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COUPLING EXPERIMENTAL DATA AND CMAQ TO INVESTIGATE THE CARBON MONOXIDE FIELD FORMATION IN THE NORTH-WEST REGION OF RUSSIA

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1. INTRODUCTION

The tropospheric burden of carbon monoxide, like that of many other trace gases, has been increasing due to human activities, although its upward trend ceased around 1995. The lifetime of CO is of the order of a few months only, and the significance of CO in atmospheric chemistry lies mainly in its competition with many other gaseous pollutants—importantly the greenhouse gas CH4—for the hydroxyl radical (OH, CO + OH → CO2 + H). Increased CO emissions cause higher CO burdens and more reaction with OH, leaving less OH for cleansing the troposphere of other reduced gases. In the background troposphere, about one third of all OH is removed by CO that reacts rapidly with OH (contributing to the latter's very short lifetime of 1 second only) [1].

Until recently, we were mostly informed about tropospheric CO concentrations via surface measurements. Now, results from remote sensing and an increasing number of aircraft flights give improved global coverage and some vertical resolution. Since the launch of the MOPITT [2] satellite instrument, followed by SCIAMACHY [3], AIRS [4] and others, we have a much better picture of large scale continental pollution plumes. The vertical resolution of satellite based remote sensing is limited to several km at best, and vertical profiles coordinated with satellite overpasses are needed to better define vertical variability. Before the satellite, surface and aircraft measurements are combined, their relative calibration must be accurately determined.

Analysis of data on carbon monoxide in the atmosphere shows significant temporal and spatial variations of both concentration fields and column amount. In this study, we applied CMAQ to investigate the main factors which influence the CO distribution in the North-West region of Russia during August-September 2002, the period, when intensive forest fires were detected in this area.

2. OBSERVATIONAL AND MODEL DATA

2.1. Observational data

Experimental data include:
1. ground-based measurements of CO total column (NDACC, Network for Detection of Atmospheric Composition Change [5]; three Russian stations [6,7]);
2. measurements of CO concentrations (NOAA ESRL/GMD CCGG Cooperative Air Sampling Network [8]);
3. satellite measurements of CO (MOPITT, Measurements of Pollution in the Troposphere) over model domain.

2.2. Model Set-Up

CMAQ V4.6 was run for a 750x750km domain (with 27 km resolution) which included North-west region of Russia (fig. 1). A 12 day simulation was performed for 2-13 of September 2002. AVHRR [9] observations detected intensive forest fires near St.Petersburg during this period (fig. 2).

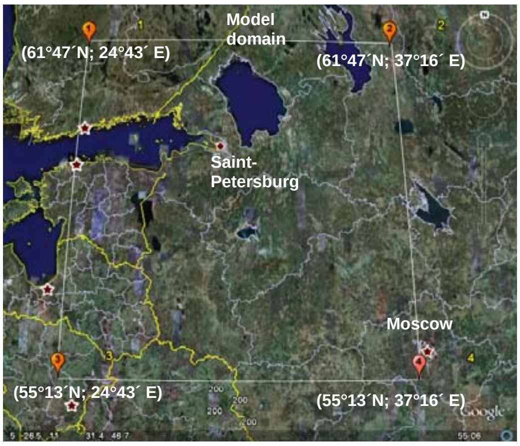

Fig.1. CMAQ model domain.

The meteorological fields necessary for the CMAQ simulations were prepared by MM5 (ds083.0 FNL 2.5x2.5 12-hourly analyses) and MCIP. Two most intensive sources indicated in fig.2 were processed by SMOKE V2.3. are of $3{\cdot}10^6$ ton/yr and $1{\cdot}10^6$ ton/yr accordingly. Emissions were already estimated using total column amount measurements near St. Petersburg and HYSPLIT dispersion model [10].

Both forest fires were in smoldering phase. Location of the centers of the sources: (57°.10´ N; 31°.15´E) and (58°.57´ N; 30°.28´E). Preliminary approximations of CO emission from considered fires.

Boundary and initial conditions were based on analysis of ground-based measurements of CO

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Fig.2. Location of the forest fires (September, 04 2002).

Fig.3. Location of the measurement sites used for IC/BC set-up.

concentration and total column amount (observational stations are indicated in fig. 3) . Two types of boundary conditions were used (fig. 4 and 5): 1. "undisturbed" (measurements for the period of July-September 2002 were used); 2. "disturbed" (measurements for the period of 20 August – 15 September 2002 were used).

For the north boundary conditions observations from the following stations were used: Ny-Alesund, Norway (Spitsbergen), NDACC and NOAA (concentrations and total column amounts); Kiruna, Sweden, NDACC (total column amounts); Pallas-Sammaltunturi, Finland, NOAA (concentrations).

Boundary conditions for the west boundary were set using data from: Harestua, Norway, NDACC (total column amounts); Baltic Sea, Poland, NOAA (concentrations); Bremen, Germany, NOAA (concentrations). South and East boundaries: Zvenigorod, Russia, Institute of Atmospheric Physics RAS, (total column amounts); Obninsk, Russia, Institute of Experimental Meteorology and NOAA (concentrations and total column amounts).

Initial conditions were based on measurements at Harestua, Norway, NDACC (total column amounts); Baltic Sea, Poland, NOAA (concentrations); Bremen, Germany, NOAA (concentrations) and St. Petersburg, Russia, St. Petersburg State University (total column amounts).

3. ANALYSIS OF CMAQ RESULTS AND OBSERVATIONAL DATA

CMAQ model results were compared with observations. CMAQ data were extracted for the 27-km grid cell corresponding to the monitoring site near St. Petersburg. Different scenarios were considered to investigate model's sensitivity to various boundary conditions (disturbed and undisturbed) and various emission rates (low bound: 8220 ton/day / 2740 ton/day; and upper bound: 13 times higher). Model results (fig. 6) indicate that emissions from forest fires should be 13 times higher than the default value to match the observations.

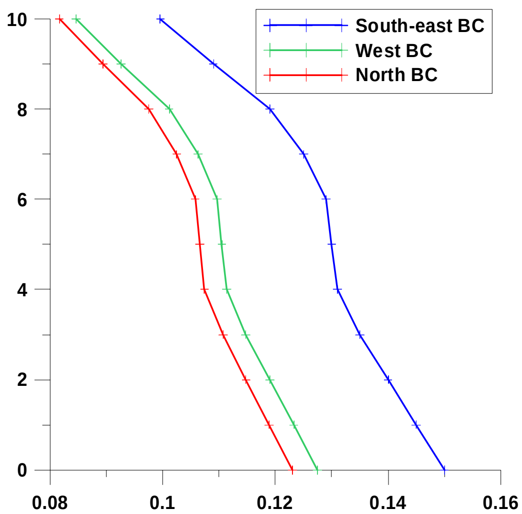

Fig.4. CO concentration vertical profiles for undisturbed boundary conditions.

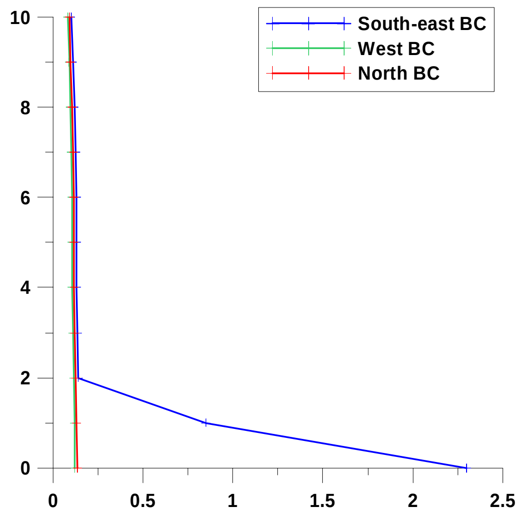

Fig.5. CO concentration vertical profiles for disturbed boundary conditions.

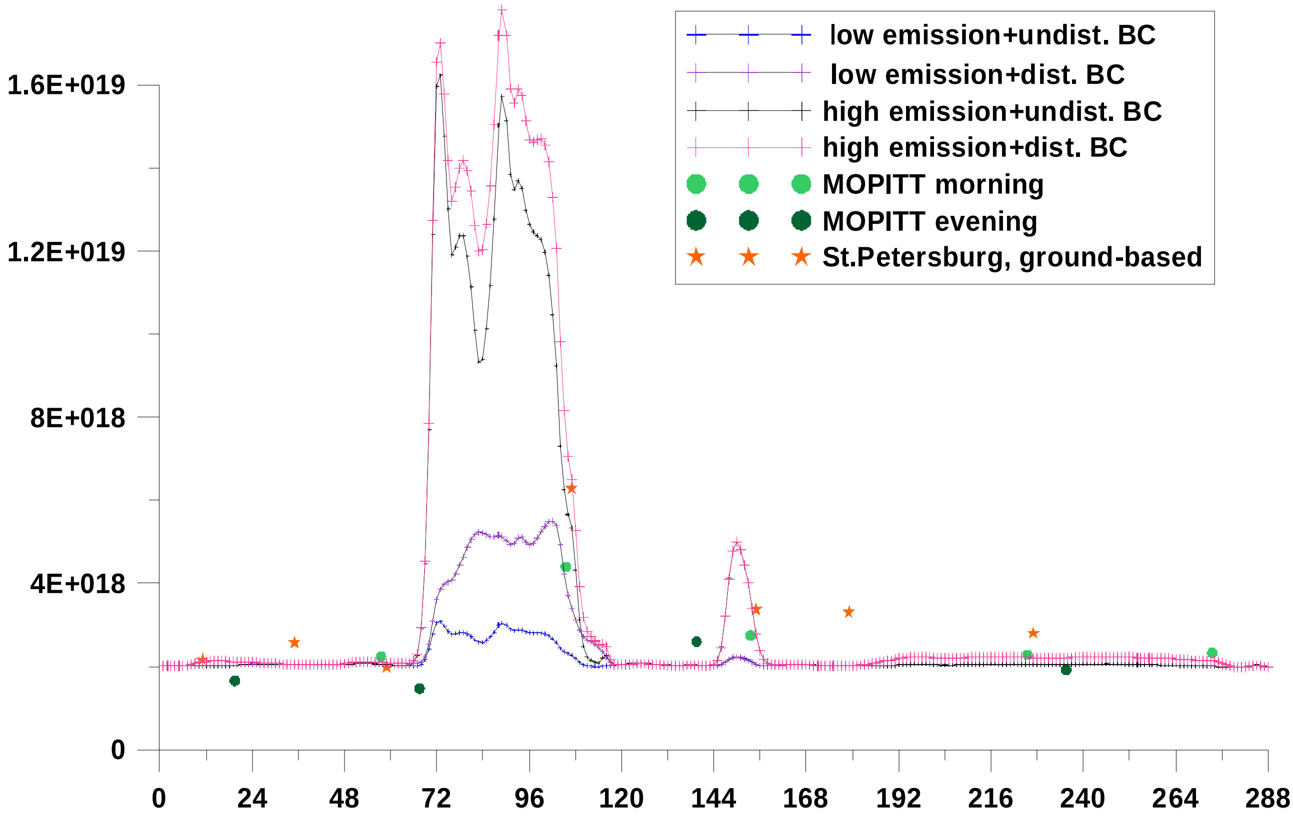

Fig.6. Comparison of CO total column amounts obtained from CMAQ and from observations (ground-based and satellite).

Fig.7 shows comparison of model results simulated for "high emission + disturbed BC" (pink scenario in fig.6) with MOPITT data. "Missing data" areas in MOPITT observations (fig.7) usually correspond to the areas with high values of CO total column amount simulated by CMAQ. We suggest that fire smoke is interpreted by MOPITT as cloud cover. There are significant systematic differences between MOPITT and CMAQ data on CO total column amount. MOPITT has low sensitivity to the CO concentration in 2.4μm channels (which are used for CO measurements in the boundary layer), therefore the near-surface data are not reliable.

4. SUMMARY

The CMAQ modeling system has been used to simulate concentration fields of CO in the North-West region of Russia. Simulation period: September 2002, when intense forest fires occurred. Modeling domain: 750x750km. Resolution: 27km.

CMAQ results were compared with the following observations:

1. satellite data from MOPITT;
2. ground-based measurements from NDACC and St. Petersburg University, Russia.

Our analysis indicated that spatial patterns of simulated and observed CO total column amount (TCA) are similar.

However, there are differences in TCA values, which can be explained by several factors:

1. Information cannot be derived from satellite measurements when clouds are present;
2. Fire smoke is interpreted by MOPITT as cloud cover, therefore – many pixels contain missing data;
3. The sensitivity of MOPITT instrument is low (only one channel, 2.4mm channels are not reliable);
4. A lot of missing data from satellite measurements make it difficult to compare the results for specific time periods.

However, using CMAQ modeling system in combination with satellite and ground-based data allowed us to improve emission estimates. In this study, matching CMAQ results with observations suggested that emissions are significantly higher than the default preliminary values.

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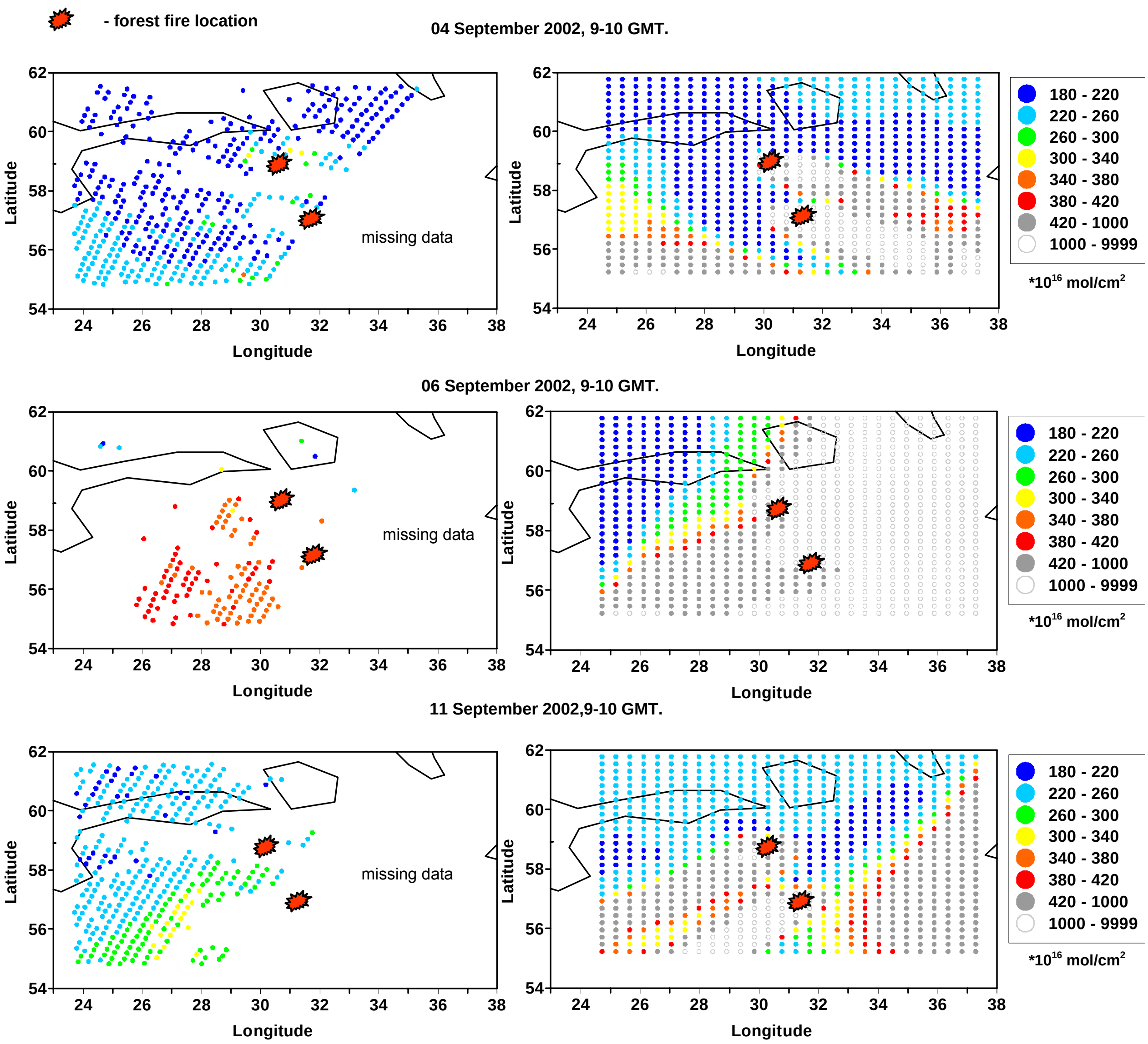

Fig.7 Comparison of CO total column amount derived from MOPITT measurements (left panels) and obtained from CMAQ model simulations (right panels).