Atmos. Meas. Tech. Discuss., 8, C3356–C3426, 2015
www.atmos-meas-tech-discuss.net/8/C3356/2015/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Long-term variability of aerosol optical thickness in Eastern Europe over 2001–2014 according to the measurements at the Moscow MSU MO AERONET site with additional cloud and NO₂ correction" *by* N. Y. Chubarova et al.

N. Y. Chubarova et al.

natalia.chubarova@gmail.com

Received and published: 19 October 2015

Questions and answers on referee's comments: We would like to thank very much Dr. Thomas Eck and anonymous referee for the valuable comments to the paper. In accordance with the comments we substantially changed the manuscript, and especially the section 3.1.1 ("Additional cloud-screening procedure and its effect on aerosol climatology"), and the section 3.3 ("Long-term AOT trends in Moscow and their possible reasons"). Several additional Figures have been also added . Since the EMEP database has been recently updated, we have included the larger data series of aerosol precursor emissions in the section 3.3 and some numbers were slightly changed. We also added some additional material in the analysis of natural factors influence on the evaluated AOT trends. However, the application and the analysis of the additional data do not change the conclusions. For better understanding where the changes had been made, in addition, we have added the preliminary updated version of the manuscript together with the Figures in the supplementary attachment.

Here we combined our answers to the comments of both reviewers, which are given together with their remarks.

The answers to Dr. Thomas Eck comments: 1.1. Although it was not expected to be a part of this draft of the paper, it is worth mentioning here that the upcoming AERONET Version 3 database utilizes a new monthly mean climatology of total columnar NO2 from the OMI satellite sensor data and that these values are significantly higher than those of the SCIAMACHY database for the Moscow_MSU_MO site.

Response: Yes, I think this is important to mention. We included this point in the text in the Discussion section.

Changes in the text: The new text is the following at the old variant page 7860 line 5 , or line 653 in new numeration: "It should be emphasized that the upcoming AERONET Version 3 database utilizes a new monthly mean climatology of total columnar NO2 from the OMI satellite sensor data and that these values are significantly higher than those obtained from the SCIAMACHY database for the Moscow MSU MO site (T. Eck, personal communication)."

1.2. The authors have additionally screened all AOT data where the cloud fraction exceeds nine tenths (i.e. overcast sky conditions) from March through October as determined by human visual observations. This threshold is dropped to greater than 6 tenths cloud cover for the other months (November through February). In my opinion this is a very general and non-rigorous method to screen clouds from data of direct sun observations, especially so for the November-February months. The human observation interval is hourly at this station, therefore measurements made automatically by AERONET sun-sky radiometers at 15 minute intervals (nominal) may be taken in gaps in cloud cover that occur between human observer estimates. Additionally, the relatively low cloud cover threshold selected for Nov – Feb of >60% does not account for the fact that at times the sun may be un-obscured by clouds when most of the rest of the sky is cloudy.

1.2. Response: I agree that it is much better to apply TSI or any other type of fish-eye camera, or MPL data. However, every method has its own shortcomings. You are right, of course, that this method is not rigorous but according to our additional analysis shown in the updated version of the paper, the method nevertheless works well and it may be helpful, especially for studying the historical data sets. We have made a lot of clarifications and added more material especially in the section 3.1.1. We also included several additional Figures to describe the approach applied in this study. Some additional checks were made for estimating the % of filtering the “good” measurements during November-February due to the application of more rigorous threshold NA<6. The analysis of additional dataset on solar disk conditions has revealed only 0.5% of wrong removed cases, which were taken in winter with more strict filter NA<6. Using 1-minute resolution direct solar irradiance measurements we tried to check the possibility of CIMEL measuring in cloud gaps. We added a Figure which demonstrates that usually measurements were not made in cloud gaps. All the analysis has been now included in the text. The parts of the changed text is given below.

1.2. Changes were made in the Section 2 (“Data description”) , directly in 3.1.1 Section. Note, that several Figures were also added. In the section 2 we added the description of the collocated solar disk conditions observations, which were in addition used for checking the proposed cloud filtering: “For the improvement of these procedures, in addition, we used visual cloud observations with 1 hour resolution. The uncertainty

of visual cloud amount measurements is about 1 or 2 cloud fraction (in tenth) according to (Handbook, 1989), however, the conditions with overcast or zero cloudiness are observed accurately by any observer. In addition, the dataset of hourly solar disk condition observations, which are performed simultaneously with cloud observations, was used in the analysis. This is a standard type of observations at the actinometrical stations in Russia. Using this characteristic we can distinct the conditions, when solar disk (SD) is free from clouds, or when SD is obscured by thin clouds (shadows can be seen at ground), or when SD can be seen but there are no shadows at ground, or when SD can not be seen due to relatively high cloud optical thickness. These SD conditions are noted with “2”, “1”, “0” and “P” marks, respectively”.

In the section 3.1.1 the changes were made after the sentence “Will it significantly affect the aerosol climatology?”(line 124 in the new numeration): “For evaluating the upper layer cloud contamination in the AERONET dataset different approaches are used. In the recent studies the ground-based MPLNET, as well as satellite CALIPSO and MODIS datasets were used for evaluating the cirrus AOT contamination (Chew et al., 2011, Huang et al., 2012). According to MPLNET data the AOT bias due to unscreened cirrus cloud presence is about 0.03-0.06 with the occurrence of 23-34% depending on the method of the estimation over the tropical region in Singapore (Chew et al., 2011). Huang et al. (2012) evaluated the susceptibility percentage of AERONET level 2.0 AOT retrievals to cirrus contamination using different types of measurements. According to MPLNET cirrus flags this value varied from zero to ïA¿4%, according to the collocated Calipso cirrus flags - from 1% to 33%, and according to the MODIS cirrus flags - from 0.4% to 18% changing significantly over the globe due to the different occurrence of cirrus clouds. However, satellites have relatively low overpass frequency over AERONET sites - one-two time a day – for MODIS, and 16-day repeating cycle - for CALIPSO (Huang et al., 2012). The MPLNET application for cirrus flags has the problem with viewing geometry difference between the sunphotometer and the MPL. In addition, the MPL signals are extremely weak at the altitudes H> 10 km, where cirrus clouds may be observed. It was also mentioned in (Huang et al., 2012) that the

MPL noise level dramatically increases during daytime especially at noon, when there are most favourable conditions for AERONET-MPLNET matchup from the point of view of the closeness of viewing geometries. The influence of the cloud contamination on aerosol properties was also discussed in (Uliumdzhieva et al., 2005) for Moscow conditions. In this paper the application of the standard cloud visual observations as an additional cloud-screening filter was proposed. We used one-hour resolution cloud observations for additional filtering of quasi-simultaneous Cimel observations at the same site. The application of cloud filter means the elimination of all AOT measurements for the entire hour interval. We showed there that the existing standard cloud-screening algorithm works perfectly, when aerosol measurements are contaminated by optically thin low layer cloudiness, which is characterized by large triplet variations. These variations are used as a parameter in the standard cloud-screening algorithm developed by Smirnov et al. (2000). However, if the cloud blocking the Sun is thin and uniform, the triplet variation can be small and the contaminated AOT measurements pass through the filter. Mainly the cirrus clouds are characterized by these properties. However, in general, according to the International Cloud Atlas (1987) other types of clouds may be also characterized by these properties as well. They include different forms of Cirrostratus, and even Altostratus translucidus clouds, which relate to the middle cloud level. In this publication all types of the cloudiness, which can induce the potential contamination of AOT will be combined under the term “upper cloudiness”. Since low cloudiness is effectively filtered out by the standard cloud-screening algorithm we proposed to apply simple total cloud amount (NA) filter, which is sensitive to the existence of upper cloudiness. In this context NA value (together with the application of standard cloud-screening procedure) provides the information about the potential existence of high and middle layer cloudiness, since the standard AERONET cloud-screening algorithm successfully removes the cases contaminated by low level clouds or the cases with strong signal variations. However, the application of different NA thresholds may provide the different samples and as a result, different statistics. As an example, in Fig.1 we demonstrate the effects of utilizing the different additional NA filters for AOT at 500nm (AOT500) and Angstrom exponent (ïĄą) datasets for the central months of the seasons. One can see a similar tendency of the AOT decrease in all seasons after removing the cases with the threshold of NA<9, which includes the conditions with almost total cloud amount of NA= 9-10. Additional testing on solar disk conditions has revealed that all eliminated cases in this sample belong to the situations, when solar disk was covered by clouds (SD=1 or SD=0). In April after eliminating the almost overcast cloud conditions with NA<9 threshold there is no further changes in AOT500 samples with more strict NA cloud threshold. At the same time the sample is dramatically (more than twice) reduced (from 229 to 113 in NA<3 sample). In July we also see a slight decrease in AOT500 in the NA<9 sample and, in addition, a significant growth of Angstrom exponent. Note, that in July and October we see even a slight increase in the AOT500 for almost clear sky conditions (the sample with NA<3). In January there is a pronounced decrease in AOT500 with reducing cloud amount. The lowest AOT500 values and largest Angstrom exponent are observed in the NA<3 sample. The application of the 24-hour Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) NOAA model backward trajectory analysis (Draxler and Hess, 1998) for all the cases in January has revealed for the NA<3 sample the prevalence of the northern (North-West, North, North-East) advection (80% of cases) characterized by low AOT (Chubarova, 2009), compared with 40% of cases for partially cloudy conditions (3<NA<9). At the confidence level of P=80% several of these dependencies are statistically significant (see the error bars in Fig.1). Balancing between the substantial decrease in case number and the accuracy of the retrievals of aerosol properties we showed that the best results were obtained when the cases with Ntotal.<9 during March-October period with almost overcast cloudiness and cloud contaminated solar disk conditions data were removed. For November –February conditions the filter threshold is more strict (Ntotal<6) since solar elevation in Moscow is low (hnoon<25ïĆř) at this time and a well-known effect of significant visual cloud amount increase towards the horizon plays a vital role. Smaller cloud threshold during winter time may induce filtering out so called “good” AOT cases. The additional analysis of solar disk conditions has revealed only 12 cases from 2521 cases (about 0.5%), which were not contaminated by clouds (SD=2) and were incorrectly removed from the sample. All of them were observed in February (18.02.2011 – 4 cases, 3.02.2003- 5 cases, 11/02/2007 - 3 cases). However, even these three days are presented in the final sample since during these days there were other sun photometer measurements at smaller NA. In addition, we analyzed the 1-minute resolution direct solar measurements with the standard Russian actinometer (WMO, 1986) for studying the possibility of AERONET direct solar irradiance observations in cloud gaps,. These data were used for estimating the standard transparency coefficient at air mass m=2 according to (Evnevich, Savikovsky, 1988): p_2=(S_h/1.367)ˆ((sinh+0.205)/1.41), (1) where S_h - is the measured value of direct shortwave irradiance, h - solar elevation.

The p2 coefficient is widely used for assessing the variation of the transparency of the atmosphere (Ohvril et al., 2009). Using this equation we evaluated the integral optical thickness as τ=-lnâĄą(p_2). This characteristic was used as the indicator for the analysis of possible AERONET CIMEL sun photometer measurements in cloud gaps. The 1-minute resolution data provide us the time series to check that CIMEL 15-minute resolution measurements can be observed in these cloud gap conditions. Since the duration of sunphotometer measurement is about 1 minute we can compare them with variations in ïĂă1-minute resolution tau ïĂăvalues around the CIMEL observation. The condition with cloud gap should relate to the tau lowest optical thickness in the moment of CIMEL observations within a few minutes around. Note, that we used the tau data series only as an indicator of high frequency solar irradiance signal variations and we do not consider their absolute value. This characteristic was used as the indicator for the analysis of possible AERONET CIMEL sun photometer measurements in cloud gaps. The 1-minute resolution data provide us the time series to check whether CIMEL 15-minute resolution measurements can be observed in these cloud gap conditions. Since the duration of sunphotometer measurement is about 1 minute we can compare them with 1-minute resolution tau ïĂăvariations around the CIMEL observation. The condition with cloud gap should relate to the tau lowest optical thickness in the moment of CIMEL observations within a few minutes around. Note, that we used the tau data series only as an indicator of high frequency solar irradiance signal variations and we do not consider their absolute values. For illustrating this phenomenon, Fig.2 presents the diurnal variations of 1- minute resolution tauïĂň AERONET level 2.0 AOT500 and Angstrom exponent data series in conditions with cloud contamination during the two days – February 27th, 2005 and February 1st, 2006 with different cloud conditions. Weather conditions on February 27th, 2005 were characterized by the presence of Cirrus, Cirrocumulus and Altocumulus clouds. Solar disk was covered by thin clouds (SD=1), the (NA) cloud amount was equal to 10. We can see that on February 27th, 2005 AOT500 observations do not correspond to the smallest values of tau around ïĂňïĂăand, hence, there were no cloud gaps conditions during AOT500 measurements (Fig.2a). The similar results were obtained on February 1st, 2006, when during the entire day Cirrus clouds with NA=10 and NA=6 were observed (Fig.2b). The morning conditions were characterized by thin overcast cloudiness with NA=10, SD=1 and low AOT500. During the day the cloud amount decreased (NA=6) but we see the gradual increase in cloud optical thickness obscuring the Sun, which is in agreement with τ data series, and with the decrease in Angstrom exponent. Note, that even SD=0 conditions were observed after noon time. The tau time series with 1-minute resolution around AERONET AOT500 measurements do not demonstrate any local decrease, but just more uniform distribution. Of course, there can be cloud gap conditions during AERONET AOT measurements, however, on average, the cloud optical thickness contamination may induce much larger effect on aerosol climatology than removing of few “good” cases. By this example we also illustrate the necessity of the additional more strict cloud screening in winter with the threshold at least equal to NA=6.”

Another changes were made at line 235 (new numeration) after the sentence “In some years the monthly mean difference can even exceed 0.1 (for example, in February 2005 and October 2012).”: “A detailed analysis was made to understand the reasons of these large discrepancies with the standard AERONET AOT dataset. Fig.4 shows the comparison between the standard daily mean AOT500 and Angstrom exponent data and their values after additional cloud filtering. In February 2005 additional cloud filtering provided full elimination of measurements during 03.02.2005 and 27.02.2005, which were characterized by smaller Angstrom exponent and extremely high AOT on 27/02/2005. According to the additional checks we found that all these cases were observed in solar disk conditions with SD=1 or SD=0. The additional cloud filtering also provides a slight increase in Angstrom exponent during the other days in February 2005, that also indirectly confirms the elimination of cloud-contaminated cases. In October 2012 the application of the additional cloud filter provided removal of high AOT500 values on 04/10/2012 due to the existence of overcast cloud conditions with SD=1 and SD=0. However, since the Angstrom exponent was not small the cloudiness was rather thin on 04/10/2012. During the other days in October the difference in both aerosol characteristics obtained before and after the additional cloud filtering was negligible."

Another changes were made after the sentence, "Note, that small day number with aerosol observations in winter due to cloudy conditions results in large relative changes of the removed day number even when only 1-2 days are removed from the initial statistics." at line 267 (new numeration): " These values are in a qualitative agreement with the cirrus susceptibility percentage tests of AERONET level 2.0 AOT retrievals against the CALIPSO vertical feature masks (Huang et al.,2012), but they differ from the similar assessments against Micro-Pulse Lidar data shown in the same paper. However, the application of the Micro-Pulse Lidar data for evaluating the cirrus cloud contamination over Tropical area (Chew et al., 2011) has revealed much higher susceptibility percentage (about 23-34%) of the AOT sample, which is in qualitative accordance with our data for winter months. However, several recent studies indicated that clouds can have a real impact on AOT. These mechanisms of aerosol/cloud interaction include aerosol hygroscopic growth, increasing aerosol concentration due to air convergence, and new particles formation in the presence of clouds (Su et al., 2008, Jeong , Li, 2010, Eck et.al. 2012, Eck et.al. 2014). In addition to well known hygroscopic growth of particles there is a mechanism of the gas-to-particle conversion, which occurs more

intensively in the aqueous phase in cloud droplets due to the oxidation of gases (SO2, NOx, SOA) (Eck et al., 2014). Due to this mechanism in the presence of convective cloudiness the formation of new aerosol particles may observe providing higher aerosol loading during the periods with higher cloud amount in the vicinity of clouds (Eck et.al. (2014). The same mechanism can also provide lower Angstrom exponent values. Another mechanism of simultaneous variations in both aerosol and cloud amount is the changes in meteorology conditions, when, depending on advection and circulation features one can obtain synchronous changes in AOT and cloud amount, which do not interact with each other. A good example of this effect is the noticeable dependence of AOT and Angstrom exponent on various cloud filters for January. As discussed above, according to the results of 24-hour HYSPLIT backward trajectory analysis the collocated changes in AOT and cloud amount (value of cloud filter, see Fig.1) are likely observed due to the changes in advection. However, during warm period according to our long-term dataset we did not obtain the AOT-cloud amount dependence, except 100% contaminated cases when the NA<9 threshold filter has been applied. We should also mention that after applying this filter, which removes the data when solar disk was blocked by cloud, we do not remove any cases with particular convective cloudiness development, except those, which have been removed by the standard AERONET cloud-screening algorithm. However, it will be interesting to compare the results with the coming AERONET version 3 dataset, where a modification of standard cloud-screening algorithm will be applied to the data according to the method described in (Eck et al., 2014)."

Also corresponding changes were made in Discussion section and Conclusions (please, look at the full version of the text in the supplementary attachment).

1.3. The authors make a repeated point of suggesting that the elimination of a local AOT maximum in February in the AERONET monthly mean AOT climatology is evidence that their cloud screening is effective and valid. However,careful examination of the AOT data for the month of February for all years shows very high AOT values in

2005 and 2006 that are associated with large Angstrom exponents (AE 440-870 1.4 in 2006), therefore suggesting that the authors additional cloud screening may have removed actual aerosol observations of fine mode aerosol in February.

1.3. Response. Thank you for the comment. We decided to pay more attention to the analysis of February 2005 and 2006 data in the text and have shown that the eliminating of these data was right. Please, look at the changes in the text and look also at the additional Figures:

1.3. Changes in the text after the sentence "In some years the monthly mean difference can even exceed 0.1 (for example, in February 2005 and October 2012), at line 236 (new numeration)": "A detailed analysis was made to understand the reasons of these large discrepancies with the standard AERONET AOT dataset. Fig.4 shows the comparison between the standard daily mean AOT500 and Angstrom exponent data and their values after additional cloud filtering. In February 2005 additional cloud filtering provided full elimination of measurements during 03.02.2005 and 27.02.2005, which were characterized by smaller Angstrom exponent and extremely high AOT on 27/02/2005. According to the additional checks we found that all these cases were observed in solar disk conditions with SD=1 or SD=0. The additional cloud filtering also provides a slight increase in Angstrom exponent during the other days in February 2005, that also indirectly confirms the elimination of cloud-contaminated cases. In October 2012 the application of the additional cloud filter provided removal of high AOT500 values on 04/10/2012 due to the existence of overcast cloud conditions with SD=1 and SD=0. However, since the Angstrom exponent was not small the cloudiness was rather thin on 04/10/2012. During the other days in October the difference in both aerosol characteristics obtained before and after the additional cloud filtering was negligible.

1.4. Another aspect of the evidence that the authors present for the effectiveness of their cloud screening is the increase by 0.03 to 0.09 in monthly average Angstrom Exponents (AE) after the additional screening. Although this is undoubted true in

many cases (since cloud contamination decreases AE), it is unknown how much of the lower AE in the additionally screened data may be due to cloud processing of particles and hygroscopic growth of particles in the presence of clouds. These well-known physical mechanisms are not mentioned at all in the manuscript, and some discussion of this should be added in the revision. Also missing in the current manuscript is some discussion of the fact that gas-to-particle conversion occurs much more rapidly in the aqueous phase in cloud droplets therefore some of the higher AOT observed during the cloudier time periods may be due to creation of new particles from oxidation of gases such as SO2 to sulfate in the cloud droplets (also formation of nitrates and SOA), in addition to hygroscopic growth (see Jeong et al. (2010; JGR), http://onlinelibrary.wiley.com/doi/10.1029/2009JD013547/full Su et al. (2008), Eck et al. (2014) for example). These physical mechanisms also need to be discussed in the current paper.

1.4. Response. We are sorry for missing these interesting papers and have added the discussion on the possibility of aerosol particle increase in the vicinity of clouds We combined both mechanisms in the discussion.

1.4. The changes in the text: We added the discussion in the 3.11 section at line 274 (new numeration): However, several recent studies indicated that clouds can have a real impact on AOT. These mechanisms of aerosol/cloud interaction include aerosol hygroscopic growth, increasing aerosol concentration due to air convergence, and new particles formation in the presence of clouds (Su et al., 2008, Jeong , Li, 2010, Eck et.al. 2012, Eck et.al. 2014). In addition to well known hygroscopic growth of particles there is a mechanism of the gas-to-particle conversion, which occurs more intensively in the aqueous phase in cloud droplets due to the oxidation of gases (SO2, NOx, SOA) (Eck et al., 2014). Due to this mechanism in the presence of convective cloudiness the formation of new aerosol particles may observe providing higher aerosol loading during the periods with higher cloud amount in the vicinity of clouds (Eck et.al. (2014). The same mechanism also provides lower Angstrom exponent values.

In the Section 4 some discussion was also added at line 616 (new numeration):

There can be some another drivers of increasing the AOT due to clouds as was obtained in several recent publications both from ground-based and satellite dataset due to aerosol humidification growth, cloud processing, or new particle formation in clouds (Quass et al., 2010, Eck et al., 2014). As a result, we may consider two competing phenomena – the effects of cloud contamination on aerosol retrievals and the possible changes in aerosol properties due to various processes in the vicinity of clouds. We should note that since it is not possible to distinct these two processes without special field experiments as it was made in (Eck et al., 2014), our revised aerosol climatology relates to the classical way of aerosol properties evaluation, when these aerosol/cloud interaction processes are not accounted for. However, the existence of the additional cloud contaminated cases with solar disk blocking conditions almost in all cases, except 0.5% in winter, biased the aerosol climatology. Their removal provides the better quality dataset.

1.5. Finally regarding the cloud screening issue, the authors mention that the AERONET cloud screening is effective for cumulus type clouds but not always for cirrus clouds. I agree with this assessment and this is well known, having been documented in other studies of cloud contamination of AERONET data. However, although the authors mention cirrus contamination they do not apply any checks specific to cirrus clouds, only to total cloud fraction.

1.5. Response. We combined all the clouds in the term “upper cloud” which we used throughout the new version of the manuscript. I guess, that the particular type of cloudiness is not very important, since this is a qualitative characteristics. The most important factors is the optical thickness (ability to measure the direct irradiance by sun photometer), and the homogeneity of the optical layer. We have also included the discussion on this point in the paper.

1.5. The changes in the text concerning this point were made from line 149 (new

numeration) after the sentence “We showed there that the existing standard cloud-screening algorithm works perfectly, when aerosol measurements are contaminated by optically thin low layer cloudiness, which is characterized by large triplet variations”

“These variations are used as a parameter in the standard cloud-screening algorithm developed by Smirnov et al. (2000). However, if the cloud blocking the Sun is thin and uniform, the triplet variation can be small and the contaminated AOT measurements pass through the filter. Mainly the cirrus clouds are characterized by these properties. However, in general, according to the International Cloud Atlas (1987) other types of clouds may be also characterized by these properties as well. They include different forms of Cirrostratus, and even Altostratus translucidus clouds, which relate to the middle cloud level. In this publication all types of the cloudiness, which can induce the potential contamination of AOT will be combined under the term “upper cloudiness”. Since low cloudiness is effectively filtered out by the standard cloud-screening algorithm we proposed to apply simple total cloud amount (NA) filter, which is sensitive to the existence of upper cloudiness. In this context NA value (together with the application of standard cloud-screening procedure) provides the information about the potential existence of high and middle layer cloudiness, since the standard AERONET cloud-screening algorithm successfully removes the cases contaminated by low level clouds or the cases with strong signal variations.”

1.6. It is very puzzling that the authors did not reference the papers of Chew et al. (2011; in Atmospheric Environment) and Huang et al. (2012; in JGR), since these papers specifically investigated the cirrus contamination of AERONET AOT data by using lidars that can identify high altitude cirrus clouds. The authors should examine these two papers and include discussion of them in the revised manuscript.

1.6. Response. We are also very grateful for the references to these papers. We have added the discussion in the text and sorry for not including them in the previous version.

1.6. The changes in the section 3.1.1 from line 124 after the sentence “Will it significantly affect the aerosol climatology?”: For evaluating the upper layer cloud contamination in the AERONET dataset different approaches are used. In the recent studies the ground-based MPLNET, as well as satellite CALIPSO and MODIS datasets were used for evaluating the cirrus AOT contamination (Chew et al., 2011, Huang et al., 2012). According to MPLNET data the AOT bias due to unscreened cirrus cloud presence is about 0.03-0.06 with the occurrence of 23-34% depending on the method of the estimation over the tropical region in Singapore (Chew et al., 2011). Huang et al. (2012) evaluated the susceptibility percentage of AERONET level 2.0 AOT retrievals to cirrus contamination using different types of measurements. According to MPLNET cirrus flags this value varied from zero to ïĄ¿4%, according to the collocated Calipso cirrus flags - from 1% to 33%, and according to the MODIS cirrus flags - from 0.4% to 18% changing significantly over the globe due to the different occurrence of cirrus clouds. However, satellites have relatively low overpass frequency over AERONET sites - one-two time a day – for MODIS, and 16-day repeating cycle - for CALIPSO (Huang et al., 2012). The MPLNET application for cirrus flags has the problem with viewing geometry difference between the sunphotometer and the MPL. In addition, the MPL signals are extremely weak at the altitudes H> 10 km, where cirrus clouds may be observed. It was also mentioned in (Huang et al., 2012) that the MPL noise level dramatically increases during daytime especially at noon, when there are most favourable conditions for AERONET-MPLNET matchup from the point of view of the closeness of viewing geometries.

We also included some analysis in the Discussion section in the following paragraph at line 631 (new numeration): The application of an additional cloud filter results in significantly decrease in day number up to 7-20% during warm period, and 25-45% during the cold period because of higher occurrence of overcast upper layer cloudiness and the application of more strict filter NA<6. We should also note that the susceptibility percentage of contaminated cases is in the qualitative agreement with the data shown in Chew et al., (2011) as well as with the results obtained in (Huang et al. 2012) for the collocated AERONET/CALIPSO and AERONET/MODIS measurements.

1.7. Abstract, lines 6-8: Please mention here that these are monthly averages of AOT. 1.7. Response Done. 1.7. Changes in the text: The application of cloud correction according to hourly visual cloud observations provides a decrease in monthly averages of aerosol optical thickness (AOT) at 500 nm of up to 0.03 compared with the standard dataset.

1.8. Abstract, lines 20-22: The way this sentence is written it suggests that Carbon Monoxide (CO) is an aerosol precursor gas, however it is not. Please rewrite this sentence to clarify.

1.8. Response: We agree that the mentioned CO trends are confusing in the sentences which contain the information about the precursors and decided to remove this part from the abstract. Please, note that the numbers of trend values have been slightly changes due to the updated EMEP dataset. 1.8. Changes in the text: “The pronounced negative AOT trends of about -1–5%yr-1 have been obtained for most months, which could be attributed to the negative trends in emissions (E) of different aerosol precursors of about 135 Ggyr-2 in ESOx, 54 Ggyr-2 in ENMVOC, and slight negative changes in NOx over European territory of Russia.”

1.9. Page 7845, line 14: There are about 400 AERONET sites, not 200 as stated here.

1.9. Response: Thank you. Sorry. Changed:

1.9. Changes in the text: “Aerosol Robotic Network (AERONET) (http://aeronet.gsfc.nasa.gov/) has been in operation since the middle of 1990s (Holben et al., 1998) with currently more than 400 sites continuously working all over the world.”

1.10. Page 7846, line 1: “. . .where the aerosol network is rare” should be modified to “. . .where the aerosol network is sparse”

1.10. Response: Done. 1.10. Changes in the text: “These records provide a reliable dataset for studying long-term variability of aerosol properties in the Eastern Europe, where the aerosol network is sparse."

1.11. Page 7846, lines 16-17: ". . .full field CIMEL. . ." should be ". . .full field of view CIMEL. . ." 1.11. Response: Thank you. Done. 1.11 Changes in the text: "The procedure of aerosol measurements by CIMEL AERONET sun/sky photometer and the inversion algorithms were described in numerous publications (http://aeronet.gsfc.nasa.gov/new_web/publications.html). MSU MO site utilizes the 1.2ïĆř full field of view CIMEL CE318 sun and sky photometer."

1.12. Page 7846, line 23: 670 nm should be 675 nm 1.12 Response: Sorry for the misprint. Done. 1.12. Changes in the text: "Both sun and sky-radiance in the channels 440, 675, 870 and 1020nm are utilized in the inversion algorithm developed by Dubovik and King [2000], which provides several important aerosol products (volume size distribution, refractive index, single scattering albedo, phase function, etc.)."

1.13. Page 7847, line 23: Please mention here the altitude of the NO2 measurements made with the APNA-360 and the data sampling time interval at the Moscow MSU MO site.NO2 has significant diurnal variability since it is a short-lived gas so it would also be useful to know the data sampling interval. 1.13. Response: Yes, this is important. We added the information in the text: 1.13 Changes in the text: "For quantifying the NO2 content, we used in-situ long-term 1 minute resolution measurements of NO2 concentrations by APNA-360, Horiba Inc. (Elanski et al., 2007) at the Moscow MSU MO at the altitude of about 3.5 meters from ground since 2002."

1.14. Page 7848, lines 8-15: Please be very clear that the observer indicates total cloud cover in increments of tenths and that when you say N>9 you really mean that only when N=10 (which is overcast total cloud cover) are the AOT observations screened as cloud contaminated. Also clarify that for November-February when you change the total cloud cover threshold to N>6 you are saying that for N=7, 8, 9, and 10 visual cloud fractions that you then eliminate all AOT measurements for the entire hour

interval.

1.14. Response: Thank you. We added the clarifications in this section for the threshold NA<9 which include the elimination of cases at NA=9 and NA=10. All other thresholds are determined in similar way.

1.14. Changes in the text: As an example, in Fig.1 we demonstrate the effects of utilizing the different additional NA filters for AOT at 500nm (AOT500) and Angstrom exponent (ïĄą) datasets for the central months of the seasons. One can see a similar tendency of the AOT decrease in all seasons after removing the cases with the threshold of NA<9, which includes the conditions with almost total cloud amount of NA= 9-10.

In the previous paragraph (from line 145) we now mentioned that : "The application of cloud filter means the elimination of all AOT measurements for the entire hour interval."

1.15. Page 7848, lines 25-27: Please add 'monthly mean' so that this sentence reads 'In some years the monthly mean difference can even exceed 0.1. . ." Yes, we added some additional analysis to these cases and included "monthly mean" in the text. The text now is as follows: 1.15. Response: Done. 1.15. Changes in the text: "In some years the monthly mean difference can even exceed 0.1 (for example, in February 2005 and October 2012)"

1.16. Also you should give some details about the two months that you mention here, October 2012 and February 2005. It should be noted that the data you eliminated for October 2012 had relatively high Angstrom Exponent, therefore suggesting that you may have removed good AOT observations dominated by fine mode particles, and also that there were only 3 days of AERONET Level 2 AOT data for Oct 2012, even before you eliminated any data.

1.16. Response: Thanks for the useful comment. We have added the additional Figure and the analysis of the cases in the text. It is clearly seen that we have now only

3 days for Octobre 2012. Please, see the details in the text below. 1.16. Changes in the text: At line 236 9 new numeration): "A detailed analysis was made to understand the reasons of these large discrepancies with the standard AERONET AOT dataset. Fig.4 shows the comparison between the standard daily mean AOT500 and Angstrom exponent data and their values after additional cloud filtering. In February 2005 additional cloud filtering provided full elimination of measurements during 03.02.2005 and 27.02.2005, which were characterized by smaller Angstrom exponent and extremely high AOT on 27/02/2005. According to the additional checks we found that all these cases were observed in solar disk conditions with SD=1 or SD=0. The additional cloud filtering also provides a slight increase in Angstrom exponent during the other days in February 2005, that also indirectly confirms the elimination of cloud-contaminated cases. In October 2012 the application of the additional cloud filter provided removal of high AOT500 values on 04/10/2012 due to the existence of overcast cloud conditions with SD=1 and SD=0. However, since the Angstrom exponent was not small the cloudiness was rather thin on 04/10/2012. During the other days in October the difference in both aerosol characteristics obtained before and after the additional cloud filtering was negligible."

1.17. Page 7849, lines 7-10: You say that water vapor is overestimated in the high cloud fraction observations, but I think this is likely a misinterpretation. It is expected that for high cloud fraction observations that total column water vapor would be higher than average. This is well known since clouds are liquid water droplets or ice crystals that have a relatively short lifetimes since they are continually evaporating or forming in a high relative humidity environment. Clouds also often form in converging air that contains higher moisture content.

1.17. Response: Thank you for providing the reasons of possible changes in water vapour content. I agree that it can be different reasons for this phenomenon. We have included the additional discussion on this point. 1.17. Changes in the text at line 259 (new numeration): "However, there can be another reason for this phenomenon:

the air convergence may create favorable conditions with higher relative humidity and water vapor content in the atmosphere, and, hence, the existence of clouds (see, for example, Jeong, Li, 2010). However, these processes should be studied further."

1.18.Page 7850, line 8: Please give the distance in kilometers between the Moscow and Zvenigorod sites in this sentence.

1.18. Response: Done. 1.18. Changes in the text at line 314 (new numeration): "Our aerosol comparisons in urban and background conditions (Chubarova et al., 2011) also demonstrated the existence of the residual NO2 contamination over Moscow, which can be seen in a specific character of AOT spectral difference between the parallel measurements in Moscow and in Zvenigorod background conditions at the distance of 55 km (see Fig.3 and the discussion in Chubarova et al., 2011)."

1.19. Page 7851, lines 6-7: Please add some information in this sentence on why the NO2 lifetime is longer in winter (i.e. photochemistry, etc.).

1.19 Response: We have added some clarifications. 1.19. Changes in the text a tline 349 (new numeration): "One can see that the maximum NO2 content is observed in February and elevated NO2 values are recorded in December-March period due to higher emissions from power stations during the heating season and larger NO2 life time in winter conditions due to decreasing of the photodissociation rates at higher zenith angles (Brasseur and Solomon, 1986)."

1.20. Page 7852, lines 10-11: Please elaborate what second derivative you are referring to in this sentence, the 2nd derivative of ln AOT versus ln WL or something else, and also provide a reference to clarify.

1.20 Response: Thanks a lot. It was really unreadable. Done. 1.20. Changes in the text at line 382(new numeration): "Both procedures lead to decreasing in the second derivative of logarithm of AOT versus logarithm of wavelength (Eck et al., 1999) and may affect the inverse RT solution in the AERONET algorithm (Dubovik, King, 2000), especially in case, when OTNO2 values are close to aerosol optical thickness."

1.21. Page 7853, lines 25-28: In Figure 4 you have plotted the AOT versus wavelength in linear scales but the standard and most common analyses typically plot these in logarithmic coordinates (both x- and y-axes). You say that the revised AOT spectra are smoother (in linear coordinates), but are they also smoother when plotted in logarithmic space? 1.21. Response: Yes, even in logarithmic space the dependence is getting smoother for most months, except June, July, and August, when the effect is negligible due to smaller OT(NO2)/AOT ratio. The determination coefficient for linear regressions is slightly higher (see the Table below for Rˆ2 in log-log space), except mentioned months. This is not the important difference, of course. Much more important is that the absolute value of Angstrom exponent changes substantially after the additional NO2 correction. We added the discussion in the text. Table month determination coefficient Rˆ2 with NO2 correction determination coefficient Rˆ2 without additional NO2 correction absolute difference 1 0.992 0.982 0.010 2 0.995 0.990 0.005 3 0.997 0.994 0.003 4 0.998 0.997 0.002 5 0.997 0.995 0.002 6 0.998 0.996 0.002 7 0.999 1.000 0.000 8 0.998 0.999 -0.001 9 0.999 1.000 0.000 10 0.999 0.997 0.002 11 0.998 0.994 0.003 12 0.995 0.991 0.005

Note that this Table is not included in the text. It is given just for clarifying our response.

1.21. The changes in the text at line 427 (new numeration): "The revised spectral dependencies for most months especially in cold period, when NO2 to AOT ratio is high, are characterized by more smooth spectral character due to the influence of spectral NO2 correction. This correction also induces slightly higher determination coefficient when obtaining Angstrom exponent within 440-870nm in logarithmic space coordinates."

1.22. Page 7855, line 21 and Table 2: You mention December AOT is this sentence, but is the data sampling sufficient in that month to make any definitive conclusions? In Table 2 you show only 4 days of data for the month of December (total for all years!),

while all other months have much more data, ranging from 33 days to 290 days. If the month of December truly has only 4 days of AOT data (revised data set) then I would suggest that you remove any discussion of differences in the December monthly means since a 4-day data sample is not statistically robust enough to make any useful conclusions.

1.22. Response: We agree, of course, that 4-day sample is too small to make the conclusions. We made additional clarifications in this text pointing out on this small sample. However, in this very place, I suppose, it is not necessary to remove December from the analysis, since we showed there the effects of NO2 and cloudiness filters, which improved the final dataset.

1.22. Changes in the text after the sentence at line 451 (new numeration): "It should be noted, that we have very small AOT statistics in December due to high cyclonic activity with cloudy weather. Moreover, the application of the restriction on air mass m>5, which can be observed in December even at noon conditions in Moscow, provides further elimination of the level 2.0 data. So the obtained climatological values should be taken with caution. However, after the additional corrections even this small dataset demonstrates reasonable AOT values, which are in agreement with the statistics obtained for January (next winter month) conditions (see Table 2).

1.23. Page 7857, line 20: Similar to the abstract, the way this paragraph is written it suggests that carbon monoxide (CO) is an aerosol precursor gas, however it is not. Please revise to clarify. 1.23. Response: Yes, thank you. Sorry for poorly written text. 1.23. The changes in the text at 529 (new numeration): "In addition, the CO emissions, which do not influence directly on the secondary aerosol generation but may characterize the intensity of pollution from the transportation sources, has also a pronounced negative trend of about 69 Gg yr-2. This negative trend also confirms the complex character of the atmosphere cleanup. There is also a tendency of NOx decrease over European part of Russia, especially during the last years but it is not statistically significant. Some negative tendency is observed in emissions of the particulate matter with the diameter less 2.5 ïĄ■m (PM2.5).” 1.24. Page 7858, lines 7-10: Similar to above, in page 7857, you suggest in this sentence that CO plays a role in aerosol generation. Please explain or clarify here. 1.24. Response: Yes, thank you. We changes the text and we also added the effects of NOx in the text according to the updated EMEP dataset. 1.24. The changes in the text: “As a result, we assume that negative trend in AOT is observed likely due to the decrease in anthropogenic emissions of SOx and NMVOC over European part of Russia, which play a significant role in the second aerosol generation, especially during warm period. Some important role can also play the decrease in NO2 emission during the last years since 2010.”

1.25. Page 7859, lines 4-7: You suggest here that the standard AERONET data set overestimates AOT at 500 nm by up to 0.03 due to cirrus contamination at the Moscow_MSU_MO site. However your additional data screening is done only based on total cloud fraction, so you cannot specify that cirrus clouds were the only reason. Of course your statement also does not consider that AOT may actually be higher in some high cloud fraction observations due to aerosol humidification growth, cloud processing, or new particle formation in clouds. Large-scale convergence that results in cloud formation may also in some cases result in higher aerosol concentrations and higher AOT in the presence of clouds (see Quass et al. (2010; ACP).

1.25. Response: We agree that the application of total cloud amount provide the filtering not only the cirrus clouds. We have added the discussion on this account in the text (please, look at our response to your previous comment above) . Yes, we do not consider the increase in AOT due to mentioned physical and chemical processes. That is why we also added some discussion on this point in the text.

1.25. Changes in the text: The changes were made in the Section 4 at line 616 (new numeration): “There can be some another drivers of increasing the AOT due to clouds as was obtained in several recent publications both from ground-based and satellite dataset due to aerosol humidification growth, cloud processing, or new particle formation in clouds (Quass et al., 2010, Eck et al., 2014). As a result, we may consider two competing phenomena – the effects of cloud contamination on aerosol retrievals and the possible changes in aerosol properties in the vicinity of clouds. We should note that since it is not possible to distinct these two processes without special field experiments as it was made in (Eck et al., 2014), our revised aerosol climatology relates to the classical way of aerosol properties evaluation, when these aerosol/cloud interaction processes are not accounted for. However, the existence of the additional cloud contaminated cases with solar disk blocking conditions almost in all cases, except 0.5% in winter, biased the aerosol climatology. Their removal provides the better quality dataset. ”

1.26. Page 7859, lines 7-10: Please also add that your much more strict filter of human observed cloud fraction >60% in winter also guarantees that more cloud filtering is done in winter and therefore highly likely to have larger differences with the standard AERONET values.

1.26. Response: Yes, I agree. We have added the suggested changes. 1.26. Changes in the text in the section 4 “Discussion” at line 626 : “The relative AOT500 difference between the standard dataset and the dataset with the additional cloud-screening has minimum in summer (5%) and maximum up to 20-30% for winter months when the occurrence of upper cloudiness is high, and AOT values are low. The larger AOT difference in winter can be also attributed to more cloud filtering (with strict filter of human observed cloud fraction ïĆş60%) and therefore highly likely larger differences with the standard AERONET values” 1.27. Page 7859, lines 11-18: For the record, the AERONET project would never apply such a general and non-rigorous cloud screening as suggested in this sentence, based on human visual observations of total cloud cover. Additionally, cirrus clouds (and other types of clouds as well) are often discontinuous in space therefore a station as far as 60 km from an AERONET sun-sky radiometer may often have a significantly different cloud fraction. Additionally in this paragraph you seem to ignore the fact that your cloud-screening test is based on only total cloud fraction, not cirrus amount.

1.27 . Response: We have changed this part of the text and removed some of the mentioned points. The term “Cirrus clouds” was changed on “upper cloudiness” throughout the text. 1.27. The changes in the text at line 637(new numeration): Since the effects of the proposed additional cloud-screening are distinct, its application may help in obtaining the better quality aerosol datasets by different users, especially for the old, historical records. However, we admit that this is not a rigorous assessment. But this additional correction could be very useful in different applications. It is also possible to verify the current aerosol datasets using the cloud data from the automatic total sky imagers, which have been already in operation at several sites (O’Neill et al., 2003, Jeong, Li, 2010) or the application of the collocated lidar measurements (Chew et al, 2011, Huang et al., 2012).

1.28 Page 7859, lines 17-18: The citation in this sentence given as (Radiation in cloudy atmosphere, 1984) should be (Feigelson, 1984).

1.28. Response. Dr. Eva Feigelson was the Editor of this book, which was written by different authors. (see http://www.springer.com/br/book/9789027718037). So it would have been necessary to leave it as is, however, we had removed the part of the text with this reference after its editing.

1.29. Page 7860, lines 13-15: Please be careful about generalizing here, since the 675 nm channel is in the visible and does not have any NO2 absorption, so the only changes in that wavelength are due to your revised cloud screening, therefore similar to the near infrared spectral range.

1.29. Response: Thank you. To clarify this point we have added the additional sentence with necessary clarifications. 1.29. Changes in the text at line 664 (new numeration): “The total difference in annual mean AOT values due to the additional account for cloud and NO2 corrections is about 0.04 in UV, 0.02 in visible, and 0.01 in near-infrared spectral range. Note, that the NO2 correction mainly concerns the 340, 380, 440, and 500nm AOT channels and the retrievals of Angstrom exponent.”

1.30. Figure 8 caption: Again you list carbon monoxide as one of the “main aerosol precursors”, therefore this caption needs to be corrected or CO (Carbon Monoxide) removed from the graph.

1.30. Response: We made the correction to the Figure caption. Now it is Figure 11. 1.30. Changes in the Caption: “Fig.11. Interannual variations in emissions of main aerosol precursors (SOx, NOx, NMVOC), CO, and particulate matter (PM2.5) according to WebDab - EMEP database over European part of Russia (a), relative changes in 50% quantile AOT500 and in SOx and NOx emissions over European part of Russia and directly over Moscow (b).”

The answers to the questions of the second anonymous referee:

2.1. Since Atmos. Meas. Tech. Journal is dedicated to the publication and discussion of advances in measurement techniques, all technical and methodological aspects should be described with minimal detail. In this framework there is a clear imbalance in the treatment given to the two issues of technical and methodological character which are discussed in the first part of the paper. The main concern is the methodology used for a second cloud screening since the largest correction of AOT climatology for Moscow clearly comes by removing AERONET data coincident with the presence of cirrus clouds (see Figure 5). I interpreted the screening cloud using cloud cover, it only applies to the AERONET observations in which there are only cirrus clouds, otherwise the approach used would be wrong because in this case valid AERONET observations would be removed of arbitrary manner which would affect the aerosol climatology of unknown way.

2.1. Response: We took into account for the remarks of the reviewer and included more discussion on cloud filtering in the text. We have included also several additional Figures as the illustration for the applicability of the proposed method. We have also added the discussion on the term “cirrus” clouds. As we already mentioned, from our point of view a particular type of cloudiness is not very important since this is a qualitative characteristics. To pass the standard cloud-screening threshold the cloud should be optically thin to direct irradiance transmission, and uniform enough during the triplet series of measurement. Not only Cirrus clouds relate to this definition but Cirrostratus, and even Altostratus translucidus clouds. We combined all the clouds which might contaminate the AOT observation in one term “upper cloud”, which we used in the new version of the manuscript.

2.1. The changes in the text concerning this point were made after the sentence from line 148 (new numeration):

“These variations are used as a parameter in the standard cloud-screening algorithm developed by Smirnov et al. (2000). However, if the cloud blocking the Sun is thin and uniform, the triplet variation can be small and the contaminated AOT measurements pass through the filter. Mainly the cirrus clouds are characterized by these properties. However, in general, according to the International Cloud Atlas (1987) other types of clouds may be also characterized by these properties as well. They include different forms of Cirrostratus, and even Altostratus translucidus clouds, which relate to the middle cloud level. In this publication all types of the cloudiness, which can induce the potential contamination of AOT will be combined under the term “upper cloudiness”. Since low cloudiness is effectively filtered out by the standard cloud-screening algorithm we proposed to apply simple total cloud amount (NA) filter, which is sensitive to the existence of upper cloudiness. In this context NA value (together with the application of standard cloud-screening procedure) provides the information about the potential existence of high and middle layer cloudiness, since the standard AERONET cloud-screening algorithm successfully removes the cases contaminated by low level clouds or the cases with strong signal variations. However, the application of different NA thresholds may provide the different samples and as a result, different statistics. As an example, in Fig.1 we demonstrate the effects of utilizing the different additional NA filters for AOT at 500nm (AOT500) and Angstrom exponent (ïĄą) datasets for the central months of the seasons. One can see a similar tendency of the AOT decrease

in all seasons after removing the cases with the threshold of NA<9, which includes the conditions with almost total cloud amount of NA= 9-10. Additional testing on solar disk conditions has revealed that all eliminated cases in this sample belong to the situations, when solar disk was covered by clouds (SD=1 or SD=0). In April after eliminating the almost overcast cloud conditions with NA<9 threshold there is no further changes in AOT500 samples with more strict NA cloud threshold. At the same time the sample is dramatically (more than twice) reduced (from 229 to 113 in NA<3 sample). In July we also see a slight decrease in AOT500 in the NA<9 sample and, in addition, a significant growth of Angstrom exponent. Note, that in July and October we see even a slight increase in the AOT500 for almost clear sky conditions (the sample with NA<3). In January there is a pronounced decrease in AOT500 with reducing cloud amount. The lowest AOT500 values and largest Angstrom exponent are observed in the NA<3 sample. The application of the 24-hour Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) NOAA model backward trajectory analysis (Draxler and Hess, 1998) for all the cases in January has revealed for the NA<3 sample the prevalence of the northern (North-West, North, North-East) advection (80% of cases) characterized by low AOT (Chubarova, 2009), compared with 40% of cases for partially cloudy conditions (3<NA<9). At the confidence level of P=80% several of these dependencies are statistically significant (see the error bars in Fig.1). .”

2.2. It is well known that the AERONET algorithm may fail with homogenous and persistent cirrus blocking the sun. In other cases the cloud screening is quite effective. Unfortunately the description of the cloud screening methodology is quite short and does not provide the necessary details to understand how it has been designed and performed (see specific comments).

2.2. Response: Thank you for the comment. We fully agree with the opinion of the reviewer and give much more details about this additional cloud filtering in the new version of the paper. The most changes concern the Section 3.1.1 but there are some respective changes in the section “Discussion ‘ and in “Conclusions”. We have attached the updated version of the manuscript as supplementary material for better understanding the changes which have been made in addition to the text shown below. Some clarification on what is solar disk conditions is given in the section “Data description” . 2.2. Changes in the text: The changes were made in “data description section :from line 94 (new numeration): “In addition, the dataset of hourly solar disk condition observations, which are performed simultaneously with cloud observations, was used in the analysis. This is a standard type of observations at the actinometrical stations in Russia. Using this characteristic we can distinct the conditions, when solar disk (SD) is free from clouds, or when SD is obscured by thin clouds (shadows can be seen at ground), or when SD can be seen but there are no shadows at ground, or when SD can not be seen due to relatively high cloud optical thickness. These SD conditions are noted with “2”, “1”, “0” and “P” marks, respectively.”

Below is the changed Section 3.1.1 without Figures which can be found in the attachments and in the supplementary materials in the body of the text.

"3.1.1.Additional cloud-screening procedure and its effect on aerosol climatology Since the aerosol measurements are carried out in automatic regime, a special cloud-screening procedure was developed for an automatic removal of cloud contaminating aerosol measurements (Smirnov et al., 2000). In the standard AERONET algorithm the data, which successfully pass the cloud screening procedure, are assigned to the level 1.5. After the second calibration and some additional visual checks the data are assigned to the final level 2.0. However, sometimes even the final dataset could “suffer” from the effects of thin homogeneous upper cloudiness contamination (O’Neill et al., 2003). As was mentioned in this paper “the strategy of the AERONET cloud screening was liberal; to interfere as little as possible with coarse mode events such as dust incursions and thus to accept the inevitability of some thin homogeneous cloud data, being admitted into the database.” Hence, the question remains, how important can be this effect. Will it significantly affect the aerosol climatology? For evaluating the upper layer cloud contamination in the AERONET dataset different approaches are

used. In the recent studies the ground-based MPLNET, as well as satellite CALIPSO and MODIS datasets were used for evaluating the cirrus AOT contamination (Chew et al., 2011, Huang et al., 2012). According to MPLNET data the AOT bias due to unscreened cirrus cloud presence is about 0.03-0.06 with the occurrence of 23-34% depending on the method of the estimation over the tropical region in Singapore (Chew et al., 2011). Huang et al. (2012) evaluated the susceptibility percentage of AERONET level 2.0 AOT retrievals to cirrus contamination using different types of measurements. According to MPLNET cirrus flags this value varied from zero to ïĄ¿4%, according to the collocated Calipso cirrus flags - from 1% to 33%, and according to the MODIS cirrus flags - from 0.4% to 18% changing significantly over the globe due to the different occurrence of cirrus clouds. However, satellites have relatively low overpass frequency over AERONET sites - one-two time a day – for MODIS, and 16-day repeating cycle - for CALIPSO (Huang et al., 2012). The MPLNET application for cirrus flags has the problem with viewing geometry difference between the sunphotometer and the MPL. In addition, the MPL signals are extremely weak at the altitudes H> 10 km, where cirrus clouds may be observed. It was also mentioned in (Huang et al., 2012) that the MPL noise level dramatically increases during daytime especially at noon, when there are most favourable conditions for AERONET-MPLNET matchup from the point of view of the closeness of viewing geometries. The influence of the cloud contamination on aerosol properties was also discussed in (Uliumdzhieva et al., 2005) for Moscow conditions. In this paper the application of the standard cloud visual observations as an additional cloud-screening filter was proposed. We used one-hour resolution cloud observations for additional filtering of quasi-simultaneous Cimel observations at the same site. The application of cloud filter means the elimination of all AOT measurements for the entire hour interval. We showed there that the existing standard cloud-screening algorithm works perfectly, when aerosol measurements are contaminated by optically thin low layer cloudiness, which is characterized by large triplet variations. These variations are used as a parameter in the standard cloud-screening algorithm developed by Smirnov et al. (2000). However, if the cloud blocking the Sun is thin and uniform, the triplet variation can be small and the contaminated AOT measurements pass through the filter. Mainly the cirrus clouds are characterized by these properties. However, in general, according to the International Cloud Atlas (1987) other types of clouds may be also characterized by these properties as well. They include different forms of Cirrostratus, and even Altostratus translucidus clouds, which relate to the middle cloud level. In this publication all types of the cloudiness, which can induce the potential contamination of AOT will be combined under the term “upper cloudiness”. Since low cloudiness is effectively filtered out by the standard cloud-screening algorithm we proposed to apply simple total cloud amount (NA) filter, which is sensitive to the existence of upper cloudiness. In this context NA value (together with the application of standard cloud-screening procedure) provides the information about the potential existence of high and middle layer cloudiness, since the standard AERONET cloud-screening algorithm successfully removes the cases contaminated by low level clouds or the cases with strong signal variations. However, the application of different NA thresholds may provide the different samples and as a result, different statistics. As an example, in Fig.1 we demonstrate the effects of utilizing the different additional NA filters for AOT at 500nm (AOT500) and Angstrom exponent (ïĄą) datasets for the central months of the seasons. One can see a similar tendency of the AOT decrease in all seasons after removing the cases with the threshold of NA<9, which includes the conditions with almost total cloud amount of NA= 9-10. Additional testing on solar disk conditions has revealed that all eliminated cases in this sample belong to the situations, when solar disk was covered by clouds (SD=1 or SD=0). In April after eliminating the almost overcast cloud conditions with NA<9 threshold there is no further changes in AOT500 samples with more strict NA cloud threshold. At the same time the sample is dramatically (more than twice) reduced (from 229 to 113 in NA<3 sample). In July we also see a slight decrease in AOT500 in the NA<9 sample and, in addition, a significant growth of Angstrom exponent. Note, that in July and October we see even a slight increase in the AOT500 for almost clear sky conditions (the sample with NA<3). In January there is a pronounced decrease in AOT500 with reducing cloud amount.

The lowest AOT500 values and largest Angstrom exponent are observed in the NA<3 sample. The application of the 24-hour Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) NOAA model backward trajectory analysis (Draxler and Hess, 1998) for all the cases in January has revealed for the NA<3 sample the prevalence of the northern (North-West, North, North-East) advection (80% of cases) characterized by low AOT (Chubarova, 2009), compared with 40% of cases for partially cloudy conditions (3<NA<9). At the confidence level of P=80% several of these dependencies are statistically significant (see the error bars in Fig.1). Balancing between the substantial decrease in case number and the accuracy of the retrievals of aerosol properties we showed that the best results were obtained when the cases with Ntotal.<9 during March-October period with almost overcast cloudiness and cloud contaminated solar disk conditions data were removed. For November –February conditions the filter threshold is more strict (Ntotal<6) since solar elevation in Moscow is low (hnoon<25ïĆř) at this time and a well-known effect of significant visual cloud amount increase towards the horizon plays a vital role. Smaller cloud threshold during winter time may induce filtering out so called “good” AOT cases. The additional analysis of solar disk conditions has revealed only 12 cases from 2521 cases (about 0.5%), which were not contaminated by clouds (SD=2) and were incorrectly removed from the sample. All of them were observed in February (18.02.2011 – 4 cases, 3.02.2003- 5 cases, 11/02/2007 - 3 cases). However, even these three days are presented in the final sample since during these days there were other sun photometer measurements at smaller NA. In addition, we analyzed the 1-minute resolution direct solar measurements with the standard Russian actinometer (WMO, 1986) for studying the possibility of AERONET direct solar irradiance observations in cloud gaps,. These data were used for estimating the standard transparency coefficient at air mass m=2 according to (Evnevich, Savikovsky, 1988): p_2=(S_h/1.367)ˆ((sinh+0.205)/1.41), (1) where S_h - is the measured value of direct shortwave irradiance, h - solar elevation.

The p2 coefficient is widely used for assessing the variation of the transparency of the atmosphere (Ohvril et al., 2009). Using this equation we evaluated the integral optical thickness as τ=-lnâĄą(p_2). This characteristic was used as the indicator for the analysis of possible AERONET CIMEL sun photometer measurements in cloud gaps. The 1-minute resolution data provide us the time series to check whether CIMEL 15-minute resolution measurements can be observed in these cloud gap conditions. Since the duration of sunphotometer measurement is about 1 minute we can compare them with 1-minute resolution tau ïĂăvariations around the CIMEL observation. The condition with cloud gap should relate to the tau lowest optical thickness in the moment of CIMEL observations within a few minutes around. Note, that we used the tau data series only as an indicator of high frequency solar irradiance signal variations and we do not consider their absolute values. For illustrating this phenomenon, Fig.2 presents the diurnal variations of 1- minute resolution ïĄťïĂň AERONET level 2.0 AOT500 and Angstrom exponent data series in conditions with cloud contamination during the two days – February 27th, 2005 and February 1st, 2006 with different cloud conditions. Weather conditions on February 27th, 2005 were characterized by the presence of Cirrus, Cirrocumulus and Altocumulus clouds. Solar disk was covered by thin clouds (SD=1), the (NA) cloud amount was equal to 10. We can see that on February 27th, 2005 AOT500 observations do not correspond to the smallest values of tau aroundïĂňïĂăand, hence, there were no cloud gaps conditions during AOT500 measurements (Fig.2a). The similar results were obtained on February 1st, 2006, when during the entire day Cirrus clouds with NA=10 and NA=6 were observed (Fig.2b). The morning conditions were characterized by thin overcast cloudiness with NA=10, SD=1 and low AOT500. During the day the cloud amount decreased (NA=6) but we see the gradual increase in cloud optical thickness obscuring the Sun, which is in agreement with τ data series, and with the decrease in Angstrom exponent. Note, that even SD=0 conditions were observed after noon time. The tau ïĂătime series with 1-minute resolution around AERONET AOT500 measurements do not demonstrate any local decrease, but just more uniform distribution. Of course, there can be cloud gap conditions during AERONET AOT measurements, however, on average, the cloud optical thickness contamination may induce much larger effect on aerosol climatology

than removing of few “good” cases. By this example we also illustrate the necessity of the additional more strict cloud screening in winter with the threshold at least equal to NA=6. Using this approach we obtained a revised dataset with additional visual cloud-screening over the whole 2001-2014 period of observations in Moscow. Figure 3a,b shows the absolute and relative differences between the standard monthly mean aerosol optical thickness at 500nm (AOT500) and additionally cloud-screened AOT500 values, as well as the differences in water vapor content, Angstrom exponent, and variation in day number over the whole period of measurements. One can see a substantial systematically overestimation of monthly mean aerosol optical thickness at 500nm in the standard AERONET dataset up to 0.03 for several months. For all months (except September and November) the error is higher than 0.01, which corresponds to the uncertainty of AOT measurements (ïĄě=0.01, depicted by the line in Fig. 3). In some years the monthly mean difference can even exceed 0.1 (for example, in February 2005 and October 2012). A detailed analysis was made to understand the reasons of these large discrepancies with the standard AERONET AOT dataset. Fig.4 shows the comparison between the standard daily mean AOT500 and Angstrom exponent data and their values after additional cloud filtering. In February 2005 additional cloud filtering provided full elimination of measurements during 03.02.2005 and 27.02.2005, which were characterized by smaller Angstrom exponent and extremely high AOT on 27/02/2005. According to the additional checks we found that all these cases were observed in solar disk conditions with SD=1 or SD=0. The additional cloud filtering also provides a slight increase in Angstrom exponent during the other days in February 2005, that also indirectly confirms the elimination of cloud-contaminated cases. In October 2012 the application of the additional cloud filter provided removal of high AOT500 values on 04/10/2012 due to the existence of overcast cloud conditions with SD=1 and SD=0. However, since the Angstrom exponent was not small the cloudiness was rather thin on 04/10/2012. During the other days in October the difference in both aerosol characteristics obtained before and after the additional cloud filtering was negligible. Due to existing AOT seasonal change the relative difference in AOT500 has a noticeable minimum in summer (5%) and the increase up to 20-30% during winter months when the occurrence of upper cloudiness is high and AOT is low. There is also discernible underestimation of Angstrom exponent in the standard AERONET dataset due to the influence of close to neutral scattering on large cloud droplet, which contaminate AOT values which are used for the Angstrom exponent evaluation. The relative bias in Angstrom exponent has also some tendency towards higher underestimation (from -1-2% to -6%) in the standard product in cold period. Both positive AOT difference and negative Angstrom exponent difference clearly indicate the reliable elimination of cloud contaminated cases after the application of additional cloud filter. It is interesting that water vapor content W is also overestimated in cloud contaminated conditions up to 0.05-0.07 cm (or 15-20%) during winter months possibly due to the additional absorption by ice and water particles. However, there can be another reason for this phenomenon: the air convergence may create favorable conditions with higher relative humidity and water vapor content in the atmosphere, and, hence, the existence of clouds (see, for example, Jeong, Li, 2010). However, these processes should be studied further. After the application of an additional cloud filter the day number significantly decreases (see Fig.3b): up to 7-20% during warm period, and 25-45% in cold period due to higher occurrence of overcast upper layer cloudiness and the application of more strict filter NA<6. Note, that small day number with aerosol observations in winter due to cloudy conditions results in large relative changes of the removed day number even when only 1-2 days are removed from the initial statistics. These values are in a qualitative agreement with the cirrus susceptibility percentage tests of AERONET level 2.0 AOT retrievals against the CALIPSO vertical feature masks (Huang et al.,2012), but they differ from the similar assessments against Micro-Pulse Lidar data shown in the same paper. However, the application of the Micro-Pulse Lidar data for evaluating the cirrus cloud contamination over Tropical area (Chew et al., 2011) has revealed much higher susceptibility percentage (about 23-34%) of the AOT sample, which is in qualitative accordance with our data for winter months. However, several recent studies indicated that clouds can have a real impact on AOT. These

mechanisms of aerosol/cloud interaction include aerosol hygroscopic growth, increasing aerosol concentration due to air convergence, and new particles formation in the presence of clouds (Su et al., 2008, Jeong , Li, 2010, Eck et.al. 2012, Eck et.al. 2014). In addition to well known hygroscopic growth of particles there is a mechanism of the gas-to-particle conversion, which occurs more intensively in the aqueous phase in cloud droplets due to the oxidation of gases (SO2, NOx, SOA) (Eck et al., 2014). Due to this mechanism in the presence of convective cloudiness the formation of new aerosol particles may observe providing higher aerosol loading during the periods with higher cloud amount in the vicinity of clouds (Eck et.al. (2014). The same mechanism also provides lower Angstrom exponent values. Another mechanism of simultaneous variations in both aerosol and cloud amount is the changes in meteorology conditions, when, depending on advection and circulation features one can obtain synchronous changes in AOT and cloud amount, which do not interact with each other. A good example of this effect is the noticeable dependence of AOT and Angstrom exponent on various cloud filters for January. As discussed above, according to the results of 24-hour HYSPLIT backward trajectory analysis the collocated changes in AOT and cloud amount (value of cloud filter, see Fig.1) are likely observed due to the changes in advection. However, during warm period according to our long-term dataset we did not obtain the AOT-cloud amount dependence, except 100% contaminated cases when the NA<9 threshold filter has been applied. We should also mention that after applying this filter, which removes the data when solar disk was blocked by cloud, we do not remove any cases with particular convective cloudiness development, except those, which have been removed by the standard AERONET cloud-screening algorithm. However, it will be interesting to compare the results with the coming AERONET version 3 dataset, where a modification of standard cloud-screening algorithm will be applied to the data according to the method described in (Eck et al., 2014)."

2.3. Although the correction of NO2 is a minor correction compared with that performed with a second cloud screening, it is described in more detail. In this last case, and from a methodological point of view, authors should better assess the new findings in relation to those reported in the paper of Chubarova and Dubovik (The sensitivity of aerosol properties retrievals from AERONET measurements to NO2 concentration over industrial region on the example of Moscow) published in 2004.

2.3. Response. Yes, thank you. We have added the additional material in the text following your comment. 2.3. Changes in the text at line 407 (new numeration): Large NO2 content has also the influence on the retrievals of other aerosol characteristics, which are not considered in this study. However, according to the previous cases study analysis we showed the pronounced effects of NO2 on the retrievals of single scattering albedo, which can increase up to 0.02 when the ratio OTNO2/AOT at 440nm is about 10% (Chubarova and Dubovik, 2004). The influence of NO2 on the retrievals of aerosol size distribution is also pronounced with the artificial bias towards smaller particles with overestimating the fine mode fraction of about dV/dlnr= 0.02 ïĄ■m3/ïĄ■m2 at r=0.05-0.065 ïĄ■m and the decrease over 0.01-0.03 ïĄ■m3/mm2 at 0.11-0.15 ïĄ■m for typical air pollution conditions (Chubarova and Dubovik, 2004). In overall, the fine mode fraction due to accounting for NO2 content changes on 1-5%. We should note that in (Chubarova and Dubovik, 2004) only few cases (n=14) were analyzed while in this study we considered the NO2 effects on AOT climatology over the whole period of measurements. In addition, in (Chubarova and Dubovik, 2004) the evaluation of the NO2 content was made using the model vertical profile according to the global 3-D GEOS-CHEM model (Martin et al., 2002), while in this paper we applied the NO2 profile in the low troposphere using the parameterizations obtained according to the in-situ NO2 measurements up to 350meters and photochemical model directly for Moscow conditions.

2.4. Concerning the NO2 climatology it is clear that AOT corrections are needed in megacities and major urban sites where NO2 values are significantly higher than those of the SCIAMACHY data base, although we must emphasize that this correction is, in most cases, lower than the AOT measurement uncertainty (0.02). However, the methodology used in this study does not seem the most appropriate since it is limited

to replacing the AERONET SCIAMACHY-based NO2 climatology by an "improved" NO2 climatology based on observations, when in the second part of the paper an AOT trend analysis is approached. Why not address the correction of the AOT series with observed daily/monthly/annual NO2 data series? NO2 varies significantly in very short periods, and clear negative trends in large urban areas have been reported (i.e. Schneider et al., (2015) and Hilboll et al. (2013), the latest referenced in the paper).

2.4.Response: We followed here the methodology used in the standard AERONET algorithm, which utilizes monthly mean NO2 estimates, which then are interpolated on a daily scale. We used the monthly mean climatology since, as we mentioned in the paper according to Hiboll et al., (2013), no statistically significant columnar NO2 trend is observed in Moscow. (see 7858 line 7 in the old variant of the paper). We have added the important word – “statistically” there. We also added the results of (Schneider, 2015) as well. The NO2 correction is not very large compared with the AOT uncertainty level therefore the slight negative changes would not produce any significant changes in AOT signal. Since we are interested in the paper in long-term trend this is not necessary to correct AOT on daily basis. The daily correction is a very challenging and it requires the application of another algorithm.

2.4. Changes in the text at line 373(new numeration): We added some clarification in the 3.1.2 result section after the paragraph “The estimated NO2 . . ..” as well as follows:. “The effect of NO2 content on the AOT retrievals is not very large and since there is no statistically significant trend in NO2 content over Moscow according to our data as well as according to satellite retrievals (Hiboll et al., 2013, Shneider et al., 2015), we suggest to account only for monthly mean NO2 values.”

However, in the future we should possibly take the corresponding satellite data for this purpose after their rigorous validation. We added this point in the Discussion section at 652-653 line: “The NO2 correction over other megacities can be also made according to long-term satellite NO2 retrievals but after their rigorous validation”

2.5. The joint analysis of AOT data series and SOx, CO and NOx emissions is performed with annual mean values. This is a simple approach that can be misleading, especially when the authors previously identified that the statistically significant trends of mean and daily maxima AOT500 values are observed in April, May and September.

2.5. Response: Yes, we agree that the monthly mean data on emissions will be much better. Unfortunately, they are unavailable. Concerning the trends – we have the negative (or zero) tendency in AOT trend for most months, except November and the statistically significant negative AOT trends for several months. Since having the same tendency for most months, we think that that we can consider the annual dataset on emissions. In the new version we added the additional analysis on possible reasons in AOT trends. Please, look at our response to your comments below and the updated text in this Section. 2.5. Changes in the text from line 522 (new numeration) "There can be several natural or anthropogenic reasons for these negative AOT trends. In order to study the effect of anthropogenic emissions we used the officially reported emission data from the Centre on Emission Inventories and Projections WebDab – EMEP database (http://www.ceip.at/status_reporting/2014_submissions/). Fig 11a presents temporal variations in emissions of different main aerosol precursors over the European part of Russia, which can affect the secondary aerosol generation in Moscow. One can see a statistically significant at P=95% decrease in SOx emission of about 135 Gg yr-1 per year (or 135 Gg yr-2), the negative trend in emission of Non-methane volatile organic compound (NMVOC) of about 54 Gg yr-2. In addition, the CO emissions, which do not influence directly on the secondary aerosol generation but may characterize the intensity of pollution from the transportation sources, has also a pronounced negative trend of about 69 Gg yr-2. This negative trend also confirms the complex character of the atmosphere cleanup. There is also a tendency of NOx decrease over European part of Russia, especially during the last years but it is not statistically significant. Some negative tendency is observed in emissions of the particulate matter with the diameter less 2.5 ïĄ■m (PM2.5). The comparison of temporal variability of main aerosol precursors over the European part of Russia and in Moscow is shown in Fig.11b. There we also present the trend in annual 50% quantile AOT500, which is not sensitive to the extremely high aerosol loading during the Moscow 2002 and 2010 fire episodes. One can see the absence of local changes in SOx in Moscow compared with a distinct negative trend in SOx up to -6.5% a year over the European part of Russia, which can be observed due to changes in fuel from coal to gas. In Moscow this change of fuel has been made earlier, at the end of 1980s. Note also, that the high median AOT values in 2006 correspond well with the elevated emission of SOx both in Moscow and at the whole European part of Russia as well as the elevated emission of NOx in Moscow. The last years are characterized by a decrease in NOx emission both in Moscow and at the European part of Russia possibly due to improving the quality of petrol standards. As a result, we assume that negative trend in AOT is observed likely due to the decrease in anthropogenic emissions of SOx and NMVOC over European part of Russia, which play a significant role in the second aerosol generation, especially during warm period. Some important role can also play the decrease in NO2 emission during the last years since 2010. However, natural AOT variations should be also taken into account. For example, since the AOT spatial distribution is characterized by a significant decrease from south-east to the north in Europe (Chubarova, 2009) natural AOT interannual variability can be observed due to the year-to-year variability of different air mass advection. We tested this effect and its possible influence on interannual AOT variability for the months with statistically significant negative trends - April, May, and September. For this purpose we compared the results obtained over the whole period of observations and over the last 5 years since 2010, when low 50% quantile AOT500 values were observed (see Fig.10b). For this purpose we used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) to generate the 24-hour backward trajectories for the days with AOT measurements in April, May and September at the altitude H=500m for 12:00 UTC. Since Moscow is located close to the center of the European Plain and there is almost the same probability of air parcel arriving from different directions, we combined the results in the standard wind diagram and compared the change in the relative number of cases in different directions over the whole period of measurements (2002-2014) with that over the last years (2010-2014). We will consider that the significant difference in circulation pattern occurs, when the change in relative number of cases over a particular direction exceeds 5%. In addition, we calculated the mean daily AOT500 for the air masses coming from different directions. Fig.12 presents the obtained wind diagrams as well as the mean daily AOT500 diagrams over these two periods. One can see that in most cases there is no significant difference in wind diagrams between 2010-2014 and 2002-2014 periods for all three months. The exception was observed in May with small prevailing of air mass advection from the East (+7%), accompanied by slightly lower AOT (difference in AOT500=-0.02), and in September with small prevalence of the air mass advection from the North (+6%) accompanied by slightly higher AOT values (difference in AOT500=+0.03). Lower AOT500 values during the last 2010-2014 period were observed almost at all the directions of air mass advection with the difference of about 0.02ïĆÿ0.14 in April, 0.02ïĆÿ0.10 in May and 0.03ïĆÿ0.18 in September. The increase in AOT500 higher than 0.01 was only observed in conditions with South-West air mass advection in April (difference in AOT500=+0.09), which occurrence is small, and in September with North and East air mass advection when there was a slight difference in AOT500=+0.03. Hence, we can state that there were no significant changes in circulation pattern during the last years. Note, that the data from September, 2002, when the intensive forest fires were observed and AOT500 was unusually high, were not used in this analysis. Wet aerosol deposition, regulated by precipitation, can also play an important role in year-to-year AOT variability. In addition, the enhancement of the dynamic stability of the atmosphere can be an effective factor leading to the stagnation of air and, hence, to the aerosol accumulation. As a parameter characterizing the atmospheric instability we used the convective available potential energy (CAPE) (Barry and Chorley, 1998). The CAPE data from the ERA-Interim re-analysis over Moscow (36-38âĄřE, 55-56âĄřN) were taken for the days, when the aerosol measurements were made. As a result, multiple regression analysis has been applied for studying the relationship of monthly mean AOT500 with temperature (as an indicator of air advection), precipitation, wind speed, wind direction and CAPE characteristics according to the Moscow dataset over the whole period of measurements. However, the analysis revealed the absence of any significant AOT correlation with any of the characteristics considered. This means that natural factors might not be responsible for the negative AOT trend in the Moscow area. In addition, we compared the changes of meteorological parameters, AOT500, the annual emissions of main aerosol precursors and PM2.5, observed during the last 2010-2014 period with their values for the whole dataset 2002-2014. We have to analyze the existing 2002-2013 dataset for emissions and assume, that they do not vary within the year, since the monthly resolution data are not available. All data were normalized against their means. Fig.13 shows error bars interval at confidence level P=95% of relatively changes over 2002-2014 in monthly mean AOT500, air temperature, precipitation, CAPE, as well as NMVOC, NOx, PM2.5, SOx emissions and, for comparison, the mean relative changes of these characteristics over the 2010-2014 period for April, May and September. One can see that the mean negative changes in emissions during the last 2010-2013 period were significantly higher than those over the whole period while the relative changes in meteorological factors demonstrate different signs, except the precipitation, which slightly increases in all months. Their mean relative changes lie mainly within the error bars interval at P=95%, except air temperature in May, and CAPE in September. However, for the other months these parameters have even the opposite sign, which might means the random character of their change. Hence, we should state that the effect of the negative trend in emission likely have the main influence on negative AOT500 trend, which was observed over Moscow. There are some slight changes in meteorological regime and advection, but they seems to be not very important."

Đąhanges in the text from line 694 (new numeration): To understand the cause of the negative trends we used the officially reported emission data from WebDab – EMEP database (http://www.ceip.at/status_reporting/2014_submissions/). According to these data we showed that the decrease in AOT in 21 century can be observed due to statistically significant at P=95% negative trends in SOx emission of about 135 Gg yr-2, in NMVOC emission of about 54 Gg yr-2, which can affect the secondary aerosol generation. We found that the high median AOT values in 2006 correspond well with the elevated emission of SOx both in Moscow and at the European part of Russia, as well as with NOx – in Moscow. The last years are characterized by the decrease in NOx emission both in Moscow and at the European part of Russia possibly due to improving the quality of petrol standard. However, the NO2 trend in Moscow and over European part of Russia is not statistically significant. We also studied the possible effect of natural factors in interannual AOT variability. According to the 24-hour NOAA HYSPLIT model backward trajectory analysis at 500m AGL for 12h UTC we obtained the wind diagrams and the distribution of daily AOT500 at different directions of the air mass advection for the months with statistically significant negative AOT trends (April, May, September). However, no significant difference in wind diagram is observed over 2010-2014 compared with the 2002-2014 period for all three months except the small increase (+7%) in conditions with the East air mass advection, accompanied by slightly smaller AOT in May, and the small increase (+6%) of air mass advection from the North with slightly higher AOT values in September. At the same time we see a significant drop in AOT500 values almost at all directions, except South-West air mass advection in April, which occurrence is small, and in conditions with North and East air advection in September. No statistically significant correlation was obtained in monthly mean AOT relationship with different meteorological parameters and CAPE. The analysis of relative changes in different characteristics obtained during the last years against the whole period of observations has revealed that mean negative changes in emissions of aerosol precursors over the 2010-2013 period were significantly higher than those over the whole period, while the relative changes in meteorological factors demonstrate different signs, except the precipitation, which slightly increased in all months. However, its changes are not statistically significant. This means the importance of the anthropogenic factor (negative emissions of aerosol precursors) for attributing the negative AOT trend in Moscow.

2.6. Why not pay attention to explain trends in these months taking into account that

atmospheric processes driving AOD are not the same throughout the year? Related to this, somewhat surprisingly, the authors claim that no significant correlation was statistically obtained in AOT relationship with different meteorological parameters and CAPE. Probably there is no correlation working with annual averages, but it is difficult to admit that in certain months/seasons, meteorological conditions do not affect AOT values. It is clear that the synoptic patterns may be subject to important interannual variations in certain months/seasons affecting local/regional meteorology around Moscow (i.e. precipitation, affecting wet deposition, wind and mesoscale convective processes, affecting dust resuspension. . .) and aerosol long-range transport from very different regions. Surely, the negative emissions of aerosol precursors play an important role in the AOT negative trend found in Moscow, as stated by the authors, but they should further investigate the role played by air masses transport and, for that, the study should be performed for each month/season, at least in months when a clear negative trend in AOT is observed (April, May and September).

2.6. Response: Yes, of course, when analyzing the possible influence of meteorological factors on AOT, we have used the data on monthly scale. In the new version of the text we have added two additional Figures and included the additional discussion on possible influence of natural variability. I agree that meteorological factors can play the important role in AOT interannual variability. I mentioned in my previous paper the importance of synoptic processes (see, for example, discussions in Chubarova AMT, 2009). However, even after additional analysis we still obtained the same conclusion. As we have already mentioned, we have added some additional analysis including the analysis of backward trajectories for the months with statistically significant trend and some other analysis to clarify the obtained results.

2.6. Changes in the text were made in the whole Section 3.3 (please, look at our previous response) : The changes from line 498 (new numeration): In April and September statistically significant negative trends were also obtained for 50% quantile AOT500. So we can state that the most significant AOT decrease is observed in spring and fall periods, however, negative tendencies are observed almost throughout a year. . The changes from line 556 (new numeration) “For this purpose we used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) to generate the 24-hour backward trajectories for the days with AOT measurements in April, May and September at the altitude H=500m for 12:00 UTC. Since Moscow is located close to the center of the European Plain and there is almost the same probability of air parcel arriving from different directions, we combined the results in the standard wind diagram and compared the change in the relative number of cases in different directions over the whole period of measurements (2002-2014) with that over the last years (2010-2014). We will consider that the significant difference in circulation pattern occurs, when the change in relative number of cases over a particular direction exceeds 5%. In addition, we calculated the mean daily AOT500 for the air masses coming from different directions. Fig.12 presents the obtained wind diagrams as well as the mean daily AOT500 diagrams over these two periods. One can see that in most cases there is no significant difference in wind diagrams between 2010-2014 and 2002-2014 periods for all three months. The exception was observed in May with small prevailing of air mass advection from the East (+7%), accompanied by slightly lower AOT (difference in AOT500=-0.02), and in September with small prevalence of the air mass advection from the North (+6%) accompanied by slightly higher AOT values (difference in AOT500=+0.03). Lower AOT500 values during the last 2010-2014 period were observed almost at all the directions of air mass advection with the difference of about 0.02ïĆÿ0.14 in April, 0.02ïĆÿ0.10 in May and 0.03ïĆÿ0.18 in September. The increase in AOT500 higher than 0.01 was only observed in conditions with South-West air mass advection in April (difference in AOT500=+0.09), which occurrence is small, and in September with North and East air mass advection when there was a slight difference in AOT500=+0.03. Hence, we can state that there were no significant changes in circulation pattern during the last years. Note, that the data from September, 2002, when the intensive forest fires were observed and AOT500 was unusually high, were not used in this analysis.”

At line 591 (new numeration): In addition, we compared the changes of meteorological parameters, AOT500, the annual emissions of main aerosol precursors and PM2.5, observed during the last 2010-2014 period with their values for the whole dataset 2002-2014. We have to analyze the existing 2002-2013 dataset for emissions and assume, that they do not vary within the year, since the monthly resolution data are not available. All data were normalized against their means. Fig.13 shows error bars interval at confidence level P=95% of relatively changes over 2002-2014 in monthly mean AOT500, air temperature, precipitation, CAPE, as well as NMVOC, NOx, PM2.5, SOx emissions and, for comparison, the mean relative changes of these characteristics over the 2010-2014 period for April, May and September. One can see that the mean negative changes in emissions during the last 2010-2013 period were significantly higher than those over the whole period while the relative changes in meteorological factors demonstrate different signs, except the precipitation, which slightly increases in all months. Their mean relative changes lie mainly within the error bars interval at P=95%, except air temperature in May, and CAPE in September. However, for the other months these parameters have even the opposite sign, which might means the random character of their change. Hence, we should state that the effect of the negative trend in emission likely have the main influence on negative AOT500 trend, which was observed over Moscow. There are some slight changes in meteorological regime and advection, but they seems to be not very important.

Specific comments:

Page 7845; Line 7: Some basic references should be provided for each satellite platform. Acronyms should be described.

2.7. Response: We described the acronyms, however, we included only the general reference to the publication, where all the satellite instruments are discussed. We did so, since the aim of our paper does not connect with satellite retrievals. 2.7. The changes in the text at line 37 (new numeration):: “Different aerosol characteristics are possible to obtain from satellite instruments, i.e. Advanced Very High Resolution

Radiometer (AVHRR), Moderate Resolution Imaging Spectroradiometer (MODIS), Advanced Along-Track Scanning Radiometer (AATSR), medium-spectral resolution, imaging spectrometer MERIS, Polarization and Directionality of the Earth's Reflectances (POLDER), Ozone Monitoring Instrument (OMI), Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), Sea-Viewing Wide Field-of-View Sensor (SeaWiFs), Multi-angle Imaging Spectroradiometer (MISR), etc. (IPCC, 2013)."

2.8. Page 7845; Line 10: Some basic references should be provided for each aerosol network. Acronyms should be described. 2.8. Response: We added the description of the acronyms. Concerning the references, we think is better for a potential reader to look further using the direct link to the WMO web page (http://www.wmo.int/pages/prog/arep/gaw/aerosol.html), where all the networks are described. 2.8. Changes in the text at line 44(new numeration) : "Ground-based aerosol networks such as Global Atmosphere Watch Precision Filter Radiometers (GAW-PFR), AErosol RObotic NETwork (AERONET), observation network SKYNET, Siberian system for aerosol research (SibRad), Micro-Pulse Lidar System (MPLNET) provide high quality aerosol measurements (http://www.wmo.int/pages/prog/arep/gaw/aerosol.html)."

2.9 Page 7845; Line 14: ".. is equipped by Cimel sun/sky..." should be "... is equipped with Cimel sun/sky..."

2.9 Response: Done. Thank you.

2.9 The new variant: "AERONET is equipped with CIMEL sun/sky photometers,. . .."

2.10 Page 7846; Lines 26-27: Notice that the uncertainty of 0.01 corresponds to a Master instrument. The uncertainty of a filed instrument, as that of Moscow MSU MO AERONET site, is 0.02 in the visible range (Eck et al., 1999).

2.10 Response: According to the paper (Eck et al., 1999) "the uncertainty of Master instrument is about 0.002-0.005 increasing to 0.01-002 (wavelength dependent) for

field instruments". In (Holben et al., 2001) there was an additional clarification that the uncertainty of 0.01 belongs to visible and infrared channels and 0.02- to UV. We added the reference to (Holben et al., 2001). Since Dr. Thomas Eck (the author of the mentioned paper) was the first reviewer of this manuscript and he did not make any comments on this account, we feel that the uncertainties which were given in the paper are right. 2.10 The new variant of the text is the following at line 87 (new numeration): "The uncertainty of aerosol optical thickness measurements does not exceed 0.01 in visible range and 0.02 - in UV spectral range (Eck et al., 1999, Holben et al., 2001)."

2.11. Page 7847; Section 3.1.1. This issue was already addressed by Chubarova et al (2011) using similar criteria and even then with little details. Authors should explain whether the cloud screening was performed only for cirrus or for all type of clouds. They must clarify whether they applied the cloud screening using quasi-simultaneous Cimel-Cloudiness observations at the same site, and then corrected daily averaged AOT values are obtained. They must also address the fact that although there is presence of cirrus, the Cimel direct sun measurements may not be affected by them (cirrus are not blocking the sun) causing valid Cimel data removing. How can this affect the corrected AOT database?

2.11. Response: Thank you for a helpful comment. We have added additional analysis including several additional Figures for clarifying the obtained results (see the attached full updated text with Figures and separately all the figures in the attachment). We discussed in the text the possibility of using the total cloud amount filter since the ability to measure direct sun and the applied standard-cloud screening provide us the situation with possible residual contamination by optically thin homogeneous cloudiness. Please, look at the response and the changed text to your previous general comments 2.1 and 2.2. In addition, we have replaced the term "cirrus cloud" by "upper layer cloudiness" (or "upper later thin homogeneous clouds") throughout the text for better clarification since there are a lot of different types of upper layer cloudiness which can affect the AOT measurements. In addition, we tested our dataset on solar disk conditions, which have revealed that all the data during warm period and almost all (but 0.5%) in winter were under the influence of cloud contamination. Please, look at the text below and the changed text applied to your general comment (2.1, 2.2). In addition, we clarified that application of the cloud screening procedure. 2.11 Changes in the text from 143 (new numeration): We used one-hour resolution cloud observations for additional filtering of quasi-simultaneous Cimel observations at the same site. The application of cloud filter means the elimination of all AOT measurements for the entire hour interval. We showed there that the existing standard cloud-screening algorithm works perfectly, when aerosol measurements are contaminated by optically thin low layer cloudiness, which is characterized by large triplet variations. These variations are used as a parameter in the standard cloud-screening algorithm developed by Smirnov et al. (2000). However, if the cloud blocking the Sun is thin and uniform, the triplet variation can be small and the contaminated AOT measurements pass through the filter. Mainly the cirrus clouds are characterized by these properties. However, in general, according to the International Cloud Atlas (1987) other types of clouds may be also characterized by these properties as well. They include different forms of Cirrostratus, and even Altostratus translucidus clouds, which relate to the middle cloud level. In this publication all types of the cloudiness, which can induce the potential contamination of AOT will be combined under the term “upper cloudiness”. Since low cloudiness is effectively filtered out by the standard cloud-screening algorithm we proposed to apply simple total cloud amount (NA) filter, which is sensitive to the existence of upper cloudiness. In this context NA value (together with the application of standard cloud-screening procedure) provides the information about the potential existence of high and middle layer cloudiness, since the standard AERONET cloud-screening algorithm successfully removes the cases contaminated by low level clouds or the cases with strong signal variations. However, the application of different NA thresholds may provide the different samples and as a result, different statistics. As an example, in Fig.1 we demonstrate the effects of utilizing the different additional NA filters for AOT at 500nm (AOT500) and Angstrom exponent (ïĄą) datasets for the central months of the seasons. One can see a similar tendency of the AOT decrease in all seasons after removing the cases with the threshold of NA<9, which includes the conditions with almost total cloud amount of NA= 9-10. Additional testing on solar disk conditions has revealed that all eliminated cases in this sample belong to the situations, when solar disk was covered by clouds (SD=1 or SD=0). In April after eliminating the almost overcast cloud conditions with NA<9 threshold there is no further changes in AOT500 samples with more strict NA cloud threshold. At the same time the sample is dramatically (more than twice) reduced (from 229 to 113 in NA<3 sample). In July we also see a slight decrease in AOT500 in the NA<9 sample and, in addition, a significant growth of Angstrom exponent. Note, that in July and October we see even a slight increase in the AOT500 for almost clear sky conditions (the sample with NA<3). In January there is a pronounced decrease in AOT500 with reducing cloud amount. The lowest AOT500 values and largest Angstrom exponent are observed in the NA<3 sample. The application of the 24-hour Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) NOAA model backward trajectory analysis (Draxler and Hess, 1998) for all the cases in January has revealed for the NA<3 sample the prevalence of the northern (North-West, North, North-East) advection (80% of cases) characterized by low AOT (Chubarova, 2009), compared with 40% of cases for partially cloudy conditions (3<NA<9). At the confidence level of P=80% several of these dependencies are statistically significant (see the error bars in Fig.1). Balancing between the substantial decrease in case number and the accuracy of the retrievals of aerosol properties we showed that the best results were obtained when the cases with Ntotal.<9 during March-October period with almost overcast cloudiness and cloud contaminated solar disk conditions data were removed. For November –February conditions the filter threshold is more strict (Ntotal<6) since solar elevation in Moscow is low (hnoon<25ïĆř) at this time and a well-known effect of significant visual cloud amount increase towards the horizon plays a vital role. Smaller cloud threshold during winter time may induce filtering out so called “good” AOT cases. The additional analysis of solar disk conditions has revealed only 12 cases from 2521 cases (about 0.5%), which were not contaminated by clouds (SD=2) and were incorrectly removed from the sample. All of them were observed in February (18.02.2011 – 4 cases, 3.02.2003- 5 cases, 11/02/2007 - 3 cases). However, even these three days are presented in the final sample since during these days there were other sun photometer measurements at smaller NA.

2.12. Moreover, authors should explain the limitations of visual observations, as these observations are subjective, and performed by several observers may have very different observation skills.

2.12. Response: Yes, you are right that the accuracy of visual cloud amount measurements is not good and is about 1 or 2 of cloud cover values expressed in tenth. However, this is not a problem to distinct the situation of overcast conditions. In addition, we used solar disk condition dataset to test the additional cloud-screening measurements. We added the discussion on the quality of the visual measurement in the text.

2.12. Changes in the text at line 92 (new numeration): “For the improvement of these procedures, in addition, we used visual cloud observations with 1 hour resolution. The uncertainty of visual cloud amount measurements is about 1 or 2 cloud fraction (in tenth) according to (Handbook, 1989), however, the conditions with overcast or zero cloudiness are observed accurately by any observer. In addition, the dataset of hourly solar disk condition observations, which are performed simultaneously with cloud observations, was used in the analysis. This is a standard type of observations at the actinometrical stations in Russia. Using this characteristic we can distinct the conditions, when solar disk (SD) is free from clouds, or when SD is obscured by thin clouds (shadows can be seen at ground), or when SD can be seen but there are no shadows at ground, or when SD can not be seen due to relatively high cloud optical thickness. These SD conditions are noted with “2”, “1”, “0” and “P” marks, respectively.”

2.13. Page 7850, Line 13: The following paper: Chubarova, N. E., Larin, I. K., and Lezina, E. A.: Experimental studies and modeling of nitrogen dioxide variations in the lowest troposphere layer in Moscow, Newsletter Moscow State University, series 5,

Geography, 5, 11–18, 2010, in which is based the evaluation of NO2 content, is not easy to be accessed. It might be included as supplement information. What is the NO2 uncertainty from in-situ observations + modeling?

2.13. Response: We added some additional clarification and some additional information of the method applied directly in the section with the description of the NO2 correction. Please, look at the changes below:

2.13. Changes in the text from line 319 (new numeration): “For accounting the NO2 amount up to the height of 350m we utilized the developed parameterizations of its content within 350m according to in-situ long-term NO2 measurements in the boundary layer from ground to 350m in several points of Moscow (at the Ostankino tower and at the top of Moscow State University Building) during summer and winter conditions to account for possible differences in meteorological factors like boundary layer altitude, temperature and photochemistry effects. These data were combined with the results of photochemical model, which had been adapted to the available experimental data on different chemical constituents and meteorological conditions in the boundary layer. Input model parameters include spectral flux of solar radiation; absorption cross sections and quantum yields of photodissociation products; rate constants of chemical reactions; the altitude temperature profiles, turbulent diffusion coefficients, concentrations of some atmospheric components, and meteorological parameters which were measured during the experiments [Chubarova et al., 2010]. The applied 1-D photochemical model calculated the vertical profiles with 50-meter resolution up to 20 km and takes into account for several hundreds of chemical reactions for 100 components. We also used the temperature profiles from Microwave Temperature Profiler MTP5 (Kadygrov et al., 2003) up to 600 m for the evaluation of the diffusion coefficients to account for the different boundary layer conditions. As a result, various weighting coefficients for summer and winter conditions were evaluated for different layers: 0-350m, 350-1000m, and 1000-2000 m. According to these data we obtained two regimes of NO2 vertical distribution typical for Moscow conditions within the low 2 km layer.”

2.14.. Page 7850; Line 13-Page 7851; Line 2; Detail, please uncertainties in NO2 estimation related with Boundary Layer height assumptions. 2.14. Response: As it is seen from the response to the previous comment we used in addition the information from collocated records from MTP5 instrument for assessing the information about the boundary layer. According to these data we inferred the turbulent coefficients for modeling the NO2 concentration up to 2 km. 2.14. change in the text at line 332: “We also used the temperature profiles from Microwave Temperature Profiler MTP5 (Kadygrov et al., 2003) up to 600 m for the evaluation of the diffusion coefficients to account for the different boundary layer conditions. As a result, various weighting coefficients for summer and winter conditions were evaluated for different layers: 0-350m, 350-1000m, and 1000-2000 m. According to these data we obtained two regimes of NO2 vertical distribution typical for Moscow conditions within the low 2 km layer.”

2.15. Page 7851; Lines 20-26; Errors of the means should be added in estimated NO2 optical thickness. OT(NO2) values are lower (no higher) than the uncertainty for AOT in wavelengths > 440 nm (0.02).

2.15. Response: We have added the error of the means to the estimated NO2 and made some additional changes in the text: 2.15. Changes in the text at line 363(new numeration): “The estimated NO2 optical thickness (OTNO2) in different CIMEL spectral channels is shown in Fig.5b. The most pronounced effects of OTNO2=0.02-0.03ïĆś0.003 are observed for 380 and 440nm channels due to the strongest NO2 absorption there..”. . ... And from line 367 (new numeration): “It should be emphasized that the added OTNO2 values are close to the uncertainty threshold of aerosol optical thickness evaluation of ïĄ¿0.02 at 340nm and are usually higher or comparable with the uncertainty threshold for AOT at other wavelengths especially, in winter and spring conditions, which should be necessary to take into account.”

2.16. Page 7857; Lines 22-23; Authors should discuss the fact that no trend is observed in PM2.5 concentration when they attribute the negative trend of AOT500 to negative emissions of aerosol precursors (basically of anthropogenic origin), since PM2.5 mainly

accounts for anthropogenic aerosols.

2.16. Response: Sorry for a misprint in the previous version of the text. It was not the PM2.5 concentration but PM2.5 emissions, which do not depend on aerosol precursors. We do not study the trends in PM 2.5 in this paper. Note, that due to updated EMEP database we were able to add some data and the numbers of trends have been slightly changed.

2.16 Changes in the text at line 534 (new numeration): Some negative tendency is observed in emissions of the particulate matter with the diameter less 2.5 ïĄ■m (PM2.5).

2.17. Page 7858; Lines 2-4; Although the assessment of AOT trends has been addressed in General Comments, authors should avoid simplistic and misleading arguments, like this one. In 2009 emissions of SOx and NOx, similar to 2006, were registered but, instead, AOT values were significantly lower than in 2006. Why?

2.17. Response: We agree that this analysis in the previous variant of the paper was simplistic. However, this is a challenging task to explain the AOT trends. And sometimes we have to use the qualitative approach . Concerning 2009: I guess that in 2009 the large SOx and NOx emissions were observed only in Moscow while over European territory the SOx emission was small and declining. (Please, look at Fig. 8b). We have added the analysis using the two additional Figures Please, look at our response to the General comment (No 2.6) and at the revised text attached as supplementary material. . 2.17. Changes in the text: The changes in the text were made in the section 3.3, Section 4 (Discussion) and in Conclusions from line 556 (new numeration): "For this purpose we used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) to generate the 24-hour backward trajectories for the days with AOT measurements in April, May and September at the altitude H=500m for 12:00 UTC. Since Moscow is located close to the center of the European Plain and there is the same probability of air parcel arriving from different directions, we combined the results in the standard wind diagram and compared the change in the relative number of cases in different directions over the whole period of measurements (2002-2014) with that over the last years (2010-2014). We will consider that the significant difference in circulation pattern occurs, when the change in relative number of cases over a particular direction exceeds 5%. In addition, we calculated the mean daily AOT500 for the air masses coming from different directions. Fig.12 presents the obtained wind diagrams as well as the mean daily AOT500 diagrams over these two periods. One can see that in most cases there is no significant difference in wind diagrams between 2010-2014 and 2002-2014 periods for all three months. The exception was observed in May with small prevailing of air mass advection from the East (+7%), accompanied by slightly lower AOT (difference in AOT500=-0.02), and in September with small prevalence of the air mass advection from the North (+6%) accompanied by slightly higher AOT values (difference in AOT500=+0.03). Lower AOT500 values during the last 2010-2014 period were observed almost at all the directions of air mass advection with the difference of about 0.02ïĆÿ0.14 in April, 0.02ïĆÿ0.10 in May and 0.03ïĆÿ0.18 in September. The increase in AOT500 higher than 0.01 was only observed in conditions with South-West air mass advection in April (difference in AOT500=+0.09), which occurrence is small, and in September with North and East air mass advection when there was a slight difference in AOT500=+0.03. Hence, we can state that there were no significant changes in circulation pattern during the last years. Note, that the data from September, 2002, when the intensive forest fires were observed and AOT500 was unusually high, were not used in this analysis. From line: 591 (new numeration): In addition, we compared the changes of meteorological parameters, AOT500, the annual emissions of main aerosol precursors and PM2.5, observed during the last 2010-2014 period with their values for the whole dataset 2002-2014. We have to analyze the existing 2002-2013 dataset for emissions and assume, that they do not vary within the year, since the monthly resolution data are not available. All data were normalized against their means. Fig.13 shows error bars interval at confidence level P=95% of relatively changes over 2002-2014 in monthly mean AOT500, air temperature, precipitation, CAPE, as well as NMVOC, NOx, PM2.5, SOx emissions and, for comparison,

the mean relative changes of these characteristics over the 2010-2014 period for April, May and September. One can see that the mean negative changes in emissions during the last 2010-2013 period were significantly higher than those over the whole period while the relative changes in meteorological factors demonstrate different signs, except the precipitation, which slightly increases in all months. Their mean relative changes lie mainly within the error bars interval at P=95%, except air temperature in May, and CAPE in September. However, for the other months these parameters have even the opposite sign, which might means the random character of their change. Hence, we should state that the effect of the negative trend in emission likely have the main influence on negative AOT500 trend, which was observed over Moscow. There are some slight changes in meteorological regime and advection, but they seems to be not very important. "

from line 703(new numeration): "We also studied the possible effect of natural factors in interannual AOT variability. According to the 24-hour NOAA HYSPLIT model backward trajectory analysis at 500m AGL for 12h UTC we obtained the wind diagrams and the distribution of daily AOT500 at different directions of the air mass advection for the months with statistically significant negative AOT trends (April, May, September). However, no significant difference in wind diagram is observed over 2010-2014 compared with the 2002-2014 period for all three months except the small increase (+7%) in conditions with the East air mass advection, accompanied by slightly smaller AOT in May, and the small increase (+6%) of air mass advection from the North with slightly higher AOT values in September. At the same time we see a significant drop in AOT500 values almost at all directions, except South-West air mass advection in April which occurrence is small, and in conditions with North and East air advection in September. No statistically significant correlation was obtained in monthly mean AOT relationship with different meteorological parameters and CAPE. The analysis of relative changes in different characteristics obtained during the last years against the whole period of observations has revealed that mean negative changes in emissions of aerosol precursors over the 2010-2013 period were significantly higher than those over the whole period, while the relative changes in meteorological factors demonstrate different signs, except the precipitation, which slightly increased in all months. However, its changes are not statistically significant. This means the importance of the anthropogenic factor (negative emissions of aerosol precursors) for attributing the negative AOT trend in Moscow. "

from line 739 (new numeration): "The interannual changes in aerosol properties reveal distinct negative trends, which are statistically significant in April, May and September. We show that the main reason for the AOT decrease could be negative trends in emissions of different aerosol precursors over European Plain according to the WebDab – EMEP database. We showed that the AOT negative trend can be observed due to a noticeable decrease in SOx, NMVOC emissions at the European Plain as well as due to the additional decrease in NOx during the last years. The analysis of variability in natural factors has not revealed their significant influence on negative AOT trends. However, further studies will be helpful for understanding the role of specific emissions and their interaction with changing weather conditions. "

2.18 Figure 1 Caption; Units of water vapour in Figure 1a is cm?

2.18 Response Yes. Thank you. We added the clarification in Fig.1.

2.18 Changes in the text: “Fig.1 . The absolute (a) and relative (b) difference of monthly mean standard level 2.0 data on aerosol optical thickness at 500nm (AOT500), Angstrom exponent and water vapour with the dataset after additional cloud correction. The standard uncertainty of AOT measurements is shown in Fig.1a. Relative changes in day number removed after additional cloud correction is shown in Fig1b. Moscow, 2001-2014 period. Note, that in Fig.1a the difference in water vapour is given in cm and other characteristics are dimensionless.”

2.19. Figure 1 Caption; Notice that AOT uncertainty marked (0.01) corresponds to a Master AERONET instrument not to an AERONET field photometer (0.02 in this case).

2.19 Response: Please, look at our detailed answer to your previous specific comment, which concerns the similar issue. We left the uncertainty as is, since in (Holben et al., 2001) there was the clarification on this account. Reference to this paper is now added in the text.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/8/C3356/2015/amtd-8-C3356-2015-supplement.pdf

---

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 7843, 2015.

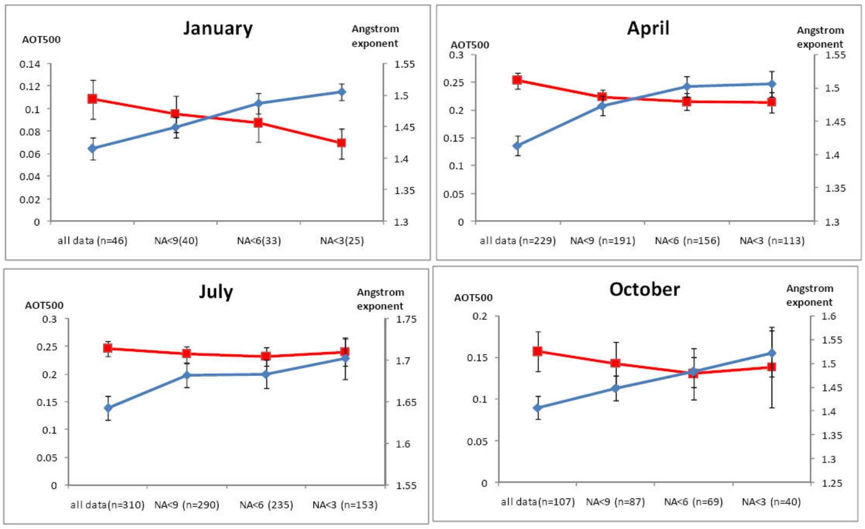


Fig.1. Mean aerosol optical thickness at 500nm (AOT500) and Angstrom exponent within 440-870 nm spectral range in different samples with various total cloud amount (NA) thresholds for the central months of the seasons. Moscow, 2001-2014. Number of days with measurements for each sample is given in brackets. Note, that the error bars are shown at the confidence level P=80%.

Fig. 1.

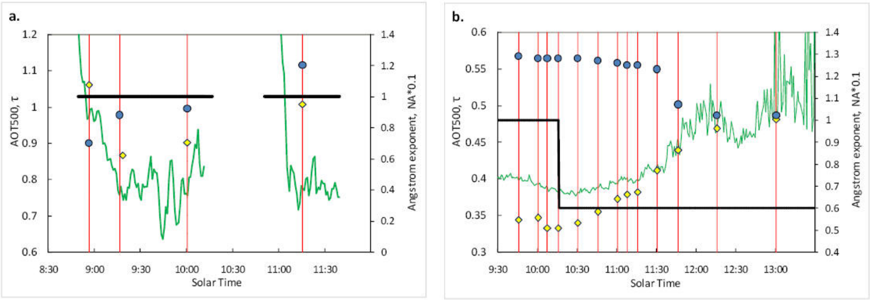


Fig.2. The time series in AOT500 (diamonds), integral optical thickness τ (green line), Angstrom exponent (circles), and total cloud amount NA*0.1 (black lines) during February 27th, 2005 (a) and February 1st, 2006 (b) in contaminated cloud conditions. Note, that τ is given only as an indicator of aerosol/cloud stability conditions during the AERONET measurements. See further details in the text.

Fig. 2.

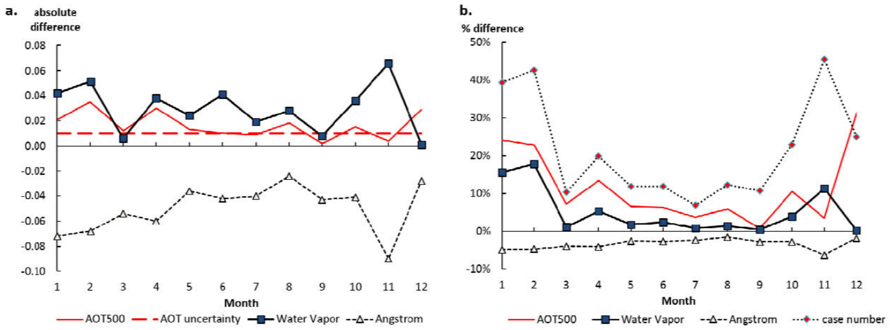


Fig.3. The absolute (a) and relative (b) difference of monthly mean standard level 2.0 data on aerosol optical thickness at 500nm (AOT500), Angstrom exponent and water vapour with the dataset after additional cloud correction. The standard uncertainty of AOT measurements is shown in Fig.2a. Relative changes in day number removed after additional cloud correction is shown in Fig2b. Moscow, 2001-2014 period. Note, that in Fig.2a the difference in water vapour is given in cm and other characteristics are dimensionless.

Fig. 3.

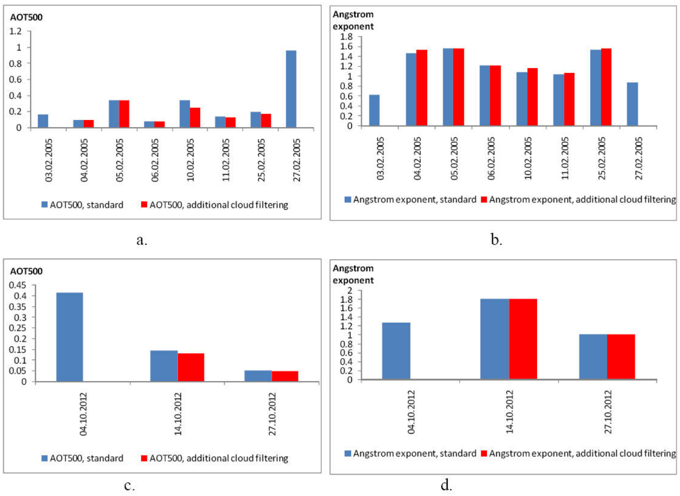


Fig.4. Standard AERONET AOT500 (a,c) and Angstrom exponent (b,d) daily means and their values after the application of the additional cloud screening for the months with large monthly mean AOT bias from the standard AERONET dataset (February, 2005(a,b), October, 2012(c,d)). Note, that the absence of the red columns (revised dataset) for several days means full elimination of the aerosol measurements after additional cloud checking. The solar disk was obscured by Cirrus, Cirrocumulus, Cirrostratus clouds during both days with SD=1, and SD=0. The halo was detected. See further description in the text.

Fig. 4.

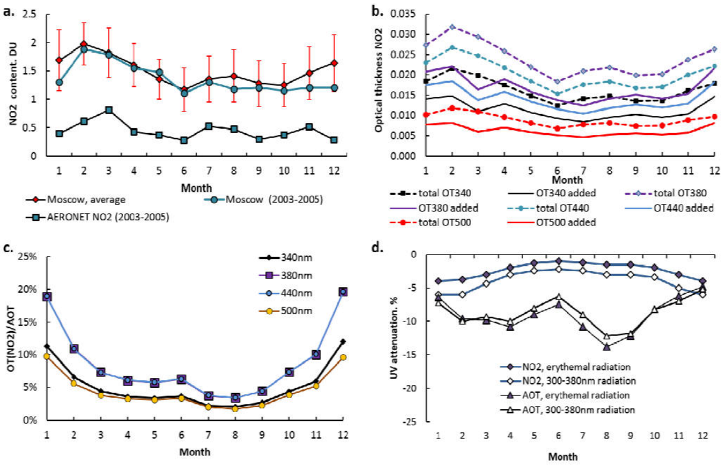


Fig.5. The seasonal distribution of mean NO_2 content over 2002-2013 and 2003-2005 periods obtained according to (Chubarova et al., 2010) and the NO_2 retrievals applied in the standard AERONET algorithm (a); monthly mean total and additional optical thickness (OT) of NO_2 at different wavelengths (b); monthly mean ratio OT(NO_2)/AOT at different wavelengths (c); relative attenuation of erythemal radiation and UV radiation 300-380nm due to NO_2 and AOT at noon time conditions according to the results of 8-stream DISORT method (d). Moscow.

Fig. 5.

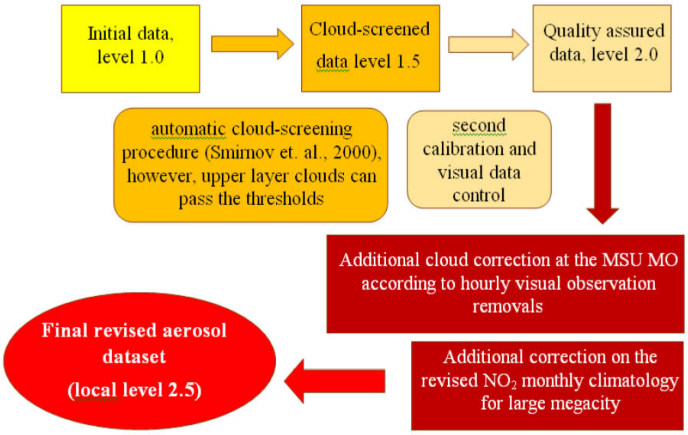


Fig.6. The scheme of the updated AERONET data proceeding with additional cloud and NO_2 correction used at the Moscow MSU MO.

Fig. 6.

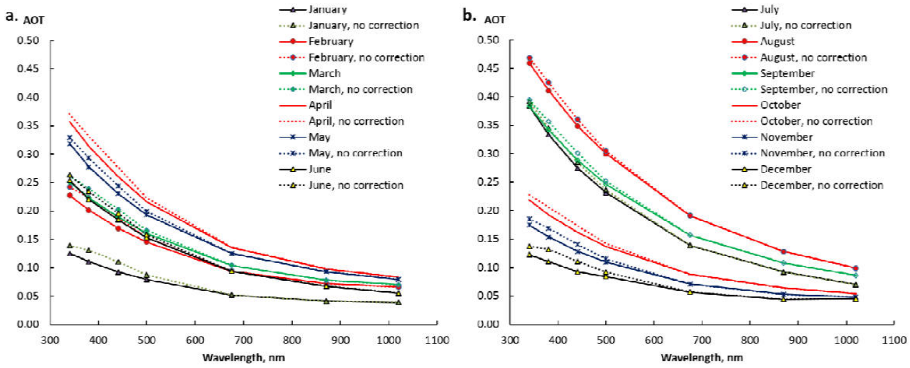


Fig.7. Spectral dependence of monthly mean AOT according to the standard and the revised AERONET dataset with the additional cloud and NO_2 correction. Moscow, 2001-2014 period.

Fig. 7.

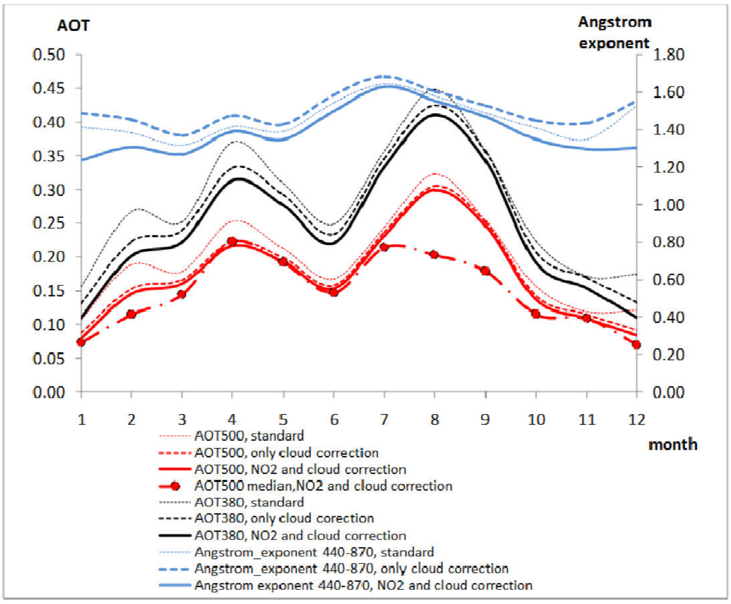


Fig.8. Seasonal variation of monthly mean aerosol optical thickness at 380 and 500 nm, median AOT at 500 nm, and Angstrom exponent according to the standard AERONET level 2.0 dataset, the data after additional cloud correction, and the final revised dataset. Note, that the additional correction of cloud and NO_2 has different sign for Angstrom exponent. Moscow, 2001-2014 period.

Fig. 8.

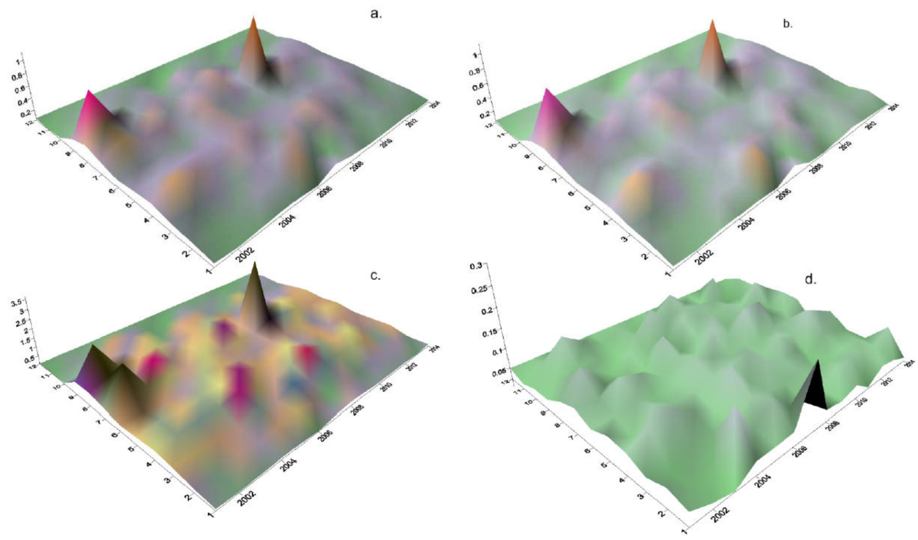


Fig.9. 3-D distribution of the revised monthly mean AOT500 (a), 50% quantile AOT500 (b), daily AOT500 maximum (c) and daily AOT500 minimum (d). Moscow.

Fig. 9.

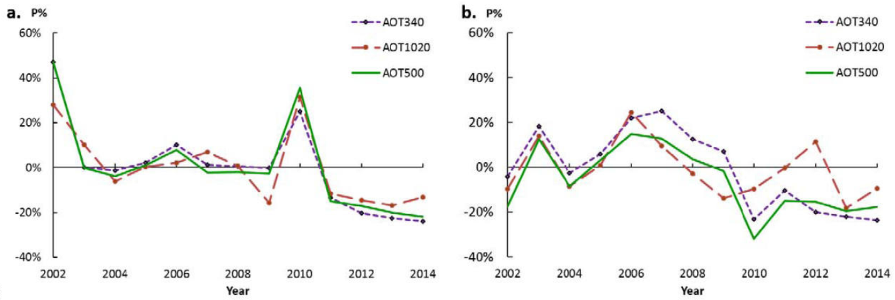
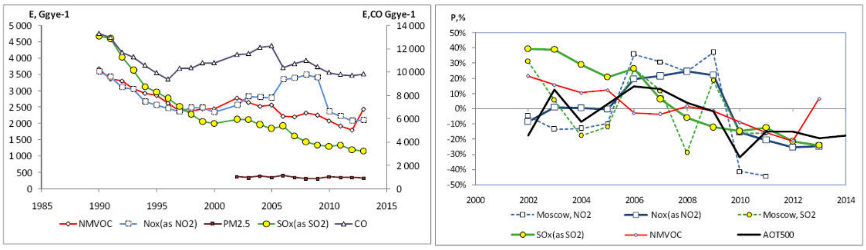


Fig.10. Interannual variations of the revised annual mean (a) and 50% quantile (b) AOT at several wavelengths. Moscow.

Comment: the annual 50% quantile AOT is estimated from monthly 50% quantile AOT values. For consistency the 2001 data were not used since the measurements have been in operation only from August

Fig. 10.



a. b.

Fig.11. Interannual variations in emissions of main aerosol precursors (SOx, NOx, NMVOC), CO, and particulate matter (PM2.5) according to WebDab - EMEP database over European part of Russia (a), relative changes in 50% quantile AOT500 and in SO_x and NO_x emissions over European part of Russia and directly over Moscow (b).

Fig. 11.

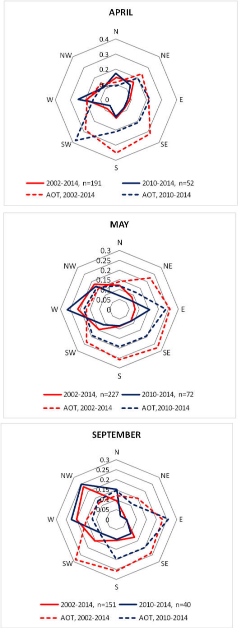


Fig.12. The wind diagram (in unit fraction) over the whole period of observations (2002-2014) and over the 2010-2014 period (solid lines) and distribution of AOT500 at different wind directions over the same periods (dashed line) (in AOT units) for the months with statistically significant negative trends. Wind directions were obtained according to the NOAA HYSPLIT model 24- hour backward trajectory analysis at 500m AGL for 12h UTC.

Fig. 12.

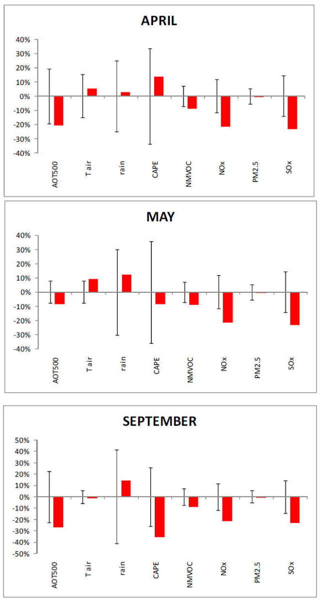


Fig.13. Error bars interval at confidence level P=95% of relatively change in monthly mean AOT500, air temperature (Tair), precipitation (rain), CAPE, as well as different emissions (NMVOC, NOx, PM2.5, SOx) from the WebDab – EMEP database and mean relative changes of these characteristics over 2010-2014. All the data were normalized against their mean values over the whole period of observation. For homogeneity reasons we do not include September 2002 in the analysis due to the large effect of smoke aerosol from forest fires, and April 2012, since it was a problem with sun photometer records. Note, that the emissions data are available only up to 2013.

Fig. 13.