



Assessment of urban aerosol pollution over Moscow megacity by MAIAC aerosol product.

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Abstract. We estimated the distribution of aerosol optical thickness (AOT) with a spatial resolution of 1 km over Moscow megacity using MAIAC aerosol product based on MODIS satellite data (Lyapustin et al., 2018) for the warm period of year (May-September). AERONET (Aerosol Robotic Network)-based validation near the city centre at Moscow_MSU_MO and over Moscow suburbs at Zvenigorod revealed that MAIAC AOT at 0.47 μm is in agreement with AERONET AOT though underestimated by 0.05-0.1 for $\text{AOT} < 1$ and overestimated for smoke conditions with $\text{AOT} > 1$. The MAIAC AOT biases were almost the same for the Moscow_MSU_MO and Zvenigorod AERONET sites, which indicated that MAIAC effectively removed the effect of the bright urban surface in the city centre. For the ground-based measurements, the annual median AOT difference between Moscow_MO_MSU and Zvenigorod (ΔAOT) varied within -0.002-+0.03 with statistically significant positive bias for most years and an average ΔAOT of ~ 0.02 . According to MAIAC dataset, ΔAOT varied within ± 0.01 and was not statistically significant. The ΔAOT started decreasing recently due to intensive urban development of the territory around Zvenigorod and the decrease of pollutant emissions in Moscow, which is mainly caused by the environmental regulations. According to the MAIAC dataset, the most pronounced spatial AOT difference over the territory of Moscow was observed at 5% quantile level, where it reached 0.05-0.06 over several locations and could be attributed to the stationary sources of aerosol pollution, for example, power plants, or aerosol pollution from roads. The difference between the maximum and the mean AOT for different quantiles, except the 95% quantile, within the Moscow region, was about 0.02-0.04 which could be attributed to the local aerosol sources. The application of the MAIAC algorithm over the whole Moscow region has revealed a decreasing AOT trend over the centre of Moscow and an increasing trend over the “New” Moscow territory which experienced an intensive build-up and agricultural development in the north and the south parts of this district, respectively.



1 Introduction

Atmospheric aerosols are the suspended particulate components of the atmosphere, which are produced directly from the emissions of particulate matter of different origins and generated from gaseous precursors. The variety of chemical and physical processes of aerosol formation provides a large diversity of their microphysical and optical properties. A significant variation of aerosol properties has been observed in the industrial urban areas. One of the key aerosol optical characteristics is the aerosol optical thickness (AOT), whose spatial and temporal variations have been studied using satellite and ground-based data in numerous papers (Koelemeijer et al., 2006, Schaap et al., 2008, Chubarova, 2009, Bovchaliuk et al., 2013, Putaud et al., 2014, Chubarova et al., 2016, etc.). Over the Europe, a permanently elevated aerosol loading was observed over several industrial regions with particularly high values found over Netherlands, Belgium, the Ruhr area, the Po-valley, the Northern Germany and the former East Germany, Poland, and parts of Central European countries. Elevated aerosol loading is generally correlated with suspended particulate matter associated with the poor air quality (van Donkelaar et al., 2015, Beloconi et al., 2018).

Large cities with their high road density and industrial enterprises are the source of aerosol pollution, which includes black carbon, sulphate, nitrate and ammonium aerosol components as well as primary and secondary organic aerosols (POA and SOA) (IPCC, 2013). And the urban aerosol is dominated by the fine mode particles (Kaufman et al., 2005). Several recent studies reported an analysis of AOT based on ground-based and satellite data over Moscow and Warsaw urban areas (Chubarova et al. 2011, Zawadzka et al, 2013, Kislov, 2017). The Moscow megacity (55°45'N, 37° 37'E) is one of the largest urban agglomerations in the world with population of 12.6 million according to the Federal Statistics Service (on January 1st, 2019) with industrial enterprises and technologies in the field of mechanical engineering and metalworking, energy and petrol chemistry, light and food industries, construction materials and an intensive residential development (Kulbachevski, 2018). In 2012, the Moscow megacity has expanded to include a “New” Moscow region mostly to the south-west. As a result, its territory has increased from 1091 to 2511 km² (<https://www.mos.ru/en/>).

Previously, the urban aerosol pollution in Moscow has been studied using concurrent observations by the AERONET Cimel sun-photometers located in the Moscow city and in the suburbs (Zvenigorod). This study revealed an average AOT at 0.5 μm of ~0.19 of which 0.02 was apportioned to the urban sources, and a tendency of lower single scattering albedo (higher absorption) in Moscow (Chubarova et al., 2011). The urban AOT difference between the city of Warsaw and suburban conditions of Belsk was estimated as 0.02 (at 0.5 μm) based on sun photometers' data (Zawadzka et al., 2013). However, the use of only two contrasting ground-based sites does not allow assessing the detailed spatial distribution of AOT and estimating an integrated urban aerosol loading even at high quality of the AOT measurements. This task can be solved by using the high quality satellite AOT retrievals.



The analysis of the results obtained from the Visible Infrared Imaging Spectrometer (VIIRS) (Jackson et al., 2013) showed that the central part of the Moscow city has a significantly higher AOT at 0.55 μm (by about 0.1) than that in the suburbs (Zhdanova, Chubarova, 2018). Such a significant difference, as discussed in this paper, has probably originated from the uncertainty in evaluation of the urban surface reflectance in the VIIRS aerosol algorithm (Liu et al., 2014). The assessment of the aerosol pollution in Moscow using the mid-visible range AOT from the MODIS data (collection 5.1) with a $1^\circ \times 1^\circ$ spatial resolution during the warm period of 2000-2013 showed that the difference in AOT due to urban effects can reach up to 0.08 if compared to AOT obtained over the green areas to the north of 58°N or to the south of 53°N (Kislov, 2017). However, the spatial resolution and the uncertainties of the AOT retrievals used in this study did not allow determining the detailed spatial features of AOT distribution. The MAIAC aerosol product (Lyapustin et al., 2018), based on MODIS data, has some advantages over the standard MODIS algorithms: it overcomes empirical assumptions related to surface reflectance and provides AOT at high 1 km spatial resolution. MAIAC uses the minimum reflectance method, implemented dynamically, to separate atmospheric and surface contributions. The sliding window technique, accumulating a time series of data for up to 16-days, provides a necessary surface characterization via dynamic retrieval of the spectral bidirectional reflectance distribution function (BRDF) (Lyapustin et al., 2018). A good knowledge of surface BRDF allows MAIAC to minimize effects of both surface brightness and view geometry on MAIAC AOT as compared to the standard MODIS Dark Target (DT) and Deep Blue (DB) products (e.g., Mhawish et al., 2018; Jethva et al., 2019). Thus, the objective of this paper is to test the MAIAC aerosol retrievals against the high-quality AERONET measurements in the Moscow area (for the urban and suburban sites) and to evaluate the trends and spatial features of the urban aerosol pollution over the Moscow megacity for the time period from 2001 to 2017.

2. Datasets and methodology

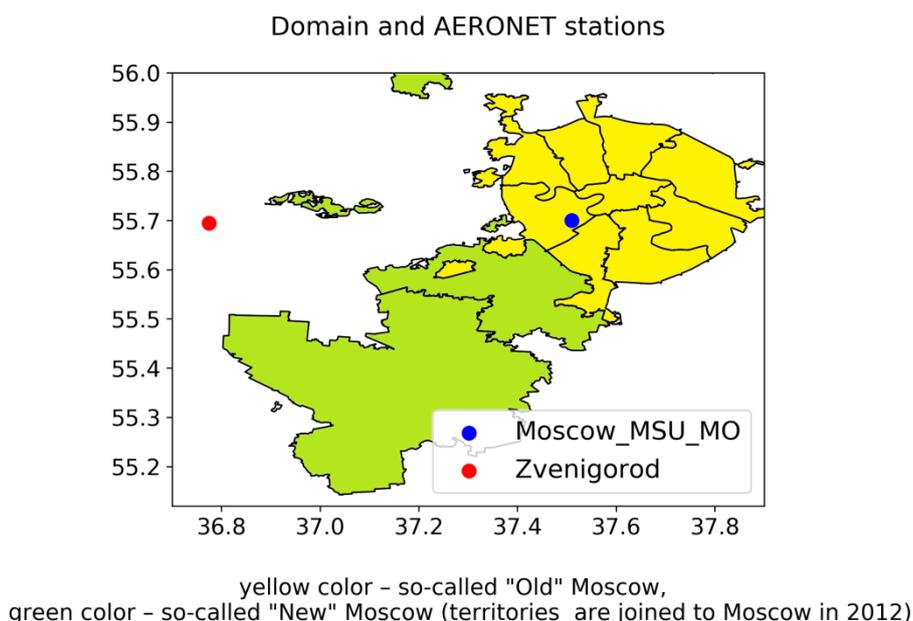
A new MODIS satellite product - MCD19A2 Collection 6 (MAIAC aerosol product) with 1 km spatial resolution was used to estimate spatial-temporal distribution of AOT over the Moscow region (<https://search.earthdata.nasa.gov/search>). MCD19A2 product provides a suite of atmospheric parameters and view geometry, including: column water vapor, AOT at 0.47 and 0.55 μm , AOT uncertainty, fine mode fraction over water, smoke injection height (m above ground), AOT QA (Quality Assurance), AOT model at 1km, and a view geometry suite at 5 km (cosine of solar zenith angle, cosine of view zenith angle, relative azimuth angle, scattering angle, and glint angle). Each parameter within each MCD19A2 Hierarchical Data Format 4 (HDF4) file contains a third dimension that represents the number of orbit overpasses. We used the data for the warm snow-free time period from May to September over the 2000-2017 years. The geographical location of the Moscow region corresponds to the MODIS granule h20v03. MAIAC retrieves AOT at 0.47 μm and provides an additional value at the standard wavelength 0.55 μm calculated according to the aerosol model used. MAIAC uses 8 different regional aerosol models tuned to the AERONET (Aerosol Robotic Network, (Holben et al., 1998)) climatology. It also detects absorbing dust and smoke aerosols and provides dust/smoke mask in the QA. The smoke test relies on a relative increase in aerosol absorption at MODIS wavelength 412 nm compared to 470–670 nm owing to multiple scattering and enhanced



absorption by organic carbon released during biomass burning combustion (Lyapustin et al., 2012). A detailed description of the MAIAC aerosol algorithm can be found in (Lyapustin et al., 2018). Only AOT values with the highest quality were used in the presented analysis (QA.QA_AOT = Best_Quality).

The data from the two sites equipped with the Cimel sun/sky photometers of the AERONET project (Holben et al., 1998) were used for the validation of the satellite AOT retrievals, as well as for determining the features of the AOT temporal-spatial distribution over the territory of Moscow megacity. They included the measurements of the Observatory of Moscow State University (Moscow_MSU_MO site, 55.70695° N, 37.52202° E) over the 2002-2017 period and Zvenigorod scientific station of Institute of Atmospheric Physics, Russian Academy of Sciences (Zvenigorod site, 55.695° N, 36.775° E) over the 2006-2017 period. The first site is located within the city, at a distance of about 8 km from the city centre, the second - the upwind suburban area about 50 km west from the city centre. The AERONET measurements at level 2, version 3 (Giles et al., 2019) were used with the additional cloud-screening using ground-based visual cloud observations at the Meteorological Observatory of Moscow State University, as described in (Chubarova et al., 2016). Additionally, we used AERONET estimates of fine mode fraction (O'Neill et al., 2003). The study domain and the location of the AERONET sites are shown in Figure 1.

In addition, we used the EMEP ('European Monitoring and Evaluation Programme') grid archive (http://www.ceip.at/new_emep-grid/01_grid_data) for assessing the spatial-temporal distribution of aerosol precursor gases emissions to explain the spatial features of the AOT distribution. We analysed the main precursor gases NO_x, SO_x, NMVOC, NH₃, along with particulate matter concentrations (PM_{2.5} and PM₁₀).



110 **Figure 1. Study domain and location of AERONET sites.**



3. Results

3.1 Validation of satellite AOT retrievals against ground-based data.

The MAIAC aerosol algorithm was successfully validated over various geographic regions: over bright desert surfaces
115 (Sever et al., 2017), over South Asia (India) (Mhawish et al., 2019), over mountainous areas (Emili et al., 2011), across
South America (Martins et al., 2017), and over North America (Jethva et al., 2019). Mhawish et al., (2019) gave a detailed
comparison of MAIAC data with standard MODIS algorithms and ground-based data, and studied the accuracy of product as
a function the sensor (MODIS on Terra or Aqua), the underlying surface, aerosol model, and scanning geometry. According
to (Mhawish et al., 2019), the MAIAC AOT error is about 15%. At high AOT, MAIAC underestimates AOT, especially in
120 MODIS Aqua record (Mhawish et al., 2019). However, on average, the AOT MAIAC data are characterized by smaller
errors compared to the two operational MODIS algorithms: Dark Target (Levy et al., 2013) and Deep Blue (Hsu et al.,
2013).

We averaged AERONET data to 1-hour resolution and calculated AOT at 0.47 μm from available AERONET AOT at 0.44
 μm and Angstrom exponent (0.44-0.87 μm) in this study. MAIAC AOT data were spatially averaged with a 5-km circle
125 centred at the Moscow_MSU_MO and Zvenigorod sites and also averaged within 1 hour to have robust estimates.
Correlations are plotted separately for the Terra and Aqua data, and together for the data from the two satellites (Fig. 2). As
can be seen in Fig. 2, the satellite AOT at 0.47 μm retrievals for Moscow_MO_MSU and Zvenigorod are underestimated by
about -0.05 for the values less than 1, and overestimated in conditions of high aerosol loading in Moscow.

The overestimation of the AOT MAIAC occurs in cases of forest fires, when MAIAC detects smoke. This is clearly seen in
130 Fig.3, where the cases of detected smoke are shown by an orange color. Overall, this error is in contrast to the typical
biomass burning conditions when MAIAC usually underestimates AOT (e.g., see Lyapustin et al., 2018). The
underestimation is caused by the fact that MAIAC C6 algorithm keeps using the same background model in cases of
detected smoke which usually has higher absorption for fresh smoke aerosol (Dubovik et al., 2002). On the contrary, the
Moscow smoke of 2010 was largely a result of smoldering peat fires producing larger particle size and lower absorption
135 (Chubarova et al., 2012, Sayer et al., 2014), the combination for which led to the AOT overestimation.

Statistical estimates of the quality of the AOT at 0.47 μm retrievals relative to the ground-based AERONET data are
presented in Table 1. It is worth noting that the errors of the MAIAC AOT are similar to both Moscow_MSU_MO and
Zvenigorod sites which indicates that the bias is model-related while the contribution of bright urban underlying surface is
effectively taken into account in the MAIAC algorithm.

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Table 1. Statistical estimates of the uncertainties in AOT MAIAC retrievals for the TERRA and AQUA data separately, for the TERRA and AQUA measurements within 1 hour (AQUA/TERRA), and together for the data from the two satellites (TERRA and AQUA) against ground-based AERONET data at the MOSCOW_MO_MSU (2001-2017) and ZVENIGOROD (2006-2017) sites. RMSE - root mean square error, MAE - mean absolute error, BIAS - mean error, N - case number.

MOSCOW_MSU_MO, all AOT				
	TERRA	AQUA	AQUA/ TERRA	TERRA and AQUA
RMSE	0.24	0.23	0.17	0.22
MAE	0.12	0.1	0.09	0.11
BIAS	0	-0.02	-0.02	-0.02
N	181	130	99	410
MOSCOW_MSU_MO, AOT<1				
RMSE	0.1	0.09	0.1	0.1
MAE	0.07	0.07	0.07	0.07
BIAS	-0.05	-0.05	-0.05	-0.06
N	171	124	94	389
ZVENIGOROD, AOT<1				
RMSE	0.07	0.09	0.08	0.08
MAE	0.05	0.07	0.05	0.06
BIAS	-0.03	-0.06	-0.04	-0.04
N	77	61	48	186

MOSCOW_MO_MSU

ZVENIGOROD

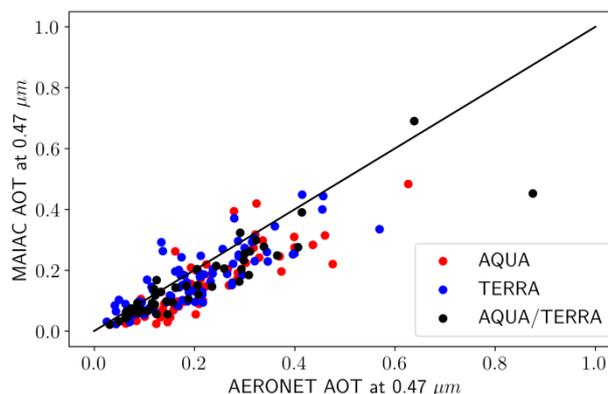
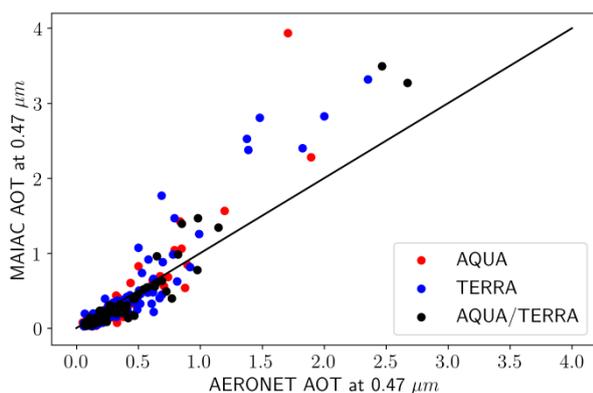
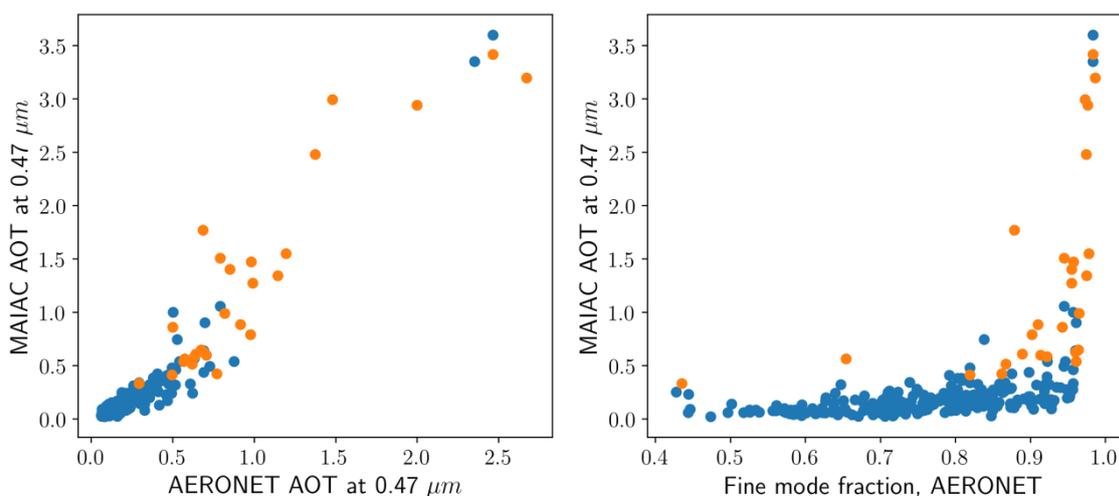


Figure 2. Correlations between MAIAC AOT at 0.47 μm and AERONET AOT at 0.47 μm for Moscow_MSU_MO and Zvenigorod AERONET sites for TERRA, AQUA and their joint overpasses within 1 hour (AQUA/TERRA).

Comment: the absence of high AOT values at Zvenigorod site is explained by technical problems with the instrument and the absence of the AERONET data at level 2 version 3 in 2010, when intensive forest fires took place.



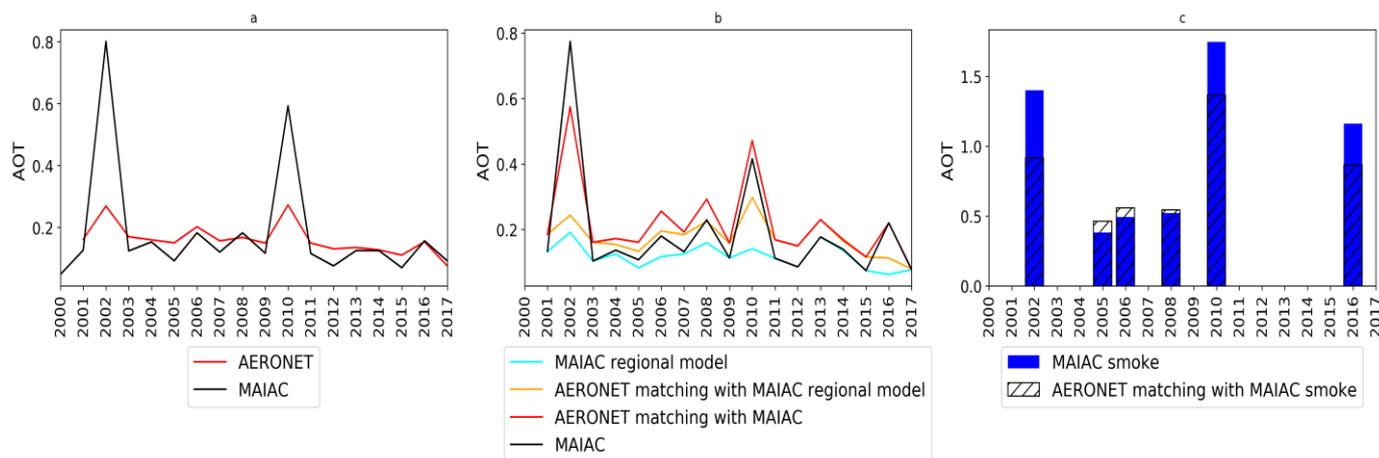
155 **Figure 3.** MAIAC AOT at 0.47 μm against AERONET AOT (left) and MAIAC AOT at 0.47 μm against fine mode
fraction AOT AERONET (right) according to the regional MAIAC aerosol model (blue color) and in cases of smoke
detection (orange color). Moscow, 2001-2017.

3.2 Temporal AOT changes in Moscow according to ground-based and satellite data

160 We studied temporal AOT changes using MAIAC AOT retrievals and AERONET long-term measurements collocated in
time over Moscow_MSU_MO site during a warm May-September period. Fig. 4a shows the time series of AOT at 0.55 μm
built for all available Moscow_MSU_MO AERONET and MAIAC data. One can see a satisfactory agreement between the
satellite and ground-based observations with the exception of 2002 and 2010 years. The highest AOT were observed in 2010
and 2002 years due to the effects of smoke aerosols from peat and forest fires in Moscow region (Chubarova et al, 2011). In
165 2016 the smoke aerosol advection was also observed from the Siberia area (Sitnov et al., 2017) providing an intermediate
AOT maximum. Fig.4b shows year-to-year variability of AOT at 0.55 μm only for matching within 1 hour
Moscow_MSU_MO AERONET and MAIAC data, and for the cases, when MAIAC regional aerosol model has been
applied. One can see a better agreement between MAIAC AOT and corresponding AERONET AOT data in year-to-year
variations. There is a clearly seen decrease in AOT during the last years according to both the MAIAC (when regional model
170 was used) and the AERONET data. The yearly means difference between AERONET and MAIAC data (AOT MAIAC –
AOT AERONET) is -0.03 for the all matching data and -0.05 for the matching data with MAIAC regional aerosol model
estimates. Fig.4c presents the AOT variations only for the cases of the smoke detection. It is seen that the AOT MAIAC
overestimation is taken place only for the cases with high AOT > 1.



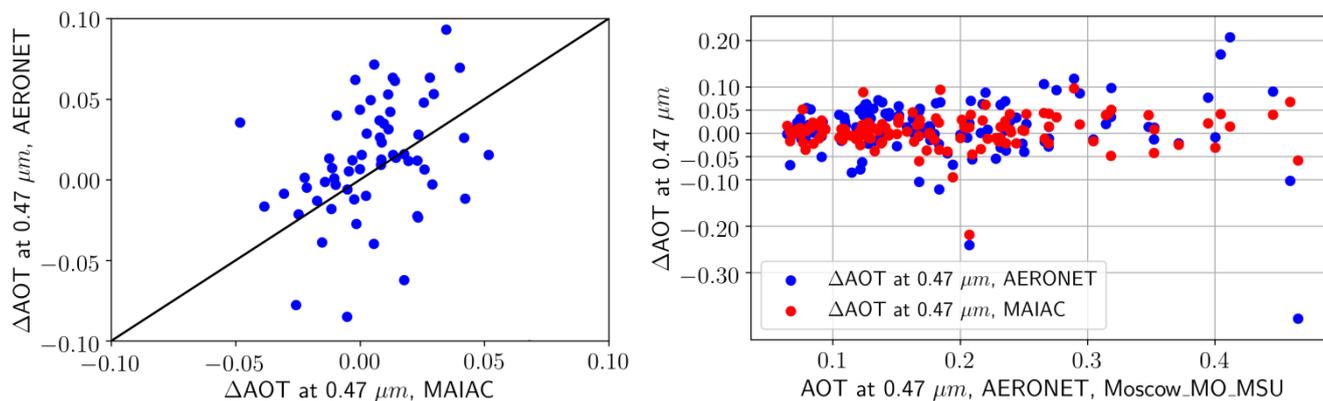
175 Thus, MAIAC AOT reproduces the absolute AOT values and the long-term AOT decrease in Moscow for the regional
 aerosol model while in case of smoke aerosol detection there is a significant overestimation of the annual AOT mean.
 Therefore, for the further analysis of urban aerosol pollution, we used only the AOT MAIAC retrievals with its attribution to
 the regional model for removing large smoke aerosol effects, which are also characterized by significant spatial
 inhomogeneity.



180 **Figure 4.** The year-to-year variations of AOT at 0.55 μm (May-September, mean values) according to AERONET
 (Moscow_MSU_MO) and MAIAC data: a) all available AERONET and MAIAC data, b) matching AERONET and
 MAIAC data for all cases and for regional aerosol model only, c) AOT MAIAC in cases of smoke detection and
 matching AERONET data.

185 **3.3 AOT urban effect according to ground-based and satellite measurements over Moscow_MSU_MO and
 Zvenigorod AERONET sites.**

Let us consider, how accurately MAIAC can reproduce the urban aerosol effect, which we evaluate as the difference
 between Moscow_MSU_MO and Zvenigorod AOT ($\Delta\text{AOT} = \text{AOT}(\text{MOSCOW_MO_MSU}) - \text{AOT}(\text{ZVENIGOROD})$). It
 should be noted that two sites are close enough to each other, so they are influenced by the medium- and long-range
 190 transport similarly. Note, that Zvenigorod site has an upwind location. Fig.5 shows the correlation between the urban aerosol
 effect from MAIAC and from hourly-averaged AERONET data. The ΔAOT values obtained from both ground-based and
 satellite data lie within the range of $-0.1 \dots 0.1$. The correlation is not high, but the range of scatter is low. It should be noted
 that the ΔAOT between Moscow_MSU_MO and Zvenigorod based on satellite and ground-based data generally correspond
 to each other. The ΔAOT between the city and the suburbs can be both positive and negative: ΔAOT varies from -0.4 to 0.21
 195 according to ground-based data and from -0.22 to 0.1 according to satellite data (see Fig.5b).



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a **b**

Figure 5. Correlation between ΔAOT at $0.47 \mu m$ ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$) obtained from the satellite and ground-based data (a) and ΔAOT at $0.47 \mu m$ as a function of AOT at $0.47 \mu m$ obtained from Moscow_MSU_MO dataset (b).

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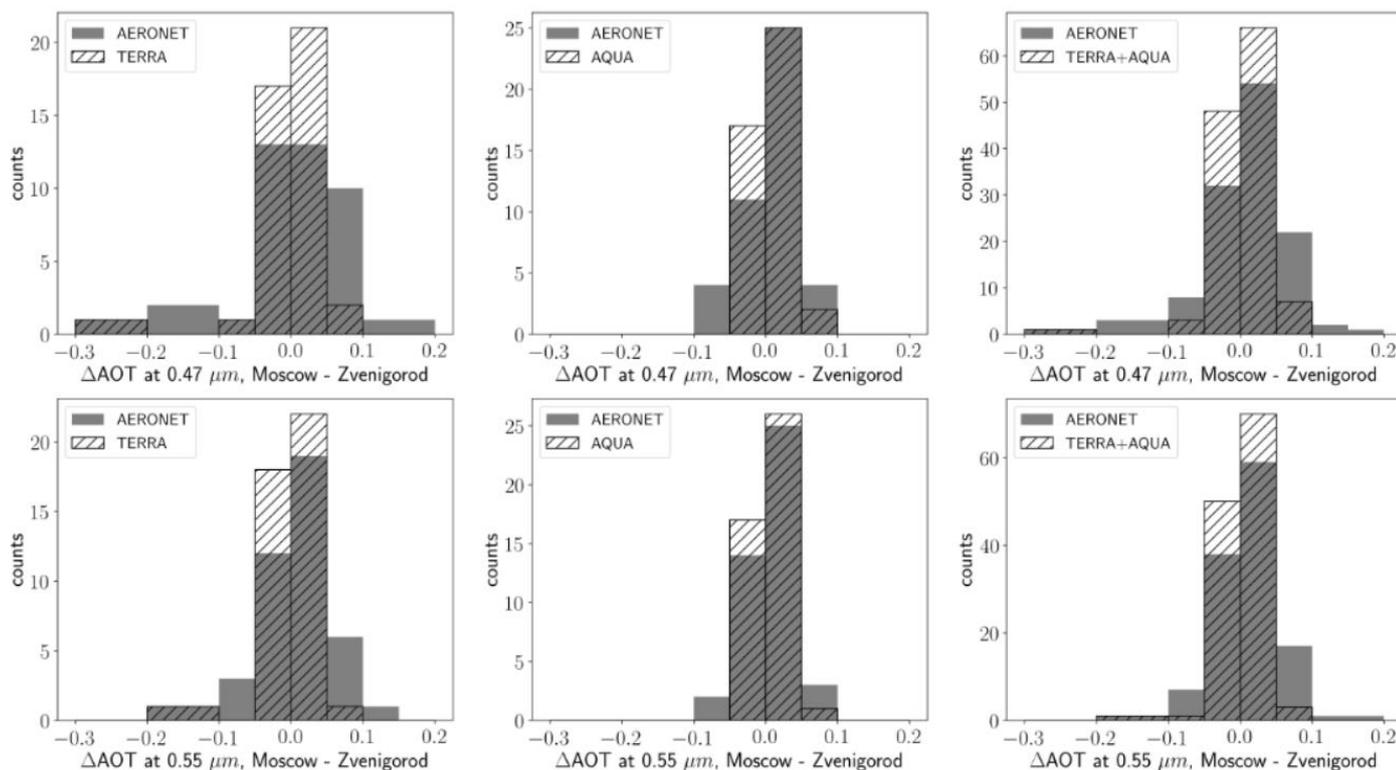
For characterizing variations in ΔAOT we analyzed frequency distributions according to ground-based and satellite data. We calculated ΔAOT separately for Terra and Aqua datasets for evaluating to some extent diurnal variability of ΔAOT . Frequency distributions of ΔAOT at 0.47 and $0.55 \mu m$ separately for the Terra and Aqua data, and together for the data from the two satellites are shown in Fig.6. The highest repeatability of ΔAOT is in the range of 0-0.05. For the Aqua AOT retrievals, which are closer to noon, the predominance of positive ΔAOT is more pronounced. In overall, the ΔAOT at 0.47 values lie within the $[0, 0.05]$ bin in 57% of cases for the Aqua and in 50% - for the Terra datasets.

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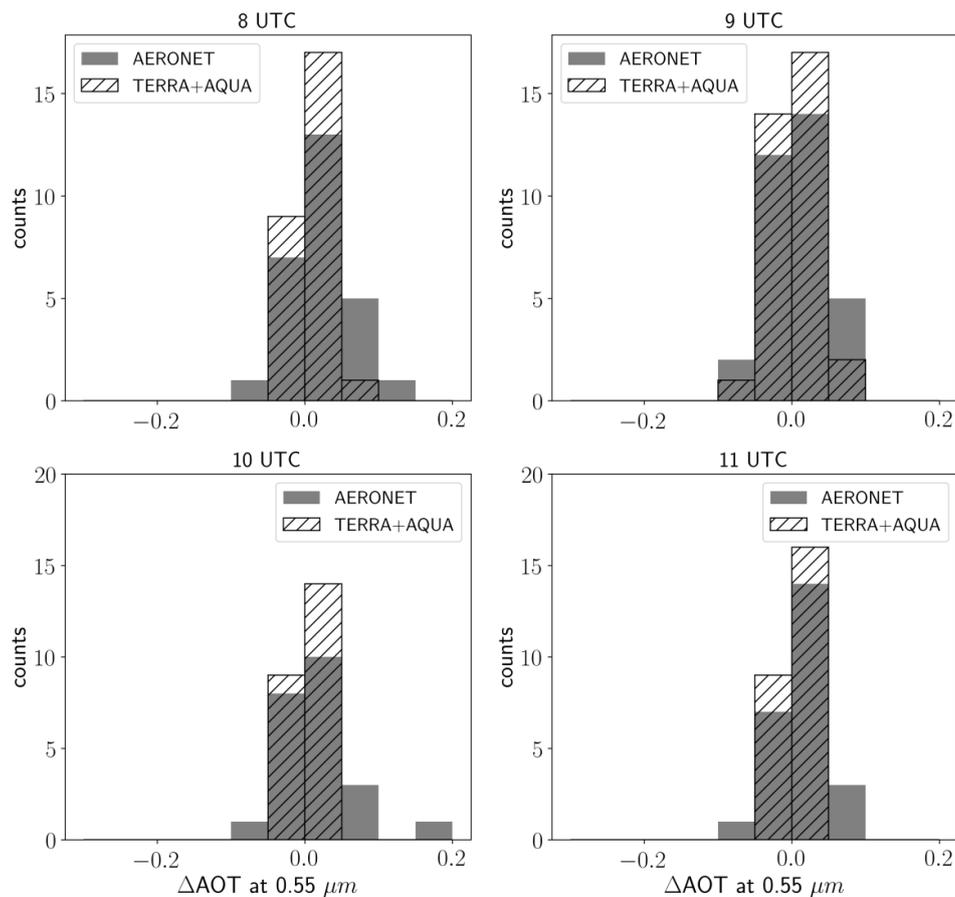
Frequency distributions of the ΔAOT for different morning hours are shown in Fig.7. One can see a similar picture for AERONET and MAIAC ΔAOT distributions with higher frequency at $[0, 0.05]$ bin. However, AERONET data has higher ΔAOT range compared with that for the MAIAC AOT. The diurnal variations of the ΔAOT according to satellite and ground-based data are also shown in Fig.8. The MAIAC ΔAOT at $0.47 \mu m$ are close to zero at the level of median values and do not exceed 0.01. The inter-quantile range of the ΔAOT at $0.47 \mu m$ is smaller for satellite data as compared to ground-based data. Satellite and ground-based ΔAOT at $0.47 \mu m$ are consistent with each other in the diurnal pattern.

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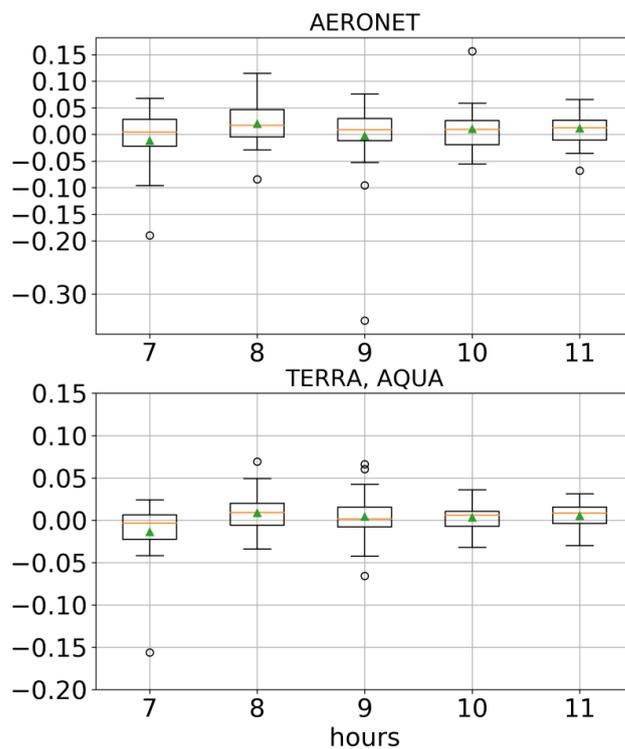


225 **Figure 6.** Frequency distribution of ΔAOT ($\Delta AOT = AOT_{\text{Moscow_MO_MSU}} - AOT_{\text{Zvenigorod}}$) at $0.47 \mu\text{m}$ (upper) and $0.55 \mu\text{m}$ (low) separately for the Terra (left column) and Aqua (middle column) datasets, and together for the data from the two satellites (right column) with frequency distribution for matching ground-based AERONET data, (2006-2017, without the data of 2009 because of technical problems at Zvenigorod AERONET site). Number of satellite and ground-based matchups is 125.



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Figure 7. Frequency distribution of ΔAOT at $0.55 \mu m$ ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$) for the TERRA and AQUA data, and ground-based data for different hours (UTC). (2006-2017, without the data of 2009 because of technical problems at Zvenigorod AERONET site). Number of satellite and ground-based matchups is 125.



235 **Figure 8.** Daily variations of the ΔAOT at $0.47 \mu m$ ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$), UTC time. The median
is in the centre, the box is the first (Q1) and the third (Q3) quartiles, the whiskers are $Q3 + 1.5 * (Q3 - Q1)$ and $Q1 - 1.5$
* $(Q3 - Q1)$, green triangles – means, points – outliers; (2006–2017, without the data of 2009 because of technical
240 problems at Zvenigorod AERONET site). Number of satellite and ground-based matchups is 125.

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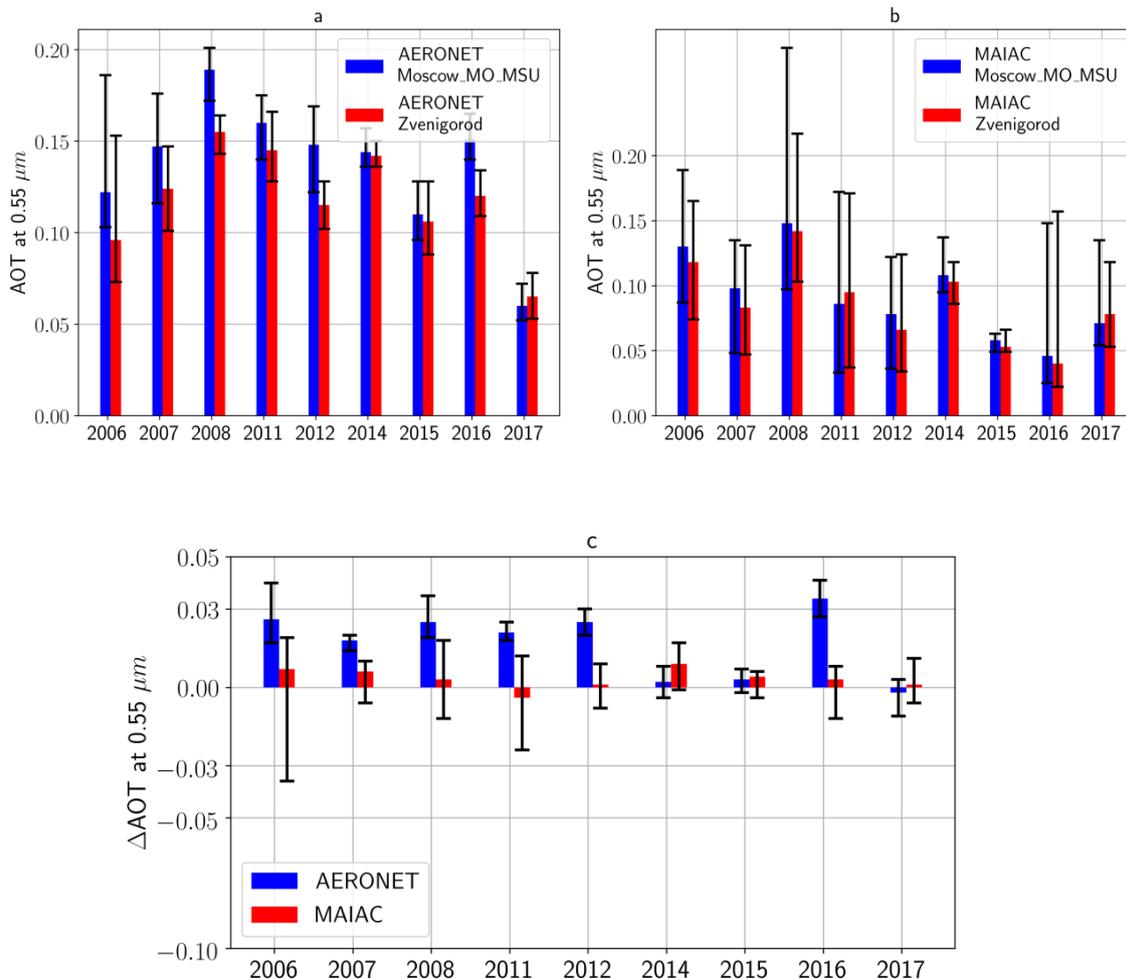


For evaluating temporal Δ AOT changes we analysed variations in annual (warm period) AOT means in Moscow and
250 Zvenigorod. The interannual variations of AOT at $0.55 \mu\text{m}$ means are shown in Fig. 9 according to AERONET and MAIAC
datasets for the 2006-2017 period. For several years the Δ AOT according to AERONET measurements are statistically
significant at 95% confidence level reaching 0.02-0.03 (median value is 0.02), while the MAIAC Δ AOT are close to zero
and not statistically significant for all years. The Δ AOT according to ground-based AERONET observations are positive
and higher before 2012. The confidence intervals for MAIAC data are much larger than the confidence intervals for
255 AERONET data because of small numbers of satellite matchups.

We excluded AOT for 2009, 2010, and 2013 years in the dataset. The AOT at $0.55 \mu\text{m}$ was significantly higher in
Zvenigorod compared to Moscow in 2009, probably, due to technical problems. Note, that most of the Zvenigorod data
during the warm period of 2009 were not included in the previous version 2 AERONET (an email, Alexander Smirnov,
personal communication, Aug. 2019). In 2010, the AOT values were strongly affected by extremely high smoke aerosol
260 loading (Chubarova et al., 2012), which was characterized by significant spatial heterogeneity. The data of 2013 year were
excluded because of lack of sufficient number of MAIAC observations to obtain robust estimates.

In general, almost for all years we see a tendency of AOT decreasing in Moscow both for the AERONET datasets and
satellite retrievals. Similar but less pronounced negative trend of AOT is observed in Zvenigorod. In addition, in the recent
years (2013-2017), excluding the 2016 year due to the influence of AOT spatial inhomogeneity of Siberian forest fires, the
265 Δ AOT becomes smaller and, moreover, negative (Fig.9c). We should note that a significant increase in vehicular traffic near
the Zvenigorod site, located 150 m away from a road, during past 25 years has resulted in the growth of the surface aerosol
air pollution level by about 2-3 times (Kopeikin, et al., 2018), which can lead to the total AOT increase there.

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275 **Figure 9. a) Year-to-year variations of May-September AOT at 0.55 μm medians (a) - according to all matching AERONET Moscow_MSU_MO and Zvenigorod data (N=1492), (b) - according to the MAIAC data (N= 264), and (c)-**
ΔAOT according to matching datasets. Error bars are given at 95% confidence level.

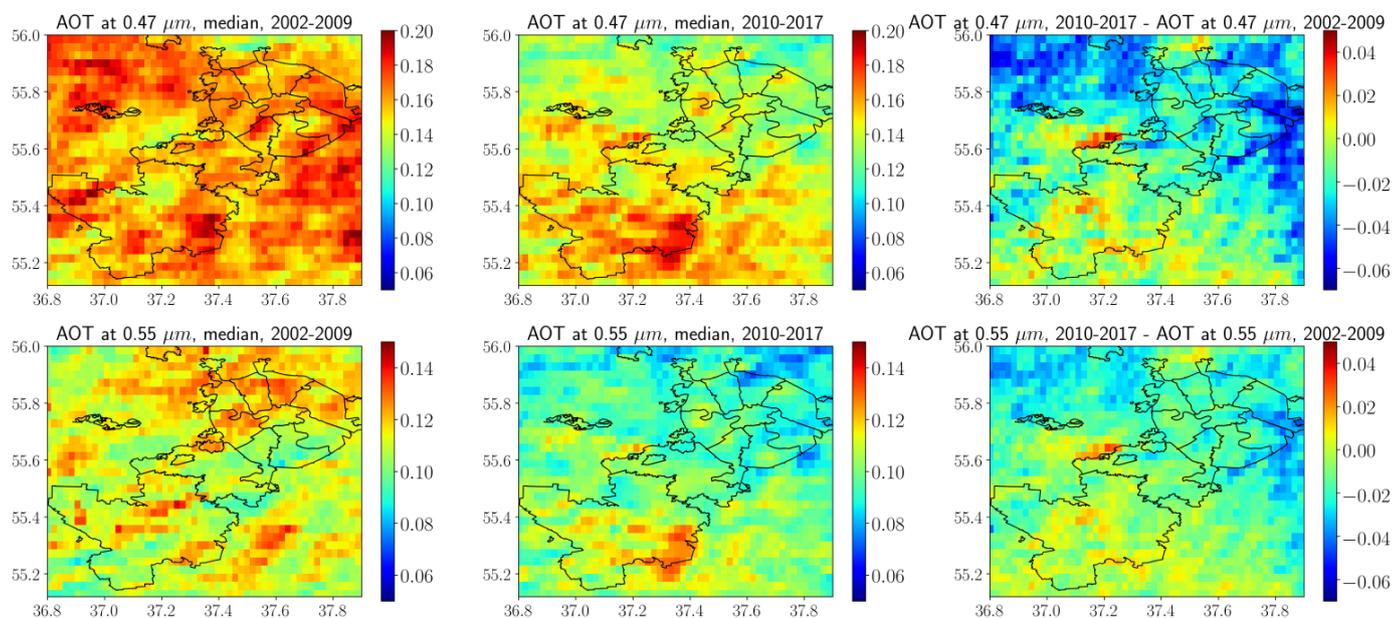


3.4 The spatial AOT distribution over Moscow region and its change in time.

Figure 10 presents the median AOT values for the two time periods (2002–2009 and 2010–2017), which show a decrease in
285 AOT over the territory of “Old”¹ Moscow and an increase over the territory of “New” Moscow. This AOT decrease is
consistent with the negative AOT tendency in AOT over Moscow_MSU_MO and Zvenigorod according to AERONET and
MAIAC data (see the discussion above).

Spatial changes of AOT over “Old” Moscow and “New” Moscow may be explained by the emissions of urban pollutants -
aerosol precursors, and, to some extent, could be associated with the uncertainties in evaluation of the type of underlying
290 surface (for example, the temporal changes in reflectance over the urban development).

Concerning the possible effect of surface changes, we should note that the MAIAC algorithm provides a dynamic
characterization of the surface reflectance properties and spectral ratios required for aerosol retrieval, and should catch
temporal surface changes associated with urban development (Lyapustin et al., 2018). In addition, the change in the
underlying surface types was analysed using the standard MODIS MCD12C1 Collection 6 product
295 (Majority_Land_Cover_Type_1), which has a spatial resolution of 5 km. The analysis has showed that there was no
significant increase in the urban underlying surface over the period 2001–2016. The number of grid cells occupied by the
urban development increased only by 6% over the north of “New” Moscow territory.

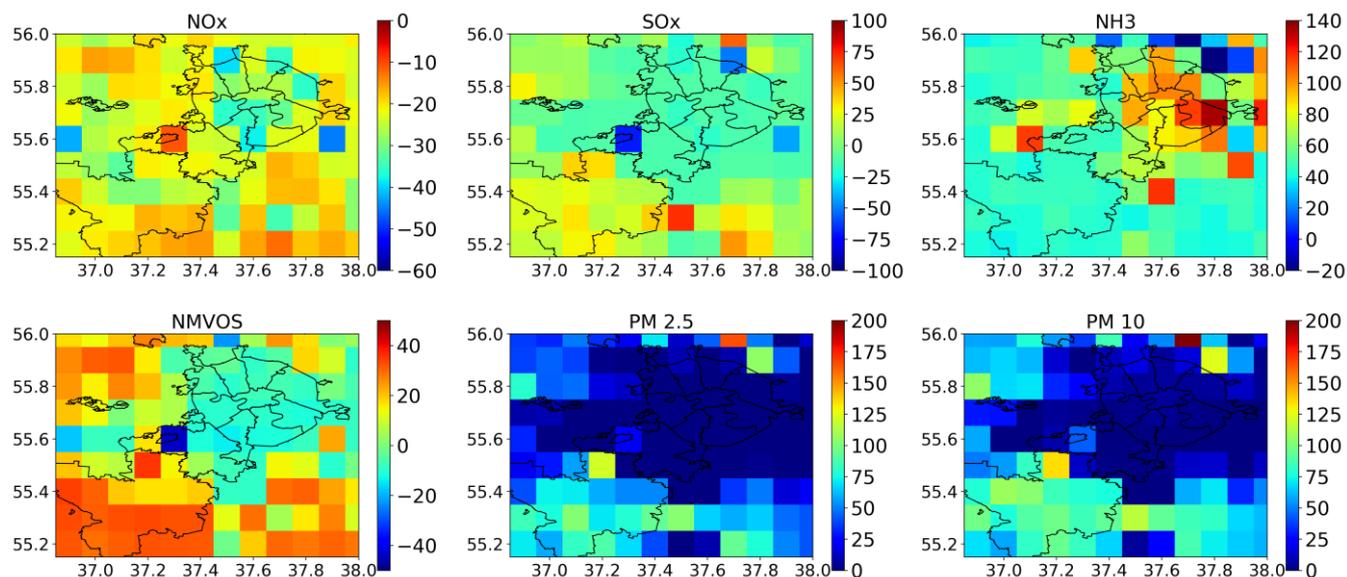


300 **Figure 10.** AOT at 0.47 μm and AOT at 0.55 μm median values for the 2002-2009 and 2010-2017 periods and their
differences.

¹ By “Old” Moscow we mean the territory of Moscow before inclusion of the “New” Moscow area in 2012.



We have also determined the change in emissions of aerosol precursors for the period 2011-2016 relative to the period 2003-2009 according to the EMEP grid archive (Fig.11). NO_x emissions were characterized by a decrease of about 30% over the territory of Moscow. NO_x emissions from motor vehicles decreased over the considered territory on average by 17%. The decrease of SO_x emissions was on average 14% over the territory of “Old” Moscow and, at the same time, the SO_x emissions increased over the territory of “New” Moscow by about 43%. Emissions of NH_3 over the territory of Moscow were increasing, on average by 81%. Emissions of Non-methane volatile organic compound (NMVOC) over the territory of “Old” Moscow was decreasing by about 6%, and at the same time, there was an increase in emissions of NMVOC over the south-west of the considered domain, up to 43%. There was an increase in suspended particles over the territory of “Old” Moscow (+ 16% PM_{10} and + 6% for $\text{PM}_{2.5}$) and much larger growth in PM (approximately in 2 fold) over the territory of “New” Moscow. However, in recent years there has been a decrease in suspended particles relative to the level of 2010 year.



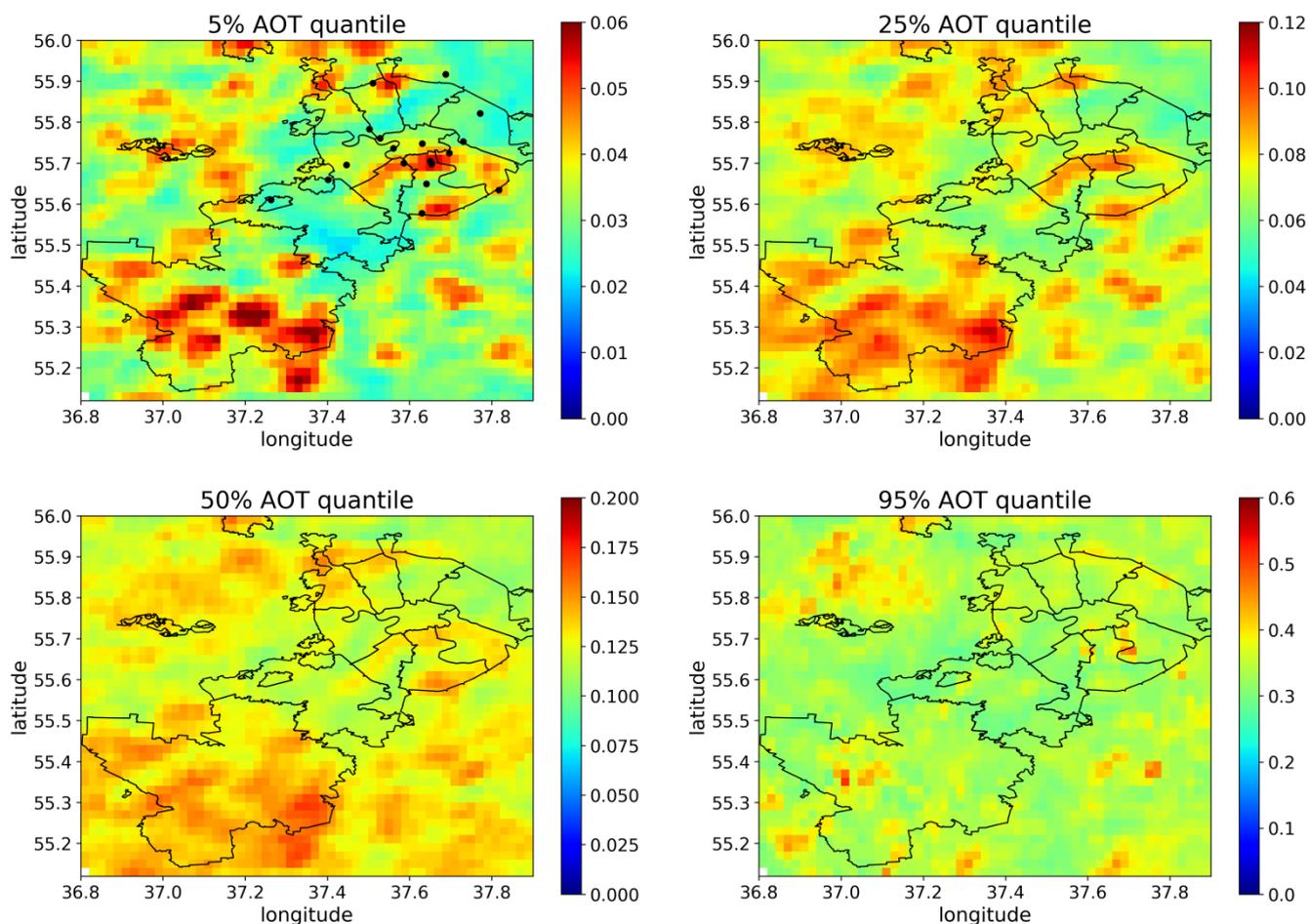
315 **Figure. 11. Ratio of emissions of gases and particle matter averaged over the 2016-2011 period to the emissions averaged over the 2003-2009 period, in percentages. EMEP dataset (http://www.ceip.at/new_emep-grid/01_grid_data)**

320 The obtained results are consistent, for example, with the data in (Chernogaeva et al., 2019), according to which over the past 10 years, pollutant emissions have decreased in Moscow, which is caused mainly by environmental regulations (Kulbachevski et al., 2018), and increased in the Moscow region. Thus, the higher AOT values over the territory of “New” Moscow can be explained by higher aerosol precursors emissions over this area than those over “Old” Moscow.



We also applied the quantile analysis to the spatial AOT fields obtained from the MAIAC algorithm. In addition to the
325 mentioned elevated mean AOT values over the territory of “New” Moscow, relatively high AOT at 0.47 μm 50% quantile
values are observed at the south-western and southern administrative districts of “Old” Moscow, probably due to highways
and industrial enterprises, thermal power plants (Fig.12). One can see the most pronounced spatial difference in AOT at 5%
quantile level, where the difference over several locations may reach 0.05-0.06 in some cases and can be attributed to the
stationary sources of aerosol pollution over “Old” Moscow. Table 2 presents mean and maximum values of AOT quantiles
330 separately for the territories of “Old” and “New” Moscow. One can see that over local points the difference between
maximum AOT and mean AOT values comprises about 0.02-0.04 for different quantiles, except 95% quantile, which can be
attributed as the local urban aerosol effect observed in Moscow. In Figure 12, we can see that there are several spatial
patterns in AOT distribution over the territory of “Old” Moscow. These patterns associate with the influence of industry and
road emissions, which produce the spatial changes in AOT over the territory of “Old” Moscow of about 0.03 for wavelength
335 0.47 μm and of 0.02 for wavelength 0.55 μm .

Over the territory of “New” Moscow, areas with elevated AOT values link with experiencing intensive city build-up and
agricultural activities at the north and at the south parts of this district, respectively.



340 **Figure.12. Quantiles (5%, 25%, 50%, 95%) AOT at 0.47 μm over Moscow megacity, 2001-2017. Black points in**
upper left map are thermal power plants according to the «System Operator of the United Power System» data
(<https://www.so-ups.ru>)

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Table 2. Mean and maximum of AOT quantiles (5%, 25%, 50%, 95%) over the “Old” Moscow and “New” Moscow territories.

quantile	“Old” Moscow		“New” Moscow	
	AOT at 0.47 μm (mean/max)	AOT at 0.55 μm (mean/max)	AOT at 0.47 μm (mean/max)	AOT at 0.55 μm (mean/max)
5%	0.03/0.06	0.02/0.04	0.04/0.06	0.02/0.04
25%	0.07/0.1	0.05/0.07	0.08/0.11	0.05/0.08
50%	0.12/0.15	0.08/0.11	0.13/0.17	0.09/0.12
95%	0.34/0.50	0.24/0.36	0.33/0.52	0.23/0.37

355 We also estimated the AOT difference depending on the distance from the city centre. Frequency distribution of AOT at 0.47 μm differences averaged over the two areas, bounded by circles with a radius of 15 km and 50 km centred in the Moscow city centre consisted of 33% of cases in the range of [-0.02;0] and 60% of cases in the range of [0, 0.02]. This finding is also consistent with ground-based data.

4. Discussion and conclusions

360 The MAIAC AOT (MODIS product MCD19A2) was used for the analysis of the urban aerosol pollution and its dynamics over the Moscow megacity. MAIAC AOT was validated against two AERONET sites located near the centre of Moscow (Moscow_MSU_MO) and in the suburban region (Zvenigorod). The validation showed a good overall agreement between the ground-based and satellite data, though MAIAC underestimated AOT by 0.05-0.1 for typical conditions (AOT<1). Statistical analysis showed a similar MAIAC AOT performance for the two sites, i.e. RMSE = 0.1, MAE = 0.07, BIAS = -
 365 0.06 for Moscow_MSU_MO and RMSE = 0.08, MAE = 0.06, BIAS = -0.04 for Zvenigorod. The obtained estimates are consistent with the global MAIAC AOT validation over the land, e.g. RMSE=0.06-0.08 and BIAS= -0.01- -0.03 over the North and South American continents (Lyapustin et al. 2018).

On average, the MAIAC AOT product reproduces the absolute AOT values and the AOT decrease since 2012 observed in the AERONET data, and shows a robust performance in urban environments with higher land surface reflectance. These
 370 results are in agreement with other studies, such as Sever et al. (2017) which showed that the pollution from industrial zone could be identified with MAIAC AOT data even over bright semi-deserts of the Dead Sea area.

In high AOT conditions (AOT>1) observed during the Moscow forest and peat fires of 2010, MAIAC showed an overestimation of AOT. This result is in contrast to the typical biomass burning conditions when MAIAC usually underestimates AOT by ~10-20% (e.g., see Lyapustin et al., 2018). MAIAC C6 algorithm lacks a specialized smoke aerosol



375 model with higher absorption and keeps using the regional background aerosol model in cases of detected smoke, which usually has a higher absorption (Dubovik et al., 2002), in particular for the fresh smoke. Atypically, the Moscow 2010 smoke was mostly generated by the slow smouldering peat burning which produces a relatively large particle size and a low absorption (Chubarova et al., 2012, Sayer et al., 2014). The combination of these properties of smoke particles not accounted for in the MAIAC algorithm may have resulted in the observed AOT overestimation. In general, we found that MAIAC
380 smoke detection was a good indicator of forest and peat fires in the Moscow region. Ability of the MAIAC algorithm to confidently capture both fresh and transported smoke in the aerosol type parameter has also been confirmed in Veselovskii et al. (2015).

To evaluate the urban aerosol effect, we analysed the spatial difference between simultaneously measured AOT at Moscow_MSU_MO and at Zvenigorod ($\Delta\text{AOT} = \text{AOT}(\text{MOSCOW_MO_MSU}) - \text{AOT}(\text{ZVENIGOROD})$), which was
385 produced from both AERONET and MAIAC datasets. AERONET measurements showed that the annual median ΔAOT varied within -0.002 to $+0.03$ with statistically significant positive bias for most years and the average difference of ~ 0.02 . A similar result was reported for the urban conditions of Warsaw (Zawadzka et al., 2013), where ΔAOT between Warsaw and Belsk was estimated as ~ 0.02 (at 500 nm) and 0.03 (at 550 nm) according to the AERONET and the standard MODIS aerosol product, respectively. According to Figure 9, MAIAC also showed a positive AOT difference ~ 0.01 between
390 Moscow and Zvenigorod for all years except 2011 (in 2017 both AERONET and MAIAC showed a negative difference) but it was not statistically significant due to higher noise in the MAIAC retrievals compared to the direct AERONET measurements. In comparison, a similar assessment using standard MODIS aerosol algorithm showed $\Delta\text{AOT} = 0.03$ (Chubarova et al., 2011). Note, that similar analysis between centre of Berlin city and its suburbs resulted in a much higher $\Delta\text{AOT} = 0.08$ (Li et al, 2018). Such difference seems to be too high and could be explained by the urban bias of the standard
395 MODIS collection MYD04_3K (3km AOT product) caused by the brighter underlying surface.

Both AERONET and MAIAC show the decreasing trend of the urban aerosol effect (ΔAOT) since 2012, which is consistent with the increase of pollutant emissions over Zvenigorod and their decrease over Moscow during the last years according to the EMEP archive (see Figure 14).

Analysis of the spatial distribution of MAIAC AOT at $0.47 \mu\text{m}$ shows higher values over the highways and main roads and
400 industrial enterprises and over the territory of “New” Moscow at the 5%, 25% and 50% quantile levels with 0.05-0.06 difference against lowest values. The largest local difference in AOT is observed in the clean conditions at 5% quantile. Hence, our results confirm the statement in (Chudnovsky et al., 2013) that “low pollution days require higher resolution aerosol retrievals to describe spatial AOT heterogeneity in urban environment”, which resulted from MAIAC-based study over the Boston area. The higher AOT over the territory of “New” Moscow can be explained by the increased aerosol
405 precursor emissions from intensive construction and agricultural activities. The difference between the maximum and the



mean AOT values for different quantiles, except 95% quantile, within the Moscow region, is about 0.02-0.04 which can be attributed to the local urban aerosol effects.

Thus, the application of the new MAIAC algorithm provides a reliable instrument for assessing the spatial features distribution of urban aerosol pollution and allows us to evaluate the level of local urban aerosol effect of about 0.02-0.04 in
410 visible spectral range over Moscow megacity as well as its temporal dynamics, which has a tendency of AOT decreasing over the “Old” Moscow and increasing over the “New” Moscow territories.

Data availability

The MODIS product data - MCD19A2 Collection 6 (MAIAC aerosol product) and MCD12C1 Collection 6 product were obtained from <https://search.earthdata.nasa.gov/search>. Grid archive of aerosol precursor gases emissions and particulate
415 matter concentrations is available at http://www.ceip.at/new_emep-grid/01_grid_data. The AERONET version 3 data from the Moscow_MO_MSU and Zvenigorod sites are available from the AERONET data repository at <https://aeronet.gsfc.nasa.gov>.

Author contribution

EYuZ and NYC designed the study and wrote the paper with essential contributions from AIL. EYuZ was responsible for
420 data collection and visualization. Data analysis was performed by EYuZ and NYC.

Competing interests

The authors declare that they have no conflict of interest.

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