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Study of the Factors Determining Anomalous Variability of Carbon Dioxide Total Column Amount over St. Petersburg

M. V. Makarova, V. S. Kostsov, and A. V. Poberovskii

St. Petersburg State University, ul. Ul'yanovskaya 1, Petrodvorets, St. Petersburg, 198504 Russia

e-mail: zaits@troll.phys.spbu.ru

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Abstract—Results of spectroscopic measurements of the carbon dioxide total column amount near St. Petersburg during forest fires in the period from August to September 2002 are analyzed. The HYSPLIT model is used to calculate air-mass trajectories and CO distribution on a mesoscale in this period. The HYSPLIT model simulations and measurements of carbon dioxide total column amount yield an estimate of the specific intensity of CO emission in a Pskov forest fire on August 28–September 8, 2002, equal to 0.17–0.26 kg m^2. This estimate can be used for an estimation of the integral CO emission from fires in northwestern Russian forests and for model simulations of atmospheric CO concentration fields. The estimate of the CO emission from forest fires that is obtained from ground-based measurements can also be made on the basis of satellite measurements if they contain information on CO in the lower tropospheric layers (0 to 2 km).

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1. INTRODUCTION

Carbon monoxide (CO) is an important gas constituent controlling the oxidizing ability of the troposphere (through the reaction with OH). CO is an indicator of anthropogenic air pollution and influences the ozone concentration in the troposphere [1]. Because of the important role that carbon monoxide plays in tropospheric chemistry, its study remains an urgent problem. MOPITT [2], SCIAMACHY [3], and other programs are in progress, in which the emphasis is on CO measurements in the troposphere. In 2005, a special working group was created to coordinate experimental and theoretical investigations of atmospheric carbon monoxide [4]. Measurements of CO are being continued at the international networks of ground-based stations, the NOAA and NDACC. The NOAA network has been conducting local gas-chromatographic measurements of surface concentrations of different gases, including CO, for more than 15 years [5]. The Network for the Detection of Atmospheric Composition Change (NDACC, the former NDSC) began measurements of characteristics of the aerosol and gas composition in the stratosphere and troposphere in 1991. For retrieval of CO total column amount and elements of the vertical distribution of CO concentration, high-resolution spectra recorded with a Fourier spectrometer are used at the NDACC stations [6]. In Russia, spectroscopic measurements of CO total column amount in the atmosphere are conducted at the Institute of Physics of St. Petersburg State University [7], the Institute of Atmospheric Physics of the Russian Academy of Sciences [8], and the stitute of Experimental Meteorology [9]. A program of complex research is being conducted on the gas and aerosol composition of the atmosphere with the use of the mobile TROICA (Transcontinental Observations into the Chemistry of the Atmosphere) observatory [10].

Experimental information is used for analysis of the spatial and temporal variability of CO in the atmosphere and is more or less assimilated into atmospheric models of different spatial scale and resolution [4]. A combined analysis of the experimental data and modeling results makes it possible to discern the most significant factors that determine the variability of carbon monoxide in the atmosphere and to evaluate the validity of transport-photochemical models and of their forecasts. This is especially important because the modern models supplemented with network and satellite measurements have approached the possibility of a real-time forecast of the chemical air composition on a regional scale (e.g., MOSAGE, BOLCHEM, CHIMERE, and other models) [11]. The inverse-modeling methods, which are based on the minimization of mismatches between measured and simulated CO values, make it possible to determine the intensities of various sources of carbon monoxide more accurately. An example is the coupled use of the MOZART model and the results of the MOPITT satellite experiment for 2000–2003 [12].

The paper is focused on analysis of the variability of carbon dioxide total column amount near St. Petersburg during forest fires in August 2002. The HYSPLIT model is used to calculate air-mass trajectories and CO dispersion in the atmosphere from forest fires in the Pskov and Novgorod regions. As a result of simulation of the evolution of a CO plume from sources of

Table 1. Spectroscopic measurements of CO total column amount (Q) near St. Petersburg from August 28 to September 9, 2007

Date	Q, $\times 10^{19}$ molecules/cm^2
28.08.2002	0.393 ± 0.008
02.09.2002	0.217 ± 0.003
03.09.2002	0.258 ± 0.01
04.09.2002	0.198 ± 0.008
06.09.2002	0.63 ± 0.05
08.09.2002	0.338 ± 0.01
09.09.2002	0.331 ± 0.01

specified location and area with consideration for CO total column amount measurements directly in the plume, an estimate has been made for the intensity of CO emission from the source under consideration (in the given case, a Pskov forest fire on September 4–6, 2002). The possibility of applying this approach to the evaluation of CO emissions from forest fires if satellite measurements of lower tropospheric CO are available is discussed.

2. EXPERIMENTAL DATA

Spectroscopic measurements of carbon dioxide total column amount (which will be further denoted by Q) have been continually conducted at the Institute of Physics, St. Petersburg State University, since 1995 [7]. The accumulated data set makes it possible to perform analysis of the variability of this atmospheric constituent on different temporal scales, from the diurnal cycle to the trend [7]. Carbon monoxide exhibits large seasonal variations with a maximum in February to March, which have a range of about 25% of the mean Q. Variations in Q on a scale of a few days are usually no more than 5%; however, much larger variations were recorded on some occasions. The error in daily means of the total column amount is 1–4% [7]. Analysis of such cases permits the detection of the most important factors responsible for the observed CO variations. Note that the variations discussed in the paper usually last two to five days, a period that is far shorter than the lifetime of CO in the atmosphere [1]. Therefore, chemical processes in the atmosphere are unlikely to be the cause of these changes.

3. HYSPLIT MODEL

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model is a complete numerical system created by a joint effort of NOAA and Australia's Bureau of Meteorology [13]. HYSPLIT computes simple air-parcel trajectories and simulates the dispersion and deposition of a pollutant particle in the atmosphere (with additional dispersion modules). The HYSPLIT model can be run both via the Internet (through the READY system at the site http://www.arl.noaa.gov.ready) or on a PC (by downloading the HYSPLIT file and meteorological databases from the same site). The web version of the model has been configured with some limitations to avoid computational saturation of the server.

4. ESTIMATION OF THE EMISSION INTENSITY OF CARBON MONOXIDE FROM FOREST FIRES

Analysis of ground-based spectroscopic measurements of CO total column amount, which were conducted under specific atmospheric conditions and supplemented with the modeling results (HYSPLIT) and with necessary satellite and meteorological information, makes it possible in some cases not only to discern the factors that have affected the measurement results but also to obtain quantitative estimates of these factors. The period of vigorous forest fires of late August to early September 2002 with intense CO emissions into the atmosphere, which is treated in the paper as a remarkable example of the relation of the anomalous variability of carbon dioxide total column amount to advective processes, provides an opportunity to evaluate the intensity of the CO sources operating at that time.

Consider in detail the changes in carbon dioxide total column amount Q that were observed near St. Petersburg from August 28 to September 9, 2002. The period was chosen because of the presence of strong sources of carbon monoxide—severe forest fires in Russia (in Siberia as well as in European Russia). Under such conditions, advection would significantly affect the observed Q values. In August to September 2002, they were increased because of a large number of forest fires across Russia (in both Europe and Siberia). Spectroscopic CO measurements in late August and early September 2002 span one day of observation (September 6) directly in a smoke plume from forest fires in the Leningrad, Pskov, and Novgorod oblasts. Knowledge of the location of forest fires with the HYSPLIT dispersion module permits the simulation of the spread of a pollutant plume in the atmosphere. Comparison of the modeling results with measurements of the total carbon dioxide in a plume provides an estimate for the intensity of CO emission from forest fires.

Ground-based spectroscopic measurements were conducted in a selected period for seven days: August 28 and September 2, 3, 4, 6, 8, and 9; their results are given in Table 1. A record value $(0.63 \pm 0.05) \times 10^{19}$ molecules/cm^2 (in the entire period of spectroscopic measurements in the area of St. Petersburg) was reported on September 6, 2002. The total column amount of CO on August 28 and September 8 was about half as large as the record value. Even much lower values of the total column amount were mea-

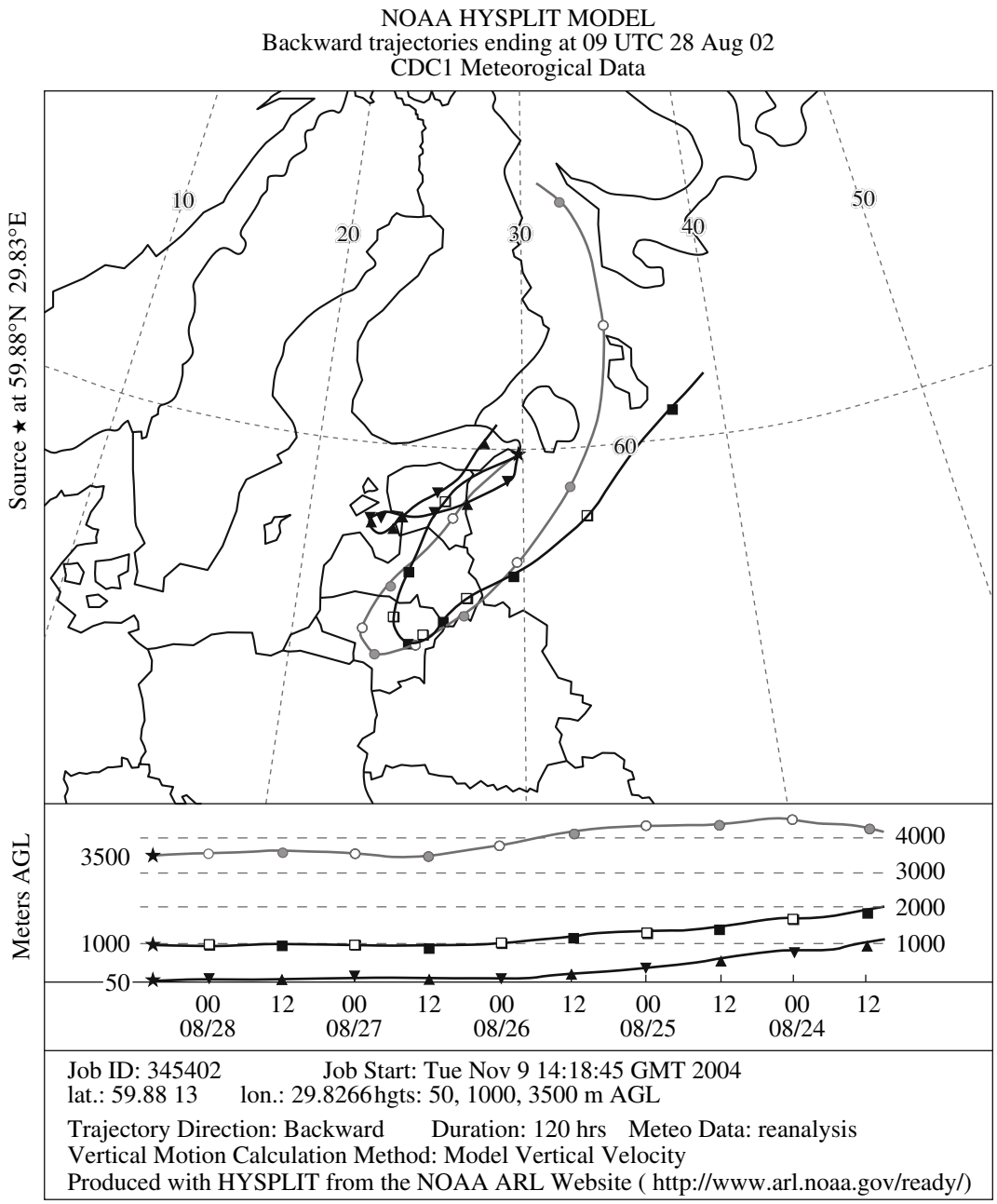

Fig. 1. Backward trajectories of air-mass motion, August 28, 2002.

sured on September 2, 3, and 4. However, they cannot be called the background values (in comparison with, for example, Q measurements during the same period of 2000 or 2001).

The HYSPLIT model was used to obtain five-day backward trajectories of air-mass motion for all of the selected days with measurements at the site where spectroscopic measurements of Q were made (Petrodvorets, St. Petersburg, 59.88°N, 29.83°E) at altitudes of 50, 1500, and 3000 m above sea level. Hereinafter, the term "backward trajectory" means a curve that is described by a moving elementary air parcel before its arrival at a given geographic point. For brevity, backward trajectories of air-mass motion will be further reduced to backward trajectories.

Changes in the direction of air-mass motion that induced such a significant change in Q occurred twice in the period from August 28 to September 8: between August 29 and September 1 and on September 5. The air that reached St. Petersburg on August 28 had accumulated biomass burning products from the areas affected by forest fires in Europe. This is most clearly demonstrated by backward trajectories (Fig. 1), which have the shape of a loop located over the continent. As

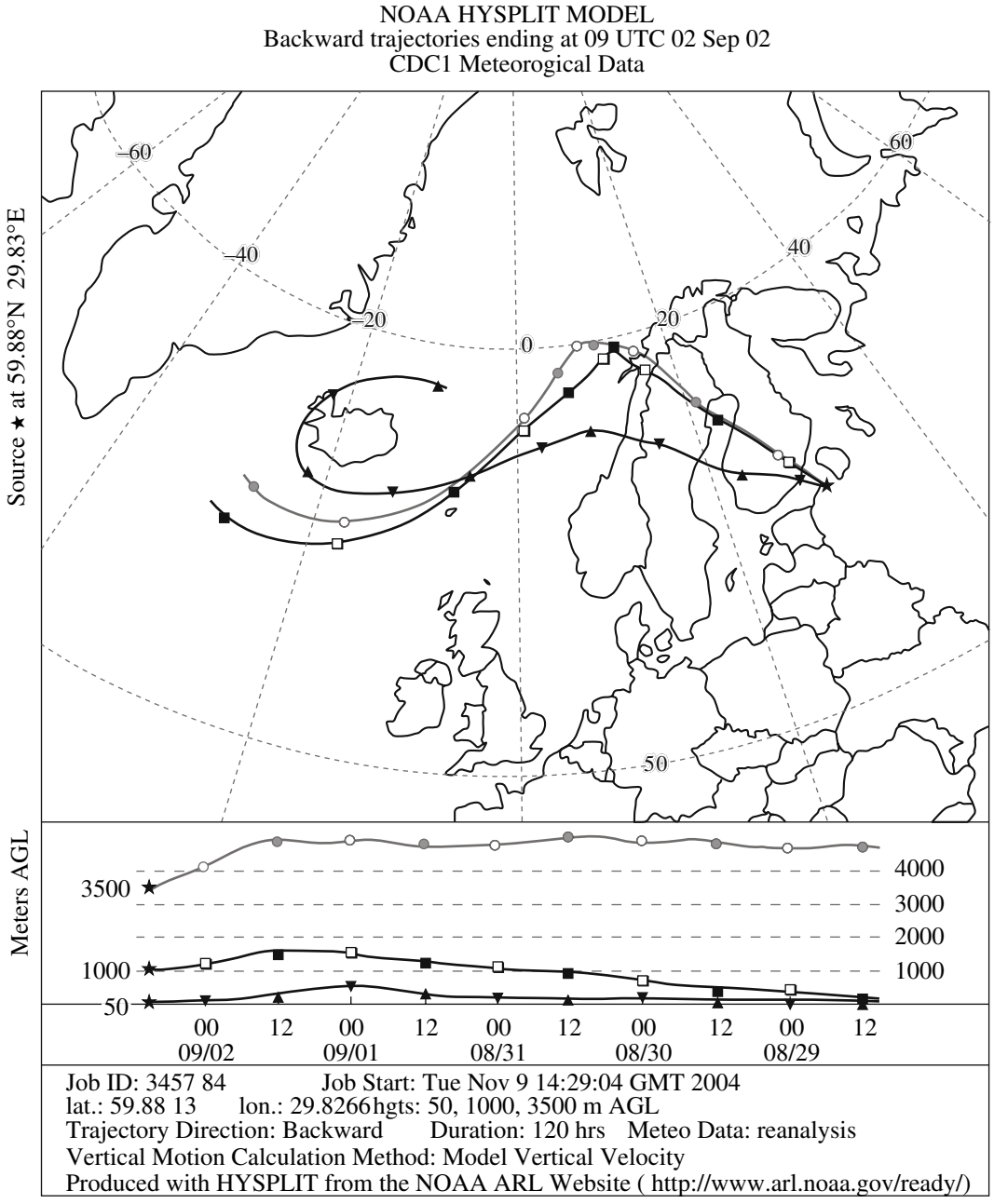

Fig. 2. Backward trajectories of air-mass motion, September 2, 2002.

a consequence, the CO total column amount values on that day (Table 1) are the highest ever recorded over the entire period of measurements: 36% above the annual maximum, which is a February mean of 0.29×10^{19} molecules/cm^2.

The change in the direction of air-mass motion during August 29–September 1 led to a twofold decrease in the total column amount of atmospheric carbon monoxide. The difference in total column amount between August 28 and September 2 was 0.18×10^{19} molecules/cm^2. On September 2, 3, and 4, the relatively clean air was mainly transported from Scandinavia and the North Atlantic (backward trajectories for September 2 are shown in Fig. 2). The conditions under which measurements were taken on September 6 can be called extreme: St. Petersburg and its suburbs were located in smoke plumes from forest fires in the Leningrad, Pskov, and Novgorod oblasts. This can be seen in the NOAA AVHRR satellite image taken on September 4, 2002, where not only the outbreaks of forest fires in northwestern Russia are tracked but also the smoke-plume direction (northwest) is clearly seen (Fig. 4) [14]. The most severe

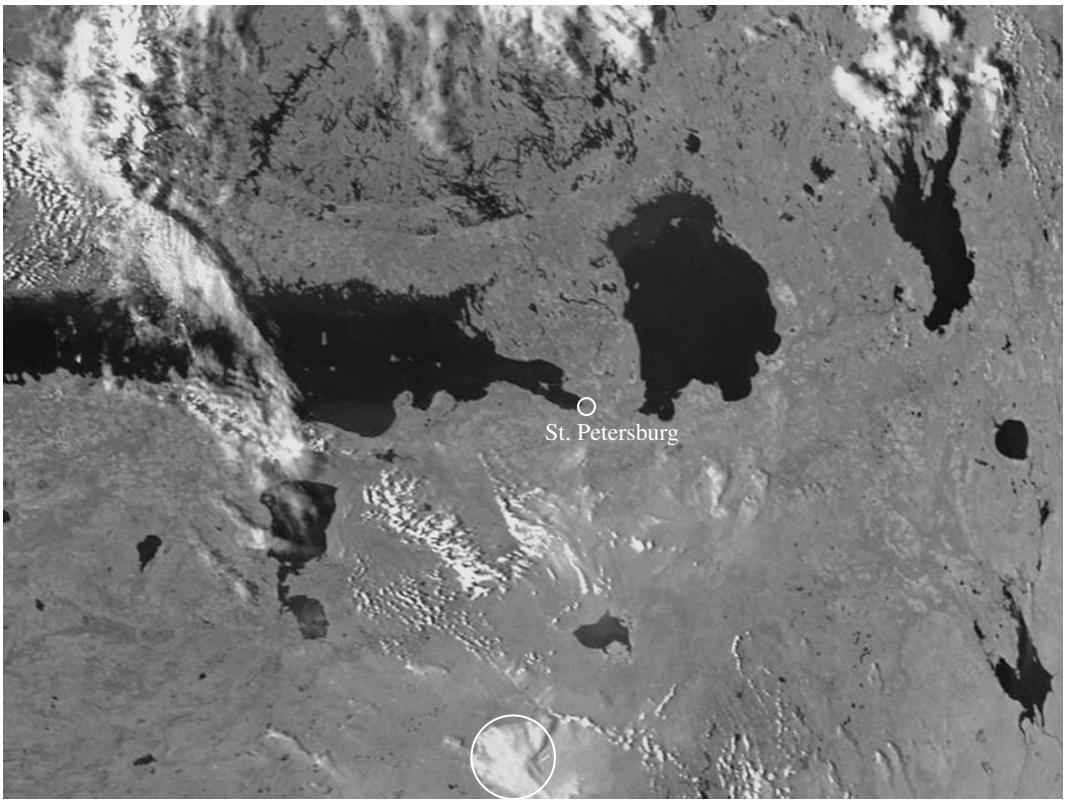

Fig. 3. NOAA AVHRR satellite image (September 4, 2002) with forest fires in northwestern Russia.

outbreak of fire (referred to as the Pskov fire) is shown by a circle. According to satellite images, its area was about 1000 km^2. The evolution of a plume from this forest fire was modeled for September 5–6. The calculation of the spread of the pollutant plume in the atmosphere was performed with the HYSPLIT dispersion module. A set of parameters was specified, which included the date and time, the simulation period, the location of a pollutant source, its intensity in arbitrary units, etc. [13]. Results of calculation of the dispersion of combustion products on September 6, 2002, averaged over a three-hour period (from 13:00 to 16:00) are shown in Fig. 4. The results suggest that the plume covered St. Petersburg and its suburbs. Unfortunately, there are no satellite images of the smoke plume on September 6 at our disposal.

With the use of the backward trajectories and the results of simulation of the spread of a pollutant plume in the atmosphere, it is possible to classify measurements of CO total column amount for the period from August 28 to September 9, 2002, by dividing them into groups with respect to scales and the distance from the influencing sources (Table 2). The first group included Q caused mainly by global-scale sources. The second group was supplemented by regional sources (forest fires in European Russia). The third group was composed of direct fire-plume measurements (mesoscale, local scale) against the background of the contributions of sources from the first two groups. Note that, in the presence of intense CO sources, air-mass transformation processes lead to a change in the total column amount of CO by about 1.5–2 times. The difference between the Q values for the third and the second group is ~0.28×10^{19} molecules/cm^2, and it can be regarded an estimate of CO in the plume of a forest fire (denoted as w_f) for September 6.

Results of simulation of the evolution of CO plumes from sources with given locations and areas

Table 2. Classification of measurements of CO total column amount (Q) for the period from August 28 to September 9, 2002

Group	Date	Average Q, molecules/cm^2	Note
1	Sept. 2–4	$(0.22 \pm 0.02) \times 10^{19}$	Background
2	Aug. 28, Sept. 8–9	$(0.35 \pm 0.03) \times 10^{19}$	Impact of regional sources (forest fires in European Russia)
3	Sept. 6	$(0.63 \pm 0.05) \times 10^{19}$	Measurements in a plume of forest fire

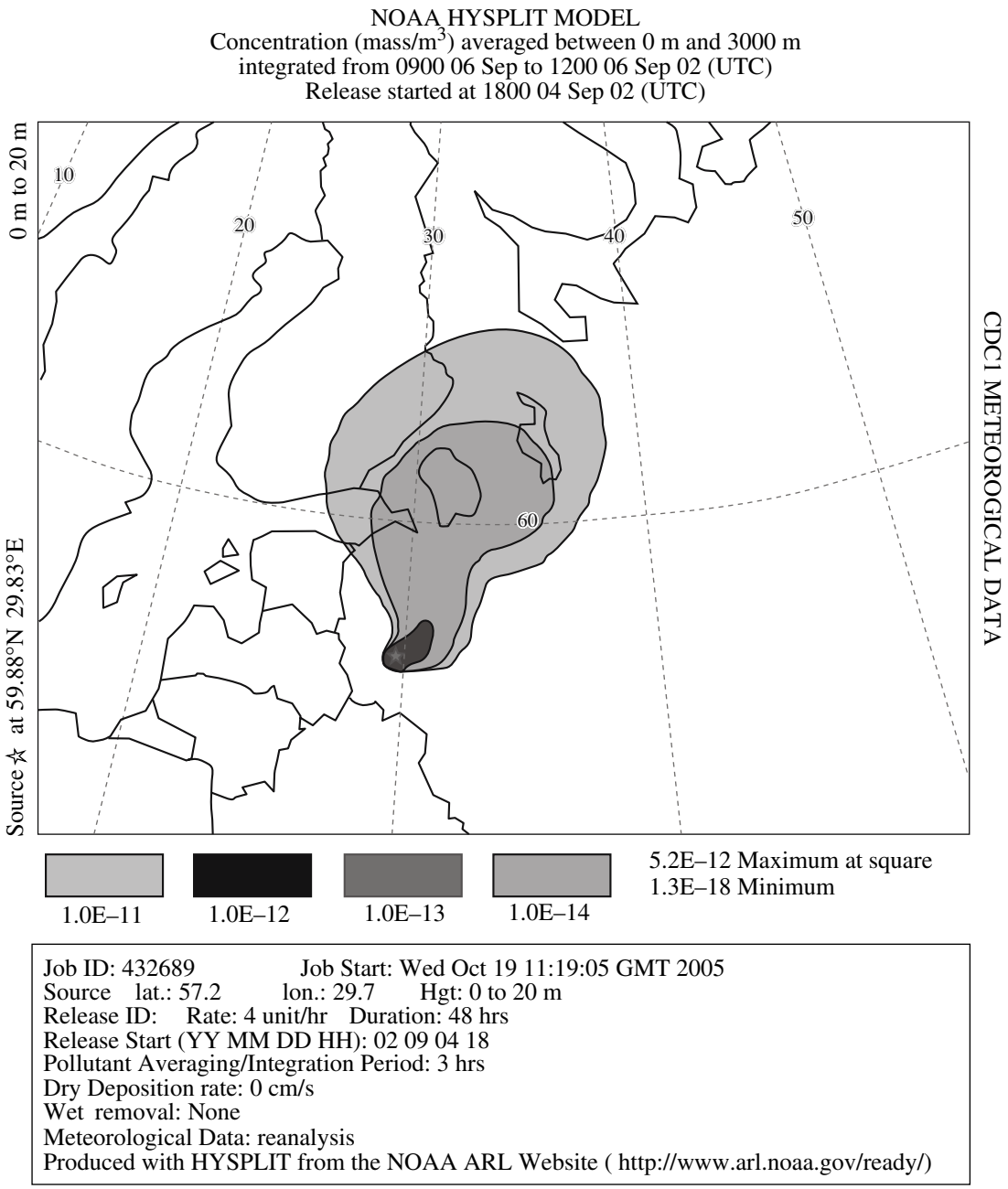

Fig. 4. Results of simulation of the spread of biomass burning products with the HYSPLIT dispersion module, September 6, 2002 (average over the period from 13:00 to 16:00).

and the availability of CO measurements directly in the plumes make it possible to evaluate the intensity of CO emission from a particular CO source. Such an approach was used for the Pskov forest fire (August 28 to September 8, 2002). The calculation involved a number of estimates and approximations. Because of the heavy smoke-filled air, the Pskov fire was classified as a forest ground fire or a peat fire [15].

The dispersion module of the HYSPLIT model computes the spread of pollutant only for a point source. Therefore, the Pskov fire was assumed to be a point source. The CO integral emission (Rm) in model simulations can be chosen arbitrarily because a mass unit in HYSPLIT is an arbitrary quantity. In our case, Rm = 4 mu/h, where mu is a mass unit. The concentration of a pollutant (CO) calculated in the dispersion model is expressed in mass units per cubic meter [13]. In forest ground or peat fires, almost the entire plume with combustion products is located in the lower tropospheric layer from 0 to 3 km [16] because, unlike crown fires, forest ground or peat fires produce no intense convection. The mean concentration in the

model was computed for this altitude range (0–3 km). Different shades of gray in Fig. 4 show areas with different mean pollutant concentrations (Cm). It is easy to see that St. Petersburg is located in the area with concentrations from 10^{-12} to 10^{-13} mu/m^3. Calculations showed that, for the place of spectroscopic measurements, the mean additional CO concentration in the layer 0–3 km in the plume from the Pskov forest fire was Cm $\approx 3 \times 10^{-13}$ [mu/m^3].

The available experimental estimate of CO total column amount in the plume of forest fires (w_f = 0.28×10^{19} molecule/cm^2) makes it possible to determine the additional concentration Ce in the 3-km layer (but now in absolute units):

$$\text{Ce} = w_f/3\ \text{km} = 0.093 \times 10^{14}\ \text{molecules/cm}^3$$

$$(\text{or } 2.7 \times 10^{-4}\ \text{g/m}^3).$$

With the use of the proportion

$$\text{Re/Ce} = \text{Rm/Cm}$$

and the known values of Ce, Rm, and Cm, it is possible to determine the experimental value of the integral emission rate Re for the Pskov forest fire. Our experimental estimate of the rate of CO emission from the entire area of the Pskov forest fire is Re = 3.6×10^9 g/h. The intensity of CO emission from a unit area of a forest fire (S is a fire area) is

$$I = \text{Re}/S \approx 3.6\ \text{g/(h m}^2).$$

Estimates of CO emissions (from forest fires) are given in the literature mainly in the form of the specific emission M, which means the total amount of CO released by a unit area of forest during the fire. Therefore, in order to make comparison with independent results, it is necessary to know the period of burning of a forest massif. In ground forest or peat fires, the burning period T of a forest (peatbog) massif is normally 2 to 3 days [17]: T = 48–72 h. In our case, this is confirmed by the fact that the active burning of the Pskov forest fire, already visible by satellite on September 4, continued at least until September 6. Thus,

$$M = IT \approx 0.17\text{–}0.26\ \text{kg/m}^2.$$

Published estimates of M for Northern Hemisphere midlatitude forest fires are shown in Table 3 for comparison [17, 18].

According to these data, the range of independent estimates of M for Northern Hemisphere midlatitude forests is 0.124–0.425 kg/m^2. The estimates of other authors differ by a factor of more than 3. It can be seen that our result (0.17–0.26 kg/m^2) is the closest to M = 0.213 kg/m^2 [18]. Taking into account the fact that a smoldering stage dominates in ground forest fires, we can conclude that $M \sim 0.2$ kg/m^2 is suitable for use in computations of the integral CO emission from forest fires in the high latitudes of the Northern Hemisphere.

Table 3. Carbon oxide specific emission M in forest fires

Reference	M, kg/m^2	Commentary
Present study	0.17–0.26	Ground forest fire (smoldering dominates)
Donald R. Cahoon Jr. et al. [18]	0.124	Active flaming stage
Donald R. Cahoon Jr. et al. [18]	0.213	Smoldering stage
Jane Liu et al. [17]	0.425	Without division into flaming and smoldering stages

Note that our value of M is half the estimate (0.425 kg/m^2) given in [17].

Analogous estimates of CO emissions from forest fires can be derived from satellite measurements of the CO total column amount (or lower tropospheric CO). The CO fields retrieved from satellite measurements permit not only the evaluation of the intensity of CO emission from severe fires but also a visual comparison of the simulated spread of the CO plume (HYSPLIT) with areas of anomalous CO values. However, satellite experiments (MOPITT, SCIAMACHY) do not yet permit the derivation of CO data with certainty in the tropospheric layer 0–2 km. Since this layer exhibits the largest CO variations due to ground-based sources (e.g., forest fires), the use of satellite data in the approach described above may result in significant errors.

CONCLUSIONS

1. Results of spectroscopic measurements of the CO total column amount content near St. Petersburg during forest fires in August 2002 have been analyzed. With the HYSPLIT model, air-mass trajectories have been calculated and CO dispersion on a mesoscale in the measurement period has been determined.

2. On the basis of the HYSPLIT model computations and measurements of Q, the specific intensity of CO emission in the Pskov forest fire of August 28 to September 8, 2002, was estimated at 0.17–0.26 kg/m^2. This estimate can be used for calculation of the integral CO emission from forest fires in northwestern Russia and for model computations.

3. An estimate of the CO emission from forest fires determined from ground-based measurements can also be applied to satellite measurements if they contain information about CO in the lower troposphere (0–2 km).

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