# ON THE ESTIMATION OF THE EFFECTS OF CLOUDS AND VOLCANIC ERUPTIONS IN THE CALCULATION OF THE CHARACTERISTICS OF UV AND VISIBLE RADIATION FIELDS IN RADIATION-CONVECTIVE AND PHOTOCHEMICAL MODELS OF THE ATMOSPHERE

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*A simple and efficient parameterization of clouds that makes it possible to calculate the solar radiation fluxes and photodissociation constants with the help of any two-flux method for given cloud optical thickness ($\tau_c \geq 8$) or cloud albedo is proposed. The changes in the flux and photolysis constants in the period after a volcanic eruption as well as in the presence of clouds are estimated for a wide spectrum of values of the sun's zenith angle, cloud albedo, and underlying surface.*

## 1. INTRODUCTION

The calculation of the solar radiation fluxes in the UV and visible (UVV) ranges with detailed spectral resolution is an integral part of radiation-convective and photochemical models of the atmosphere (RCMA and PCMA). Different modifications of the two-flux method, which combines speed with accuracy and versatility, are employed for this purpose.

Recent investigations[1–3] show that the gas composition and thermal regime in different layers of the atmosphere are substantially altered by clouds and the product of volcanic eruptions. The character of these changes is in many ways connected with the transformation of the UV and visible radiation fields, which determine how quickly the atmosphere is heated as well as the photodissociation constants of the optically active minor gaseous components of the atmosphere.

The question of accounting for cloud and volcanic nonuniformities when solving the equation of radiation transfer in the atmosphere was studied in Refs. 1, 2, and 4. In this paper we obtain a number of estimates of the effect of such nonuniformities on solar radiation fluxes and the rates of some key photodissociation reactions.

## 2. CHARACTERISTICS OF UV RADIATION TRANSFER IN THE POSTVOLCANIC PERIOD

The transport of large quantities of sulfur dioxide ($SO_2$) up to altitudes of 20–30 km has been found to be one of the important factors which, together with the increase in the aerosol optical thickness, are responsible for the disturbance of the optical state of the stratosphere after a volcanic eruption. It is well known that this compound strongly absorbs radiation at wavelengths below 310 nm. In our calculations the volcanic cloud was simulated by specifying constant mixing ratios equal to 0.05 and 0.1 ppm in the layer 20–30 km. The numerical experiments performed (Fig. 1) make it possible to draw a number of conclusions regarding the character of the photodissociation of optically active gases in the post eruption period.

1. The transport of sulfur dioxide up to stratospheric altitudes is important for the photodissociation regime of gases that are optically active at wavelength below 310 nm.

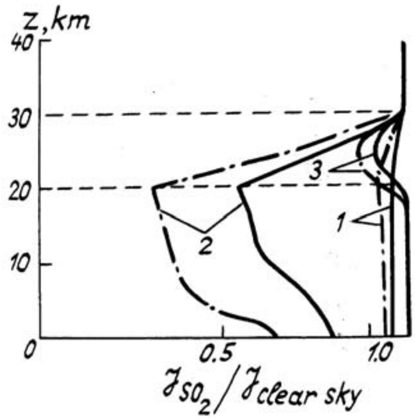


*FIG. 1. The photodissociation constants in the presence of a volcanic cloud with respect to the values for a purely molecular atmosphere. The volcanic cloud occupies the layer 20–30 km. The mixing ratios of $SO_2$ are 0.05 ppm (solid line) and 0.1 ppm (dot-dashed line).*
*1) The reaction $O_3 + h\nu \rightarrow O_2 + O(^1D)$;*
*2) the reaction $O_2 + h\nu \rightarrow O + O$;*
*3) the reaction $HNO_3 + h\nu \rightarrow OH + NO_2$.*

2. The values of the photolysis are most strongly affected in the volcanic layer 20–30 km (Fig. 1). Below this layer, however, this effect

diminishes rapidly owing to the altitude redistribution of the contribution of different parts of the spectrum to photodissociation so as to increase the relative role of quanta with $\lambda \geq 300$ nm, where absorption by $SO_2$ drops rapidly.

## 3. CHARACTERISTICS OF THE TRANSFER OF UW RADIATION IN THE PRESENCE OF CLOUDS

**Cloud parameterization**. In the standard method of calculating the UVV solar radiation fields in the atmosphere in the presence of aerosol, including cloud aerosol, the physical and optical characteristics of the aerosol particles — size distribution, composition, etc. — are taken into account in solving the radiation transfer equation.[5,6] This approach has the drawback that it is unwieldy. The dependence of the radiation field on the parameters of the cloud particles is very complicated, and this makes it difficult to gain a qualitative understanding of the character of radiation effects of clouds. In addition, this approach cannot be used to take clouds into account within the framework of the Isaksen-Luther method,[7,8] often employed in the radiation blocks of modern PCMA and RCMA.

In many cases a different technique, in which the cloud layer as a whole is parameterized, is preferable. In this approach a single macro-characteristic is employed: the cloud albedo $A_c$.

The parameterization proposed in this work is based on the ideas incorporated in Refs. 2 and 9. Following Refs. 2 and 9, we shall regard a cloud as an infinite layer confined between the levels $i_c$ and $i_c + 1$ of a plane-parallel atmosphere. As the calculations of Ref. 9 show, the albedo of a cloud whose optical thickness is fixed increases as the angle of incidence of the initial flux of solar radiation increases. For this reason we introduce, as done in Ref. 9, the albedo $A_c^0$ for the direct flux, determined by the sun's zenith angle $\Theta_0$, and the albedo $A_c^*$ for the diffuse radiation.

We shall define the reflectance and transmittance of the cloud layer for direct and diffuse flux as follows:

$$R_c^0 = A_c^0 r_{i_c}; \quad (1)$$

$$R_c^* = A_c^* r_{i_c}; \quad (2)$$

$$T_c^0 = (1 - A_c^0) t_{i_c}; \quad (3)$$

$$T_c^* = (1 - A_c^*) t_{i_c}, \quad (4)$$

where the factor $r_{i_c} = t_{i_c} = e^{-\tau_{a,i_c}/\bar{\mu}}$ describes the absorption by atmospheric gases in the layer $(i_c,\ i_c+1)$; in the near-UV and visible region $\tau_{a,i_c}$ is determined by the optical thickness of ozone $\tau_{a,i_c} = \tau_{O_3,i_c}$; and, $1/\bar{\mu} = 1.66$ is the average secant (diffuseness factor).

Any two-flux method (for example, the δ-Eddington method,[10] the Isaksen-Luther method,[7,8] etc.) can be used to calculate the solar radiation fields in the presence of clouds.

The reflectances and transmittances $R_i^0$, $R_i^*$, $T_i^0$, $T_i^*$ for $i > i_c$ as well as $R_i^*$ and $T_i^*$ T for $i < i_c$ can be calculated for a purely molecular atmosphere, while the coefficients $R_{i_c}^0$ $R_{i_c}^*$, $T_{i_c}^0$, $T_{i_c}^*$ can be found from expressions (1)–(4) (the δ-Eddington and other methods), or from analogous relations for the actinic fluxes (Isaksen-Luther method):

$$\tilde{R}_c^0 = 2\mu_0 R_c^0; \quad (5)$$

$$\tilde{R}_c^* = R_c^*; \quad (6)$$

$$\tilde{T}_c^0 = 2\mu_0 T_c^0; \quad (7)$$

$$\tilde{T}_c^* = T_c^*; \quad (8)$$

where $\mu_0 = \cos\theta_0$ and $\theta_0$ is sun's zenith angle.

To calculate the fluxes below the clouds ($i < i_c$) the fact that, as shown in Ref. 9, the radiation below an optically thick ($\tau_c \geq 8$) cloud can be regarded as purely diffuse, i.e., for $i < i_c$ the direct fluxes $F_i^0 = 0$, must be taken into account.

**Discussion.** The effect of clouds on the solar radiation flux as estimated with the help of the above-described Isaksen-Luther parameterization for clouds in an absorbing and scattering atmosphere can be compared with the help of Fig. 2 with the results of Ref. 9 which were obtained in the approximation of a conservative (purely scattering) isolated (the atmosphere is optically transparent) cloud by the δ-Eddington method starting from the parameters of a cloud aerosol.

In the visible part of the spectrum the agreement id virtually complete (the discrepancy for small values $A_c^0$ in Fig. 2c can be explained by the fact that the parameterization of the cloud in calculating the subcloud fluxes is not suitable in the case of an optically thin cloud in calculating the subcloud fluxes is not suitable in the case of an optically thin cloud ($\tau_c \leq 8$; see Ref. 9)). As the wavelength decreases the optical thickness of the atmosphere becomes significant, and this results in a significant reduction of the fluxes, for $A_g = 0$, however, the UV flux above an optically thin ($A_c^0 \leq 0.175$) cloud is somewhat higher, owing to scattering, that the fluxes in the visible region of the spectrum (Figs. 2a and 2b).

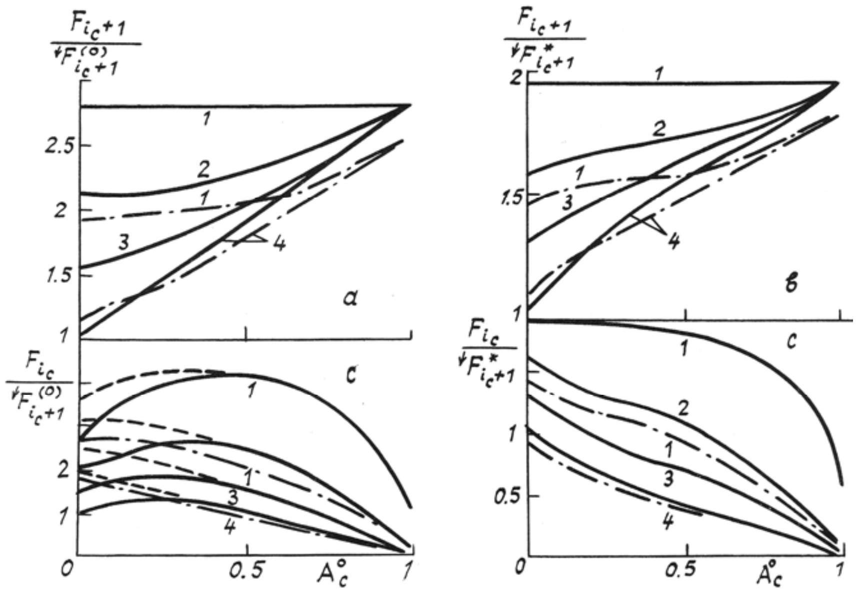

*FIG. 2. The total monochromatic actinic fluxes above and below a cloud with respect to the starting flux, $\theta_0 = 20$, as a function of the cloud albedo $A_c^0$ for underlying surface albedo $A_g = 0.95$ (1), 0.6 (2), 0.3 (3), and 0 (4). a) above the, cloud, the initial flux is collimated; b) above the cloud, the initial flux is isotropic; c) below the cloud, the initial flux is collimated; d) below the cloud, the starting flux is isotropic. The solid lines show the results of Ref. 9 (conservative insulated cloud; δ-Eddington method); the dashed curves ($\lambda = 550$ nm) and dot-dashed curves ($\lambda = 290$ nm) show the results of this work. In Figs. 2a, 2b, and 2d the solid and dashed lines coincide.*

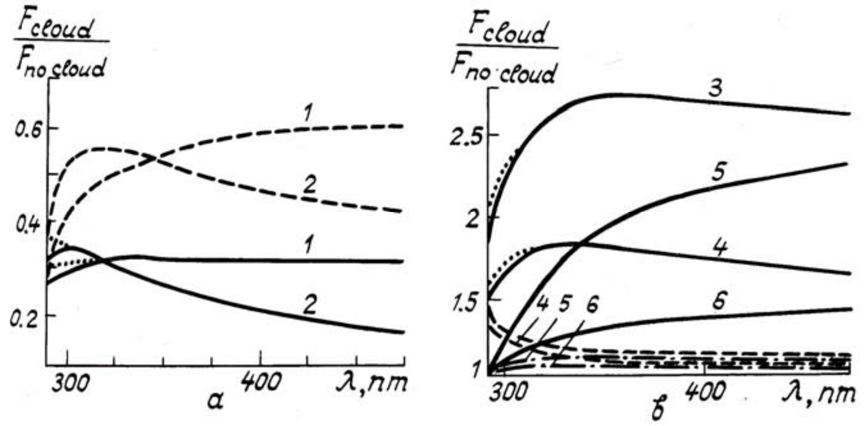

*FIG. 3. The total monochromatic actinic fluxes above and below a cloud with respect to fluxes in a clear atmosphere as a function of the wavelength with $A_c^0 = 0.8$ below (a) and above (b)* the cloud. *1 — $z = 0$, $\theta_0 = 20°$; 2 — $z = 0$, $\theta_0 = 70°$; 3 — $z = 2$, $\theta_0 = 20°$; 4 — $z = 2$, $\theta_0 = 70°$; 5 — $z = 10$, $\theta_0 = 20°$; 6 — $z = 10$, $\theta_0 = 70°$; $A_g = 0$; (solid line), 0.75 (dashed and dot-dashed lines), and* 0 *(dotted lines); the ozone concentration in the cloud layer is 50% lower than outside the cloud layer*

Figure 3 shows the ratio of the fluxes in the presence of a cloud to the fluxes in a clear atmosphere for $z = 0$ km (the underlying surface), $z = 2$ km (the top boundary of the cloud), and $z = 10$ km as a function of the wavelength of the radiation. At $z = 0$ km, $\theta_0 = 20°$ (curve 1) as well as $z = 10$ km (5, 6) the spectral behavior of the flux ratios is easily explained by the increase in the absorption in the UV range. The situation is somewhat more complicated, however, for the ratios of the subcloud fluxes at $\theta_0 = 72°$ (2) and the fluxes at the top boundary of the cloud (3, 4). To illustrate the situation more clear we shall consider the case $A_g = 0$.

The weak maximum (curve 3) at $\lambda \sim 350$ nm is explained by the contribution of fluxes reflected by the cloud and scattered in the atmosphere above the cloud. The maxima 2 and 4 at $\lambda \sim 330$ nm are of special interest. The fact that the cloud can attenuate the UV fluxes more effectively than fluxes in the visible part of the spectrum was established by comparing the ratios of the constants of the photolysis reaction $O_3 + h\nu \rightarrow O_2 + O(^1D)$ ($J^*(O_3)$) and $NO_2 + h\nu \rightarrow NO + O$($J(NO_2)$) for an atmosphere with and without clouds by both computational[6] and experimental[11] methods. The parameterization given by the expressions (5)–(8) makes it possible to explain this clearly and simply.

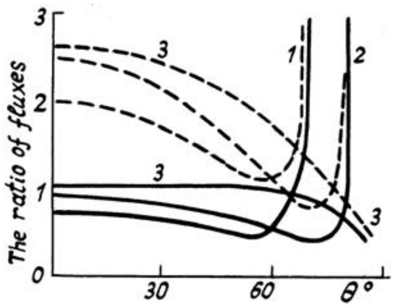


*FIG. 4 The ratio of the total monochromatic actinic fluxes at the underlying surface to the fluxes in a purely absorbing atmosphere as a function of the sun's zenith angle. $\lambda$ = 300 nm (1), 310 nm (2), and 550 nm (3). $A_g$ = 0 (solid lines) and 0.75 (dashed lines).*

One can see from Fig. 4 that in the troposphere the direct radiation makes the main contribution to the fluxes in the visible part of the spectrum while fluxes in the near-UV region are virtually purely diffuse.

Thus for an optically thick cloud, zero underlying surface albedo, and large values of $\theta_0$ the ratios of the fluxes at the underlying surface can be written as follows:

$$k_{UV/B,0} \equiv \frac{\left[F_{0(\text{cloud})} / F_{0(\text{no cloud})}\right]_{UV}}{\left[F_{0(\text{cloud})} / F_{0(\text{no cloud})}\right]_{B}} \cong \frac{\left[\tilde{T}_c^*\right]_U}{\left[\tilde{T}_c^0\right]_B} \cong \frac{1 - A_c^*}{2\mu_0\left[1 - A_c^0\right]}. \quad (9)$$

Since for large $\theta_0$: $2\mu_0 < 1$ and $A_c^0 > A_c^*$ the ratio (9) is always greater than unity in this case.

Analogously the ratios of the fluxes above the cloud can be written as follows:

$$k_{UV/B,i_c+1} \equiv \frac{\left[F_{i_c+1(\text{cloud})} / F_{i_c+1(\text{no cloud})}\right]_{UV}}{\left[F_{i_c+1(\text{cloud})} / F_{i_c+1(\text{no cloud})}\right]_{B}} \cong \frac{1 + \left[\tilde{R}_c^*\right]_{UV}}{1 + \left[\tilde{R}_c^0\right]_B} \cong \frac{1 + A_c^*}{1 + 2\mu_0 A_c^0}. \quad (10)$$

For large $\theta_0$ this relation is likewise somewhat greater than unity. Thus for $\theta_0 = 70°$, $A_g = 0$, and $A_c^0 = 0.8$ the expression (9) and (10) give the values $k_{UV/B,0} = 1.9$ and $k_{UV/B,i_c+1} = 1.1$ (compare with the values obtained from computer calculations of the flux ratios under the same conditions: $k_{300/550.0} = 2.1$ and $k_{300/550.2} = 1.1$). The decrease in the flux ratios for $\lambda \le 300$ nm is caused by a sharp increase of the absorption by ozone in this part of the spectrum. However the real values of the fluxes at $\lambda \sim 300$ nm could be somewhat higher because of the reduction in the ozone concentration owing to the heterogeneous annihilation of ozone on cloud particles, which, according to the estimates of Ref. 12, reaches 50% (see Fig. 3).

The results of comparing the effect of the clouds on the rate of photolysis of $NO_2$, which dissociates primarily in the short-wavelength part of the visible range, and $O_3$, which in the troposphere dissociates into $O(^1D)$ atoms owing to the near-UV radiation, completely agree with the flux characteristics found above (Fig. 5). At small zenith angles $[J(NO)]_{\text{cloud}} / [J(NO_2)]_{\text{no cloud}}$, is greater than $[J^*(O_3)]_{\text{cloud}} / [J^*(O_3)]_{\text{no cloud}}$ in the entire range of altitudes, while for large $\theta_0$ the reverse ratio holds below the cloud and near its top boundary. The reduction of the ozone content can increase insignificantly the value of $[J^*(O_3)]_{\text{cloud}}$ (Fig. 5).

## 4. CONCLUSIONS

The possibility of including cloud-induced and postvolcanic radiation effects in two-flux methods for calculating the UVV solar radiation fields in the atmosphere, which are employed in RCMA and PCMA, was studied. A parameterization, whose chief attributes are its simplicity and versatility and which makes it possible to calculate the fluxes and the photodissociation constants with the help of any two-flux method with fixed optical thickness ($\tau_c \ge 8$) or cloud albedo, was proposed for the cloud layer.

The main results can be formulated as follows:

– the sharp increase in the sulfur dioxide concentration at stratospheric altitudes during the postvolcanic period can result in a significant reduction of the photolysis rates for gases that are optically active at $\lambda \le 3000$ nm within the volcanic cloud, but below this layer the effect rapidly diminishes;

– the changes in the solar radiation fluxes and the photodissociation constants in the presence of clouds were estimated for different values of the sun's zenith angle $\theta_0$ as well as the cloud albedo and underlying surface albedo; and,

– it was shown that clouds affect differently the solar radiation fluxes in the UV and visible spectral ranges; namely, it was found that both near the top boundary of the cloud and below the cloud for small $\theta_0$ ($\theta_0 \le 70°$) the ratio $F_{cloud}/F_{nocloud}$ was smaller for the UV radiation, while for large $\theta_0$($\theta_0 \ge 70°$) the ratio was smaller for the visible radiation.

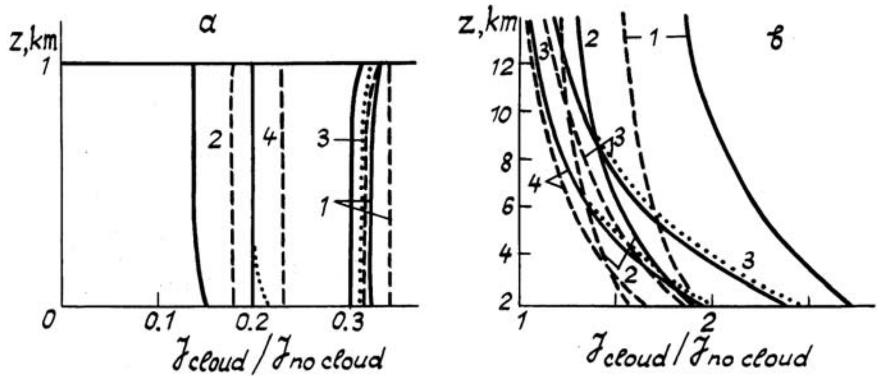


*FIG. 5. The ratio of the photodissociation constants above and below a cloud to the their values for a clear atmosphere, $\tau_c$ = 64, below (a) and above (b) the cloud. 1 – $\theta_0$ = 20°; $A_c^0$ = 0.83, J($NO_2$); 2 – $\theta_0$ = 70°; $A_c^0$ = 0.89, J($NO_2$); 3 – $\theta_0$ = 20°; $A_c^0$ = 0.83; J*($O_3$); 4 – $\theta_0$ = 70°; $A_c^0$ = 0.89, J*($O_3$). $A_g$ = 0 (solid line), 0.25 (dashed line), 0 (dotted line); the zone content in the cloud layer is 50% lower than outside the layer.*

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