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Assessment of the Influence of Air Invasions from the Upper Troposphere on the CO Total Column Amount in the St. Petersburg Region

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Abstract—The influence of air invasions from the upper troposphere on the CO total column amount is studied on the basis of spectroscopic measurements of the CO total column amount, backward trajectories of air-mass motions (the HYSPLIT model), and meteorological data. It is shown that the observed invasions of substratospheric and upper-troposphere air masses determine the minimum CO total column amount in late January–late March. The invasion of air masses from the upper troposphere can result in a decrease in the CO total column amount to 30% (of its mean values). Using January 31, 2000, as an example, we show the influence of the invasion of Arctic air masses from the upper troposphere on the CO total column amount in the St. Petersburg region: the results of measurements of the CO total column amount in the St. Petersburg region and at the Kiruna polar station (NDACC) are in agreement to within 1% if the vertical transport of air masses is taken into account. Thus, for a correct combined analysis of measurement data on the CO total column amount for different observation stations, it is necessary to use data on air-mass trajectories.

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INTRODUCTION

Carbon oxide (CO) is a chemically active component that affects the three most important constituents of the Earth's atmosphere: tropospheric ozone (O_3), methane (CH_4), and carbon dioxide (CO_2). The hydroxyl radical (OH) is the main sink of carbon oxide and methane in the troposphere; therefore, carbon oxide simultaneously controls the CH_4 content and the oxidation power of the troposphere (OH). The oxidation of CO can result in the formation or destruction of tropospheric ozone, depending on the content of nitric oxides. For example, in the presence of a sufficiently high content of nitric oxide (NO), the reactions of CO oxidation result in the formation of tropospheric ozone [1, 2]. Concentrations sufficient for the formation of O_3 can be reached in industrially developed regions in the middle latitudes of the Northern Hemisphere and in the upper troposphere in the tropical latitudes (where a significant amount of NO is formed due to lightning). Under low concentrations of nitric oxide in the troposphere (which are more characteristic of the Southern Hemisphere), another chain of reactions occurs with the participation of CO, which leads to the destruction of tropospheric O_3 [1]. In both cases, the final product of CO oxidation is carbon dioxide (the most important greenhouse gas).

Atmospheric dynamic processes have a significant influence on both CO local concentrations and CO total column amounts in the atmosphere. In many cases, the origin of air masses provides an explanation for observed variations in the gas composition of the atmosphere [3–8]. Thus, for example, the anomalously high values of the CO total column amount in the atmosphere over St. Petersburg in late summer–early fall are due to the arrival of CO enriched air masses from the regions of major forest fires. Under such conditions, the processes of air-mass transformation especially strongly affect the CO total column amount in the atmosphere [9]. A significant increase (~25% of annual means) in the atmospheric CO amount during the cold seasons is due to the high intensity of anthropogenic CO sources and low concentrations of the hydroxyl radical in the troposphere (the main sink of CO). When transported eastward or northeastward, air masses accumulating emissions from land sources (for example, St. Petersburg) can result in an additional increase in the CO total column amount by 20–30% (of the means characteristic of these seasons). The background values of CO are usually recorded when Atlantic air masses arrive [9].

Along with advection, the vertical transport of air masses also affects the gas content in the atmosphere [10]. The cells of atmospheric circulation and the related vertical motions of air masses result in significant variations in the concentration of atmospheric trace gases [6]. The periods during which upward air flows transport pollutants from the lower troposphere to the higher layers are clearly observed at high-altitude stations [11]. The cases of increased O_3 concentration due to the invasion of stratospheric and substratospheric air masses in the lower troposphere have been recorded at the Kislovodsk Scientific Station of the Institute of Atmospheric Physics of the Russian Academy of Sciences (~2070 m above sea level, 43°44′ N, 42°43′ E) [12].

STATEMENT OF THE PROBLEM

The main objective of this study is to assess the effect of air invasion from the upper troposphere on variations in the CO total column amount for St. Petersburg. This study is based on spectroscopic measurements of the CO total column amount [13, 14], analysis of air-mass trajectories (the HYSPLIT model) [15], and meteorological data [16, 17]. A combined analysis of a set of daily means of the CO content for the St. Petersburg region (1997–2005) and air-mass backward trajectories has made it possible to select the periods (seven) that illustrate the effect of the vertical transport of air masses on the CO total column amount. In most cases, these are downward air flows observed mainly during cold seasons. Three of the seven cases were recorded in late January–February: January 31, 2000; February 21, 2003; and February 21, 2004. Note that intense invasions of substratospheric and upper-troposphere air masses are a relatively rare event. During these periods, one can expect a decrease in the CO concentration in the lower troposphere and, as a consequence, a decrease in the measured total column amounts of CO.

Optical (spectroscopic) measurements of the CO total column amount require certain weather conditions: clear skies or breaks in clouds. Therefore, the choice of the most illustrative examples that demonstrate the effect of a concrete dynamic process on the CO total column amount is more difficult than in the case of continuous measurements (for example, local gas-chromatographic measurements) [3, 11, 12]. It is necessary that, during the period of air-mass replacement, such measurements be taken over several days. Specific days of measurements during this period must be distributed so that it is possible to observe the effect of the considered atmospheric dynamic process on the total column amount of the gas.

RESULTS AND DISCUSSION

Let us consider in more detail the results of measurements of the CO total column amount in the atmosphere over the St. Petersburg region, which were obtained from late January to late March in 1997–2005 (see Fig. 1). This two-month period corresponds to the maximum of the annual cycle of the CO total column amount [13, 18] and is characterized by a relatively slight (as compared to other months) interannual variability of the means of the CO total column amount. This is clearly demonstrated by the annual cycles of the CO total column amount for 1997–2004 (see Fig. 2) [18]. The only exception is 1999, when, in January–February, anomalously high values of the CO total column amount were recorded [13]. Over 9 yr, from late January to late March, the measurements were taken over 114 days. For the indicated period, the mean value of the CO total column amount is $w_m = 0.290 \times 10^{19}$ mol/cm^2 and the standard deviation $\sigma = 0.025 \times 10^{19}$ mol/cm^2. The minimum $(0.221 \pm 0.003) \times 10^{19}$ mol/cm^2 and maximum $(0.37 \pm 0.01) \times 10^{19}$ mol/cm^2 values (daily means) were recorded on February 21, 2004, and on February 27, 1999, respectively. The number of high values that go beyond the corridors $w_m \pm \sigma$ and $w_m \pm 2\sigma$ significantly exceeds that of low values (see Fig. 1). This is related to the fact that the measurements were performed at a land station, so that the observed CO total column amount depends on the intensity of CO sources.

Over the 9 yr of measurements, the three lowest daily means (for late January–late March) were recorded when air masses arrived from the upper troposphere: on January 31, 2000; February 21, 2003; and February 21, 2004 (see Fig. 1). The distinctive feature of these three days is a rapid decrease in the CO total column amount as compared to the neighboring days (by 14% for January 31, 2000, and February 21, 2003, and by 30% for February 21, 2004). Without analysis of supplementary meteorological data and air-mass trajectories, such cases can be treated as blunders in taking measurements and in processing the data. The most significant decrease in the CO total column amount was observed on February 21, 2004, and was due to the invasion of upper-troposphere air masses formed over the south Atlantic. It is of interest to analyze the three cases; however, we think that the period January 29–February 1, 2000, is the best for the following detailed analysis.

Spectroscopic measurements of the total column amounts of CO and H_2O in the atmosphere over the St. Petersburg region were taken on January 29 and 31 and on February 1. The results of these measurements are given in Table 1. The total column amount of water vapor for January 30 was obtained from the results of meteorological sounding in the town of Voeikovo (marked "ms" in the table). It follows from Table 1 that, in the St. Petersburg region, significant decreases in the total column amounts of CO and water vapor were observed on January 31. The differences in the results of CO measurements for January 29–31 and February 1–January 31 are 14 and 13% of the mean total column amount, respectively. These values are

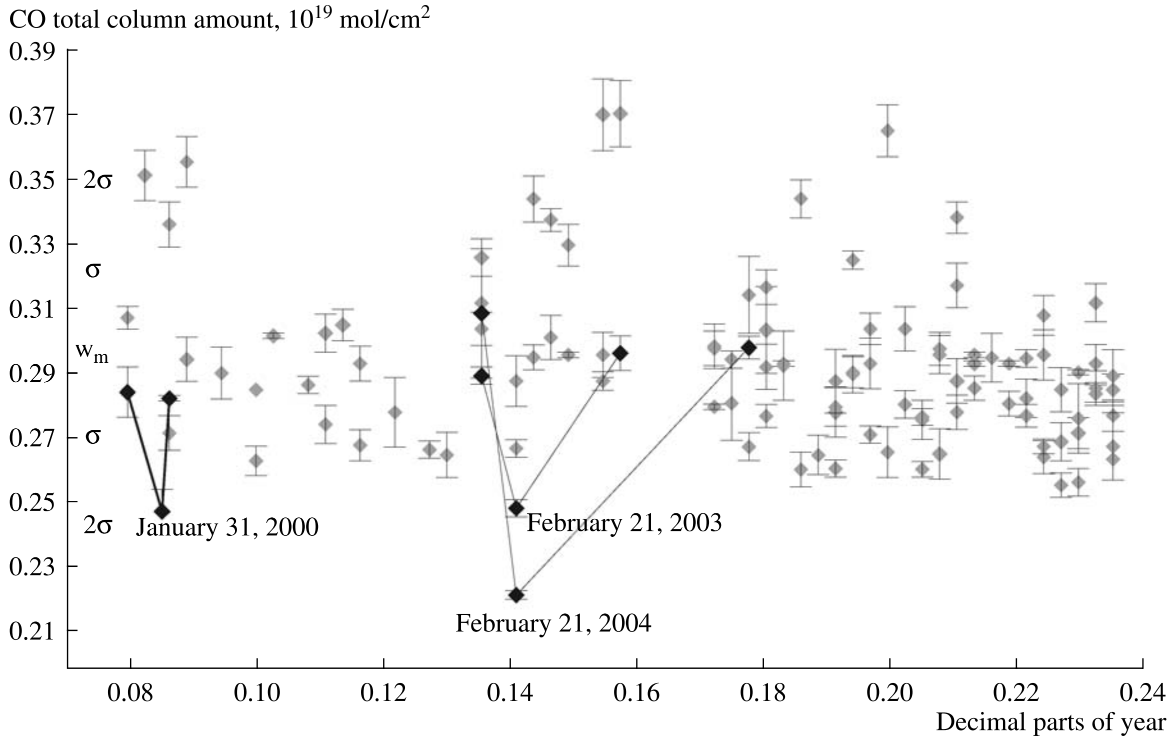

Fig. 1. Measured CO total column amounts for the St. Petersburg region (late January–late March, 1997–2005).

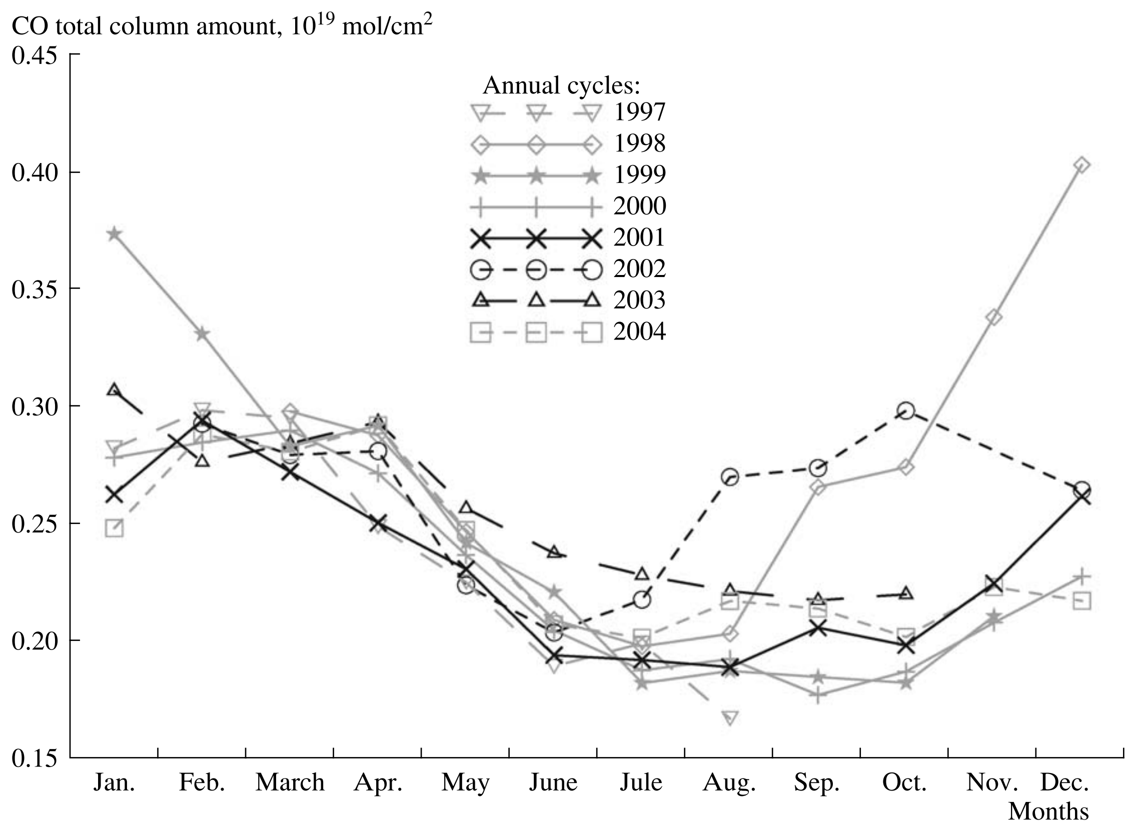

Fig. 2. Annual cycles of the CO total column amount for 1997–2004.

Table 1. CO and H_2O total column amounts for the St. Petersburg (S) and Kiruna (K) regions

Date, station	Vertical motions of air masses	CO total column amount, 10^{19} mol/cm^2	H_2O total column amount, g/cm^2
January 28, 2000 (K)	–	0.2331 ± 0.0005	
January 29, 2000 (S)	–	0.284 ± 0.005	0.51
January 30, 2000 (S)	–	–	0.70 (ms)
January 31, 2000 (S)	downward airflows	0.244 ± 0.004	0.20
February 1, 2000 (S)	–	0.282 ± 0.005	0.36
February 2, 2000 (K)	–	0.2453 ± 0.0005	

comparable to the amplitude of seasonal variations in atmospheric CO (~25%).

Figure 3 gives the 5-day air-mass backward trajectories (HYSPLIT) to the St. Petersburg region (Petrodvorets), 59°88′ N, 29°83′ E) for January 31, 2000. The horizontal and vertical projections of the air-mass trajectories are shown at the top and bottom of Fig. 3, respectively. The end points of the trajectories (Petrodvorets, 59°88′ N, 29°83′ E) in the graphs are marked by asterisks. The downward air motions are clearly seen at the bottom of Fig. 3: on January 31, 2000, Arctic air masses arrived from the upper troposphere. No intense vertical air motions were observed on the neighboring days—January 29 and 30 and February 1; this is clearly seen in Fig. 4, which gives the results of calculating the air backward trajectories for January 29, 2000.

Arctic air masses usually invade the middle latitudes in the rear of cyclones (behind cold fronts). Let us outline the synoptic situation of the period under consideration and dwell upon some features of interest. To this end, the AT 500 maps of absolute baric topography were analyzed [17], which follow the motion of the north Atlantic cyclone through Scandinavia and farther northeastward. St. Petersburg proved to be under its influence on January 29–31. A decrease in pressure, a rise of air temperature to positive values (1.4°C), overcast skies, and precipitation (see Table 2, which gives meteorological data obtained at the station in the town of Voeikovo [16]) suggest that, on January 30, St. Petersburg was in the warm sector of the cyclone. In the nighttime from January 30 to 31, the passage of the cold front (accompanied by heavy precipitation in the form of a snow shower, a decrease in the air temperature, and a change in the wind direction) was observed, which was followed by an Arctic air invasion accompanied by clearing of the sky, which was observed even at 15:00 (see Table 2) and which made it possible to take spectroscopic measurements. Figure 5 presents a family of maps plotted for 15:00 (local time), January 31, 2000 [17]. The regions of low temperatures and humidity in the form of "tongues," which are located in the rear of the cyclone and result from the invasion of cold dry Arctic air masses, are clearly seen on the AT 500 and 850 maps (air temperature) and the AT 700 map (relative humidity). According to ground-based meteorological data, the effect of downward dry cold airflows was noticeable even since 06:00. At 12:00, the skies started clearing, and, at 15:00, relative humidity reached its minimum –54% (although the air temperature contin-

Table 2. Meteorological observations on January 31, 2000, at the station in the town of Voeikovo (St. Petersburg, No. 26063, 59°58′ N, 30°18′ E)

Local time	Air temperature, °C	Pressure at sea level, hPa	Relative humidity, %	Dew-point temperature, °C	Wind direction and velocity, ° and m/s	Cloudiness, amount	Altitude and form of the lower boundary of cloudiness	Phenomena (KN-01 code)
00:00	+2.0	974	97	+1.6	200, 3	10	450 m (Ns)	(02) Cloudiness without changes
03:00	0.0	978	97	–0.4	261, 5	10	800 m (Ns)	(26) Snow shower (previous hour)
06:00	–1.0	983	74	–4.6	280, 6	10	1250 m (Ac)	(85) Moderate snow shower
09:00	–3.0	986	75	–6.4	270, 4	9	800 m (As)	(02) Cloudiness without changes
12:00	–3.0	988	64	–8.2	279, 5	3	3000 m (Ci)	
15:00	–3.0	990	54	–10.1	311, 3	1	3000 m (Ci)	
18:00	–5.0	994	63	–10.3	270, 1	0	3000 m (Ci)	
21:00	–6.0	996	79	–8.7	261, 2	0	3000 m (Ci)	

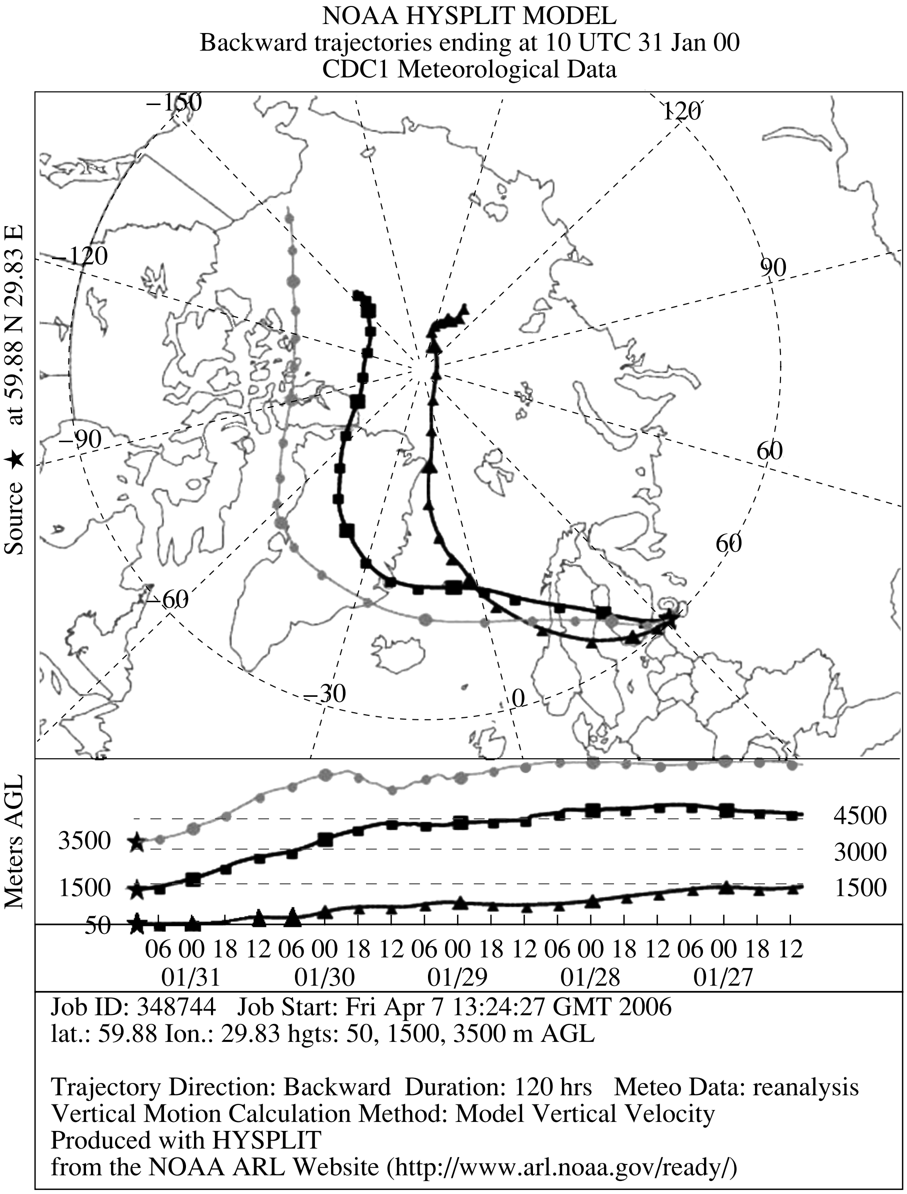

Fig. 3. Air-mass backward trajectories for January 31, 2000.

ued to decrease until 18:00) [16]. Note that, on January 31, the spectroscopic measurements of the CO total column amount were performed from 13:00 to 16:00, when St. Petersburg was in the center of the Arctic tongue.

Let us estimate the variations in the CO total column amount that were bound to occur owing to the arrival of air masses from Arctic tropospheric layers higher than 3 km (when air with a lower concentration of CO from the upper layers replaces the lower-troposphere continental air rich in carbon oxide). To this end, we use the NDACC data on the CO total column amount [14, 19]. The two of the NDACC stations—Ny-Alesund and Kiruna—are of interest. These stations are located northwest of St. Petersburg in the regions from which air masses entered the city on January 31, 2000. Only at the Kiruna station (67°84′ N, 20°41′ E, 419 m above sea level) were the measurements taken during the period closest to that under consideration (January 29–February 1, 2000): data on the CO total column amount are available for January 28 and February 2, 2000. The measurement results for these days are given in Table 1, and the mean value of the CO total column amount is 0.239×10^{19} mol/cm^2. The vertical distribution of CO taken from the AFGL86 model [20] and brought to the value of its total column amount for the Kiruna station (0.239×10^{19} mol/cm^2) is shown in Fig. 6 (no data on the vertical distribution of the CO concentration for the selected period for the St. Petersburg and Kiruna

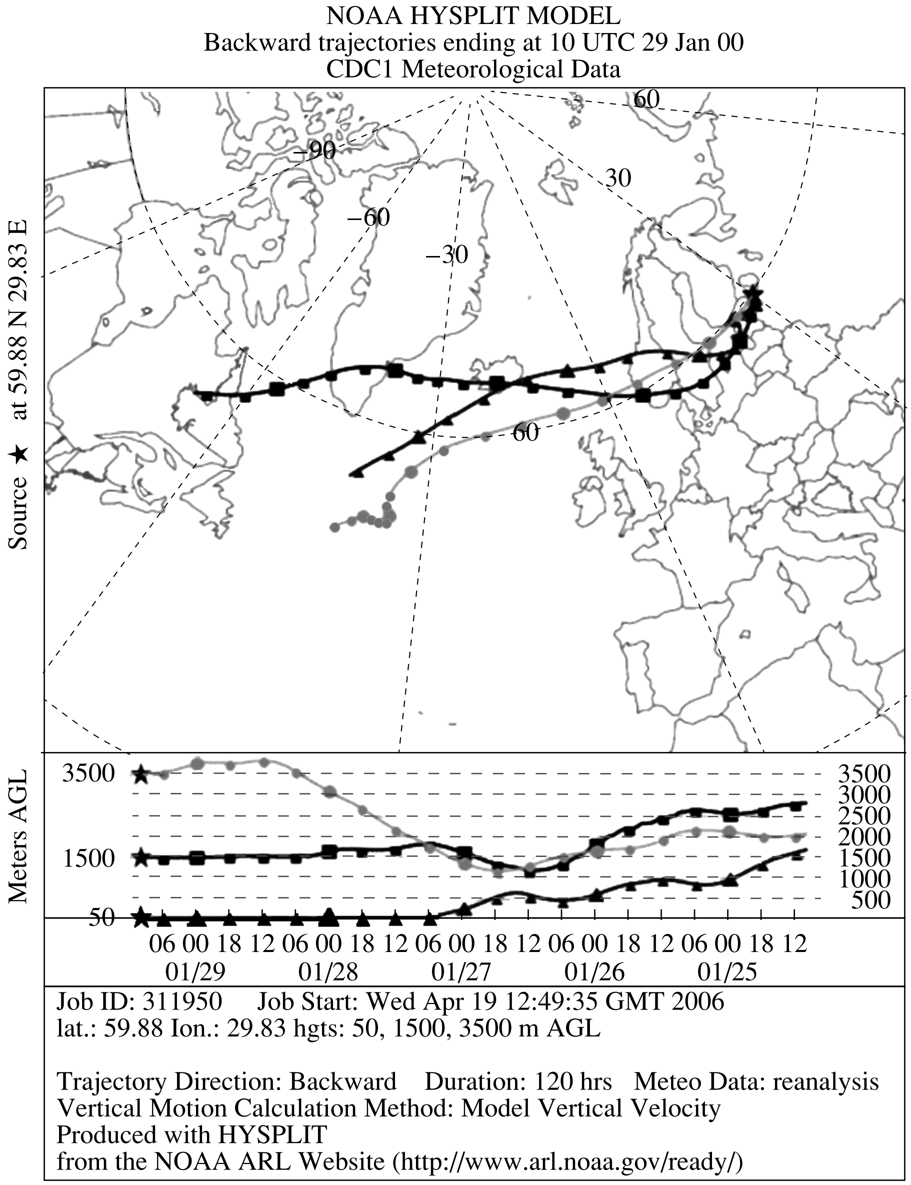

Fig. 4. Air-mass backward trajectories for January 29, 2000.

regions are available: measurements of the CO vertical distribution in the atmosphere are still of sporadic character or on close access). If the height from which air masses that entered the St. Petersburg region on January 31, 2000, is determined with the aid of backward trajectories, the local profile of the CO concentration can easily be constructed for the St. Petersburg region. The value of the CO concentration (for January 31 in the St. Petersburg region), for example, at a height of 1 km, will be determined by Arctic air masses that arrived from a height of approximately 3.5 km above sea level (see Fig. 6). Let us similarly transform the values of the CO concentrations for all heights in the troposphere. In Fig. 6, the arrows show schematically how the vertical distribution of the CO concentration was formed for St. Petersburg (from the profile for Kiruna). Thus, the CO concentration profile for St. Petersburg is obtained by transforming the mean profile [20] on the basis of data on vertical air motions for the region and time of interest.

The CO total column amount calculated for the new specific profile (for St. Petersburg) is 0.242×10^{19} mol/cm^2 and is a “theoretical” estimate for the CO total column amount for January 31, 2000, which differs from the measurement result for January 31 (0.244×10^{19} mol/cm^2) by ~1%. The agreement between the results can be considered very good, and this justifies the validity of the proposed method. If we were to use a line of reasoning identical to that in the previous section but start with the mean total column

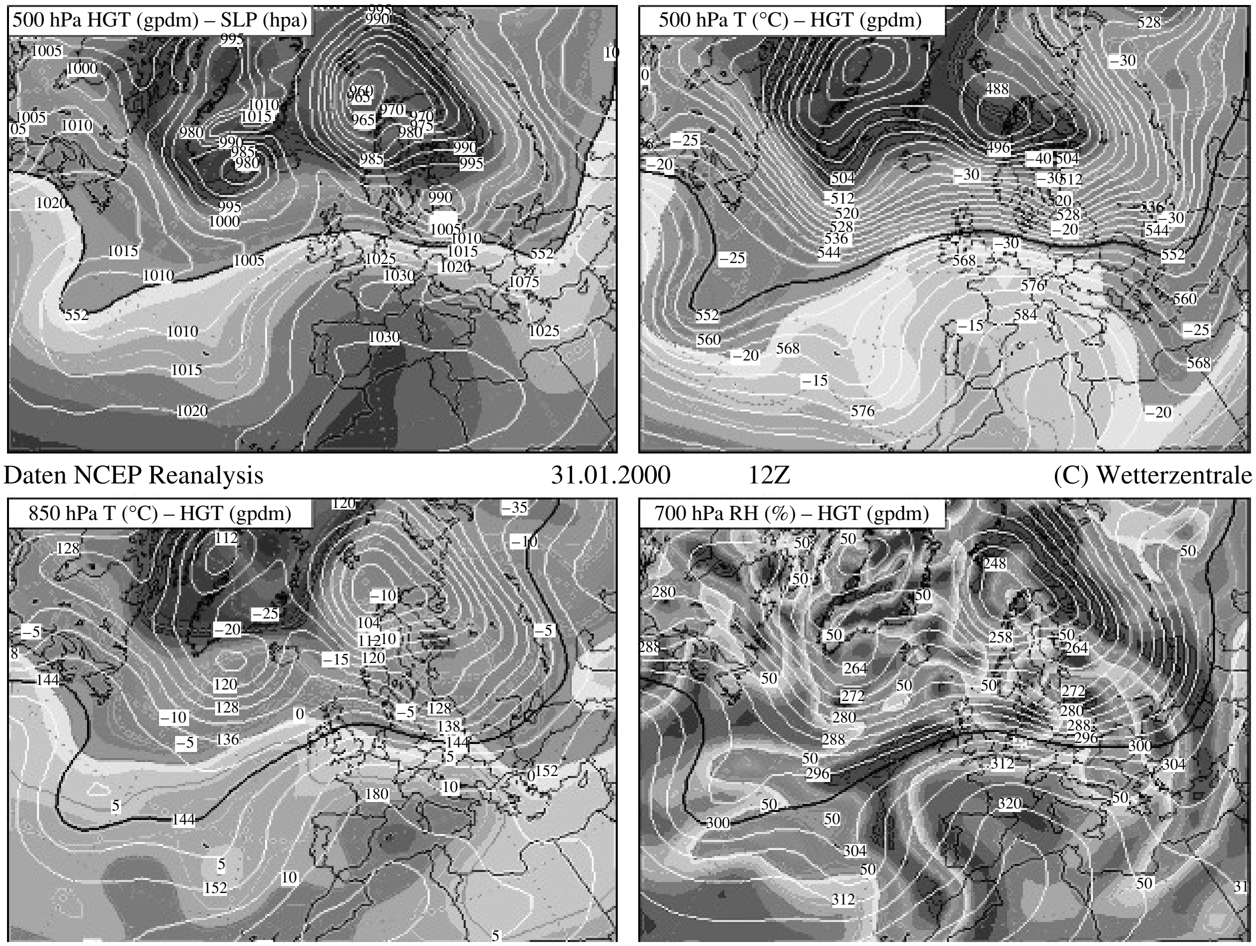

Fig. 5. Family of maps (AT 500 with pressure at sea level, AT 500 and 850 with air temperature, and AT 700 with relative humidity) for 15:00 (local time), January 31, 2000 [17].

amount of CO for St. Petersburg (0.283×10^{19} mol/cm^2) instead of its total column amount for Kiruna, the "theoretical estimate" would be ~0.269×10^{19} mol/cm^2, which is 10% higher than the experimental value of the CO total column amount (0.244×10^{19} mol/cm^2).

Thus, when air masses arrive from the upper troposphere, the CO total column amount depends to a great extent on the air basin in which these air masses were formed. In the cold seasons, in the Arctic air, the CO content in tropospheric layers higher than 3.5 km is approximately 8% less than that for the St. Petersburg region. The low total column amount of CO for January 31, 2000, is caused not only by the descent of air masses from the upper layers but also by their origin (in this case, by their Arctic origin). This should be taken into account to obtain correct results in comparing data on the CO total column amount obtained at different stations (for example, St. Petersburg and Kiruna).

CONCLUSIONS

Based on the results of spectroscopic measurements, simulated air backward trajectories, and meteorological data, the study of the influence of air invasions from the upper troposphere on the CO total column amount in the atmosphere over the St. Petersburg region has shown the following.

1. For late January–late March, the minimum total column amounts of CO recorded in the course of 10-yr (1995–2005) measurements were observed during invasions of air masses from the upper troposphere (January 31, 2000; February 21, 2003; and February 21, 2004).

2. The invasion of air masses from the upper troposphere in the middle latitudes can cause a significant decrease in the CO total column amount (to 30% of its mean values). In the case of the observed downward airflow, variations in the CO total column amount depend not only on the heights from which this airflow arrives but also on the air basin in which this air mass is formed. These variations can reach about 15% for the invasion of Arctic air masses (from the upper tro-

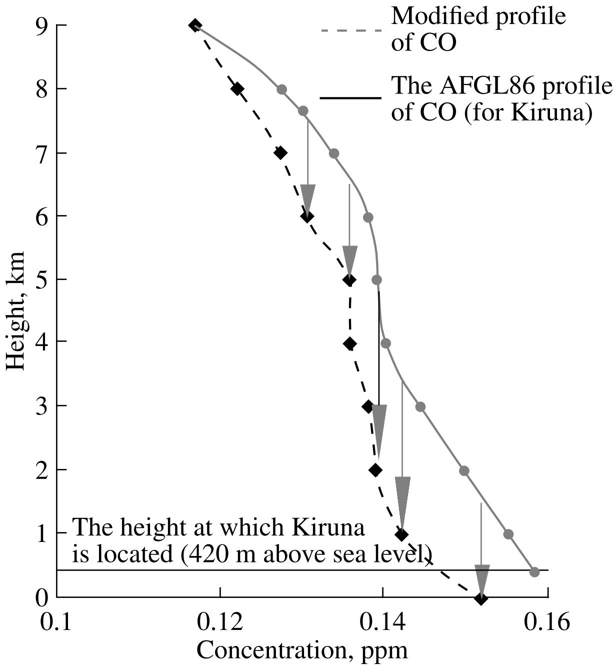

Fig. 6. Profiles of the CO concentration in the troposphere. The solid line denotes the AFGL86 profile corresponding to the CO total column amount for the Kiruna station; the dashed line corresponds to the profile for St. Petersburg; and the arrows denote downward airflows that form the vertical distribution of the CO concentration in the St. Petersburg region.

posphere) and 30% for the invasion of Atlantic air masses.

3. Analysis of the arrival of Arctic air masses on January 31, 2000, has shown that the results of measurements of the CO total column amount in the St. Petersburg region and at the Kiruna station (NDACC) are consistent to within 1% if the vertical transport of air masses is taken into account. In tropospheric layers higher than 3.5 km, the CO total column amount for the Arctic air basin (during the cold seasons) is 8% lower than that for the St. Petersburg region.

4. In a comparative study and combined analysis of the results of measuring the CO total column amount at different observation stations, it is necessary to take into account information on the origin of air masses. Consideration for the vertical motions of air masses and advection processes makes it possible to explain discrepancies observed sometimes in the values of the CO total column amount.

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