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Numerical simulation of modern methane emissions and their influence on climate variability and gas composition of the **Arctic atmosphere**

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Abstract. The impact of Arctic methane hydrate emissions on the gas composition and climate is studied by using a chemistry-climate model. Model runs are carried out using two methane emission scenarios (with Arctic methane emissions increased by 5 and 10 times, respectively, relative to the reference level of emissions of 10 Tg/year). The methane hydrate emissions have a strong impact on the concentrations of methane and other gases. Temperature distributions in the polar and nonpolar regions in the troposphere and stratosphere are obtained. The influence on the climate variability and gas composition of the atmosphere is estimated.

1. Introduction

Now much attention has been given to the impact of non-carbon dioxide greenhouse gases on climate change. Atmospheric methane, being one of the most important greenhouse gases, on the other hand, plays an important role in the chemical balance of the lower and middle atmosphere. Methane determines tropospheric ozone formation and affects hydroxyl radical content, which is the main oxidizer of the troposphere [1-5]. In the stratosphere, methane is a major source of water vapor and, therefore, affects the catalytic ozone destruction [1-2].

An important feature of CH_4 is that it is not formed in the atmosphere and the only source of atmospheric methane is its surface emissions [6]. At the same time, methane surface emissions are determined by both natural processes and anthropogenic factors. Considering the climatic effects of methane, it is necessary to take into account that the intensity of natural biogenic methane emissions greatly depends on climate and promptly responds to its changes, which leads to the formation of feedbacks between climate changes and the methane content in the atmosphere [7]. In particular, in the Arctic zone, where accumulated a huge amount of methane in the permafrost, the continental shelf and the gas hydrates and where climate change is faster than in the other regions, the melting of permafrost and the destruction of gas hydrates due to climate change can lead to release of large amounts of methane. This can lead to further strengthening of climate change which, in turn, can affect the increase in methane emissions from melting permafrost and the destruction of gas hydrates [8].

Gas hydrates are unstable with increasing temperature and lowering pressure, which can lead to the release of a significant part of the methane accumulated in them during climate warming [9]. As a

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result, the amount of methane emitted from gas hydrates may become comparable to emissions from other globally significant sources. In the future, with increasing temperature, the role of Arctic gas hydrates as a source of methane on a global scale will only increase, which necessitates theoretical and experimental studies to assess the power of methane emissions from gas hydrates and the consequences of their impact on climate change and atmospheric composition. At the same time, due to the long lifetime in the atmosphere (on average, about 10 years), methane variability affects the radiative and chemical properties of the atmosphere not only in the source zone, but also in other regions and altitudes, up to the impact on global effects.

Volodin E.M. [10] used a general circulation model developed at the Institute of Numerical Mathematics of the Russian Academy of Sciences (INM RAS) without chemistry included to study the influence of an additional source of Arctic methane on the interhemispheric asymmetry of the CH_4 concentration and climate [10]. The results of numerical experiments with additional methane emissions of 50 or 100 Tg/year in the northern high latitudes demonstrate that the differences in the methane concentrations between the Arctic and the Southern Hemisphere are 14 and 17%, respectively, a value that is higher than the observed 10% [10]. The spatial distribution of the warming generated by an additional methane emission is determined only by the average amount of methane contained in the atmosphere and has almost no dependence on where the source of methane lies. An additional methane emission of 4000 Tg/year causes the global mean surface temperature to rise by about 1.5° [10]. This work is an extension of [10] with the chemistry-climate interaction added.

In addition to the chemical and climatic feedbacks associated with methane emissions increase into the atmosphere in the Arctic zone as a result of climate change, there are also purely chemical feedbacks formed due to the fact that the intensive melting of methane hydrates and the growth of their content in these zones can greatly reduce hydroxyl radicals, because methane is one of its main destroyers. As a result, the methane content may increase even more, since the main destruction of atmospheric methane occurs as a result of chemical interaction with OH [1,2]. On the other hand, the effect of reduction of hydroxyl radicals on methane itself can have an impact on other gases, since the self-cleaning ability of the atmosphere is mainly determined by the content of hydroxyl radicals, OH.

Numerical simulations using chemistry-climate models (CCMs) that take into account the interaction of physical and chemical processes in the troposphere and stratosphere are best suited to assess the impact of potential increases in methane emissions on changes in the temperature regime and gas composition of the atmosphere. In this paper, a chemistry-climate model of the lower and middle atmosphere is used to estimate the effect of possible enhanced methane emissions from Arctic gas hydrates on changes in ozone and other atmospheric gases and the temperature of the troposphere and stratosphere.

2. Model description and numerical experiment structure

This study was carried out using a numerical chemistry-climate model of INM RAS and Russian State Hydrometeorological University (RSHU) [11]. The INM RAS – RSHU CCM covers the entire globe with a horizontal resolution of 4 × 5 degrees of latitude-longitude and from the earth's surface up to the mesopause. The model chemical scheme takes into account the variability of 74 minor gases, among which there are both organic and inorganic compounds, long-lived or short-lived. The model takes into account advection processes, chemical transformations, turbulence, dry and wet deposition, and phase transitions. The chemical scheme includes 174 chemical reactions and 51 photodissociation processes. Surface emissions of modeled gases are based on EDGAR, GISS NASA, GEIA, and MEGAN databases. In particular, the biogenic (termites, wetlands, decomposition of organic matter in oceans, forest fires) and anthropogenic (industrial emissions, transport, oil and gas production, energy, agricultural activity and its waste, biomass burning) methane surface emissions are used to set the model lower boundary condition.

To estimate the impact of possible increased methane emissions from Arctic gas hydrates on the gas composition and structure of the atmosphere, the INM RAS – RSHU CCM was run for 15 years starting in 2015. At the same time, methane emissions from Arctic gas hydrates were increased by 5

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and 10 times compared to a background level of 10 Tg/year. The results of the calculations averaged over the period from 11 to 15 years of the model calculations were compared with the calculations based on the baseline scenario in which methane emissions from gas hydrates remained at the present level.

3. The results

Figure 1 demonstrates the percentage change in the methane concentration due to increased Arctic methane emissions by 5 and 10 times. Tropospheric increases in methane are consistent with enhanced Arctic emissions. At the same time, the largest increase, as expected, is revealed in the Arctic, i.e. near the zone of enhanced emissions. In the upper stratosphere, there is also an increase in the methane content, in order of magnitude corresponding to the tropospheric increase as much as below and above. Moreover, in the polar regions of the middle stratosphere there is the smallest increase in the methane content. The latter effect points to complex nonlinear effects of methane emissions that are observed across the globe, and the most unusual results are recorded in the opposite part of Earth's atmosphere.



Figure 1. Modeled percent change in methane concentration in response to enhanced methane emission from Arctic gas hydrates by (a) 5 times, (b) 10 times.

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Figure 2. Modeled percent change of hydroxil radical OH concentration in response to enhanced methane emission from Arctic gas hydrates by (a) 5 times, (b) 10 times.

Hydroxyl radical OH is the main chemical methane loss in the atmosphere and, on the other hand, OH is largely destroyed by methane. As shown in Fig.2, with a rise in Arctic methane emissions as a result of possible melting of gas hydrates due to climate changes, the tropospheric hydroxyl radical OH significantly reduced in the troposphere, which corresponds to the known fact of destruction of OH molecules by methane molecules. At the same time, in the stratosphere, on the contrary, there is an increase in the hydroxyl content, which is probably a consequence of an increase in the stratospheric water vapor, which is a source of hydroxyl radicals. In the troposphere, the influence of methane on the water vapor content is insignificant due to the dominant role of other sources of water vapor.

In the polar regions during the polar night, when there is no formation of hydroxyl radicals under the influence of solar radiation, the influence of methane on the OH content is very different from other latitudes. At the same time, the tropospheric effects of reducing hydroxyl radicals are amplified during the polar night, as the destruction of methane is preserved, and in the stratosphere the effects differ in the Northern and Southern Hemispheres. In the Southern Hemisphere the content of hydroxyl radicals is mainly reduced during the polar night both in the troposphere and stratosphere, which can be explained by the destruction of hydroxyl methane and in the absence of solar radiation, and lack of water vapor production from methane, whereas in the Northern Hemisphere in the stratosphere an increase in methane is often observed, indicating the influence of complex chemical-dynamic feedbacks. It should be noted, however, that the concentration of hydroxyl radicals during the polar night is negligible and OH has little effect on the atmospheric chemistry in the absence of the sun.

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Figure 3. Modeled percent change in ozone concentration in response to enhanced methane emission from Arctic gas hydrates by (a) 5 times, (b)10 times.

Methane contributes to the production of tropospheric ozone. Thus, its increase as a result of the melting of Arctic hydrates should contribute to an increase in the ozone content in the troposphere. The results of the model experiments presented in Fig.3 demonstrate that this effect is available, and it becomes most noticeable with a tenfold increase in the methane emissions from Arctic gas hydrates. However, it is noticeable that the effect of methane increase on the tropospheric ozone is not linear. It is expressed, on the one hand, in the absence of geographical consistence between the regions of methane and ozone increase (compare to Fig.1), and, on the other hand, in the absence of a significant change in the ozone reaction between scenarios with fifth and tenth increase in the methane emissions. This may be due to the influence of other gases, in particular, nitrogen oxides, on the ozone production due to methane fluxes from Arctic gas hydrates. Therefore, the effect of methane on the tropospheric ozone is less than would be expected on the basis of a significant increase in the Arctic methane emissions.

In the stratosphere, the impact of Arctic methane emissions on ozone is negligible, with the exception of the lower Antarctic stratosphere. In most of the stratosphere, methane has little effect on ozone, despite increases in the water vapor and hydroxyl radicals (Fig.2). This effect is a consequence of the determining role of nitrogen catalytic cycles of the stratospheric ozone depletion, and the increase in hydroxyl radicals leads more to the binding of nitrogen radicals than to an increase in ozone depletion in hydrogen catalytic cycles. In the lower Antarctic stratosphere, where the formation of Antarctic "ozone holes" is developing, the increase in methane content is likely to contribute to even greater denitrification of the polar stratosphere by binding nitrogen gases, which increases the intensity of ozone depletion in chlorine and bromine cycles. Accordingly, unlike other regions of the stratosphere, the ozone content in Antarctica is significantly reduced with enhanced methane emissions in the Arctic.

Figure 4 shows the temperature percentage change due to an increase in the Arctic methane emissions by 5 and 10 times. There are some areas where there is temperature decrease or increase. In Antarctic spring and summer in the lower stratosphere (between altitudes of 15 km and 45 km and between altitudes of 11 km and 21 km) Arctic methane emissions increase by 5 times is observed. The

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other scenario when the Arctic emission increase is by 10 times shows more intense and wide temperature decrease. A temperature decrease by 2 % is observed from \sim 10 km to 27 km in winter and from 12 to 44 km in spring. Besides, it is important to note that in spring and summer a temperature increase is observed. It is situated about 6-7 km higher than the temperature decrease.

In the Arctic stratosphere, the temperature is increasing from September to November. In the troposphere, the temperature is stable (no decrease and no increase). Thus, when setting scenarios for increasing methane hydrate emissions, an increase in temperature is observed in relation to the current state (at the current level of emissions). It should also be noted that warming in the stratosphere in winter with an increase in methane hydrate emissions is 10 times more intense than the increase in methane hydrate emissions by 5 times - both in terms of the temperature change and spatial resolution - the focus of a positive temperature increase; the summer period is characterized by a decrease in the size and intensity of the tropospheric focus of warming.

In the Antarctic, the temperature decrease is more intense when the Arctic methane emissions increase by 10 times than by 5 times. The Arctic decreasing temperature is observed from September to February with the Arctic methane emissions increase by 10 times. If the Arctic methane emissions increase by 5 times, the decrease is observed from September to November.



Figure 4. Modeled percent change of temperature in response to enhanced methane emission from Arctic gas hydrates by (a)5 times, (b) 10 times.

4. Conclusions

Numerical experiments with a chemistry-climatic model of the lower and middle atmosphere were carried out to assess the effect of a probable increase in methane emissions from the melting of Arctic gas hydrates due to climate warming on the gas composition and temperature of the atmosphere.

The results of the numerical experiments have shown that the increase in methane emissions affects the composition and structure of the atmosphere not only in the emission zone, i.e. in the Arctic, but also in some other regions of the Globe. The effects in the troposphere and stratosphere are different, IOP Conf. Series: Earth and Environmental Science **386** (2019) 012020 doi:10.1088/1755-1315/386/1/012020

and the methane emissions may have a great impact on the atmospheric gas composition and temperature, especially in the polar regions. Moreover, in Antarctica, i.e. in the opposite region of the Globe, changes in both the gas composition and the temperature of the atmosphere, which are most significant and different from those in the other regions, are recoded. The results of the model experiments demonstrate the nonlinearity of the effect of methane emissions from the Arctic gas hydrates on the gas composition and temperature regime of the atmosphere.

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