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Simulation of seasonal geographic patterns of methane using observation data*

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The three-dimensional global climatic model is employed as a tracer transport model to the study of atmospheric distributions of methane. Methane is input at appropriate surface grid points. The 4-year model experiment was run using sparse observed data at 19 sampling sites for the period 1984–1987, by which time a stable north-south gradient in the atmospheric methane and seasonal variations are obtained in the model. Verification of the climatic model as a tracer transport model indicates good agreement in the northern mid-latitudes with the known experiments of the 3D model synthesis of the global methane cycle.

1. Introduction

Methane (CH_4) absorbs outgoing infrared radiation over the range of wave lengths in the vicinity of 7.7 microns and, thus, it plays an important role in the thermal radiation balance of the Earth's atmosphere. It is assumed that 15 per cent increase in the methane concentration from 700 ppbv in the pre-industrial period to more than 1700 ppbv today has occurred from anthropogenic emissions. Methane plays also the major role in the atmospheric chemistry, being the net sink for the tropospheric hydroxyl radical OH. Now all the main sources of CH_4 and sinks have been established. However, yet there are uncertainties in source strengths. The expansion and the specification of our knowledge of the geographic and seasonal distribution of sources and sinks of CH_4 are necessary, if we want to develop an effective strategy of our actions to lower methane fluxes to the atmosphere.

It is accepted to consider three main sources of methane emissions:

- the biological activity under anaerobic conditions (natural wetlands, rice paddies, enteric fermentation in ruminants, etc.);
- transmission losses in the process of production and consumption (natural gas, petroleum, coal, etc.);
- burning and decomposition of biomass.

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One of the main ways of defining CH_4 source strengths is, in fact, the direct methane flux measurements. Unfortunately, methane emissions depend on many meteorological and biological parameters and vary widely in space and time. It makes the problem of interpolation (analysis) of measurements, derived from the surface sampling sites, to other points over the Earth's surface, very difficult. By virtue of it it is possible to expect that solution of this problem will have large errors in some areas, where there are no regular observations. The continuous records of measurements of CH_4 from the global network were made by the National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL). The data set of the measurements from the network of flask sampling sites between 1983 and 1993 represents the most complete set of methane concentrations in the surface layer of the atmosphere. The long-standing observations of climatic important gases, including methane, are a basis to examine the role of atmospheric motions (advection, convection) and its sources and sinks. Measurements are limited in space and time. The global characteristics of the distribution of gas components can be derived with the help of the 3D model of the general atmospheric circulation. By the present time, using climatic models there have been carried out studies on the reproduction of the global distribution of greenhouse gases, such as CO_2, chlorofluorocarbons ($CFCl_3$, CF_2Cl_2) [1, 2]. There have been performed numerical simulations of the global methane cycle, reproducing accurately enough the locations of sources and sinks of CH_4, its seasonal cycles [3].

In this paper, on the basis of the global climatic model [4], complemented by the tracer-methane transport scheme, an attempt has been made to assimilate the available data on concentration of CH_4, obtained from the extensive network of surface stations for the period of 1984–1987. The analysis made on the basis of this model has shown that it reproduces the main observed features of the large-scale transport of methane, such as the north-south gradient, the interannual trend and seasonal variations. This provides a basis for the further improvement and development of the model, including complex interactions with surface and subsurface environments, for greater understanding of the global methane budget and its sources and sinks.

2. Model

The numerical simulation of the global methane distribution in the atmosphere has been done with the 3D climatic model of the atmospheric dynamics, in which the trace gas transport was included [4].

The continuity equation governing the transport of the atmospheric tracers may be written in the following general form:

$$\frac{\partial \rho c}{\partial t} + \text{3DADV} = \text{SOURCE}. \tag{1}$$

The complete 3D advection 3DADV may be divided into three components:

$$\text{3DADV} = \text{HORADV} + \text{VERADV} + \text{VERCONV},$$

where HORADV denotes the horizontal advection of tracer, VERADV is the vertical advection of tracer, VERCONV is the tracer transport due to the vertical convection. The vertical convection (subgrid vertical fluxes) consists of the two parts:

$$\text{VERCONV} = \text{CLOUDCON} + \text{VERDIFF},$$

where CLOUDCON is the tracer transport by convective clouds, VERDIFF is the turbulent transport of tracer which depends on the static stability of the atmosphere. All the listed mechanisms of the tracer transport, except for CLOUDCON, are reproduced by the general circulation model ECSib [4]. The absence in the model of such an important mechanism of transport as CLOUDCON is its lack as a transport model (this disadvantage is supposed to be removed by introduction of the new parameterization of convective cloudiness like "mass–flux"), but even in this case it is possible to obtain the reasonable climatic distribution of methane in the atmosphere as a result of modeling.

Changes of the horizontal winds, temperature, the surface pressure are determined by solving of the conservation equations for momentum, mass, and energy. These equations together with the hydrostatic assumption and the equation for conservation of water substance form a closed system to define the climatic characteristics of quasi-stationary atmospheric motions. It should be noted that the given system of equations has the following integrated laws of conservation in time: mass, complete energy, angular momentum, potential enstrophy, specific humidity. The equations are presented in the spherical geometry with σ-coordinate in vertical. In the model except for the boundary conditions, the distribution of geopotential height is determined, and the ocean surface temperature, the geographical distribution of the ice cover, the latitudinal distribution of the solar angle of declination are set, and the temperature and humidity of soil are calculated.

The physical block of the model includes schemes for parameterization of subgrid-scale processes: radiative heating, turbulent and convective exchanges, condensation of water vapor.

The spatial difference method for the numerical solution of the model equations is based on Arakawa's C-grid [5], which approximately conserves the discrete analogues of basic integrated invariants of the system [6]. The numerical integration in time has been done with application of the semi-implicit scheme [7]. The model has a horizontal resolution of 4° latitude ×

5° longitude, and the vertical structure includes 15 σ-levels, four levels of which are located in the boundary layer, seven – in the troposphere and four – in the stratosphere. The results of the numerical integration of the general circulation atmosphere model have shown that the major features of the global climate can be realistically simulated [8].

The methane transport equation has been included in the given climatic model which in the flux form is as follows:

$$\frac{\partial c}{\partial t} = -\nabla_\sigma(\vec{V} \cdot c) - \frac{g}{p_s}\frac{\partial}{\partial \sigma}(\dot{\sigma} c) + S, \tag{2}$$

where S is methane sources and sinks, c is the volumetric mixing ratio.

The vertical methane fluxes in the surface air layer are parameterized on the basis of the Monin–Obukhov similarity theory in much the same way as the vertical fluxes of water vapor by the analytical approximations which are different in conditions of non-neutral stability [4]. Above the constant flux layer the vertical fluxes are parameterized by the mixing length theory, i.e.,

$$M^T = \rho^2 \frac{g}{p_s} K_M \frac{\partial c}{\partial \sigma}, \tag{3}$$

where the vertical diffusion coefficient K_M, as well as earlier, is set equal to the vertical diffusion coefficient for water vapor.

The tendency caused by turbulent methane fluxes is obtained by solving the diffusion equation

$$\frac{\partial c}{\partial t} = -\frac{g}{p_s}\frac{\partial}{\partial \sigma}(M^T) \tag{4}$$

with application of the implicit scheme in time. Similar to the conservation equation for water vapor, the continuity equation for methane is integrated using a centered-difference scheme and after each time step, the negative mixing ratios are excluded according to the vertical diffusion scheme [7].

3. Experiment

The numerical experiment in the modeling of the global atmospheric tracer-methane was carried out as follows.

The objective was to simulate on the basis of climatic model the global distribution and seasonal variations of the CH_4 mixing ratios within the model year, using the monthly-averaged surface concentrations for the 4-year observational period. This has provided an excellent check of the ability of the model realistically to describe the redistribution of trace gas and to define the agreement of model predictions with measurements.

The data set of the monthly-averaged CH_4 concentrations represents the arithmetic mean of weekly measurements from 19 sampling sites, located

between 82° N and 90° S [9]. Stations included in the NOAA/CMDL network are reference sites for the long-term monitoring of pollutants CO_2, CH_4, N_2O, which are essential for the climate formation. Most of the measurement sites are in remote locations, far from continental sources and sinks. The three stations (Barrow, Station "M", South Pole) from the NOAA/CMDL network have been singled to present spatial and temporal changes of CH_4.

The sampling site at Barrow (71°19′ N, 156°36′ W, 11 m above mean sea level (MSL)) is located on the Arctic coastal seashore of Alaska. The analysis of air samples for CH_4 was begun in April 1983. The annual mean CH_4 mixing ratio at Barrow rose from 1724.1 parts per billion by volume (ppbv) in 1984 to 1806.5 ppbv in 1992. The average rate of increase over the period of record was equal to 11.1 ppbv/year [9]. On the basis of the data for 1984–1987 it has been established that all of the northernmost NOAA/CMDL sites, including Barrow and Station "M", have recorded the major seasonal minima of the CH_4 concentrations during the summer (usually in July).

The Station "M" (66°00′ N, 2°00′ E, 4.5–9 m above MSL) is located in the open North Atlantic Ocean west of Norway. The annual mean atmospheric CH_4 mixing ratio at this station rose from 1708.5 ppbv in 1984 to 1793.8 ppbv in 1992. The average growth rate over the period April 1983 – December 1992 was 10.7 ppbv/year [9]. The seasonal variation of CH_4, observed at Station "M", is characterized by the minimum of mixing ratios during the summer months. Primary and secondary seasonal maximum occurred during the late winter and early spring months.

The annual mean CH_4 mixing ratio at the NOAA/CMDL South Pole (89°59′ S, 24°48′ W, 2810 m above MSL) increased from 1576.0 ppbv in 1984 to 1666.9 ppbv in 1992. Over the period of record (February 1983 – December 1992), the average CH_4 growth rate was 11.5 ppbv/year [9]. Over the 4-year period (1984–1987) both the mixing ratio and the phase and amplitude of the seasonal cycle were indistinguishable, within the limits of the measurement precision, at the three southernmost sites in the NOAA/CMDL network: South Pole, Palmer Station, Cape Grim.

The sampling sites (Barrow and Station "M") represent the northern polar latitudes which are subject to natural and anthropogenic methane sources. As all the three stations are removed from methane sources and sinks, this allows isolating the impact of dynamic processes on the CH_4 distribution throughout the height of the atmosphere. As to all the data set from 19 NOAA/CMDL stations, their major feature is the existence of the north-south gradient of the atmospheric methane with the difference of 150 ppbv (for the annual mean concentration) between the extreme northern and the extreme southern stations. For the high southern latitudes, a seasonal cycle of methane has been revealed with a maximum in the autumn (September–October) and with a minimum in February. The typical amplitude of a seasonal cycle was $\sim$ 30 ppbv. In the northern latitudes, a

seasonal cycle is more complex and is caused by seasonally varying natural sources. It is known that almost 75% of methane sources are in the northern hemisphere. The obtained annual mean of the CH_4 concentration contained the increasing trend, on the average, 0.5–1% between 1984–1987.

The initial values of the CH_4 concentration used as the input data at regular gridpoints were calculated by application of the correction method from the observed mixing ratios at surface stations [10].

The major principle of the numerical analysis is the following. If there are N stations giving contributions to the concentration at the gridpoint, then the concentration at this gridpoint will be

$$c_i^{(n+1)} = \frac{\eta^{(n)} c_i^{(n)} + \sum_{j=1}^{N} [f(\rho)\mu c]_j}{\eta^{(n)} + \sum_{j=1}^{N} [f(\rho)\mu]_j}, \tag{5}$$

where $f(\rho)$ is the factor taking into account the density of stations surrounding a particular station; ρ is the number of stations within a radius of influence; μ is the weighting function; n is the number of modifications; $\eta^{(n)}$ is the sum of weights for n-th modification; $c_i^{(n)}$ is the analyzed field for the n-th modification, used as preliminary field for the $(n+1)$-th modification. The shape of the weighting function, μ, and the influence function, $f(\rho)$, were determined according to the approximations presented in work [10]. After the five-fold application of formula (5) the error of the analysis was no more than 2% when comparing the observed concentrations with interpolated concentrations at any station.

4. Model results

Probably, one of the main results of the performed research is that the climatic model including the methane transport allows us to interpret rather sparse ground-based data. There have been simulated reasonable spatial and temporal structures of CH_4 fields on the basis of surface measurements.

The presented maps (Figures 1–4) of the annual mean surface CH_4 concentration for the period 1984–1987 not only show the growth of the content of the atmospheric methane for this period but actually give the information about its major sources and sinks. Figure 5 shows the simulated field of the CH_4 concentration, at $\sim$ 960 mb, averaged over the 4-year period and determined relative to the global annual mean concentration. At the Earth's surface in the northern mid-latitudes the main characteristic feature of the distribution of methane is steep meridional gradients generated by the

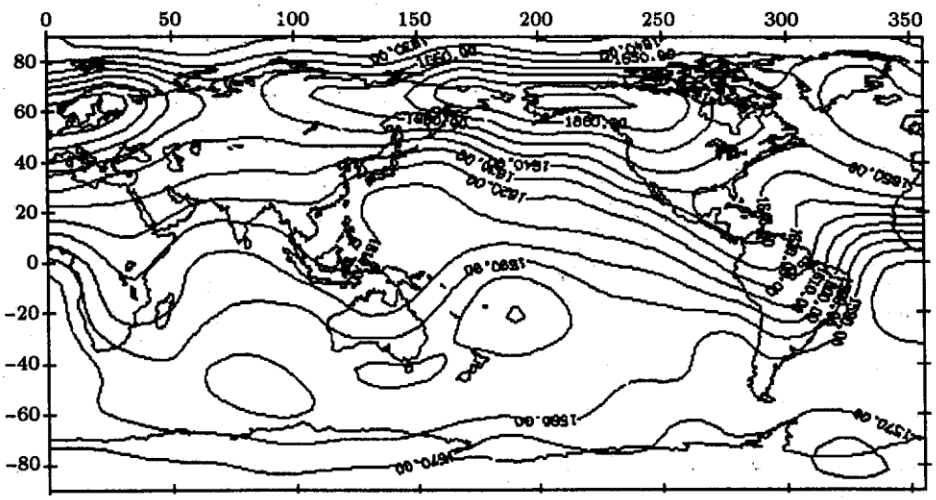

Figure 1. Analyzed global distribution of CH_4 (in ppbv) at 1000 mbar, annual average of 1984

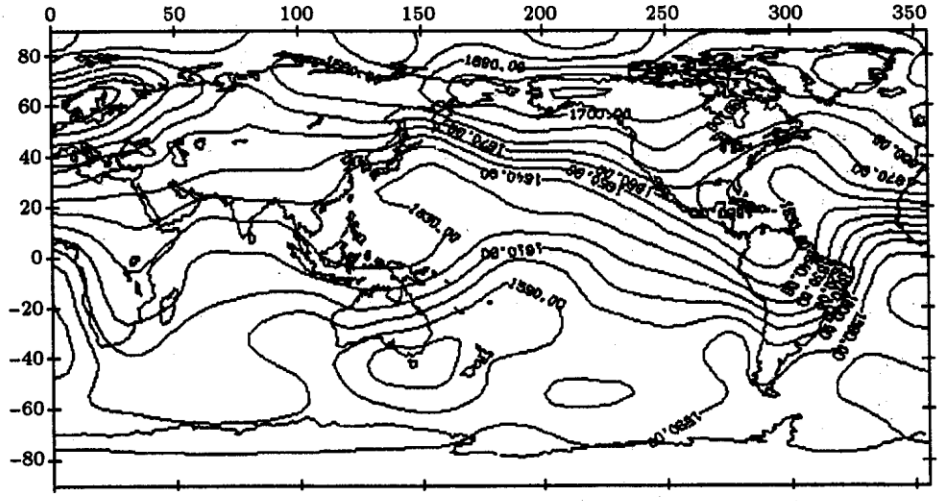

Figure 2. Same as Figure 1, except annual average of 1985

continental sources (natural – wetlands (bogs, the tundra); anthropogenic – the mine of fossil fuel (coal, gas), rice paddies, animals (ruminants), landfills). The northern and the southern tropics have appeared to be the most complex regions to simulate the methane distribution. The surface field of CH_4 points to the presence of significant gradients in the zonal direction. It is here where the global meridional transport of CH_4 is less expressed, and this is not agreed with observational data (see Figure 5).

Changes in the transport rate caused by convection in the atmospheric boundary layer, the change of the global circulation intensity and seasonality

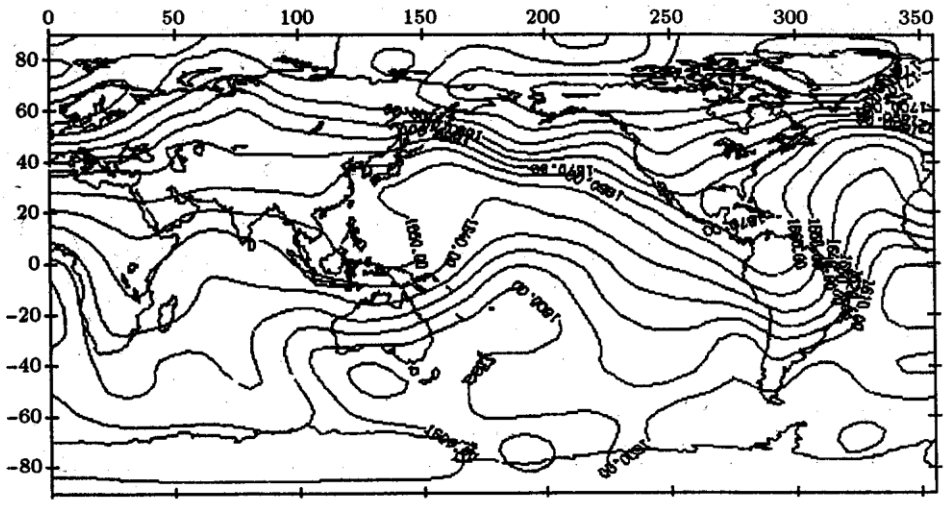

Figure 3. Same as Figure 1, except annual average of 1986

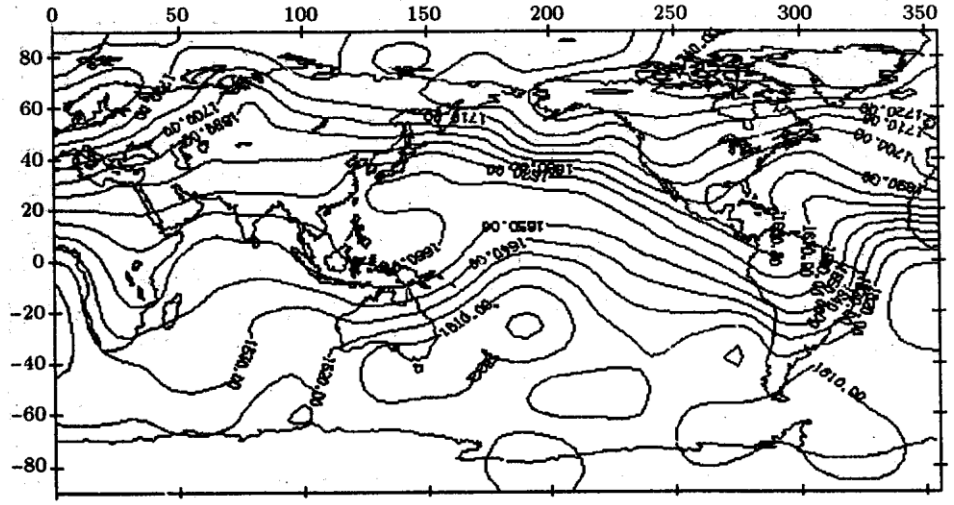

Figure 4. Same as Figure 1, except annual average of 1987

of the natural methane sources result in essential seasonal variations of the CH_4 concentrations. The difference between concentrations at $\sim$ 1000 mb and in the middle troposphere ($\sim$ 500 mb) is represented in Figure 6. The presence of negative vertical gradients in the northern hemisphere over the continental source regions is clearly seen. As for the southern hemisphere, the chemical destruction of methane (the reaction with the hydroxyl radical OH in the troposphere) is the only active source/sink term which in the absence of other processes would result in a vertical profile that increases with height.

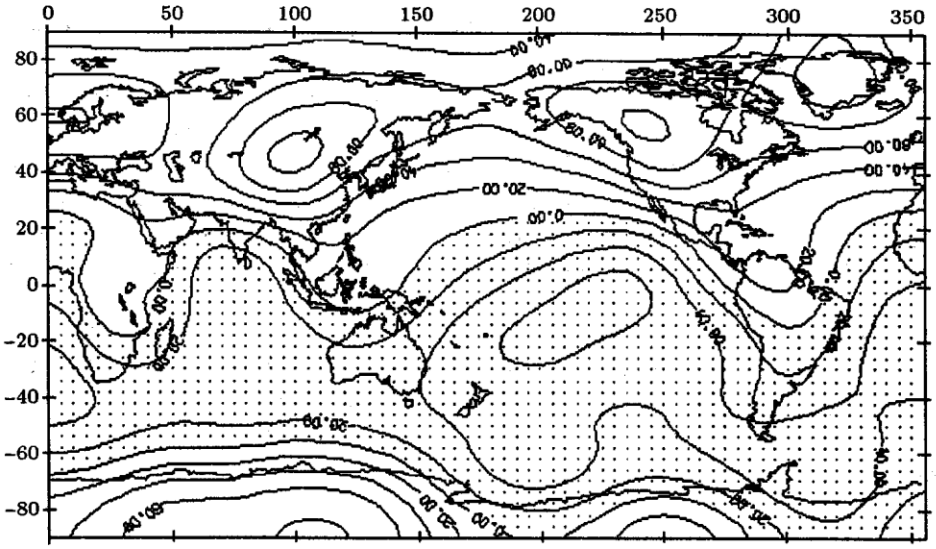

Figure 5. Simulated global distribution of CH_4 concentration defined relative to the global annual mean (in ppbv) at 960 mbar, annual average of model year

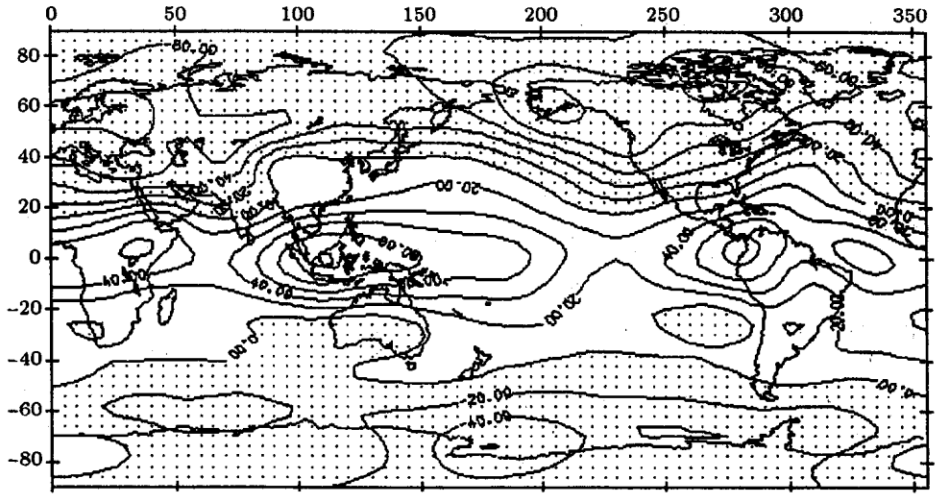

Figure 6. Simulated global distribution of the annual mean CH_4 concentration differences (in ppbv) between 500 and 1000 mbar

5. Conclusions

The surface measurements data for the CH_4 concentration are analyzed on the basis of the climatic model including the large-scale methane transport. The results of the 3D modeling represent that as the response to the observed CH_4 concentration in the atmospheric boundary layer, the model

simulation reproduces significant gradients over ground-based source regions in the northern mid-latitudes.

The presence of the characteristic features, such as seasonal cycles, the increasing trend and the north-south latitudinal gradient in the surface concentrations for the period 1984–1987 actually give the information about the methane sources and sinks at mid-latitudes. The same features are an opportunity for a more complete and adequate description of the processes leading to the methane emission to the atmosphere, its chemical destruction.

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