quad (10)$$

where $c = 0{\cdot}097 \pm 0{\cdot}009$ and $d = 0{\cdot}68 \pm 0{\cdot}04$. The scatter of points may reveal differences in aerosol size distributions. Eq. (10) predicts that as the aerosol content of the atmosphere tends to zero, the ratio S_d/S_t should tend to 0·097, a value which is close to the theoretical ratio of 0·072 for a model atmosphere containing 2 cm precipitable water.

(f) *Ratio of scattering to absorption*

Measurements of direct and diffuse radiation at the Earth's surface can be used to estimate the relative contributions of scattering and absorption to the total attenuation of radiation by aerosol. If $\theta(z)$, a function of zenith angle, is the ratio of downward to total scattering by aerosol and $\alpha(z)$ is the ratio of total scattering to absorption, then downward

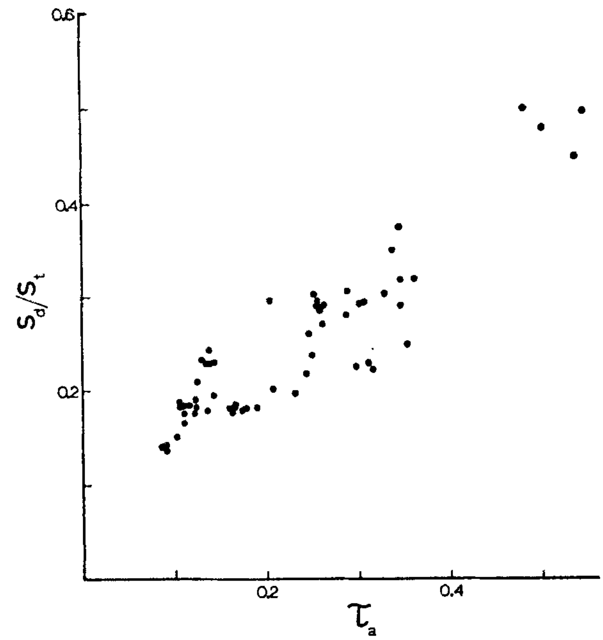


Figure 9. Variation of ratio of diffuse to total radiation S_d/S_t on cloudless days (z <60°) with τ_a.

scattering is responsible for a fraction $\alpha\ \theta/(\alpha + 1)$ of the total attenuation. For solar zenith angles less than 60°, radiation scattered forwards by aerosol is recorded as a contribution to the downward diffuse flux at the ground. The difference between the measured diffuse flux $S_d(\tau, z)$ which depends on τ and z, and the estimated flux for an aerosol free atmosphere containing the same amount of water vapour $S_d(0, z)$, should be a fraction $\alpha\ \theta/(\alpha + 1)$ of the total attenuation of the direct beam referred to a horizontal surface. In symbols, the relation is

$$S_d\,(\tau, z) - S_d\,(0, z) = [\alpha\ \theta/(\alpha + 1)]\ [S_p\,(0, z) - S_p(\tau, z)] \cos z \quad . \quad (11)$$

When τ is between 0·1 and 0·3 and z is less than 60°, the relation between the radiative fluxes and τ is almost linear and can be expressed by equations of the form

$$S_p\,(\tau, z) = S_p\,(0, z) - e(z)\,\tau \quad . \quad . \quad . \quad (12a)$$

$$S_d\,(\tau, z) = S_d\,(0, z) + f(z)\,\tau \quad . \quad . \quad . \quad (12b)$$

Substituting these values in Eq. (11) gives

$$\alpha(z) = \{[\,e(z) \cos z\ \theta\,(z)/f(z)] - 1\}^{-1} \quad . \quad . \quad . \quad (13)$$

Values of e and f were determined by calculating S_p and S_d as functions of τ_a (see Figs. 11 to 13) and θ was calculated from the ratios of downward to upward scattering given by G. D. Robinson (1962). Robinson's ratios were ' a very rough estimate ' based on atmospheric measurements by Waldram (1945). However, because forward scattering is usually an order of magnitude larger than back scattering, θ is close to unity and the values of α derived from Eq. (13) are insensitive to error in the ratios adopted for θ.

Fig. 10 shows that the ratio of scattering to absorption decreased rapidly as sec z increased from 1·1 to 1·5 and approached a value of 0·5 as sec z approached 2.

Blackwell *et al.* (1954) analysed radiation at Kew on clear days and found that α decreased from 1·7 to 0·25 when sec z increased from 1·23 to 4·8 and the corresponding points lie on either side of the curve in Fig. 10. Roach's (1961) measurements of attenuation made from an aircraft over the English channel gave an average value of α of about 4 for

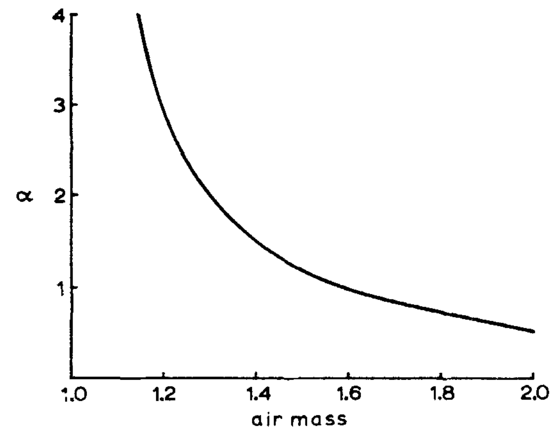


Figure 10. Dependence on air mass number (sec z) of the ratio α of scattering to absorption by aerosol.

values of sec z between 1·8 and 1·16. G. D. Robinson (1962) used data from a number of stations to show that the mean value of α was about 0·5 when sec z was between 1·15 and 3. Further analysis of his material, however, shows significant differences between sites. At most inland sites, α decreased with increasing sec z but at the island sites of Lerwick and Malta α increased with sec z. Some of these inconsistencies may be the result of errors in the measurement of diffuse radiation with solarimeters exposed beneath a shade ring (see Section 4(e)). Values of α based on such measurements will tend to be anomalously small because the flux of diffuse radiation is underestimated and the direct flux is overestimated.

Rasool and Schneider (1971) treated what they regarded as an extreme case of an absorbing aerosol. The single-scattering albedo defined as the ratio of total scattering to total attenuation i.e. $\alpha/(\alpha + 1)$ was 0·9 at 0·55 μm. This ratio corresponds to $\alpha = 9$.

Shettle (1972) analysed measurements of ultra-violet irradiance during a photochemical smog. His results are best fitted by $\alpha = 15$. He also quotes unpublished calculations by Twitty showing that α ranged from 4 to 12 for three aerosol size distributions. In view of the increased importance of scattering at short wavelengths these results are not inconsistent with the largest values reported here for the whole spectrum.

5. Generalizations

(a) *Daily radiation totals*

To establish the dependence of daily radiation totals on aerosol content, computer programs were written to calculate three quantities:

(i) the irradiance of the solar beam $S_p(\tau, z)$, for specific values of τ, assuming 1·0 cm precipitable water, 0·3 cm ozone, a solar constant of 1353 W m^{-2} and a solar radius vector of unity;

(ii) corresponding values of the total and diffuse radiation on a horizontal surface $S_t(\tau, z)$ and $S_d(\tau, z)$ (from Eq. (10) and the identity $S_t = S_d + S_p \cos z$);

(iii) daily integrals of S_p, S_t and S_d from irradiances calculated every fifteen minutes throughout the hours of daylight.

Eq. (10) is not valid for zenith angles exceeding 60°. To estimate total and diffuse fluxes at larger zenith angles, it was assumed that the ratio $[S_d(\tau, z)/S_t(\tau, z)] \div [S_d(0, z)/S_t(0, z)]$ was independent of z and was equal to the (constant) value established for $z < 60°$. The error introduced by this arbitrary assumption is small at latitudes less than 60° in summer because relatively little energy is received on a horizontal surface when z exceeds 60°, e.g. only about 10 per cent of the daily total radiation on 22 June at 52°N is received when z exceeds 60°. The error is much larger during winter months which were excluded from the analysis.

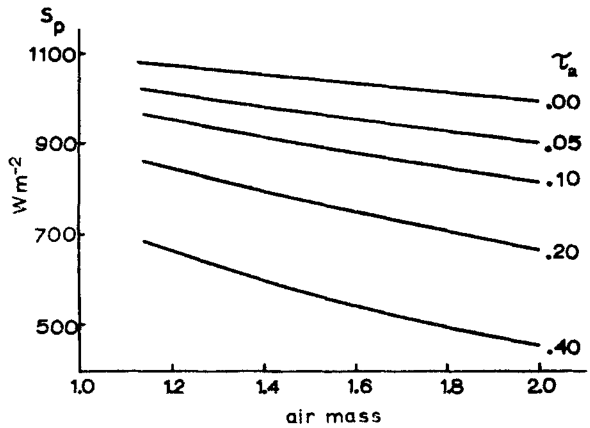


Figure 11. Dependence of S_p on air mass number and τ_a.

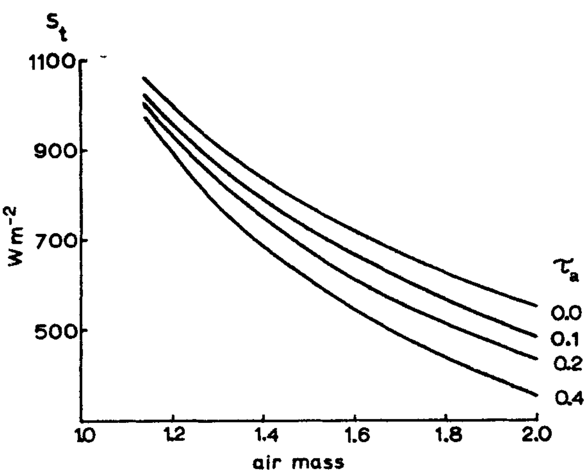


Figure 12. Dependence of S_t on air mass number and τ_a.

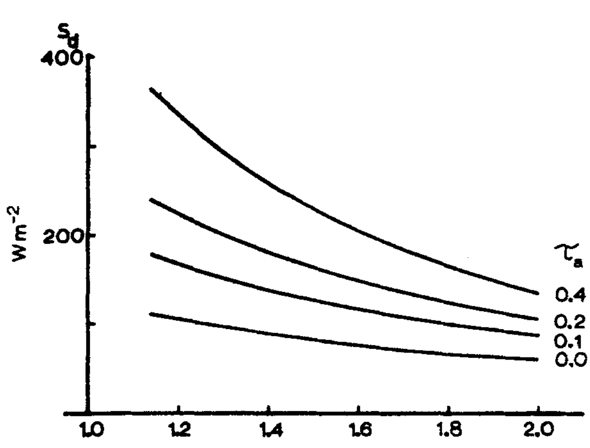


Figure 13. Dependence of S_d on air mass number and τ_a.

Representative Figures are presented graphically (Figs. 11 to 13) and as Tables (Appendix). More detail can be derived from the programs which are available from the authors. Because multiple scattering would increase losses of radiation at large zenith angles, the figures represent the maximum possible insolation for each value of τ.

The maximum daily insolation measured at Kew since 1957 is 30·9 MJ m^{-2} on 29 May 1966. This flux corresponds to $\tau \simeq 0{\cdot}1$ in good agreement with minimum values at Sutton Bonington. The maximum at Aberporth, 32·4 MJ m^{-2} on 9 June 1961, corresponds to $\tau \simeq 0{\cdot}04$. The evidence from Aberporth, Strontian, and Valentia on the west coast of Ireland (Unsworth and Monteith, in preparation) suggests that the minimum background aerosol content over the North Atlantic corresponds to $\tau \simeq 0{\cdot}05$, consistent with the factor which Houghton (1954) used for attenuation by ' dust '. Over many parts of Britain, the average value of τ is probably about 0·2. The loss of energy attributable to smoke and other industrial products can be estimated roughly by comparing entries in the tables for these two values of τ. At latitude 55°N, the loss of *total* radiation ranges from 18 per cent in March (possibly an underestimate for reasons discussed above) to 10 per cent in June. The loss in *direct* radiation on a horizontal surface, relevant to the distribution of radiant flux in buildings and within crop canopies, ranges from 36 to 29 per cent. The loss of visible direct radiation will be greater than the value calculated for the whole solar spectrum (Eq. (9)) but some of the visible flux scattered out of the direct beam will contribute to the diffuse component. Further measurements are needed to establish whether the relations between scattering and absorption described in Section 4(*e*) can be treated as independent of wavelength.

(*b*) *Geographical differences of* τ_a

Total and diffuse radiation are recorded at nine Meteorological Office stations in Britain and are reported as monthly averages in the *Monthly Weather Report*. Previous analysis of these measurements revealed unexpected differences in the attenuation of radiation at different sites. For example, the amount of direct solar radiation per hour of bright sunshine was 24 per cent higher at Aberporth than elsewhere in Britain (Monteith 1966). By combining these climatological measurements of radiation and sunshine duration with predictions for a model atmosphere, it is possible to derive monthly averages of the transmission coefficient τ_a.

Let N be the duration of sunshine registered by a Campbell-Stokes recorder on a completely cloudless day. In summer, values of N at British stations are usually about 1½ hours shorter than the corresponding duration of daylight because the cards used in the recorder do not start to burn until the irradiance of the direct solar beam exceeds a threshold which Cox (1972) established as about 200 W m^{-2}. Using this figure and taking average monthly values of precipitable water and ozone from Bannon and Steele (1960) and N. Robinson (1966) respectively, monthly mean values of $N(\tau)$ were calculated for four stations as functions of τ_a. The maximum values of sunshine duration for summer months at Sutton Bonington are consistent with estimated values for $\tau_a \simeq 0{\cdot}1$. For larger values of τ_a, N decreases almost linearly with increasing τ_a and can therefore be expressed as

$$N(\tau) = N(0{\cdot}1)(1 - g\,\tau_a) \qquad . \quad . \quad . \quad (15)$$

where $N(0{\cdot}1)$ is the value of N when $\tau_a = 0{\cdot}1$. The coefficient g, depending on season and on the latitude of the site, is about 0·7 for most British stations in summer (i.e. an increase of 0·1 in τ_a decreases the duration of sunshine by 7 per cent).

The direct flux received on a horizontal surface during a cloudless day or $\Sigma\, S_s(\tau)$ was also calculated as a function of τ and expressed as

$$\Sigma\, S_s(\tau) = \Sigma\, S_s(0{\cdot}1)(1 - h\,\tau_a) \qquad . \quad . \quad . \quad (16)$$

where the coefficient h is about 1·5. A mean irradiance for the solar beam $\bar{S}_s$ can be defined as $(\Sigma\, S_s)/N$ where ΣS_s is expressed in J m^{-2} and N is in seconds. This quantity can be written in the form

$$\bar{S}_s(\tau) = \bar{S}_s(0{\cdot}1)(1 - h\,\bar{\tau}_a)/(1 - g\,\bar{\tau}_a) \quad . \quad . \quad . \quad (17)$$

Solving Eq. (17) for $\bar{\tau}_a$ gives

$$\bar{\tau}_a = \{\bar{S}_s(\tau) - \bar{S}_s(0{\cdot}1)\}/\{g\,\bar{S}_s(\bar{\tau}) - h\,\bar{S}_s(0{\cdot}1)\} \quad . \quad . \quad (18)$$

To find $\bar{\tau}_a$, a value of $\bar{S}_s(\tau)$ was calculated from climatological records by subtracting the average daily value of diffuse radiation for a given month $\Sigma\,S_d$ from the corresponding total radiation $\Sigma\,S_t$. Then

$$\bar{S}_s = (\Sigma\,S_t - \Sigma\,S_d)/n$$

where n is the daily average of bright sunshine for the month.

TABLE 1. MONTHLY MEAN VALUES OF $\bar{\tau}_a$ BASED ON RADIATION DATA FROM 1966-1970

	$\bar{\tau}_a$			
	Lerwick	Eskdalemuir	Aberporth	Kew
April	0·11	0·26	0·20	0·36
May	0·23	0·33	0·33	0·42
June	0·19	0·30	0·28	0·43
July	0·24	0·32	0·22	0·41
August	0·20	0·28	0·29	0·39
September	0·12	0·15	0·17	0·28
Six-month mean	0·18	0·27	0·25	0·38

Table 1 shows values of $\bar{\tau}_a$ estimated from this analysis for four stations in six months. The main source of error in the estimates of $\bar{\tau}_a$ is probably the effect of thin high cloud which can reduce solar irradiance without affecting the record of sunshine duration. If this happened often, $\bar{\tau}_a$ would be overestimated.

The salient features of Table 1 are:

(i) the similarity of $\bar{\tau}_a$ at Aberporth and Eskdalemuir. The 5° difference of latitude between these stations was probably not accounted for properly in a previous analysis which implied that the insolation at Aberporth was anomalously large (Monteith 1966);

(ii) the values of $\bar{\tau}_a$ at Lerwick are smaller in every month than the values at Aberporth and Eskdalemuir and are 0·07 less on average;

(iii) the values of $\bar{\tau}_a$ at Kew are larger in every month than the values at Aberporth and are 0·13 more on average.

The geographical differences of $\bar{\tau}_a$ are consistent with the latitude and position of the four stations. Lerwick is far enough north to avoid frequent exposure to air masses with a long trajectory over the continent and winds from the south-east are relatively infrequent at this station and elsewhere in Britain. Eskdalemuir is occasionally exposed to smoke from Glasgow and Tyneside (McIntosh 1957) but values of $\bar{\tau}_a$ between 0·1 and 0·2 are probably common both at Eskdalemuir and at Aberporth. At Kew, on the other hand, local sources of pollution must be responsible for the relatively large values of $\bar{\tau}_a$.

The inferences from Table 1 and the material in the Appendix were combined with conclusions from the more detailed study at Sutton Bonington to give a broad picture of the relative importance of different sources of aerosol for the attenuation of solar radiation in Britain. Table 2 shows estimates of S_p, S_t and S_d for a solar zenith angle of 45° (air mass 1·4), solar radius vector of unity, 1 cm precipitable water and 0·3 cm ozone. Values of τ were rounded off to the nearest 0·05 to avoid an unjustified impression of precision and the relative importance of aerosol at different types of site was shown by expressing each irradiance as a percentage of the values for a 'clean' site with no local sources of atmospheric pollution. The figures re-emphasize one of the main conclusions from the measurements at Sutton Bonington that the attenuation of solar radiation by aerosol is determined mainly by the origin of the prevailing air mass and that local sources of aerosol are less important at most sites. Estimates of radiative flux for the same values of τ but for other solar angles can be derived by reference to Figs. 11 to 13.

TABLE 2. CHARACTERISTIC VALUES OF τ_a AND CORRESPONDING IRRADIANCE AT $z = 45°$

Description of Site	Air mass	τ_a	S_p (W/m^2)	Per cent of *maximum* S_p at cleanest site	S_d (W/m^2)	Per cent of *minimum* S_d at cleanest site	S_t (W/m^2)	Per cent of *maximum* S_t at cleanest site
No aerosol		0·00	1040		80		780	
Northerly island site or west coast of Ireland, minimum pollution from natural land sources (e.g. soil particles, spores) and negligible smoke	polar	0·05	980	100	105	100	755	100
	average	0·20	800	81	165	157	685	91
	continental	0·35	650	66	215	205	630	83
Rural or coastal site exposed to natural pollution and small amounts of smoke: no large towns within 20 miles	polar	0·10	920	94	125	119	730	97
	average	0·25	745	76	180	171	660	87
	continental	0·40	605	62	235	224	620	82
Urban site within or close to a large town (say, population exceeding 100,000)	polar	0·25	745	76	180	171	660	87
	average	0·40	605	62	235	224	620	82
	continental	0·55	490	50	285	272	585	78

ACKNOWLEDGMENT

The work reported in this paper formed part of a project supported by the Natural Environment Research Council.

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Appendix

Values of daily total and diffuse insolation, ΣS_t and ΣS_d respectively, were calculated in a computer program which numerically integrated irradiances S_t and S_d calculated over a time increment of 15 minutes. The number of sunshine hours N which would be recorded by a Campbell-Stokes recorder was also calculated assuming that burning of the card ceased when S_p was less than 200 W m^{-2} (Cox, 1972). Tables A1-A3 gives figures for ΣS_t, ΣS_d and N for three latitudes (52, 55 and 58°N) on three dates (21 March, 1 May and 22 June corresponding to solar declinations of 0, 15 and 23·4° respectively) for values of τ_a ranging from 0 to 0·4. For all these calculations, monthly average values of precipitable water (interpolated from Bannon and Steele, 1960) and ozone (from N. Robinson 1966) were used, and the solar radius vector and declination were taken from List (1966).

TABLE A1. Variation of insolation and sunshine hours with τ_a for latitude 52°N

	Latitude 52°N								
	21 March			1 May			22 June		
τ_a	ΣS_t (MJm^{-2})	ΣS_d (MJm^{-2})	N (hours)	ΣS_t	ΣS_d	N	ΣS_t	ΣS_d	N
0·00	17·5	2·1	12·0	28·0	3·4	14·5	33·0	4·0	16·3
0·05	16·4	2·6	11·0	26·7	4·3	13·8	31·8	5·0	15·3
0·10	15·4	3·1	10·5	25·6	5·0	13·3	30·6	6·0	14·8
0·20	13·7	3·8	9·8	23·7	6·4	12·5	28·7	7·7	13·8
0·30	12·3	4·3	9·0	22·2	7·6	11·8	27·2	9·2	13·0
0·40	11·2	4·7	8·5	21·0	8·6	10·8	26·1	10·6	12·3

TABLE A2. Variation of insolation and sunshine hours with τ_a for latitude 55°N

	Latitude 55°N								
	21 March			1 May			22 June		
τ_a	ΣS_t (MJm^{-2})	ΣS_d (MJm^{-2})	N (hours)	ΣS_t	ΣS_d	N	ΣS_t	ΣS_d	N
0·00	16·1	2·0	12·0	27·0	3·3	14·8	32·8	4·0	16·8
0·05	14·9	2·4	11·0	25·7	4·1	14·0	31·4	5·0	15·8
0·10	13·9	2·8	10·5	24·5	4·8	13·5	30·2	6·0	15·3
0·20	12·2	3·4	9·8	22·5	6·1	12·5	28·2	7·6	14·3
0·30	10·8	3·8	9·0	21·0	7·2	11·8	26·6	9·0	13·3
0·40	9·7	4·1	8·0	19·8	8·2	11·0	25·3	10·4	12·5

TABLE A3. Variation of insolation and sunshine hours with τ_a for latitude 58°N

	Latitude 58°N								
	21 March			1 May			22 June		
τ_a	ΣS_t (MJm^{-2})	ΣS_d (MJm^{-2})	N (hours)	ΣS_t	ΣS_d	N	ΣS_t	ΣS_d	N
0·00	14·6	1·8	11·8	26·0	3·2	15·0	32·1	3·9	17·8
0·05	13·5	2·2	11·0	24·7	4·0	14·3	30·8	4·9	16·5
0·10	12·5	2·5	10·5	23·5	4·7	13·8	29·5	5·8	15·8
0·20	10·8	3·0	9·5	21·5	5·9	12·8	27·4	7·4	14·8
0·30	9·4	3·3	8·5	19·8	6·8	11·8	25·7	8·8	13·8
0·40	8·3	3·5	7·8	18·5	7·7	11·0	24·5	10·0	12·8