Estimate of the influence of trace gases on long-wavelength radiation in the atmosphere

V.N. Krupchatnikov and L.I. Kurbatskaya

Institute of Computational Mathematics and Mathematical Geophysics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk

Received February 2, 2000

An estimate of the sensitivity of radiation fluxes and heating inflows, caused by the Earth's surface and long-wavelength radiation, to variations of trace-gas concentrations in the atmosphere is presented in this paper.

Trace-gas components (TGC), small amounts of which are universally present in the atmosphere, are active absorbers of IR radiation. These gases are O_3, CO_2, CH_4, and N_2O. The most active absorber among them throughout the spectral range from 4 to 120 μm is water vapor (H_2O). Ozone absorbs in the range $\lambda = 9.6$ μm. CO_2 plays an important part in radiation balance; it is well mixed in the atmosphere; its concentration increases every year and now amounts to 345 cm/m^3. The absorption of IR radiation takes place mainly within the 15 μm spectral range.

The next two gases in importance in IR absorption are CH_4 and N_2O. Methane is the carbon-containing component which is generated near the Earth's surface and is then gradually transported upwards to altitudes where it oxidizes, thus becoming a source of carbon monoxide and formaldehyde. Methane is not generated in the atmosphere chemically; instead its sources are the biosphere and the lithosphere. The CH_4 concentration at present is about 1.7 cm/m^3; it is well mixed in the troposphere, but beyond the tropopause its concentration decreases sharply because of oxidation and participation in reactions with other gases. The amount of methane in the atmosphere increases slowly from year to year by 1.2–1.5%. Nitrous oxide N_2O is generated in soil, particularly at higher temperatures and as a result of fertilizing with nitrogen salts. The N_2O concentration in the troposphere amounts to about 0.31 cm/m^3 and increases by 0.3% per year. IR absorption by CH_4 and N_2O takes place in the 7.25–8.15 μm spectral range.

In recent years, the problem of CO_2 increase in the atmosphere due to anthropogenic factors has received much attention. Quantitative analysis of the impact of CO_2 increase has led to the conclusion of a strengthening of the greenhouse effect and an elevation of temperature. Studies of the problem based on various climate models reduce to an examination of the sensitivity of the global climate to a doubling, quadrupling, and so on of the CO_2 concentration. Such studies are exemplified by the work of C.A. Wilson and J.F. Mitchel,[1] which present results of numerical experiments with the 11-layer model (AGC) which incorporates a model of the mixed upper ocean layer and a thermodynamical model of ice.

One of their experiments shows that a CO_2 doubling, followed by relaxation to a new stable equilibrium, leads to an elevation of the annual average temperature in the troposphere by ~ 5°C and to a decrease in the surface area covered with ice. In addition, global cloudiness was lowered by 3% at the expense of middle-layer clouds. This process was accompanied by some increase in cloudiness in the top and bottom layers at polar latitudes. The distribution of precipitation in the Northern hemisphere in summer also changed: in the high latitudes the precipitation increased while in the mid-latitudes above the continents it decreased. Obviously, the mechanism of formation of anomalies in the tropospheric mean temperature δT, surface temperature δT_s, cloudiness, *etc.*, is determined by anomalies in the impact functions such as fluxes of evident and latent heat near the surface, vortex fluxes of nonstationary perturbations, and radiative heating due to the presence of greenhouse gases.

In this work an attempt is made to describe the last of the above three factors leading to anomalies. In addition to CO_2, which traditionally is treated as the most important greenhouse gas, CH_4 and N_2O are considered, and their influence on IR radiation is estimated. To do this, we use the radiation model of a plane-parallel atmosphere in local thermodynamic equilibrium. The equations for the upwelling $F_i^{\uparrow}(p)$ and downwelling $F_i^{\downarrow}(p)$ fluxes can be written as[2,3]

$$F_i^{\uparrow}(p) = \{B_i(g) - B_i(p_s)\}\, T_i(p, p_s) + B_i(p) - \int_{p_s}^{p} T_i(p, p')\, \{dB_i(p')/dp'\}\, dp', \quad (1)$$

$$F_i^{\downarrow}(p) = \{B_i(top) - B_i(p_{top})\}\, T_i(p, p_{top}) + B_i(p) - \int_{p}^{p_{top}} T_i(p, p')\, \{dB_i(p')/dp'\}\, dp', \quad (2)$$

$$F_{i,\,net}(p) = F_i^{\uparrow}(p) - F_i^{\downarrow}(p),$$

$$H(p) = (g_0/C_p)\, d\{\sum_i F_{i,\,net}(p)\}/dp,$$

0235-6880/00/08 738-03 $02.00

where $F_i^{\uparrow}(p)$ and $F_i^{\downarrow}(p)$ are the upwelling and downwelling fluxes for the ith spectral range, $B_i(p)$ is the Planck function, p_s is the pressure at the Earth's surface, p_{top} is the pressure at the top of the atmosphere, $T_i(p, p')$ is the transmittance of the layer located between p and p', g denotes the ground layer, *top* denotes the top of the atmosphere, g_0 is the acceleration due to gravity ($g_0 \cong 9.80665$ m/s^2, $C_p = 1004$ J/kg deg), and $H(p)$ is the rate of radiative cooling.

$$T_i(p, p') = \exp\left[-\frac{\bar{S}_i u}{\sigma_i}\left(1 + \frac{\bar{S}_i u}{\pi' \gamma_{L_i}}\right)^{-1/2}\right], \qquad (3)$$

where $\bar{S}_i$ is the mean line intensity, σ_i is the mean spacing between lines, γ_{L_i} is the half-width of the Lorentz profile, and u is the amount of absorbing matter.

The transmittance $T_i(p, p')$ depends on three parameters: $\bar{S}_i/\sigma_i$, γ_i/σ_i, and u. The amount of absorbing matter u is found as

$$u = \int_z^{\infty} \rho \, dz, \qquad (4)$$

where ρ is the density of the substance under study. The parameters $\bar{S}_i/\sigma_i$ and γ_i/σ_i, listed in Table 1, are taken from Ref. 4 and augmented by data for CH_4 and N_2O from Ref. 5.

In the case of overlapping absorption bands of different gases, the mixture transmittance is found by the multiplication law. The H_2O absorption function in the continuum range of the 8–12 μm window is described by the relation[2]

$$T(p, p') = \exp\{[-k_1\Phi_1(\theta)p + k_2\Phi_2(\theta)e]u\}, \qquad (5)$$

where $k_1 = 0.1g^{-1}$ cm$^2 \cdot$ atm^{-1}, $k_2 = 20g^{-1}$ cm$^2 \cdot$ atm^{-1} (at $\theta_0 = 263$ K);

$$\Phi_1 = (263/\theta)^{-1.5}; \quad \Phi_2 = (263/\theta)^{6.5}; \qquad (6)$$

where e is the absorption elasticity and u is the mass fraction of H_2O (g/g).

The long-wavelength spectrum in the radiation model is divided into 17 spectral intervals with the transmittance T_i in each given by formula (5). These transmittance values are then used in equations (1) and (2) to find the upwelling and incident fluxes. Along a vertical, an 18-level model is considered which sets the boundary conditions at the top and bottom boundaries of the atmosphere. Numerical experiments were carried out based on the available data on the altitude distribution of the temperature, relative humidity, pressure, and ozone concentration. The total amount of CO_2 was taken to be constant whereas the CH_4 and N_2O amounts were varied. To estimate the influence of TGC's on long-wavelength radiation, the following combinations were used: 1) greenhouse gases absent; 2) H_2O; 3) CO_2; 4) CH_4; 5) N_2O; 6) $H_2O + CO_2$; 7) $H_2O + CO_2 + CH_4$; 8) $H_2O + CO_2 + CH_4 + N_2O$; 9) $H_2O + CO_2 + 2CH_4 + N_2O$; 10) $H_2O + CO_2 + CH_4 + 2N_2O$.

Table 1. Parameters of the statistical band model in the IR range

Range, cm^{-1}	$\bar{S}/\sigma$, cm$^2 \cdot$ g^{-1}	$\pi\gamma_i/\sigma$
Rotational band of H_2O		
40 – 160	7210.30	0.182
160 – 280	6024.80	0.094
280 – 380	1614.10	0.081
380 – 500	139.03	0.080
500 – 600	21.64	0.068
699 – 720	2.919	0.060
720 – 800	0.386	0.059
800 – 900	0.0715	0.067
15-μm band of CO_2		
582 – 762	718.7	0.448
6.3-μm band of H_2O		
1200 – 1350	12.65	0.089
1350 – 1450	134.47	0.230
1450 – 1550	632.9	0.320
1550 – 1650	331.2	0.296
1650 – 1750	434.1	0.452
1750 – 1850	136.0	0.359
1850 – 1950	35.65	0.165
1950 – 2050	9.015	0.104
2050 – 2200	1.529	0.116
6 – 9.2-μm band of CH_4 and N_2O		
1060 – 1180 (N_2O)	27.30	0.39
1180 – 1240 (CH_4, N_2O)	90.40, 3240	0.17, 0.40
1240 – 1300 (CH_4, N_2O)	1325.9, 1616.7	0.24, 0.87
1300 – 1360 (CH_4, N_2O)	2800.1	0.28, 0.45
1360 – 1320 (CH_4)	109.4	0.39
1420 – 1680 (CH_4)	10.9	0.138

The results show that in the absence of trace gases the flux $F_s^{\downarrow}$ incident on the Earth's surface is zero; the upwelling flux $F_{top}^{\uparrow}$ at the top of the atmosphere is equal to $(\sigma T_s^4 \times 0.996)$. Table 2 is divided into two parts. The top part lists downwelling and upwelling fluxes for individual absorbing components, and the bottom one – these same fluxes for combinations of these components.

Table 2. Downwelling $F_s^{\downarrow}$ and upwelling $F_{top}^{\uparrow}$ fluxes for individual trace gases and combinations

Gas	$F_s^{\downarrow}$(W/m^2)	$F_{top}^{\uparrow}$
H_2O	265	361
CO_2	76	412
CH_4	16	446
N_2O	11	452
$H_2O + CO_2$	309.3	322
$H_2O + CO_2 + CH_4$	313.2	315.9
$H_2O + CO_2 + CH_4 + N_2O$	315.6	313.9
$H_2O + CO_2 + 2CH_4 + N_2O$	316.6	312
$H_2O + CO_2 + CH_4 + 2N_2O$	317.0	312.5

Table 3. Rates of radiation cooling $\partial T/\partial t$ (deg/day) for individual trace gases and combinations as functions of p (mB)

$\partial T/\partial t$ p, mB	H_2O	CO_2	CH_4	N_2O	$H_2O + CO_2$	$H_2O + CO_2 +$ $+ CH_4 + N_2O$	$H_2O + CO_2 +$ $+ 2CH_4 + N_2O$
2.27	− 0.3	− 1.15	− 0	0	− 1.47	− 1.473	− 1.479
18.70	− 0.4	− 1.20	− 0	0	− 1.56	− 1.56	− 1.589
52.50	− 0.24	− 0.70	0	0	− 0.95	− 0.95	− 0.958
96.70	− 0.21	− 0.34	0.016	0.014	− 0.56	− 0.55	− 0.554
156	− 0.44	− 0.05	0.042	0.022	− 0.49	− 0.46	− 0.452
223	− 1.12	− 0.104	0.048	0.025	− 1.23	− 1.20	− 1.195
297	− 1.90	− 0.177	0.037	0.020	− 2.7	− 2.06	− 2.059
378	− 2.04	− 0.208	0.022	0	− 2.19	− 2.19	− 2.195
458	− 1.74	− 0.217	0	− 0	− 1.84	− 1.85	− 1.850
542	− 1.5	− 0.202	− 0.014	− 0.030	− 1.58	− 1.59	− 1.579
624	− 1.48	− 0.206	− 0.038	− 0.057	− 1.41	− 1.40	− 1.390
703	− 1.41	− 0.198	− 0.062	− 0.085	− 1.26	− 1.24	− 1.224
777	− 1.45	− 0.193	− 0.086	− 0.115	− 1.23	− 1.20	− 1.187
844	− 1.62	− 0.196	− 0.112	− 0.144	− 1.34	− 1.30	− 1.290
901	− 1.87	− 0.216	− 0.144	− 0.173	− 1.56	− 1.52	− 1.503
947	− 2.11	− 0.238	− 0.175	− 0.198	− 1.78	− 1.73	− 1.714
980	− 2.48	− 0.308	− 0.218	− 0.219	− 2.16	− 2.12	− 2.106
998	− 3.43	− 0.518	− 0.263	− 0.232	− 3.26	− 3.22	− 3.200

As can be seen, the strongest absorbers of long-wavelength radiation are H_2O and CO_2. Inclusion of CH_4 and N_2O leads to an increase in the incident flux and a decrease in the upwelling one. Doubling of CH_4 or N_2O amplifies this effect. At the same time, Table 2 demonstrates the nonmonotonic character of the influence of variations of the CH_4 and N_2O concentrations on atmospheric cooling. To find the cause of this phenomenon, we consider a spectral range $\Delta\nu_i$, that includes the necessary spectral lines of the gases under study. Let T_i^k be the transmittance of the mixture; then the cooling associated with the upwelling radiation at a level z is

$$\frac{dF_i^{\uparrow}}{dz} = \pi B_i(z)\,\frac{dT_i^k}{dz}\,. \qquad (7)$$

Similarly, for the downwelling radiation

$$\frac{dF_i^{\downarrow}}{dz} = -\,\pi B_i(z)\,\frac{dT_i^k}{dz} \qquad (8)$$

and for the efficient radiation

$$H_i^k = \frac{dF_{\text{net},\,i}^k}{dz} = -\,\pi B_i(z)\,\frac{dT_i^k}{dz}\,. \qquad (9)$$

We add one more gas having fundamental lines in the same spectral range to the mixture of k gases. The efficient cooling H_i^{k+1} in this case is

$$H_i^{k+1} = \pi B_i(z)\,\frac{dT_i^{k+1}}{dz}\,, \qquad (10)$$

where $T_i^{k+1} = T_i^k\,T_i^1$. If adding the gas causes a decrease of cooling, i.e., $H_i^{k+1} < H_i^k$, then it follows from (8) and (9) that

$$\frac{dT_i^{ki}}{dz} - \left(\frac{T_i^k}{1 - T_i^{k+1}}\right)\frac{dT_i^{k+1}}{dz} < 0. \qquad (11)$$

The derivative of the transmission function in (10) is found from formula (5)

$$\frac{dT_i^k}{dz} = C^k T_i^k \quad \text{and} \quad \frac{dT_i^{k+1}}{dz} = C_i^{k+1} T_i^{k+1}, \qquad (12)$$

where C^k and C^{k+1} depend on the ratio of the mixtures q^k and q^{k+1}, the absorption line half-widths γ_i^k, and γ_i^{k+1}, and the mean line intensities S_i^k and S_i^{k+1}. After substituting for the derivatives of the transmission function in (10) using relations (11) we have

$$\frac{T_i^{k+1}}{1 - T_i^{k+1}} > \frac{C^k}{C^{k+1}}\,. \qquad (13)$$

When treating particular models of gaseous mixtures, the quantities C^k and C^{k+1} are calculated and criterion (13) can be easily tested.

Table 3 demonstrates that introducing CH_4 and N_2O results in a diurnal lowering of the cooling in the troposphere by ~ 0.02°. Doubling of the CH_4 concentration leads to a diurnal decrease of the cooling below 549 mB by ~ 0.02°.

It follows from the above-said that when modeling possible climate variations for regions with elevated CH_4 and N_2O concentrations, these gases should be taken into account in the radiation block of the AGC model.

Acknowledgments

The work was partially supported by the Russian Foundation for Basic Research (Grants No. 98–05–65318 and No. 00–05–65459a).

References

1. C.A. Wilson and J.F. Mitchell, J. Geophys. Res. **92**, 315–343 (1987).
2. W.T. Roach and A.A. Slingo, Quart. J.R.M.S. **105**, No. 445, 603–615 (1979).
3. R.M. Goody, Quart. J.R.M.S. **78**, 165–169 (1952).
4. C.D. Rodgers and C.D. Walshaw, Quart. J.R.M.S. **92**, No. 391, 67–93 (1966).
5. E.V. Rozanov, Yu.M. Timofeev, and V.A. Frolkis, Izv. Akad. Nauk SSSR, Ser. Fiz. Atmos. Okeana **17**, No. 4, 384–390 (1981).