

*We thank the reviewer for the important comments, which help to improve the manuscript.*

### **The answers on the anonymous Referee Comment #1.**

1/. Explain the statistical test you used. How did you consider the uncertainty in AERONET-retrieved AOD? It is 0.01-0.02 (Eck et al. 1999). You say that the difference of 0.02 is statistically significant. Please give the details behind this conclusion, considering the AERONET uncertainty which is about the same size than this difference. The AERONET-uncertainty should be considered and discussed throughout, regarding AOD differences, Angstrom Exponent, SSA.

*All the important information has been included in the manuscript. The new version of the manuscript has much more details on different points: uncertainties, statistical approach, etc. In the section 2 “Data and methods of the analysis” several parts have been added:*

Direct Sun measurements are made with 1.2° full field of view at 340, 380, 440, 500, 675, 870, 940 and 1020 nm every 15 minutes during daytime (Holben et al., 1998). These measurements are used to compute aerosol optical thickness except that for 940 nm channel, which is used to estimate the total water content W. The uncertainty of AOT measurements does not exceed 0.01 in visible range and 0.02 in UV spectral range (Eck et al., 1999) for field instruments. Direct Sun measurements also provide the data for estimating the Angstrom exponent from spectral values of AOT by the least square method. The sky radiance measurements at 440, 675, 870, and 1020 nm in conjunction with the direct Sun measurements are used to retrieve different microphysical, optical and radiative aerosol characteristics (Dubovik and King, 2000). The accuracy of individual aerosol retrievals is analyzed in (Dubovik et al., 2000, 2002). It was shown there that for high aerosol loading ( $AOT_{440} > 0.4$ ), the retrievals of the single scattering albedo have the uncertainty of about 0.03 while at AOT at 440nm less than 0.2 the accuracy level drops down to 0.05-0.07. The error in aerosol volume size distribution within the intermediate particle size range ( $0.1 \mu m \leq r \leq 7 \mu m$ ) does not exceed 10% for practically all situations (Dubovik et al., 2000).

.....

The analysis of the differences between the aerosol data at the two sites were made by the standard approach by estimating the significance of the difference of an aerosol parameter between two samples. We showed that the differences of all the parameters has passed successfully the Kolmogorov- Smirnov test on normal distribution (Afifi, and Azen, 1979) and , hence, the standard t-Student criterion (ts) can be applied for calculation of the confidence level  $\delta$  for each mean value of an aerosol parameter as follows:

$$\delta_{AP} = \pm \frac{(ts \cdot \sigma_{AP})}{\sqrt{N-1}} \quad (1)$$

where

$\sigma_{AP}$  – the standard deviation of the difference  $\Delta P$  obtained between the CIMEL measurements at the two sites,  $P$  is the parameter analyzed;

$N$ - number of cases;

$ts$  –  $t$ -statistics for the Student’s distribution.  $ts = 1.96$  when  $N \rightarrow \infty$  at 95% significance level.

In addition, since the aerosol optical thickness is the object of the main interest, we have fulfilled additional analysis accounting for the uncertainty of measurements using the following expression:

$$\delta\Delta AOT = \sqrt{\left(\frac{ts \cdot \sigma_{\Delta AOT}}{\sqrt{N-1}}\right)^2 + (\delta AOT_1)^2 + (\delta AOT_2)^2} \quad (2)$$

where

$\delta AOT_1$  and  $\delta AOT_2$  are the typical systematic instrumental errors (0.01 in visible range) of Moscow and Zvenigorod instruments.

$\sigma_{\Delta AOT}$  – standard deviation of the difference  $\Delta AOT$  obtained between the CIMEL measurements at the two sites.

*This test also provides the statistical significance of the difference in AOT as it is written now in the section “Results”:*

The mean overall difference in AOT is about 0.02, which is statistically significant at the 95% level. The additional test with account for the uncertainty of AOT CIMEL measurements also shows the significance of the differences for all the seasons....

*The discussion on size distribution uncertainties have been included:*

A comparison between the mean aerosol size distributions over Moscow and over Zvenigorod shows a considerably higher concentration of coarse mode particles in Moscow, especially, near radius of 5  $\mu\text{m}$  (about 31%) that is much higher than the uncertainty of measurements (Fig. 7). Also a higher concentration of fine mode particles near 0.1  $\mu\text{m}$  is observed. The difference in fine mode concentration should be studied further, because of possible  $\text{NO}_2$  contamination in Moscow, as discussed above, which can be attributed to an artificial increase in fine mode particles.

*The SSA comparisons with account for SSA uncertainties were discussed in the section “Results” as well:*

Since the inversion method requires the threshold of  $\text{AOT}_{440} > 0.4$  and typical AOT's in Moscow are relatively low (mean  $\text{AOT}_{440} \sim 0.23$ ), there are only few cases in SSA retrievals. It is necessary to mention that this is a quite typical situation for high latitude boreal zone. Over these areas relatively high AOT values are observed mainly in smoke aerosol conditions. Therefore, in addition, we used other thresholds ( $\text{AOT} > 0.3$ ,  $\text{AOT} > 0.2$ ,  $\text{AOT} > 0.1$ ) and all AOT statistics to analyze SSA for larger number of cases at different aerosol loading. The mean SSA in Moscow and its difference with Zvenigorod at different AOT thresholds are shown in Fig. 9 and in Table 2. One can see the absence of the difference in SSA between Moscow and Zvenigorod at large  $\text{AOT} > 0.4$ . At the same time there is a tendency of SSA decreasing in Moscow with AOT decrease. The difference can reach  $d\text{SSA} = -0.03$  when considering all available measurements of the Dataset 2 (see Table 2). However, even this difference is equal to the uncertainty of the SSA retrievals, while SSA retrievals at  $\text{AOT}_{440} < 0.4$  have even the larger uncertainty of measurements (Dubovik et al., 2000). Taking this into consideration, the obtained difference can be considered only as a preliminary result.

2/ The descriptions were occasionally difficult to follow. Just as an example, in lines 15-15 of the block 5473. You say that the difference in M1 can be under-estimated due to the different number of observations. Earlier you say that the difference is based on pairs of quasi-simultaneous measurements. If

you have pairs, why do you have different number of observations in the analysis. And even if you do have, for some reason, why it should result in underestimation, in other words why the effect would be systematic?

*In the text the M1 dataset corresponds to the standard AERONET monthly mean dataset of level 2 obtained separately for Moscow and Zvenigorod. Monthly mean values have been calculated using different number of observations at the sites. M2 dataset is our more accurate approach based on the strict pairs of observations. Our aim was to compare the differences between the two approaches. See the revised text below:*

As a result, this dataset (Dataset 1) contains the pairs of quasi-simultaneous measurements at the Moscow and Zvenigorod sites. Total number of the hourly averaged cases is about 1200. In order to examine the quality of the Dataset 1 we compared the monthly mean differences in AOT taken from this dataset with the differences in AOT taken directly from the AERONET website for Moscow and Zvenigorod sites. Fig. 1 presents the comparison between the AOT differences obtained by the two methods: the standard AERONET method (marked as the M1 method) and the more accurate approach, which has been described above (marked as the M2 method).

One can see that the more accurate second approach (M2) provides the absence of negative monthly mean AOT differences compared with the results of the M1 method. These negative AOT differences correspond to the situations, when Moscow AOT's are smaller than those in Zvenigorod. So the absence of negative monthly mean AOT differences in M2 approach is more reasonable, since Moscow should provide some additional emission of aerosol particles. Overall, the application of the more accurate method provides the difference of  $\pm 0.05$  with the standard approach for monthly mean AOT values.

3/ Regarding the section 3,

- You go directly to the differences, it would be informative to have first seasonality of AOD (for one site or both) to get an idea about the AOD variability too, not only about the differences. Also, is there seasonality in the prevailing wind directions?

*The new version of the text includes the brief analysis of the seasonal behavior of AOD and the reference to the paper with the detailed study of the AOD climatology (see below). Their absolute values have been included in Table 1. Concerning the prevailing wind direction, typically we have prevailing westerlies but the direction can change significantly in each season due to very often changing synoptic processes.*

Table 1 shows mean aerosol characteristics in Moscow and the statistics of the differences between Moscow and Zvenigorod ( $dP = P_{\text{Moscow}} - P_{\text{Zvenigorod}}$ ) for various characteristics observed in different seasons. The seasonal changes in AOT in visible spectral range has a pronounced minimum in winter of about 0.1, while in other seasons it is about 0.2 (for the detailed analysis of the AOT climatology see, for example, Chubarova (2009)).

There are two type of speculative conclusions in the Results -section: 1) some of them could be further assessed, 2) some of them should be better explained/justified. About the first category, you mention that SCIAMACHY resolution may play a role. It would be very easy to take OMI, with better resolution, and focus on Moscow, to get evidence if this is the case.

The standard AERONET analysis for correcting NO<sub>2</sub> in the atmosphere uses the SCIAMACHY climatology of NO<sub>2</sub> for the period 2003-2005 with the spatial resolution of 0.25 x 0.25 degrees. (see [http://aeronet.gsfc.nasa.gov/new\\_web/publications.html](http://aeronet.gsfc.nasa.gov/new_web/publications.html)).

According to it, for example for the 2008-2009 period, the mean NO<sub>2</sub> content in Moscow was 0.44 DU and in Zvenigorod 0.29 DU (correspondingly about  $1.18 \cdot 10^{16}$  mol/cm<sup>2</sup> and  $0.78 \cdot 10^{16}$  mol/cm<sup>2</sup>). So the total difference is about  $0.4 \cdot 10^{16}$  mol/cm<sup>2</sup> or 0.15 DU.

We did not take OMI data because of still not excellent OMI spatial resolution (13x24 km at nadir). Instead, we took the available climatology of the total NO<sub>2</sub> content obtained from ground-based direct spectral measurements at each site. According to these climatology obtained in (Elokhov, A.S. and Gruzdev, A.N., 1998, Measurements of column contents and vertical distribution of NO<sub>2</sub> at Zvenigorod Scientific Station. Proceedings of SPIE, 3583, pp. 547–554 and Gruzdev, A. N. and Elokhov, A. S.(2010) 'Validation of Ozone Monitoring Instrument NO<sub>2</sub> measurements using ground based NO<sub>2</sub> measurements at Zvenigorod, Russia', International Journal of Remote Sensing, 31: 2, 497 — 511) the typical total NO<sub>2</sub> content in Zvenigorod is about  $4 \cdot 10^{15}$  mol/cm<sup>2</sup>. In some days (~ 20% in summer) the pollution advection affects the NO<sub>2</sub> increase up to  $7 \cdot 10^{15}$  mol/cm<sup>2</sup>. Thus, we can have **the absolute maximum** NO<sub>2</sub> content there of about  $0.7 \cdot 10^{16}$  mol/cm<sup>2</sup> (or ~0.3 DU) while this value corresponds to the **mean NO<sub>2</sub> content (0.29 DU)** according to SCIAMACHY( see above).

At the same time in Moscow according to the similar measurements and approach only tropospheric NO<sub>2</sub> content shows high mean NO<sub>2</sub> content (around  $1.5\text{--}2 \cdot 10^{16}$  mol/cm<sup>2</sup> or 0.5-0.7 DU) reaching up to  $4 \cdot 10^{17}$  mol/cm<sup>2</sup> (Ivanov V.A., A.S. Elokhov, O.V. Postilyakov, I.B. Belikov Preliminary results of boundary layer nitrogen dioxide integral content in Moscow area.2010 In the proceedings of "Current problems in remote sensing of the Earth from space", N 7 V.1. p. 92-98. ). Adding the stratospheric NO<sub>2</sub> content (~ 0.1 DU) we obtain mean NO<sub>2</sub> content of about 0.6-0.8 DU. So the approximate difference between the sites should be around 0.3-0.5 DU if we take even the absolute maximum NO<sub>2</sub> content at Zvenigorod. In the standard AERONET algorithm the difference (as mentioned above) is only 0.15 DU that is too low. This proves our idea that the difference in AOT is due to not full account for the NO<sub>2</sub> content in the current AERONET version 2 algorithm.

*This additional information was added to the text.*

*The paragraph now is rewritten as follows:*

... The spectral dependence of the AOT average difference between Moscow and Zvenigorod is shown in Fig. 3. One can see the existence of a quite noticeable maximum at 380- 440 nm, which can be attributed to the additional effects of higher NO<sub>2</sub> content in Moscow (Chubarova et al., 2008), which possibly is not fully accounted for in the AERONET dataset (see a similar shape in NO<sub>2</sub> absorption coefficients in Fig. 3). This difference can correspond to an additional NO<sub>2</sub> content of about 0.3 DU in Moscow and can be seen both in clear sky and all-sky conditions. We should mention that the standard AERONET version 2 algorithm uses the SCIAMACHY NO<sub>2</sub> climatology for the period 2003-2005 with a spatial resolution of 0.25°x0.25° for correcting NO<sub>2</sub> content in the atmosphere (see [http://aeronet.gsfc.nasa.gov/new\\_web/publications.html](http://aeronet.gsfc.nasa.gov/new_web/publications.html)). According to this climatology, the mean difference in NO<sub>2</sub> content between the sites is about

0.15 DU (or  $0.4 \cdot 10^{16}$  mol/cm<sup>2</sup>). The analysis of the climatology of the direct NO<sub>2</sub> measurements by spectral instruments have shown much higher difference of 0.3-0.5 DU (Elokhov and Gruzdev, 1998, Gruzdev and Elokhov, 2010, Ivanov V.A. et al., 2010) which corresponds much better to our estimates.

The SCIAMACHY NO<sub>2</sub> retrievals, which are used for NO<sub>2</sub> correction in the AERONET algorithm, can be lower in Moscow, to some extent, due to comparatively large space averaging, which combines both clean and polluted areas.

- About the second type of conclusions (just one example), discussion in lines 20-28 in the block 5475 about the dominance of natural processes, only based on the correlation in Angstrom exponent, is not clear and with obvious evidence.

*This means that since we consider Zvenigorod as a clean site, high correlation in AOT with Moscow data means the prevailing of non-urban aerosols. The correlation in Angstrom parameter values also confirms that in most cases we have similar size distribution. The statement has been smoothed and some changes were made in addition.*

*New version:*

..The correlation between the Angstrom exponent values together with high correlation between the AOT's can mean that for Moscow conditions the natural processes are likely the dominating factor in transformation of the aerosol particle size distribution."

- Figure 10 is explained in Conclusions, why not in Results? Also, it is not clear how exactly the "temporal lag" was accounted for. What data and how used.

*The text has been changed. Now Figure is in the section "Results". We agree that the description in the text is not sufficient. Some additional material has been added:*

In order to account for the possible effects of the temporal lag in AOT the correction to the AOT's was applied using the obtained linear regression equation between dAOT500 and dW:

$$dAOT500 = 0.21 dW + 0.02, \quad r = 0.52 \quad (3)$$

Fig. 5 presents frequency distribution of the initial dAOT500 dataset and the dataset corrected on the air transport temporal lag. As a result, we obtained an increase in the occurrence of positive dAOT (more than 75% of cases compared with the 72% calculated using the initial dataset), the decrease in dAOT standard deviation from 0.05 to 0.04, and the same average difference of about 0.02. It is clearly seen that the removal of this factor does not change significantly the mean results but it certainly leads to even more pronounced aerosol pollution effects with smaller number of negative dAOT cases.

- Here, as in many places more generally, the reader was not provided with the sufficient details. The lack of sufficient details is the major weakness in the current form the manuscript. Once those will be given and the descriptions and conclusions will be clarified, it will be possible to evaluate the manuscript.

*We tried to include as many clarifications in the new version of the manuscript as possible. The new paragraphs have been added in the different parts of the text including the definition and clarification of aerosol radiative forcing:*

Aerosol radiative forcing (ARF) at the top of the atmosphere (TOA) is used for characterizing the impact of aerosol on the temperature regime. Since the standard AERONET radiation products include the calculation of  $ARF$  (Garcia et al., 2008), we used this characteristic to estimate the influence of the large city on its changes. The AERONET aerosol radiative forcing is defined as the difference between the global solar irradiance with and without aerosol at the top and at the bottom of the atmosphere.

$$ARF_{TOA} = -(F^{\uparrow a}_{TOA} - F^{\uparrow o}_{TOA}) \quad (4)$$

$$ARF_{BOA} = (F^{\downarrow a}_{BOA} - F^{\downarrow o}_{BOA}), \quad (5)$$

where  $F^a$  and  $F^o$  are the broadband fluxes at the top (TOA) and at the bottom (BOA) of the atmosphere with and without aerosols. However, when we speak about the ARF difference at the top of the atmosphere between the two sites, the resulting value will be the same as if considering the net fluxes, which are usually used in the ARF analysis (see, for example, Yu et al., 2006). According to the statistics shown in Table 2, mean  $ARF$  at the top of the atmosphere is about  $-0.9 \pm 0.6 \text{ W/m}^2$ .