Recent changes of atmospheric composition in background and urban Eurasian regions in XXI-th century

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Abstract. An analysis of the CO and CH\textsubscript{4} total column (TC) as well as aerosol optical depth (AOD) data in background and urban Eurasian regions for different time-periods and seasons from 1998 to 2018 years is presented. Trend estimates based on long-term spectroscopic datasets of OIAP RAS for Moscow, Zvenigorod (ZSS, Moscow province), Beijing (joint site of OIAP RAS and IAP CAS) and NDACC stations located in Eurasia are compared between themselves and with similar assessments obtained from satellite data. The comparison of satellite and ground-based trend estimates was provided for the days of synchronous measurements only. Analysis results of the satellite observations of AIRS v6 of CO and CH\textsubscript{4} TC and MODIS AOD data are confirmed by ground-based trend estimates. Significant decrease of anthropogenic CO in the megacities Moscow (2.9±0.6%/yr) and Beijing (1.2±0.2%/yr) for autumn months of 1998−2018 was found according to ground-based spectroscopic observations. In spite of total anthropogenic CO emission decrease (for Europe and China) and the decrease of wild-fires emissions in Central North Eurasia (0−90° E, 42−75° N) in 2008−2018 we found CO TC stabilization or even increasing for summer and autumn months of 2008−2018 in background regions of Northern Eurasia. Decrease of AOD over Central and Southern Europe as well as over China (1−5%/yr) was observed since 2007. Since 2007-2008 an increase in CH\textsubscript{4} TC positive trend values over Northern Europe as well as for tropical belt of Eurasia was obtained.

1. Introduction

A numerous publications report about decrease in anthropogenic emissions of some greenhouse gases and pollutants (e.g., CO, NO\textsubscript{X} and aerosols, including “black carbon”, BC) in European countries as well as in South-East Asian countries excluding India [1-5]. Negative trends in the column and surface concentration of these impurities were observed in most of urban and background areas of the Northern hemisphere and associated in these studies with a decrease in anthropogenic emissions. Carbon monoxide CO isn’t a greenhouse gas, and in the process of studying the interrelation of climate change with changes in the atmospheric composition in recent years CO has gone as if by the wayside. However, CO has a number of important properties: it largely determines the concentration of the hydroxyl (OH) that is the main atmospheric oxidant, and thus plays an important role in all atmospheric photochemistry; it leads to the destruction of one the most important greenhouse gas ozone in the background atmosphere [6-8].

The surface concentration of carbon monoxide in cities is one of the most important parameters for air quality monitoring, due to the CO toxicity, as well as the possibility of the formation of dangerous concentrations of surface ozone at elevated concentrations of CO in conditions of photochemical smog in large cities or in wildfires areas [8,9]. The associations of anthropogenic CO emissions with emissions of other greenhouse gases and pollutants such as CO\textsubscript{2}, NO\textsubscript{X}, aerosols are well known [5, 8,
The global natural and anthropogenic CO emissions are approximately comparable. The main anthropogenic source of CO is the combustion processes, including fuel combustion. One of the most important non-permanent sources of CO is wildfires, which emissions can reach 25% of global emissions [11,12]. The main natural source of atmospheric CO is the oxidation of methane and non-methane hydrocarbons [7,8].

An example of modern long-term tendencies in megacities and background regions during different time-periods and seasons is presented at the Fig 1. Methane is the second greenhouse gas (GHG) after CO$_2$ by the integral significance of the greenhouse effect and the first one by the greenhouse effect per molecule [13,14]. The growth rate of methane concentration in the atmosphere is significantly higher than the growth rate of carbon dioxide concentration, especially after 2007 [1,3,15].

The content of aerosols in the atmosphere is one of the most important parameters of air quality. Aerosols have a significant impact on climate and its changes through radiation forcing. Emissions and concentrations of some aerosols (“black carbon” aerosol, smoke aerosol) in regions with high anthropogenic load or areas of wildfires are closely connected with emissions and concentrations of many atmospheric impurities, in particular CO [5].

Our previous studies focused mainly on variations and trends in CO and CH$_4$ total column in the cities and background areas [5,16,17]. In the recent paper [18] positive trends of CO TC in the autumn months in the unpolluted outskirts of Moscow (Zvenigorod) and St. Petersburg (Petrov) in the period of 2007-2014 were noted simultaneously with a significant anthropogenic emission decrease in different megacities (as example, Moscow and Beijing) in the same period. An attempt to investigate the change in trends of CO and CH$_4$ TC in different regions of Eurasia was made in [19]; it was found that the increase in CO TC in the summer and autumn months in Northern Eurasia isn’t directly related to either anthropogenic emissions or emissions from wildfires. However, analyzed time-period in these studies was relatively short and the further verification of trend estimates is needed.

The present study aimed to clarify obtained positive trend dynamics of CO TC, to study the trend dynamics of aerosol optical depth (AOD) and CH$_4$ TC, and to compare the trend estimates obtained for different impurities, different areas and methods. For this purpose, ground-based measurements (spectrometers of OIAP RAS and NDACC, Network for the Detection of Atmospheric Composition Change) were used; daily data of orbital spectrometers AIRS v6 and MODIS/Terra were used to study the spatial distribution trends of CO, CH$_4$ and AOD. To assess the impact of Eurasian wildfires on the obtained trend values the average annual emissions of combustion products from GFED v4.1 were calculated [20]. Trends of CO emissions from wild-fires in boreal areas of Eurasia are presented in Table 1.

| Years       | CNE     | ENE     |
|-------------|---------|---------|
| 2003-2018   | -3.07±1.44 | 1.18±0.67 |
| 2007-2018   | -5.17±2.72 | 5.24±2.01 |

### Table 1. CO emission trends (%/yr) from wildfires for Central North Eurasia (CNE, 10º–90º E, 42º–75º N) and Eastern North Eurasia (ENE, 90º–180º E, 42º–75º N) according GFED v4.1.

#### 2. Methods and instrumentations

2.1. OIAP RAS and NDACC long-term ground-based spectroscopic measurements of CO and CH$_4$ TC

The ground-based observations of CO and CH$_4$ TC were carried out at 4 sites of A.M. Obukhov Institute of Atmospheric Physics (OIAP RAS) (Moscow, ZSS, ZOTTO and Beijing – joint IAP CAS and OIAP RAS observation stations) by absorption spectroscopy method with using identical grating spectrometers of medium resolution (0.2 cm$^{-1}$), [21, 22, 5]. At Petrov site the ground-based FTIR observations were carried out by Saint-Petersburg State University (SPBSU), see details in [23,24].
We also analyzed CO TC datasets from Thule, Kiruna, Harestua, Ny-Alesund, Bremen, Zugspitze and Jungfraujoch NDACC stations. Locations and specifics of stations are presented in Table 2. The details about the European NDACC stations could be found in [25] http://www.ndsc.ncep.noaa.gov/sites/).

Specifics of observations and site location are presented in Table 2.

**Table 2.** Information about the ground-based stations of CO and CH$_4$ spectroscopic measurements (location, used observation periods, a number of measuring days per year, etc.).

| Site (number), Analyzed time intervals (years) | Typical season, amount of observation days per year for CO/CH$_4$ | Coordinates, °N/°E/height above sea level | Affiliation, country, region |
|---------------------------------------------|---------------------------------------------------------------|----------------------------------------|-------------------------------|
| ZSS, 1972–2018                             | Round the year, 70–90/70–90                                   | 55.7°/36.8°/200 m                     | OIAP, Russia, Moscow province |
| Beijing, 1998–2018                          | October–November, 15–20/0                                     | 40.0°/116.4°/80 m                     | OIAP/IAP CAS, China, Beijing |
| Peterhof, 2003–2016                         | Round the year, 60–80/50–70                                  | 59.9°/29.8°/20 m                      | NDACC/SPbSU, Russia, Peterhof |
| Thule, 2003–2018                            | April–September, 20–70/10–50                                 | 76.5°/68.7°/30 m                      | NDACC, Greenland             |
| Kiruna, 2003–2018                           | Round the year, 50–100/50–100                                 | 67.8°/20.4°/420 m                     | NDACC, Sweden                |
| Harestua, 2003–2015                         | Round the year, 30–60/30–60                                  | 60.2°/10.8°/600 m                     | NDACC, Norway                |
| Ny-Alesund, 2003–2018                      | March–October, 20–40/20–40                                   | 78.9°/11.9°/15 m                      | NDACC, Norway, Spitsbergen   |
| Bremen, 2003–2018                           | Round the year, 10–30/10–30                                  | 53.1°/8.8°/30 m                       | NDACC, Germany               |
| Zugspitze, 2003–2018                       | Round the year, 30–50/30–50                                  | 47.4°/11.0°/2964 m                    | NDACC, Germany, Alps         |
| Jungfraujoch, 2003–2018                     | Round the year, 20–50/30–50                                  | 46.5°/8.0°/3850 m                     | NDACC, Switzerland, Alps     |

2.2. **Ground-based AERONET datasets of AOD**

Estimates of AOD trends were provided by using of ground-based observations of Eurasian AERONET-network sites, diurnal AOD data for 500 Nm wavelength, Level 1.5, [26] http://aeronet.gsfc.nasa.gov/) and orbital MODIS Terra 1°x1° diurnal AOD data for 550 Nm of Level 3 collection 5.1 (http://modis.gsfc.nasa.gov/). AERONET L 1.5 was chosen in accordance with the largest number of daily data in comparison with L2.0. AERONET sites location see p. 3.1 table 4.

2.3. **AIRS orbital data of CO and CH$_4$ Total Content**

The AIRS (Atmospheric InfraRed Sounder) spectrometer was launched aboard the Aqua satellite on the 4th May, 2002. The main element is the orbital grating spectrometer which records the atmospheric absorption spectra of Earth's infrared radiation in the spectral range of 3.75 to 15.4 microns [27] twice a day from ascending and descending orbits, covering more than 80% of the Earth's surface. In the current study, we used the data of the third level AIRS v6 (Level 3 v6, resolution 1°x1°, only daily measurements of CO and CH$_4$, (http://disc.sci.gsfc.nasa.gov/AIRS/documentation/v6_docs/v6releasedocs1/V6_L3_Product_User_Guide.pdf) from an ascending orbit (i.e. around 12: 30-13:30 local time for each point). The number of days of measurements per year for each 1°x1° cell is about 300-320 in the middle latitudes of Eurasia.

2.4. **Satellite MODIS datasets of AOD**
MODIS/Terra data (550 Hm, Level 3 collection 5.1) are available from 2003 year, http://modis.gsfc.nasa.gov/. MODIS AOD data aren’t available for winter months for middle- and high-latitude regions; therefore AOD so called “annual” trends obtaining from MODIS for all Eurasian regions relate to season from March to October approximately (excluding sub-tropical and tropical ones, where “yearly-averaging” of AOD diurnal means are correct).

2.5. Comparison of satellite and ground-based data
Before obtaining of satellite trend distributions, the comparison of satellite data (diurnal means in spatial resolution 1°x1° for AIRS CO and CH₄ TCs and MODIS AOD products) and ground-based data was produced [16-18]. Best correlation of orbital diurnal CO TC data with ground-based ones was obtained for AIRSv6 (R²~0.7-0.8 and slope~1 for CO TC for linear type of the regression dependences) especially under background conditions and for MODIS/Terra for AOD (R²~0.6-0.8, slope~0.51-0.96) that is in a good agreement with other studies [28-30].

In present work, the linear trends of all measured parameters for all three impurities were compared on synchronous measurement days (i.e., there were used only days for which both ground-based and satellite spectrometer data are available). Trend estimates based on satellite and ground data obtained by this method are in good agreement for all points and all impurities (see below table 3 and 4). Thus, we consider the trend distributions obtained from observations of orbital complexes are sufficiently representative for both background and urban areas.

2.6. Trend evaluation method
Trends were calculated separately for seasonally/yearly averaged means for both ground-based and satellite data; estimates for every case were produced for linear type of approximation. Trend distributions based on AIRS v6 satellite data were calculated similarly and are based on averaged data for each 1°x1°cell.

3. Results and discussion

3.1. Comparison of satellite and ground-based data
Preliminarily, for all three impurities and for each sites of ground-based observations, the trend estimates obtained from ground-based and orbital complexes were compared on the same days for orbital and ground-based measurements. Satellite and ground-based estimates of CO and CH₄ and AOD trends are in good agreement for almost all selected items, see table 3 and 4.

It should be noted that the statistical significance of trend estimates obtained by this method is relatively low (5−30 days for seasonal estimates, and 15−110 for yearly-averaged estimates, depending on the point, year and impurity). Therefore, while the estimates in table 2 partly (partially) reflect overall long-term trends, we recommend to consider these estimates as a preliminary result.

The Table 3 is intended primarily to prove the representativeness of satellite trends and their spatial distributions.

| Ground-based observations: sites, years after 2000 | Ground-based | AIRS |
|--------------------------------------------------|--------------|------|
| Season | Trend CO, %/year | Trend CH₄, %/yr | Trend CH₄, %/yr |
| Year | Season | Year | Year | Year |
| ZSS | 03−18 | 08−18 | Sep−Nov | -1.25±0.10 | -0.18±0.01 | -0.77±0.11 | -0.59±0.03 |
| | | | | -0.63±0.03 | 0.77±0.04 | -0.68±0.03 | 0.04±0.00 |
regions are presented at Fig. 1. The typical results of CO TC interannual variations and trend estimates for urban and background sites, coordination, °N./E., years AERONET MODIS/Terra Trend, %/yr Trend, %/yr Year Season Year Season

| Sites          | 03–16 07–16 | Oct–Nov | 03–16 07–16 | Sep–Nov | -1.80±0.54 | -1.33±0.91 | -0.57±0.29 | -0.30±0.35 |
|----------------|-------------|---------|-------------|---------|------------|------------|------------|------------|
| Peterhof       |             |         |             |         | -0.53±0.29 | 0.07±0.40  | -1.06±0.28 | -0.40±0.43 |
| Thule          |             |         |             |         | -1.65±0.16 | -0.60±0.05 | -0.73±0.04 | -0.15±0.01 |
| Kiruna         |             |         |             |         | -0.90±0.05 | -0.32±0.02 | -0.71±0.03 | -0.38±0.02 |
| Harestua       |             |         |             |         | -0.25±0.01 | 0.94±0.05  | 0.03±0.00  | 0.56±0.02  |
| Ny Al.         |             |         |             |         | -1.09±0.47 | -0.47±0.69 | -0.82±0.38 | -0.40±0.54 |
| Bremen         |             |         |             |         | -0.76±0.05 | -0.92±0.01 | -0.76±0.05 | -0.83±0.04 |
| Zugspitze      |             |         |             |         | -0.50±0.02 | -0.32±0.02 | -0.66±0.03 | -0.68±0.04 |
| Jungfr.        |             |         |             |         | -0.62±0.03 | -0.28±0.01 | -0.93±0.05 | -0.70±0.07 |

Table 4. Aerosol optical depth (AOD) trends according AERONET and MODIS/Terra observations

3.2. Trend estimates of CO and CH₄ Total Content and AOD
The typical results of CO TC interannual variations and trend estimates for urban and background regions are presented at Fig. 1.
Figure 1. Variations of CO Total Content and its trends according OIAP spectroscopic observations for 2000-2017 time-period: a) - yearly averaged values for Moscow and rural Zvenigorod (ZSS, Moscow province) b) - seasonal averaged CO TC for Zvenigorod

For the period 2003-2018, yearly averaged CO TC decreased at the rate of 0.1-1.5%/year over almost the entire territory of Eurasia, see Fig. 2a. These results are confirmed in [28,31] and in our previous studies [17-19]. The greatest rate of CO reduction is typical for the Beijing region and Central China (up to 1.5% /yr), which corresponds to our estimates for Beijing (from 1.2 to 1.5%/yr) obtained by the ground-based spectroscopic measurements [5,18,19].

However, for the period after approximately 2007-2008, the rate of the CO decrease over almost all of Eurasia regions slows down (exclude China). We note that the significant differences between the seasonal trend distributions from the yearly-averaged one can be related especially to the summer-autumn period.

For the summer (June-August) and autumn (September-November) seasons 2008-2018, Fig.2b and 2c respectively, an increase in CO TC was obtained for the middle and high latitudes of Eurasia, including Northern Europe. In Central Siberia, the rate of CO increase reached 1.0% / year (in the summer period 2008-2018), which can be explained by wildfires impact in some regions of Siberia. The positive trend of emissions from fires in Eastern Siberia in 2008-2018 is confirmed by GFED data on the Eastern North Eurasia domain (90°−180° E, 42°−75° N), see Table 1. In the Western Eurasia (domain Central North Eurasia, 10°−90° E, 42°−75° N) the trend of emissions from wild-fires in this period is negative according GFED v4. 1. It is known that the greatest intensity of wildfires in Central Europe and Northern Eurasia is usually observed for the period from May to August, and the lifetime of CO in the atmosphere does not exceed 3 months for the winter period and 10-15 days for the summer [6,8], and yet, over a large area of Eurasia, positive autumn trends were obtained in the autumn months after 2008 (Fig. 2c).

CO TC trends positive dynamic (the difference between 2009-2018 and 2003-2009 trends) was obtained for almost all of Eurasian territory (Fig. 2d).
Figure 2. Distribution of CO total column trends, according AIRS v6 data:
  a) - for the 2003-2018, yearly averaged means;
  b) - for summer months of 2008-2018;
  c) - for autumn months of 2008-2018;
  d) – trend dynamics of yearly averaged CO TC: the difference between trend (2009-2018) and (2003-2009).

Similarly, trend of CH₄ TC estimates were derived from AIRS v6 data, and then compared with the estimates based on NDACC station measurements (Table. 2). The rate of increase in the methane total column for 2003-2018 was 0.1–0.4 % / year, depending on the region, which doesn’t contradict the
GAW global assessment and WDCGG estimates (an increase in methane concentrations of 0.3%/yr), [1] and http://ds.data.jma.go.jp/gmd/wdcgg/. Fig. 3 presents the trends of CH$_4$ obtained for the 2003-2018 (Fig. 3a) and for the years 2008-2018 (Fig. 3b). Many researchers, explaining the beginning of the rapid increase in methane concentrations after 2007, refer to the processes of warming and increasing natural emissions of methane from northern marshes, river estuaries and tundra [3,15,32]. However, we’d like to note that in the polar regions of East Eurasia, trends of CH$_4$ TC haven’t changed significantly after 2007; the greatest changes have occurred in the Southern regions of Eurasia and North Europe, Fig. 3b and tab.3. The near zero growth of methane emissions obtained in the Eastern regions of the Arctic is consistent with the results of the study of regional methane emissions [33].

**Figure 3.** Distribution of CH$_4$ total column trends according to AIRS v6 data:

- a) - for the 2003-2018, yearly averaged means;
- b) - the same for 2008-2018;
- c) - trend dynamics of CH$_4$ TC: the difference between trend (2009-2018) and (2003-2009)

MODIS AOD data for middle and high latitudes for the period from about November to March (depending on latitude) aren’t available, and MODIS measurement coverage during the winter months provides only for the subtropical and tropic areas. Therefore, the "years-averaged" distributions of
AOD trends presented in latitudes North of 50 ° N cover the period from about April to October (Fig. 4a,b).

**Figure 4.** Distributions of aerosol optical depth (AOD) trends over Eurasia according MODIS/Terra:

- **a)** - for the 2003-2017 period, yearly averaged means;
- **b)** – the same for 2007-2017;
- **c)** - for the autumn months of 2003-2017;
- **d)** - for the autumn months of 2007-2017.
It should be noted that the growth of AOD for "early-averaged" values and for the summer months over Siberia is associated with summer wildfires in this region. However, we obtained that the AOD decline accelerates over Europe as well as over China, after 2007, for the annual average (Fig. 4a, b), as well as for seasonal (for summer and autumn, Fig. 4c, d) estimates. This fact is confirmed by ground-based AERONET data (Table 4). The trends of AOD coincide with the CO TC trends in regions with high anthropogenic load or in areas of wild-fires (AOD and CO growth in Eastern and Central Siberia in the summer months, and decline of both in the China at any season), but differ for Europe. Remind that in the autumn months after 2007-2008 in Southern Europe, the trends of CO TC are close to zero (Fig. 2c), while in the same season and period and same areas a significant decrease in AOD (1-2%/year) was observed, Fig. 4 g.

4. Conclusions
The analysis of long-term series of ground-based and satellite spectroscopic measurements of carbon monoxide, methane and aerosol optical thickness were presented. According to the ground-based spectroscopic measurements and the AIRS satellite spectrometer, trend estimates in the Total Column (TC) of CO and CH$_4$ for the Eurasia domain (0−180° E, 0-85° N) for different time periods and seasons were provided. Similar estimates using data from the AERONET ground-based network and the MODIS/Terra orbital sensor were obtained for aerosol optical depth (AOD). A good agreement between the satellite and the ground-based estimates was obtained. In the megacities of Moscow and Beijing the decline in the CO TC was observed, as before 2008 and after, at the rate of 1.2-2.4%/yr, depending on the city, season and analyzed time interval. The yearly averaged CO TC in the background regions of Northern Eurasia for the period 2003-2018 as a whole decreased at the rate of 0.05−1.5 %/yr depending on the region, and CH$_4$ TC increased at the rate of 0.16–0.65 %/year. After 2007-2008 CO and CH$_4$ trends in uncontaminated areas of Eurasia have changed. In summer and autumn months, an increase in CO TC in most mid-and high-latitude regions of Europe and Asia, including the Arctic regions was obtained, as well as an increase in the growth rate of CH$_4$ in Northern Europe and in tropical and subtropical Eurasian regions. The decrease of AOD in Southern and Central Europe in the summer and autumn months of 2007-2017 at the rate of 0.8-2.0 %/year is consistent with negative trends in emissions from fires in Europe and Western Siberia and isn’t consistent with the positive CO TC trends in Europe. One of possible reasons of such dynamics of trends CO and CH$_4$ content in the atmosphere could be changes in the global photochemical system occurring against the global climate change, in particular, changes in the source/sink ratio for trace atmospheric gases.

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