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ARTICLE

Atmospheric Carbon Dioxide and Climate

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ABSTRACT

Atmospheric radiative fluxes are evaluated for the line-by-line model of spectral lines in considering the atmosphere as a weakly nonuniform plane layer and altitude profiles of its parameters are taken from the model of standard atmosphere. Concepts of molecular spectroscopy are combined with the local thermodynamic equilibrium for greenhouse gases and with information from HITRAN data base for parameters of radiative transitions. In addition, the energetic balance of the Earth allows one to determine the radiative flux from clouds. As a result, the algorithm is worked out for evaluation of the atmospheric radiative flux toward the Earth depending on its composition. We below concentrate on the change of atmospheric radiative fluxes as a result of doubling of the concentration of CO_2 molecules. It is shown that the change of the global temperature in this case according to the above algorithm in 5-6 times exceeds that followed from climatological models which are based on old spectral data, rather than those from HITRAN data base. These codes ignore overlapping of spectral lines of atmospheric radiators.

1. Introduction

The participation of the greenhouse effect in the energetic balance of our planet was understood two hundred years ago [1,2]. Then atmospheric emission in the infrared spectrum range increases the radiative flux to the Earth and increases the global Earth's temperature which is the Earth's surface temperature is averaged over the globe and time. Three basic greenhouse components are H_2O and CO_2 molecules, as well as water microdroplets which constitute clouds.

Starting from the Arrhenius paper [3] in 1896, connection between the concentration of atmospheric carbon dioxide and the Earth's climate causes the main attention to the greenhouse problem, especially, if this results from the human activity. The contemporary version of infrared (IR) atmospheric emission for outside radiation was presented in [4-6] that considers emission and absorption of atmospheric molecules. Recently [7] the author formulated the problem of IR atmospheric emission toward the Earth with accounting for cloud radiation. We below consider the theory of IR atmospheric radiation to the Earth and connect its results with those of adjacent problems.

2. Methods

We now consider methods which allow us to compose the algorithm to calculate the radiative flux from the atmosphere to the Earth. For the analysis of emission of molecular components, it is necessary to modify the classical molecular spectroscopy which was constructed as a direction of quantum mechanics at the beginning of 20

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century (for example [8,9]). Classical molecular spectroscopy considers infrared radiation of molecules as transitions between some vibration and rotation molecular states, so that selection rules determine the connection between initial and final vibration and rotation states for radiative transitions of a given molecule symmetry.

In molecular spectroscopy, rates of radiative transitions in molecules are expressed through the Einstein coefficients of these transitions. Spectra of molecules have the discrete character, but spectral lines for each radiative transitions are broaden due to interactions in gases where radiative molecules are located. Contemporary molecular spectroscopy is based on data banks, as HITRAN data bank for molecules [10-13]. Then the rate of a given radiative transition is expressed through four molecular parameters, namely, ω_j, the frequency of the center of a j-th transition, S_j, the intensity of this transition that is proportional to the first Einstein coefficient, ν_j, the width of the spectral line, and E_j, the excitation energy from the ground molecule state to a lower transition state [11].

Considering the atmosphere as a weakly nonuniform flat gaseous layer located over the Earth surface, and combining classical molecular spectroscopy with local thermodynamic equilibrium at each point of this layer, one can calculate the radiative fluxes to the Earth for the line-by-line model [14], i.e. for each frequency, at a given mixture of emitted molecular gases and a given altitude profile for the density of each greenhouse gas and the temperature. In this case we use the model of standard atmosphere [15] for the space distribution of carbon dioxide molecules and water molecules. But this algorithm is suitable for any space distribution of emitted components and is useful in meteorological codes where atmospheric radiation may be added to transport of mass, temperature and humidity. In this case the altitude profiles of the temperature and density of radiative components are required also.

One can model atmospheric radiation toward the Earth as emission of a flat layer. If this gaseous layer is optically thick and its temperature is independent of an altitude, the partial radiative flux through its boundary is given by the Planck formula

$$I_\omega(T)=\frac{\hbar\omega^3}{4\pi^2c^2\left[\exp\left(\frac{\hbar\omega}{T}\right)-1\right]} \tag{1}$$

If the optical thickness of the layer is restricted, the radiative flux at a given frequency is given by [16]

$$J_\omega(T)=I_\omega(T)g(u_\omega) \tag{2}$$

In this way the opaque factor $g(u_\omega)$ is defined for a given optical thickness u_ω of the layer according to formula [16]

$$g(u_\omega)=\int_0^1 d\cos\theta\int_0^{u_\omega}\exp\left(-\frac{u}{\cos\theta}\right)du=\int_0^1\cos\theta d\cos\theta\cdot\left[1-\exp\left(-\frac{u_\omega}{\cos\theta}\right)\right], \tag{3}$$

where θ is an angle between the direction of photon motion and the boundary perpendicular. In the limit of optically thick layer $u_\omega >> 1$ the opaque factor is $g(u_\omega)=1$, and formula (2) is transformed in formula (1).

It is necessary to take into account that the temperature varies, as an altitude is changed. For this one can divide the troposphere in some layers with an identical temperature and account a nonuniformity of the layer by numerical evaluation, as it is made in [5]. In our approach [17,18] we expand the total radiative flux over a small parameter and reduce the radiative process to that of an equilibrium layer with the temperature T_ω of an effective layer that is located at an altitude h_ω, that is

$$T_\omega=T(h_\omega) \tag{4}$$

In particular, in the case of an optically thick layer the effective layer is defined such, that its optical thickness is 2/3 [17,18]

$$u(h_\omega)\equiv\int_0^{h_\omega}k_\omega(h)dh=\frac{2}{3} \tag{5}$$

Here $k_\omega(h)$ is the absorption coefficient at a given frequency and an indicated altitude. The procedure of determination of the radiative temperature T_ω in a general case is described in [7] in detail.

The above analysis relates to molecular components of the atmosphere. Then, considering emission of molecules to be noncoherent, we summarize radiative fluxes from all the molecules and obtain the radiative temperature T_ω for the total radiative flux of a molecular gas. The separate problem is insertion in this scheme clouds or small atmospheric particles that is absent in climatological codes. Among models of cloud emission, we choose finally such, that clouds on a certain altitude absorb infrared radiation entirely. This means that the optical thickness of clouds is high, and they emit infrared radiation. As a result, we have for the radiative flux from the atmosphere [7]

$$J_\omega=I_\omega(T_\omega)g(u_\omega)+I_\omega(T_{cl})\left[1-g(u_\omega)\right], \tag{6}$$

where T_{cl} is the cloud temperature.

For realization of formula (6), it is necessary to use profiles for distribution of atmospheric radiators. As for radiating molecules, this information follows from the model of standard atmosphere or other similar sources. But it is difficult to obtain the same information for clouds consisting of water microdroplets because they are not formed in motionless air. Therefore determination of the

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amount of condensed water in the atmosphere is problematic.

In order to solve this problem within the framework of the model of standard atmosphere, we add to the above information the energetic balance of the atmosphere which was composed at first by NASA [19]. In particular, the NASA data for the Earth's energetic balance was published in the author book [20] of 1975. Subsequently other versions of the Earth's energetic balance were represented by World Meteorological Organization (WMO) [21,22]. But these fluxes coincide with previous ones within the limits of a few percent.

The total flux of infrared radiation toward the Earth integrated over frequencies must coincide with that followed from the energetic balance of the Earth and its atmosphere. This requirement allows one to determine the position of clouds. In particular, for the model of the standard atmosphere, the effective altitude of cloud location is approximately 3.4 km, and the effective temperature of cloud emission is $T_{cl} = 266K$, whereas the temperature of the Earth's surface is equal $T_E = 288K$ for this model. Thus, the above scheme allows one to determine the parameters of the radiative flux from the atmosphere to the Earth.

3. Results

We use the above scheme to determine various radiative fluxes of the atmosphere. The connection between direct and reverse radiative processes is governed by the Kirchhoff law [23] or the principle of detailed balance for these processes. This allows one to use the absorption coefficient k_ω as the parameter of the absorption process in the analysis of atmospheric emission. The absorption coefficient of molecules of a given type is given by

$$k_\omega = N \sum_j S_j(T) a_{\omega-\omega_j} \tag{7}$$

Here N is the total number density of molecules of a given type, S_j is the intensity of the spectral line for a given transition, and the distribution function of photons over frequencies for air at atmospheric and nearby pressures is determined for the impact mechanism of broadening of spectral lines

$$a_{\omega-\omega_j} = \frac{\nu_j}{2\pi\left[\left(\omega-\omega_j\right)^2 + \left(\nu_j/2\right)^2\right]}\ , \tag{8}$$

where ω_j is the frequency of the spectral line center, and ν_j is the spectral line width. Next, the intensity of spectral lines is given in the HITRAN tables for a certain temperature. The transition to another temperature proceeds on the basis of formula

$$S_j(T) = S_j(T_o) \cdot \exp\left(\frac{\varepsilon_j}{T_o} - \frac{\varepsilon_j}{T}\right) \tag{9}$$

Table 1. Radiative fluxes of greenhouse atmospheric components to the Earth and their contributions to the total radiative flux absorbed by the Earth [7]

Component	Flux, W/m^2	Portion, %
H_2O-molecules	166	51
H_2O-droplets (clouds)	96	29
CO_2-molecules	58	18
CH_4-molecules	4	1
N_2O-molecules	3	1

Thus, these formulas with using the HITRAN data allow us to determine the absorption coefficient and then the atmospheric radiative fluxes to the Earth according to a described scheme. The values of the radiative fluxes toward the Earth are presented in Table 1. In addition, atmospheric molecules and clouds are sources of infrared emission in a different frequency range. According to evaluations within the framework of this scheme [7], approximately 95% of the radiative flux at frequencies below 800cm^{-1} is created by water and carbon dioxide molecules, while 84% of this flux at frequencies above 800cm^{-1} is due to water microdroplets of clouds.

Let us consider the greenhouse effect which consists in change of the global temperature as a result of variation of the atmosphere content. Usually as the measure of this effect is taken the doubling of the concentration of CO_2 molecules, and below we compare variations of radiative fluxes as a result of this variation. We denote by ΔJ the variation of the total radiative flux as a result of doubling of carbon dioxide concentration, and by $\Delta J(H_2O), \Delta J(CO_2), \Delta J_{cl}$ the corresponding changes of the radiative fluxes created by water molecules, carbon dioxide molecules and clouds correspondingly. From the definition of the total radiative flux the following equation is fulfilled

$$\Delta J = \Delta J(H_2O) + \Delta J(CO_2) + \Delta J_{cl} \tag{10}$$

Table 2 contains the changes of these radiative fluxes in indicated frequency ranges.

Table 2. Changes of radiative fluxes from the standard atmosphere to the Earth as a result of doubling of the concentration of CO2 molecules in the infrared spectrum range [7]. These changes are defined in the text and are

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expressed in W/m^2

Frequency range, Δω	ΔJ (CO2)	ΔJ (H2 O)	ΔJcl	ΔJt
580 - 600	0.96	-0.89	-0.04	0.03
600 - 620	0.81	-0.74	-0.03	0.04
620 - 640	0.63	-0.61	0	0.02
640 - 660	0.15	-0.14	0	0.01
660 - 680	0.18	-0.18	0	0
680 - 700	0.21	-0.20	0	0.01
700 - 720	0.12	-0.03	-0.03	0.06
720 - 740	0.64	-0.05	-0.39	0.20
740 - 760	1.07	-0.10	-0.68	0.29
760 - 780	0.56	-0.02	-0.40	0.14
780 - 800	0.25	-0.02	-0.17	0.06
800 - 850	0.15	-0.03	-0.08	0.04
900 - 950	0.20	0	-0.16	0.04
950 - 1000	0.76	-0.01	-0.53	0.22
1000 - 1050	0.18	0	-0.13	0.05
1050 - 1100	0.37	0	-0.26	0.11
total	7.24	-3.02	-2.90	1.32

As it follows from Table 2, doubling of the concentration of atmospheric carbon dioxide causes an increase of the radiative flux due CO_2 molecules by 7.2W/m^2, whereas radiative fluxes due to water molecules and water microdroplets decrease by 3.0W/m^2 and 2.9W/m^2 correspondingly, and the change of the total radiative flux is 1.3W/m^2. In addition, the contribution of 30% to the change of the radiative flux due to CO_2 molecules is created by vibration transitions at wave lengths near 9.4μm and 10.6μm which are used in CO_2 lasers. The contribution of this frequency range to the total radiative flux due to carbon dioxide molecules is about 2%.

In addition to Table 2, we give in Figure1 the average radiative flux ΔJ_c which is created by CO_2 molecules at frequencies below indicated ones and the corresponding total flux ΔJ_t. These fluxes are defined according to formulas

$$\Delta J_c = \int_0^{\omega} \Delta J(CO_2) d\omega, \quad \Delta J_t = \int_0^{\omega} \Delta J d\omega, \tag{10}$$

where $\Delta J(CO_2)$ and ΔJ are defined in formula .

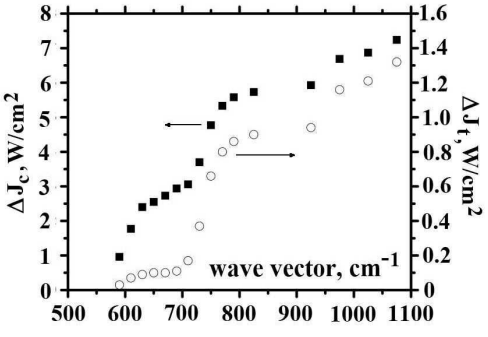

Figure 1. Change of the radiative fluxes as a result of doubling of the concentration of CO_2 molecules up to an indicated frequency due to emission of CO_2 molecules (ΔJ_c) and the change of the total radiative flux (ΔJ_t)

Figure 1 includes the frequency ranges which are responsible for formation the flux changes under consideration. The change of the radiative flux due to CO_2 molecules ΔJ_c is formed mostly (approximately 80%) inside the absorption band of the CO_2 molecule (mostly near the boundary of this absorption band) that ranges from 580 cm^{-1} up to 760cm^{-1}. The frequency range near the left boundary of the absorption band of the CO_2 molecule does not give the contribution to the change of the total radiative flux toward the Earth ΔJ_t because of a strong absorption by water molecules at such frequencies. Hence, an increase of the radiative flux due to carbon dioxide molecules is compensated by the decrease of the radiative flux due to water molecules at these frequencies.

One can transform the change of the total radiative flux as a result of doubling of carbon dioxide concentration $\Delta J = 1.3$ W/m^2 into the change of the global temperature ΔT in the standard method through the climate sensitivity [24]. As a result, one can obtain

$$\Delta T = (0.6 \pm 0.3)\ \mathrm{K} \tag{11}$$

A large error results from a high sensitivity of this change to variation of parameters. In addition, there is an uncertainty in determination of shift of the cloud temperature as a result of an increase of the global temperature.

4. Discussions

We now analyze the above results. In spite of a low accuracy, the change of the global temperature due to doubling of the atmospheric concentration of CO_2 molecules (11) differs dramatically from that obtained on the basis of climatological computer codes [25]

$$\Delta T = (3.0 \pm 1.5)\ \mathrm{K} \tag{12}$$

We show the reason of this description. Climatological codes are based on studies before a middle of 20 century [3,26-28] when information about radiative parameters was restricted. Then interaction between spectra of carbon dioxide molecules and other greenhouse components (water molecules and clouds) was ignored. As a result, the change of the radiative flux due to carbon dioxide molecules was equated with the change of the total radiative flux from the atmosphere. In reality, an increase of the carbon dioxide concentration, which leads to an increasing radiative flux due to CO_2 molecules, causes simultaneously a decreasing radiative flux owing to water molecules and water microdroplets of clouds. Under the contemporary atmosphere content, the ratio of changes for the flux due to carbons

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dioxide and the total one is 5-6. Correspondingly, climatological models with such computer codes give the change of the global temperature which exceeds the real one in 5-6 times.

A certain contribution to the error of the global temperature change (11) follows from model assumptions in the scheme used. In order to estimate this error, we compare in Table 3 values of change of the global temperature ΔT for model approaches which are used by the author in the course of construction of the above algorithm. The first model [29,30] where it is used the average absorption coefficient of water molecules and water microdroplets over the total spectrum, and the average absorption coefficient of carbon dioxide molecules over oscillations, the second model [31,32] takes the same water parameters and the accurate absorption coefficient is used. In the third model [33,34] the accurate absorption coefficient of water molecules on the basis of HITRAN data base is used, and the altitude distributions for water molecules and water microdroplets are taken identical. In the above scheme [8] we account for clouds to be located starting from certain altitudes. The values of ΔJ coincide for these four approaches with the accuracy of 20%.

Note one more peculiarity of Table 3. For first three models we restrict by the frequency range below 800 cm^{-1} only, because this range gives the main contribution to the total radiative flux due to CO_2 molecules. But we convince subsequently that the range of laser spectral lines above 900cm^{-1} gives the contribution of 30 % to the change of the radiative flux due to CO_2 molecules. Therefore we use in Table 3 the factor 1.3 for results of first three models.

Table 3. Change of the global temperature at doubling of the carbon dioxide concentration

Model	ΔT, ^{0}C	References
1	0.5	[29, 30]
2	0.4	[31,32]
3	0.5	[33, 34]
4	0.6	[7]

The physical picture of evolution of the global temperature may be added by NASA monitoring of atmospheric parameters. Careful measurements of the concentration of atmospheric CO_2 molecules in Mauna Loa observatory (NASA) in Hawaii [35,36] show the variation of this value from 316ppm up to 411ppm starting from 1959 up to now [37,38]. Because this observatory is located far from sources and absorbers of carbon dioxide, and a time of residence of a CO_2 molecule in the atmosphere is (4-5) years, these measurements may be considered as the global variation of the concentration of atmospheric carbon dioxide.

One more parameter of the atmosphere, that characterizes its state as a whole, is the global temperature that is the temperature of the Earth surface which is averaged both over time and globe. The problem is that fluctuations of global temperature at each point of the Earth as a result of such averaging over time are measured in degrees. However, it is possible to reduce the fluctuations in the case of comparison of the temperature differences at the same point and time of day and season, but in different years, and with subsequent averaging. This concept [39] allows one to reduce the temperature fluctuations up to (0.1-0.2) K.

The change of the global temperature from 1985 up to now, where the correlation is observed between the change of the global temperature ΔT and the change of the concentration of atmospheric carbon dioxide, we have [40-42]

$$\Delta T = (0.6 \pm 0.1)\ \mathrm{K} \tag{13}$$

In spite the temperature evolution for oceans and land, as well as for the Northern and Southern Hemispheres, are different [43], one can take (13) as the global temperature change for last 35 years. This leads the change of the global temperature as a result of doubling of carbon dioxide concentration in the real atmosphere [44]

$$\Delta T = (2.5 \pm 0.3)\ \mathrm{K} \tag{14}$$

Comparing this with formula, one can find that the contribution to the change of the global temperature due to accumulation of carbon dioxide in the contemporary atmosphere is approximately 25%.

We also summarize the results of evaluations under consideration. We represent the algorithm of calculation of the radiative flux from a weakly nonuniform gaseous layer toward its surface. This algorithm is used for calculation the radiative flux from the atmosphere to the Earth's surface for the model of standard atmosphere. We use a local thermodynamic equilibrium for atmospheric air and its molecular admixtures, as well as clouds which provide a high optical thickness of the atmosphere and are located at a certain altitude. In addition, we assume radiation to be noncoherent, so that the radiative flux at a given frequency is a sum of fluxes from individual radiators. As a result, the total radiative flux at a given frequency is expressed through the radiative temperature of molecular radiators, the opaque factor for molecules and the effective altitude of emitting clouds or the radiative temperature of water microdroplets which constitute clouds. The cloud temperature follows from the requirement that the total radiative flux toward the Earth summarized over frequencies is

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equal to that from the Earth's energetic balance.

In calculation the molecular radiative parameters, we combine molecular spectroscopy with information followed from HITRAN data base. This algorithm allows us to determine the partial radiative flux at each frequency and the contribution of each greenhouse component. The contribution to the total radiative flux for the real (standard) atmosphere is 51% due to water molecules, 29% due to water microdroplets (clouds), 18% due to CO_2 molecules, and 2% due to CH_4 and N_2O molecules. In addition, 98% of the flux of infrared radiation at wavelengths below 12.5 μm is created by H_2O and CO_2 molecules, whereas 85% of the radiative flux at wavelengths above 12.5 μm is due to clouds.

If the concentration of atmospheric CO_2 molecules is doubled without a change the other atmospheric parameters, the change of the radiative flux to the Earth due to CO_2 molecules is 7.2 W/m^2, whereas the change of the total radiative flux with accounting for screening fluxes from other components is 1.3 W/m^2 that corresponds to the global temperature change of 0.6 ± 0.3 K. Usually climatological models do not account for the interaction between greenhouse that leads components to a six times larger temperature change. One can compare the latter value with results of NASA programs for the analysis of evolution of the carbon dioxide concentration and the global temperature. From this it follows that doubling of the concentration of atmospheric CO_2 molecules is accompanied by the change of the global temperature according to (14). Thus, in the real atmosphere only a forth part of the global temperature change occurs due to variation of the concentration of CO_2 molecules results from the greenhouse effect involving these molecules.

5. Conclusion

In this paper we apply the algorithm [7] for evaluation of atmospheric radiative fluxes toward the Earth to the problem of the change of the global temperature as a result of doubling of carbon dioxide concentration in the atmosphere. This algorithm includes the combination of thermodynamics and molecular spectroscopy of atmospheric air with usage the contemporary data for radiative IR transitions in molecules from HITRAN data base within the framework of the line-by-line model, as well as the energetic balance of the Earth and its atmosphere for radiative fluxes of clouds. Though we apply this algorithm to the global temperature on the basis of the model of standard atmosphere, it may be applied to the analysis of radiative fluxes from a local atmosphere if altitude profiles of the atmospheric temperature and number densities of greenhouse components are known. In this paper we take this algorithm for evaluation of the global temperature due to doubling of the carbon dioxide amount in the atmosphere.

The main conclusion of the above analysis is the importance of interaction between greenhouse components (H_2O and CO_2 molecules, water microdroplets of clouds), since elementary radiators of the atmosphere are simultaneously absorbers. Hence, an increase of the amount of one greenhouse component which leads to growth of the radiative flux due to this component, causes simultaneously the screening for radiation of other components, i.e., the radiative flux owing to other components decreases. This fact was not taken into account in first studies of this problem [3,26-28] because of restricted information about radiative transitions in molecules, though it was discussed [27,28].

In particular, the analysis on the basis of information of fifties [27,28] gave that overlapping of spectra of H_2O and CO_2 molecules leads to a decrease of the global temperature by 20%. The above evaluations on the basis of information from HITRAN data base show that this change is in 5-6 times. Unfortunately, some climatological codes do not account for this fact and suggest strongly heightened values of the change of the global temperature due to an increasing amount of carbon dioxide in the atmosphere. Just these values are used in the Paris agreement of 2015 on climate [45]. We above determine the above change of the global temperature according to which the greenhouse effect due to CO_2 molecules is approximately 25% of the total change of the global temperature under contemporary atmospheric conditions. As it follows from this, the basis of the Paris climatic agreements [45] is wrong.

It would note indicate the danger of a wide propaganda about the role of carbon dioxide in the future climate. In particular, the propaganda of European media in interest of some financial groups convinces European habitants that the most danger in futures follows from injection of carbon dioxide in the atmosphere as a result of combustion of fossil fuels. Indeed, a contemporary increase of the carbon dioxide amount in the atmosphere is a result of human activity which changes the carbon equilibrium between the atmosphere, land and oceans. The careful investigations are required, as the above NASA programs for atmospheric carbon dioxide and global temperature, which allow one to understand a real state of affairs in order to conserve our planet for the man.

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