THE ROLE THAT TRACE SUBSTANCES PLAY IN THE RADIANT ENERGY BALANCE OF THE ATMOSPHERE

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RESUMEN

Los gases y las partículas y/o líquidos suspendidos en la atmósfera existen en cantidades muy pequeñas, sin embargo, absorben, tanto radiación solar como terrestre. En consecuencia generan divergencias del flujo radiante que influyen en el perfil vertical de la temperatura.

En este trabajo se presenta una comparación de la eficiencia de los diferentes absorbedores H_2O vapor de agua $(H_2O)_2$, CO_2,O_3 y aerosoles.

ABSTRACT

Trace substances in the atmosphere absorb radiation in the solar and terrestrial wavelength range. In consequence there will be radiant flux divergences changing the vertical temperature profile.

This paper presents a comparison of the efficiency of the different absorbers H_2O vapour $(H_2O)_2$, CO_2, O_3 and aerosol particles.

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Trace substances effect atmospheric heating or cooling in various ways. Emphasis is to be placed upon two basic processes:

a) The reflection of the solar and sky radiation by the surface of the earth and the cloud tops as well as the scattering of solar radiation on air molecules and aerosol particles (haze) determine the albedo of the planet earth. It is the percentage of solar radiation that is turned back to space (the global value of the planetary albedo is approximately 30%). This value may be altered as a function of the surface albedo, the cloud cover and the number of aerosol particles: An increase of the albedo of the planet earth yields an increase of radiant loss of the system earth – atmosphere; a decrease of the albedo yields a decrease of radiant loss and consequently results in heating of the whole system.

b) Absorption and emission of short and long wave radiation by trace elements in the atmosphere cause radiant flux divergences. Taking into account the specific heat of the air as well as its density, it is possible to give a conspicuous illustration of the heating and cooling rates during specific intervals of time.

Freed-back effects between the phenomena mentioned under a) and b) and between other meteorological ones have ordinarily to be expected and e.g. have to be taken into account for investigations into climatic change.

Trace substances are gaseous, solid or liquid particulate matter suspended in the atmosphere; in comparison with the mass of dry air they exist in an extremely small quantity, in general less than 1%. However, they have a great bearing in many fields of meteorology, e.g. in cloud physics and the influence on the radiative, i.e. the energy transfer in the atmosphere which have repercussions on the general circulation. We have to distinguish between trace gases and the solid and liquid traces (aerosol particles). The very differentiated effects they have on the radiant energy balance of the atmosphere are partly still open to question.

The following description of these effects only touches upon the commonly known features as for instance the dual influence of the trace gases ozone, carbon dioxide and water vapour resulting in a heating of the atmosphere through absorption of radiation and cooling through emission.

It is the goal of this paper to deal with the widely ignored effects of the water vapour dimeres and above all of the aerosol particles.

Tracer Gases

Ozone (O_3)

The near ultraviolet solar radiation of wavelengths less than 0.29 μm does not reach the surface of the earth; it is almost completely absorbed by the ozone at the top of the ozone layer (stratopause). The bearing of this absorption process is not so much the decrease of the total of solar radiation reaching the ground – amounting to a few percent only – but it is the strong heating of the stratosphere on the one hand and the shielding of the near ultraviolet radiation from the ground on the other hand. The latter biological aspect is of greatest importance. Ozone has several radiative absorption bands, e.g. the Chappuis bands in the visible absorb some percent of the solar radiation. The infrared bands, e.g. centered at 9.6 μm, can be neglected for considerations of tropospheric radiation.

The Carbon Dioxide (CO_2)

In spite of its absorption bands in the near infrared there is no great influence on the radiant energy balance because the intensity of the solar radiation is rather small in that portion of the solar spectrum.

Whereas CO_2 absorbs the total of electromagnetic radiation emitted from the earth's surface, i e., the terrestrial radiation, at wavelengths greater than 15 μm. On the other hand, this energy is reemitted by the CO_2 downward to the surface of the earth.

This downward flux is temperature – dependent and contributes to the atmospheric counterradiation. This CO_2 absorption ranges in between 12.5 μm and 18 μm; at this upper boundary it coincides with the water vapour absorption. Thus, a change in the CO_2 content of the atmosphere results in a change of the atmospheric counterradiation as well as the terrestrial radiation: An increase of the CO_2 content increases the atmospheric counterradiation and decreases the terrestrial radiation. If this influence of CO_2 is considered individually, it infers a heating of the surface of the earth.

In fact, there is fairly good evidence that the atmospheric CO_2 content has increased by almost 10% since the end of the last century due to the gradual increase in the combustion of fossil CO_2 since the beginning of the large-scale industrialization. A doubling of the CO_2 content would result in an increase of the mean temperature of the surface of the earth by 2°(SMIC, 1971). Such a change in amount would be barely greater than the natural variations due to climatic variations during the last centuries. This, however, holds only under the condition that feed-back processes are not taken into consideration; for a temperature increase due to an increase in CO_2 content would result in changes of the water vapour content as well as the cloud cover. A change in the amounts of these two parameters of the order of magnitude of only 1% may compensate for the temperature increase due to the obvious increase in CO_2 content (Möller 1963). Therefore, it is not admissible to apply merely formal computational results of a general heating of the surface of the earth due to the increase in the CO_2 content to draw conclusions on potential climatic variations. This caution is generally recommended in the consideration of any variations of trace elements in space and time.

The Water Vapour and the Water Vapour Dimeres

Due to its low concentration the water vapour has to be classified under the atmospheric trace gases. In contrast to the CO_2 content, the water vapour content undergoes very great temporal as well as local variations which are induced by meteorological conditions. Furthermore, there is evidence of great regional differences due to climatic peculiarities. Since the water vapour strongly influences the radiant energy balance, the variations in the water vapour content are reflected in very great temporal and local variations of the water vapour induced modifications of the radiation field. The absorption of solar radiation by water vapour is strongest due to the absorption bands between 0.7 μm and 2.7 μm: The amount of incident solar radiation absorbed ranges in between 7% and 20% resulting in a heating of the atmosphere. In addition to that, the terrestrial radiation is greatly attenuated due to the strong absorption bands of the water vapour in the infrared (5 – 8 μm and $\geqslant 13$ μm). Moreover, these water vapour absorption bands jointly with the carbon dioxide absorption band near 15 μm cause the atmospheric counterradiation. This greenhouse effect has a great bearing not only on the microclimate but also on the macroclimate.

Fig. 1 (according to Geleyn and Korb, 1975) gives an example of the cooling rates due to the emission and the absorption by the three main absorbent gases, namely ozone, carbon dioxide and water vapour, in the infrared; the values were based upon (Manabe and Möller, 1961) as well as (Dopplick 1971), however, they have recently been revised (Geleyn and Korb, 1975) in the light of new data and with improved mathematical approaches. It is obvious that within the troposphere, it is mainly the water vapour that effects radiant flux divergences resulting in marked cooling. Whereas the same effect in the stratospheric radiant energy balance is mainly brought about by CO_2 and O_3. It must be emphasized that because of the coincidence of the absorption bands of water vapour and CO_2 near 15 μm the joint effect of the three absorbers ("all absorbents") must not be figured out in an additive way.

Besides these cooling rates, there are heating rates due to the absorption of the incident short-wave solar radiation (Fig. 2). The computations have been based upon average conditions of the trace gases and upon a surface albedo of $A = 0.1$ as well as a solar elevation of 15°; the latter represents the mean solar elevation of the entire globe of the earth during a 24-hour day (Geleyn and Korb, 1975). The albedo plays an important role: It causes the rays to pass through the atmosphere repeatedly due to the reflection by the surface of the earth (prolonged path of absorption). This figure shows that in the troposphere in the short wave portion of the electromagnetic spectrum, too, it is again the water vapour which is the main gaseous absorbent causing temperature variations due to radiant flux divergences.

Not before recently, the importance of a special type of absorption in the window between 8 μm and 12 μm has been disclosed: This absorption depends on the partial pressure of the water vapour e. In Grassl (1974), is given proof of this so-called "e-type-absorption" in the atmosphere by actinometric measurements of the clear sky. The influence on the radiant flux divergences, however, is felt only in the tropics because of their higher water vapour content. E.g. in the latitude of 15° cooling rates within the lowest 1.5 km of the atmosphere of almost 2° per day are brought about amounting to approximately 50% of the total effect of trace elements. The computations for the the Fig. 2 have been basen upon the assumption of rather small water vapour contents ranging between 1.8 cm PPW (surface) and 0.2 cm PPW (at a height of 3 km); the normal values for the tropics range are in between 5 cm PPW and 10 cm PPW. The e-type-absorption shows a linear increase with the water vapour content. The numerical values listed in the table Ia according to Grassl (1974), are valid for average water vapour profiles. The effect of the water vapour dimeres on the radiant energy balance reaches its maximum in the tropics due to their high water vapour content.

By taking into account the absorption by water vapour dimeres it was possible to clarify two hitherto discrepancies in the behaviour of two quantities which are of importance for the heating balance of the atmosphere: At first the great deviations between the measured and calculated long wave radiant flux divergences in the lowest kilometers of the tropical atmosphere; secondly, the actinometric measurements in the window between 8 μm and 12 μm in the tropics by means of satellites yield surface temperatures which are by several degrees Centigrade lower than those obtained by direct measurements, due to the influence of the water vapour dimeres. It is presumed that this absorbent is a dimerous water vapour molecule $(H_2O)_2$, therefore the term "dimere water vapour absorption".

The Remainder of Trace Gases

These trace gases have absorption bands at various wave lengths, however, their absorptivity is so ineffective that their influence on the radiant energy balance can be neglected.

But these trace gases influence the radiant energy balance indirectly in two ways: a) N_2O, CO and H_2 influence the ozone balance, b) sulfur dioxide, SO_2, is converted by oxidation into sulfate particles. The latter are important constituents of the atmospheric aerosol; their influence on the radiant energy balance will be discussed in the next paragraph.

Trace Substances in the Form of Aerosol Particles

Absorption or emission by aerosol particles takes place within the entire electromagnetic spectrum from 0.3 μm up to about 20 μm as it is shown in Fig. 3 according to Fischer (1973), Fischer (1975) and Thudium (1975). Many papers, e.g. Bullrich *et al.* (1973), deal with the number, size, chemical structure, physical properties and optical characteristics of the aerosol particles as functions of the relative humidity.

The following discussion of the influence of the aerosol particles on the radiant energy balance refer to the measurements of average data which characterize specific average turbidity conditions of the atmosphere.

An appropriate measure of the atmospheric turbidity is Linke's turbidity factor T with regard to the wave length 0.55 μm: $T = \frac{a_A + a_R}{a_R}$ with a_R representing the extinction of light of the whole atmosphere by the air molecules and a_A representing the extinction of light by the aerosol particles.

The computations have been based upon the following assumptions: The real part n of the complex index of refraction m being 1.5 and the imaginary part k characterizing the absorption being 0.01 ($m = n - ki$).

The influence of the Aerosol Particles on the Short Wave Spectrum

Some years ago it has been postulated that there was one more absorbent constituent of the atmosphere whose absorptivity in the solar spectrum almost equaled that of the water vapour. This assumption has been confirmed: The additional absorbent is the aerosol.

The problem has two aspects:

a) The aerosol particles absorb radiation thus effecting radiant flux divergences which can be expressed as heating rates per day. This heating increases with the increase of aerosol content (turbidity and k).

b) The aerosol particles scatter radiation in all directions, also back to space. The latter portion increases with increasing turbidity; this would result in a cooling effect of the whole system earth-atmosphere, however, this is counteracted by the aerosol absorption thus diminishing the portion of radiant energy scattered back to space through the upper edge of the atmosphere. This counteraction increases with increasing turbidity and increasing surface albedo both of which augment the number of multiple scattering processes thus increasing the absorption: The final result is a decrease of the albedo of the planet Earth.

There are well known papers (Mitchell, 1971, Yamamoto and Tanaka, 1972) describing the computations following these aspects. New quantitative results on this interaction Eschelbach (1973) have been obtained by solving the equation of radiative transfer and by applying newly measured values of the absorption of radiation by aerosol particles.

Fig. 4 illustrates the aspect a): It shows the radiant flux divergences expressed as daily heating rates due to the aerosol absorption in the wave length range from 0.4 μm through 0.8 μm for average aerosol, i.e. average turbidity conditions. The computational results are valid for a day in the last third of April at 50°N. Near the surface, the heating amounts to 0.6° per day. There can be an increase by the factor 2 with increasing turbidity. The computations covering the entire solar spectrum have proved that the aerosol absorption is of the same order of magnitude as the water vapour absorption. This has been indicated in the Fig. 2.

Fig. 5 illustrates the aspect b): It shows the planetary albedo R as a function of the turbidity factor T in the visible spectrum ($\lambda = 0.55$ μm) for a zenith distance of the sun, $\vartheta = 37°$, and for three different values of surface albedo in the case of a cloudless sky. The computations have been based upon a mean imaginary part of the complex index of refraction (which characterizes the absorption) $k = 0.01$ (Fischer, 1975); this mean value has been obtained from measurements.

It can be seen that if the surface albedo A is missing (A = O), the planetary albedo R slightly increases in spite of increasing turbidity. However, as soon as with A = 0.18, the planetary albedo R starts decreasing with increasing turbidity. Higher values of the surface albedo A, e.g. A = 0.8, markedly reduce the planetary albedo R due to the aerosol absorption, i.e. the portion of incoming solar radiation that is scattered back to space by the aerosol particles is decreased with increasing surface albedo.

The Influence of the Aerosol Particles on the Infrared Radiation

The above described absorption of short wave radiation by the aerosol particles results in a decrease of the planetary albedo on the one hand and a heating of the atmosphere on the other hand. Recent investigations (Fischer and Grassl, 1975) (Volz, 1972) have revealed that there is also radiant emittance effected by the aerosol particles due to their strong absorption bands in the window near 9 μm. Thus, the above discussed heating effect is counteracted by atmospheric cooling because of this radiant emittance. The latter effect has been taken into account for average aerosol conditions in the Fig. 2 and Table 1b.

The Net Radiation Effected by the Aerosol Particles

The above described counteraction of heating and cooling due to absorption as well as emission of radiation results in a net effect as follows: The computations published in Fischer and Grassl (1975), which are valid for average aerosol conditions ($T = 2$, $k = 0.01$) and for a relative humidity of 35% in the geographical latitude of 50°, yield a net effect of 0.2° per day. This is a small amount, and in humid tropical regions it is negligible in comparison with the absorption effect by the water vapour dimeres. In middle and northern latitudes, however, it can be estimated that a possible increase of turbidity and aerosol absorption would enhance the net heating to become approximately 1° per day.

Summing up, the statement can be made that the influence exerted by the aerosol particles –integrated over all latitudes– increases with increasing turbidity being especially enhanced when the surface albedo surpasses the value of 18%. In most of the cases, the influence of the aerosol particles effects a heating of the atmosphere and a simultaneous slight cooling of the surface. Is is therefore deemed important to control both the increase of the aerosol content as well as of the surface albedo with special emphasis to a possible man made influence.

The Influence of the Aerosol Particles on the Radiant Energy Balance of Clouds

The influence of aerosol particles suspended within clouds is of both direct and indirect nature: At first, an increase in the number of aerosol particles in clean air increases the number of small droplets; over the continents this is evident from the increase in the reflectivity of the clouds, i.e. the clouds are shining. Secondly, an increase in the number of aerosol particles increases the multiple scattering within the clouds, i.e. the paths of the light rays are prolonged considerably resulting in a considerable increase of absorption by the aerosol particles. Therefore, the influence of both the cloud droplets as well as the aerosol particles results in a temperature increase. The absorption within the clouds sets an upper limit to the albedo of clouds of about 0.8.

Computations valid for average solar elevations and average aerosol content yield heating rates of approximately 1.5° per day within a stratus cloud of a thickness of 1000 m, according to Geleyn and Korb (1975) as it can be seen in the Fig. 6. In this figure, the ordinate to the left represents the vertical distribution of the relative humidity. The abscissa represents the heating rates per day.

The curves are valid for a stratus cloud of a thickness of 1 000 m without and with taking into account the aerosol absorption for two different zenith distances of the sun, namely $\vartheta = 60°$ and $\vartheta = 72.5°$; while the surface albedo A = 0.1 and the liquid water content of the cloud is $\rho^{(2)} = 0.01$ g per cm^3 according to Geleyn and Korb (1975).

It is common pratice to introduce for the computation the "single scattering albedo" ω_o, which is the ratio of radiative scattering a_s to radiative extinction $a = a_s + a_a$ of the atmospheric aerosol particles. Hence, with a_a representing the radiative absorption,

$$\omega_o = \frac{a_s}{a_s + a_a}$$

If there is no absorption, $\omega_o = 1$.
The Table 2 according Grassl (1975), shows that a decrease of ω_o, that means an increase of T and k which is not uncommon over the continents, can raise the heating to 4° per day.

It is not yet known what further indirect implications on the cloud development might be derived from this heating in the case of increasing relative humidity.

The Role that the stratospheric Aerosol Plays in the Radiant Energy Balance

Conspicuous anomalous coloured luminosities observed in the western twilight sky as for instance the purple light (reddish afterglow), indicate even to the naked eye that there must be haze in the stratosphere, i.e. that quite a number of aerosol particles must be suspended in the upper atmosphere. This number can be assumed to be multiplied, e.g. after volcanic eruptions; this reduces the radiative transmissivity, often for several years, because the aerosol particles stay much longer in the stratosphere than in the troposphere. However, noteworthy losses of solar radiation reaching the ground occur only either due to several consecutive severe volcanic eruptions or due to a giant eruption like that of the Krakatao. As for the stratosphere itself, the eddition of aerosol particles always effects a heating due to radiative absorption, in the short wave as well as the long wave spectrum.

Table 1a

pressure	75°N		45°N		15°N	
in mb	in °C/day	in % of total	in °C/day	in % of total	in °C/day	in % of total
990	0.008	3.1	0.56	28.7	2.26	61.0
900	0.009	1.1	0.31	22.8	1.77	54.0
700	0.008	0.7	0.05	4.2	0.29	16.1

Table 1 b

990	0.069	26.5	0.125	6.4	negligible	
900	0.049	6.1	0.060	4.4		
700	0.008	0.7	0.015	1.3		

Cooling rates in the window spectral region as percentage of the total cooling.
1 a e-type (water vapour dimeres)-absorption.
1 b aerosol, turbidity factor T = 3, according to Grassl (1974).

Table 2

ϑ	$\omega_0 = 0.999$	*0.998*	0.996	0.995
60°	0.55	1.10	2.22	2.80
53°	0.70	1.33	2.62	3.30
37°	0.90	1.89	3.75	4.70

Heating rates per day within a stratus cloud according to Grassl (1975).
The value of $\omega_0 \approx 0.998$ represents the normal conditions.
The value of $\omega_0 = 0.995$ is valid for regions of severe air pollution.
ϑ zenith distance of the sun.

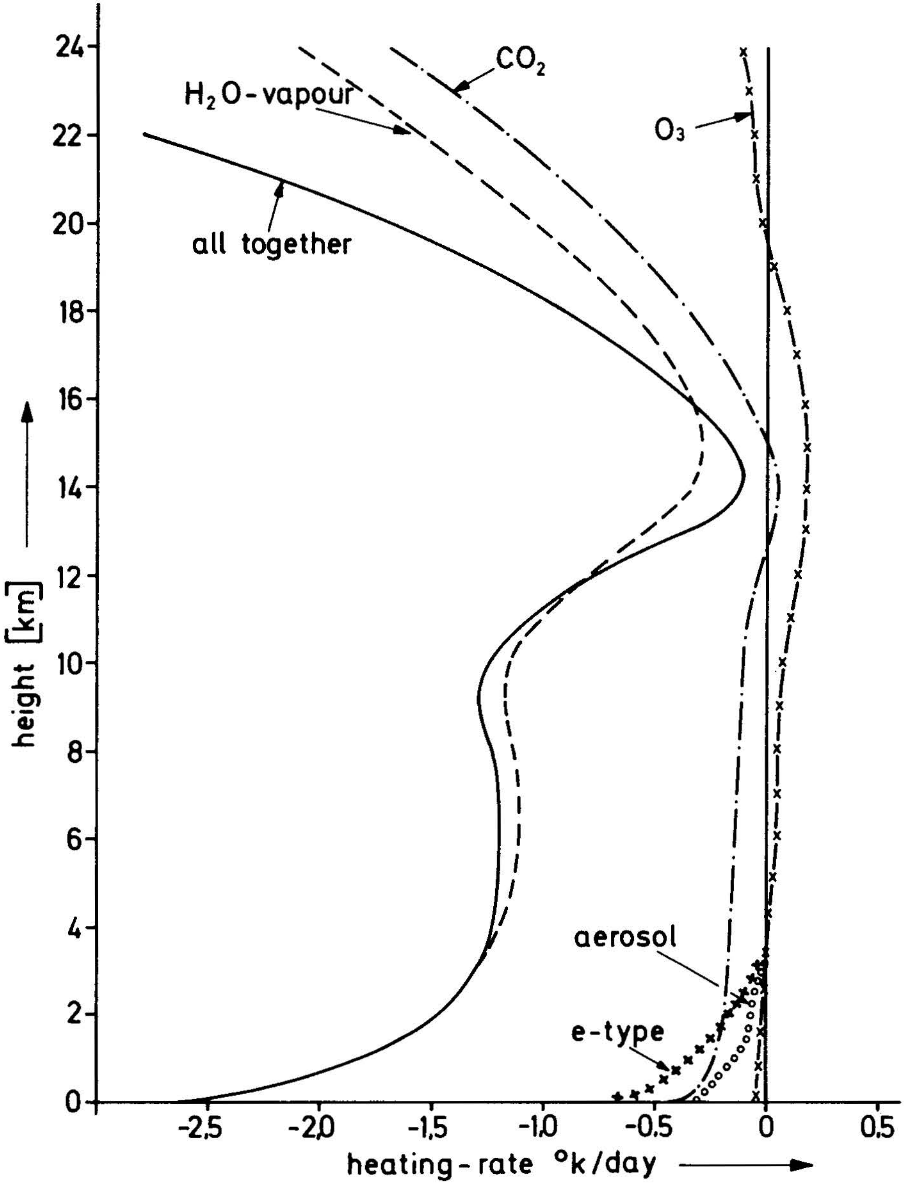

Fig. 1. Long wave radiant flux divergence in °K per day in a cloudless atmosphere caused by the trace gases water vapour, CO_2 and O_3. The effect of the water vapour dimere (e-type)-absorption and the aerosol absorption in the lower atmosphere is included. Water vapour content near the surface 1.8 cm ppw, in 3000 m altitude 0.5 ppw.

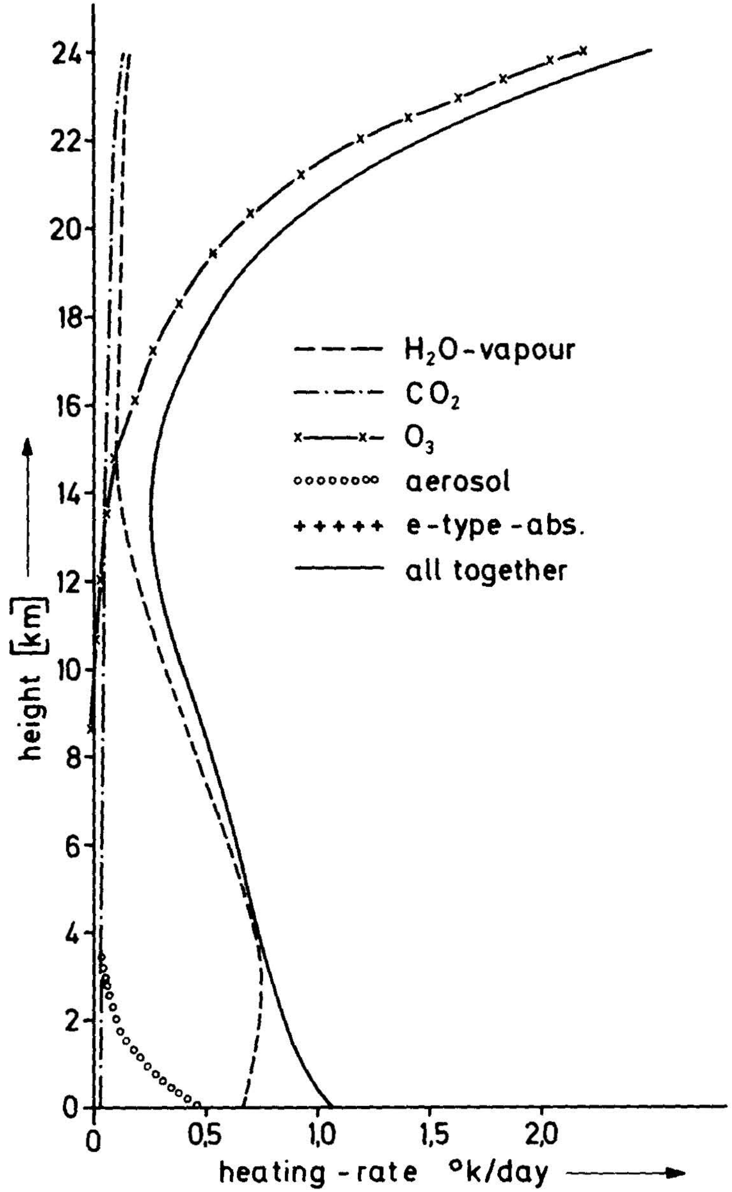

Fig. 2. as Fig. 1, but short wave radiant flux divergence. Zenith distance of the sun $\vartheta = 75°$, surface albedo A = 0.1, according Geleyn and Korb (1975).

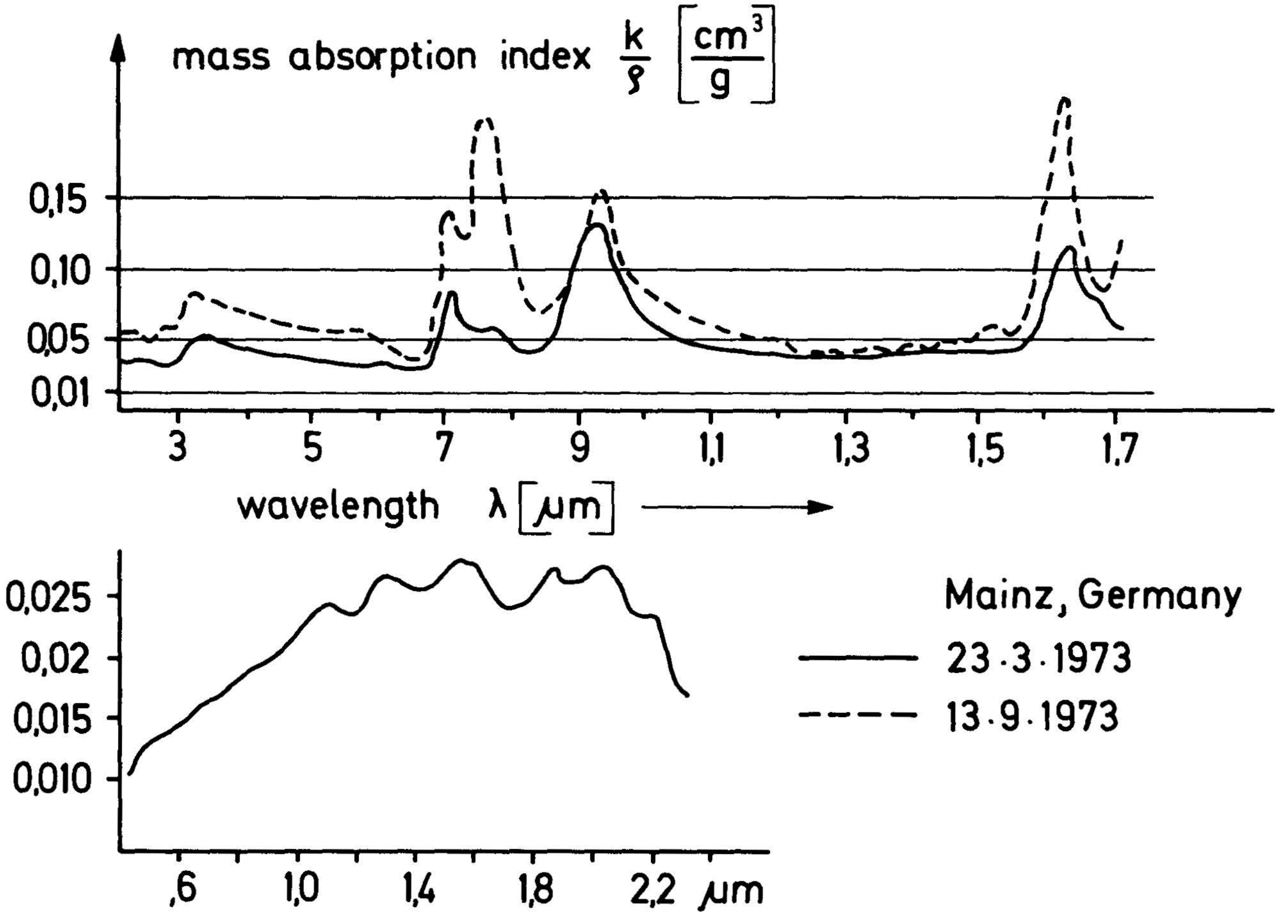

Fig. 3. Mass absorption index $\frac{k}{\rho}$ vs wavelength of aerosol particles sampled by impactor at Mainz, according Fischer (1973 and 1975). ρ density of the aerosol particles, Measurements according to (16), $\rho \approx 2.2$.

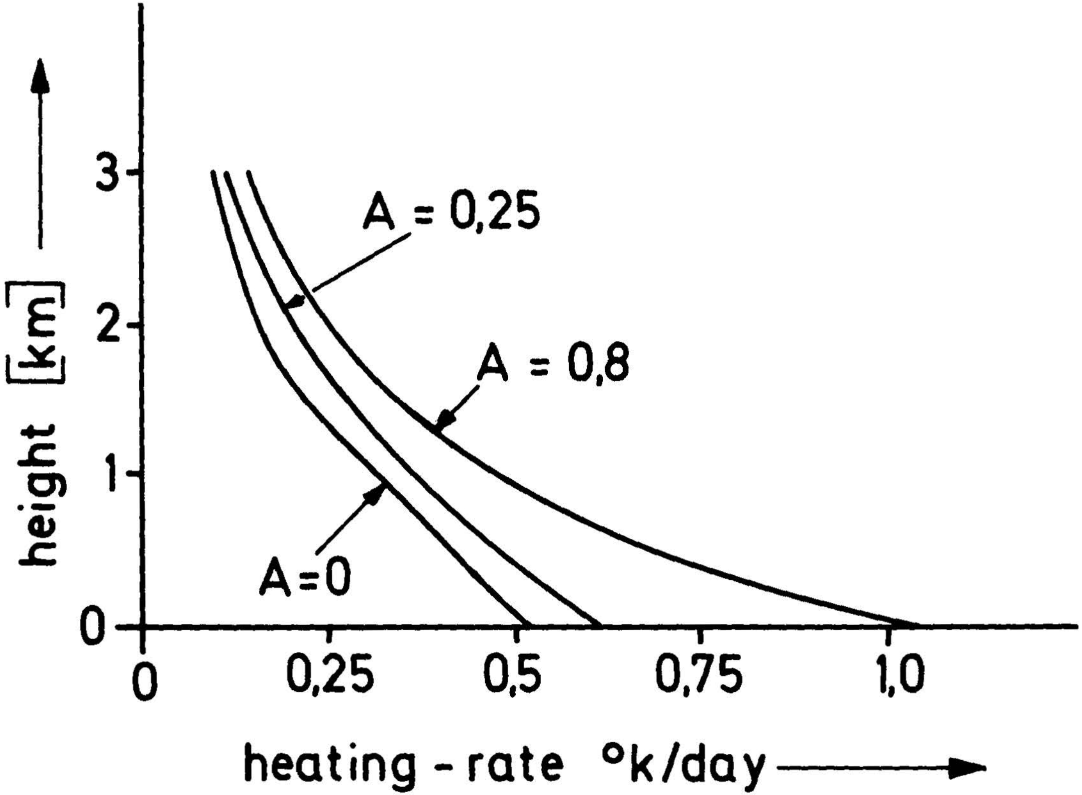

Fig. 4. Absolute heating for a day in the last third of April at 50° north latitude as function of the height in km. Parameters: $m = 1.5 - 0.02\ i$, $T = 4$, $A = 0, 0.25, 0.8$. The spectral interval $0.45 < \lambda < 0.85\ \mu m$, according Eschelbach (1973).

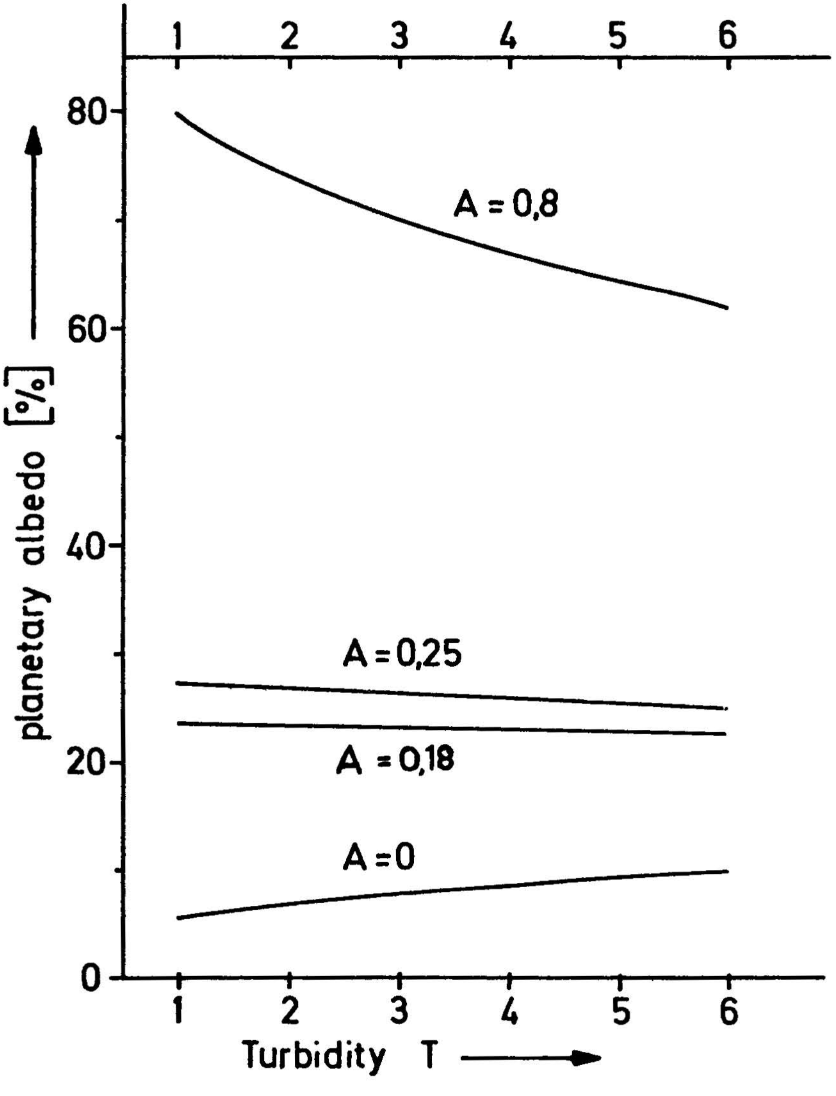

Fig. 5. Planetary albedo R in % as function of turbidity factor T. Wavelength λ = 0.55 μm, m = 0.15 – 0.02 i.
Surface albedo A = 0, 0.18, 0.25, 0.8, zenith distance of the sun ϑ = 37°, according Eschelbach (1973).

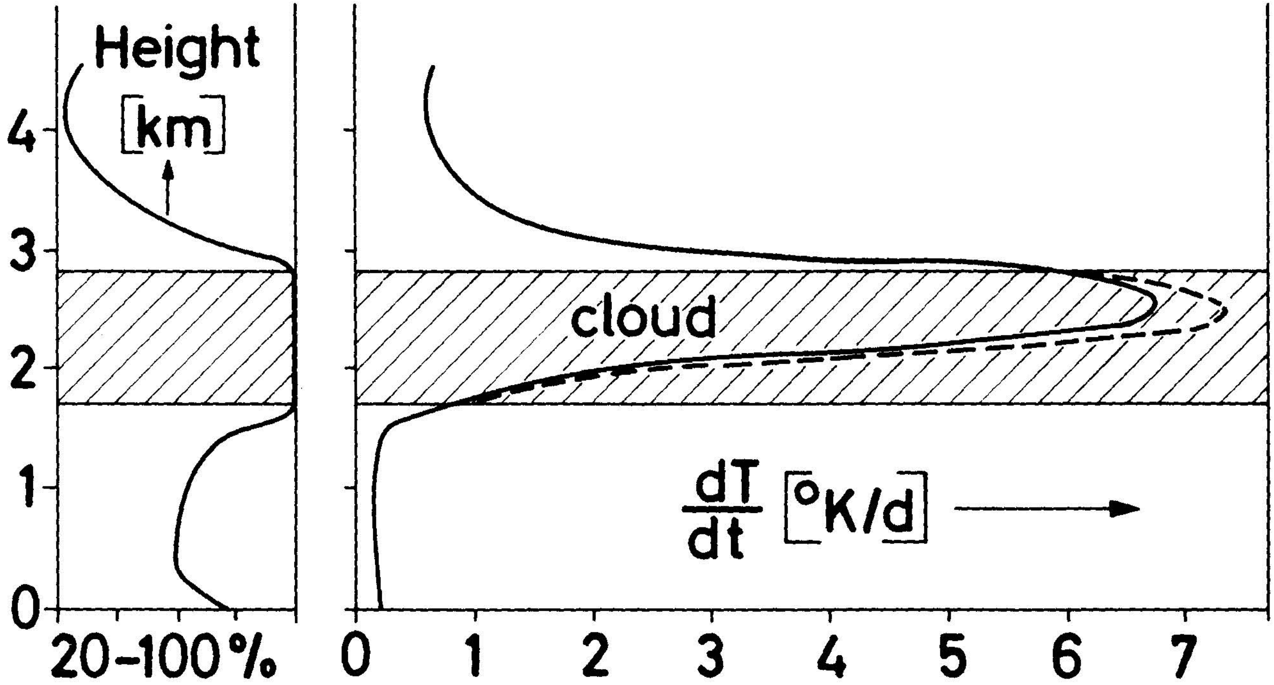

Fig. 6. Absorption of solar radiation in clouds.
Left: relative humidity, right: heating °K/day.
— no aerosol absorption k = O;
- - -Aerosol absorption k = 0.01, ω_o = 0.999, A = 0.1, $\rho^{(2)}$ = 0.01 g cm^{-3}, according Geleyn and Korb (1975).

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