

**METHANE IN THE SURFACE LAYER OF THE ATMOSPHERE:
CURRENT CONTENT, LONG-TERM TRENDS,
AND INTRA-ANNUAL VARIABILITY**

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Abstract. The article deals with the empirical analysis of series of monthly mean concentrations of methane in the near-surface layer of the atmosphere from the global network of monitoring stations. They operate within the Global Atmosphere Watch (GAW) under the auspices of the World Meteorological Organization (WMO). The data is freely available at the World Data Center for Greenhouse Gases GAW/WMO (WDCGG) on its website <https://gaw.kishou.go.jp/>. The temporal coverage is from the 1980s. Data series from 69 stations are considered, of which 22 stations represent global background conditions. The rest of the stations are regional. Long-term trends in concentrations and the intra-annual (inter-monthly) deviations of monthly mean concentrations from long-term trends were studied. The multi-year trend was estimated using a series of 12-month running averages. To exclude systematic differences in methane concentrations, these series were adjusted to the series for the high-latitude Arctic station Alert (82° 30' N, 62° 21' W). The analysis showed that long-term trends are non-linear (in particular, a known pause in the growth of methane levels in 1999-2006 is observed), but are similar at most stations under consideration. Exceptions are six regional stations classified as “abnormal” in terms of methane. Possibly, this abnormality is due to the influence of certain sources of methane (anthropogenic or natural). Long-term trends at the rest of the stations just slightly differ from the average trend for the global stations. The series of intra-annual (inter-monthly) deviations of monthly mean concentrations from long-term trends for many stations (even those located at very significant distances from each other) show high correlative similarity. However, this similarity manifests itself at an optimal time shift from 5 months towards earlier dates up to 6 months towards later dates. The results of the analysis are consistent with the assumption that the intra-annual variability in methane concentration is largely driven by seasonal factors that are significantly related to latitude, such as vertical mixing in the atmosphere and

destruction in the troposphere in reactions with hydroxyl. The root-mean-square values of intra-annual (inter-monthly) fluctuations in methane concentration depend significantly on latitude. In general, the higher the latitude is, the greater is the value. The maximum values are reached in the latitudinal belt within 45-50° N, and further to the North the values decrease. This feature of intra-annual fluctuations in the level of methane content may be explained, among other things, by significant inter-seasonal fluctuations in anthropogenic methane emissions occurring at the indicated latitudes in the countries with developed economies located in North America and Western Europe. The correlations of the series of intra-annual (inter-monthly) fluctuations of the monthly mean concentrations of CH₄ and CO₂ were estimated as rather high, about 0.8, at optimal time shifts, which is observed both at some polar stations and at tropical ones. This confirms the assumption that natural seasonal biogeochemical and geophysical processes play a significant role in the formation of intra-annual (inter-monthly) deviations of methane and carbon dioxide content in the near-surface layer from long-term trends. These processes include vertical mixing of air, CO₂ absorption on the Earth's surface, and destruction of methane in the troposphere in reactions with hydroxyl.

Keywords. Earth's atmosphere, near-surface layer, methane, monitoring data, global analysis, current concentrations, long-term trends, intra-annual variability, spatial patterns, correlative similarity.

Introduction

We owe the greenhouse effect on Earth mainly to five gases: water vapor H₂O, carbon dioxide CO₂, methane CH₄, nitrous oxide N₂O, and ozone O₃. Methane, constituting an insignificant part of the atmospheric air (currently about 2 ppm), has a powerful effect on the Earth's climate. With a time horizon of 100 years for GWP (Global Warming Potential), methane enhances the greenhouse effect about 30 times more efficiently than CO₂ per unit of emission into the atmosphere (Myhre et al., 2013, p. 8SM-39). Anthropogenic sources such as rice growing, animal husbandry, and biomass combustion account for about 50% of global annual methane emissions from the Earth's surface into the atmosphere (Kirschke et al., 2013).

Systematic measurements of the content of greenhouse gases in the near-surface layer of the atmosphere began in the second half of the 20th century. In 1958, C.D. Keeling and his team began measuring the CO₂ content at the Mauna Loa monitoring station (Semenov, 2018). This led to a number of important conclusions (Keeling et al., 2005). Since then, the network of monitoring stations that measure the content of greenhouse gases in the atmosphere has become global. At the international level, these stations operate within the Global Atmosphere Watch (GAW) under the auspices of the World Meteorological Organization (WMO). They are located on the continents and islands in the ocean and characterize the content of greenhouse gases in the near-surface layer of the atmosphere.

The station observational data are stored at the World Data Center for Greenhouse Gases GAW/WMO (WDCGG). Its resource <https://gaw.kishou.go.jp/> also contains data on CH₄ levels which are analyzed in this article.

The purpose of this work is to carry out a global analysis of the current methane content in the near-surface layer, long-term trends, and intra-annual variability based on the data from the global network of GAW monitoring stations on the methane content in the near-surface layer of the atmosphere.

Data and methods

This paper analyzes 69 station series of data on the monthly mean methane content in the near-surface layer of the atmosphere presented on the resource <https://gaw.kishou.go.jp/> (publicly available data, downloaded on 23.03.2020). The initial measurements were carried out by various research teams in the course of *in situ* measurements or subsequent laboratory analysis of air samples taken in special containers.

The global distribution of stations is shown in Fig. 1. Twenty-two of them are classified as global stations. They represent the global background methane content, as they are not under the direct influence of certain CH₄ emission sources. The series of other regional stations characterize regional levels and trends.



Figure 1. Global distribution of GHG monitoring stations the methane data from which are used in this work; the criterion for abnormality of regional stations will be discussed below

To accomplish the analysis of the current methane content, long-term trends, and intra-annual variability of methane content in the surface layer of the atmosphere, this study uses the above data. The analysis was carried out in accordance with the methodology proposed in (Semenov, Kuzovkin, 2020). In the calculations, the programming language *R* was employed (The *R* Project for Statistical Computing, <https://www.r-project.org/>). The main methodological points are given below in brief.

To estimate long-term changes in CH₄ content for a station, its series of 12-month running averages $\{B(n)\}$ is used. It is constructed from the initial series $\{A(n)\}$ of monthly mean methane concentrations at the station. For a given month n , respective 12-month average is calculated using the value of the n -th month, 6 months prior, and

6 months later. The calculation is only performed if all these initial data are available. When calculating the average, the data of the first and last months are taken into account with a factor of 0.5, and the rest terms with a factor of 1.

To compare long-term trends at different stations, instead of the series $\{B(n)\}$ we used their analogs, namely, the series adjusted to the reference station Alert (82.5° N; 62.34° W), due to the following reasons. Different points in the geographic space differ in the level of anthropogenic emission of methane into the atmosphere, the rate of its chemical runoff, and transport processes in the troposphere. This leads to systematic differences in methane levels, which should be excluded when assessing the trends. For this, for each station, the average difference between the values of $B(n)$ at this station and those at the Alert station was calculated (of course, for the months for which these values are available for both stations). Then this correction was added to each element of the series $\{B(n)\}$ for the given station. The resulting series is denoted $\{B^*(n)\}$. The calculated corrections were subsequently used to characterize systematic differences in methane levels at Alert and other stations.

The choice of the Alert station as a reference one is conventional. This high-latitude station obviously represents the global background and has a fairly complete set of observational data (collected since 1985).

In nature, seasonal variations are superimposed on long-term changes in the methane content in the atmosphere. Indeed, not only emissions (natural and anthropogenic) from the Earth's surface into the atmosphere have seasonal components but also the processes of horizontal transfer and vertical mixing, as well as destruction in reactions, in particular, with hydroxyl (Kiselev, Karol, 2019). Therefore, besides analyzing long-term changes, it is important to assess seasonal variations in methane content. For this purpose, here the series $\{C(n)\}$ is used, which is obtained by subtracting the corresponding series $\{B(n)\}$ from the series $\{A(n)\}$. The series $\{C(n)\}$ characterizes intra-annual (inter-monthly) seasonal fluctuations.

Series $\{C(n)\}$ for different stations are compared according to their root-mean-square value (the analog of the oscillation amplitude) and according to the "optimal" time shift. For an ordered pair of series, this indicates a time shift by k months of the second series relative to the first one, i.e., transition to the series $\{C(n+k)\}$ at which the correlation of the series reaches its maximum. In this case, when searching for the maximum, the values of k from (-5) to 6 are considered. For $k > 0$, this will be called a shift by k months against time (i.e., towards earlier dates), and if $k < 0$, by $|k|$ months along time (i.e., towards later dates). Such estimates of the optimal shift can be made not only for a pair of series of methane content, but also for a pair of series for different gases, e.g., for methane and carbon dioxide.

Results and discussion

Multi-year trends and absolute content levels

The results of calculating the average of long-term trends (represented by the series $\{B^*(n)\}$) for global stations (GB, characterizes the global background change in methane content) and regional stations (RB, reflects the average regional

change in methane content) are shown in Fig. 2. It can be seen that the RB and GB curves are very close (the differences do not exceed 6 ppb); the regional stations indicate, on average, the same long-term methane trend as the global stations. The similarity of the long-term trends in methane content is possibly explained by the relatively rapid mixing of methane in the lower atmosphere compared to its lifetime: the mixing time is not more than a few months (Eliseev, 2018; Kiselev, Karol, 2019), while the lifetime is about 10 years (Voulgarakis et al., 2013; Eliseev, 2018).

Note that this trend is essentially non-linear. In particular, a pause in the growth of methane levels in 1999-2006 indicated by many authors (for example, (Dlugokencky et al., 2009)) is seen.

In Fig. 2, we also mark with background colors the maximum and minimum individual deviations from the average change for all the stations (for the global stations from GB, and for the regional stations from RB). The difference between the maximum and minimum values of an individual deviation is not more than 12 ppb for global stations. In general, for regional stations it is basically not more than 18 ppb, which approximately corresponds to 1% of the modern values of the methane content in the near-surface layer of the atmosphere.

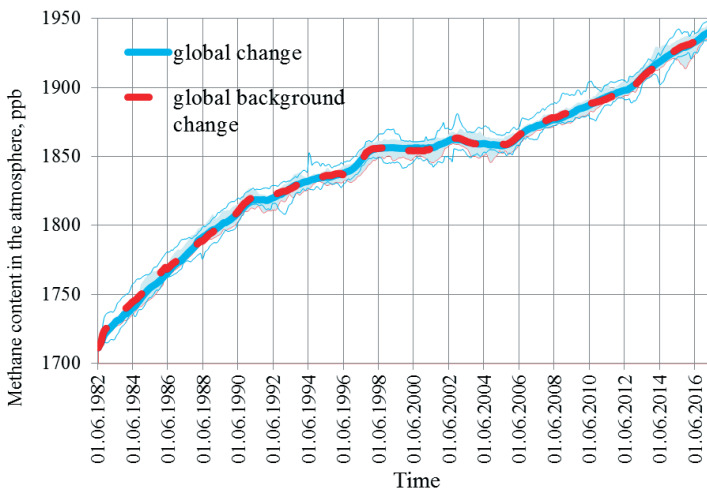


Figure 2. Average long-term changes in CH_4 content in the near-surface layer of the atmosphere (ppb): red for a group of global stations (GB) and blue for a group of regional stations (RB); the difference between maximum and minimum deviations for individual stations from the average change for respective group is shown in pink and blue, respectively

However, for the following six stations, the deviations are higher: Seretta, Southern Great Plains, Park Falls, Constanta, Tateno, Tae-ann Peninsula. We qualify these stations as “abnormal” in terms of methane. Possibly, this abnormality is due to the influence of certain sources of methane emissions. Fig. 3 shows the average long-term changes in CH_4 content in ppb in the near-surface layer of the atmosphere for the whole group of regional stations (RB, blue) and individual changes for “abnormal” regional stations. The figure shows that deviations from the average are substantial for such stations. We exclude these stations from further analysis.

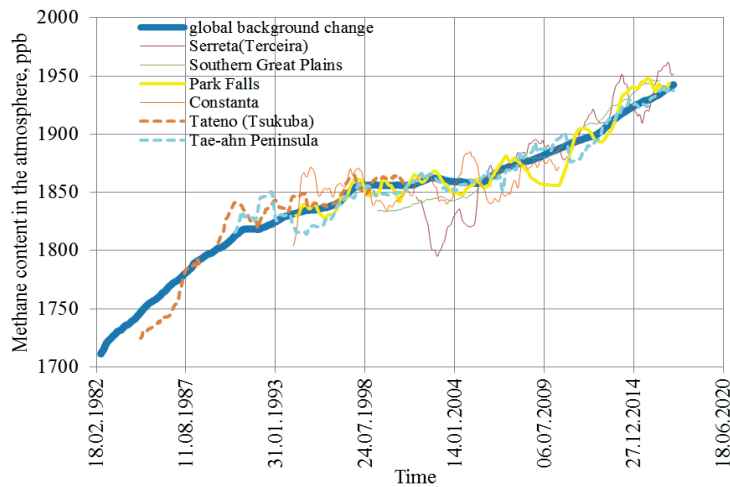


Figure 3. Average long-term changes in the content of CH_4 in the near-surface layer of the atmosphere (ppb) for the whole group of regional stations (RB, blue) and individual changes for “abnormal” regional stations

The results of calculation of systematic differences in methane content at the stations and the Alert reference station are given in Fig. 4. The figure shows that at the continental stations located in North America and Eurasia, especially in the regions with developed economies, the long-term CH_4 levels significantly exceed the values at the Alert station in most cases. In the Southern Hemisphere, the multi-year CH_4 levels are lower than at Alert. With decreasing latitude, differences become more noticeable. In the extratropical zone, they reach 152 ppb, which is about 8-9% of the average methane level for the entire long-term observation period.

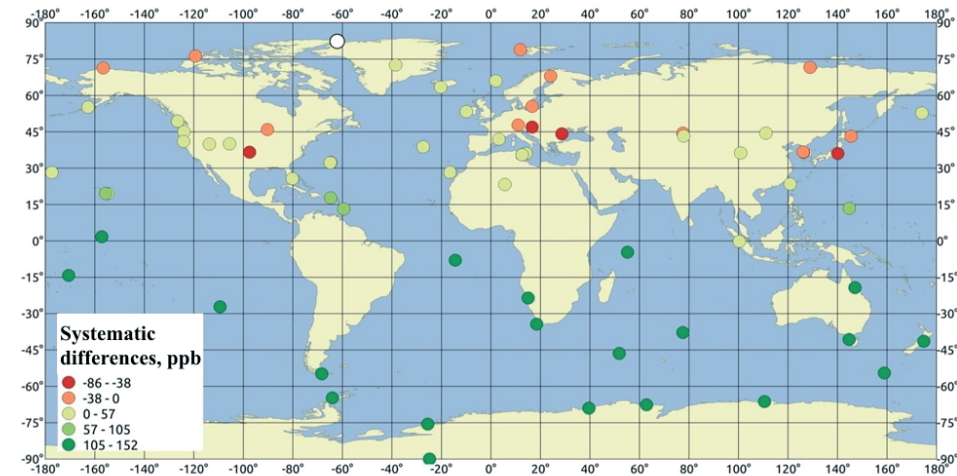


Figure 4. Systematic differences in long-term methane levels at Alert and other stations

There are many possible reasons for this dependence. In particular, the Northern Hemisphere locates more substantial sources of anthropogenic methane emissions

than the Southern Hemisphere due to the higher level of its economic development. This is also true for natural emissions. In the Southern Hemisphere, the ocean occupies a larger part of the Earth's surface than in the Northern Hemisphere, and oceanic methane emissions are small compared to terrestrial ones (Ciais et al., 2013; Kirschke et al., 2013), which come, for example, from huge swamp systems.

Intra-annual variability

Intra-annual (inter-monthly) fluctuations in methane content at a station are represented by the series $\{C(n)\}$, $C(n) = A(n) - B(n)$. The following indices were calculated:

- the root-mean-square value of $\{C(n)\}$ in relative units, namely, in relation to the value for Alert;
- the optimal time shift relative to the Alert station (see "Data and Methods" section).

The results are shown in Fig. 5 and 6.

As seen in Fig. 5, the root-mean-square value is noticeably dependent on latitude. In general, the higher the latitude is, the greater the value is. The maximum values are reached within the latitudinal belt 45° - 50° N, and further to the North the values decrease. A similar dependence was obtained earlier for carbon dioxide (Semenov, Kuzovkin, 2020). This pattern of intra-annual fluctuations in the level of methane content may be explained, among other factors, by significant inter-seasonal fluctuations in anthropogenic methane emissions at the indicated latitudes in the countries with developed economies located in North America and Western Europe.

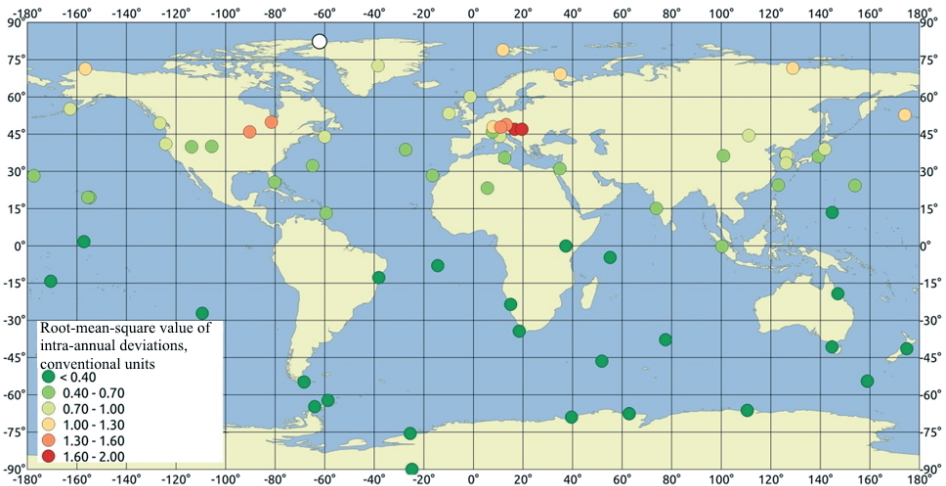


Figure 5. Root-mean-square value of intra-annual deviations of CH_4 content from the long-term trend at different stations; the unit is taken as the root-mean-square-value for Alert (indicated by the white circle)

The results of calculating the optimal time shift for various stations in relation to the reference station Alert are shown in Fig. 6. As seen from the figure, in most of the Northern Hemisphere the intra-annual oscillations are in phase, i.e., the time shift in relation to the Alert station is zero. However, for a number of stations in the

temperate zone (within 30-60° N), the values of the optimal shift are negative, i.e. “methane events” occur earlier than at the Alert station. This is possibly due to significant anthropogenic component of methane emissions in this latitudinal belt. The seasonal distribution of this component substantially differs from the seasonal distribution of the natural component. The optimal time shift in the Southern Hemisphere is positive and increases generally with decreasing latitude. This is consistent with the following concept: intra-annual fluctuations in methane content at stations in the Southern Hemisphere are mainly driven by seasonal processes largely determined by latitude, namely, vertical mixing and destruction of methane in reactions with hydroxyl.

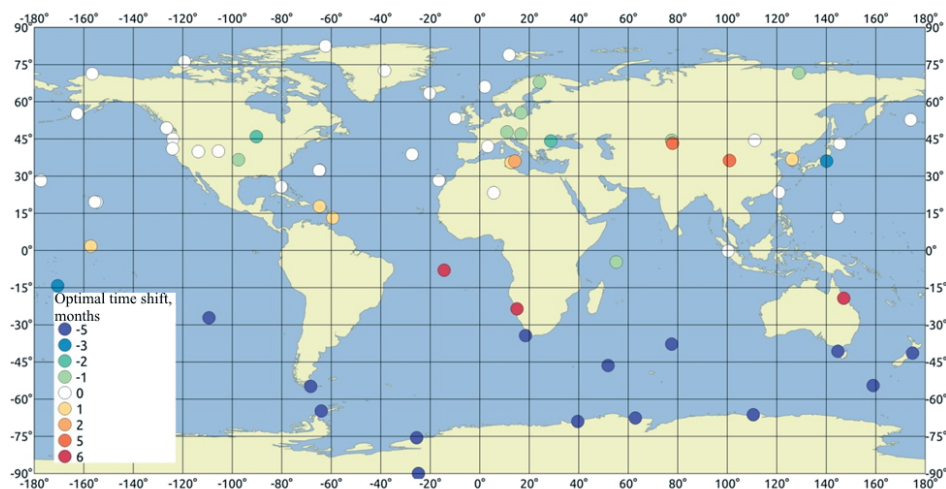


Figure 6. The optimal time shift (months) of the series of intra-annual (inter-monthly) deviations of methane content from the long-term trend for different stations in relation to Alert

For CH_4 , as well as for CO_2 (Semenov, Kuzovkin, 2020), the $\{C(n)\}$ series characterizing the intra-annual (inter-monthly) deviations of the monthly mean levels of methane from the long-term trend can demonstrate high correlations taking into account the optimal time shift. To illustrate this, Fig. 7 shows such series for three stations: Alert, Guam, and Easter Island. These stations are located at a considerable distance from each other. Alert is an Arctic station (82.50° N, 62.35° W, 210 m above sea level). Guam is a station located in the tropical zone of the Northern Hemisphere in the Philippine Islands (13.43° N, 144.77° E, 2 m a.s.l.). Easter Island is a Pacific station in the Southern Hemisphere located roughly at halfway from Chile to Tahiti (27.17° S, 109.42° W, 41 m a.s.l.). Nevertheless, taking into account the optimal time shift of (-5) for the Easter Island station, the $\{C(n)\}$ series of these stations show a similar pattern of changes over time, and correlations are quite high: 0.86 (Alert and Guam) and 0.93 (Alert and Easter Island).

Theoretically, the seasonal factor associated with the solar radiation flux plays a significant role in the formation of intra-annual changes in the levels of methane and carbon dioxide. Its seasonal enhancement stimulates both vertical mixing in the atmosphere, which contributes to the decrease in the content of these gases in the near-surface layer, and the removal of CO_2 from the atmosphere due to photo-

synthesis by plants, as well as the destruction of CH_4 in reactions with hydroxyl. As a result, certain similarity could be expected in the $\{C(n)\}$ series for these gases.

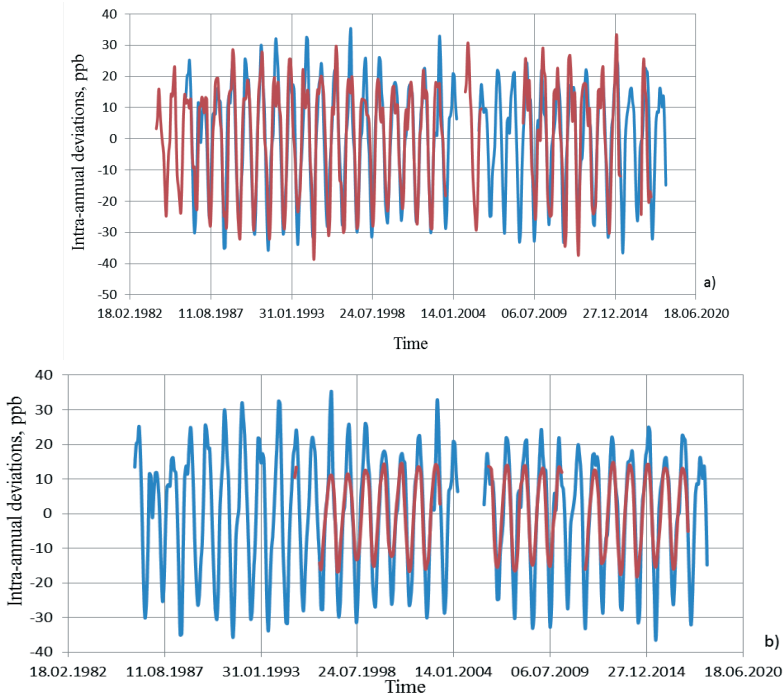


Figure 7. Series $\{C(n)\}$ of intra-annual (inter-monthly) deviations of methane concentration (ppb) from the multiyear trend for stations a) Alert and Guam and b) Alert and Easter Island; a 5-month time shift towards later dates was applied to the series for Easter Island

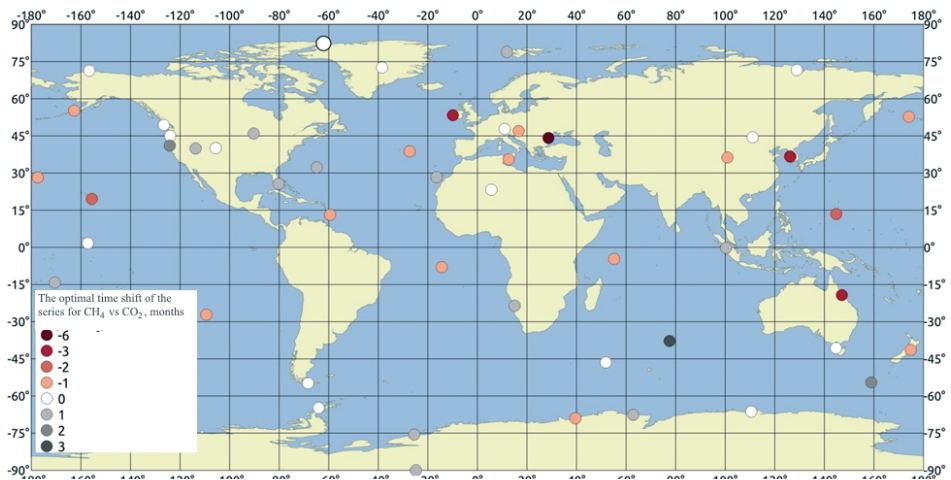


Figure 8. The optimal time shift of the series of intra-annual (inter-monthly) deviations of the methane content from the long-term trend in relation to respective series for CO_2

Fig. 8 shows the optimal time shift of the series of intra-annual (inter-monthly) deviations of a gas content from the long-term trend for CH_4 in relation to CO_2 .

As seen in Fig. 8, at most stations the optimal time shift between the series representing intra-annual fluctuations in the levels of methane in relation to respective series for carbon dioxide is not more than a month in the absolute value. The places where this does not take place are within or near the areas of industry and/or energy production.

Table 1 presents correlations of series $\{C(n)\}$ of intra-annual (inter-monthly) deviations of methane content from the long-term trend and respective series for CO_2 at the optimal time shift of CH_4 series in relation to CO_2 series for different stations.

Table 1. Correlations of series $\{C(n)\}$ for CH_4 and CO_2 at the optimal time shifts of the methane series relative to respective series for CO_2

Station name	Time shift	Correlation	Latitude, °	Longitude, °
Alert	0	0.77	82.50	-62.34
Zeppelin Mountain (NyÅlesund)	0	0.72	78.91	11.89
Summit	0	0.73	72.58	-38.48
Tiksi	0	0.64	71.59	128.92
Barrow (AK)	0	0.72	71.32	-156.61
Cold Bay (AK)	-1	0.47	55.20	-162.72
Mace Head	0	0.51	53.33	-9.90
Shemya Island	-1	0.59	52.72	174.10
Estevan Point	-1	0.50	49.38	-126.54
Hohenpeissenberg	0	0.53	47.80	11.01
Hegyhatsal	0	0.82	46.95	16.65
Park Falls (WI)	-1	0.48	45.93	-90.27
Ulaan Uul	0	0.78	44.44	111.09
Constanta (Black Sea)	0	0.30	44.17	28.68
Trinidad Head (CA)	1	0.46	41.05	-124.15
Niwot Ridge - T-van (CO)	-1	0.68	40.05	-105.59
Wendover (UT)	0	0.70	39.90	-113.72
Serreta (Terceira)	-1	0.49	38.77	-27.38
Tae-ahn Peninsula	2	0.12	36.73	126.13
Anmyeon-do	-1	0.72	36.54	126.33
Mt. Waliguan	6	0.43	36.29	100.90
Lampedusa	1	0.74	35.52	12.63
Tudor Hill (Bermuda)	-1	0.69	32.27	-64.88
Izaña (Tenerife)	-1	0.68	28.31	-16.50
Sand Island	-1	0.72	28.22	-177.37
Key Biscane (FL)	-1	0.73	25.67	-80.20
Assekrem	-1	0.71	23.27	5.63
MaunaLoa (HI)	-3	0.70	19.54	-155.58
Guam (Mariana Island)	-2	0.84	13.43	144.78
Ragged Point	-1	0.83	13.17	-59.43

Christmas Island	-1	0.61	1.70	-157.17
Bukit Kototabang	0	0.37	-0.20	100.32
Mahé	3	0.40	-4.67	55.17
Ascension Island	0	0.65	-7.97	-14.40
Samoa (Cape Matatula)	-3	0.14	-14.25	-170.56
Cape Ferguson	-2	0.40	-19.28	147.06
Gobabeb	0	0.57	-23.57	15.03
Easter Island	-3	0.32	-27.17	-109.42
Cape Point	1	0.68	-34.35	18.49
Amsterdam Island	0	0.67	-37.80	77.54
Cape Grim	1	0.72	-40.68	144.69
Baring Head	1	0.46	-41.41	174.87
Crozet	1	0.75	-46.43	51.83
Macquarie Island	1	0.73	-54.50	158.94
Ushuaia	2	0.79	-54.85	-68.31
Palmer Station	1	0.80	-64.77	-64.05
Casey	1	0.82	-66.28	110.52
Mawson	1	0.76	-67.60	62.87
Syowa	1	0.75	-69.01	39.58
Halley	1	0.73	-75.57	-25.50
South Pole	0	0.78	-90.00	-24.80

As seen in Table 1, the correlations of $\{C(n)\}$ series for CH_4 and CO_2 at optimal time shifts are indeed substantial, up to about 0.8. Moreover, this is observed both at the polar (for example, Casey, Alert) and tropical stations (for example, Guam), which supports the assumption that natural seasonal biogeochemical and geophysical factors play a significant role in the formation of intra-annual (inter-monthly) deviations of methane and carbon dioxide content in the near-surface layer from long-term trends. They are associated with the processes of vertical mixing of gases, absorption of CO_2 on the Earth's surface, and destruction of methane in the troposphere in reactions with hydroxyl.

Conclusions

The empirical analysis of monitoring data reflecting the changes was carried out in this work. It showed that the long-term trend in the level of methane content in the near-surface layer of the Earth's atmosphere in recent decades is non-linear and almost uniform at different points of the geographic space. Stabilization of methane levels in 1999-2006 was observed globally. Their intra-annual variability has significant correlative similarity manifesting itself after the application of time shifts, which generally reflects differences in calendar seasons for different latitudes. The highest absolute levels of methane content and the amplitude of its intra-annual fluctuations are geographically confined to the most economically developed regions.

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