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Time Variability of the Total Methane Content in the Atmosphere over the Vicinity of St. Petersburg

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Abstract—The results of measuring the methane content in the entire atmospheric thickness over the St. Petersburg region are given for 1991–2007. It is shown that, within this period, the mean annual cycle of the total methane content is characterized by its maximum values in December–January and its minimum values in June–August when the annual-cycle amplitude amounts to ~3.6%. In this case, the annual variations in the total methane content may differ significantly from the mean annual cycle obtained in some years. A statistically significant linear trend of the total CH_4 content has not been revealed for 1991–2007. The obtained values of the linear-trend index have opposite signs in the winter and summer months (positive for January 0.6 ± 0.2%/year and February 0.4 ± 0.2%/year and negative for July 0.3 ± 0.2%/year and August 0.2 ± 0.1%/year). This fact suggests the tendency for an increase in the amplitude of the annual cycle of the total CH_4 content. The results of a spectral analysis of a series of data on the total CH_4 content show that, for 1991–2007, the following harmonics are pronounced with a confidence of 95%: 12 months (annual harmonic), 32 months (quasi-biennial oscillations), and 55 months (4.5 years), which are also pronounced in the series of meteorological parameters and total ozone content.

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1. INTRODUCTION

The most important greenhouse gases of the Earth's atmosphere include water vapor, carbon dioxide, methane, and nitric oxide. Compared to the preindustrial period, the atmospheric contents of CO_2 and CH_4 have increased approximately two times. Currently, the relative contribution of methane to radiation forcing amounts to about 20% [1]. Methane plays a noticeable role in the tropospheric and stratospheric chemistries: in the troposphere, its reactions with hydroxyl and ozone are important; in the stratosphere, methane serves as a source of water vapor and hydrogen and as a sink of chlorine [2].

The local ground-based measurements of the concentration of methane are performed at the NOAA/GMD (United States) and AGAGE (the international network supported by NASA) stations [1]. At the stations of the Network for Detection of Atmospheric Composition Change (NDACC), the regular spectroscopic measurements of the total content of methane (and, in some cases, the elements of the vertical profiles of CH_4) are performed on the basis of the high-resolution IR spectra of direct solar radiation [3]. In Russia, the spectroscopic method of measurements has been employed at the Institute of Atmospheric Physics, Russian Academy of Sciences (IAP RAS), since 1974 [4, 5] using spectrometers that have an average spectral resolution. Currently, regular ground-based spectroscopic measurements of the total methane content are taken at three stations in Russia: at the Research Institute of Physics (RIP) of the St. Petersburg State University (in the vicinity of St. Petersburg), at the IAP Zvenigorod Scientific Station (ZSS), and at the Institute of Experimental Meteorology (IEM, Obninsk) [6, 7].

The spectroscopic measurements of the total CH_4 content have been taken in the vicinity of St. Petersburg (the Peterhof station located 25 km from the center of the city; 59.9° N, 29.9° E, 20 m above sea level) since 1991 [8–10]. The instruments used and the methods of interpreting the measurement results are described in [8, 9]. In this paper, the results obtained from measurements of the total methane content over 15 years (from January 1991 to 2007) are analyzed.

2. ANALYSIS OF TIME VARIATIONS IN THE TOTAL METHANE CONTENT

2.1. General Characteristic of Time Variations in the Total Methane Content

The results of measurements of the total methane content given in Fig. 1 contain about 1100 daily means of the total content of CH_4. The number of day-

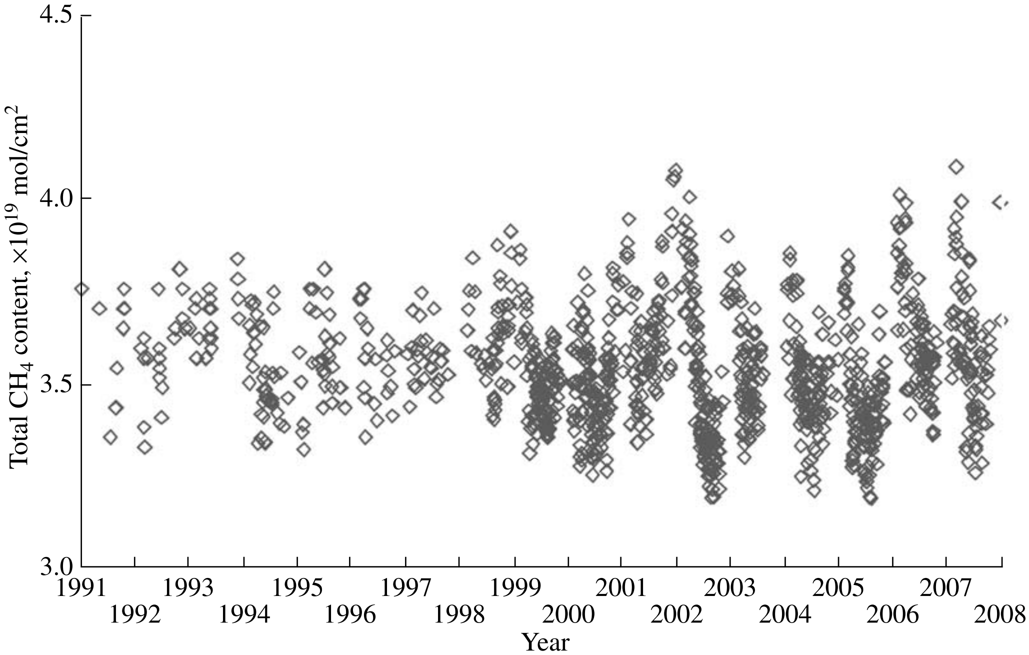

Fig. 1. Daily means of the total CH_4 content in the vicinity of St. Petersburg.

light measurement runs has increased since 1998 (compared to the previous period) due to the measurements carried out even in the cases of short-term sky clearings, not because of an increase in the number of sunny days. The characteristic property of the spectroscopic measurements is their dependence on weather conditions (clear skies or cloud gaps sufficient to take measurements are necessary). In addition, in fall and winter, the number of measurement runs is reduced due to a decrease in daylight hours. Note that the influence of both weather conditions and the period of daylight hours on the number of measurement runs is characteristic of any spectroscopic measurements.

The daily mean of the total CH_4 content was calculated as a weighted average of the individual measurements of the total CH_4 content during a day. The quantity inversely proportional to the rms disagreement between the measured and calculated spectra of solar radiation was used as the weight of an individual measurement. This quantity is an objective assessment of the quality of the solution of the inverse problem in determining the total content of gas. The random error of the individual measurement of the total methane content amounts to 4–6% [8]. The error of the daily mean of the total CH_4 content usually amounts to 1–3% and depends on meteorological conditions [8–10].

Table 1 gives the extreme (maximum w_{max} and minimum w_{min}) values of the total CH_4 content; its average (w_{avr}), median (w_{med}), and rms deviation (σ); and the coefficients of asymmetry μa and excess μe. According to the results of measurements of the total CH_4 content in the atmosphere over the vicinity of St. Petersburg, the minimum daily mean was observed on July 27, 2005, and the maximum daily mean was observed on February 21, 2007.

The meaning of some of the statistical characteristics is clearly seen in Fig. 2, which gives a histogram of the recurrence frequency of the total CH_4 content values. The values of w_{med} and w_{avr} marked by the vertical lines are very close (under a symmetrical distribution of random quantity, the median coincides with the mean). The positive coefficient of asymmetry implies a deviation of the distribution given in Fig. 2 towards large values (this is manifested in a longer "tail" in the region of the large values of the total CH_4

Table 1. Statistical characteristics of the series of the total CH_4 content

$w_{max} \times 10^{19}$ mol/cm^2	$w_{min} \times 10^{19}$ mol/cm^2	$w_{avr} \times 10^{19}$ mol/cm^2	$w_{med} \times 10^{19}$ mol/cm^2	$\sigma \times 10^{19}$ mol/cm^2	μa	μe
4.09	3.19	3.54	3.52	0.16	0.2	0.5

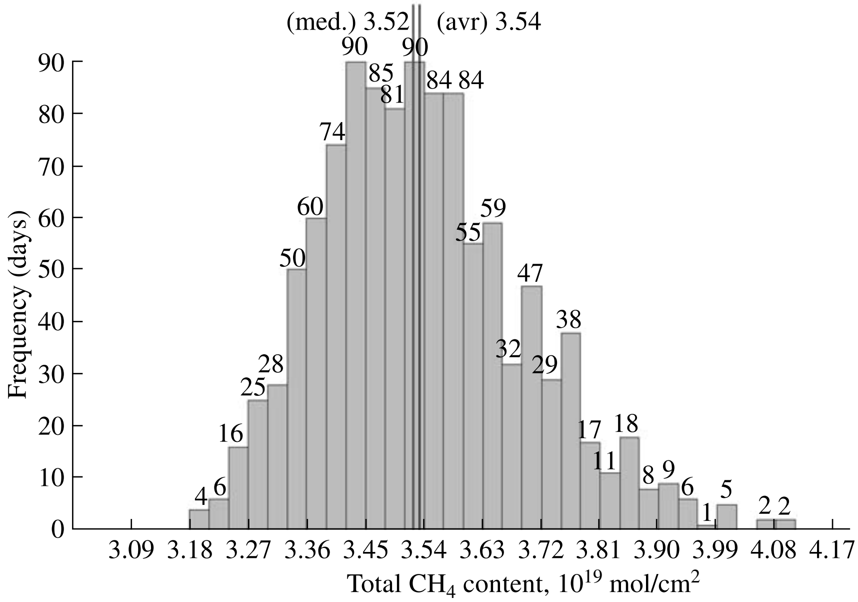

Fig. 2. Histogram of the recurrence frequency of the total CH_4 content values.

content). The excess coefficient μe is small, but its positive value suggests a more peaked distribution than is normal.

2.2. Annual Variations in the CH_4 Content

The time variations in the total CH_4 content include its periodic seasonal, long-term, and irregular variations. To characterize seasonal variations in the total CH_4 content in the vicinity of St. Petersburg, Fig. 3 is given, which demonstrates the mean annual cycle for the entire period of observations (1991–2007) and some examples of the annual cycles for different years of observations (2001, 2002, 2006, and 2007).

The obtained mean annual (also called seasonal) cycle of the total CH_4 content for 1991–2007 is characterized by its maximum values in December–January and its minimum values in June–August, and its amplitude amounts to ~3.6%. It follows from Fig. 3 that the character of annual variations for some years can differ significantly from the obtained mean annual cycle. The seasonal variations observed in 2007 are close to the mean annual cycle, but they have a higher total content of CH_4 in February–March and a pronounced minimum in July. In 2006, the local maximum of the total CH_4 content was observed in May–July and its minima were observed in April and September. In 2001 and 2002, the minima of the total CH_4 content were observed in April and August, respectively. The amplitude of the annual cycle for a certain year (see Fig. 3) may differ significantly from its mean (~3.6%). Thus, for example, in 2002, the amplitude of the annual cycle amounted to ~8%. The differences in the character of seasonal cycle for different years are associated with the natural year-to-year variability of the intensity of the sources and sinks of CH_4, which affects the amount of methane in the atmosphere [11]. For example, a deep minimum in the summer of 2002 was most likely caused by dry and hot weather conditions; this resulted in a decrease in the CH_4 emission due to the drying up of wetlands (the powerful natural sources of methane in the northwestern region).

The observed interannual variability of the mean annual cycle of the total CH_4 content is characterized by the rms deviations (σ_m) calculated for each month

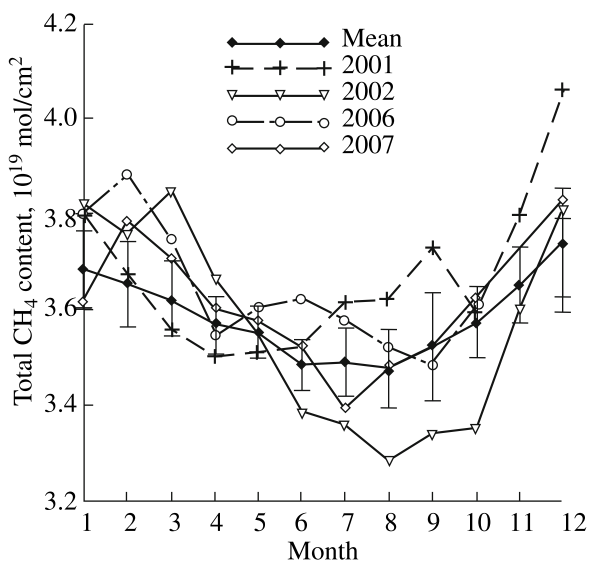

Fig. 3. Annual variations in the total methane content in the vicinity of St. Petersburg.

Table 2. Indices of the linear trend (r) of the total CH_4 content for two periods of observations

Month	r, % per year	
	1991–2002	1991–2007
January	–	0.6 ± 0.2
February	0.3 ± 0.6	0.4 ± 0.2
March	–0.2 ± 0.6	0.1 ± 0.3
April	–0.3 ± 0.7	–0.2 ± 0.3
May	–0.6 ± 0.6	–0.1 ± 0.2
June	–0.2 ± 0.5	–0.2 ± 0.2
July	–0.2 ± 0.6	–0.3 ± 0.2
August	–0.4 ± 0.5	–0.2 ± 0.1
September	–	–0.4 ± 0.2
October	–	–0.5 ± 0.1

and given in Fig. 3. For St. Petersburg, the values of σ_m from April to August are lower (the minimum is observed in May–June) than for the fall–winter period. This is partially due to the small number of observation days in fall and winter and to the strong variability of the total CH_4 content in the cold period (which may be caused by an increase in the intensity of anthropogenic sources in fall and winter).

The surface mixture relations CH_4 (q_0) for the NOAA CMDL nearest (in latitude) stations (Baltic, 55.5° N, 16.7° E, and Iceland, 63.2° N, 20.2° W) have seasonal variations which are similar (in character) to the total CH_4 content variations for St. Petersburg [12]. However, the amplitudes of the annual cycle for the surface mixture relations at the NOAA CMDL stations do not exceed 1.5–2% [12].

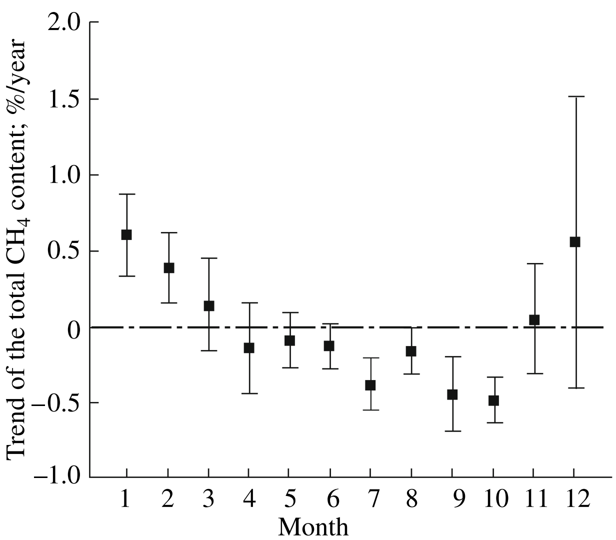

Fig. 4. Indices of the linear trend r for different months over the period 1991–2007.

2.3. Long-Term Variations (Trends) in the CH_4 Content

To estimate the long-term variations (trends) in the total CH_4 content (r, %/year), its monthly means were used. This made it possible to eliminate the necessity for excluding the seasonal cycle of the total methane content and to perform calculations only for the months provided with a sufficient amount of ground-based measurement data. Table 2 gives the values of r for two periods (1991–2002 and 1991–2007). In these calculations, the linear regression (the least-squares method) was used for the months best provided with data. Figure 4 graphically shows the monthly dependence of the index of the linear trend r (for 1991–2007) and the errors in estimating r.

Here it is important to emphasize that, for 1991–2002 (see Table 2), no statistically significant linear trend has been revealed: the errors in determining r exceed its values. For 1991–2007, in January–February and in July–October, the values of r are higher than their errors. The indices of the linear trends for these months have opposite signs: positive for January–February and negative for July–August. The obtained values of r do not allow one to draw an unambiguous conclusion about the trend of the total methane content for the period under consideration. The value of the correlation coefficient calculated for the whole series of the daily means of the total CH_4 content (implying a correlation between the file of daily means and time) implies the absence of a significant linear trend for the whole series of observations.

The dissimilar signs of r for the winter ($r > 0$) and summer ($r < 0$) months suggest an increase in the amplitude of the annual cycle of the total CH_4 content in the atmosphere over St. Petersburg in 1991–2007. This increase may be associated with a decrease in the lifetime of methane in the atmosphere [11, 13, 14]. A global redistribution of the sources of methane (and the increasing influence of Asia) is assumed to be the biggest cause of these phenomena. This conclusion is based, for example, on the calculations of the CH_4 lifetime which were performed with a global three-dimensional model of the atmosphere [14, 15].

For comparison, let us present published estimates of the trends of the total CH_4 content obtained from ground-based spectroscopic measurements at other stations. According to the results of long-term (over 30 years) observations at the ZSS, the increase in the total CH_4 content amounted to ~0.5%/year over the period 1974–2007, and, in 2001–2007, this increase went down to ~0.2%/year [16].

The results of the measurements (taken at the IEM) of CH_4 in the surface air layer and in the entire atmospheric thickness show that, within the linear approximation, the increase in the CH_4 concentration in the surface air amounted to approximately 0.003 ppm/year (~0.2%/year) over the period of January 1998–

December 2001 [6]. The increase in the CH_4 concentration in an atmospheric column amounted to about 0.008 ppm/year (~0.5%/year) over the period of May 1998–April 2001 [6].

At the NDACC stations closest to St. Petersburg [17], a slight positive linear trend was observed over the period 1995–2005 (Table 3). The maximum value of the trend was observed at the Harestua station (located in the same latitude as St. Petersburg), and its minimum value was observed at the Zugspitze high-altitude station. It is seen that the value of the trend of the total CH_4 content depends on the geographical location of the stations.

Out of eight stations (St. Petersburg, Zvenigorod, and six NDACC stations), the minimum index of the linear trend was observed for St. Petersburg (lack of increase) in 1991–2007, and its maximum index was observed for Obninsk in 1998–2001. It should be noted that here we are dealing only with a qualitative comparison of the trends obtained at these stations, because both the time periods and the methods used in estimating long-term variations in the total CH_4 content are different.

A large amount of data on surface methane concentrations have been obtained at the ground stations of two observation systems (NOAA/GMD and AGAGE). These stations (usually oceanic or coastal) are located far from the intensive sources of methane. The instrumentation used at these stations is characterized by high accuracy. The results of measurements show that, over the past 25 years, the methane content has increased approximately by 25%; however, in recent years, the rate of its increase has significantly decreased. In the late 1970s and early 1980s, *r* amounted to 1%/year; in 1999–2005, *r* had low values close to zero [12]. As for the total methane content, regional variations are observed in the trends of the surface concentration of methane.

Table 3. Trend estimates obtained at the NDACC stations for 1995–2005 [17]

NDACC station	Latitude	Height, km	Trend of the total CH_4 content, %/year
Ny-Alesund	79° N	0.02	0.14 (±0.08)
Kiruna	68° N	0.4	0.35 (±0.08)
Harestua	60° N	0.6	0.40 (±0.06)
Zugspitze	47° N	2.96	0.12 (±0.05)
Jungfraujoch	47° N	3.58	0.17 (±0.03)
Izana	28° N	2.36	0.14 (±0.09)

2.4. Spectral Analysis of the Total Content of CH_4

The total content of CH_4 was spectrally analyzed on the basis of a series of the monthly means obtained within the observation period 1991–2007. The data on the monthly means of the total methane content were a 161-month-long series. The data for each month were obtained through the averaging of daily means. In this case, the algorithm of the modified classical Fourier analysis was used to study the time series which contain nonuniformly located omissions [18–20]. This method has a number of advantages (in particular, a higher accuracy than the classical Fourier analysis), especially in analyzing harmonics, the periods of which are comparable to a series length. The significance of harmonics was assessed with the Brooks–Carruthers method [21]. It should be noted that, in the presence of omissions (1–9 months), an increase in the noise component in the spectral region with periods of two to ten months was noted in the sets of observational data [18–20].

The following algorithm was used to make a spectral analysis for a series of the monthly means of the total CH_4 content [18, 19]:

(1) The spectral composition of the initial series (having omissions) was analyzed with the modified classical Fourier analysis. The periods and amplitudes of the basic harmonics were determined.

(2) The data omissions were filled with a 12-month harmonic with a known phase (on the basis of the known mean annual cycle), and the spectral composition of the filled series was analyzed.

(3) On the basis of a comparison between the results of an analysis of the initial and filled series, spectral components stably pronounced in both the series were revealed.

The results of the Fourier analysis are given in Fig. 5, where the spectral range under analysis is divided into three regions: (1) from 4 to 10 months (Fig. 5a), (2) in the vicinity of the annual harmonic from 10 to 15 months (Fig. 5b), and (3) from 15 to 80 months (Fig. 5c).

In Figs. 5a, 5b, and 5c, the figures indicate the periods of the observed basic harmonics and the level above which the amplitudes are regarded as significant with the indicated confidence probability.

As a result of the analysis made for the period 1991–2007, the following spectral components can be revealed with a confidence of 80%: 8, 12, 13, 18, 26, 32, and 55 months. Only the harmonics 12, 32, and 55 months are reliable at a level of 90%. For the entire period of observations, the maximum values of spectral density are concentrated in the vicinity of 12, 24–38, 44–70 months.

The annual harmonic of the total CH_4 content is most pronounced; its amplitude amounts to about 3% of the mean of the total CH_4 content (over the entire period of observations), which is in sufficiently good agreement with the estimate obtained earlier (3.6%)

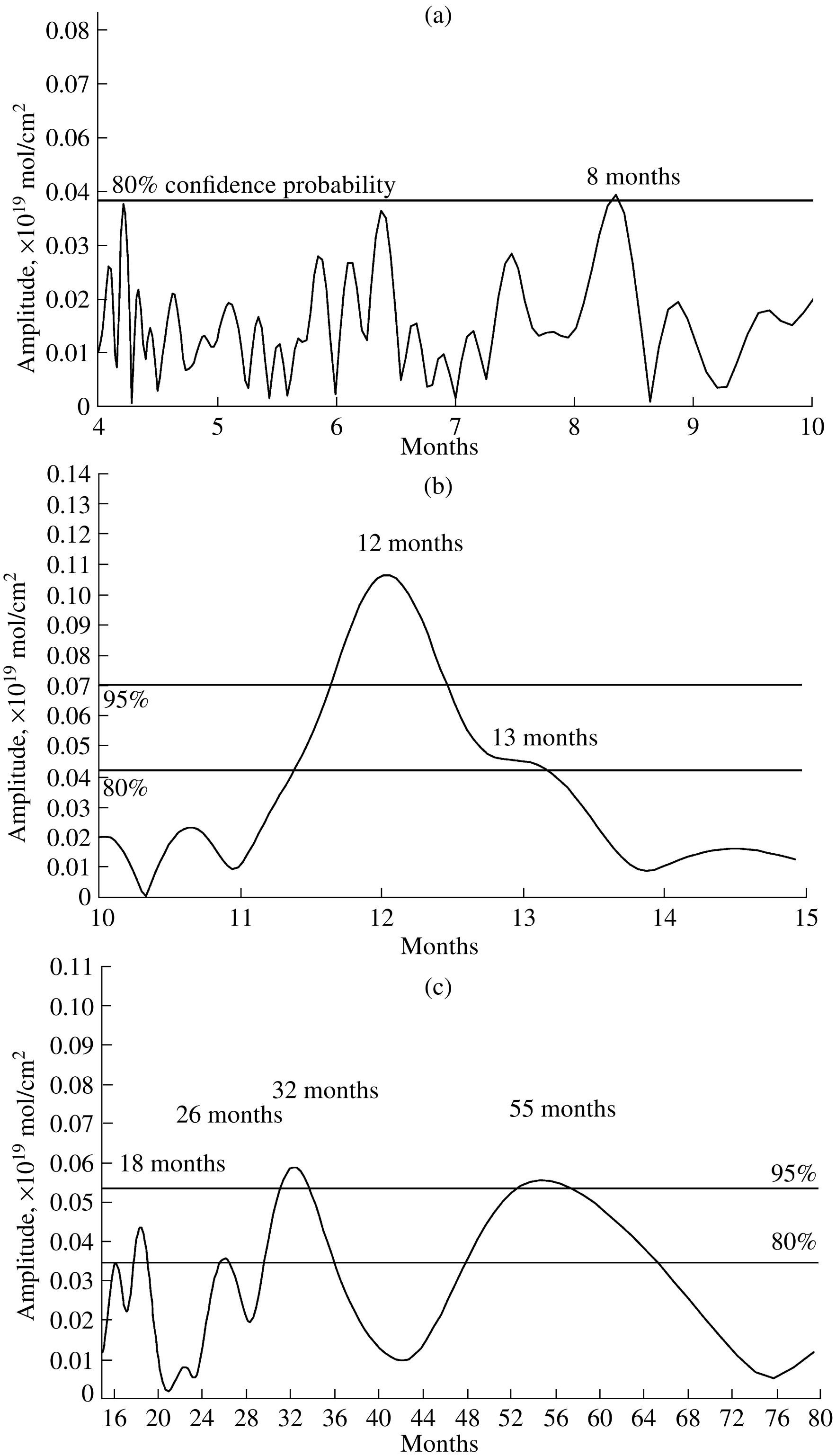

Fig. 5. (a) Spectral composition of the total CH_4 content series (1991–2007), periods of 4 to 10 months; (b) spectral composition of the total CH_4 content series (1991–2007) in the vicinity of the annual harmonic, periods of 10 to 15 months; and (c) spectral composition of the total CH_4 content series (1991–2007), periods of 15 to 80 months.

for the amplitude of the mean annual cycle. Not only the annual harmonic, but also a number of other harmonics with periods of about 8, 13–15, and 18 months can be observed in meteorological series, as well as in the series of the total O_3 content [18, 19]. When analyzing the series of the total content of CH_4 with a confidence probability of 80%, oscillations corresponding to periods of 8, 13, and 18 months were revealed.

The obtained oscillations with periods of 26 and 32 months belong, apparently, to quasi-biennial oscillations (QBOs). The amplitude of the most intensive period of the QBOs (32 months) for the total CH_4 content amounts to 1.7% of its mean (3.54×10^{19} mol/cm). Such oscillations are manifested as singlet, doublet, and triplet oscillations, for example, in the series of the total O_3 content in different latitudes [19, 22].

There are also long-period oscillations with a period of 55 months in the spectrum (4.5 years), which is close to a doubled period of the quasi-biennial harmonic. Their amplitude is also noticeable and amounts to ~1.6%. In literature [19, 23], the manifestation of the quasi-five-year oscillations (a period of 4–6 years) is noted in the form of the recurrence of the El Niño phenomenon, in meteorological series, and in the series of the total ozone content.

3. BASIC RESULTS AND CONCLUSIONS

The following results and conclusions have been obtained and drawn on the basis of an analysis of a series of ground-based spectroscopic measurements of the total CH_4 content (1991–2007).

(1) The mean annual cycle of the total methane content over the period 1991–2007 is characterized by its maximum values in December–January and its minimum values in June–August. The character of annual variations for individual years may differ significantly from the obtained mean annual cycle. The amplitude of the mean annual cycle of the total methane content amounts to ~3.6%.

(2) No statistically significant linear trend of the total CH_4 content has been discovered for the 1991–2007 period. The indices of the linear trends obtained for January–February and July–August have opposite signs: positive for January (0.6 ± 0.2%/year) and February (0.4 ± 0.2%/year) and negative for July (–0.3 ± 0.2%/year) and August (–0.2 ± 0.1%/year). This fact suggests that the tendency for an increase in the amplitude of the annual cycle of the total CH_4 content was observed in 1991–2007.

(3) For the monthly means of the total CH_4 content throughout 1991–2007, the following spectral components were revealed with a confidence of 95%: (a) a 12-month harmonic mainly determining the annual cycle of the total CH_4 content, (b) a 32-month harmonic belonging to quasi-biennial oscillations, and (c) oscillations with a period of 55 months (4.5 years).

At a confidence probability of 80%, the following periods are additionally manifested: 8, 13, 18, and 26 months.

Many of the revealed periods are manifested not only in the total CH_4 content but also in the series of meteorological parameters and in the series of the total O_3 content. This suggests that the basic mechanisms determining the spectral composition of the variability of these geophysical parameters are common.

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