

ON SHORTWAVE RADIATION ABSORPTION BY TROPOSPHERIC AEROSOLS. I. "CLEAR" AND CONTINENTAL AIR MASSES

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On the basis of observations of shortwave radiation fluxes at different levels in the troposphere, the radiant energy influx has been determined in air masses of different types. Part I of this paper presents the results obtained in relatively "clear" air masses and in continental air masses (of desert and anthropogenic origin).

Investigations of descending- and ascending hemispheric radiation fluxes (illumination from above and below a horizontal surface) at various levels in the Earth's atmosphere make it possible to calculate the radiant energy influx at various atmospheric levels and, hence, to determine experimentally the spectral absorptivity of atmospheric aerosols. It is with this in mind that the above investigations have been made in the course of the "KENEX", "POLEX", and "GAREX" experiments, and others¹⁻⁴. To date, the laboratory of shortwave radiation of Leningrad State University has accumulated a rather sizable experimental database that enables one to make certain generalizations. It is not large enough to fully classify atmospheric aerosols by their absorptivity. However, right now, particular groups of air masses could be picked out from those observed which could provide a basis for future classification. This is the problem dealt with in the present paper.

CONVENTIONALLY "CLEAR" AIR MASSES

The determination of the radiant energy influx in a cloud-free atmosphere is a highly involved experimental problem, as one must calculate relatively small second differences of the observed descending $F^\downarrow(\lambda)$ and ascending $F^\uparrow(\lambda)$ radiation fluxes at the upper (1) and lower (2) boundaries of the observed atmospheric layer:

$$\begin{aligned} b_{12}(\lambda) &= [F_1^\downarrow(\lambda) - F_1^\uparrow(\lambda)] - [F_2^\downarrow(\lambda) - F_2^\uparrow(\lambda)] = \\ &= B_1(\lambda) - B_2(\lambda), \end{aligned} \quad (1)$$

where $B_1(\lambda)$ and $B_2(\lambda)$ are the radiant energy budgets at the upper and lower boundaries of the layer, respectively; $b_{12}(\lambda)$ is the radiant energy influx into the atmospheric layer between levels (1) and (2). It is clear that the error in determining the radiant energy influx in the atmosphere is about twice as large as the measurement error in the descending and ascending radiation flux. Therefore, if upon using the standard technique of photoelectric measurements under field

conditions the limiting random measurement error in the radiation flux is of the order of 1–15%, then the minimum determinable radiant energy influx will be about 2–3% of the fluxes involved. Only the use of a special measurement technique enabling one to take a uniform series of measurements, and hence to apply statistical methods of observational data processing, would enable one to reduce this estimated lower limit of the radiant energy influx somewhat. Thus, at the very outset, it should be pointed out that such air masses whose radiant energy influx is no greater than the threshold indicated above will be referred to as "clear" air masses.

Such air masses were observed in the first experiments of the "KENEX" program⁵. Figure 1 shows the relative radiant energy influx in a tropospheric layer at altitudes from 300–900 m to 4200 m obtained on July 17, August 4 and October 25, 1970. As is seen from this figure, on August 4 the air mass was conventionally "clear" since the radiant energy influx in the atmosphere was at most equal to the measurement error. It was a day of observations after a period of heavy rains with apparently resulted in either "washing out" aerosol particles from the troposphere or "wetting" them, which markedly reduced their absorptivity.

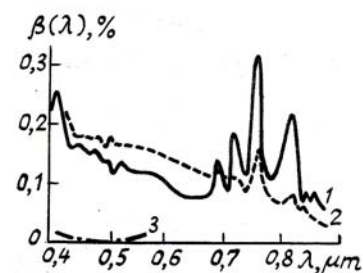


FIG. 1. Relative radiant energy influx in the tropospheric layer from 0.85 to 4.2 km recovered from the observations of June 17 (2), August 4 (3) and October 25, 1970 (1).

The radical modernization of spectrophotometric equipment accomplished in 1977–1983 and the use of

a measuring technique enabling one to carry out statistical processing of the measurements results⁶ reduced the minimum detectable radiant energy influx in the atmosphere by approximately a factor of 3 or 4, and made it equal to about 0.5–1% of the measured radiation flux⁷. Figure 2 presents the spectral behavior of the relative radiant energy influx in the atmospheric layer from the surface (50 m altitude) up to 5500 m (the influx was recalculated for a layer of 1000 mbar thickness; the quantities measured were half the plotted values), as obtained on May 22nd, 1973 over Lake Ladoga. The radiant energy influx in the wavelength range 400–700 μm is about 2–4%, which somewhat exceeds the threshold indicated above, but is essentially of the same order. The arctic air mass under observation was also "clear" in the sense mentioned above. It seems fairly clear that the total radiant energy influx can be measured quite reliably with a relative error of about 50%), while one can hardly discuss the spectral distribution of this quantity taking into account the measurement errors (except for a certain increase in the influx near the ozone absorption band).

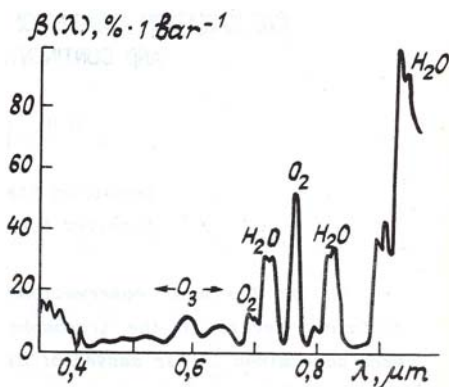


FIG. 2. Relative radiant energy influx over the Lake Ladoga in the layer of the troposphere from 0.05 to 5.5 km, recovered from observations of May 22, 1983.

Summing up the foregoing discussion, one can conclude that such situations can occur in the Earth's atmosphere when shortwave radiation absorption by tropospheric will be less (August 4th, 1970) than or comparable (May 22nd, 1983) to the error of its measurements. The experimentally determined spectral variations of spectral variations of the value are, of course, statistically insignificant.

CONTINENTAL AIR MASSES CONTAINING "DRY" AEROSOL

Radiant energy influx in the troposphere was first successfully determined during expedition work in the "KENEX-70" program on October 25th, 1970 over a desert surface, under near dust-storm conditions. The results of these studies were thoroughly discussed in Ref. 2. We note here just the main features and spectral behavior, as well as the altitude dependence of

shortwave radiation absorptivity of "dry" desert (sand, as will be shown below) aerosol.

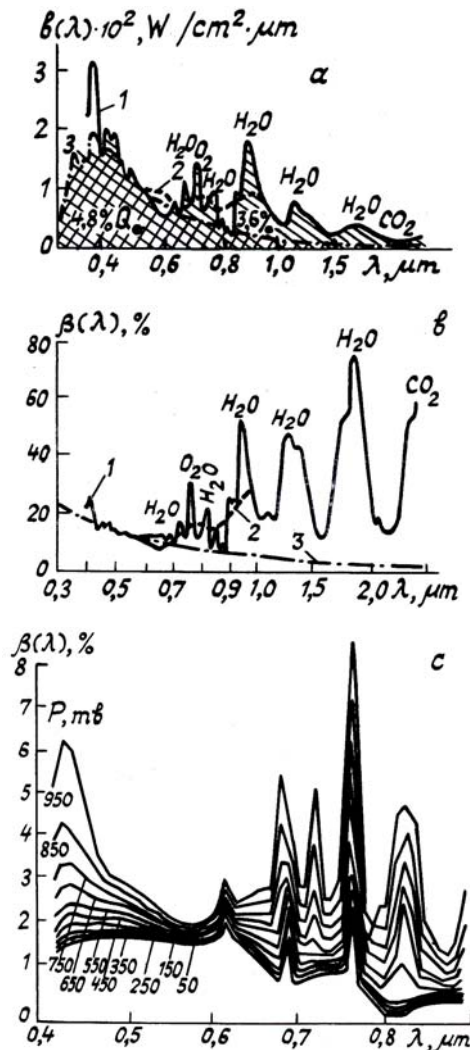


FIG. 3. Absolute (a) and relative (b) radiant energy influx over a desert in the tropospheric layer from 0.3 to 8.4 km on October 25, 1970; spectral curves of relative radiant energy influx (c) in layers with Δp = 100 mb at various levels in the atmosphere.

Figures 3a and 3b show absolute and relative radiant energy influx in the atmospheric layer from 0.3 to 8.4 km observed on October 25th, 1970 over Repetek station. We shall begin the analysis with the data presented in Fig. 2.

Figure 3b shows the spectral behavior of relative radiant heat influx $\beta(\lambda) = b(\lambda) / F_1^\downarrow(\lambda)$ (in percent). This curve explicitly shows the dominant role of molecular absorption in the relative radiant energy influx into the troposphere. If aerosol absorption near $\lambda = 0.4 \mu\text{m}$ is about 20%, then in the vicinity of $\lambda = 0.6 \mu\text{m}$ it is already reduced to 10%, and as the wavelength increases, it is reduced still more. At the centers of the molecular absorption bands, however,

corresponding values turn out to be 60% at $\lambda = 1.4 \mu\text{m}$, 80% at $\lambda = 1.8 \mu\text{m}$, etc. One can also see in Fig. 3b the spectral behavior of the imaginary part of the refractive index of hematite, the substance that gives desert sand its reddish color. It follows by comparing this curve with the spectral dependence of $\beta(\lambda)$ that the spectral behavior of aerosol absorption is similar to that of the imaginary part of the complex refractive of hematite. The averaged spectrum of aerosol absorption for spectral regions away from molecular absorption bands can be approximated by a λ^{-1} dependence. The curve of the averaged aerosol absorption, which is approximated by a λ^{-1} dependence, is plotted as curve 2 of Fig. 3b. This approximation is to be considered as one possible description of the spectral behavior of aerosol absorption in the near infrared region.

Curve 3 in Fig. 3a has been obtained by multiplying the smoothed curve of relative radiant energy influx (approximated by $\sim \lambda^{-1}$) by the energy distribution in the spectrum of the radiation incident on the upper boundary, which makes it possible to obtain the spectrum of the absolute averaged radiant heat flux into the troposphere due to absorption of radiant energy by aerosols. The area under this curve, which is equal to $0.095 \text{ cal/cm}^2\text{min}$. (4.8% of the solar constant), characterizes the total absorption of radiation by aerosols in the wavelength range under consideration. The area enclosed between this curve and the observed radiant energy influx curve is equal to $0.075 \text{ cal/cm}^2\text{min}$. (3.8% of the solar constant). It defines the total absorption of radiation by gas components of the atmosphere.

Thus, it happens that although molecular absorption plays the main role in the relative radiant heat influx, the spectrum of the source of radiation (the Sun) entering the troposphere is such that the absolute radiant energy influx due to absorption of radiation by aerosol and gas components of the atmosphere turns out to be of the same order of magnitude. The total absorption calculated using the data presented in Fig. 3a is equal to $0.17 \text{ cal/cm}^2\text{min}$ (8.5% of solar constant) with an error of about $0.04 \text{ cal/cm}^2\text{min}$.

The validity of the data obtained can be verified by comparing this value with independent actinometric (pyranometric) measurements: the influx into the layer 0.3 to 8.4 km from actinometric (integrated measurements is 0.204 ± 0.040 ; the influx into the layer 0.3–8.4 km, using spectral measurements and estimates of absorption in the range from 2.4 to $3.0 \mu\text{m}$, is $0.195 \pm 0.040 \text{ cal/cm}^2\text{min}$.

Such a good agreement between integrated radiant energy fluxes obtained independently is undoubtedly evidence of reliable spectral measurements. The validity of the data is also confirmed by the fact that the observed absorption by aerosols can be fully accounted for by the number of aerosol particles of a given chemical composition that were observed on the day the optical measurements were made.

Figure 3b shows the changes with height in the relative radiant energy influx into layers with

$\Delta p = 100 \text{ mb}$. In regions away from molecular absorption band, the radiant energy influx is due to aerosol absorption. In the lower layers of the atmosphere, the influx increases with decreasing wavelength. Obviously, this can be explained by the spectral behavior of the imaginary part of the refractive index of hematite. In the upper layers of the atmosphere, $\beta(\lambda)$ becomes independent of the wavelength, i.e., the aerosol becomes grey (apparently it is dust of cosmic origin or aerosol brought in from other regions). Thus, the aerosol generated by the desert surface rises only as high as the tropopause. The stratospheric aerosol is of different origin.

Results similar to those obtained in Repetek on October 25, 1970 were then also obtained in other experiments carried out in the same year, 1970⁵, in 1971⁸, and in 1974 in the vicinity of ATEP⁹ and others.

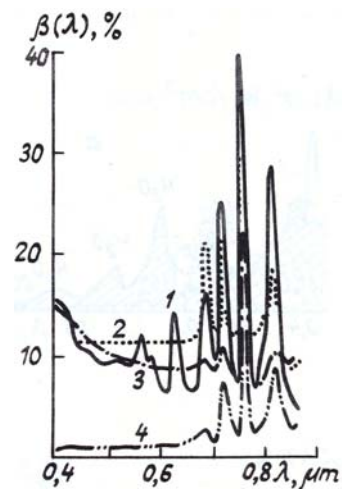


FIG. 4. Absolute radiant energy influx in the layer of the troposphere from 0.5 to 7.2 km as observed:

1 – in the vicinity of Zaporozh'e on August 22, 1972; 2 – in the vicinity of Rustavi on December 4, 1972; 3 – over Donetsk on August 9, 1975; 4 – over Rylsk on August 3, 1975.

Another type of "dry" continental aerosol repeatedly observed in the experiments was aerosol of anthropogenic origin, observed in air masses over industrial regions. Figure 4 presents the relative radiant energy influx into the entire layer of the atmosphere up to 4.2 km obtained in the vicinity of the town of Rustavi on December 4, 1972¹⁰, in Zaporozh'e and Dnepropetrovsk on August 22, 1972¹¹, in Rylsk on August 3, 1975 and in Donetsk on August 9, 1975¹². Examination of the figure shows that in industrial regions, the relative radiant energy influx can achieve the same values as in desert regions, i. e., at maximum in the blue region of the spectrum they can amount to 15–17%. At the same time, the spectral behavior of the influx proves to be less clear-cut namely: over Rustavi and Rylsk it was practically neutral. The curves rise in the blue region of the spectrum over Zaporozh'e and Dnepropetrovsk, and over Donetsk this might be due to the fact along with the

smoke from metallurgical plants, ferric oxides having the same spectral behavior as the imaginary part of the refractive index of hematite are emitted into the atmosphere. Besides, it should be noted that soot itself may be reddish as well.

Thus, as a result of these experiments, the absorptivity of continental "dry" aerosols of two kinds was investigated: desert (sand) and anthropogenic (smoke or dust). In both cases, very high integrated absorption was observed, sometimes reaching the total absorption by all the molecular components of the atmosphere. The spectral behavior of the radiant energy influx due to absorption of radiation by desert (sand) aerosol and by some kinds of anthropogenic (smoke) aerosols has a maximum of the imaginary part of the refractive index of ferric oxides (hematite, limonite and others) in the blue region of the spectrum. In a number of instances, anthropogenic aerosols (apparently sand and some kinds of smoke aerosols) absorb shortwave radiation nonselectively.

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