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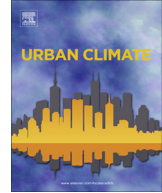
Air quality and CO emissions in the Moscow megacity

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A R T I C L E   I N F O

A B S T R A C T

Moscow with a population of 12.1 million people is the largest city in Europe. An up-to-date monitoring network organized in Moscow in the early 2000's has made it possible to estimate air quality and the concentrations of key pollutants emitted into the atmosphere. In this work, air-quality estimates obtained earlier were corrected using new observational data. As a result, Moscow ranks among clean megacities in the world. The emissions of CO were calculated on the basis of data on the surface concentration, vertical profile, and total column of CO. The surface concentration of CO was measured at 25 stations uniformly spaced on the territory of Moscow. The vertical profiles of CO were measured at the Ostankino TV tower. Data on the CO total column were obtained from long-term measurements using infrared solar absorption spectroscopy at two sites located in the center of Moscow and in Zvenigorod (53 km to the west of the center of Moscow). The annual emissions of CO from the Moscow megacity were estimated at 870 ± 200 Gg yr^{-1} for 1992–2008 and 680 ± 160 Gg yr^{-1} for 2002–2008. These values are slightly lower than those given for Moscow in the global inventories EDGAR and IPCC-AR-4.

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1. Introduction

Moscow is the northernmost megacity in the world. The territory of Moscow is officially bounded by the Moscow Automobile Ring Road (MARR) with a mean radius of approximately 17 km and it

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slightly extends beyond this boundary in both northern and southern directions (Fig. 1). The area of Moscow is officially about 1000 km^2, and, according to the 2002 census, its population was 10.3 million people. However, over the last 25 years, unbuilt areas inside and outside this ring road were rapidly developed. The territory of Moscow goes beyond the formal boundaries. It is almost circular in form with a mean radius of 24 km and an area of 1810 km^2. Such a structure of this megacity was supported by observational heat-island data obtained in the course of the 2000–2010 TROICA (TRanscontinental Observations into the Chemistry of the Atmosphere) expeditions using a mobile railroad laboratory (Elansky et al., 2009, 2012). This laboratory crossed the Moscow megacity in different directions, and obtained data showed that the air temperature and the concentrations of pollutants within the atmospheric surface layer significantly increased at a distance of, on average, 25–35 km from the center of Moscow (Elansky et al., 2014a). In 2005, the population of the Moscow megacity was approximately 12.1 million people, and the mean population density amounted to 6690 inhabitants km^{-2}. The Moscow megacity is located on a plain at minimum and maximum heights of 110 and 255 m above sea level, respectively.

In the late 1980s and early 1990s, because of the disintegration of the Soviet Union and economic crisis, most of Moscow industrial enterprises and almost all metallurgical and machine-building plants stopped working. The number of circulating vehicles decreased. In the late 1990s, the number of vehicles, especially private cars, started gradually to increase. From 1989 to 2011, the total number of cars registered in Moscow increased from 0.450 million to 4.5 million.

In the early 1990s, the State Air Pollution Monitoring (SAPM) network working in Russian cities partially ceased to operate. In Moscow about 10 stations continued to measure the surface concentrations of O_3, NO_2, SO_2, and aerosols. These measurements were taken from one to four times a day, which did not allow one to obtain reliable daily mean values. A large number of breaks brought uncertainties also in obtaining both monthly and yearly means. Because of corrections often introduced into the SAPM data, air-pollution estimates obtained on the basis of SAPM data significantly differed (e.g.,
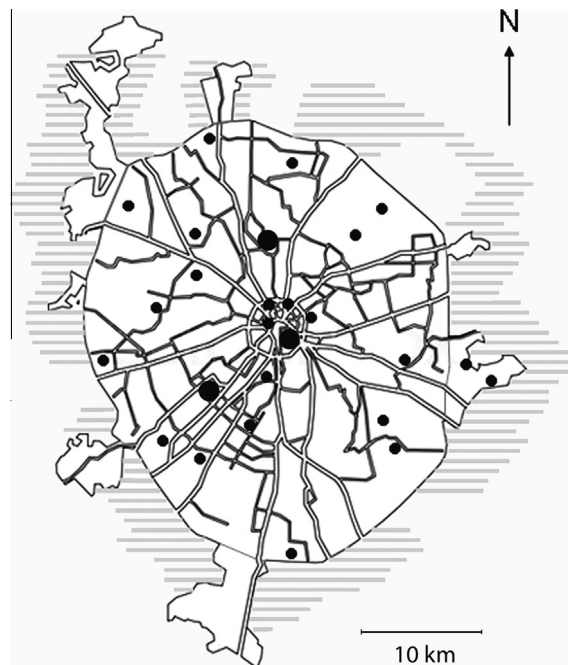
Fig. 1. The official territory of Moscow city and surrounding urban areas (shading) forming the megacity. Black circles mark the Mosecomonitoring sites. Large circles show the position of the Obukhov Institute of Atmospheric Physics (center), Ostankino tower (North), and Moscow State University (South–West).

Bezuglaya et al., 1993; UNCSD, 2001; Gurjar et al., 2008; Bezuglaya and Smirnova, 2008; State Report, 2009). Only in the 2000s, an up-to-date monitoring network was established in Moscow that made it possible to obtain reliable data for studying atmospheric pollution over Moscow. The estimates of air quality and CO emissions obtained for Moscow on the basis of these new data are given in this work.

2. Measurements

In the late 1990s, the Moscow Ecological Monitoring (MEM) network was organized by the Moscow Government's Department of Natural Resources. In the 2000s, the number of stations entering into this network increased up to 25. These stations are more or less uniformly spaced on the territory of Moscow (Bulletin, 2008; Gorchakov et al., 2011; Mosecomonitoring, 2011) (Fig. 1). The surface concentrations of air pollutants, including O_3, NO_x, CO, PM10 are measured at these stations. The concentrations of O_3, NO_x, and CO have been measured at different heights at the Ostankino TV tower since 2006.

Observations of the composition of the atmosphere are carried out at the ecological site of the Obukhov Institute of Atmospheric Physics (OIAP) in collaboration with the Meteorological Observatory of the Moscow State University (MSU station). This station is located on the territory of the University in southwestern Moscow (52.42° N, 37.31° E) at a distance of 7 km from its center (Fig. 1). The station is furnished with instruments to measure surface gas concentrations, aerosol properties, vertical air-temperature profile within a layer of 0–600 m, the integral content of NO_2 in the entire vertical atmospheric column and in the atmospheric boundary layer, and other parameters (Elansky et al., 2006, 2009, 2011). The periodicity of measurements varies from 1 min (O_3, NO_x, UV-radiation fluxes, and other parameters) to 5 min (NO_2 content in a vertical column).

The observations of atmospheric constituents are carried out at the OIAP building (OIAP station) located in the center of Moscow. The measurements of CO total column (CO TC) using infrared solar absorption spectroscopy have been taken since the late 1970s (Dianov-Klokov and Fokeeva, 1982). In the last few years, the integral content of NO_2 in the atmospheric boundary layer and both temperature and wind stratifications within a layer of 0–600 m have also been measured here using a sodar and microwave technique (Kallistratova and Coulter, 2004; Kadygrov and Pick, 1998; Kallistratova, 2008).

The vertical wind profile is regularly measured with sodars at two other sites located 1 km apart on the MSU territory (Yushkov, 2008; Lokoshchenko et al., 2012). The direct observations of air temperature and wind profile are carried out at the Ostankino TV tower (Bulletin, 2008; Mosecomonitoring, 2011).

Standard meteorological measurements in Moscow are taken at the MSU Meteorological Observatory and a few meteorological stations of the Russian Hydrometeorological Service (Isaev et al., 2003; Chubarova et al., 2011). The vertical temperature profile by microwave technique is measured at three sites: MSU and OIAP stations and in the town of Dolgoprudnyi located on the outskirts of Moscow (Kadygrov, 2009; Kuznetsova et al., 2004, 2012).

Outside the megacity, the composition of the atmosphere and its stratification are observed at the OIAP Zvenigorod station situated in a clean forest area 53 km to the west of the center of Moscow (55.70° N, 36.78° E). The longest CO TC data series has been obtained here since 1974. A detailed description of used instruments, methods of CO TC retrieving, and unique (in length) data series can be found in (Yurganov et al., 2002, 2011; Rakitin et al., 2011).

Additional data on the composition of Moscow's atmosphere and polluted air flows coming into and out of this megacity have been obtained due to observations from the mobile railway laboratory running around and through Moscow (TROICA experiments, Elansky et al., 2009, 2011, 2012, 2014a).

Thus, since the early 2000s, reliable data have been available, which make it possible to compare the air quality in Moscow with that in other megacities. Some published estimates of the atmospheric status in Moscow, which were obtained in the 1980s–1990s from irregular measurements often with uncalibrated instruments, can now critically be revised. The annual emissions of CO from the Moscow megacity are estimated on the basis of CO concentrations measured in the surface layer and at the Ostankino high-altitude TV tower and on the basis of CO TC measurements.

3. Air quality in the Moscow megacity

The concentrations of primary pollutants characteristic of Moscow's air basin are noticeably lower than those in most other megacities of the world. According to the 2002–2010 observational data obtained at the MSU station, the averaged (over the 2002–2010 period) concentrations of O_3, NO_2, CO, and SO_2 amount to 31.4, 41.6, 690, and 4.0 $\mu g\, m^{-3}$, respectively (in details the temporal variability of key gases is described by Elansky et al., 2014b). Some peak concentrations of polluting gases and aerosols against the general low mean background were observed only in the anomalously hot summers of 2002 and 2010, and they were caused by both forest and peatbog fires in the vicinity of Moscow (Elansky et al., 2011). However, even under these conditions, the concentration of ozone – the main indicator of air pollution and photochemical activity – with maximum hourly mean values of 268 $\mu g\, m^{-3}$ for 2002 and 255 $\mu g\, m^{-3}$ for 2010 did not exceed its highest levels for such megacities as Mexico (576 $\mu g\, m^{-3}$), Sao-Paolo (403 $\mu g\, m^{-3}$) Bogota (348 $\mu g\, m^{-3}$), and New York (272 $\mu g\, m^{-3}$) (see Bang Quoc Ho, 2012). In other years, the maximum concentrations of O_3 in Moscow did not exceed 98 $\mu g\, m^{-3}$.

The O_3, NO_2, CO, and SO_2 concentrations averaged over the 2002–2008 period of observations in the surface air layer over the MEM and MSU stations amount to 32.4, 40.2, 790, and 4.4 $\mu g\, m^{-3}$, respectively (Mosecomonitoring, 2011; GAW, 2012; Elansky et al., 2014b). In addition to the MSU station, the concentration of O_3 was measured at 13 MEM stations located within the Moscow megacity, the concentrations of CO and NO_2 were measured at 24 stations, and the concentration of SO_2 was measured at 8 stations. All mean concentrations of the pollutants for the MSU and MEM stations almost coincide except for significantly higher CO mean concentrations obtained at some MEM stations which are located in the vicinity of highways.

In 1993, in the center of Moscow (OIAP station), during the period of both transport and industrial collapses, the annual mean concentrations of O_3 and NO_2 amounted to 38.7 and 48.2 $\mu g\, m^{-3}$, respectively (Elansky and Smirnova, 1995); these values are close to those averaged over the 2002–2010 period. In 1965–1990, the SAPM network recorded as a rule higher NO_2 surface concentrations. The highest NO_2 concentration 87 $\mu g\, m^{-3}$ was recorded in 1990 just before the economic collapse (Bezuglaya and Smirnova, 2008). Since 1992 the annual mean concentration of NO_2 obtained at the SAPM network did not exceed 45 $\mu g\, m^{-3}$ up to 2008 (State Report, 2009).

Gurjar et al. (2008) proposed a multi-pollutant index (MPI) considering the combined level of the three criteria pollutants NO_2, SO_2, and TSP in view of the WHO Guidelines for Air Quality (2000). Using this index, they estimated the quality of ambient air in 18 megacities and ranked them by their pollution level. Moscow proved to be among the most polluted megacities of the world, and the annual mean concentration of NO_2 (170 $\mu g\, m^{-3}$) for Moscow significantly exceeded that for the rest of megacities. This absolutely unreal concentration of NO_2 and the annual concentration of SO_2 (15 $\mu g\, m^{-3}$) were taken from M.S. Myagkov's personal communication.

Table 1, which is a corrected version of the table from (Gurjar et al., 2008), gives the NO_2, SO_2, and PM10 concentrations averaged over the 2002–2008 observation period according to data obtained at the MSU station and the MEM network. Table 1 also gives the new MPI values calculated for Moscow using the formula from (Gurjar et al., 2008):

$$\text{MPI} = 1/n \left\{ \sum_{i=1}^{n} [(\text{AC}_i - \text{GC}_i)/\text{GC}_i] \right\}$$

where AC_i is the concentration of the i-th pollutant, GC_i is the maximum concentration according to WHO recommendations (2000), and n is the number of pollutants used in calculations.

The MPI was calculated for TSP = 150 $\mu g\, m^{-3}$. This value seems to be extremely high. Analyzing multiyear observational data obtained at the SAPM, Bezuglaya and Smirnova (2008) showed that Moscow is one of the cleanest industrial cities in Russia in the 1990–2000s, the annual mean TSP for Moscow did not exceed 50 $\mu g\, m^{-3}$, while the TSP averaged over this period for Russian cities was about 120 $\mu g\, m^{-3}$ (the same results are presented in State Report, 2009).

However, because, during this period, only a few stations operated in Moscow and the requirements for the quality of data were not strict, the TSP means obtained at the SAPM network for the

Table 1

Annual mean concentrations of SO$_2$, NO$_2$, particle masses (TSP and PM10), MPI, and rank for megacities. These megacities are ranked by air quality on the basis of recalculations with consideration for real observational data obtained at the MSU and MEM stations. Rank given by Gurjar et al. (2008) is shown in square brackets.

Megacity	Population in mln, (2005)	TSP (PM10), μg m^{-3} [rank]	SO$_2$, μg m^{-3} [rank]	NO$_2$, μg m^{-3} [rank]	MPI	Rank (Gurjar et al., 2008)
Tokyo	33.4	40 [16]	19 [10–13]	55 [10]	−0.27	17 [16]
Seul	23.1	84 [14]	44 [5]	60 [7]	0.10	12 [–]
Mexico	22.0	201 [10]	47 [4]	56 [9]	0.52	9 [10]
New-York	21.8	27 [19]	22 [8]	63 [6]	−0.23	15 [14]
Mumbai	21.1	243 [9]	19 [10–13]	43 [13]	0.39	10 [11]
Delhi	20.8	405 [4]	18 [14–15]	36 [16]	0.92	6 [7]
São Paulo	20.3	53 [15]	18 [14–15]	47 [11]	−0.29	18 [17]
Shangnai	18.6	246 [8]	53 [3]	73 [4]	0.87	7 [8]
Los Angeles	17.9	39 [17]	9 [17]	66 [5]	−0.25	16 [15]
Jakarta	16.9	271 [7]	35 [7]	120 [2]	1.24	5 [5]
Osaka Kobe	16.6	34 [18]	19 [10–13]	45 [12]	−0.37	19 [18]
Cairo	15.8	593 [1]	37 [6]	59 [8]	1.93	3 [3]
Kolkata	15.4	312 [6]	19 [10–13]	37 [15]	0.59	8 [9]
Karachi	14.6	668 [1]	13 [18]	30 [17]	1.81	4 [4]
Dhakka	13.6	516 [3]	120 [1]	83 [3]	2.40	1 [1]
Buenos Aires	13.5	185 [11]	20 [9]	20 [18]	−0.01	13 [13]
Beijing	12.4	377 [5]	90 [2]	122 [1]	2.01	2 [2]
Rio de Janeiro	12.2	139 [13]	15 [16]	60 [7]	0.11	11 [12]
Moscow, 2000	11.9	150 [12]	5.1 [19]	38.4[14]	−0.09	14 [6]
Moscow, 2000	11.9	35 (PM10)	5.1 [19]	38.4 [14]	−0.06	14[6]
Moscow, 2002–2008	12.1	32 (PM10)	4.4 [19]	40.2 [14]	−0.10	14 [6]

Note: The population of the megacities for 2005 is taken from the CIESIN, 2005; the MSU and MEM stations data are averaged over the 2002–2008 period and are extrapolated to 2000. Data on the masses of all particles (TSP) (from Gurjar et al., 2008) and particles with a size of less than10 μm (PM10) (from observations at the MSU and MEM stations) were used in calculations of the MPI for Moscow. The WHO standards (GC$_i$): TSP = 90 μg m^{-3}, PM10 = 20 μg m^{-3}, SO$_2$ = 50 μg m^{-3}, and NO$_2$ = 40 μg m^{-3} (WHO, 2000).

entire territory of Moscow seem unreliable. For example, Bezuglaya and Smirnova (2008) gave for Moscow the annual mean TSP = 27 μg m^{-3} for 2006, which is noticeably lower than the mean of PM10 for Moscow in the same year. Therefore, in Table 1, we left TSP = 150 μg m^{-3} (used by Gurjar et al., 2008) as the highest possible value for 2000.

The measurements of PM10 began in 2002 at the MSU station and in 2004 at the MEM network. The mean value of PM10 obtained at the MSU and MEM stations for 2004 amounted to 33 μg m^{-3} (Gorchakov et al., 2007). In the following years, the value of PM10 slowly decreased. The mean value of PM10 for Moscow (10 stations) amounted to 28 μg m^{-3} for May 2007–April 2008 (Bulletin, 2008) and 25 μg m^{-3} for 2009 (Gorchakov et al., 2011). The PM10 value obtained at the MSU and MEM stations and averaged over the 2002–2008 period amounts to 32 μg m^{-3}. Since Gurjar et al. (2008) gave air-quality indices for megacities for 2000, the PM10 value was also linearly extrapolated to 2000. Taking into account the fact that there were no rapid changes in the infrastructure of Moscow in the 2000s, the PM10 extrapolation to 2000 yields approximately 35 μg m^{-3}.

It follows from a comparison of the MPIs for the megacities that, after corrections, Moscow rapidly changed its ranks by some indicators and by the air-pollution index. Correspondingly, the ranks of the rest of the megacities also changed. Additional changes were caused by the inclusion of Seoul in Table 1, whose MPI was calculated using data from Bang Quoc Ho (2012).

The distinctive feature of Moscow is that the content of SO$_2$ in its air basin is low. By this indicator, among all 19 megacities, Moscow is the cleanest city (rank 19). This is due to complete electrification and gasification of the domestic sector as well as gasification of central heating and replacement of many boiler rooms by new three up-to-date thermal power plants (TPPs) during the last two decades. By the NO$_2$ content, Moscow also moved from the 1st place to the 14th one. In spite of the fact that Moscow's autopark has rapidly increased in the last years, no more than 60% of vehicles participate in the traffic on the roads because of a large number of vehicles formally registered but not circulating

throughout the city and a low road capacity. The replacement of outdated vehicles by new ones with a decreased exhaust of NO_x also favored the stabilization of pollutant emissions from vehicles in the 2000s.

Among the three components, the mass aerosol concentration (TSP or PM10), which significantly exceeds the WHO standard, makes the largest contribution to the pollution of Moscow's air basin. The sources of aerosols are heavy traffic, dirt roads, waste grounds without vegetation, and industrial zones many of which are abandoned.

The air-pollution index calculated for Moscow (using TSP = 150 μg m^{-3} and corrected SO_2 and NO_2 concentrations) amounts to −0.09 for 2000. Thus, among other megacities of the world, Moscow proves to be on the 14th instead of the 6th place by air pollution (Table 1). If PM10 is used instead of TSP, the pollution index increases up to −0.06, and Moscow remains on the 14th place. In the following years, the urban-air composition varied only slightly. On average, over the 2002–2008 period, the MPI decreased to −0.10. When compared to other megacities (for 2000), this value also corresponds to the 14th place (it is evident that some other megacities could have also improved their air quality in the 2000s). Surely the concentration of CO should also be taken into account in calculating the MPI. However, it is a difficult task to collect consistent data on the world's megacities for this purpose. Since we have the MSU and MEM stations, at which the atmospheric content of CO is continuously measured, we calculated CO emissions on the basis of observational data on the surface concentration of CO (methodology 1) and on the total content of CO in an atmospheric vertical column (methodology 2).

4. Estimating the emission of CO from measurements of its surface concentration

Fig. 2 gives the yearly average concentrations of CO obtained at the MSU and MEM stations over the May 2007–April 2008 period versus distance from the center of Moscow (Bulletin, 2008; Mosecomonitoring, 2011). Such a spatial distribution of CO concentrations is characteristic of Moscow over the entire 2002–2008 period. The concentrations of CO vary mainly from 0.60 to 1.0 mg m^{-3}. Their mean value (for 2002–2008) is 0.79 mg m^{-3}. The minimum CO concentration 0.15 mg m^{-3} was recorded at two stations located in the northwestern sector outside the Moscow megacity's boundaries (these stations are not shown in Fig. 1).

It follows from Fig. 2 that, in the distribution of CO concentration there is no maximum over Moscow's center. The Moscow megacity located on a plain has a good natural ventilation, and enhanced CO concentrations are observed when air flows leave Moscow but not over its center. This was clearly confirmed by the results of measurements taken from the mobile railway laboratory running around the megacity (TROICA experiments) (Elansky et al., 2010). At the same time, no increase in the CO concentration is noticeable in the eastern direction, although westerlies are dominating. This is explained by the fact that industrial enterprises are located mainly on the eastern periphery of Moscow. Their emissions, in general, slightly affect the air quality in the megacity under westerlies, but they significantly increase the concentration of CO in Moscow's western sector under easterlies. Due to this, the spatial distribution of annual mean CO concentrations can be considered almost uniform.

According to multiyear observational data obtained at the Meteorological Observatory and Moscow's meteorological stations, the annual mean wind velocity in an atmospheric surface column (at a height of 10 m) is 2.5 m s^{-1} (Isaev et al., 2003). The measurements of vertical wind profiles with a sodar and at the Ostankino TV tower yield slight wind changes up to heights of 150–200 m. At these heights, the yearly mean wind velocity ranges within 3.5–4.5 m s^{-1}. Above this level, both daily and seasonal variations increase, and the wind velocity increases approximately up to 6 m s^{-1} at a height of 300 m (Yushkov, 2008; Lokoshchenko et al., 2012).

The vertical distribution of CO in the urban atmosphere is determined by vertical thermal stratification and mixing processes. Their basic characteristics are given in (Kuznetsova et al., 2004, 2012; Yushkov and Kouznetsova, 2008) on the basis of the 2000–2009 temperature profile measurements. In winter, the heat island over Moscow propagates only up to a height of 100 m. The thermal stability in the center of Moscow is significantly weaker than on its outskirts. The temperature gradient γ within an air layer of 0–100 m exceeds the dry-adiabatic gradient γ_a in 88% of cases for the center
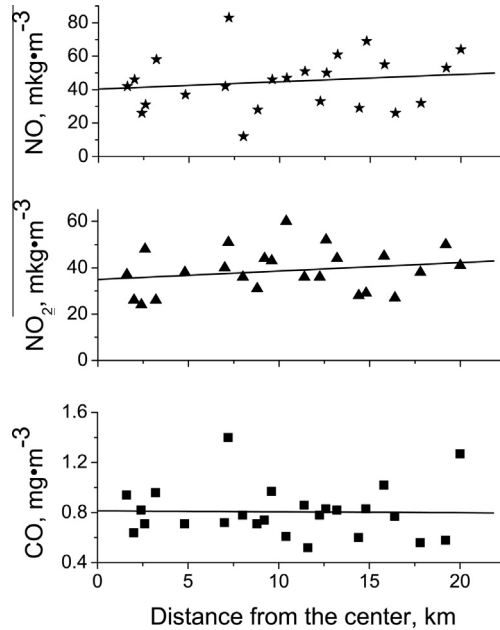
Fig. 2. Annual mean (May 2007 – April 2008) surface NO, NO_2, and CO concentrations at the MEM and MSU stations versus the distance from the center of Moscow.

of Moscow and in 45% of cases for its outskirts. The recurrence of temperature inversions amounts to 2% for the center of Moscow and 25% for its outskirts. Thus, air mixing occurs mainly within a layer of 0–100 m and more intensively over the center of Moscow. In spring, the mean depth of the air layer affected by the Moscow megacity is 150 m. The air mixing is more intensive than in winter. In summer, the urban effect spreads, on average, to a height of 250–300 m. The atmospheric surface layer over the center of Moscow is weakly stable and close to isothermal, however, its stability increases towards Moscow's outskirts. The recurrence of surface inversions is 3% for the center and 32% for outskirts. In fall, windy weather dominates, and the difference between data obtained in the center of Moscow and on its outskirts decreases when compared to that for the rest of the seasons. Unstable stratification still persists in the center, and stable stratification dominates on outskirts. Outside Moscow's boundaries, air mixing occurs only in the daylight hours and for a short time period. Here, the mixing-layer height is 100–150 m.

The annual mean temperature stratification of the boundary layer over Moscow is characterized by instability dominating over its center. Here, intensive air mixing occurs throughout an atmospheric layer extending, on average, from the surface up to a height of 250–300 m. The vertical air mixing becomes less intensive from the center of Moscow towards its outskirts, where it occupies a layer extending, on average, from the surface up to a height of 220 m.

The same results were obtained from an analysis of pollutant-concentration data obtained from measurements at the Ostankino TV tower. Fig. 3 gives the monthly mean concentrations of CO, NO, and NO_2 that were measured at heights of 10, 130, 248, and 348 m from May 2007 to April 2008 (Bulletin, 2008; Mosecomonitoring, 2011). The maximum surface concentrations of CO and NO for August–October coincide with the peak of traffic intensity before the first fall of snow in November. The NO_2 maximum in September–October is weakly pronounced because of seasonal minimum of the surface concentration of ozone, on which the photochemical formation of NO_2 is dependent (Zvyagintsev et al., 2010a,b; Elansky et al., 2014a,b). Both CO and NO concentrations are significantly lower at a height of 130 m. During the cold season, the concentration of CO decreases to its almost background values. At heights of 130 and 248 m, the concentrations of both CO and NO and their
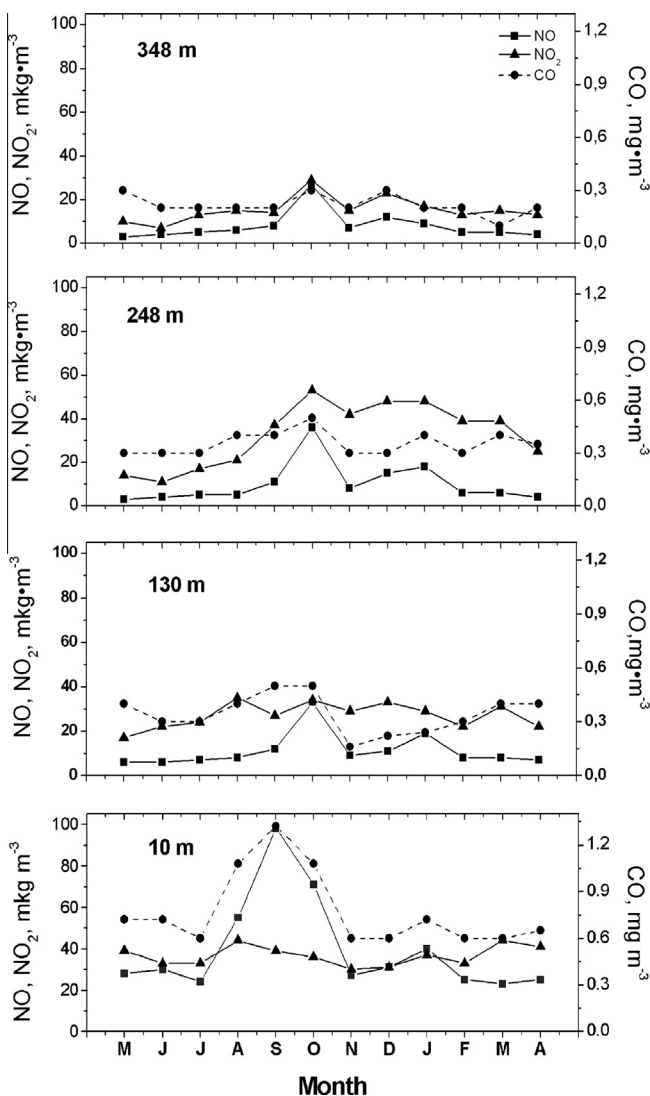
Fig. 3. Monthly mean concentrations of NO, NO$_2$, and CO at Ostankino tower (levels 10, 130, 248, and 348 m) between May 2007 and April 2008.

seasonal variability are almost the same. Thus, within a layer of 130–248 m, the height distribution of these pollutants is almost homogeneous throughout the year. Above 248 m, both CO and NO concentrations decrease.

During the cold season, the NO$_2$ concentration is very high at a level of 248 m. This may be caused by nitrogen oxides emitted from the pipes of thermal power plants during the heating season (October–mid-April). The basic city demands for heat are provided by 15 TPPs running on natural gas. The height of their pipes ranges between 100 and 200 m, and, thus, the products of combustion of natural gas (NO and, to a less extent, NO$_2$ and CO) are emitted into the atmosphere above the basic mixing layer. Very high concentrations of NO$_2$ at a height of 248 m during the heating season imply that at this layer ozone is undepleted and an active transformation of NO into NO$_2$ takes place, i.e., the height 248 m is close to the upper boundary of the urban effect on atmospheric composition.

Both seasonal and annual mean vertical CO, NO, and NO$_2$ profiles obtained at the Ostankino TV tower are given in Fig. 4. Seasonal differences in the profiles of CO vanish away at a height of about 200 m, and a rapid decrease in its concentration occurs above 248 m. Such a structure of the vertical CO profile completely corresponds to the mixing conditions which follow from an analysis of temperature stratification. The NO profile also demonstrates good mixing up to heights of 100–150 m, however, during the cold season, intensive emissions from TPPs exert their influence up to a height of 348 m. The same is true for the NO$_2$ profile, but, in addition, it shows a weak destruction of O$_3$ at heights between 130 and 248 m during the cold season, which implies that, during this season, the atmosphere in this layer becomes immune from urban effects. Thus, one may conclude that, for Moscow, within a year, the upper boundary of the atmospheric layer, within which CO of urban origin is concentrated, is at a height of 180–250 m. The annual mean position of this upper boundary is approximately at a height of 220 m. This relatively small thickness of the air layer affected by the Moscow megacity is associated with the location of Moscow on a plain at a high latitude of 55.6° N and with a low annual mean temperature of 6.6 °C. Even under the conditions of anticyclonic blocking and smoke generation by peatbog fires in the anomalously hot summer of 2010, the thickness of the atmospheric polluted layer over Moscow's outskirts amounted to no more than 340 m (Yurganov et al., 2011).

Table 2 gives the annual means of the basic parameters and their uncertainty limits used in calculating CO emissions from Moscow for 2002–2008. The territory of Moscow is almost a circle with a mean diameter of 48 km. Within the mixing layer, whose annual mean height is assumed to be 220 m, the vertical distribution of CO is considered uniform. The mean (over 2002–2008) surface concentration of CO amounts to 0.79 mg m^{-3}. As the minimum (background) concentration of CO outside the megacity is about 0.15 mg m^{-3}, the urban contribution is assumed to be 0.64 mg m^{-3}. By a mean wind velocity of 3.7 m s^{-1} in a layer of 0–220 m, the time during which air masses remain over the megacity amounts to approximately 3.6 h. On scales of a few hours, CO can be treated as a conservative pollutant. From these conditions it follows that the yearly average CO emission per unit of urban area amounts to 342 Mg km^{-2} yr^{-1}. The probable uncertainty limits vary between 225 and 490 Mg km^{-2} yr^{-1}. The yearly mean CO emission from the entire area of Moscow megacity amounts to 620 Gg yr^{-1} at an uncertainty of 475–760 Gg yr^{-1}. Minimum values of the integral emission of CO and its emission per unit area were obtained for the largest size of the megacity 52 km in diameter and a minimum wind velocity of 3.4 m s^{-1} and their maximum values were obtained for the megacity with a diameter of 44 km and a wind velocity of 4.0 m s^{-1}.
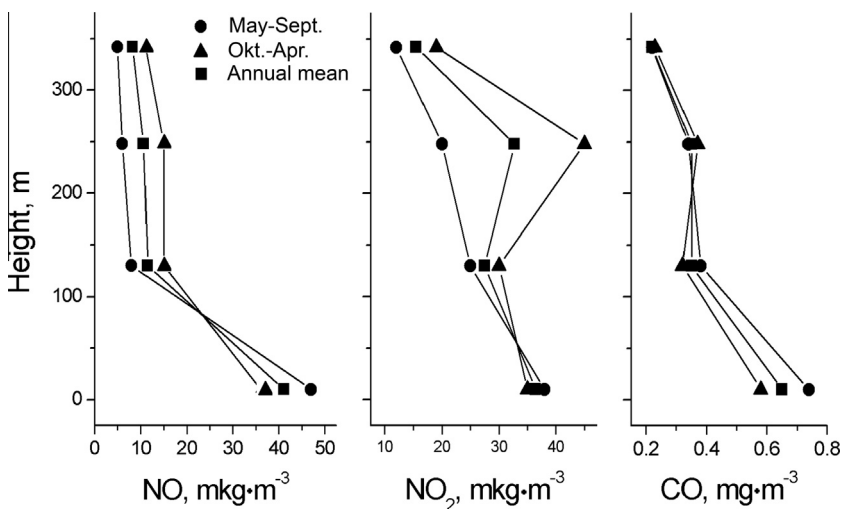

Fig. 4. The average vertical profiles of NO, NO$_2$, and CO concentrations at Ostankino tower for warm and cold seasons, and for the whole period of observations between May 2007 and April 2008.

Table 2
Annual mean CO emission from Moscow megacity, parameters for its calculation, and their uncertainties.

No.	Parameter	Mean values	Limits of uncertainty
1.	Diameter of megacity, km	48	44–52
2.	Area of megacity, km^2	1810	1520–2123
3.	Height of mixing layer, m	220	180–250
4.	Mean wind velocity in mixing layer, m s^{-1}	3.7	3.4–4.0
5.	Time of air mass exchange in the city, h	3.60	3.05–4.25
6.	Urban contribution to CO concentration, mg m^{-3}	0.64	0.60–0.68
7.	CO emission per unit area, Mg km^{-2} yr^{-1}	342	225–490
8.	Integral CO emission from Moscow megacity, Gg yr^{-1}	620	475–760

## 5. Estimating CO emissions from column measurements

The CO total content in a vertical column of the atmosphere (CO TC) over the Moscow megacity has been measured using infrared solar absorption spectroscopy at the OIAP station (Moscow's center) since 1992 (Rakitin et al., 2011). This methodology was proposed by Stremme et al. (2013) for estimating CO emissions from Mexico City. The methodology is not sensitive to the depth of mixing layer, when compared to that described above.

To estimate the spatial distribution of CO emissions and their diurnal evolution, we used CO TC data (Zvenigorod station), data on the integral content of CO in the atmospheric boundary layer (Ostankino TV tower), and data on the diurnal variations of CO surface concentration (MSU station).

In megacities, daily variations in CO depend on CO emissions from urban sources, inhomogeneous spatial CO distribution, turbulent diffusion, and air advection. Uncertainties in estimating the influence of these factors were analyzed by Stremme et al. (2013) for Mexico City. The greatest uncertainty exists with respect to the effect of turbulent diffusion, because it is difficult to quantitatively estimate its contribution. However, one can minimize this contribution by choosing an optimal observation period. For Mexico City, such a period falls between 11:15 and 13:15 (LT), when turbulence is not yet developed, the wind velocity is low, and the influence of the inhomogeneous spatial distribution of CO is minimal due to the Sun's high position. In Moscow with its densely built-up area, large-scale enterprises standing almost idle since the early 1990s, the transport sources of CO are basic. Because of a radial structure of Moscow's road network, the transport emissions are more or less uniformly distributed over the megacity. The thickness of a polluted air layer over Moscow is much less than that over Mexico City (Molina and Molina, 2002; Molina et al., 2010). Therefore, measurement results are less sensitive to changes in the sunbeam azimuth. Moreover, in averaging a large data volume, CO TC variations caused by the advection of air masses from different directions can be treated as noise with a higher probability than in the case of Mexico City. Due to all these circumstances, the morning hours 8:30–10:30 (LT) are most favorable for measuring the CO TC and estimating the CO emission for Moscow.

Rakitin et al. (2011) gives the averaged diurnal evolution of CO TC for Moscow for the 1992–2008 and 2003–2008 observation periods. The main body of data was obtained under solar anticyclonic conditions and on weekdays only. For low wind velocity, undeveloped convection, and weak turbulent mixing during these morning hours (8:30–10:30), the direct emissions of CO determine the CO TC growth. In fact, the CO TC growth is not linear, because the influence of advection does not vanish completely. Approximating the growth rate of CO TC by the linear function yields an error of about 15%. The mean growth rate of CO TC between 8:00 and 10:00 amounted to $(0.175 \pm 0.025) 10^{18}$ mol cm^{-2} h^{-1} for 1992–2008 and $(0.135 \pm 0.020) 10^{18}$ mol cm^{-2} h^{-1} for 2003–2008. This growth rate of CO TC corresponds to urban emissions for the place of measurements: 81.2 ± 14.5 kg km^{-2} h^{-1} for 1992–2008 and 62.5 ± 11.2 kg km^{-2} h^{-1} for 2003–2008.

In order to extrapolate hourly emissions to the whole day, it is necessary to know the CO TC diurnal cycle. Measurements of CO TC are taken only during daylight hours; therefore, in order to estimate the diurnal evolution of the emissions, one can use observational data on the surface concentration of CO and its vertical profile, obtained at the Ostankino tower. Fig. 5 gives the annual mean diurnal cycle of
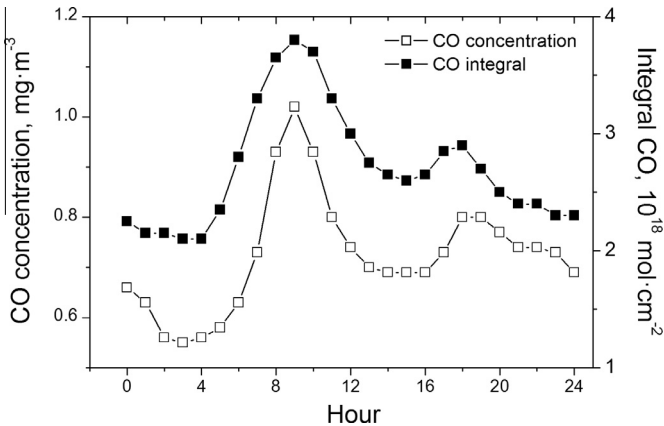
Fig. 5. The average diurnal variations of surface CO concentration at the MSU station in 2002 and of integral CO content in 0–348 m layer at Ostankino tower (May 2007 – April 2008).

CO for 2002 taken from Elansky et al. (2006). The diurnal CO variations observed in Moscow coincide completely with those observed in Mexico City (Stremme et al., 2013) except absolute values. Since we have the integral content of CO in a layer of 0–348 m (Fig. 5), we can decrease an error in estimating the temporal extrapolation factor proposed by Stremme et al. (2013). As a result, the emission of CO per day exceeds the emission per hour (from 9:00 to 10:00) by 17.9 times. The error of the extrapolation was estimated at 10% using the methodology proposed by Stremme et al. (2013). Thus, for the given observation site, the daily CO emission was estimated at 1450 ± 315 kg km^{-2} day^{-1} for 1993–2008 and 1120 ± 240 kg km^{-2} day^{-1} for 2003–2008. Extrapolating these values to annual ones, we have 530 ± 115 Mg km^{-2} yr^{-1} for 1992–2008 and 410 ± 90 Mg km^{-2} yr^{-1} for 2003–2008. This latter extrapolation does not introduce an additional random error, because the initial values of the CO TC growth rate within a time period of 8:30–10:30 were obtained from its annual mean.

The CO TC was measured at the OIAP station on working days. An analysis of the results of continuous observations of the surface concentration of CO and its vertical profile at the Ostankino tower yields a 6% annual mean decrease in the CO TC for Sundays when compared to other days of week. Uncertainties associated with the effect of Sundays introduce an error of no more than 2% into the estimate of the annual emission of CO. After introducing corresponding corrections, the density of CO flux over the center of Moscow amounts to 525 ± 115 Mg km^{-2} yr^{-1} for 1992–2008 and 405 ± 90 Mg km^{-2} yr^{-1} for 2003–2008.

A daily CO-emission flux was obtained for the center of Moscow. In order to extend its values to the entire megacity, it is necessary to know the spatial distribution of CO TC. Since satellite measurements (Yurganov et al., 2011) underestimate the CO content in a highly polluted surface air layer over Moscow (the same conclusion for NO$_2$ was drawn by Elansky et al. (2011)), we estimated this distribution using data on the surface concentration and integral content of CO in an atmospheric layer of 0–348 m. The features of the megacity's infrastructure were also taken into account. As was shown above, the spatial distribution of CO concentration inside the MARR was, on average, almost homogenous (Fig. 2). Good agreement between the values of the integral content of CO in the boundary layer (Ostankino tower) and CO TC (Moscow's center) leads us to the same conclusion.

The results of comparison of CO TC data simultaneously obtained in Moscow and Zvenigorod makes it possible to estimate the size of region affected by urban CO sources. Rakitin et al. (2011) showed that the effect of Moscow's CO sources is pronounced at Zvenigorod only in 3% of the total number of observation days, and they came to the conclusion that Zvenigorod is a background station.

There is good agreement between CO TC data obtained from both satellite and ground-based measurements at Zvenigorod (Yurganov et al., 2011). Since the satellite measuring area $1° \times 1°$ was located at a distance of 30–100 km to the west of Moscow's center, one can assume that the boundary

Table 3

Averaged annual CO emissions per unit area and from the entire area of the Moscow megacity for different observation periods.

Observational data	CO surface concentration	CO total column	
Period of observation	2002–2008	1992–2008	2003–2008
Emission per unit area (Mg km^{-2} yr^{-1})	340 ± 120	550 ± 120[*]	425 ± 95[*]
Total emission (Gg yr^{-1})	620 ± 150	870 ± 200	680 ± 160

[*] Local annual CO emission flux per unit area in Moscow's center.

of Moscow's influence is located between Moscow and Zvenigorod, on average, at a distance of 30–35 km from Moscow's center. Studying the degree of changes in atmospheric composition over the Moscow region from the mobile railway laboratory (TROICA experiments) yielded the same results (Elansky et al., 2014a). In addition, it was shown that the concentration of CO over Moscow's territory bounded by the MARR is close to constant. Outside the MARR CO TC decreases in conformity with changes in both building and highway densities. Therefore, we can assume that, outside the MARR (with a mean radius of 17 km), the urban contribution to the CO TC linearly decreases by 1/3 at the megacity's boundary which is 24 km away from the center. In this case, the territory over which the CO TC exceeds its background values is assumed to be circular and the area of this territory is 1810 km^2. In fact, Moscow's territory is slightly extended along railway roads and main highways, which is reflected in increased CO concentrations observed at a distance up to 30–35 km from the center of Moscow (TROICA experiments, Elansky et al., 2014a). Using the method by Stremme et al. (2013), one can estimate an error in extrapolating CO TC to the entire area of Moscow (1810 km^2). The upper limit of this error does not exceed 15%. The annual mean emissions of CO into the atmosphere from the entire area of the Moscow megacity amount to 870 ± 200 Gg yr^{-1} for 1992–2008 and 680 ± 160 Gg yr^{-1} for 2003–2008.

6. Discussion and conclusions

The air-quality indices given for Moscow in Table 1 characterize a new state of the urban infrastructure formed by the late 1990s. Unfortunately, the SAPM network operating since 1965 could not provide reliable data on the mean pollution level for Moscow during the period of economic crisis. This can easily be seen from comparison between observational data obtained at the SAPM network (see State Reports 2002–2008) and at the MEM and OIAP stations (Mosecomonitoring, 2011; Bulletin, 2008; Elansky, 2006, 2014b). The main reason is that the objectives of the SAPM and MEM networks were different. The main task of the SAPM network was to detect toxic-pollutant concentrations hazardous to human health. Therefore, stations entering into the SAPM network were located in the zones within which industrial enterprises polluting the atmosphere and motorways with heavy traffic were located. The MEM network was organized in order to provide air-quality information for the entire territory of Moscow, including residential areas, parks, and pieces of idle land.

SAPM data used by M.S. Myagkov (Gurjar et al., 2008) yielded an overestimation of the MPI. These data correspond to the peak concentrations of SO_2, NO_2, and TSP. Their very high concentrations were sometimes recorded at the MEM, MSU, and OIAP stations. A significant increase in the concentration of NO_x was noted during intense turbulent mixing in the rear of the cold front (Lokoshchenko and Elansky, 2006). Under these conditions, the emissions of TPPs could reach the land surface. Very high SO_2 concentrations of up to 40 $\mu g\, m^{-3}$ were recorded at some stations in anomalously cold January 2006 (the air temperature decreased to $-38\,°C$) when almost all TPPs used reserved liquid fuel because of lack of natural gas. High pollutant concentrations were also recorded when a blocking anticyclone was over Moscow. In the summer of 2002 and 2010, the influence of such anticyclones was enhanced due to forest and peatbog fires. In these years, the hourly mean of PM10 reached 250–350 $\mu g\, m^{-3}$ (Gorchakov et al., 2007, 2011), SO_2 and NO_2 in 2010 reached 41.2 and 407 $\mu g\, m^{-3}$, respectively; these are the highest values for the 2000s (Elansky et al., 2011).

By 2008, a decrease in the recurrence of extreme situations caused by both anthropogenic and natural factors became apparent in the form of a weak negative trend in the annual mean concentrations

of SO_2 and PM10 (Table 1). However, a significant increase in the number of vehicles (despite ecologically clean models) resulted in an increase in the atmospheric content of NO_2. Nevertheless, the MPI value for Moscow has almost remained unchanged since 2000.

The estimates of CO emissions according to the 2003–2008 CO TC observations are close to their values calculated on the basis of the surface concentration of CO for 2002–2008 (Table 3). Although the existing difference is within the accuracy, nevertheless it can be justified, because estimating the annual mean emission of CO from its surface concentration does not take into account emissions from the high pipes of TPPs. Therefore, we think that the value 680 ± 160 Gg yr^{-1} is more accurate for 2002–2008. For a longer CO TC observation period of 1992–2008, the mean emission of CO is noticeably larger. The differences in estimates obtained for different observation periods can be explained by the negative trend of the CO concentration in the mixing layer over the megacity. In 2002–2008 (except the anomalous summer of 2002), the surface concentration of CO at MSU decreased with a rate of about 2.5% yr^{-1} (Elansky et al., 2014b). In the 2000s, the average negative CO concentration trend (about 2% yr^{-1}) was recorded for all Russian cities (State Report, 2009). According to Rakitin et al. (2011), the urban portion of CO TC also decreased with a rate of 0.6% yr^{-1} from 1996 to 2008. The main reasons for this decrease were the modernization of Moscow's heating system, the replacement of old vehicles by newer models with a high-temperature fuel combustion and low CO emissions, and a low road capacity (no more than 60% of all registered vehicles can circulate throughout the city).

Taking into account that the CO TC trend in Moscow is almost linear, one can assume that the average CO emission 870 ± 200 Gg yr^{-1} for 1992–2008 approximately corresponds to that for 2000, and 680 ± 160 Gg yr^{-1} corresponds to 2005. Gurjar et al. (2008) presented the annual emission of CO for Moscow (1324 Gg yr^{-1}) from M.S. Myagkov's personal communication. This value exceeds that obtained in this work by 1.5 times. M.S. Myagkov probably used SAPM network CO data obtained before the economic crisis under the conditions of an increased level of air pollution.

Three global anthropogenic emission inventories – EDGAR, IPCC-AR4, and RETRO – were used by Butler et al. (2008) to calculate the emissions of CO. The annual CO emissions obtained for Moscow amounted to 979, 981, and 1249 Gg yr^{-1}, respectively. All these inventories are based on both observational and model data obtained in the late 1990s and the very early 2000s. Our estimate for this period (870 ± 200 Gg yr^{-1}) coincides (within the limits of error) with those from the EDGAR and IPCC-AR4 inventories.

In the RETRO inventory, much more importance is given to domestic emissions. In Moscow, in the 1990s, due to a setback in production and an increase in the number of vehicles, transport emissions rapidly increased when compared to domestic and industrial ones; for 2000, their ratio is estimated at 80% (transport), 10% (domestic), and 10% (industrial) (Bulletin, 2008; State Report, 2009). This ratio is closest to that from the IPCC-AR4 inventory (Butler et al., 2008).

According to Stremme et al. (2013), the annual CO emission from Mexico City amounted to 2150 ± 500 Gg yr^{-1} for 2008, which exceeds that from Moscow for approximately the same period by three times. Differences between Moscow and Mexico City are not only in the sizes of these megacities and in the number of inhabitants but also in the amount of contribution made by the domestic sector to the emission of CO. This contribution amounts to 15–20% for Mexico City (Molina and Molina, 2002; Molina et al., 2010; SMA-GDF, 2010).

In Gurjar et al. (2008), data are given on the annual emissions of CO from 18 megacities; these data were obtained by different authors and published within the 1990–2004 period. According to the estimate 870 ± 200 Gg yr^{-1} obtained by us, Moscow ranks 9th after Tokyo, Beijing, Shanghai and other megacities. According to the CO emission estimates taken from the EDGAR inventory and presented in this work, Moscow ranks 10th. If we refer to the results of calculations of annual CO emissions, which were performed by Butler et al. (2008) for 32 largest cities in the world on the basis of the ED-GAR, IPCC-AR4, and RETRO inventories, Moscow ranks 17th, 13th, and 18th, respectively. Since maximum CO emissions are observed in cities, in which the quality of air (Table 1) is highest, taking into account CO concentrations in calculations of the MPI may improve the ranking of Moscow among other megacities by air quality.

The estimates of CO emissions from the Moscow megacity may be used in regional models to study the influence of the megacity on atmospheric status, on the occurrence of air-pollution events, and on climate in a region under study (see e.g. Baklanov et al., 2007; Lawrence et al., 2009). However, in

order to simulate the air quality in a city and predict its possible changes, the proper knowledge of emission sources and their location in time and space is necessary. In 2013 the Russian Academy of Sciences and the Russian Hydrometeorological Service started joint developing of a complete emission inventory to support an air quality forecasting and emission scenarios analysis for air quality management. The presented evaluation of CO emissions can be used to cross-check inventory data.

Acknowledgments

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