

## Modeling of methane flows in the atmosphere resulting from seabed fluid discharge

V.V. Malakhova

**Abstract.** Reservoirs of geological methane can occur within the sediments at any depth, right up to the seabed. Methane is also sequestered by gas hydrates. One mechanism to release methane from sediments to the ocean is through submarine mud volcanism, hydrocarbon seeps and vents. The 3D mathematical model of the dissolved gas transport by the ocean currents is used for the quantitative estimation of the scale of a possible methane flux into the atmosphere from the submarine fluid venting. The climatic large-scale currents field is obtained from the 3D global ocean dynamics model. The problem was numerically solved on a spatial grid with resolution of 3 degrees in latitude and in longitude with 24 levels on the vertical. In this case, a 3D equation of transport of dissolved methane from sources is simultaneously solved. Methane sources in this model are set taking into account a speed of a filtration methane flux. The calculated methane flux into the atmosphere exceeded 9–20 Tg/year, depending on the experiment scenario.

### 1. Introduction

Methane is an important greenhouse gas next to carbon dioxide. An increase of the methane content in the atmosphere activates the greenhouse effect, because methane intensively absorbs the Earth's heat radiation. According to modern ideas of the global warming mechanisms of the atmosphere due to the greenhouse effect, methane is responsible for 15% of the increase in the mean temperature. However, a potential contribution of methane to the climate changes is 20–27 times higher than that of carbon dioxide [1]. The yearly gain of methane in the atmosphere is 1–2%, which exceeds the carbon dioxide gain. Thus, for the last 40 years, the methane concentration has increased by approximately 30% and is now about 1750 ppb [2].

The contribution of the World ocean as a source of the methane global emission into the atmosphere is estimated by different authors within 5–50 Mt/yr at a yearly total of 460–1665 Mt/yr [1]. Thus, the contribution of the oceans is insignificant (a total of 2–5%). Other estimates show that methane flows from only the areas of their usual venting from the continental margins can give from 8 up to 65 Mt/yr [3, 4]. Such a difference can be explained by a poor knowledge of the abiogenic methane flows from the lithospheric sources, which include deposits of gas hydrates, hydrothermal midocean ridge (MOR) systems, fluid seeps, gas vents, mud volcanoes, etc. [1].

The submarine fluid venting has been studied since the twentieth century, [3]. Many approaches have been undertaken to study the methane

discharge from mud volcanoes and other cold seeps such as decomposing gas hydrates. Methane can rapidly dissolve and oxidize near to the seafloor [5]. This supports the conclusion that methane from oceanic sources contributes little to the atmospheric methane budgets, 5–15 Mt/yr, [6]. However, there is also evidence that methane may not oxidize in a higher water column for the time scales of several years to decades [5].

The recent measurements prove that the global venting of gas from the Earth's interior is rather active both from land and huge ocean areas. Geological sources of methane in the atmosphere are generally thought to be minor [7]. The yearly contribution of submarine geological sources, which earlier have never been taken into account, is estimated to be of order of 30–70 Mt/yr, [8] and 16–40 Mt/yr, [9, 10].

Geological methane is released to the hydrosphere and to the atmosphere by the two main pathways natural gas seeps and mud volcanoes. It is known that the submarine venting of fluids is characteristic of definite bottom zones, whose location and sizes are described in [11, 12]. A large majority of the fluid venting sites appear to be the result of the focused fluid migration through the sea floor, [11, 12]. The fluid discharge areas are mostly found at continental slopes. The results of numerous investigations show that gas hydrates are located close to these places immediately at the sea bottom. This connection confirms the fact that there is a continuous methane inflow for gas hydrates generation [13]. Studies of the submarine Haakon Mosby mud volcano have shown that gas hydrates at the sediment surface participate in the generation of a cold methane plume, which is distributed in the direction of bottom streams. Therefore, a methane flow into the water can reach approximately  $50 \text{ m}^3/\text{m}^2$  per year [13].

Fieldwork experiments in the East Siberian Sea and in the Laptev Sea revealed plumes with dissolved methane of a high concentration [14, 15]. It was experimentally found that the near-bottom water layer in the above areas was supersaturated with methane relative to its concentration in the atmosphere of high altitudes by 2200–3000% and relative to the surface waters by 700–2000%, which is evidence of a significant contribution of shelf waters of these seas into the methane emission into the atmosphere.

On the whole, the sea water is insufficiently saturated with methane as compared to the methane solubility all the way from the surface to the bottom; therefore, the world ocean is, in practice, an infinite reservoir for methane coming to water from the areas of the bottom fluid venting and captured by the ocean streams, as was observed in a decaying gas hydrate field in the caldera of the Haakon Mosby mud volcano in the North Sea [13], and then reaches the atmosphere. Thus, simultaneous measurements of methane were conducted in the near water atmospheric layer and in the surface water layer in the region of the East Siberian Shelf [15], which have shown that the regions of anomalously high methane concentrations in the

air (up to 8 ppm) spatially correspond to spots of high concentrations of gas dissolved in the sea water (up to 500 nmol). This work presents a model of methane transport from sources of submarine venting of the deep water fluids.

## 2. The model and experimental details

To quantitatively estimate the scale of a possible gas emission into the atmosphere from the seabed sources due to the submarine gas venting, a 3D quasi-geostrophic model of the world ocean dynamics [16] was used, supplemented with a model of dissolved methane transport by the ocean streams.

The climatic state of the ocean was found from the 3D linearized model of the world ocean dynamics, involving the seasonal variability with allowance for the actual topography of the bottom and the Arctic water reservoir.

The dissolved methane distribution was obtained from solving the advection-diffusion equation of the methane transport. A feature of this parameter is an interlatitude gradient directed from north to south with maximal concentrations in the Northern Hemisphere. Therefore, the dissolved methane distribution on the ocean surface was set as dependent solely on the latitude.

Further, the dissolved methane transport by the ocean streams from the model-set bottom sources at possible points of gas venting was studied, and the methane emission into the atmosphere was calculated.

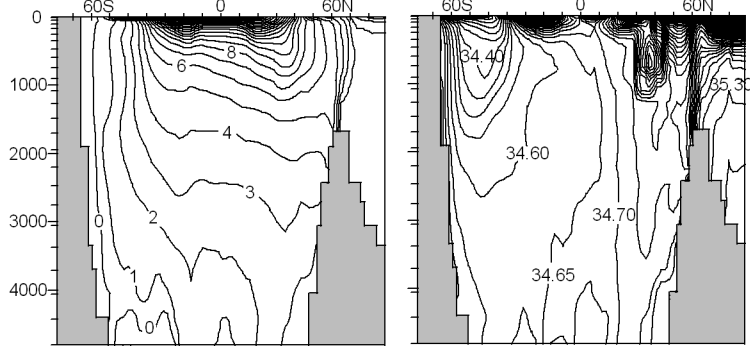
At the first stage, the initial thermohaline ocean state was determined. The problem was solved for a test site of the world ocean between  $76.5^\circ$  and  $88.5^\circ$  at a shift of  $1.5^\circ$  relative to the equator and the areas side boundary using a uniform grid with a  $3^\circ$  step in longitude and in latitude. The Mediterranean Sea was added to the simulated test site. To describe the thermocline range in greater detail, a vertical non-uniform grid with decreasing steps closer to the surface was used. Ices, located northward from  $80^\circ$  N and southward from  $70^\circ$  S, were taken into account implicitly at the cost of zero stresses of the wind friction and constant negative temperatures of the water surface.

Zonal-average temperature fields and salinity, found numerically, well agree with the observational data (Figure 1). Thus, the formation of an intermediate water mass with a lower salinity is distinctly expressed and corresponds to observations.

Beginning with this climatic state of the world ocean, the system of equations for the ocean dynamics is integrated at a time step of 10 days. Simultaneously, the 3D advection-diffusion equation of the water dissolved methane transport is solved in [17]:

$$\frac{\partial C}{\partial t} + \frac{u}{a \sin \theta} \frac{\partial C}{\partial \lambda} + \frac{v}{a} \frac{\partial C}{\partial \theta} + w \frac{\partial C}{\partial z} = \frac{\partial}{\partial z} \kappa \frac{\partial C}{\partial z} + \frac{\mu}{a^2} \Delta C, \quad (1)$$

with the boundary conditions:



**Figure 1.** Zonally averaged temperature (left) and salinity (right) fields in the world ocean, found from a numerical experiment by the model for an integration period of 5000 years

- at the ocean surface:  $C = C^*(\lambda, \theta, 0)$ ;
- at the side surface  $\Gamma$ :  $\mu \frac{\partial C}{\partial n} = 0$ ;
- at the ocean bottom  $z = H(\lambda, \theta)$ :  $\kappa \frac{\partial C}{\partial z} = 0$  (in the absence of a methane sources).

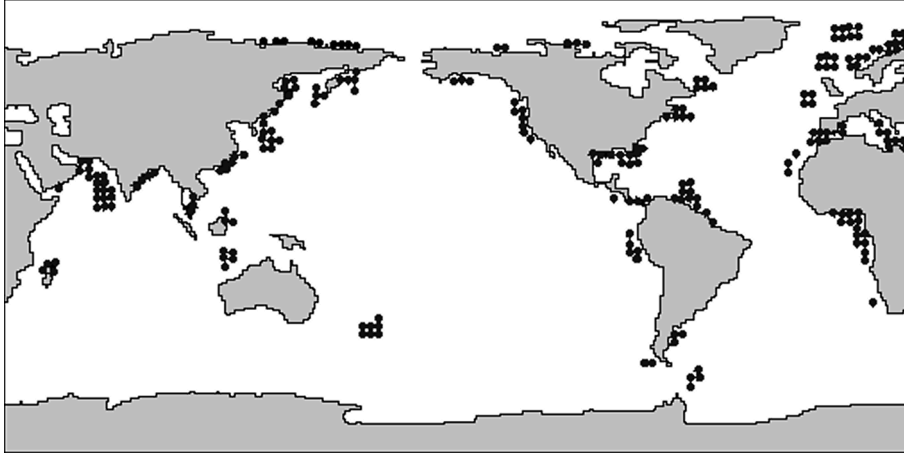
Here,  $C(\lambda, \theta, z)$  is a dissolved methane concentration;  $C^*(\lambda, \theta, 0)$  are given values of the methane concentrations at the ocean surface, which change from zone to zone between 50 ppb in the southern high latitudes up to 100 ppb in the northern high latitudes (1 ppb =  $10^{-9}$  l/l = 1 nl/l);  $u$ ,  $v$ , and  $w$  are velocity vector components by the coordinates  $\lambda$ ,  $\theta$ , and  $z$ , respectively, where  $\lambda$  is the longitude,  $\theta$  is an addition of the latitude to  $90^\circ$ , and  $z$  is directed vertically downward;  $\mu(z)$  and  $\kappa(z)$  are coefficients of the vertical and horizontal turbulent diffusion;  $a$  is the Earth's mean radius;  $t$  is time;  $H(\lambda, \theta)$  is the bottom relief;  $\Gamma$  is the regions side surface; and

$$\Delta = \frac{1}{\sin^2 \theta} \frac{\partial^2}{\partial \lambda^2} + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial}{\partial \theta}.$$

### 3. Results of the numerical experiments

As a result of geological studies of the world ocean, more than 100 areas of the submarine methane venting were found in [11, 12]. These areas are mainly located in peripheral ocean regions, covering shores, continental slopes, and near continental deep water grooves. In the model, the methane sources are set as located at the ocean bottom (Figure 2) (following [11, 12]).

Starting with the moment of establishing the climatic state of the world ocean (see Figure 1), two experiments have been conducted.



**Figure 2.** Model distribution of methane sources at the ocean bottom by data from [11,12]

In the first experiment, the model methane source was set as an increase in the methane concentration at some calculation bottom point where the gas venting was possible:  $C = C_I$  at  $z = H(\lambda, \theta)$ .

In the second experiment, the methane source was set with allowance for the methane filtering rate through a deposition layer:  $\kappa \frac{\partial C}{\partial z} = q_c$  at  $z = H(\lambda, \theta)$ .

The increased methane concentration at the near bottom points of possible sources was set equal to a 100 fold excess over the methane background value ( $C_I = 5000$  ppb).

Such an approach was confirmed by the methane concentration measurements in the ocean waters. In [18], an example of the Derugin depression is presented, where at station 228 with gas venting the methane concentration was 100-fold higher than at the neighboring station, where the methane venting was absent. Anomalously high methane concentrations of 5000–10000 and 20000 nl/l were found at the bottom layers in the area of methane bubble discharge at the Sakhalin northeast slope and in the Sea of Okhotsk. They exceed the methane background concentrations in the near bottom water by 100–1000 times and even more. In the area of methane flows from the kern of the upper depositions (100–400 cm), the methane concentration exceeded 500–1000 ml/l in the moist deposit [19]. The bottom surface in this region is presented by rounded structures 500–700 m in diameter with numerous small holes and protuberances formed of gasfluid flows from deep layers of the deposition mantle [19]. Direct geochemical tests in the region of the Haakon Mosby mud volcano gave abnormally high gas concentrations at a distance of 80 m above the volcano [13].

Studies of the shoal ocean areas have shown the methane emissions from deep water gas deposits to have a plume form, which allows methane to escape oxidation and reach the atmosphere [14]. When gas plumes are located deeper than 100 m, methane is concentrated in the water thickness in some layer up to the depth of dissolving gas bubbles. In the process of gas bubble ascending, the vertical water transport takes place in the gas plume itself, which is accompanied by a horizontal water movement to the plume center, which compensates the vertical water discharge. At the gas plume top, the density of the gaswater emulsion becomes equal to the density of the surrounding water.

This results in termination of the vertical movement and the horizontal distribution of emulsion, leading to the formation of a layer with the augmented methane content [18].

Further, the model transport of dissolved methane by the ocean streams in the world ocean is studied and the methane emission in the atmosphere is calculated. A methane flow through the ocean surface is a function of the surface water temperature  $T$  and the wind velocity  $V$  [14]:

$$F = a^2 q \int_{\Omega} 0.13 V^2 \left( \frac{\text{Sc}}{660} \right)^{-1/2} \left[ \kappa \frac{\partial C}{\partial z} \right] \Big|_{z=0} d\Omega,$$

where  $q = 0.7169 \cdot 10^3$  is a specific weight of methane and Sc is the Schmidt number:

$$\text{Sc} = 2039.2 - 120.31T + 3.4209T^2 - 0.04043T^3.$$

It is assumed that the whole of methane is dissolved in the sea water, and methane bubbles do not reach the atmosphere. Each experiment began with the same initial calculated climatic fields. The integration time of the numerical model and the methane transport equation spanned approximately 500 years. The diffusion methane flow into the atmosphere was found in each experiment.

In the first experiment, methane sources were set indirectly through the gas 100-fold increased concentration. The yearly methane emission equals 9 Tg/yr. The mean concentration of the dissolved gas increased from 50 up to 650 nl/l. The dissolved methane is transported by the ocean streams throughout the ocean.

In the second experiment, the effect of the scenario parameter, i.e., the source power, was tested. The methane source in the model was set with allowance accounting for the filtration emission rate of the methane dissolved in the porous water. For a filtration gas flow, the following formula was proposed in [13]:

$$q_c = V_f(C_p - C), \quad V_f = D/\delta,$$

where  $C_p$  is the methane solubility in the sea water,  $V_f$  is the ascending filtration rate,  $\delta = 1$  cm is the thickness of a diffusion sublayer, and  $D = 10^{-5}$  cm<sup>2</sup>/c is the coefficient of the methane molecular diffusion in the water.

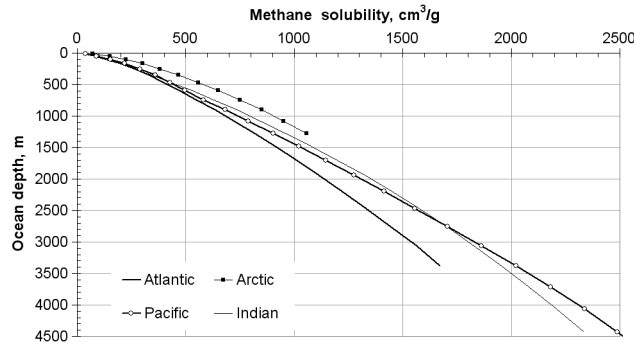
The expedient was used when a narrow diffusion sublayer was separated close to the bottom, in which the total transport was meant to be due to the molecular diffusion, only.

The methane solubility in the sea water was determined as in [20]:

$$C_p = 0.4342 \frac{\rho^{0.64}}{(T + 5.278)^{0.3882}} \text{ cm}^3/\text{g},$$

where  $\rho$  is pressure in kg/cm<sup>2</sup>, and  $T$  is temperature in °C.

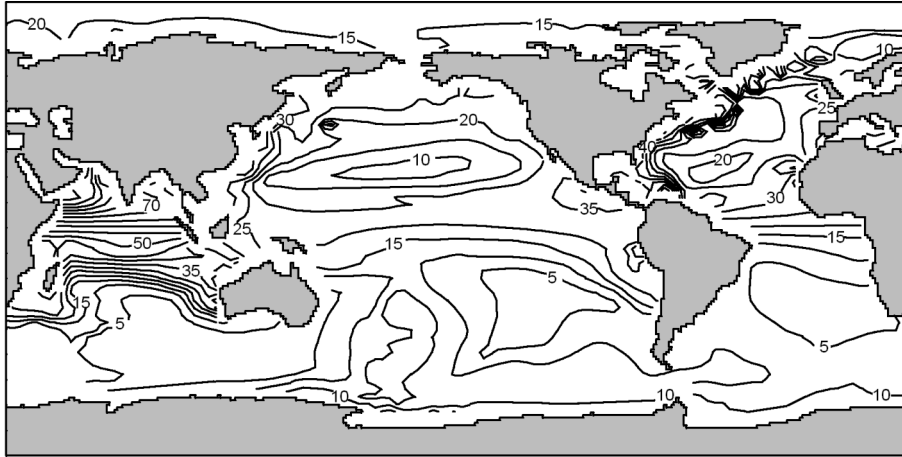
On the whole, the sea water is insufficiently saturated with methane as compared to its equilibrium values. The air-equilibrium methane concentrations are 0.05–0.08 ml/l. These values slightly increase with depth [13]. At the same time, the methane solubility already in the surface water reaches approximately 30–50 ml/l and quickly grows with depth, attaining values of 1200–2000 ml/l at the bottom (Figure 3).



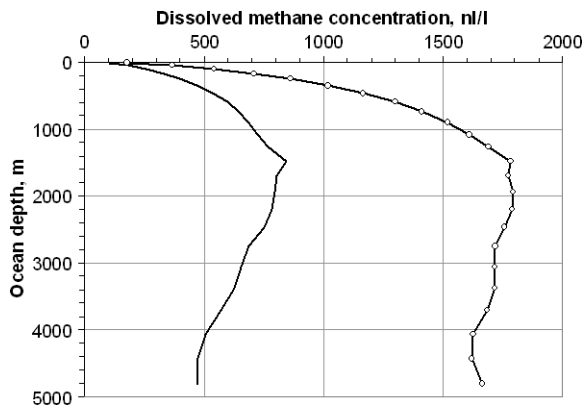
**Figure 3.** Methane solubility in sea water calculated for each ocean

At a gas concentration in water less than the solubility limit, methane is emitted into water as long as its concentration reaches a maximum magnitude under a given temperature and pressure. The gas solubility rate depends on the processes of the sea water mixing.

The source power in the second experiment depends on the gas solubility and is different at all model points. This results in increased methane concentrations in the gas venting zones located deep in the sea water, while in the shore areas, the source power decreases. The depth of the model integration was about 500 years before the establishment of the climatic state of the world ocean. In the second experiment, a maximum methane emission into the atmosphere increased, reaching 21 Tg/yr. The methane mean concentration in sea water was 1500 nl/l.



**Figure 4.** Distribution of the dissolved methane in the ocean at a depth of 250 m obtained from the second numerical experiment in 500 years of the model time. Values on isolines are in relative units (1 rel. un. = 50 nl/l)



**Figure 5.** Mean vertical profiles of dissolved methane obtained from the second numerical experiment in 50 (solid line) and in 400 years (line with a marker) of the model time

Figure 4 shows the horizontal distribution of the dissolved methane at a depth of 250 m found during the second experiment. The dissolved methane is transported by the ocean streams throughout the ocean. This process is under a strong influence of the horizontal oceanic circulation. Thus, high concentrations in the tropics of the Pacific Ocean are due to streams of the west direction. An advective methane transport in the Kuroshio region is distinctly seen as well. The Atlantic Ocean, best ventilated by streams, is most methane laden; the Indian Ocean occupies the second place. The methane concentration grows with depth and becomes maximal close to the most powerful sources. Thus, at a depth of 250 m, the dissolved methane concentration is 5300 nl/l and at a depth of 2000 m it is 12000 nl/l. The vertical average dissolved methane profiles at different time moments are given in Figure 5, where the maximum gas concentrations allow one to



trace depths of the methane venting. Therefore, the maximum methane concentrations are found at depths between 1000 and 2000 m.

#### 4. Conclusion

The results of an investigation into the methane model transport by the ocean streams in the world ocean from the seabed sources due to the submarine gas venting are presented. The model sources are set by increased background values of the dissolved methane or by setting the rates of the water dissolved methane filtration venting. The problem is solved on the test site of the world ocean with allowance for the actual bottom topography and using a horizontal three-graduate grid with 24 vertical levels. The numerical modeling gives the estimate of the methane emission into the atmosphere between 9 and 20 Tg/yr depending on the experiment scenario. The modeling results of the submarine fluid venting from gas seeps, vents, and mud volcanoes are comparable with the available published data estimates [3,4,7–9,21]. Clearly, this flux is significant and should be recognized as a component of the global carbon cycle.

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