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CHANGES IN TRENDS OF ATMOSPHERIC COMPOSITION OVER URBAN AND BACKGROUND REGIONS OF EURASIA: ESTIMATES BASED ON SPECTROSCOPIC OBSERVATIONS

ABSTRACT. The analysis of the CO and CH4 total column (TC) as well as aerosol optical depth (AOD) data in urban and background regions of Eurasia for different seasons and periods from 1998 to 2016 years is presented. Trends estimates based on long-term spectroscopic datasets of OIAP RAS (Obukhov Institute of Atmospheric Physics, Russian Academy of Sciences) for stations Moscow, Zvenigorod (ZSS, Moscow province), Zotino (ZOTTO, Central Siberia), Beijing (joint site of OIAP RAS and IAP CAS (Institute of Atmospheric Physics, Chinese Academy of Sciences)), SPbSU stations Peterhof and NDACC stations located in Eurasia were compared between themselves and with similar assessments obtained from satellite data. Significant decrease of anthropogenic CO in megacities Moscow (3.5±2.2%/yr) and Beijing (1.4±1.4%/yr) in autumn months of 1998–2016 were found according ground-based spectroscopic observations. In spite of total anthropogenic CO emissions decrease (for Europe and China) and absence of growth of wild-fires emissions in 2007–2016 we found that CO TC in background regions of Northern Eurasia has stabilized or increased in summer and autumn months of 2007–2016. Decrease of AOD over Central and Southern Europe and over China (1–5%/yr) was observed after 2007. Since 2007 an increase in CH_4 TC trends over Northern Europe as well as for tropical belt of Eurasia has been obtained. Analysis of satellite observations AIRS v6 of CO and CH_4 TC and MODIS AOD data confirmed the ground-based estimates of trends.

KEY WORDS: remote sensing, atmospheric spectroscopy, atmospheric composition, global changes, urban and background regions, Eurasia, trends

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INTRODUCTION

We present an analysis of recent changes in total content trends of important atmospheric compounds such as carbon monoxide (CO), methane (CH_4) and aerosols.

Carbon monoxide is one of major atmospheric pollutants. CO concentration largely determines the air quality in large and small cities (WMO/IGAC 2012; Elansky 2014; Elansky et al. 2018). Its emissions and concentrations are largely interrelated with anthropogenic and/or wildfires emissions and concentrations of other pollutants, as example aerosols (Rakitin et al. 2011; Golitsyn et al. 2015; Elansky et al. 2018). CO lifetime is relatively long, from 10 days in summer to 3 months in winter (Khalil et al. 1999; Novelli et al. 1998; Jacob 1999) and therefore it's a good tracer to investigate the long transport of pollutants from area with intensive sources to background regions. CO largely determines the OH and O3 concentrations and oxidation capacity of atmosphere (Jacob 1999).

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 (Myhre et al. 2013; Sonneman and Grygalashvyly 2014). Numerous works and researchers report about growth CH_4 concentrations after short period of stagnation since 2007 in almost all Earth regions; this growth is usually connected with climate changes and global warming processes. Some authors consider possible strong future methane emission growth especially in Polar regions from hydrates of Arctic shelf deposits (Shakhova et al. 2014). Also CH_4 is one of major sources of atmospheric CO (Jacob 1999).

Aerosols concentration is one of the most important characteristics of air quality. Their presence in atmosphere also influence on climatic characteristics through radiative forcing. Emissions of some kinds of aerosols: soot or black carbon, and smoke aerosols (Golitsyn et al. 2015) from fuel combustion and wild fires correlate with CO ones. Therefore, when studying the global changes in atmospheric composition we could expect an agreement in sign of CO and aerosol trends in urban and industry regions and regions of repetitive wildfires. The task of joint studying the trends of two tracers (CO and aerosols) in bulk atmosphere seems to be interesting. Both TC CO and AOD values characterize the condition of whole troposphere and their trends should poorly depend on the local sources emissions.

Numerous papers and reports devote to the about global and/or regional decrease of surface concentrations and total column (TC) of some atmospheric pollutants such as CO (Yurganov et al. 2010; WMO/IGAC 2012; Worden et al. 2013; Warner et al. 2013; Golitsyn et al. 2015), NO2 and aerosols (Hilboll et al. 2013; Coen et al. 2013; Chubarova et al. 2016), and simultaneous growth of concentration of GHGs CO_2, CH_4 etc. Climate changes take part simultaneously with changes of the atmospheric composition, both in background and urban areas (WMO/IGAC 2012; IPCC 2013).

Analysis of the CO total column (CO TC) trends in different regions of Eurasia for time-period of 1998 - 2014 was presented in our previous papers (Rakitin et al. 2011; Wang et al. 2014; Golitsyn et al. 2015; Wang et al. 2018). In preliminary results, we have paid an attention to CO TC positive trends for autumn months of 2007-2014 over rural outskirts of Moscow (ZSS, 53 km West from Moscow center) and Saint-Petersburg (Peterhof, 35 km to south-west from the St. Petersburg). In (Rakitin et al. 2016; Rakitin et al. 2017) the positive trends of CO TC were found for summer and autumn months of 2007–2015 over Russian and European spectroscopic observation stations (increase 0.5–3.6%/yr in dependence on site). This results were unclear for us, because CO global decrease from the beginning of 21-st century usually associates with reduction of anthropogenic emissions (WMO/IGAC 2012; IPCC 2013). Aim of the present study was update of our previous CO and CH_4 TC trends estimates for ground-based stations of OIAP RAS, SPbSU and NDACC and comparison of the CO trends distribution with the same for CH_4 and AOD based on satellite observations, for different Eurasian regions, seasons and time periods. In addition, we wanted to investigate a possible reason of background CO increase, perhaps connected with emissions from wildfires in Eurasia.

MATERIALS AND METHODS

OIAP RAS, IAP CAS and SPbSU measurements of TC CO and CH_4

The ground-based observation 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 observations station) by absorption spectroscopy method with using by identical grating spectrometers of medium resolution (0.2 cm-1), (Dianov-Klokov et al. 1989; Rakitin et al. 2011; Golitsyn et al. 2015). At Peterhof site the ground-based FTIR observations have been carried out by Saint-Petersburg State University (SPBSU), see details in (Poberovskii et al. 2011; Makarova et al. 2011). Specifics of observations and sites location please see at Table 1.

NDACC ground-based datasets of TC CO and CH_4

In this study, we also analyzed TC CO dataset from Thule, Kiruna, Harestua, Ny-Alesund, Bremen, Zugspitze and Jungfraujoch NDACC stations. Locations and specifics of stations are presented in Table 1. The details about the European NDACC stations could be found in (Senten et al. 2008; http://www.ndsc.ncep.noaa.gov/sites/).

Satellite AIRS datasets

Satellite data (product AIRS v6, Level 3 (L3), i.e. diurnal averaged for 1°x1° CO and CH_4 TCs, ascending data only for 2003-2016) were used to investigate spatial and temporal distributions of this species and their long-term variations (Aumann et al. 2003; Worden et al. 2013). MOPITT and AIRS CO datasets are the longest among the presently functioning orbital missions; AIRS advantage is possibility to measure both CO and CH_4 columns with frequency of 300-350 days per year for every cell 1°x1° against 50-60 measurements per year for CO only (MOPITT).

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, (Holben et al. 1998, 2001; http://aeronet.gsfc.nasa.gov/) and orbital MODIS Terra/Aqua 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 a largest number of daily data in comparison with L2.0.

Satellite MODIS datasets of AOD

MODIS/Terra and Aqua data are available from 2001 and 2003 years respectively.

MODIS AOD data aren't available for winter months for middle- and high-latitude regions; therefore AOD MODIS so called "annual" trends for all Eurasian regions excluding sub-tropical and tropical ones relate to season from April to October (approximately).

Before obtaining of satellite trends distribution the comparison between satellite data (diurnal means in spatial resolution 1^0x1^0 for AIRS CO and CH_4 total column and MODIS AOD products) and ground-based ones was produced (Rakitin et al. 2015; Rakitin et al. 2016; Wang et al. 2018). 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 another works (Worden et al. 2013; Anderson et al. 2013; Kim et al. 2016).

Trend evaluation method

Trends were calculated separately for seasonally/yearly averaged means for both ground-based and satellite data; estimates for every case are presented for linear type of approximation together with values of their 95% confidence intervals.

Results and discussion

Typical levels of anthropogenic CO and aerosols atmospheric pollution in Beijing are 2–5 times higher in comparison with Moscow ones, that is in good agreement with our previous results (Rakitin et al. 2011; Wang et al. 2014; Golitsyn et al. 2015). According to ground-based data the negative trends in CO TC were found for both megacities for different time-periods (decrease 1.7÷3.5%/yr for Moscow and 1.4÷2.3%/yr for Beijing in dependence on time-period and season), in spite of significant increase of motor vehicles in both megacities, see Fig. 1 and Table 2. Our estimates for megacities are confirmed by the conclusions of other reports (WMO/IGAC 2012; IPCC 2013; Warner et al. 2013; Worden et al. 2013, Elansky et al. 2018) about total reduction of CO anthropogenic emissions in most of world megacities.

In other hand the CO TC trends in summer and autumn seasons after 2007 year over background regions (OIAP sites ZSS, ZOTTO, SPbSU site Peterhof and Northern European NDACC stations) were positive

Table 1. Locations of ground-based spectrometers

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 (1) 2003–2016	Round the year, 70–90/70–90	55.7°/36.8°/200 m	OIAP RAS, Russia, Moscow province
Moscow (2) 2003–2016	Round the year, 70–90/ 0	55.7°/37.6°/200 m	OIAP RAS, Russia, center of Moscow
ZOTTO (3) 2008–2016	June–August, 10–15/0	60.8º/89.4º/120 m	OIAP RAS, Russia, Central Siberia
Peterhof (4) 2003–2016	Round the year, 60–80/ 50–70	59.88º/ 29.82º/20 m	SPbSU, Russia, Leningrad province
Beijing (5) 2003–2016	October–November, 15–20/0	39.97°/116.38º/80 m	IAP RAS, Russia, CAS, China, Beijing
Thule (6) 2003–2015	April–September, 20–70/10–50	76.53°/68.74°/30 m	NDACC, Greenland
Kiruna (7) 2003–2015	Round the year, 50–100/ 50–100	67.8º/20.4°/420 m	NDACC, Sweden
Harestua (8) 2003–2014	Round the year, 30–60/30–60	60.2°/10.8°/600 m	NDACC, Norway
Ny-Alesund (9) 2003–2014	March–October,20–40/20–40	78.9°/11.9°/15 m	NDACC, Norway, Spitsbergen
Bremen (10) 2003–2014	Round the year, 10–30/10–30	53.1°/8.8°/30 m	NDACC, Germany, industrial region
Zugspitze (11) 2003–2014	Round the year, 30–50/30–50	47.42°/10.98°/2964 m	NDACC, Germany, Alps
Jungfraujoch (12) 2003–2015	Round the year, 20–50/30–50	46.55°/7.98°/ 3850 m	NDACC, Switzerland, Alps

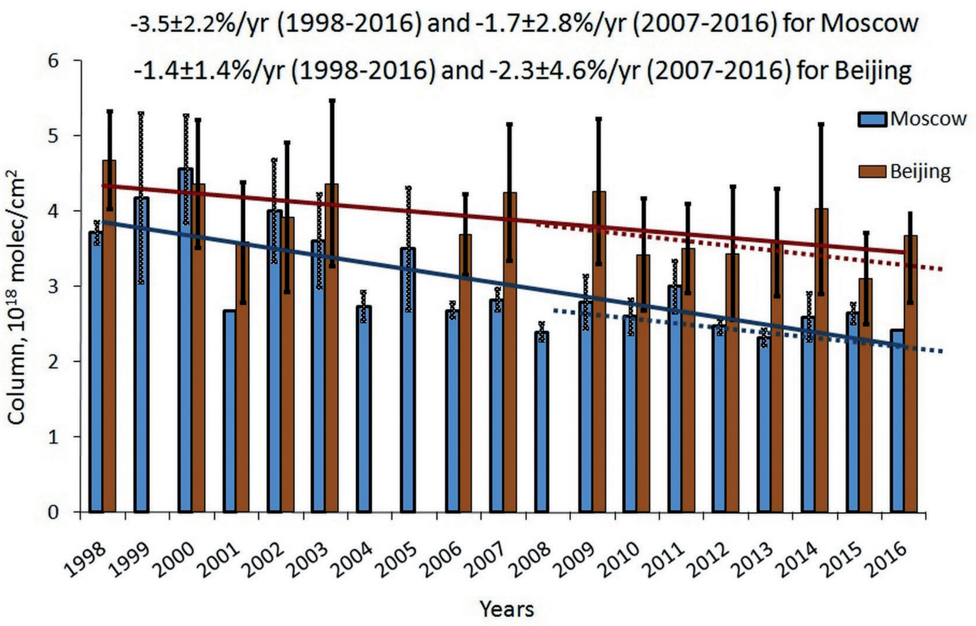

Fig. 1. CO TC means and trends for Moscow and Beijing (1998–2016, averaging for October-November)

(increase 0.4-2.4%/yr, see Fig. 2 and Table 2) according to both ground-based and satellite observation. Distributions of trends obtained from AIRS v6 satellite data is in fairly good agreement with the ground-based estimates (Table 2, Fig. 2).

According to AIRS v6 data an acceleration of CH_4 TC increase after 2007 was found for Northern Europe and tropical belt of Eurasia (Fig. 3). Significant seasonal differences in trends were not found for almost all of Eurasian regions, that confirms our previous results (Rakitin et al. 2017).

Table 2. CO total column trends for different time-periods according to annually and seasonally averaged data of ground-based and satellite measurements. Numbering of sites corresponds to Table 1. The positive seasonal average TC CO trends are marked by bold font

Ground-based observations: sites, years	Ground-based			AIRS		
	Season	Trend, %/year		Time periods	Trend, %/year	
		Season	Year		Season	Year
ZSS (1) 1998–2016 2003–2016 2007–2016	Sep-Nov	-1.25±1.51 **0.15±1.10** **0.94±1.26**	-1.33±0.76 -1.60±1.31 -0.58±1.87	2003–2016 2007–2016	-0.48±0.84 -0.21±1.54	-0.70±0.33 -0.46±0.52
Moscow (2) 1998–2016 2003–2016 2007–2016	Sep-Nov	-3.54±2.21 -2.53 ±2.39 -1.65±2.82	-2.32±0.98 -2.83±1.67 -2.11±2.47			
ZOTTO (3) 2008–2016	Jun-Aug	**~1.10**		2003–2016 2007–2016	0.50±2.06 **1.66±4.09**	-0.37±0.57 **0.02±0.96**

Peterhof (4) 1998–2016 2003–2016 2007–2016	Sep-Nov	-0.71±1.48 **0.20±0.87** **0.83±1.44**	-0.16±0.59 -0.37±0.81 -0.33±1.81	2003–2016 2007–2016	-0.39±0.72 0.26±1.04	-0.62±0.33 -0.26±0.43
Beijing (5) 1998–2016 2003–2016 2007–2016	Oct-Nov	-1.40±1.36 -1.96±2.67 -2.29±4.57		2003–2016 2007–2016	-0.81±0.84 -0.27±1.29	-1.03±0.44 -1.00±0.78
Zhule (6) 2003–2015 2007–2015	Jul-Sep	-1.13±2.88 **0.13±3.55**	-1.81±1.54 -1.34±2.93	2003–2016 2007–2016	-0.14±0.58 **0.26±1.04**	-0.34±0.27 -0.21±0.42
Kiruna (7) 2003–2015 2007–2015	Jul-Oct	-1.14±1.38 **0.47±1.33**	-1.16±0.74 -0.26±0.98	2003–2016 2007–2016	-0.48±0.75 **0.24±1.09**	-0.61±0.32 -0.27±0.38
Harestua (8) 2003–2014 2007–2014	Jul-Oct	-0.59±2.18 **1.56±3.07**	-0.79±1.29 **0.35±2.43**	2003–2016 2007–2016	-0.52±0.82 **0.10±1.27**	-0.72±0.35 -0.39±0.46
Ny Ales. (9) 2003–2014 2007–2014	Jul-Sep.	**0.17±2.15** **2.36±3.36**	-0.67±1.80 **1.07±1.72**	2003–2016 2007–2016	-0.06±0.63 **0.63±0.80**	-0.52±0.32 -0.30±0.49
Bremen (10) 2003–2015 2007–2015	Jul-Oct	-0.25±2.55 **2.08±4.68**	-0.05±1.18 -0.52±2.81	2003–2016 2007–2016	-0.57±0.68 -0.07±1.01	-0.79±0.34 -0.49±0.47
Zugspitze (11) 2003–2014 2007–2014	Jul-Oct	-0.07±1.11 **1.33±1.43**	-0.56±0.97 **0.17±1.85**	2003–2016 2007–2016	-0.63±0.71 -0.08±1.10	-0.85±0.40 -0.53±0.54
Jungfr. (12) 2003–2015 2007–2015	Jul-Oct	-0.30±0.84 **0.89±1.07**	-0.93±0.55 -0.32±0.96	2003–2016 2007–2016	-0.65±0.72 -0.04±1.08	-0.89±0.36 -0.58±0.47

According to AERONET and MODIS observations, AOD trends over Central and Southern Europe were negative (see Table 3 and Fig. 4), for all seasons (except winter) and time-periods. For Northern Eurasia AOD temporal changes were positive only in Central Siberia, that due to intensive Siberian wild fires of 2012, 2014 and 2015 years; also a decrease of AOD was obtained for China, Table 3 and Fig. 4. Our estimates for different regions are in good agreement with results of other reports and papers (IPCC 2013; Coen et. al. 2013) including regions of Moscow and Beijing (Chubarova et al. 2016; Wang et al. 2017).

Apparently from Table 2, the TC CO trends for all stations in the European part have changed a sign after 2007. Thus trends show values, which are more than 1% per year in the summer-autumn period (marked by bold font in Table 2) at the Harestua, Ny-Alesund, Bremen, Zugspitze stations. The trends also have changed at the ZSS, Peterhof, Jungfraujoch, Kiruna and Zhule stations, but the trend value is less expressed.

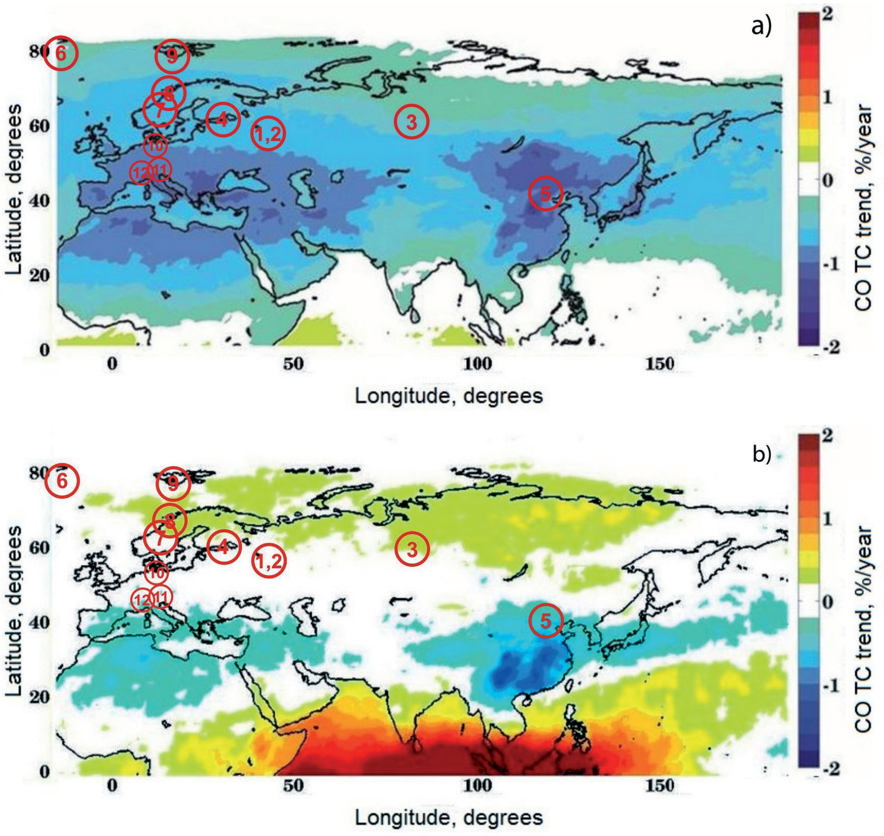

Fig. 2. Distribution of CO total column trends, according AIRS v6 data: a) - annual averaged values for 2003-2016; b) - autumnal averaged values for 2007-2016. Numbering of sites corresponds to Tables 1 and 2

CO TC and AOD positive trends after 2007 in summer months could be explained for some sites and regions by impact of Siberian wild-fires of 2012, 2014 and 2015 years. However in autumn months fires usually finish. CO life-time in troposphere varies from 1-2 weeks in summer to 3 months in winter (Novelli et al. 1998; Khalil et al. 1999; Jacob 1999). So, a significant impact of intensive summer Siberian wild-fires of 2012, 2014 and 2015 years or winter Malaysian ones (occurred in 2015) on atmosphere over Moscow and Saint-Petersburg outskirts in October-November as well as their influence on Northern European background regions seems improbable.

Therefore, such changes in summer and autumn CO trends cannot be explained by growth of wildfires impact at least in Europe where wild-fires emissions decreased for both time-periods 2007–2016 and 2003–2016 (Randerson et al. 2017; GFED v4.1; Rakitin et al. 2017). Possible reason, needing additional evidence is the changes in atmospheric photochemical system for example, can be an additional formation of carbon monoxide from methane that concentrations increased after 2007.

CONCLUSIONS

A significant decrease of CO total column and AOD in two large megacities Moscow and Beijing was found for time-period of 2007–2016. CO TC trends in summer and autumn months after 2007 changed their sign from negative to positive in almost all of background regions of Northern Eurasia. After 2007 an acceleration of increase of CH_4 TC was found for tropical and subtropical belt of Eurasia as well as for Northern Europe. Changes in AOD

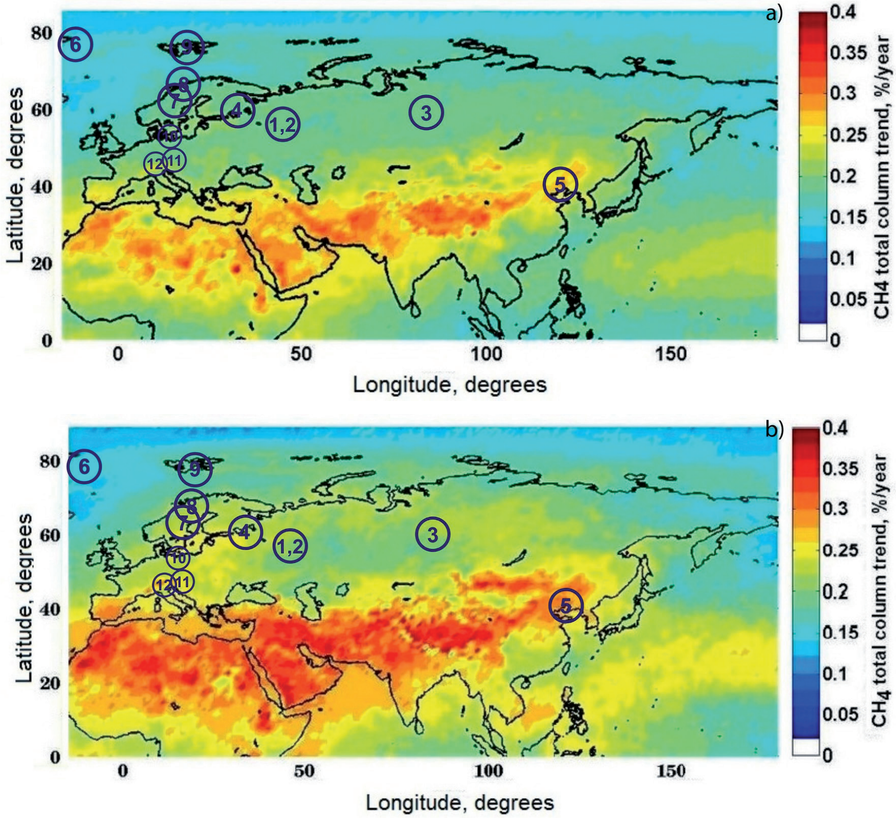

Fig. 3. Distribution of CH4 total column trends according to annually averaged AIRS v6 data: a) - for 2003–2016; b) for 2007–2016. Numbering of sites corresponds to Table 1 and 2

Table 3. Aerosol optical depth (AOD) trends according AERONET and MODIS observations (summer months and annual average, 2003–2015 and 2007–2015 time-periods)

Sites, years	AERONET		MODIS				
	Trend, %/year		Years	Trend,%/yr, Terra		Trend,% /yr, Aqua	
	Jun-Aug	Annual		Jun-Aug	Annual	Jun-Aug	Annual
Zvenigorod (1) 2003–2015 2007–2015	-0.5	-2.7	2003-2015 2007-2015	-0.3 -4.0	-1.7 -3.2	0.1 -4.7	-1.1 -5.6
Moscow (2) 2003–2015 2007–2015	-0.2 -3.4	-1.9 -4.1	2003-2015 2007-2015	-0.9 -5.1	-2.4 -4.4	-0.5 -4.5	-4.9 -1.2
Tomsk (3) 2003–2015 2007–2015		-2.3 -1.6	2003-2015 2007-2015	1.1 0.9	0.0 -1.2	1.0 2.3	-0.8 0.7

Irkutsk (4) 2004–2015 2007–2015	0.9	3.1 1.4	2004-2015 2007-2015	1.1 0.9	1.9 -2.8	0.5 -1.2	-0.4 -3.4
Yakutsk (5) 2003–2015 2007–2015	6.1 7.7	3.5 0.5	2003-2015 2007-2015	2.5 3.9	1.9 1.7	2.6 4.6	2.3 -0.03
Beijing_(6) 2003–2015 2007–2015			2003-2015 2007-2015	-0.5 -4.4	-0.1 -1.3	-0.6 -1.8	0.3 -1.3
XiangHe (7) 2004–2015 2007–2015	-2.0	0.2 -1.6	2003-2015 2007-2015	-0.4 -4.2	-0.1 -1.2	-0.6 -1.8	0.3 -1.3

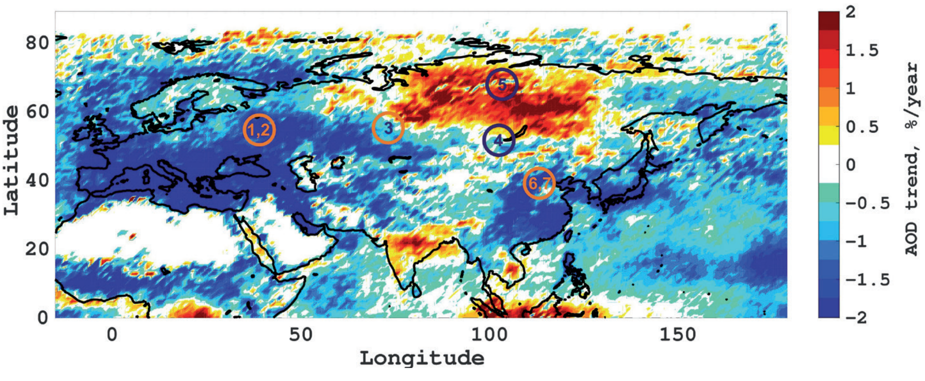

Fig. 4. Distributions of aerosol optical depth (AOD) trends (annual average) according MODIS/Terra for 2007–2016 years

trends for time-periods of 2000–2007 and 2007–2016 were insignificant. A significant decrease of AOD was obtained for almost all of Eurasian regions except of East Siberia, India and South-East Asia for both time-periods. Such pattern of changes in atmospheric composition especially in CO trends cannot be explained by growth of anthropogenic and/or wild-fires emissions. Possible reason of beginning of CO growth may be the change in the ratio of the natural sources and sinks with a significant role of atmospheric photochemical mechanisms.

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