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Annual Cycle and Long-Term Trend of the Methane Total Column in the Atmosphere over the St. Petersburg Region

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Abstract—The annual cycle and long-term trend of the methane total column in the atmosphere over the Petergof station (St. Petersburg State University) are analyzed on the basis of data obtained from Fourier-transform infrared spectrometry and EMAC-model calculations. The amplitude of the annual cycle of the total column of CH_4 amounts to 2.1 and 1.5% according to experimental and model data, respectively. For the atmospheric column-averaged mole fraction of CH_4, the amplitude of its annual cycle is smaller than that for its total column and amounts to 1.1 and 0.6% according to experimental and model data, respectively. The results of local continuous measurements of surface CH_4 concentrations showed that, in 2013, the atmospheric column-averaged mole fractions of CH_4 and the amplitudes of diurnal variations in its local concentration were characterized by the same dynamics of seasonal variations. An analysis made on the basis of simulation results showed that atmospheric conditions (under which Fourier-transform IR measurements were performed) could increase the amplitude of the annual cycle of the total column of CH_4 2.5 times when compared to the true one. The results of Fourier-transform IR measurements and EMAC-model calculations showed that, during 2009–2012, the atmospheric concentration of CH_4 increased at a rate of ~0.2% per year. If measurement data obtained in 2013 are added, this rate decreases to ~0.13% per year.

Keywords: methane trends, annual cycle, methane, FTIR measurements local measurements, chemical transport model

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INTRODUCTION

Methane ranks behind only water vapor and carbon dioxide as the most prevalent greenhouse gas. Since the beginning of the Industrial Revolution, the concentration of methane has increased by a factor of two [1]. Despite the fact that atmospheric CH_4 has actively been studied since the 1980s, up to now its sources have been known with insufficient accuracy and current variations in its atmospheric concentration demonstrate some features that call for further investigations [2]. In the early 21st century, almost zero methane trends were observed in the atmosphere [3]; however, in 2006, the concentration of CH_4 again started to rapidly increase [4]. For the 2007–2009 period, the concentration of CH_4 in the atmosphere over the Northern Hemisphere increased with a rate of ~1% per year [5] according to data obtained from measurements of its surface concentrations [6] and its total column amounts [4]. The basic reasons for the observed variability of the methane trend are assumed to be as follows: (i) emissions from mire complexes, which depend on temperature and precipitation amount, (ii) increased anthropogenic emissions, and (iii) emissions of CH_4 during permafrost melting and its release into the atmosphere from methane-hydrate reserves in the Arctic Ocean [2, 4, 7]. However, the reasons for this methane increase since 2006 are still not quite clear, and, at present, they are under active dispute in the scientific community [2, 4–7].

EXERIMENTAL AND MODEL DATA

In this work, both the annual cycle and long-term trend of atmospheric methane are studied on the basis of data obtained from observations at the Petergof monitoring station of St. Petersburg State University (SPbU) and from EMAC-model calculations (2009–2012) [9]. Both results of Fourier-transform IR (FTIR) measurements for 2009–2013 (see, for example, [8]) and data obtained in 2013 from the local monitoring of atmospheric methane were also used in this study. A Los Gatos Research GGA–24r-EP gas analyzer was used for continuous measurements of CH_4 concentrations. The error of measurements with

Table 1. Data on geographic location, measurement periods, and seasonal-cycle amplitudes for the Izana, Zugspitze, and Petergof stations

Data	Izana (TCCON), 28°18′ N, 16°29′ W, 2370 m above sea level	Zugspitze (NDACC), 47.42° N, 10.98° E, 2964 m above sea level	Petergof, 59.88° N, 29.82° E, 20 m above sea level		
Period under analysis	2001–2010	2004–2009	2009–2012		
Data series under analysis	FTIR MF	FTIR MF	FTIR MF	EMAC/FTIR MF	EMAC MF
Annual-cycle amplitude, %	0.6	0.94	1.1	0.6	0.6

this instrument amounts to (0.3–1) ppbv, depending on the accumulation time (100–5 s).

ANNUAL VARIATIONS IN THE TOTAL COLUMN AND COLUMN-AVERAGED MOLE FRACTION OF METHANE

The acronyms used in this work for the data arrays under analysis are as follows: (i) TC corresponds to the total column of CH_4; (ii) MF corresponds to the atmospheric column-averaged mole fraction of CH_4, which is obtained by dividing the TC of methane by the total number of molecules in the dry atmosphere; (iii) FTIR corresponds to data obtained from measurements using ground-based Fourier-transform IR measurements; (iv) EMAC corresponds to a complete time series generated by the EMAC model; and (v) EMAC/FTIR corresponds to a data sample from simulation results, which coincides in date and time with FTIR measurements.

Figures 1 and 2 show the mean annual cycles for the TC and MF of CH_4 for 2009–2012 according to experimental and model data and the differences between the mean annual cycles (FTIR – EMAC/FTIR) and (EMAC/FTIR – EMAC).

Total Column of CH_4

Annual variations in the TC of CH_4 according to FTIR experimental data and sampled EMAC/FTIR model data have similar forms: two local minima in March and July and two maxima in January and May. The amplitudes of the annual cycles of the TC of CH_4 for the FTIR and EMAC/FTIR data arrays amount to 2.1 and 1.5%, respectively.

The difference between these annual cycles (FTIR – EMAC/FTIR) shown in Fig. 1 varies from 0.5% (March) to 2.6% (November) and has a pronounced seasonality: it increases during fall (September–November). It is assumed that this is due to the fact that the EMAC model underestimates natural methane sources [10].

Earlier published results of our measurements with the aid of a classical solar infrared spectrometer (SIRS) [11] showed that, in 1991–2007, the mean annual cycle with an amplitude of ~3% had its maximum in December–January and minimum in June–August, which differs from the annual cycle according to the 2009–2012 Fourier-spectrometry data. However, annual cycles for individual years may differ in character and amplitude. For example, according to SIRS measurements in 2006, the maximum was observed in February and two minima were observed in April and September [11], which is similar to seasonal variations according to FTIR TC measurement data (Fig. 1). Anomalously high amplitudes of the TC annual cycle, which exceeded the mean amplitude almost two times, were recorded in 1998 and 2002 [11].

Atmospheric Column-Averaged Mole Fraction of CH_4

It follows from Figs. 1 and 2 that, in going from CH_4 TC values to column-averaged CH_4 mole fraction values, the annual cycle changes.

According to FTIR MF data, the March minimum of the annual cycle becomes more pronounced and the summer minimum almost vanishes; the maximum is observed in November instead of January (as in the case of CH_4 TC). For comparison, Fig. 3 gives the graphs corresponding to the FTIR MF annual variations for 2013 and 2009–2012. Thus, for example, in 2013, the spring minimum was observed during two months (March and April), the CH_4 concentration decreased to the spring minimum in July, and maxima were recorded in May–June and September.

The sampled EMAC/FTIR MF data array preserved the March, August, and May extrema, and the winter maximum also shifted from January to November. The annual-cycle amplitudes for CH_4 MF are smaller than those for CH_4 TC (see Table 1, column 3).

Let us compare our estimates with results published in [12, 13], where the annual CH_4 MF cycle was analyzed for the Zugspitze and Izana stations (Table 1). As the latitude increases by approximately ~30°, the amplitude of seasonal CH_4 MF variations increases by approximately 0.5%. Our model estimates for the amplitude of the annual cycle are characteristic of the Izana subtropical station rather than the Petergof station.

The annual variations expressed in percent of the mean CH_4 MF are given for the Izana and Zugspitze stations in Fig. 2 (right top axis *Y*). The annual cycle

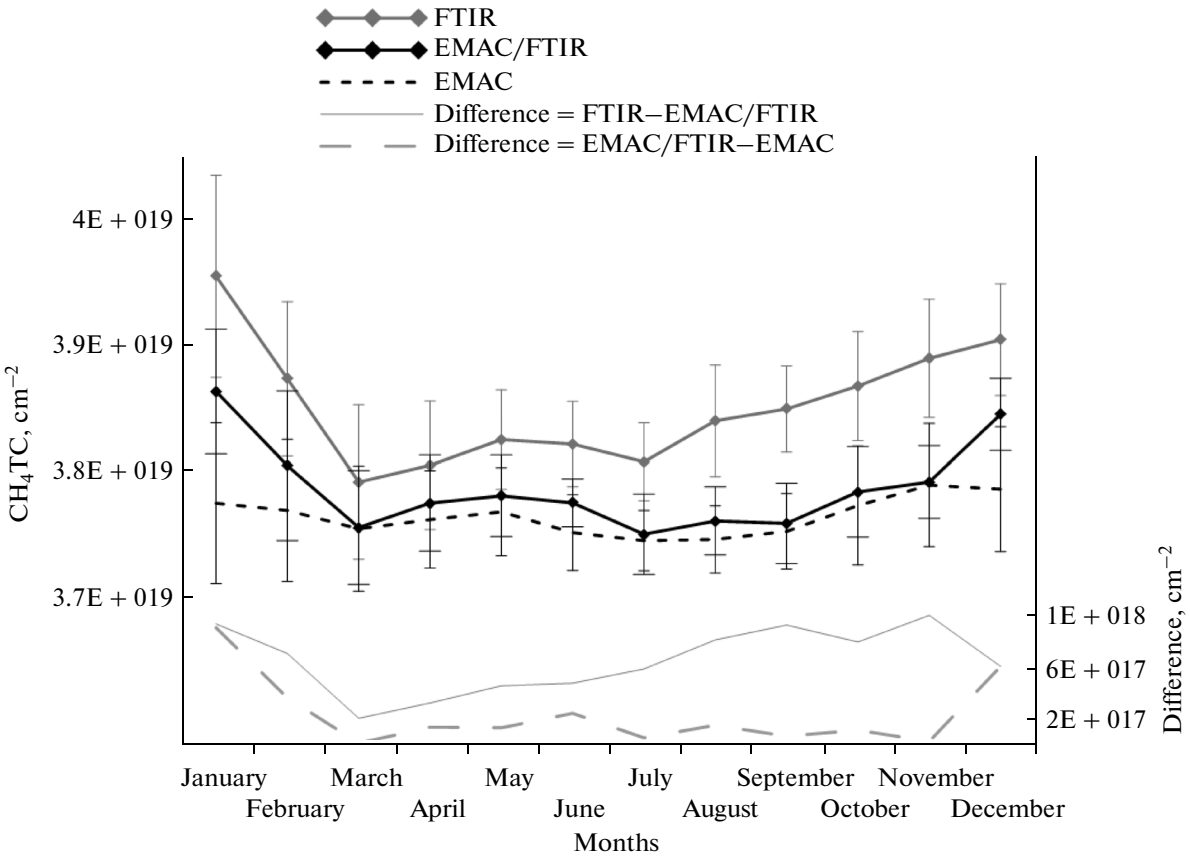

Fig. 1. Mean annual variations in the TC of CH_4 for 2009–2012 according to both Fourier-spectrometry and EMAC model data and the difference between the mean annual cycles (FTIR – EMAC/FTIR) and (EMAC/FTIR – EMAC).

was taken from Fig. 12 in [13] for the Izana subtropical station and from Fig. 11 in [12] for the Zugspitze mid-latitude station.

It follows from Fig. 2 that the annual variations for the Petergof and Izana stations, despite the difference between their amplitudes, are closer in character than those for the Petergof and Zugspitze stations. Note that the March clearly pronounced minimum is observed for all the three stations, and the maximum position is individual for each station: August for Zugspitze, October for Izana, and November for Petergof. Some differences in the annual variations during the fall–winter period may be due to insufficient measurement data obtained at our station during the cold season.

In [13], the form of the FTIR MF seasonal cycle obtained at the Izana station is explained mainly by annual variations in the tropopause height. In fact, our analysis of meteorological-sounding data obtained at the Voeikovo station (59.95° N, 30.70° E, WMO No. 26063) confirmed that the tropopause height is minimum in February–March and maximum in October–November (this coincides with the minimum and maximum of the CH_4 MF annual cycle).

However, in addition to the tropopause height, a number of other factors are responsible for the formation of the CH_4 MF annual cycle [10, 12, 15, 16]: seasonal variations in sources and sinks, stratospheric processes (in addition to the abovementioned tropopause height), and troposphere–stratosphere exchange. In addition, at least the two factors—the tropopause height and the intensity of methane emissions—have close positions of their extrema: winter–early spring and late summer–fall [17].

Despite the fact that the Petergof station is not a high-latitude station, in analyzing the results of both winter and spring CH_4 measurements it is necessary to take into account processes that occur in the polar stratosphere. In winter, the circumpolar vortex may be shifted and, for some time, cover the latitude ~60° N, which was supported by a joint analysis of the results of HCl TC measurements and data on potential vorticity for our station [18]. Moreover, as was noted in [19], for 2 months after the collapse of the circumpolar vortex, regions with vortex-air remains may be preserved in the mid-latitude stratosphere (usually higher than 50° N). Since the winter polar stratosphere is characterized by low CH_4 contents [20], this may lead to stronger variations in winter and spring CH_4 MF values recorded at

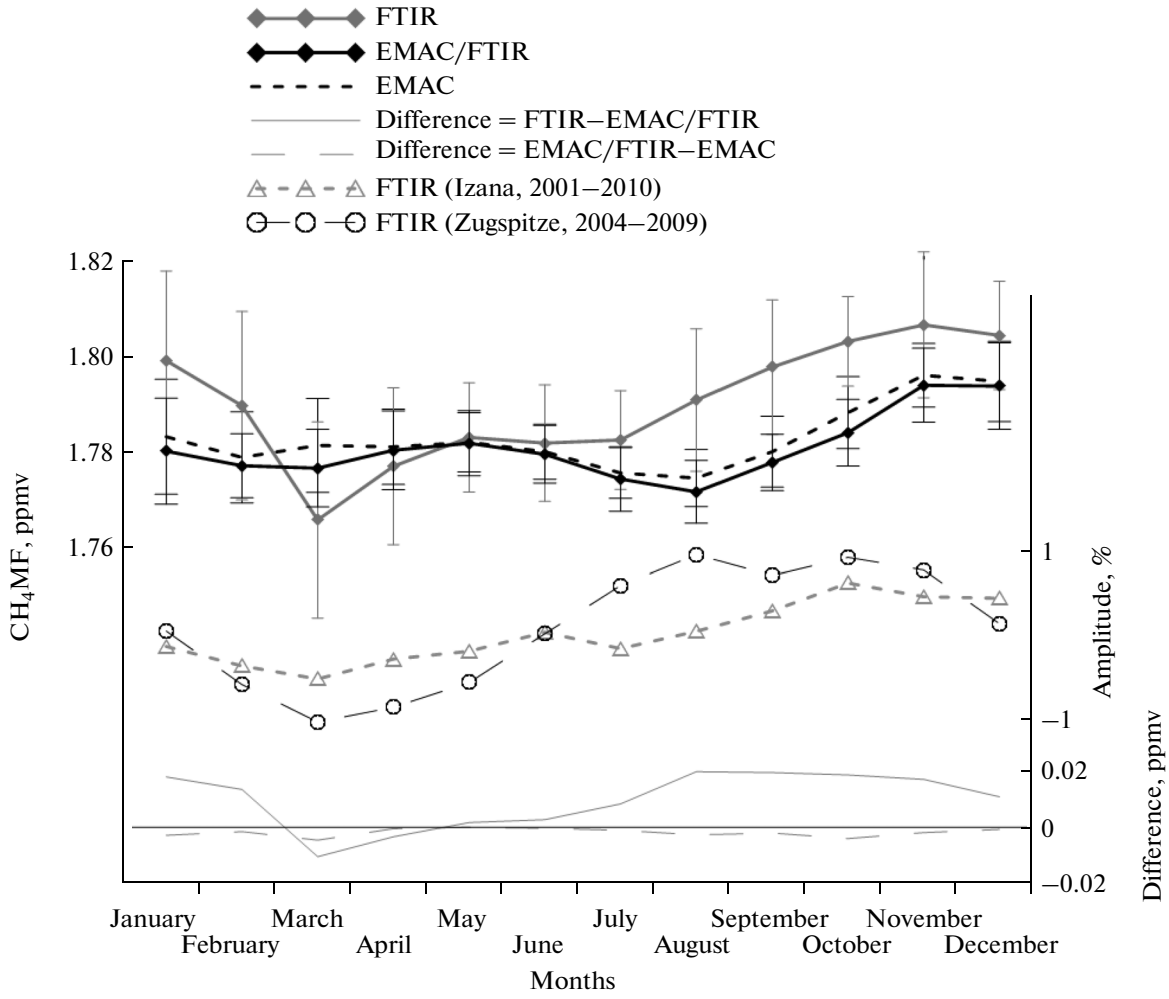

Fig. 2. Mean annual variations in the atmospheric column-averaged mole fraction of CH_4 for 2009–2012 according to both Fourier-spectrometry and EMAC model data and the difference between the mean annual cycles (FTIR – EMAC/FTIR) and (EMAC/FTIR – EMAC). Annual cycles for the Izana and Zugspitze stations.

the Petergof station, when compared, for example, to subtropical stations of the Izana type (~28° N). For concrete quantitative estimates of the influence of the circumpolar vortex on CH_4 TC and MF, it is necessary to analyze the stratospheric portion of the methane total column using additional information [21, 22]; however, this is beyond the scope of this work.

If experimental data on the main sink of methane—the concentration of OH—are not available (it is only known that the maximum concentration of OH in the troposphere is usually observed in June–July), we can try to qualitatively estimate variations in the intensity of methane emissions for our region from month to month on the basis of data obtained from local measurements of the surface concentration of CH_4 (local mole fraction (LMF) of methane), which are available at the Petergof station for January–October 2013.

Figure 3 shows the CH_4 LMF diurnal variations averaged over fortnightly intervals (i.e., there are two diurnal cycles for each month). Data for the first half of July 2013 are not available, because measurements were not performed. It is known that the minimum daytime surface concentrations of CH_4 depend on the activity of mixing processes and the height of the boundary layer rather than the intensity of methane sources [15]. At night, relatively weak vertical motions and the presence of inversions result in the accumulation of methane emitted by its sources in the lower troposphere [23].

Let us compare the 2013 annual variations in the three parameters (Fig. 4): FTIR CH_4 MF, minimum (absolute monthly minima) surface methane concentrations (CH_4 LMF), and the amplitude of the diurnal cycle of CH_4 surface concentrations (half the difference between the extrema of the approximating function—triangles in Fig. 3).

The LMF minimum values can be considered the characteristics of the regional background for the Petergof station. For the available observational data

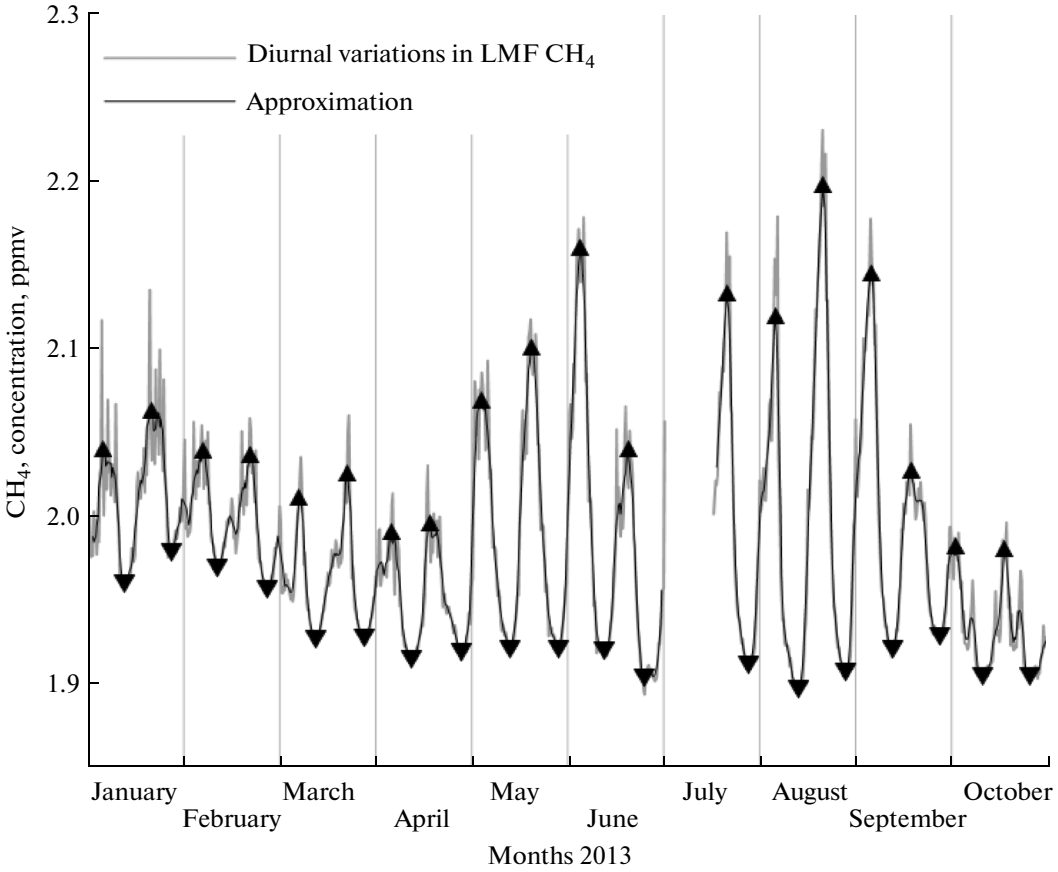

Fig. 3. Annual CH_4 variations: FTIR MF (Petergof, 2013), minimum surface concentrations (Petergof, 2013), surface concentrations (Baltic Sea, 2002–2009), diurnal maximum of surface concentrations (Petergof, 2013), and mean annual cycle for FTIR MF (Petergof, 2009–2012).

obtained during 10 months in 2013, they manifested variations that are characteristic of surface concentrations and close to the mean annual cycle (1992–2009) for the Baltic Sea station (the nearest (to Petergof) NOAA/CMDL station, 55.35° N, 17.22° E, 3 m above sea level) [24]. It follows from Fig. 4 that the seasonal variations in FTIR CH_4 MF and minimum LMF are in antiphase, while the amplitudes of the diurnal cycles show the same dynamics of their variations [10]. For example, the FTIR MF minimum observed in late June–early July 2013 was deeper when compared to the 2009–2012 mean annual cycle. It coincided with both the period of maximum OH and the dip of the LMF diurnal cycle.

Estimating the Influence of Meteorological Conditions during FTIR Measurements on the CH_4 TC and MF Annual Cycles

Let us use the results of simulation in estimating the influence of meteorological conditions on the CH_4 mean (over 2009–2012) annual cycle obtained from FTIR measurements. We will consider that, for the EMAC complete model-data array, we obtain the "true" annual cycle of atmospheric methane and, for the EMAC/FTIR sampled data, we obtain its "measured" annual cycle.

Total column

The results of an analysis showed that the mean amplitude of the EMAC/FTIR TC annual cycle is 2.5 times larger than that of the EMAC TC annual cycle (0.6%). It follows from Fig. 1 (see the lower part of Fig. 1) that the difference between the monthly means (EMAC/FTIR TC – EMAC TC) is positive during the whole year. The most significant differences between the "measured" and "true" annual cycles are observed from December to February; in this case, this difference (EMAC/FTIR TC – EMAC TC) reaches its maximum (~2.3%) in January. If the positions of the minima before and after the sample of Fourier-observation dates were preserved, the maximum shifted from November to January (see Fig. 1).

Atmospheric column-averaged CH_4 mole fraction

The annual cycles obtained from the EMAC MF and EMAC/FTIR MF data arrays (see Fig. 2) coincide in amplitude and character; the only difference is

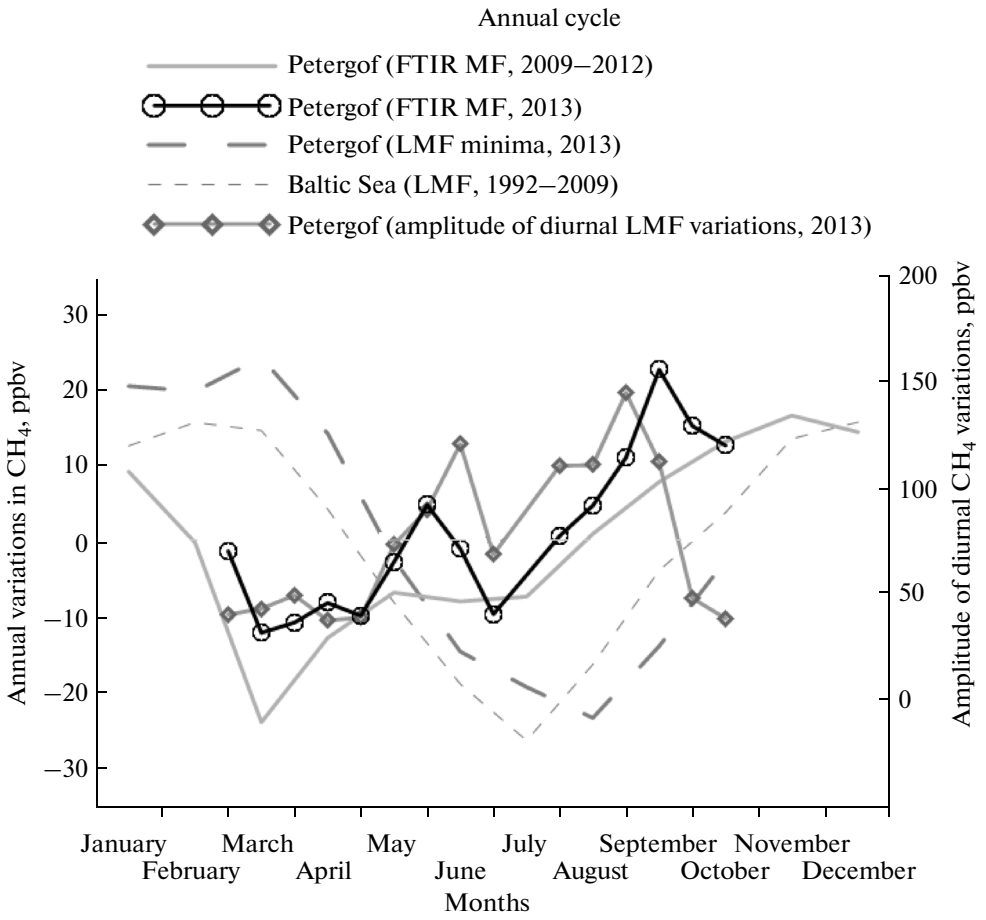

Fig. 4. Diurnal variations in the concentration of methane (Petergof, 2013).

in the absence of the March minimum for EMAC MF. Unlike the total column, the monthly means according to the EMAC/FTIR MF sampled data array are lower than those according to the EMAC MF complete data array. Their difference (EMAC/FTIR MF – EMAC MF) does not have a clearly pronounced seasonality.

The results show that the influence of meteorological conditions is especially pronounced during winter, when daylight periods are short and the number of sunny days with stable atmosphere is small. The differences are explained by the fact that the FTIR measurements of direct solar radiation are mainly performed under anticyclonic stable weather conditions (high pressure and descending and horizontally propagating air flows).

LONG-TERM VARIATIONS IN CH_4 TC

The 2009–2012 long-term methane variations were estimated on the basis of the following six data arrays: FTIR TC, EMAC/FTIR TC, EMAC TC, FTIR MF, EMAC/FTIR MF, and EMAC MF. For the EMAC complete model series, the 2009–2012 complete data array was used. For FTIR and EMAC/FTIR, we used data obtained only from March to July inclusive and in October, because the FTIR measurements were taken during these months in all four years, and, thus, we could eliminate the influence of seasonal CH_4 variations. Note that the different methods of estimating the long-term trend yield almost coincident results [5].

Table 2 gives the linear-trend values and the corresponding confidence intervals (95%). It is seen that both experimental and model estimates of the methane trend for 2009–2012 are in good agreement (~0.2% per year). The estimates of the long-term CH_4 TC variations for all data arrays (except FTIR TC) can be considered significant (according to the Student criterion, a confidence probability of 95%). Note that, unlike the total column (FTIR TC), the linear-trend estimates for atmospheric column-averaged methane mole fractions (FTIR MF) are statistically significant at the same confidence probability. This is due to the stronger variation of FTIR TC values when compared to FTIR MF values.

Table 2. Estimates of long-term (2009–2012) TC and MF variations

Long-term variations	Total column (TC)			Atmospheric column-averaged mole fraction (MF)		
Data array	FTIR	EMAC/FTIR	EMAC	FTIR	EMAC/FTIR	EMAC
Linear trend, % per year	0.18 ± 0.18	0.21 ± 0.12	0.20 ± 0.05	0.18 ± 0.12	0.21 ± 0.05	0.24 ± 0.02

Table 3. Data on the CH_4 MF trends for the Petergof, Garmisch, and Zugspitze stations

Station	Garmisch	Zugspitze	Petergof	
Measurement period	2006–2011		2009–2012	2009–2013
Trend, ppbv per year	5.1 ± 0.9	4.8 ± 1.0	3.2 ± 2.1	2.3 ±1 .4

If the 2013 FTIR measurement results are included in the analysis, the 2009–2013 trend decreases to (0.13 ± 0.11)% per year for TC and (0.13 ± 0.08)% per year for MF. This allows one to assume that the period of a rapid increase in the atmospheric content of methane, which started in 2006 and was recorded by different measuring systems [4–7], is over.

Table 3 gives the CH_4 MF trend data (in ppbv) for the three stations at which FTIR measurements were taken: Petergof, Garmisch (TCCON, 47.48° N, 11.06° E, 743 m above sea level), and Zugspitze [4]. Taking into account the fact that the measurement periods coincide incompletely and there are differences in geographic location, one can consider that the CH_4 MF trends have close values for all three stations.

CONCLUSIONS

The joint analysis of experimental and model data on atmospheric methane for the Petergof station showed the following:

(1) The annual cycle of the TC of CH_4 has two local minima in March and July and one maximum in January. The amplitude of the CH_4 TC annual cycle according to these (experimental and model) data arrays amounts to 2.1 and 1.5%, respectively. The difference between the experimental and model annual cycles manifests a pronounced seasonality, which is probably due to the fact that methane emissions from natural sources were underestimated in the EMAC model.

(2) For the atmospheric column-averaged mole fraction (MF) of CH_4, the maximum of annual cycle is observed in November; in this case, the annual-cycle amplitudes for CH_4 MF are smaller than those for CH_4 TC and amount to 1.1% for experimental data and 0.6% for model data.

(3) The 2013 seasonal variations in the atmospheric column-averaged mole fraction of CH_4 and the diurnal-cycle amplitude for the local surface concentration of CH_4 have the same dynamics of variations.

(4) An analysis on the basis of EMAC data showed that atmospheric conditions under which the FTIR measurements are performed may affect annual-cycle characteristics:

(i) The mean amplitude of the annual cycle of the TC of CH_4 for the EMAC/FTIR TC sampled data array is 2.5 times larger than that for the EMAC TC complete model data array. The difference (EMAC/FTIR TC – EMAC TC) is positive for all months and reaches its maximum (~2.3%) in January. The position of the extrema of the CH_4 TC annual cycle does not change.

(ii) The annual cycle estimated from the EMAC/FTIR MF sampled data series is lower than that estimated from EMAC FM; the annual-cycle amplitudes and character are, on the whole, preserved.

(5) The results of FTIR measurements and EMAC calculations show that the rate of increase in the atmospheric methane is about 0.2% per year (2009–2012). If the 2013 measurement data are included, this trend decreases to ~0.13% per year.

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