

(1) *comments from Referees (are marked by italics)*, (2) author's response (plain text), (3) **author's changes in manuscript** (are marked by yellow color).

General comments:

1. I think it will be great if authors provide spatial and seasonal/temporal average pattern of AOD.

Long-term AERONET measurements at Moscow (Moscow_MO_MSU site) demonstrate that seasonal variations of AOT are noticeable with maximum in April and July (median AOT at 0.5 μm are equal 0.22-0.21) and minimum in December and January (median AOT at 0.5 μm is equal 0.07). There are a few previous publications concerning AOT seasonal and temporal variations (for example, Chubarova et al. 2011, Chubarova et al., 2016). We added the discussion about AOT changes in the manuscript in subsection 2.3 (see below). In present research, we considered only warm period of year (May-September). In this period of year, AOT variations are not large (~ 0.15 -0.21).

Spatial variations of AOT are shown at Fig.12 (now Fig.11) in the first version of the manuscript. Our main objective is to discover spatial structure of AOT, to reveal possible local pollution based on MAIAC product over Moscow region.

Changes in manuscript: subsection **2.3 AERONET data**

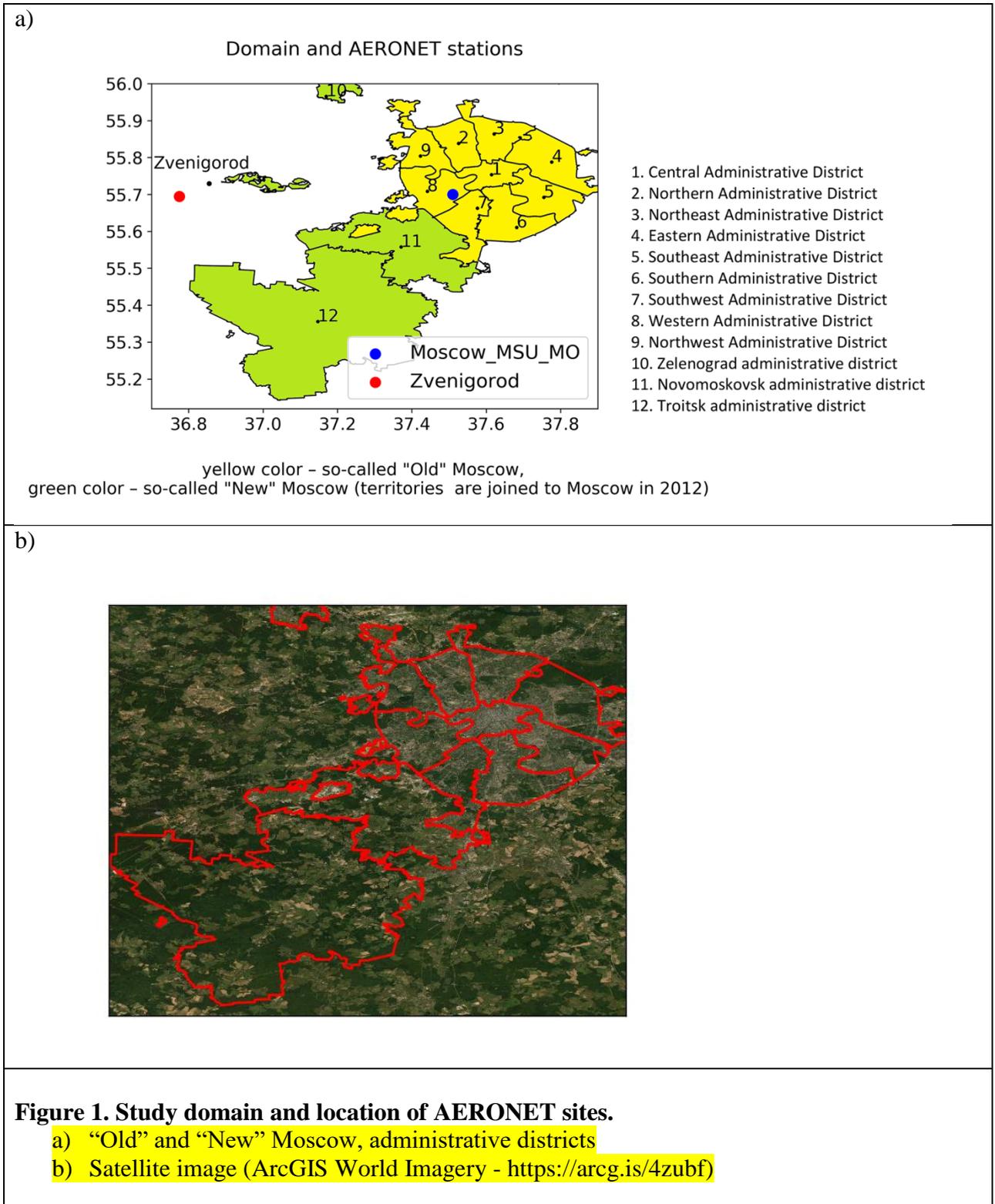
Long-term measurements at the Moscow_MSU_MO have revealed noticeable seasonal changes in AOT with maximum in April and July with median AOT at 0.5 μm of about 0.22, and minimum in December and January with AOT at 0.5 μm of 0.07 (Chubarova et al. 2011b, Chubarova et al., 2016). However, in this study we focused on snow-free period (May-September), during this period of year AOT variations are not large (~ 0.15 -0.21).

Furthermore, on AOD images all important geographic locations must be shown: suburban cities, city center, etc. Readers are not familiar with Moscow geography and it is difficult to follow authors results. Also at the beginning need to explain the differences between New Moscow and Old Moscow under section of "Study Area". And where are these regions on the map? Otherwise i discovered the differences in pollution pattern between both parts only at the end of a paper. Introduction should be devoted to the previous studies done in the subject that are the most relevant to the authors study rather than to study area explanation that should be only briefly explained.

We agree that an additional information about Moscow geography is needed. We updated the Fig.1 providing satellite image and administrate division of Moscow. We moved from Introduction to special subsection information about study area. We called "Old" Moscow is the city territory before 2012 year. In 2012, the Moscow megacity has expanded to the south-west and we called this new territory as "New" Moscow. "Old" Moscow is marked by yellow color and "New" Moscow is marked by green color in Fig.1. We also modified the Introduction, including more references. Please, see the details of changes below.

Changes in manuscript:

Fig. 1 is updated.



2. The study area, datasets and methodology

2.1 The study area

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/>). The study domain is shown in Figure 1. The Moscow city boundaries, its administrative districts and satellite image of Moscow region are shown in Figure 1.

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. Anthropogenic aerosols affect the temperature profile, play important role as a cloud condensation nuclei, impact the hydrologic cycle, through changes in cloud cover, cloud properties and precipitation (Kaufman et al., 2002, Kaufman, 2006).

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 usually correlates with a suspended particulate matter associated with the poor air quality (Wang, J. and Christopher, 2003, Hoff, Christopher, 2009, Chudnovsky et al., 2012, van Donkelaar et al., 2015). Recently a high 1 km resolution aerosol MAIAC satellite product has been used for estimating relationships between AOT and particulate matter (Chudnovsky et al., 2013b, Hu et al., 2014, Kloog et al., 2015, Xiao et al., 2017, Beloconi et al., 2018, Liang et al., 2018, Han 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 (Chubarova et al. 2011a, Kislov, 2017), Warsaw (Zawadzka et al, 2013), Córdoba (central Argentina) (Della Ceca et., 2018) urban areas.

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., 2011a). 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 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 verify the MAIAC aerosol retrievals against the ground-based AERONET measurements over 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. My additional comments relate to the analyses of AOD percentiles (Figure 12- which is interesting). Without a general/AOD average maps, I find it difficult to analyze results of AOD lower/upper percentiles. I also think that these analyses are speculative and must be very carefully presented, more as authors interpretation, as a "hint to local pollution", hint to

regional, etc with references as done in Discussion. May be including this figure in Discussion section would be better? And comparison with ground confirmation of these results? With some critical statements of these results.

In the analysis we decided not to use average AOT values, but to focus on quantile AOT analysis to avoid impact of forest and peat fires causing non-periodic strong AOT inhomogeneity. The events of big forest and peat fires strongly influence the mean AOT values. We think that using of median values is a robust way to show the AOT spatial distribution. We added lines of main roads and highways in Figure 12 (old number, now Figure 11) and now the links of enhanced AOT values with urban emissions (roads, power stations) are seen much better. We also updated Figure 12 (now Figure 11). We found that on the 5% quantile map the maximum of AOT corresponding to large areas of building construction or industry zone and farmlands. We added an additional Figure, where several points of local pollution were shown to associate with location of anthropogenic objects (road, building construction areas). We found these points by visual examination of high resolution satellite images.

Changes in manuscript:

We also applied the quantile analysis to the spatial AOT fields obtained from the MAIAC algorithm separately for the Aqua and Terra datasets and for both of them. The quantile estimates of AOT over the territory of Moscow region are shown in Figure 11 and Table 2. In addition to the 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 (see Fig.1), probably due to highways and industrial enterprises (Fig.11). The spatial changes in AOT over the territory of “Old” Moscow are about 0.03 for wavelength 0.47 μm and 0.55 μm . 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, for example, the areas of building constructions or industrial zones, which can be clearly distinguished in Fig.12. The enhanced AOT over the territory of “New” Moscow are associated with locations of farmlands, which are used in active agricultural activity providing additional aerosol emission. We determined the locations of areas of buildings constructions, industrial zones, farmlands using high resolution satellite images (WorldView-2, IKONOS).

Table 2 presents mean and maximum values of AOT quantiles for the territories of “Old” and “New” Moscow separately for the Aqua and Terra datasets and for both of them. 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 aerosol effect observed in Moscow megacity. Median AOT values according to the Terra dataset are slightly higher (by 0.01-0.02) than the Aqua dataset. The discrepancies in 95% quantile AOT estimates according to these datasets link with the different samples of Terra and Aqua observations.

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.

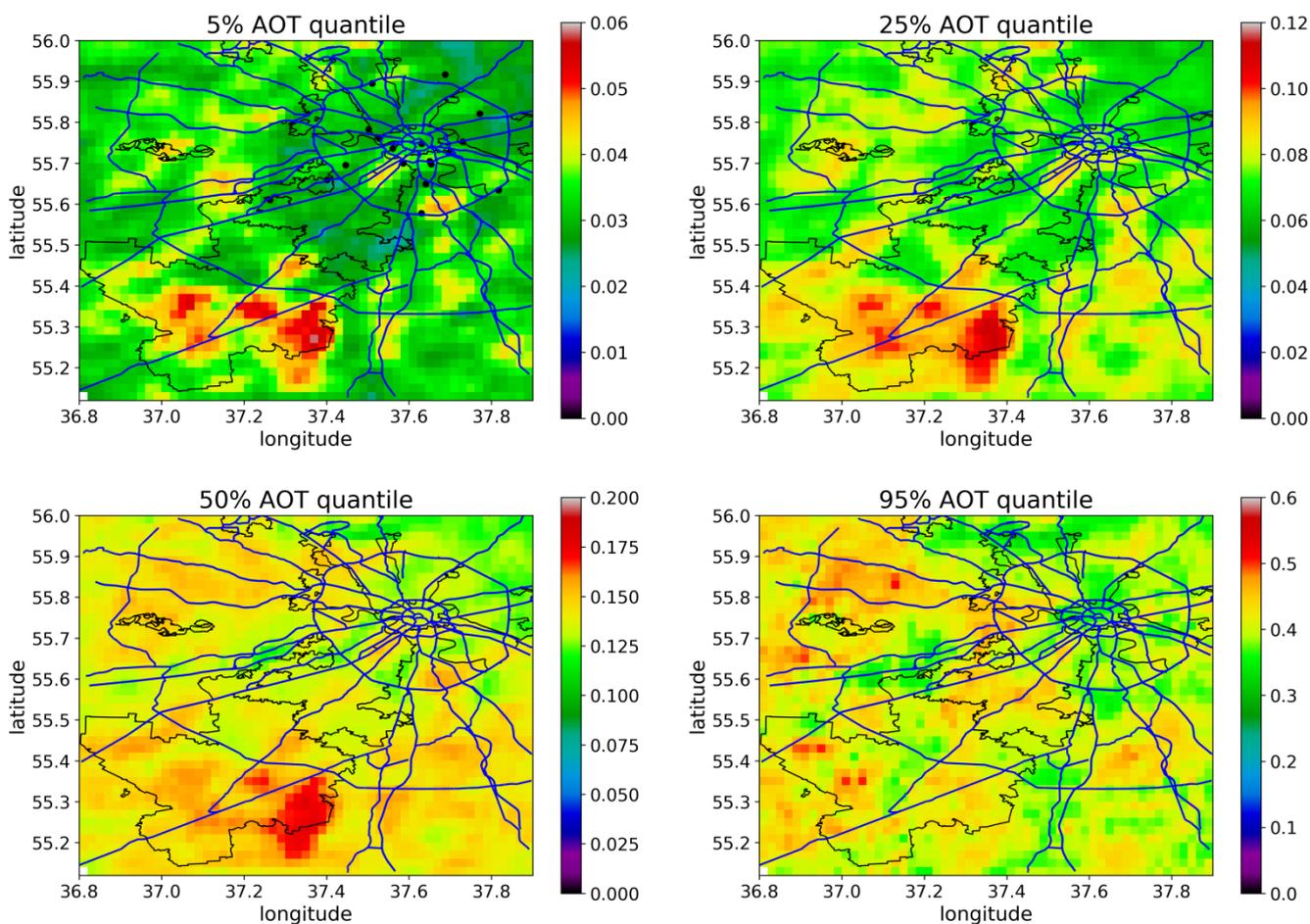


Figure.11. Quantiles (5%, 25%, 50%, 95%) AOT at $0.47 \mu\text{m}$ over Moscow megacity, 2001-2017, Aqua and Terra datasets together. Black points in upper left map are thermal power plants according to the «System Operator of the United Power System» data (<https://www.so-ups.ru/>). Blue lines are the main highways (data: OpenStreetMap - <https://www.openstreetmap.org>)

Table 2. Mean and maximum of AOT quantiles (5%, 25%, 50%, 95%) over the “Old” Moscow and “New” Moscow territories, 2001-2017.

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)
	Aqua			
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
	Terra			
5%	0.03/0.04	0.02/0.03	0.04/0.06	0.02/0.04
25%	0.07/0.09	0.05/0.06	0.08/0.12	0.06/0.08
50%	0.14/0.17	0.1/0.11	0.15/0.19	0.1/0.13
95%	0.42/0.52	0.3/0.37	0.45/0.55	0.32/0.39
	Aqua and Terra			
5%	0.03/0.05	0.02/0.03	0.03/0.06	0.02/0.04
25%	0.07/0.09	0.05/0.06	0.08/0.11	0.05/0.08
50%	0.13/0.16	0.09/0.11	0.14/0.18	0.1/0.12
95%	0.39/0.48	0.28/0.34	0.41/0.51	0.29/0.36

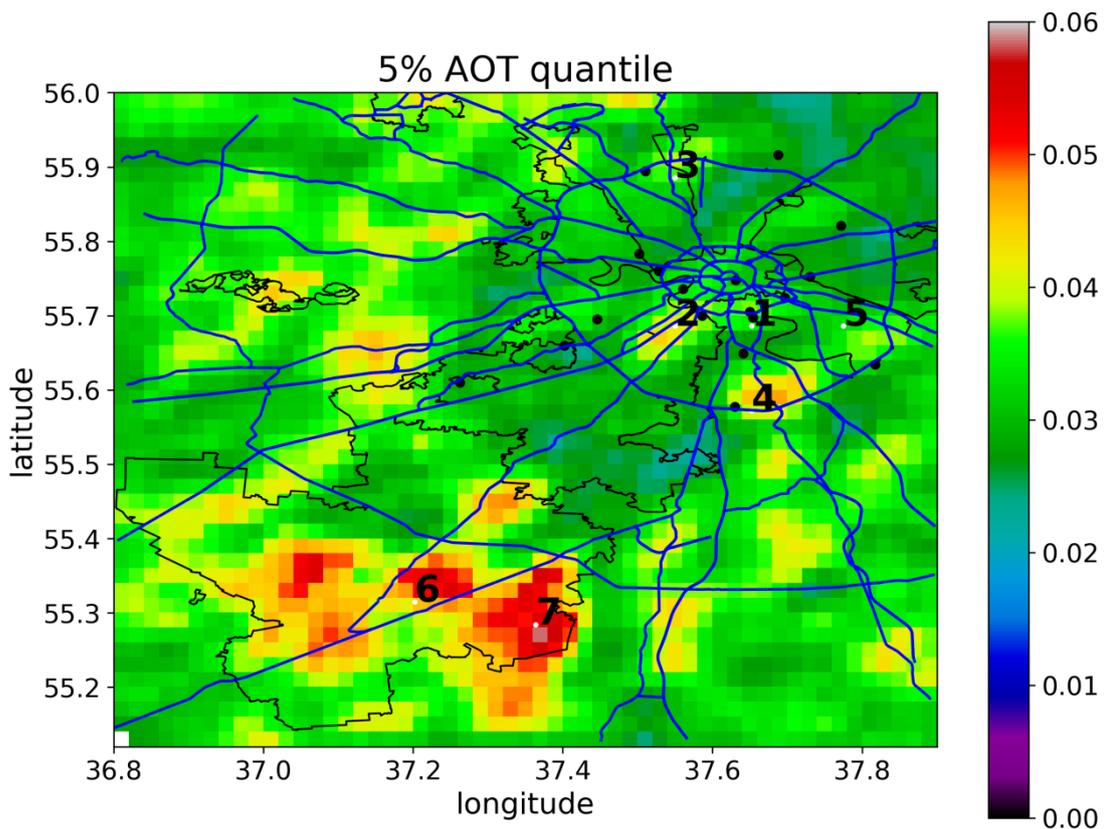


Figure 12. The 5% quantile of AOT at 0.47 μm , 2001-2017. Points on map: 1, 3, 5 – industrial zones with building construction areas, 2, 4 – highways, 6, 7 – farmlands.

3. *May be some figures can be removed as it reduces the paper clarity. Some figures are not explained and not well presented (details are below).*

We removed fig. 7, please see changes below.

Specific/minor comments:

1. Introduction:

Additional literature search is required. For example: - Line 37-39: 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). Authors need to add additional citations that originally investigate the subject. For example, the correlation between particulate matter concentrations and AOD is not a new subject and was widely discussed. As pointed out in Hoff and Christopher 2009 (review article), different geographic locations exhibit different correlations.

Look at Figure 3 in Chudnovsky et al. 2012 "Prediction of daily fine particulate matter concentrations using aerosol optical depth retrievals from the Geostationary Operational Environmental Satellite (GOES)" JAWMA V(62)
- The use of AOD in atmospheric application is excellently presented by Kaufman et al. 2002: "A satellite view of aerosols in the climate system" published in Nature.

- Line 44: Authors stated "recent studies" Although I do not find citations to 2011 or 2013 as recent studies. I searched what was done with MAIAC recently- and perhaps can be relevant- up to authors decision of course: Barnaba et al. 2018: Satellite-based view of the aerosol spatial and temporal variability in the Córdoba region (Argentina) using over ten years of high-resolution data, ISPRS Journal of Photogrammetry and Remote Sensing. And more publications can be found.

Thank you! We added additional discussion in the Introduction, please see edited text of Introduction above. Since the analysis of relationship between particulate matter and AOT is not our main scope we paid on this subject not much attention.

-Line 74: "against the high-quality AERONET measurement". I would avoid such a strong statement as "high-quality" Sometimes even AERONET provide biased measurements. I would suggest "against ground-based AOD measurements".

We did this correction, but we should mention that we use additional cloud filtering by visual observations of cloudiness for AOT AERONET data version 3, so the used data is really tend to be high-quality.

Methods: - Methodology section and data sets-all is mixed up. One needs to dig the information. Please reorganize to sub-paragraphs MAIAC AOD, AERONET data, gaseous pollution data, study area, etc. The same is for results section.

We divided the Section 2. The study area, datasets and methodology into subsections : 2.1 The study area, 2.2. MAIAC data, 2.3 AERONET data, 2.4 EMEP data.
The section Results is not changed and consists of several subsections.

Results: 1. Figure 2: authors need to provide equation for both plots, slope, intercept, r, and explain high residuals on both plots, what are possible causes.

Yes, of course, now we provided fitting equations. We updated figure 2. One can see that the correlation between AOT MAIAC and AERONET is high for Moscow_MO_MSU site, $R = 0.91-0.97$.

Changes in manuscript:

However, the correlation between the AOT MAIAC retrievals and AERONET data is high. Slopes of regressions lines are higher at the Moscow_MO_MSU site than that at Zvenigorod, since at Zvenigorod site high aerosol loading due to forest and peatbog fires has not been included in the sample.

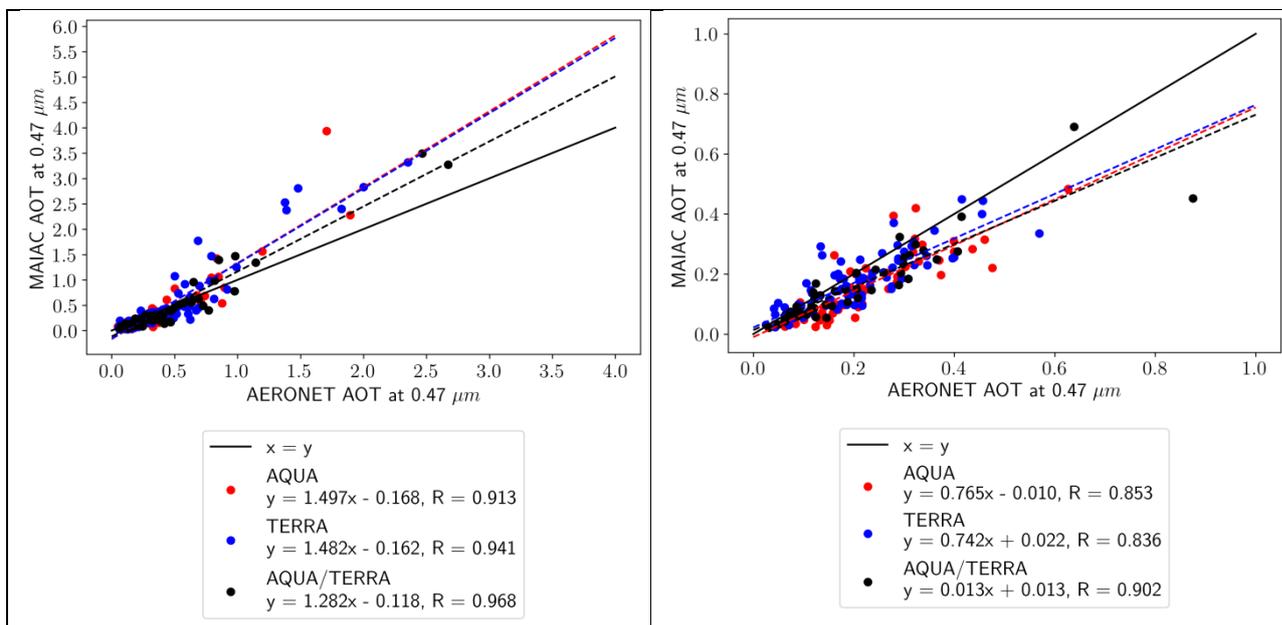


Figure 2. Correlations between MAIAC AOT at 0.47 μm and AERONET AOT at 0.47 μm for Moscow_MSU_MO and Zvenigorod AERONET sites for Terra, Aqua and their joint overpasses within 1 hour (Aqua/Terra).

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.

2. I do not understand Figure 5- it says correlation, but I do not see correlation coefficient, I do not see any pattern except of lack of it. I see a scatter plot with zero correlation. What authors wanted to present? I get puzzled.

We made changes in the manuscript: Fig.5 shows a relationship between ΔAOT from MAIAC and from hourly-averaged AERONET data. The ΔAOT values obtained from both ground-based and satellite data lie within the range of $-0.1 \dots 0.1$. 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 caption of Fig.5 has been changed.

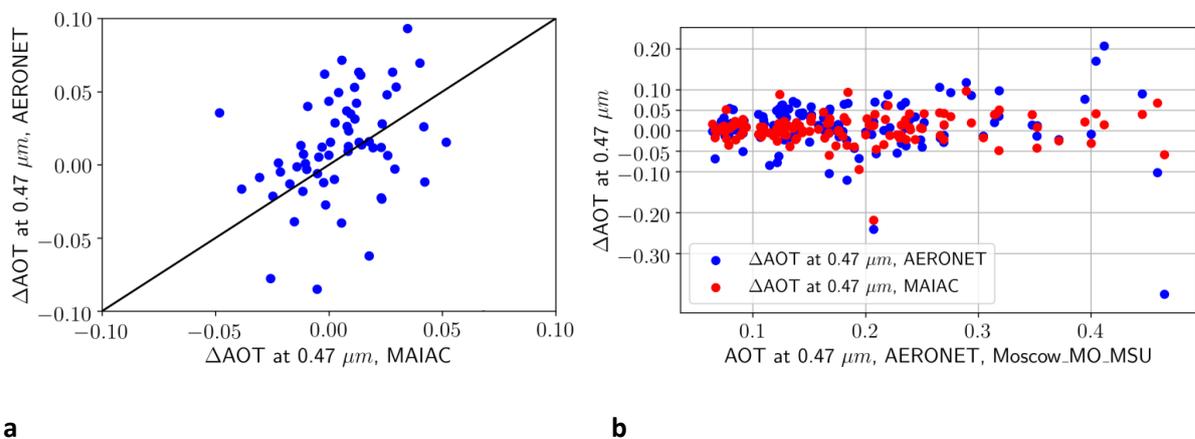


Figure 5. (a) Relationship between ΔAOT at $0.47\mu m$ ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$) obtained from the satellite and ground-based data; (b) ΔAOT at $0.47\mu m$ as a function of AOT at $0.47\mu m$ obtained from Moscow_MSU_MO dataset.

3. Figure 10: I do not understand what median AOD maps present? Why authors can't present average AOD values instead? Please justify your selection.

We chose median values to show robust unbiased AOT spatial distribution. We do not use mean AOT values to avoid impact of forest and peat fires causing non-periodic strong AOT inhomogeneity which significantly influence on the average estimates.

4. Figures 6 and 7 are not explained. Please provide explanations to your results.

We decided to remove Fig.7, because it repeat in some extent Fig.8 and changed the text as following:

For characterizing variations in ΔAOT we analysed frequency distributions according to ground-based and satellite data. In general, polar orbiting satellites demonstrate similar daily average

AOT independent of morning or afternoon orbits (Kaufman et al., 2000). However, we calculated ΔAOT separately for Terra and Aqua datasets for evaluating to some extent diurnal (in the morning and noon hours) variability of ΔAOT . Frequency distributions of ΔAOT at 0.47 and 0.55 μm 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. Fig. 6 also shows a large negative ΔAOT in cases of Terra measurements in our sample. 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.

The diurnal variations of the ΔAOT according to satellite and ground-based data are also shown in Fig.7. The MAIAC ΔAOT at 0.47 μm 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 μm is smaller for satellite data as compared to ground-based data. Satellite and ground-based ΔAOT at 0.47 μm are consistent with each other in the diurnal pattern.

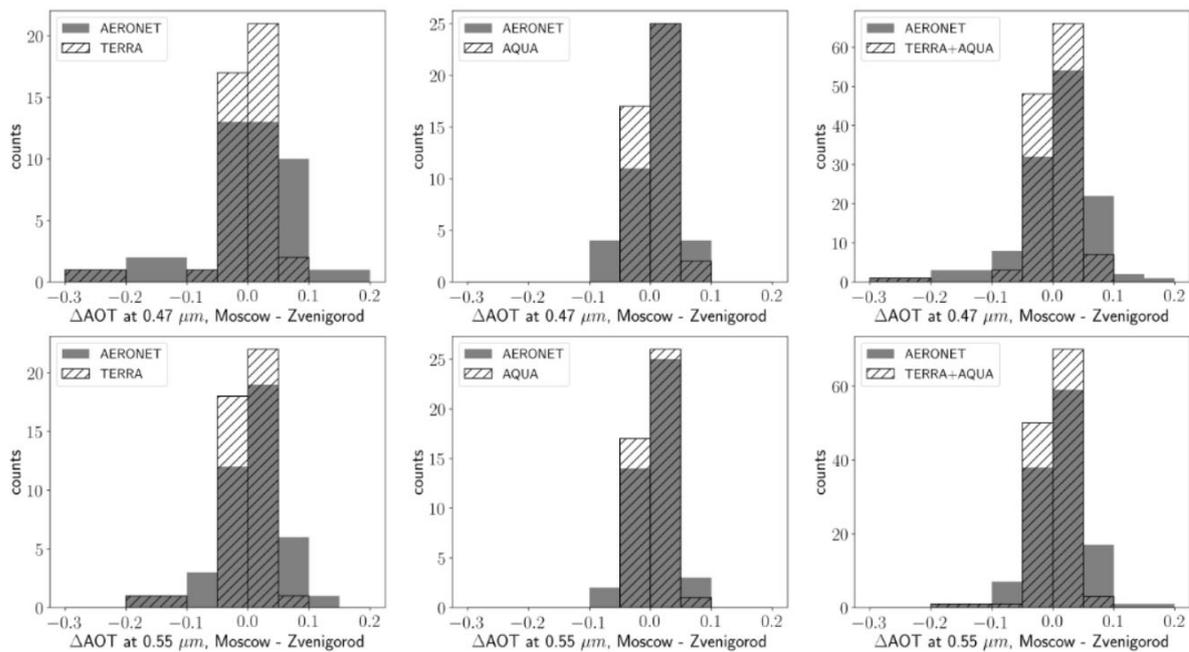


Figure 6. Frequency distribution of ΔAOT ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$) at 0.47 μm (upper) and 0.55 μm (low) separately for the Terra (left column) and Aqua (middle column) datasets, and together for the data from the two satellites (right column) with frequency distribution 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.

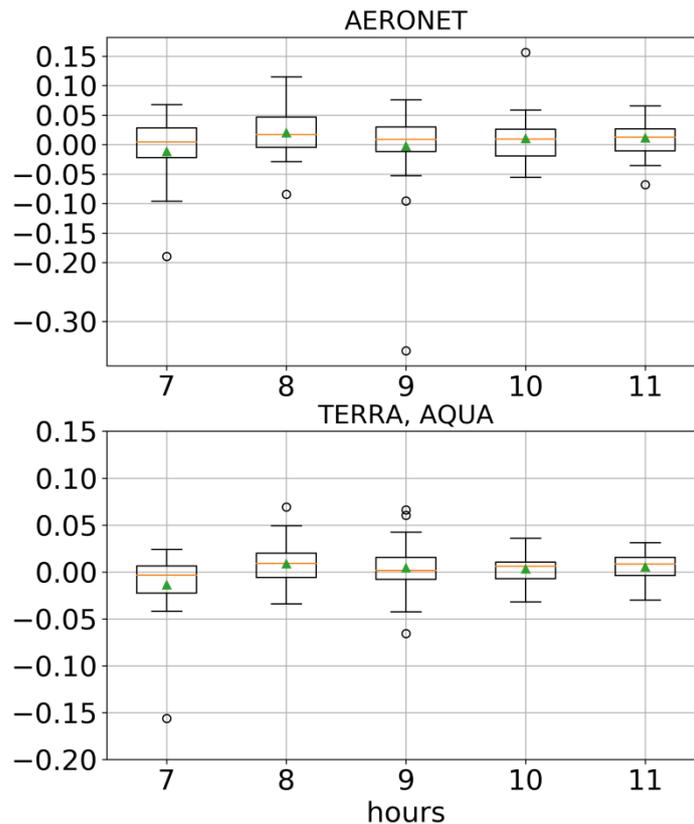


Figure 7. Daily variations of the ΔAOT at $0.47 \mu m$ ($\Delta AOT = AOT_{Moscow_MO_MSU} - AOT_{Zvenigorod}$), UTC time. The median is in the centre, the box is the first (Q1) and the third (Q3) quartiles, the whiskers are $Q3 + 1.5 * (Q3-Q1)$ and $Q1 - 1.5 * (Q3-Q1)$, green triangles – means, points – outliers; (2006-2017, without the data of 2009 because of technical problems at Zvenigorod AERONET site). Number of satellite and ground-based matchups is 125.

Discussion:

Authors need to provide discussion on points that overestimated and underestimated by MAIAC AOD retrieval at least by showing what are meteorological conditions that favor these results. I mean- analyses of residuals (from figure 2).

In this research, we paid main attention to the analysis of aerosol model used in MAIAC AOT algorithm. We showed that AOT MAIAC were overestimated for smoke conditions with $AOT > 1$ due to spatial and temporal variability of smoke properties, which can be various in different geographical regions. Cases studies of influence of meteorological conditions on AOT MAIAC product is the issue of our future research, which is now mentioned in the text.

Authors also need to state the limitations of their results and future directions in one short paragraph.

We added concluding remarks, please see below

Changes in manuscript:

Thus, the application of the new MAIAC algorithm provides a reliable instrument for assessing the spatial distribution of aerosol pollution and allows us to evaluate the level of local aerosol effect of about 0.02-0.04 in 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.

In this research we have verified the MAIAC algorithm data against ground-based data and obtained spatial and temporal variability of AOT MAIAC retrievals over Moscow region for evaluating aerosol pollution. Future studies focused on influence of different meteorological conditions on AOT MAIAC retrievals will be valuable for detection events of the extreme urban aerosol pollution and further MAIAC product validation.

All changes in the manuscript are marked by yellow color.