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Anthropogenic impact on arctic near-surface methane: observations and model simulations

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Abstract. Impact of climatically significant anthropogenic emissions to seasonal methane (CH_4) variations observed at arctic and subarctic background stations in 1999 – 2019 has been quantitatively estimated using GEOS-Chem chemical transport model. It is shown that the formation of a stable continental pollution plume from sources in Western Europe, European Russia and Siberia allows to explain up to 5.5–8.6 % of observed CH_4 surface concentration (~104–165 ppb). These atmospheric response values are several times higher than the of the observed annual methane variability amplitude (22–36 ppb), which allows to conclude that regional anthropogenic methane emissions sources play a significant role in regional CH_4 balance in arctic and subarctic areas.

1. Introduction

According to the fourth synthesis report published in 2007 by the Intergovernmental Panel on Climate Change (IPCCAR4), methane (CH_4) is the second most important (after carbon dioxide, CO_2) greenhouse gas in the atmosphere [1].

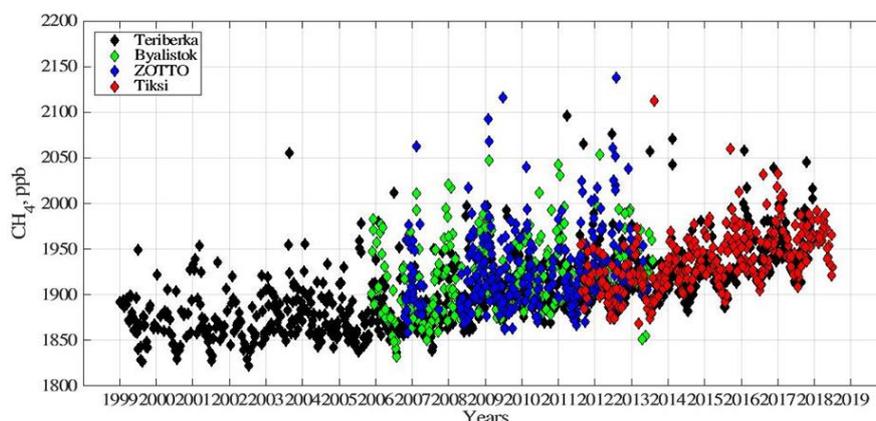


Figure 1. Surface methane concentration according to flask measurements at Teriberka, Bialystok, ZOTTO and Tiksi stations.



Insufficient data on methane content in surface air makes it difficult to quantify methane emissions from major regional anthropogenic and biogenic sources in the Northern Eurasia. This gap is partially filled by long-term observations at stations Teriberka (69.1° N, 35.1° E, 15 m above sea level (m.a.s.l.)), Bialystok (53.1° N, 23.1° E, 160 m.a.s.l.), Zotino (60.8° N, 89.4° E, 300 m.a.s.l.) and Tiksi (71.4° N, 128.5° E, 10 m.a.s.l.). The preliminary analysis of these observations data is given below. Since 2005, according to data at all stations, there has been a significant increase in surface methane concentration (see figure 1). The reasons for this increase are not completely clear. The paper provides quantitative estimates of the most important regional anthropogenic methane emissions sources contribution to observed long-term variability of methane concentration in surface air. Reliable and detailed quantitative estimates of atmospheric methane sources and sinks are needed both to interpret the observed variability and to predict future changes. Background stations observational data, as well as numerical calculations performed by chemical transport models (CTM) is used to analyze surface methane variations. In this paper we use GEOS-Chem model, which is widely applied in chemically active and greenhouse gases fields calculation.

2. Background stations and GEOS-Chem model

GEOS-Chem (<http://acmg.seas.harvard.edu/geos/>, used version 12-01) - numerical Eulerian global chemical-transport model of atmospheric composition, taking into account all major natural and anthropogenic sources and sinks of chemically active gases and aerosols [2]. Meteorological fields and surface data with a time resolution of 3 (two-dimensional fields) and 6 (three-dimensional fields) hours are taken from the GEOS-GMAO global data assimilation system (Goddard Earth Observing System - NASA Global Modeling Assimilation Office, <http://gmao.gsfc.nasa.gov/GEOS/>) and are reprojected to model grid. In this paper EDGAR 4.3.2 [3] data were used for anthropogenic CH₄ emissions in the model, WetCHARTs v1.0 (global wetland CH₄ emission model ensemble for use in atmospheric chemical-transport models) [4] data - for biogenic emissions and GFED4 – from CH₄ emissions from wildfires [5]. We used 4°×5° model grid, meteorology MERRA2, for comparison with observations were used first level output with a height ~58 m above the ground. Calculations of chemical evolution were carried out in standard mode "NO_x-O_x-hydrocarbon-aerosol" (the so-called "full chemistry" mode for the troposphere, 253 tracers, >500 reactions, time step 60 min.)

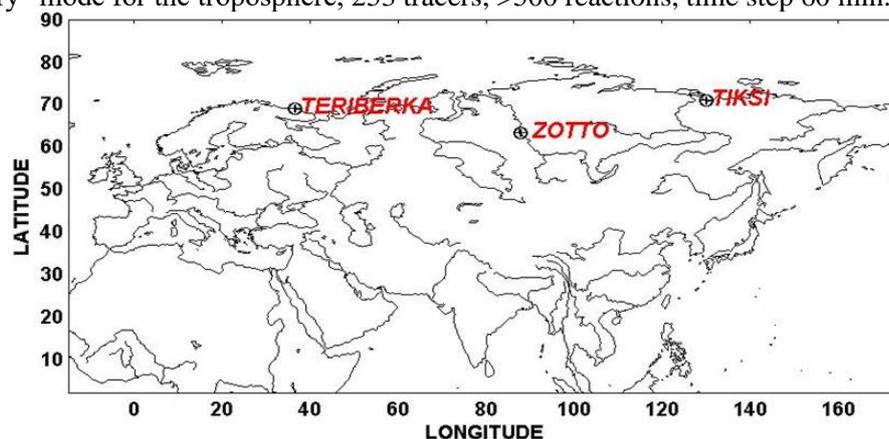


Figure 2. Atmospheric monitoring background stations used for comparison with GEOS-Chem model.

Generally, the results of calculations for all stations (see figure 2), based on monthly averaged data, was in good agreement with the observations despite the limited spatial resolution of the model. The mean bias $\delta = \bar{O} - \bar{C}$, standard error $RMSE = \left(\overline{(O - C)^2} \right)^{1/2}$, and correlation coefficient $COR = \overline{(O - \bar{O})(C - \bar{C})} / (\sigma_o \sigma_c)$ were used as consistency criteria. O and C are observed and calculated by

the model monthly mean concentrations of O_3 , σ_o , σ_c - corresponding standard deviations, and the upper line means averaging over all values.

For station Teriberka it was obtained $COR = 0.75$ (0.65 .. 0.82), $RMSE = 19.1$ (16.9 .. 22.7) ppb and $\delta = 2.2$ (-1.0 .. 5.6) ppb. The values indicated in parentheses correspond to the limits of the 95% confidence interval calculated by bootstrap method [6]. It should be noted that these values for the station was calculated on basis of flask measurements, carried out 2-10 times a month.

For station Tiksi it was obtained $COR = 0.61$ (0.44 .. 0.74), $RMSE = 22.1$ (18.8 .. 26.3) ppb and $\delta = 5.1$ (0.5 .. 10.0) ppb.

For stations ZOTTO it was obtained $COR = 0.70$ (0.39 .. 0.84), $RMSE = 23.9$ (18.8 .. 32.1) ppb and $\delta = -9.6$ (-16.1 .. -1.8) ppb.

Generally, calculation results for all three stations, based on monthly averages, was in good agreement with observational data, taking into account limited model spatial resolution. The best agreement is found for Teriberka station, the worst - for Tiksi station. Systematic deviation of calculated methane concentrations from observed in individual seasons was caused by inability of correctly reproducing the high-frequency (synoptic) CH_4 field variations, which make a significant contribution to overall CH_4 variability. Another reason might be the insufficiency of used emission data, especially in Russia. Taking into account the important role of wildfires in middle and high latitudes of Northern Eurasia as a source of large amounts of chemically active gases, including methane, uncertainties in calculation of combustion products emissions might play a major role in observed discrepancies between model calculations and observations. In addition, a significant contribution of total methane variation synoptic component to total measured signal is possible.

3. Influence of regional anthropogenic CH_4 emissions on methane concentration in arctic and subarctic stations

3.1. Model simulations scenario

Quantitative estimates of anthropogenic emissions contribution to CH_4 concentration field were made on the basis of a regional approach: there was identified one macroregion, including Western Europe, European territory of Russia (ETR), Siberia and Russian Far East (see figure 3).

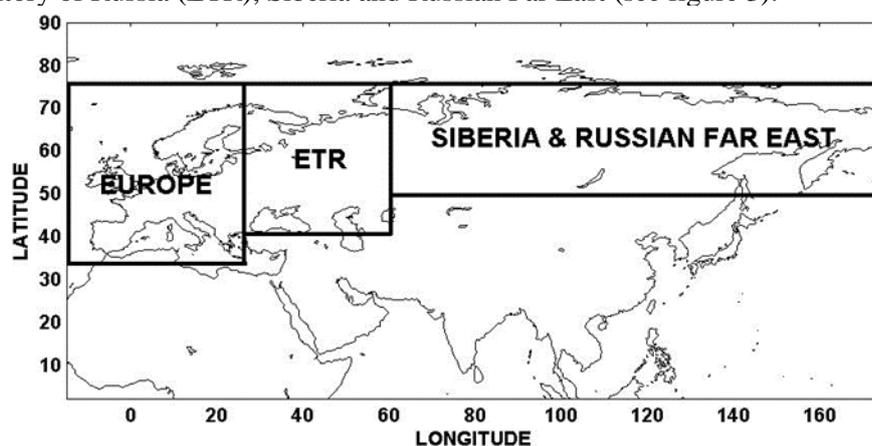


Figure 3. Geographical areas selected for the calculations: Europe (35-75N, 15-27E), European territory of Russia – ETR (41-75N, 27-60E) and Siberia & Russian Far East (49-75N, 60-180E).

The atmospheric response (AR_S) values were calculated as the difference between concentration obtained from baseline model calculation with full emissions, $\chi(CH_4)_0$, and concentration calculated by the model with reduced emissions according to used scenario, $\chi(CH_4)_S$:

$$AR_S = \chi(CH_4)_0 - \chi(CH_4)_S$$

Thus, average seasonal response fields for 2007-2018 were calculated at first model level (~ 58 m from the surface). GEOS-Chem calculations were performed for the period from July 1, 2006 from the model initial startup file with global concentration fields that played the role of initial conditions to December 31, 2018, while the first 6 months were used to accelerate the model (adaptation of atmospheric concentration fields to reduced emission fields) and were excluded from further analysis.

3.2. Calculation results

The average AR values for three stations considered earlier are shown in table 1 and summary diagram based on it in figure 4. Generally, the effect of long-range transport from regions of climatically significant CH₄ emissions in Northern Eurasia on surface methane and AR values is most significant in winter due to several factors: a seasonal increase in emissions from fossil fuels burning, an increase in methane photochemical lifetime due to a decrease in hydroxyl concentration, as well as an increase in of atmospheric tracers residence time in lower troposphere due to a decrease in role of convective transport and a higher static stability of the troposphere as a whole [7]. According to calculations results, AR value in winter period can reach 7-10% of observed methane concentration.

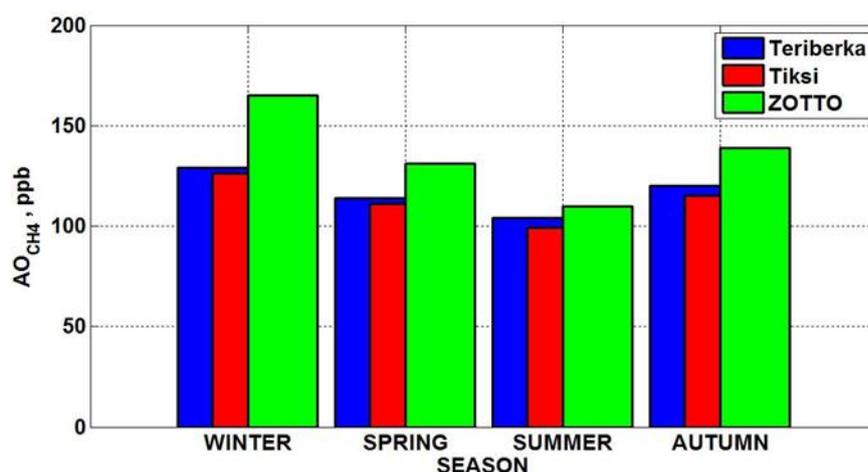


Figure 4. Summary diagram of AR_s for Teriberka, Tiksi and ZOTTO. The average values for all seasons of 2007 - 2018 are given.

Table 1. Seasonally averaged surface methane concentration values for Teriberka, Tiksi and ZOTTO observation stations (ppb). Seasonally averaged atmospheric response values to anthropogenic methane emissions for designated stations (ppb).

	Teriberka		Tiksi		ZOTTO	
	Avg.	AR	Avg.	AR	Avg.	AR
Winter	1934	129	1948	126	1948	165
Spring	1914	114	1927	111	1911	131
Summer	1912	104	1912	99	1917	110
Autumn	1922	120	1939	115	1925	139

The highest AR values (up to 165 ppb in winter) on anthropogenic methane emissions in all seasons are reached for ZOTTO. For most of the year, the station area is located in influence zone of atmospheric pollution sources in Western Europe, ETR and southern Siberia. The continental leeward plume associated with these sources forms an area of high concentrations of pollutants, including CH₄, that extends over almost the entire Northern Eurasia. The stations located on Arctic

coast are far from the axis of this plume, so anthropogenic impact on them is even less. The AO values for Teriberka are only slightly (by 3 – 5 ppb) higher than for Tiksi, which is much further to the east.

4. Summary

The impact of climatically significant anthropogenic sources of CH₄ emissions in Northern Eurasia to the observed seasonal methane variability in 1999-2019 on the Kola Peninsula (Teriberka station), in central Siberia (ZOTTO), and in northeast of Eurasia (Tiksi) were quantified, based on GEOS-Chem global chemical transport model calculations. According to results, the contribution of regional emissions to measured methane concentrations is (~104-165 ppbv), or 5.5-8.6 % of average measured at three stations annual value 1926 ppbv. The calculated values of atmospheric response are several times higher than the amplitude of observed annual methane variability (22-36 ppbv), it allows to conclude that regional sources of atmospheric methane emissions in Western Europe, in European territory of Russia and in Siberia play a significant role in regional balance of the surface CH₄ concentration in lower troposphere above the continent. In summer, the impact of regional methane emissions is slightly weaker (105 ppbv) compared to winter (~140 ppb) due to increasing the role of vertical convective exchange in lower troposphere.

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