Influence of variations in the CH_4 and N_2O concentration on long-wave radiative fluxes in the Earth's atmosphere

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It is generally accepted that the greenhouse effect in the Earth's atmosphere is caused by increasing concentrations of not only carbon dioxide, but also CH_4, N_2O, and freons. However, the inferred conclusions substantially depend on the accuracy of radiative models which is determined by the quality of spectroscopic information and correct parametrization of the equation of radiative transfer. In this paper, errors in calculation of long-wave radiative fluxes under clear-sky conditions are analyzed. In this context the problem is discussed on the necessity to take into account such greenhouse gases as CH_4 and N_2O in current radiative models.

Introduction

The role of minor gaseous constituents in radiative processes running in the Earth's atmosphere is a matter of common knowledge. One very popular hypothesis on the climate history relates the change in the temperature to variations of the carbon dioxide concentration in the atmosphere.[1] Growing pollution of the atmosphere increases the contribution of other gases, such as methane, nitrogen oxides, and freons. According to the data obtained in Refs. 2 and 3, the effect of doubling the CH_4, N_2O, CF_2Cl_2, and $CFCl_3$ concentration is similar to that of doubling the carbon dioxide concentration. Estimates made with ignorance of the atmospheric dynamics allow only tendencies in the climatic change to be revealed. To obtain more reliable information, one should take into account the contribution of these gases into radiative blocks of the models of global circulation in the atmosphere.

The demands imposed on the radiative models used in such problems are rather strong. Thus, for example, a decrease of the solar constant by 1% (~ 14 W/m^2) may lead to new Ice Age,[4] whereas doubling of the CO_2 concentration which is thought to cause significant warming of the climate produces a change in the long-wave flux of upward radiation by about 3–4 W/m^2, what also makes up about ~ 1% of the total flux of the outgoing thermal radiation.[5] Thus, the accuracy of calculation of the long-wave radiative fluxes should be at least as high as 1%.

The results of comparison of the measured data on downward fluxes of long-wave radiation under clear-sky conditions with those calculated by the line-by-line method demonstrate the accuracy currently achieved in taking into account the molecular absorption.[6] According to Ref. 6, average discrepancy between the calculated and experimental values is about 1 W/m^2, the rms deviation is about 2 W/m^2, and the maximum spread is from −4.2 to 5 W/m^2.

The specificity of climatological problems requires the models to provide high speed of calculation of the radiative characteristics. The exponential series[7–9] give some compromise between the accuracy and speed, so some techniques have been developed allowing a determination of the model parameters from the results of direct calculation. The methods have been formulated for expansion into the exponential series with a preset accuracy. The recent studies[10–12] have lifted the last restriction on this approach associated with taking into account overlapping of absorption bands. These achievements allowed a development of new methods ensuring high speed of computation and the accuracy comparable with that given by the line-by-line method (for example, in Ref. 6 the discrepancy between these methods was below 1 W/m^2). The similar estimates were obtained by us when using the software package described in Ref. 12.

Thus, we can say that the methodical error caused by parametrization does not exceed the error associated with the initial spectroscopic information. Most scientists[6,13] are inclined to believe that the main cause of errors is continuum absorption. Nevertheless, it is shown in Ref. 14 that the uncertainty in the parameters of spectral lines also plays an important role.

In this paper, based on the statistical approach, we consider the influence of the uncertainty in the spectral lines parameters included in the HITRAN–96 atlas[20] on the error of calculation of long-wave radiative fluxes, as well as, their sensitivity to uncertainties in the coefficients of continuum absorption. The influence of CH_4 and N_2O concentration variations on the radiative processes in the Earth's atmosphere is studied. Under discussion is also the necessity of accounting for these gases in current radiative models.

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Test calculations

The KD (*k*-distribution) model used in these calculations is described in detail in Ref. 12. We have analyzed the main sources of errors in this model and divided them into three groups. The first group includes the parametrization error due to presentation of the transmission functions as exponential series. The second group incorporates the errors which appear at numerical integration over height. The third group of errors is associated with uncertainty of the initial spectroscopic information. To estimate these errors, a series of test calculations has been performed (Table 1). As a result, it was found that, in general, KD agreed closely with LBL2, which employed the similar model of continuum absorption.[14] We obtained marked discrepancies for the downward fluxes at the altitudes above 50 km. To reveal the causes of these discrepancies for the isothermal atmosphere which contains only CO_2 in the 15-μm region, the downward flux intensity was calculated by the line-by-line method for narrow spectral ranges (~ 100 cm^{-1}); the method used for integration over height was the same as used in the KD model. To exclude the errors connected with the diffusion approximation, we revised the scheme of the fluxes calculation in the KD model. For faster computation of the integral exponents, the interpolation procedure was used. The estimates have shown this modification to have no effect on the speed of fluxes calculation. The discrepancy between the fluxes of the KD model and our line-by-line calculation did not exceed 0.3%. This allowed the conclusion that the main causes of discrepancy between KD and LBL2 in this case are a wide-meshed altitude grid and different quadrature formulas used. The problem of presenting transmission functions as exponential series is much studied. The error of the method of *c-k*-correlation used to calculate the transmission functions of an inhomogeneous path does not exceed 1% provided that the integration over height is correct.[15,16] That is why we focused our attention on the second group of errors.

Table 1. Comparison of the long-wave fluxes, in W/m²

Model	Surface			Tropopause			Upper boundary	Dnet1	Dnet2
	$F_1^{\uparrow}$	$F_1^{\downarrow}$	Net1	$F_2^{\uparrow}$	$F_2^{\downarrow}$	Net2	$F_3^{\uparrow}$		
3 MLS, isothermal atmosphere 250 K, CO_2 300 ppmv									
Mean	221.41	41.81	179.61	221.42	24.27	197.09	221.47	17.48	24.38
RMS	0.48	2.32	2.49	0.68	2.28	2.11	0.85	2.71	2.29
LBL1	221.56	40.27	181.29	221.56	25.06	196.49	221.55	15.20	25.06
LBL2	221.42	40.71	180.70	221.42	24.83	196.58	221.42	15.88	24.84
KD	221.53	40.78	180.74	221.53	23.87	197.66	221.53	16.92	23.87
4 MLS, isothermal atmosphere 250 K, CO_2 600 ppmv									
Mean	221.05	45.71	175.37	221.04	27.79	193.19	221.10	17.82	27.91
RMS	2.18	2.76	4.21	2.40	2.59	2.52	2.42	3.44	2.62
LBL1	221.56	44.31	177.24	221.56	29.23	192.33	221.55	15.09	29.22
LBL2	221.42	44.57	176.85	221.42	28.52	192.90	221.42	16.05	28.52
KD	221.53	44.46	177.07	221.53	27.58	193.94	221.53	16.87	27.59
19 MLS, H_2O LBL2 – continuum Clough et al., RSB – continuum Roberts et al.									
Mean	422.97	326.23	96.71	322.09	6.93	315.31	322.05	219.19	6.65
RMS	1.40	14.06	13.42	7.67	1.39	7.29	7.25	15.52	1.42
LBL1	423.54	330.40	93.14	328.22	4.90	323.32	328.06	230.18	4.74
LBL2	423.51	336.97	86.54	319.57	7.06	312.51	319.16	225.97	6.65
RSB	423.51	341.46	82.05	322.54	6.54	316.00	322.27	233.95	6.27
KD	423.60	335.34	88.26	321.14	7.11	314.03	320.91	225.77	6.88
20 MLS, H_2O, without continuum									
Mean	423.24	273.19	150.22	329.06	6.63	322.45	328.79	171.80	6.33
RMS	0.86	17.82	17.41	9.78	1.50	8.93	9.38	12.24	1.59
LBL1	423.54	261.10	162.44	336.99	4.90	332.10	336.83	169.66	4.73
LBL2	423.51	271.88	151.64	333.51	6.53	326.97	333.05	175.33	6.08
KD	423.60	271.38	152.22	334.03	6.75	327.28	333.85	175.06	6.57

Note. $F^{\uparrow}$ is the upward flux and $F^{\downarrow}$ is the downward flux; Net = $F^{\uparrow} - F^{\downarrow}$; Dnet1 = Net1 – Net2 and Dnet2 = $F_3^{\uparrow}$ – Net2; the mean is averaged data of Ref. 5; RMS is the rms deviation; LBL1 is the line-by-line calculation by Harshvardhan (Ref. 32); LBL2 is the line-by-line calculation by Fomin (Ref. 32); KD is the calculation with the use of *k*-distribution, MLS is the midlatitude summer.

The profiles of meteorological parameters are set, as a rule, on rather wide-meshed altitude grid, and for calculation of radiative cooling, it is necessary to calculate long-wave radiative fluxes. So, the simplest quadrature formulas are commonly used at almost all nodes of this grid. As a rule, two types of such formulas are used: the trapezoid formula in the case of small optical depth or the quadrature one with a weight function in the case of large optical depth.[18] (This is a standard approach for the case when a rapidly varying function is under the integral. Such a function is considered as a weight function, while the other one is described by an interpolating polynomial,[17] although Bakhvalov et al.[18] tried to assign a physical meaning to the latter quadrature formula.) The consequence of this is the use of the quadrature formulas with the residual common term which can hardly be calculated theoretically. By this reason, the most reliable estimate of the calculation accuracy is the Runge method,[17] which allows one to determine the optimum number of nodes for integration. Table 2 presents the values of the discretization step for the spectral region up to 3000 cm^{-1} at a preset accuracy ε. Taking into account the altitude distribution of absorbing gases, one can choose a non-uniform integration grid: narrow-meshed for the troposphere and wider-meshed at high altitudes. To achieve the accuracy of 0.1 W/m^2, the minimum number of nodes is about 200, whereas for the standard grid of the AFGL model[19] with 44 nodes ε ~ 6 W/m^2. To ensure the error no more than 1 W/m^2, about 100 nodes are needed. Our simulation has shown that to calculate the profiles of temperature, pressure, and gas concentration at missed points, the logarithmic interpolation should be used in case of significant variation of the meteorological parameters. In other cases the formulas of spline type are more suitable, because they allow a smooth function to be constructed by the given nodes. The procedure of interpolation contributes its own error into calculations. Therefore, an achieving of high accuracy due to the greater number of new nodes is illusive. The actually achievable accuracy of calculation of long-wave radiative fluxes is about 1 W/m^2 for the AFGL model.

Table 2. Accuracy of integration over height versus the discretization step

Absorbing constituents	Maximum integration step h, km	
	ε < 0.1 W/m^2	ε < 0.01 W/m^2
H_2O	0.19	0.056
CO_2	0.3	0.1
CH_4	1.9	0.62
$H_2O+CO_2+O_3+CH_4+N_2n$	0.18	0.048

Spectroscopic errors

To study the long-wave radiative fluxes sensitivity to uncertainties in spectroscopic line parameters, the statistical approach[21] was used by us based on the assumption that the line intensities and halfwidths are non-correlated random parameters and their mean values are not shifted. This approach has led us to the conclusion that the transmission functions with spectral resolution below 5 cm^{-1} are insensitive to uncertainties in the spectral line parameters. Therefore, the attention should be paid to systematic errors in integral intensities of spectral bands and line halfwidths. The estimates used in this paper are based on the results of detailed analysis of the HITRAN–96 data presented in Ref. 21. Of prime interest are the errors in initial spectroscopic information for three gases H_2O, CO_2, and O_3, which by 98–99% determine the long-wave radiative fluxes. The strongest 6.3-μm band of H_2O is sufficiently well studied. Relative error in the integral intensity for the spectral regions about 20 cm^{-1} wide can be estimated as 5–8%. These estimates have been obtained as a sum of errors of individual lines and, therefore, obviously are overestimated. Similar estimates were also obtained for errors in the H_2O rotational band. In the 8–12-μm window where weak lines lie, the errors in integral intensities are somewhat higher (about 10%). This is also the upper estimate obtained by summing up the errors of individual lines. In our opinion, the systematic error should be far less than the random error. The recent experiment[22] for the 6.3-μm band of H_2O indirectly confirms our assumptions. In the plots (Fig. 1) given for the 1450–1700-cm^{-1} spectral region the transmission functions error did not exceed ±0.05, and its behavior was oscillating about zero line. Since our access to experimental data is limited, we have chosen for testing the 1.4-μm band (see Fig. 1), because, similarly to 6.3-μm band, its parameters are also determined with sufficiently high accuracy. The deviations of the calculated data from the experimental ones were calculated by the formula $\delta = \frac{1}{N}\sum_{i=1}^{N} |T_i^{\text{exp}} - T_i^{\text{calc}}|$ (Table 3).

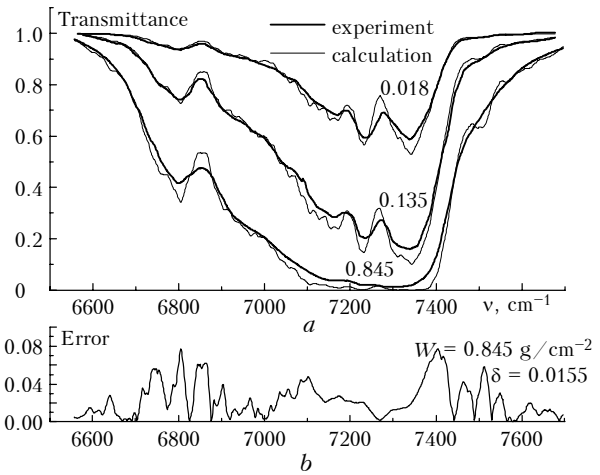

Fig. 1. Spectral transmittance of H_2O in the 1.4-μm band for three absorber masses W = 0.018; 0.135; and 0.845 g/cm^{-2}, spectral resolution of 50 cm^{-1}. The experimental data are borrowed from Ref. 23. Calculation by the direct method[24] (*a*) and the absolute error of calculation of the transmission functions (*b*).

Table 3. Experimental (T^{exp}) and calculated (T^{calc}) transmittance of the 4.3-μm band and the integral estimate of the calculation accuracy

W, g/cm^2	T^{calc}	T^{exp}	δ
0.018	0.869	0.870	0.014
0.135	0.642	0.647	0.019
0.845	0.411	0.426	0.023

The observed discrepancies between the experimental and calculated data can be explained by the systematic error of ~ 10–15% due to line intensities and halfwidths. It should be noted that Yamanouchi and Tanaka[23] presented only the effective pressure of water vapor, what contributed an additional error. Besides, the model we use for the continuum in this spectral region is, in our opinion, insufficiently correct. Therefore, the estimate of the systematic error should be even lower. It follows from the above-said that the approach used by Tjemkes et al.[25] for calculating the error as a sum of errors of individual lines gives very rough and overestimated results.

According to Ref. 21, the systematic error in the line parameters of 15-μm band of CO_2 varies from 2 to 7%; the accuracy of the data obtained for ozone is somewhat lower (the error achieves 10%). However, because the ozone molecule is heavy enough, the separation between lines is small and the lines significantly overlap. In this case the sensitivity of the transmission functions to uncertainties in line halfwidth is noticeably lower than for water vapor and CO_2. Therefore, the systematic error in the spectral line parameters in strong absorption bands can be integrally estimated for these three gases as 5–10%. For downward fluxes such errors lead to variations about 0.3–0.6 W/m^2 in the selective absorption coefficients of H_2O. The total systematic error about 10% in the CO_2, H_2O, and O_3 selective absorption having the same sign, can lead to variations in the downward flux about 1 W/m^2. This argues that the current databases afford sufficient calculation accuracy corresponding to capabilities of atmospheric experiments.

One more source of errors in calculation of long-wave radiative fluxes is the continuum absorption in macro- and microwindows. To illustrate the role of continuum, Fig. 2 presents the difference between the ascending fluxes at the upper boundary of the atmosphere and the downward fluxes at the level of the Earth's surface with considering and neglecting the continuum.

It is seen from Fig. 2, that the continuum absorption contributes not only in the 8–12-μm window, but also in the water vapor rotational band. The long-wave continuum is most pronounced for the polar latitudes.[26] In our calculations we used the model of continuum[14] which is sufficiently adequate to the experimental data obtained recently under clear-sky conditions.[6]

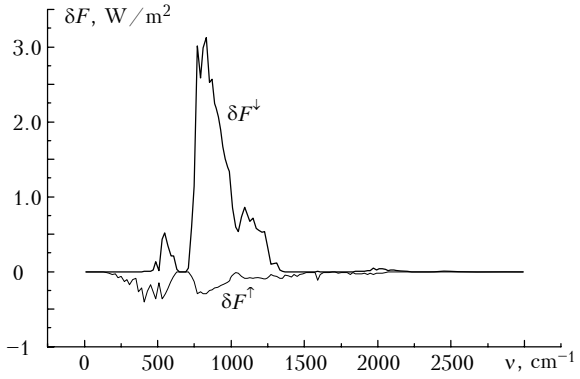

Fig. 2. Spectral contribution of the water vapor continuum absorption to the radiative fluxes: $\delta F = F_c - F$ (F_c is the radiative flux with allowance made for the H_2O continuum absorption, and F is the radiative flux with ignorance of the continuum absorption).

It should be noted that new modifications of this model[27] differ from the model from Ref. 14. The difference concerns mostly the 6.3-μm band and only slightly affects the spectrally integral fluxes (see Fig. 2). To obtain the estimates of errors in the coefficients of continuum absorption, an intercomparison of four models has been performed. Three models are a generalization of the laboratory experiments,[14,28,29] whereas Ref. 30 uses the data of field measurements. The three models are defined only in 8–12-μm region, therefore the data were intercompared there. Simulation was performed for downward fluxes, because of their strong sensitive to continuum absorption. The intercomparison for mid-latitude summer has shown that the models from Refs. 14 and 30 give close results, while the calculations by the models from Refs. 28 and 29 are close to each other, but differ from the above-mentioned results by 4–5 W/m^2. Our estimates have shown that such a spread corresponds to an error in the coefficients of continuum absorption of about 10%. The correction of the H_2O continuum temperature dependence proposed in Ref. 31 does not clarify the situation, since the discrepancies even increases. On the other hand, the atmospheric experiment described in Ref. 6 allows the conclusion that the model from Ref. 14 ensures the agreement between the experimental and calculated data with the rms error about 2 W/m^2. If this error is fully due to the continuum absorption, then the absorption coefficients calculated by the CKD model[14] are accurate to about 4–5%. The similar estimates are presented in Ref. 29. In view of the above-said, the main source of the error in calculation of fluxes is the uncertainty in the coefficients of continuum absorption. It should be especially emphasized that the contradictions between the data of observation over the intensity of outgoing[31] and downward[6] radiation remain and are to be explained.

Variations of CH_4 and N_2O concentration

The influence of the CH_4 and N_2O concentration variations on the radiative processes in the atmosphere were investigated. As expected, the increase in the concentration from 0 to 1.7 ppm for CH_4 and from 0 to 0.32 ppm for N_2O (values of the concentration corresponding to the ground conditions) leads to stronger absorption of radiation in the troposphere ~ 3 W/m^2 (tropical conditions), whereas further doubling of their concentration in the Earth's atmosphere results in relatively less absorption ~ 1.3 W/m^2. This is caused by the saturation effect in the absorption bands of these gases. Qualitatively close results were obtained for summer and winter conditions of the Northern Hemisphere mesozonal model, but their contribution decreased rapidly as the temperature decreased. Thus, for example, for winter conditions the ignorance of the gases contribution makes up 1.1 W/m^2.

An exclusion of the CH_4 and N_2O from the model for the conditions of midlatitude summer and tropics results in the error in the upward and downward long-wave radiative fluxes about 1%, whereas the error of the current methods of calculation is somewhat lower. In the case of doubling the concentration of these gases, the long-wave radiative fluxes change by no more than 0.6%. Table 4 presents the data on the variability of radiative cooling of the atmosphere. As is seen, the CH_4 and N_2O cause the atmosphere heating both in the troposphere and stratosphere. The energy contribution from these gases exceeds possible variations due to an uncertainty of the radiation absorption by water vapor. According to our estimates, these gases should be taken into account in the current radiative models. However, the effects caused by the increase in their concentration in the troposphere can hardly be taken into account nowadays, because the errors of the model for this altitude range are significantly larger. This is rather clearly illustrated in Fig. 3.

Table 4. Variability of the Earth's atmosphere radiative heating at varying concentrations of CH_4 and N_2O for the midlatitude summer, in W/m^2.

Midlatitude summer	Troposphere	Stratosphere	Entire atmosphere	Surface
Ignoring q m_4 and N_2O	− 2.74	− 0.06	− 2.80	− 0.97
Doubling of q m_4	0.47	− 0.04	0.43	0.21
Doubling of q m_4 and N_2O	1.15	− 0.09	1.06	0.52
Doubling of q O_2	3.78	− 2.64	1.14	1.56
5-% variations of the H_2O absorption coefficients				
Only selective	∓ 0.23	∓ 0.17	∓ 0.40	∓ 0.41
Taking into account the continuum	∓ 2.00	∓ 0.20	∓ 2.20	∓ 3.17

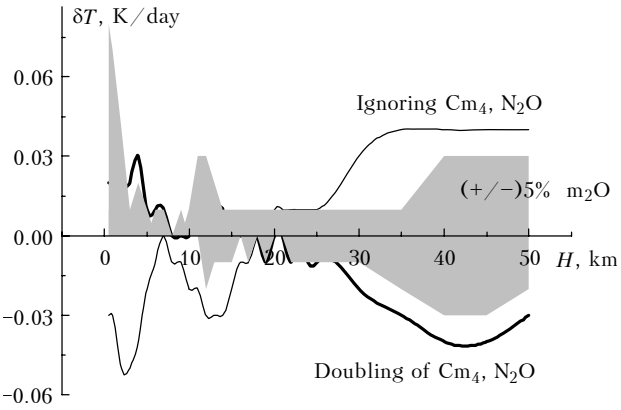

Fig. 3. Variability of the rate of radiative cooling δT (deg/day) in the case of exclusion of CH_4 and N_2O from the model and in the case of doubling of their concentration. The dashed area corresponds to 5-% variations in the H_2O absorption coefficients.

A different situation takes place in the case of doubling of the CO_2 concentration because in this case changes in fluxes magnitude in the troposphere and stratosphere are far larger than in the case of doubling concentration of CH_4 and N_2O. This fact does not contradict the statement that the CH_4 and N_2O concentration doubling produces the effect comparable with that for CO_2, if to consider the atmosphere as a whole. It is seen from Table 4 that the increase in the CO_2 concentration leads to strong heating in the troposphere and cooling in the stratosphere. These contributions to radiative processes are of different signs, therefore the integral estimates of the CO_2 energy contribution are comparable with the CH_4 and N_2O contributions which mainly causes the heating of the atmosphere and the Earth's surface.

Acknowledgments

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