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Carbon Monoxide Variations in the Antarctic Atmosphere from Ground-Based and Satellite Measurement Data

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Abstract—The results of systematic (2003–2017) measurements of the total content and height-averaged relative volume concentration of CO at st. Novolazarevskaya with a spectrometer with a resolution of 0.2 cm^{-1} are presented. The inverse problem of determining the total content of CO as well as interfering gases (H_2O and N_2O) was solved using the SFIT4 software package. A data analysis indicated that during the measurements the average total CO content at st. Novolazarevskaya was (8 ± 2) 10^{17} molecules/cm^2 and the height-averaged volume concentration was (37 ± 8) ppb. The resulting data were compared with variations in the total CO content at st. Arrival-Heights, MOPITT satellite data, and CO surface concentrations at st. Syowa. The maximum and minimum values of CO were observed in September and January–February, respectively. For all the data series considered, the trends are insignificant; in this case, an increased CO content was observed in 2010, and an increasing trend of the minimum values of CO was observed in recent years (2014–2017). Both stations (Novolazarevskaya and Arrival-Heights) are characterized by an excess of satellite data over ground-based measurement data (19% and 14%, respectively); here, a seasonal dependence of the deviation was observed with minimal deviations in December–January. Data of surface measurements of the total content at st. Novolazarevskaya and Arrival-Heights are rather well consistent, and the average deviation since 2010 was 2.4%. The average concentration of CO at st. Syowa (51 ppb) is higher than the height-averaged concentration at st. Novolazarevskaya. According to spectral, wavelet, and composite analyzes, all data series considered include fluctuations in the range of 6–45 months with almost identical periods and phase relationships.

Keywords: carbon monoxide, total content, Antarctica, ground-based and satellite measurements, spectral and cross-wavelet analysis

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INTRODUCTION

Carbon monoxide (CO) is an atmospheric trace gas with a climatic and toxic effect on the environment. The main natural sources of carbon monoxide are the oxidations of methane, chlorophyll, isoprene, and terpenes, oceans and volcanic eruptions, as well as the burning of forests and peat bogs. Carbon monoxide sinks occur in reactions of CO oxidation with OH hydroxyl, during its absorption in soils, and when transported to the stratosphere (Brimblecombe, 1988). The photochemical lifetime of a CO molecule is around 2 months; the content of carbon monoxide in the atmosphere varies significantly from season to season and spatially. The total content and vertical distribution of atmospheric CO is monitored by different satellite observation systems (Timofeev, 2016) as well as instruments of high spectral resolution available in the Network for the Detection of Atmospheric Composition Change (NDACC, https://www2.acom.ucar.edu/irwg/groups) and Total Carbon Column Observation Network (TCCON, https://tccon-wiki.caltech.edu). The local surface concentrations of CO are determined by the NOAA network of international stations—Earth System Research Laboratory (ESRL, www.esrl.noaa.gov). In Russia, the total CO content in the atmosphere is measured spectroscopically at the Institute of Atmospheric Physics, Russian Academy of Sciences (Yurganov et al., 2002), St. Petersburg State University (Makarova et al., 2011), and Research and Production Association "Typhoon" (Kashin et al., 2000). There are regular comparisons of satellite measurements of the total CO content with ground-based spectroscopic data for different regions of the Earth and measurement conditions aimed at obtaining representative satellite data (Rakitin et al., 2015, Sitnov et al., 2017, Buchholz et al., 2017).

The measurements of the total content of carbon monoxide in background conditions are important for studying the CO variations in the atmosphere. Due to the geographical location of Antarctica and the nature of atmospheric circulation, the Antarctic atmosphere is least subjected to anthropogenic impact than other regions of the Earth. Therefore, the observed variations in trace gases have primarily natural causes. Since 2003, the RPA "Typhoon" together with the Arctic and Antarctic Scientific Research Institute (AASRI) has been monitoring the gas composition (CO_2, CH_4, N_2O, CO, and H_2O) of the Antarctic atmosphere. This study describes the results of systematic (2003–2017) measurements of the total content and height-averaged relative volume concentration of CO at st. Novolazarevskaya (70.78° S, 11.82° E, 120 m above sea level). The data obtained are compared with measurements of the total content of CO at the NDACC ground-based station Arrival-Heights (77.82° S, 166.65° E), with satellite data of MOPITT (Measurements of Pollution in the Troposphere, Multispectral CO Total Column, MOP03JM_007, Daytime/Descending) as well as surface CO concentrations at the ESRL Syowa station (69.00° S, 39.58° E).

INSTRUMENTATION AND TECHNIQUES

Spectra are recorded by a spectrometric system of instruments (Kashin et al., 2000) consisting of a solar tracking system, a spectrometer, and a computer that provides control over the system as well recording and storage of data.

The system is located in a separate measuring pavilion with a rotary mirror of the tracking system installed on the roof. The solar radiation enters through a hole in the roof into the optical matching system with a spectrometer. The spectral resolution of the spectrometer is 0.2 cm^{-1}. The measurements were conducted using direct solar radiation at solar heights of more than 15°. The recording time of a spectrum is 6 min. From 2003 to 2009, the system had been repeatedly improved; in 2006, it was destroyed by hurricane wind gusts; since April 2010, no changes have been made.

The total content of CO is determined by the method of solar molecular–absorption spectroscopy. The content of carbon monoxide is measured using the CO vibrational–rotational band ν_3 with a center of 2141.3 cm^{-1}. In the atmosphere, this band is overlapped by spectral lines of the absorption bands of H_2O, CO_2, and N_2O. Therefore, the operating spectral range is chosen so that the influence of the absorption lines of interfering gases on the CO spectrum is minimal. Based on the calculation results, we choose an optimal operating band of the spectrum (from 2150.0 to 2160.0 cm^{-1}) including lines of carbon monoxide, lines of water vapor, and weak lines of carbon dioxide and nitrous oxide. The experimental spectra were bound by frequency through the R1–R3 absorption lines of CO.

Continuing and developing the previous works (Kashin et al., 2007), this study differs significantly by the calculation method: the inverse problem of determining the total content of CO and interfering gases (H_2O and N_2O) was solved using SFIT4 v0.9.4, a freeware software package of the University Corporation for Atmospheric Research (https://wiki.ucar.edu/display/sfit4/).

We have adapted this package to the MDR-23 low-resolution device installed at st. Novolazarevskaya. To this end, we developed a program for linking spectra by frequency and forming a structured input file, a program for determining the solar zenith angle, and a program for generating and visualizing output data. The HITRAN-2012 atlas (Rothman et al., 2013) was used as an atlas of spectral lines; a priori profiles of temperature, pressure, and gases were taken from the Whole Atmosphere Community Climate Model (WACCM, https://www2.acom.ucar.edu/gcm/waccm); they were given the same as for st. Syowa, which is located relatively close.

For comparison with the results of measurements at st. Novolazarevskaya, we used the data of measurements of the total content of CO by a Bruker IFS125HR high-resolution instrument at st. Arrival-Heights (FTIR CO Total Column; NDACC; ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/arrival/hdf/ftir/). The comparison with the measurement data of the MOPITT satellite instrument was performed using the Giovanni database (https://giovanni.gsfc.nasa.Gov/giovanni/). To do this, we averaged the measurement data on the total content of CO for 3° × 3° cells covering the Novolazarevskaya and Arrival-Heights stations (10–13)° East, (70–72)° S and (165–168)° E, (76–79)° S, respectively. For comparison with the measurement data of surface concentrations at st. Syowa (ESRL, Global Monitoring Division; ftp://aftp.cmdl.noaa.Gov/data/trace_gases/co/flask/surface/), the data of st. Novolazarevskaya converted for dry air to height-averaged volume concentration in ppb.

For brevity, the data of ground-based stations are denoted further in the text as NL (Novolazarevskaya), AN (Arrival-Heights), and SY (Syowa). The additional letter G (ground) indicates the results obtained by ground-based instruments and the letter S (satellite) indicates satellite data. The height-averaged volume concentration and the total content are specified by the subscripts X and TC, respectively. For example, for st. Novolazarevskaya, NL_G_X is the height-averaged volume concentration in ppb and NL_G_{TC} NL_S_{TC} is the total content CO in molecules/cm^2 according to ground-based and satellite measurements (above st. Novolazarevskaya).

RESULTS OF GROUND-BASED AND SATELLITE MEASUREMENTS

As an example, Fig. 1 shows the results of single measurements during the periods of maximum (October) and minimum (January) values of the CO concentration.

During the day, the total content and averaged concentration of CO are characterized by insignificant

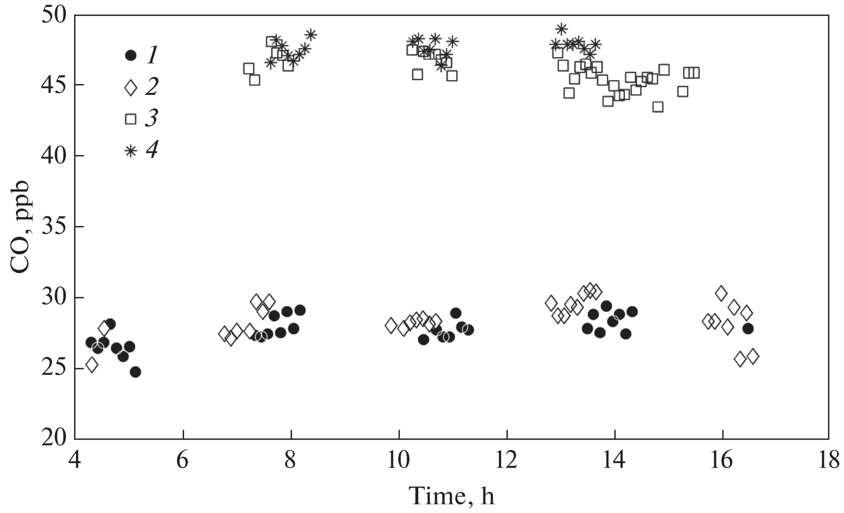

Fig. 1. Diurnal variability of CO at st. Novolazarevskaya. (*1*) January 4, 2009; (*2*) January 17, 2009; (*3*) October 11, 2018; and (*4*) October 29, 2017.

fluctuations that fit within a random instrumental error. The time intervals between measurement series are explained by concurrent measurements of other gases. The instrumental random error in determining the CO content obtained in terms of repeated measurement results during the day is ×4%. The results of calculations of the total content of CO for 2003–2007 coincide with the data given in (Kashin et al., 2007) within the error limits.

The results of CO measurements at st. Novolazarevskaya and the data of st. Arrival-Heights and st. Syowa shown in Fig. 2 clearly demonstrate seasonal variations in CO in the Antarctic atmosphere. The CO content varies from maximum values in September–October (Antarctic spring) to minimum values in January–February (end of the polar day at st. Novolazarevskaya) and begins to increase in March–April.

The data of ground-based and satellite measurements were compared using a traditional method (Fioletov et al., 2008; Visheratin, 2012), calculating the relative deviation (Δ) in percents between the two rows C1 and C2: $\Delta(i) = 100 \times (C1(i) - C2(i))/C1(i)$. The relative deviations for monthly mean values of CO are shown in Fig. 3.

Data of surface measurements of the total content at st. Novolazarevskaya and st. Arrival-Heights are in sufficiently good agreement (Fig. 3a); since 2010, the average deviation $NL\_G_{TC} - AH\_G_{TC}$ is –2.4%. As mentioned above, until 2010, the equipment at st. Novolazarevskaya had been repeatedly improved. A comparison of the data from ground-based stations and measurements by the MOPITT instrument (Figs. 3b, 3c) showed that both the Novolazarevskaya and Arrival-Heights stations are characterized by an excess of satellite data over ground-based data, constituting 19% and 14%, respectively, for average deviations over the entire period of measurements, and 12% and 16%, respectively, for the period after 2010. The growth in the average deviation at st. Arrival-Heights indicates a slight drift of long-term trends of AH_G_{TC} and AH_S_{TC}. Similar results (an excess of satellite data over data at st. Arrival-Heights by 9.5% and a drift of trends) were obtained for data for 2001–2012 by Buchholz et al. (2017). According to (Buchholz et al., 2017), the MOPITT data for the other 13 NDACC stations located at different latitudes of both hemispheres also exceed the data of ground-based stations with correlation coefficients in the range from 0.72 to 0.96. Rakitin et al. (2015) and Sitnov et al. (2017) claim that the coupling between the daily average values of CO total content and ground-based data of stations located in Eurasia is not so close. Depending on the place and conditions of measurements, the correlation coefficients for background conditions vary over a wide range from 0.28 to 0.96 (Rakitin et al., 2015) and from 0.4 to 0.8 (Sitnov et al., 2017).

One should note the following feature: the differences between satellite data and data of st. Novolazarevskaya and st. Arrival-Heights are maximal in December–January, i.e., when the Sun above Antarctica is highest above the horizon. The comparison of MOPITT satellite data with ground-based spectroscopic measurements of CO in Eurasia (Rakitin et al., 2015, Sitnov et al., 2017) also indicated a sufficiently good agreement in summer and a significant difference in winter. According to (Sitnov et al., 2017), this may be caused by an increase in the stability of the lower atmosphere in winter and, therefore, an increase in the fraction of CO content in the surface layer (MOPITT is not sufficiently sensitive to this). According to (Rakitin et al., 2015), the above-mentioned differences are possibly conditioned by more intense convective mixing in summer, which leads to a rise of

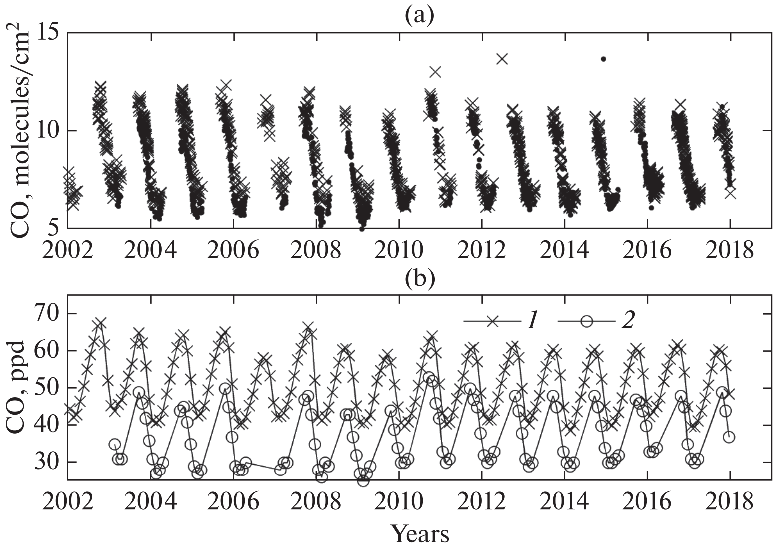

Fig. 2. Temporary variations in CO in the Antarctic atmosphere. (a) Daily average total content of CO (10^{17} molecules/cm^2) at st. Arrival-Heights (*1*) and st. Novolazarevskaya (*2*) and (b) monthly average surface concentrations at st. Syowa (*1*) and average volume concentration in the atmospheric column at st. Novolazarevskaya (*2*).

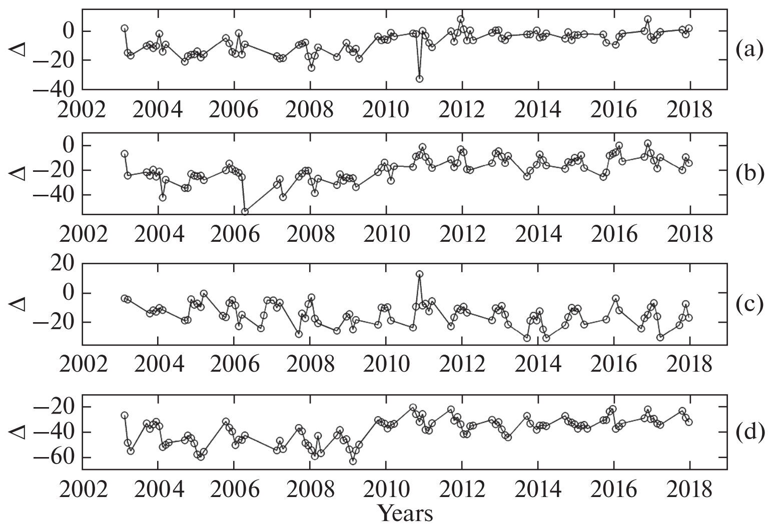

Fig. 3. Relative deviations Δ (%) between different series: (a) $NL\_G_{TC} - AH\_G_{TC}$, (b) $NL\_G_{TC} - NL\_S_{TC}$, (c) $AH\_G_{TC} - AH\_S_{TC}$, and (d) $NL\_G_X - SY\_G$.

gaseous admixtures to the middle troposphere, i.e. to heights where the sensitivity of the satellite spectrometer is satisfactory.

As expected, the measurement data in samples of surface air at st. Syowa exceed the height-average volume concentration of CO at st. Novolazarevskaya. For the entire period of measurements, the average excess is 37%. Since 2010, the average excess is 31%; here, a seasonal dependence of the deviation is also observed, with a range reaching 15–20%; however, in this case, the largest deviations are observed in December–January. Further study is required to find the reasons for the seasonal dependence of deviations.

Both ground-based and satellite data of monthly average values (except for surface concentrations of CO at st. Syowa) have significant gaps in measurements during the polar night. Therefore, these gaps were previously filled for further analysis. To do this, we determined the parameters of the linear trend and annual and semi-annual harmonics for monthly mean values using the iterative least squares method. The general expression approximating the trend, and the annual and semi-annual components of CO fluctuations has the following form:

$$Y = \mathrm{S0} + \mathrm{B\,N}/12 + \mathrm{A12}\sin(\mathrm{P12} + 2\pi\mathrm{N}/12) + \mathrm{A6}\sin(\mathrm{P6} + 2\pi\mathrm{N}/6), \quad (1)$$

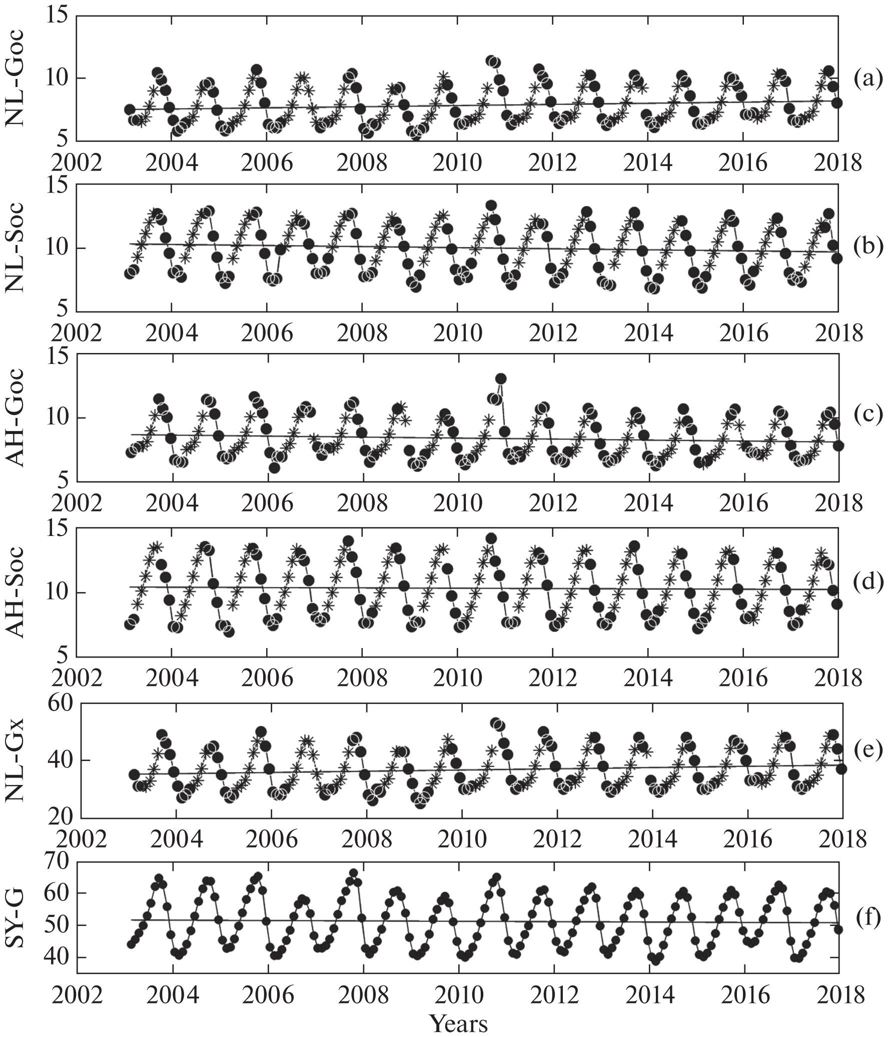

Fig. 4. Time series of monthly average values of CO. Initial data (c) and data interpolated according to (1) (crosses) with the parameters from the Table 1. NL_G_X (e) and SY_G (f) are in ppb; in the remaining panels, the average content of CO is in 10^{17} molecules/cm^2.

where A12 and A6 are the amplitudes, P12 and P6 are the phases of annual and semi-annual harmonics (radians), S0 and B are the trend parameters (per year), N is the month number starting from February 2003. A12, A6, and S0 are in ppb for the height-averaged concentration of CO and 10^{17} molecules/cm^2 for the total content. The fluctuation parameters in bold (see Table 1) are significant at a confidence level of 95%. The trend B is insignificant for all series.

The CO values calculated from (1) were added instead of gaps in the original series. Examples of series with filled gaps are shown in Fig. 4. The tempo-

Table 1. Trend and Fluctuation Parameters

Parameter	SY_Gx	NL_Gx	NL_Goc	NL_Soc	AH_Goc	AH_Soc
S0	**51.7**	**35.2**	**7.5**	**10.3**	**8.7**	**10.4**
B	−0.06	0.21	0.045	−0.04	−0.04	−0.01
A12	**10.2**	**8.80**	**1.87**	**2.50**	**1.94**	**2.85**
P12	**−2.47**	**−2.72**	**−2.77**	**−2.22**	**−2.72**	**−2.21**
A6	**1.80**	2.56	0.58	0.40	0.6	0.39
P6	**−1.60**	−1.24	−1.23	−1.50	−1.31	−0.89

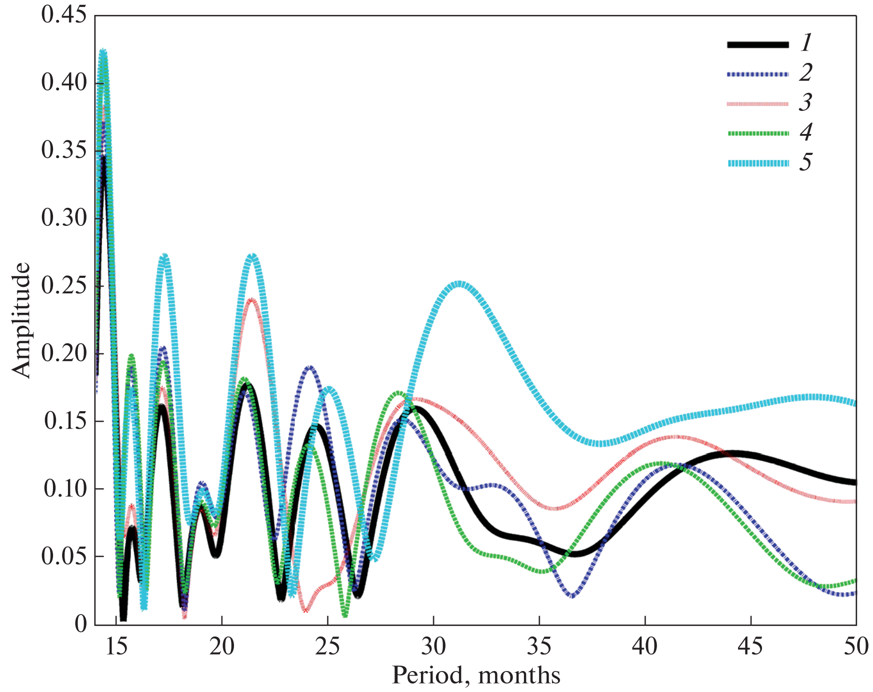

Fig. 5. Amplitude spectra of CO variations (10^{17} molecule/cm^2) of periods of 13–50 months. (*1*) NL_G_{TC}, (*2*) NL_S_{TC}, (*3*) AH_G_{TC}, (*4*) AH_S_{TC}, and (*5*) SY_G (the amplitude in ppb is reduced by 5 times).

ral variability of CO over the given time period has common features. For all the series shown in Fig. 4, the time series have a similar seasonal variation, which is also confirmed by the data given in Table 1: the phases of the annual and semi-annual harmonics for the total contents of CO coincide within the standard deviation. The maximum values of CO are characterized by an increase in 2010. According to (WMO, 2018), an increase in the surface concentration of CO in 2010 was observed in all latitudinal zones of the Earth. The increase in the Northern Hemisphere is caused by large forest fires in Russia in the summer of 2010, while the reasons for the increase in CO in Antarctica are unclear. It should also be noted that in recent years (Figs. 2 and 4) there has been an increase in the minimum concentrations of CO, which is also observed for surface values of CO in the Southern Hemisphere (WMO, 2018).

ANALYSIS OF THE SPECTRAL COMPOSITION OF CO VARIATIONS

To compare the variations of ground-based and satellite data for time periods greater than 12 months, we use spectral analysis methods. The spectral analysis was performed using the Lomb-Scargle modified Fourier transform (Scargle, 1982). This method allows for analysis of series with gaps; however, significant gaps can introduce distortions. Therefore, we used monthly average series of the same length N = 179 months from February 2013 to December 2017 with filled gaps; the linear trend was first removed from data. The results of the spectral analysis are presented in Fig. 5; the parameters of a number of harmonics are given in Table 2. The harmonics with periods of more than 15 months are insignificant even when the annual and semi-annual harmonics are excluded from the series.

Table 2. Parameters of harmonics with periods of more than 14 months

Station	Fluctuation period, months (amplitude, 10^{17} molecules/cm^2).					
NL_Goc	14.4(0.35)	17.1(0.16)	21.2(0.18)	24.4(0.15)	29.1(0.16)	44.1(0.13)
NL_Soc	14.4(0.37)	17.2(0.20)	21.0(0.17)	24.1(0.19)	28.5(0.15)	41.5(0.12)
AH_Goc	14.4(0.38)	17.1(0.17)	21.4(0.24)	–	29.1(0.17)	41.4(0.14)
AH_Soc	14.4(0.42)	17.2(0.19)	21.0(0.18)	24.1(0.13)	28.3(0.17)	40.7(0.12)
SY_G (ppb)	14.4(2.1)	17.2(1.4)	21.4(1.4)	25.0(0.9)	31.2(1.3)	47.9(0.8)

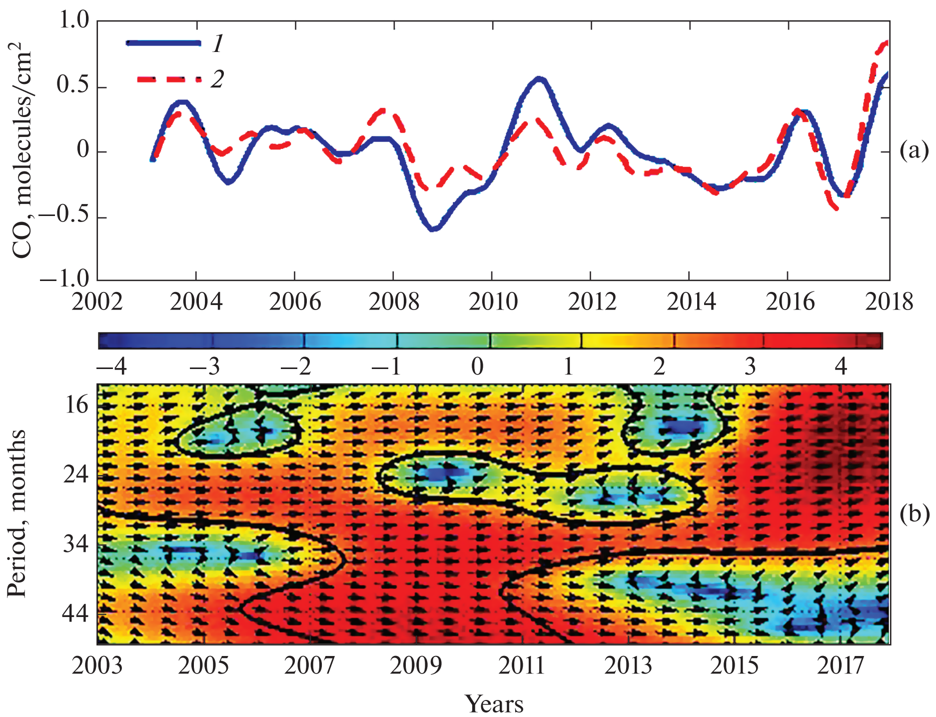

Fig. 6. Comparison of temporal variations in CO for periods of 14–50 months. (a) Composite time series and (b) results of cross-wavelet analysis: (*1*) NL_G_{TC} and (*2*) NL_S_{TC}. The color palette is in relative units, the rightward direction of arrows means synchronicity of vibrations, and the bold line identifies the regions areas with a confidence interval of more than 95%.

Nevertheless, at a qualitative level, the similarity of amplitude spectra of 13–50 months is noteworthy (the length of analyzed series is still insufficient to analyze the harmonics with larger periods).

With some difference in the amplitudes of harmonics probably reflecting the difference in measurement techniques, the periods of fluctuations according to satellite and ground-based measurements are very close. For data of st. Syowa obtained by the method of surface air sampling, the fluctuation periods of more than two years are shifted to the range of long waves. Since the Novolazarevskaya and Arrival-Heights stations are located on opposite coasts of Antarctica, this similarity of the periods of observed fluctuations in the CO field is conditioned by the effect of fluctuations in the general atmospheric circulation on circulation processes over Antarctica (Gruza et al., 2007). The spectral structure of CO variations has similarities with the spectral structure of variations in the total ozone content: the atmosphere of central Eurasia is also characterized by fluctuations with periods of 14.7, 18.2, 21.1, 23.8, 34.1, and 46 months (Visheratin et al., 2006). It seems logical to analyze the variations in the total ozone content over the Antarctic in further studies.

At the same time, the fact that the fluctuation periods from satellite and ground-based measurements of CO coincide (Fig. 5, Table 2) does not necessarily mean the coincidence of fluctuation phases (i.e., synchronicity of fluctuations) since the Fourier analysis yields fluctuation periods averaged over the entire period of analysis. As was shown earlier (Fig. 2, Table 1), the phase relationships in seasonal variations for satellite and ground-based data are the same. To estimate the phase relationships for periods of 14–50 months, we additionally used a composite method (Visheratin, 2012, Visheratin et al., 2017) and a cross-wavelet analysis (Torrence and Compo, 1998). For example, the composite time series containing the sum of fluctuations in the range of 14–50 months are shown for ground-based and satellite data for st. Novolazarevskaya (Fig. 6a).

Figure 6b shows the results of a cross-wavelet analysis. It follows from Fig. 6 that the phase relationships between ground-based and satellite data in long-period fluctuations (less than 50 months) are well consistent according to both composite and wavelet analyzes. A similar pattern is also observed for the data of AH_G_{TC} and AH_S_{TC}. The cross-wavelet analysis of Syowa and Novolazarevskaya station data showed that the differences in the periods of harmonics for more than 3 years observed from spectral analysis data (Fig. 5) appear only in the first half of the analyzed period; after 2010, the fluctuation phases are synchronized for the entire interval of 14–50 months.

CONCLUSIONS

The results of long-term measurements of the total content of CO at st. Novolazarevskaya and the results of comparison with the data of st. Arrival-Heights, st. Syowa, and MOPITT satellite data are presented.

The results of measurements of the total content of CO at st. Novolazarevskaya agree within the measurement error with similar measurements at st. Arrival-Heights. The MOPITT satellite data exceeds the surface measurement data obtained at st. Novolazarevskaya and st. Arrival-Heights by 19% and 14%, respectively; in this case, the relative deviation has a periodic character and is minimal at time periods close to the solstice. The height-averaged relative volume concentrations of CO at st. Novolazarevskaya are 37% less than the surface concentration at st. Syowa. The linear trends are insignificant for all series. There was an increase in the total content of CO in 2010 as well as an increase in the minimum values after 2014.

According to the spectral analysis, the distributions of fluctuation amplitudes and periods of more than 6 months are similar for all the series, regardless of the method and place of measurement. The wavelet and composite analyzes show that the CO variations are phased not only for annual and semi-annual harmonics, but also for fluctuations of 13–50 months.

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