Methane Emission from Bottom Sediments

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(Received January 13, 2003; in revised form February 21, 2003)

Abstract

The problem concerning the extraction of the characteristics of methane formation and transport processes from the experimental data is discussed.

INTRODUCTION

Methane is formed in bottom sediments due to the treatment of plant and animal residues with the help of microorganisms. Methane is emitted into the atmosphere by means of molecular diffusion, with the help of bubbles and through plants. The fraction of a channel depends on methane diffusion, solubility, methane generation rate in the bottom layer, plant density. Along with methane, substantial role in transport processes is played by nitrogen, which enters bottom layers from the atmosphere. In the present work, on the basis of the theory of methane emission developed in [1, 2], the goal to extract the characteristics of methane formation and transport using the experimental data is formulated.

Gas emission from sedimentary (active) layers of water reservoirs depends on a number of parameters. The most important among them are methane (gas No. 1) generation rate in the sedimentary layer (W_1) and its dependence on depth ($W_1(z)$). The exponential attenuation of methane generation rate with depth on average is most characteristic of natural conditions:

$$W_1 = W_{10}\exp(-\nu z)$$

Here W_{10} is the rate of methane generation on the surface of the sedimentary layer, ν is the parameter of generation rate attenuation, z is the depth.

Emission rate is also determined by the solubility of methane and nitrogen (gas No. 2) described by Henry's constants K_1 and K_2, respectively, by the mobility of methane and nitrogen molecules determined by diffusion coefficients D_1 and D_2, respectively, by atmospheric pressure P_0 = 1 atm, and nitrogen content of the atmosphere X_2. It is also necessary to take into account the porosity of medium.

In experimental investigations, the dependence of methane concentration on depth $C_1(z)$ is usually measured; much rarer are measurements of the dependence of nitrogen concentration on depth $C_2(z)$, the position of bubble layer h, methane flux into the atmosphere, in which the bubble flux J_{1B} can sometimes be successfully distinguished from the diffusion flux J_{1D}. The model of the active layer is shown in Fig. 1.

EXTRACTION OF THE SYSTEM'S PARAMETERS FROM EXPERIMENTAL DATA

When analyzing experimental data, it is very convenient to use dimensionless parameters. In order to distinguish between dimensionless and the corresponding dimensional

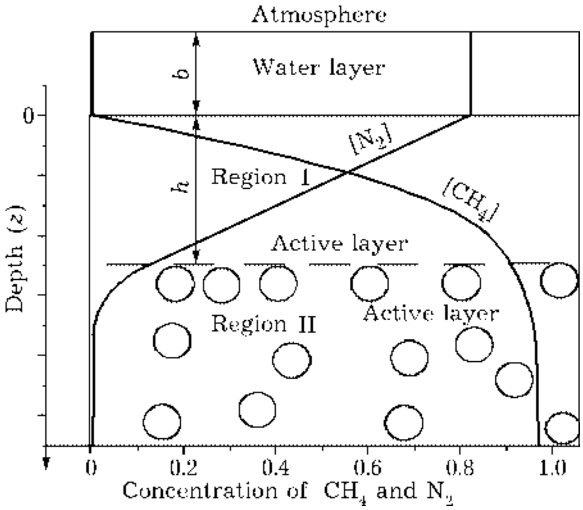

Fig. 1. Model of the active layer.

parameters, let us sign "~" for the former ones. For instance, dimensionless concentration will be written down as $\tilde{C}$. Concentrations of all the gases will be expressed through K_1P_0. This parameter is methane concentration in the sedimentary layer in equilibrium with methane concentration in the gas phase, in which methane pressure is P_0.

The rate of methane formation depending on depth z can be written down as a dimensionless parameter:

$$\tilde{W}_1(z) = W_1(z) / W_{10}$$

where W_{10} is some characteristic rate of methane generation in the system under consideration. For example, if the rate decreases with depth, we may take the rate of generation on the surface of the layer as W_{10}. At the worst, some specific value can be used as W_{10}, for example 10^{-12} mol/(cm^3 s).

Distance can be expressed in the following units:

$$l = \sqrt{K_1D_1P_0 / W_{10}}$$

For calculations, it is convenient to use the unit of 10^{-12} mol/(cm^2 s) for $K_1D_1P_0$ and 10^{-12} mol/(cm^3 s) for W_{10}. So, l is obtained in centimeters.

The distance inside the active layer will be expressed by a dimensionless value $\tilde{z}$:

$$\tilde{z} = z / l$$

Then,

$$h = \tilde{h}l$$

$$\nu = \tilde{\nu} / l$$

Similarly, in order to describe fluxes, we choose the value

$$j = \sqrt{K_1D_1P_0W_{10}}$$

Then, for example,

$$J_{1D} = \tilde{J}_{1D}j$$

When analyzing experimental data, we will assume that Henry's constants, diffusion coefficients and porosity are known. The thickness of water layer above the active layer will be taken equal to zero.

Methane generation rate and its dependence on depth

If methane generation rate is constant, then, methane concentration reaches unity with depth (in dimensionless units), while nitrogen concentration reaches zero (Fig. 2, *a*). If the generation rate attenuates with depth, methane and nitrogen concentrations will reach steady values below unity and above zero, re-

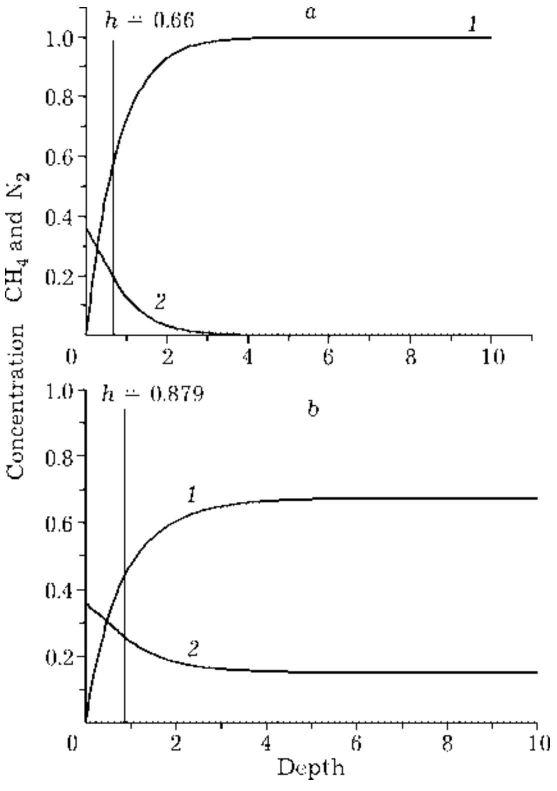

Fig. 2. Dependence of methane (*1*) and nitrogen (*2*) concentrations on depth: *a* − $\tilde{\nu} = 0$; *b* − $\tilde{\nu} = 1$. The h value indicates the position of bubble layer.

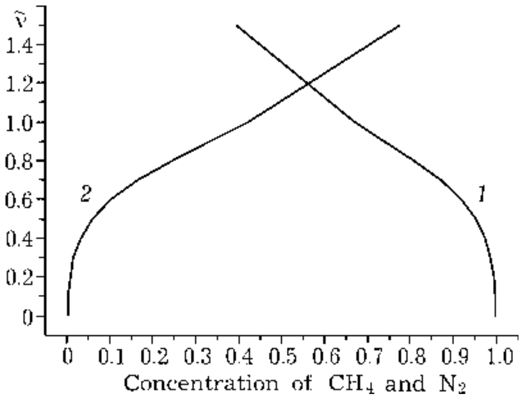

Fig. 3. Estimation of $\tilde{\nu}$ value from the steady concentrations of methane (*1*) and nitrogen (*2*).

spectively (see Fig. 2, *b*). Assuming exponential decay, one may determine the parameter of exponential attenuation by analyzing steady concentrations of methane and nitrogen. The steady concentration of methane is used for this purpose, and the ratio of the steady concentration of nitrogen to the nitrogen concentration on the surface of active layer is calculated. In principle, it is sufficient to know the steady concentration of one of the gases; however, information about two gases allows us to perform additional control of the calculated parameter $\tilde{\nu}$. This parameter can be determined using the plot shown in Fig. 3, or by measuring the molar fraction of methane X_1 in bubbles evolved into the atmosphere, because a linear connection exists between $\tilde{\nu}$ and X_1 (Fig. 4):

$$\tilde{\nu} = (1.03 - X_{1B}) / 0.453$$

In order to determine ν one should know the length l. To determine l, we may use the

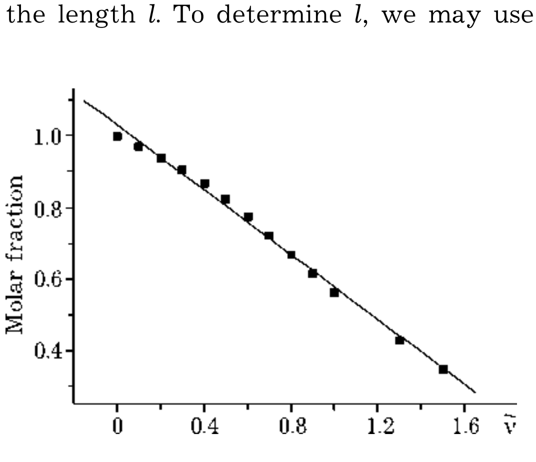

Fig. 4. Dependence of methane molar fraction in the bubble flux on $\tilde{\nu}$.

depth position of a point in which methane concentration reaches 50 % of the steady level $z_{0.5}$, or the depth of occurrence of the bubble layer h. Calculations show [2] that $\tilde{z}_{0.5}$ is equal to 0.6 with a satisfactory accuracy. Therefore, in dimensional unit,

$$z_{0.5} \approx 0.6l = 0.6\sqrt{K_1 D_1 P_0 / W_{10}}$$

Similarly, the depth of occurrence of the bubble layer is

$$h \approx 0.8l = 0.8\sqrt{K_1 D_1 P_0 / W_{10}}$$

though the accuracy of l determination is lower in case when h is measured. Knowing l and $\tilde{\nu}$ one can calculate the parameter of exponential attenuation of methane generation rate using the equation $\nu = \tilde{\nu} / l$.

Now, knowing the value of l, we may calculate methane generation rate W_{10}:

$$W_{10} = K_1 D_1 P_0 / l^2$$

Methane fluxes

Methane flux in the absence of vegetation consists of two components: diffusion flux and bubble flux. They can be distinguished from each other experimentally only in rare cases [3]. In the major part of experiments, a sum of both components is measured.

At first, let us calculate the flux using equation

$$j = lW_{10}$$

As it follows from the results obtained in [2], the diffusion flux of methane is

$$J_{1D} \approx j$$

The bubble flux can be estimated [2] with the help of the diffusion flow of nitrogen and molar fraction of methane in bubbles as

$$\tilde{J}_{1B} = \tilde{J}_{2D} X_{1B} / (1 - X_{1B})$$

The diffusion flux of nitrogen is linearly connected with X_{1B} by the equation:

$$\tilde{J}_{2D} = 0.32X_{1B} - 0.06$$

and therefore the bubble flux of methane can be calculated using the equation

$$J_{1B} = (0.32X_{1B} - 0.06)X_{1B} / (1 - X_{1B})j$$

Measurement of the fluxes allows one to bring this scheme to an additional test.

CONCLUSION

The proposed scheme allows calculating the parameters of methane generation rate and methane fluxes into the atmosphere on the basis of the data on the dependence of methane concentration on depth and on the composition of bubbles in the sedimentary layers.

Acknowledgement

The work has been performed according to the plan of Integration Program of the SB RAS and is supported by RFBR (Grant No. 01–05–64792).

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