



A proposal on accounting for the non-radiative heat fluxes in the atmospheric transfer of thermal photons

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The key parameter for radiative transfer is the *mean value* of photons' free path length and not its *variance*, which can therefore be formally set to zero. Then the photon mean free path length l counted from an arbitrary point to the point of the next collision is related to the mean free path length l_c between two consequent collisions as $l = l_c/2$. If one divides the atmosphere into n adjacent layers of thickness l_c each, then photons absorbed within layer k ($1 \leq k \leq n$) are only those photons that were emitted within the upper $(k - 1)$ -th and lower $(k + 1)$ -th neighbouring layers. A photon emitted at point z' within layer k ($0 \leq z' \leq l_c$) will reach layer $(k - 1)$ if only the angle between the vector of its propagation and the vertical axis does not exceed ϑ , where $\cos \vartheta = z'/l_c = \mu$. Radiative flux F_k emitted by layer k to the upper (+) and lower (-) hemispheres is $F_k^\pm = 2\pi I_k l_c \int_0^1 \mu d\mu = \pi I_k l_c$, where I_k is the intensity of isotropic radiation of a unit volume of the greenhouse substance in a unit solid angle. But only two thirds of this flux, $F_k \int_0^{l_c} dz' \int_{z'/l_c}^1 \mu d\mu = (2/3)F_k$, reach the neighbouring layer $(k - 1)$ and are absorbed there. The energy conservation law for layer k can be written as $H_{k+1} - H_k = -A_k$, $H_k \equiv (2/3)(F_k^+ - F_{k-1}^-)$, where $A_k > 0$ is the power of latent and sensible heat transformed into thermal photons within layer k . In the continuous representation in terms of optical depth $\tau = k/2$ ($\tau_s \equiv n/2$) these equations become $dH(\tau)/d\tau = -A(\tau)$, $dF^+(\tau)/d\tau = (3/4)H(\tau)$, $H(\tau) = F^+(\tau) - F^-(\tau)$, $A = \sum_{k=1}^n A_k = \int_0^{\tau_s} A(\tau)d\tau$, $A(\tau) = (1/2)A_k$, yielding a previously undescribed relationship between H and A . Changing variables to $x \equiv \tau/\tau_s$ and $a(x) \equiv \tau_s A(x)/A$ and performing the integration, one obtains $F^+(\tau_s) = (1 + K\tau_s)F_e$, $K = (3/4)(\alpha + \beta\gamma)$; $\alpha \equiv H(\tau_s)/F_e$, $\beta \equiv A/F_e$, $\alpha + \beta = 1$, $\gamma \equiv \int_0^1 xa(x)dx$, $\int_0^1 a(x)dx = 1$, F_e is thermal flux leaving to space. Eddington's approximation for radiative equilibrium remains formally valid for the case of non-radiative fluxes as well, if coefficient K at τ_s is changed as proposed.

A new scheme for accounting for the non-radiative heat fluxes in the radiative transfer problem for atmospheric thermal photons

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1. Radiative transfer equation: Implicit assumptions and validity for solar and laser radiation

The very notion of radiative transfer implies that there is a flux of radiative energy in a certain direction in space. Choosing the axes such that the direction of the radiative flux coincides with axis z , one obtains the well-known equation for the energy density in a unit solid angle in a unit interval of radiation frequency ν , of a ray beam that propagates at angle ϑ to z axis, the magnitude called radiation intensity $I(z, \vartheta)$:

$$\begin{aligned} \mu \frac{\partial I(z, \mu)}{\partial z} &= -\frac{1}{l(z)} \{I(z, \mu) - S(z)\}, \\ \mu \frac{\partial I(\tau, \mu)}{\partial \tau} &= +I(\tau, \mu) - S(\tau), \quad d\tau \equiv -\frac{dz}{l(z)}, \quad \mu = \cos \vartheta, \end{aligned} \quad (1)$$

where $l(z) = [\sigma n(z)]^{-1}$ is photon mean free path length, σ is the absorption cross-section (also called absorption coefficient), $n(z)$ is the volume density of the number of absorber molecules in the medium; $d\tau \equiv -dz/l(z)$ is the optical path length, that is calculated in the opposite direction compared to the direction of z axis; $S(\tau)$ is the isotropic energy density of the radiation emitted by the medium per unit optical path length flux, called source function. Putting $\mu = 0$ in (1), one obtains:

$$S(\tau) = I(\tau, 0). \quad (2)$$

It should be emphasized that equation (1) is a most general representation for the propagation of radiation within the medium, it does not assume stratification of this medium (e.g., atmosphere). The first term in the right-hand part of Eq. (1) corresponds to absorption of radiation by the medium. The magnitude by which intensity changes over one photon mean free path length $dz = l$ (i.e., over optical path length $-d\tau = 1$) is equal to intensity itself, i.e. $-dI = I$ at $-d\tau = 1$. The physical meaning of this relationship is that all radiation, on average, is absorbed along one mean free path length.

But the first two parts of Eq. (1) carry additional information about the medium where absorption occurs. It is implicitly assumed that absorption of photons is spatially uniform over the whole optical path. That is, the length of photon's free path length, which is counted from an arbitrary point in space to the point of collision with a molecule and absorption, is on average equal to l in Eq. (1). It is under this implicit assumption that one can consider practically any, however small, values of $d\tau$, down to the small magnitudes of the order of intermolecular distances in the medium.

Equation (1) allows one to meaningfully consider irreversible absorption by the medium of laser and direct solar radiation that propagates within a narrow solid angle at $\mu = 1$. Irreversibility of absorption means that the medium does not emit photons of the same length as absorbed, so that $S(\tau) = I(\tau, 0) = 0$. In this case Eq. (1) solves as

$$I(\tau) = I(0) e^{-\tau}. \quad (3)$$

Equation (1) is only physically meaningful for intensity $I(\tau, \mu)$ in an infinitely small solid angle, which is much smaller than all the other solid angles measured in the considered problem (Milne, 1930). This becomes clear from the fact that in free space, i.e. at $l = \infty$, the right-hand part of Eq. (1) is equal to zero, while the left-hand part for any finite source of radiation continues to change as the ray beam diverges in space in any finite solid angle. Therefore, Equation (1) for intensity $I(\tau, \mu)$ is physically meaningful only for irreversible absorption of laser or direct solar radiation which conform to the above criteria, i.e. propagate practically without divergence.

Relationship (3) is used to measure the magnitude of the molecular absorption cross-section σ (absorption coefficient) at different radiation frequencies with the known density n of absorber molecules. An account of the collisional pressure broadening of the contours of absorption lines has shown that after integrating over the entire contour of an isolated absorption line or a whole absorption band, absorptivity $T(n) = \int e^{-\tau} d\nu$ and emissivity $A(n) = 1 - T(n)$ are related to n in different ways dependent on the magnitude of the latter, from linear dependence at small n to square-root and logarithmic dependence at large n . The central part of the absorption contour, which widens towards the wings of the absorption profiles with growing n , ceases to depend on concentration with growing n . This is interpreted as “saturation” of absorption. All these regularities arise in consequence of the exponential decline of intensity with growing τ , Eq. (3).

To summarize,

Conventional form of the radiative transfer equation contains undiscussed implicit assumptions, which make it valid for description of only particular types of radiative transfer, like laser or direct solar radiation, which propagate with practically no angular divergence in space and are irreversibly absorbed by the medium. Transfer of thermal photons is principally different.

2. Thermal radiation in the atmosphere: Radiative equilibrium

Thermal radiation of the Earth’s surface undergoes resonance scattering when interacting with atmospheric greenhouse substances. **Thermal photons do not disappear, but change their direction.** Some part of them return back to the surface, where the energy density of thermal photons increases compared to the case when their resonance scattering in the atmosphere is absent. In the result, flux of thermal radiation diffuse to the upper atmosphere and leaves to space due to random walk of thermal photons.

Thermal photons propagate in all directions at any possible values of μ in (1). Therefore, for thermal photons Eq. (1) remains physically meaningful only for intensity in an infinitely small, physically unmeasurable, solid angle. The second term $S(\tau)$ in the right-hand part of Eq. (1) starts, as compared to the previously discussed case of irreversible absorption, to differ from intensity by a constant magnitude that is independent of intensity. That this is so is clear, in particular, from relationship (2). In the result, the first part of Eq. (1) ceases to remain on intensity; and namely this fact determines the diffusional, linear character of the propagation of thermal radiation. In this situation, isolation of intensity in the right-hand part of Eq. (1) becomes a mathematical manipulation that is critically misleading as distorting the physical meaning of the investigated processes, see Appendix 1.

To summarize,

Transfer of thermal radiation in the atmosphere does not retain any traces of the exponential behaviour of intensity as described by Eq. (3). The notions of transmissivity and absorptivity, defined with use of the exponent $e^{-\tau}$, lose their meaning. The dependencies of any physical variables on concentrations of greenhouse substances, that were based on the $e^{-\tau}$ exponent, like square-root, logarithmic, etc., vanish as well. Consequently, such dependencies cannot determine how the greenhouse effect depends on concentration of greenhouse substances.

3. Transfer of thermal radiation does not depend on the variance of photon free path length

In the presence of non-radiative fluxes of latent and sensible heat, dissipation of their energy leads to heating of atmospheric air at different heights. Via thermal energy of the chaotic motion of air molecules, the energy of non-radiative fluxes is ultimately converted, due to collisional excitation of molecules of the greenhouse substances, to the energy of thermal radiation. In this case the source function remains undetermined. It is necessary to find how it is related to the dissipation rate of the dynamic fluxes of latent and sensible heat.

This relationship can be established if one refrains completely from the consideration of equations (1) and (2). Instead, one should use the remarkable fact that the flux of propagation of particles or photons during their random walk is fully determined by the **mean** free path length of thermal photons and is independent of its variance and standard deviation. This is caused by the fact that positive and negative deviations of random free path lengths from the mean value cancel each other during random walk. Therefore, it is physically possible to perform all consideration of the transfer problem using the mathematically convenient zero magnitude of the standard deviation of photon's free path length.

Let us introduce mean length l_c of photon free path between two consecutive collisions of photon with molecules of the absorber and mean length l of photon free path from an arbitrary point in space to the first collision with a molecule of the absorber. When the variance of photon free path length \hat{l}_c between two consecutive

collisions is zero, mean length l is related to mean length l_c of photon free path between two subsequent collisions by a simple geometric rule:

$$l = \frac{l_c}{2}. \quad (4)$$

This relationship is obtained by averaging the coordinate of the initial point from which l is measured, over the whole distance between two consecutive collisions (when the variance of \hat{l}_c is zero, this distance is invariably equal to l_c , thus yielding (4)).

A different assumption regarding the variance of photon free path length, implied in Equation (1), is the assumption about uniform spatial distribution of absorber's molecules, when the probability for a photon to be absorbed by a molecule is the same for any given point in space and independent of the distance covered by the photon prior to collision. In such a case the standard deviation of \hat{l} from the mean is equal to the mean, l , while the latter (i.e. mean length of photon free path from an arbitrary point to collision) coincides with the mean length l_c of photon free path between two subsequent collisions, $l = l_c$.

A vivid daily-life example of the difference between the latter equality and Eq. (4) is provided by the dependence of mean waiting time at a bus stop on the temporal distribution of buses along the route. In the morning, when the buses come to the bus stop strictly following the timetable, in fixed time periods t_c with zero variance, mean waiting time t at the stop of a person who comes there at an arbitrary time point is equal to $t_c/2$, $t = t_c/2$. However, in the middle of the day, after having stuck in traffic jams, the bus cars re-distribute randomly along the route and arrive at the bus stop in arbitrary time periods with large non-zero variance. In such a case, if the mean speed and number of buses on the route remain the same, mean waiting time at the bus stop increases twofold and becomes equal to the mean time period t_c between two consecutive arrivals of buses at the bus stop, $t = t_c$.

4. Accounting for the sensible and latent heat fluxes from the surface to the atmosphere

Let us divide the atmosphere into n adjacent layers of depth l_c each, Fig. 1. Obviously, l_c decreases with increasing height due to the drop of air concentration; l_c is different in different layers, but for simplicity we omit the low index k (denoting layer's number) at l_c . As far as the variance of l_c is put zero, photons absorbed (and re-emitted) within any k -th layer are only those photons that originate (i.e. were emitted) within the neighbouring lower and upper layers numbered $k + 1$ and $k - 1$, respectively.

Let us now show that when the radiative flux passes through a given layer k to the next one, only $2/3$ of this flux change due to collisions with the absorber molecules located within the next layer. The remaining $1/3$ of the radiative flux experience collisions with molecules located within the same k -th layer and does not transmit radiation to the next layer, i.e. effectively it does not participate in radiative transfer.

Consider a photon emitted in the upward direction from a given point within layer k at a distance x from the border of the upper adjacent layer number $k - 1$, Fig. 2. This photon, travelling distance l_c , will reach the next $(k-1)$ -th layer if only the angle between the vertical z -axis and the direction of its movement does not exceed ϑ , where $\cos \vartheta = x / l_c = \mu$. Let I_k denote the power of isotropic radiation emitted into the upper hemisphere by a unit volume of the absorber (e.g., greenhouse gas) within a unit solid angle. Flux F_k emitted by the k -th layer of thickness l_c into the upper (+) and lower (-) hemispheres, is equal to $F_k^\pm = 2\pi I_k l_c \int_0^1 \mu d\mu = \pi I_k l_c$. Radiative flux $d\hat{F}_k(x)$,

composed of photons which are emitted within area of thickness dx located at a distance $x \leq l_c$ from the upper border of the k -th layer and which collide with the absorber molecules within the next $(k - 1)$ -th layer, is equal to:

$$d\hat{F}_k(x) = 2\pi I_k dx \int_{x/l_c}^1 \mu d\mu = F_k \frac{dx}{l_c} \left(1 - \frac{x^2}{l_c^2}\right).$$

The corresponding flux $\hat{F}_k(l_c)$ of photons emitted within the whole k -th layer and reaching the next layer is equal to

$$\hat{F}_k(l_c) = F_k \int_0^{l_c} \frac{dx}{l_c} \left(1 - \frac{x^2}{l_c^2}\right) = \frac{2}{3} F_k. \quad (5)$$

This means that the flux of photons exchanging energy between any two adjacent layers is equal to $2/3$ of the radiative flux F_k^\pm within the layer. The remaining one third of thermal photons emitted within a given layer does not leave this layer, but is absorbed within its boundaries.

The energy conservation law for the k -th layer can be written as follows:

$$\frac{2}{3} F_k^+ + \frac{2}{3} F_k^- = \frac{2}{3} F_{k+1}^+ + \frac{2}{3} F_{k-1}^- + A_k, \quad \sum_{k=1}^n A_k = A, \quad F_{\text{in}} - A = H_s, \quad F_{n+1}^+ \equiv F_s. \quad (6)$$

Here $A_k > 0$ is the additional flux of radiative energy which originates in the k -th layer after dissipation of the non-radiative fluxes of sensible and latent heat into thermal radiation, as well as after dissipation of the flux of solar radiation absorbed within this layer; A is the total flux of sensible and latent heat, as well as of solar radiation, which undergoes dissipation in the entire atmospheric column, $A \geq 0$; F_{in} is the total flux of solar radiation absorbed by the planet (including solar energy absorbed in the atmosphere), in the stationary case F_{in} is equal to the outgoing flux F_e of thermal radiation leaving the atmosphere into space; the difference $H_s \equiv F_{\text{in}} - A$ denotes the part of solar radiation which undergoes full dissipation (i.e. to thermal radiation) at the planetary surface, $H_s > 0$; F_s is the flux of thermal radiation of the surface. The boundary conditions $F_0^- = 0$, $F_0^+ = \frac{2}{3} F_1^+ = F_e$, $\left(F_1^+ - F_0^- = \frac{3}{2} F_e\right)$ take into account that thermal radiation from layer with $k = 0$ leaves into space. Taking into account that within each layer fluxes of emitted radiation are isotropic, $F_k^+ = F_k^-$, one can write (6) in the following discrete form of the diffusion equation:

$$(F_{k+1} - F_k) - (F_k - F_{k-1}) = -\frac{3}{2} A_k; \quad (7)$$

In the continuous representation, taking into account the boundary conditions, the last equation in (7) assumes the conventional diffusion form:

$$\frac{d^2 F^+(k)}{dk^2} = -\frac{3}{2} A_k, \quad \frac{dF^+(0)}{dk} = \frac{3}{2} F_e, \quad F^+(0) = F_e. \quad (8)$$

Let us now use the conventional definition of atmospheric optical depth τ and optical thickness τ_s , when height z is counted from the surface upwards:

$$\tau = \int_z^\infty \frac{dz'}{l(z')}, \quad \tau_s = \int_0^\infty \frac{dz}{l(z)}, \quad d\tau = dz/l(z) \quad (9)$$

Optical depth τ (9) at a given height z is defined as the number of layers of thickness l (see (4) and Section 3) above z . Using relationship (4) we obtain:

$$k = \frac{\tau}{2}, \quad n = \frac{\tau_s}{2}, \quad \Delta k = \frac{\Delta \tau}{2}, \quad (10)$$

where k and n are the corresponding numbers of layers of thickness l_c . In the result, we obtain

$$\frac{d^2 F^+(\tau)}{d\tau^2} = -\frac{3}{4} A(\tau), \quad \left. \frac{4}{3} \frac{dF^+(\tau)}{d\tau} \right|_{\tau \rightarrow 0} = \frac{3}{4} F^+(0); \quad F^+(0) = F_e. \quad (11)$$

$$A = \sum_{k=1}^n A_k = \int_0^{\tau_s} A(\tau) d\tau, \quad d\tau = 2 dk, \quad A(\tau) = \frac{1}{2} A(k).$$

Solution of Eqs. (11) under boundary condition $H(0) = F^+(0) = F_e$ becomes

$$F^+(\tau) = \left(1 + \frac{3}{4} \tau\right) F_e - \frac{3}{4} \int_0^\tau d\tau' \int_0^{\tau'} A(\tau'') d\tau''. \quad (12)$$

The twofold integral in (10) can be transformed to an ordinary one by integrating over τ' by parts. Introducing new variables $x \equiv \tau/\tau_s$ и $a(x) = \tau_s A(x)/A$, and $a(y) \equiv \tau_s A(y)/A$ the flux of thermal radiation at the Earth's surface can be written as

$$F_s \equiv F^+(\tau_s) = (1 + K\tau_s) F_e, \quad K = \frac{3}{4} [\alpha + \beta\gamma], \quad (13)$$

$$\alpha \equiv H_s / F_e, \quad \beta \equiv A / F_e, \quad \alpha + \beta = 1, \quad \gamma \equiv \int_0^1 x a(x) dx, \quad \int_0^1 a(x) dx = 1.$$

At $A = 0$ (the case of radiative equilibrium) we have $K = 3/4$ and the exact solution of (13) coincides with the well-known Eddington's approximation for Eq. (1).

Account of non-radiative heat fluxes at large values of τ only changes coefficient K at τ_s , which does not depend on τ_s and, hence, on concentrations of greenhouse substances. It should be stressed that the ratio F_s / F_e and, hence, the greenhouse effect **grow linearly** with τ_s , being proportional to the concentration of greenhouse substances. Growth of concentration does not lead to any saturation of the greenhouse effect.

5. Greenhouse effect dependence on concentrations of greenhouse substances

All the results obtained so far pertain to monochromatic radiation.

Due to the resonance character of interaction between thermal photons and molecules of greenhouse substances, when absorption and subsequent emission of photons does not change their frequency ω , formula (13) remains valid for any particular molecular absorption band of the greenhouse substances. Flux of thermal radiation of the surface, F_s , is closely approximated by equilibrium blackbody radiation σT_s^4 , where T_s is surface temperature, and is described by Planck's distribution $F_p(\omega, T_s)$. Surface flux F_{si} in the region of the i -th absorption band of width $\Delta\omega_i$ can be represented as

$$F_{si} = \delta_i F_s, \quad \delta_i \equiv \frac{\Delta\omega_i F_p(\omega_i, T_s)}{F_s}, \quad F_s = \sigma T_s^4, \quad \sum_{i=1}^N \delta_i = 1, \quad (14)$$

where N is the total number of spectral intervals corresponding to different absorption bands in the thermal spectrum, including spectral windows where absorption is absent, i.e. $\tau_{si} = 0$. Denoting thermal flux outgoing into space in the region of the i -th absorption band as F_{ei} one can re-write formula (13) as

$$F_s \delta_i = (1 + K \tau_{si}) F_{ei}, \quad \sum_{i=1}^N F_{ei} = F_e. \quad (15)$$

Assuming that the non-resonance collisional excitation of the absorption bands is the same over entire thermal spectrum, it is reasonable to accept that coefficient K accounting for dissipation of non-radiative energy fluxes is independent of radiation frequency ω and, consequently, of index i in (15). The relationship between total fluxes of thermal radiation at the surface, F_s , and outside the atmosphere, F_e , is then obtained from (12) and (13) to be:

$$F_s = \frac{F_e}{b}, \quad b \equiv \sum_{i=1}^N \frac{\delta_i}{1 + K \tau_{si}}, \quad \tau_{si} = \int_0^\infty \frac{dz}{l_i(z)}, \quad l_i(z) = [N_i(z) \Sigma_i]^{-1}, \quad (14)$$

where $N_i(z)$ and Σ_i are the concentration and absorption cross-section, respectively, for the i -th greenhouse substance (Σ_i is equal to the sum of intensities of all absorption lines in the i -th absorption band divided by band width $\Delta\omega_i$; Σ_i is independent of gas pressure and is not affected by pressure broadening). The value of b has the meaning of relative transmissivity of the atmosphere with respect to thermal radiation. Relative greenhouse effect can be defined as $(F_s - F_e) / F_s = 1 - b$, while the absolute magnitude of the greenhouse effect is $F_s - F_e = [(1 - b) / b] F_e$.

Thus, here we have shown that the ratio of the upward fluxes of thermal radiation at the planetary surface and outside the atmosphere in the presence of non-radiative heat fluxes is a linear function of atmospheric optical thickness τ_s , i.e. it retains the form of Eddington's approximation with a modified coefficient at τ_s . This coefficient term can be unambiguously calculated from the vertical distribution of the dissipative power of the non-radiative heat fluxes.

Note that the proposed approach is not equivalent to the use of two-stream approximations, which violate both first and second laws of thermodynamics, see Appendix 2. Neither it involves the assumption of local thermodynamic equilibrium, which, when applied in the terrestrial atmosphere, violates the second law of thermodynamics, as far as here optical thicknesses of all greenhouse substances are of the order of unity, so that absorption over different parts of the thermal spectrum deviates greatly from the uniform (grey) absorption, see Appendix 3.

APPENDICES

English text of appendices to be posted in this file after 15th of April, 2007.

In the meantime, see Russian text at

www.bioticregulation.ru/common/pdf/foton-ru.pdf