

Dear Referee #2

We thank for your useful comments, corrections and suggestions. We have corrected the text and add a new text and figures according your comments. We hope that our revisions that we have made according your suggestions improve the text of the paper.

Referee #2 Major remarks:

1. The structure of the paper is not good. Seasonal features are described several times, there are many other duplications in the text. I would recommend to re-arrange the text combining the data description, methods, and the results in the separate sections. In addition, all the data, which describe seasonal changes, should be considered together and then the authors could add the material about the interesting urban experiment and its results.

G. Milinevsky. We re-arrange the text, correct and remove duplications in the text according your suggestions to improve presentation of the results and discuss aerosol behavior adding the figures. The text describing structure of the paper is given in Introduction:

"In Section 2 we describe the instrumentation and measurement in sites/regions. The results of seasonal variation studies of aerosol parameters are given in Section 3. In Section 4 we discuss the results for individual sites and regions with conclusion remarks in Section 5."

2. I would recommend to add the POLDER measurements as a base for the analysis over the considered area or at least over the urban centers (not only over few of them). It would be important to see the comparison of the POLDER AOT retrievals with ground-based measurements in a special method description section. After this analysis, if the validation shows good results, the authors could describe the ground-based and satellite retrievals together (for example, considering their seasonal changes).

GM. We add the POLDER measurement data for the analysis over two representative urban and industrial centers (Kharkov, Dnipropetrovsk) and include Angstrom exponent (670-865 nm) values for all regions (see new Fig. 4N). To compare with AERONET data, we add Kyiv and Minsk POLDER data. The comparison of the POLDER AOT retrievals with ground-based measurements has been discussed in details in our previous paper in the ACP, 2013 (doi:10.5194/acp-13-6587-2013), mentioned by Referee. We included partly the results of that comparison in the text and provided POLDER AOD (865 nm) data in figures describing Kyiv and Minsk fine mode AOD (870 nm) for comparison (new Fig. 2N (c), (e)).

3. I agree with the first reviewer that the analysis of the data is not enough, it includes mainly the description of the Figures. This should be significantly improved.

GM. We analyse obtained results in order to improve understanding of the specific aerosol behavior in studied region. Discussion included more aerosol parameters, particularly spectral SSA and refractive index analysis for Kyiv site, fine and coarse modes of particles volume concentration separate consideration for Kyiv and Minsk, analysis of AOD and Angstrom exponent from Kyiv AERONET site, POLDER/PARASOL data analysis was extended to more number of cities etc., and the results illustrated by figures.

4. I am not a specialist in English but even I can see a lot of bad English style expressions, the absence of articles in many cases and even mistakes in grammar. This should be also improved.

GM. We correct the English trying to avoid bad English style expressions and checked articles usage and grammar.

Referee #2 Minor remarks:

1. In the Introduction it should be clearly stated what are the principally new ideas and intent of the authors in this study. What is the difference with their previous paper in the ACP, 2013. I would recommend, in addition, to give more attention to the results of POLDER data.

GM. We have added sentences describing the principally new ideas, intent of our study, and the difference with our previous paper in the ACP, 2013. We pay more attention to POLDER data (see answer to Major remark 2 above).

New sentences:

"In continuation of our previous study, we concentrated here on analysis of seasonal aerosol properties and dynamics over the urban industrial areas of Ukraine based on AERONET sites and POLDER/PARASOL satellite imaging spectroradiometer data. Unlike the previous paper (Bovchaliuk et al., 2013) we analyse the AOD variation for total AOD and separately for fine and coarse modes AOD and volume concentration, spectral SSA and refractive index for AERONET sites in Ukraine and expand analysis of POLDER data to a larger number of urban areas. The purpose of that analysis is to determine what kind of aerosol dominates in specific season to study the possible aerosol sources."

2. Since the authors stress on studying the urban effects, the additional references on aerosol urban effects might be added, the results should be compared and analyzed in the text (see, for example, Zavadzka et al. *Atm. Env.* 2012, Chubarova et al. *AMT*, 2011, others).

GM. We add new references and discuss the results of aerosol properties seasonal variability in the Eastern Europe given in these and some other papers. However we have not purpose to study directly the urban effects, rather to evaluate the average season behavior over several urban/industrial areas in Ukraine. To avoid misunderstanding we have corrected the title to better corresponding to the paper content and we concentrate on analysis of aerosol seasonal behavior over urban/industrial areas of Ukraine. New title is: "Aerosol seasonal variations over urban/industrial regions in Ukraine according to AERONET and POLDER measurements"

5. The description of aerosol ground-based measurements can be shortened. When describing Angstrom exponent the authors should mention the conditions on Junge distribution which is not often observed in reality and is necessary for reliable evaluating of Angstrom exponent. Please, make a special section on the data and method description in which all different methods are described. The sources of the aerosol can be described separately. The authors should be very cautious and describe thoroughly all specific conditions when they use the level 1.5 data. I would recommend to describe the reasons in each time since the absence of level 2 data could mean the poor final calibration of the data. But I understand that utilizing level 1.5 in single scattering albedo is sometime necessary. The authors sometimes tried to describe these aspects but irregularly and in different places of the text. Please, combine this analysis in one place.

GM. We provide a special Section 2: "Description of instrumentation and measurements sites/region" where the data and methods are described. We combine in one place the description of the data that analyzed. We understand that we should be careful analyzing the level 1.5 data

we should be careful. However we need to use level 1.5 data for analysis of single scattering albedo and refractive index because level 2.0 data for these parameters are absent. In rest cases Level 2.0 data are used. We present the level of used data, describe their errors.

On Angstrom exponent description – see our next comment.

6. Page 10736 lines 1-5. Too much information and duplications in the description of Angstrom exponent.

GM. We have shortened the description of Angstrom exponent and refer to (Kokhanovsky, 2008) [Kokhanovsky A.A. Aerosol optics. Light absorption and scattering by particles in the atmosphere. Springer and Praxis Publishing, 2008. – 146 p., Chapter 2.2, P.17 – 23.], where "the relationship between Angstrom exponent and parameters of the size distribution can be derived for any type of size distribution" including Junge distribution. So, we include the text:

“The Angstrom exponent is the parameter that is related to the aerosol particles size distribution and depends on the effective radius of particles that allows dividing aerosol by fraction: small Angstrom exponent is related to large particles and large Angstrom exponent correspond to fine mode particles. But also it is influenced by the coefficient of variance of the size distribution (Schuster et al., 2006, Kokhanovsky, 2008.”

7. Page 10736 line 10-13. Dublication.

GM. We have removed this duplication.

8. Page 10736 line 16-17 As far as I know the uncertainty of AOT latest measurements is about 0.01 in visible and in near-infrared channels and 0.02 in UV. Please, check and make a reference.

GM. Corrected, and reference to (Holben et al., 1998) has been added.

9. Page 10737 line 7-8. Another value for the AOT uncertainties?

GM. Removed.

10. Page 10737 line 17. POLDER data should be compared with the AERONET. The uncertainties of POLDER measurements should be stated.

GM. We provide the new text on comparison POLDER and AERONET data and discuss them together . New text included:

"The retrieving accuracy of aerosol parameter has been investigated over ocean (Herman et al., 2005; Goloub et al., 1999) and land. The correlation with AERONET sun photometers is 0.77–0.95 depending on the location of ground-based sites (Bréon et al., 2011; Fan et al., 2008; Bovchaliuk et al., 2013). The standard cloud screening algorithm has been used for the processing of POLDER measurements over land surfaces (Bréon and Colzy, 1999). In our investigation we used cloud free pixels. More details on methods of aerosol properties analysis using POLDER instruments data are given in our previous paper (Bovchaliuk et al., 2013).

"The detailed comparison of AERONET sun photometer data and POLDER instrument data is given for all East-European AERONET sites in Bovchaliuk et al. (2013). The comparison of POLDER AOD (865 nm) and AOD (870 nm) for fine mode particles ($r < 0.3 \mu\text{m}$) measured in Moscow, Minsk, Belsk, Kyiv, Moldova and Sevastopol AERONET sites shows good accordance

of data. In general, the correlation coefficients AOD (865 nm)/AOD (870 nm) are in a range of 0.76–0.93 except for Sevastopol site (0.63) and the standard deviations in the AERONET/POLDER comparisons are less than 0.02 for AOD values between 0.01 and 0.25 (Bovchaliuk et al., 2013)."

11. Page 10737, line 27. Fine mode aerosol can be also produced by biomass burning for example. There are a lot of publications on this account. This should be clarified in the text. The procedure of satellite data cloud screening is very important for evaluating good aerosol data. This should be described and analyzed in the text.

GM. We clarify in the text that fine mode aerosol can also be produced by biomass burning events and provide references. On the procedure of satellite data cloud screening see our comment on your minor remark 10 above.

2. Page 10738, section 4. In the analysis of seasonal changes I would recommend to combine POLDER and AERONET data.

GM. We combine POLDER and AERONET data in analysis of seasonal changes (see e.g. Fig. 2N) where it is possible.

13. Page 10738, section 4. AOT at 440 is significantly contaminated by NO₂ in the urban areas. The correction in AERONET algorithm is not full (see, for example, the discussion in Chubarova et al., 2011). The wavelength 440nm is near the maximum of the NO₂ absorption. So I would recommend to use another wavelength in the analysis, the same or close to the POLDER wavelengths, for example. This would make the analysis of the satellite and ground-based AOT data more clear and comparable.

GM. According your advice we use one more wavelength 870 nm in the analysis that close to the POLDER wavelength 865 nm and we discuss the AERONET and POLDER data together where it is possible (see e.g. Fig. 2N). We also include the text (see below) on description of NO₂ impact on AERONET data with appropriate references.

"However, it should be mentioned that the 440 nm spectral channel corresponds to the NO₂ molecular absorption band (Burrows et al., 1998). The NO₂ impact is considered in the AERONET parameters retrieval algorithm using the satellite data from OMI, SCIAMACHI, GOME instruments (see AERONET Version 2 Direct Sun Algorithm, <http://aeronet.gsfc.nasa.gov/>). But the recent satellite NO₂ data accuracy is relatively small (Boersma et al., 2004; Boersma et al., 2011). Since the NO₂ concentration in the troposphere is determined mainly by anthropogenic sources like fossil fuel combustion (Ehhalt et al., 2001) than, in a case of urban/industrial regions, we cannot exclude probable influence of NO₂ on measured AOD (440 nm) values. To estimate this influence we can use the optical remote sensing data received for urban territories in USA (Alexandrov et al., 2002a and 2002b). Though the NO₂ atmospheric extinction yearly mean values for ex. over New York (measurements at 440 nm from September 1995 till November 1996) were below 5 Dobson Units (DU), but in some days the extinction value reached 15 DU. Considering the absorption cross-sections of NO₂ (Burrows et al., 1998), the corresponding extinction optical thickness will be 0.07 for 5 DU and 0.2 for 15 DU case. This value exceeds significantly the errors of AOD measurements by AERONET sun photometers in 440 nm spectral band (Holben et al., 1998). According Rublev et al., (2004) the NO₂ concentration at 0.5 km altitude in Moscow rush time hours corresponds to extinction about 13.7 DU and AOD (440 nm) value about 0.18. Therefore, NO₂ in urban/industrial regions like considered in our paper could impact AERONET AOD (440 nm) data. From set of spectral channels of AERONET sun photometer CE318-2 the channel 870 nm is free from NO₂ influence. Moreover this spectral band corresponds to main spectral channel of

POLDER instrument. However, the relative errors of AOD (870 nm) are in 2 – 3 times greater than for AOD (440nm). Therefore, it looks appropriate to use the data from both spectral channels for evaluation of seasonal variations of aerosol optical properties in this paper."

14. In the analysis of seasonal changes, please, add the comparisons with the available climatologies over this area (see, for example, Kinne et al., "A new global aerosol climatology for climate studies", 2013, Chubarova et al. , 2009).

GM. We use for analysis the climatology in East European region discussed in supplement to our previous paper (Bovchaliuk et al., 2013) where wind fields over Eastern Europe presented for the period 2001–2011 (see acp-13-6587-2013-supplement.pdf) and compare them with the climatologies in the mentioned papers given in comment 14.

15. Page 10739 line 3. Missed reference to Fig.1 a?.

GM. We have corrected the text and add reference to new Fig 2N (a).

16. Page 10739, line 7. The use of Figures should be in a sequence form. Please, change numeration of Fig.4. Figure 4 does not prove the statement of prevailing the coarse mode aerosol in the fall and winter. Please, check. The prevalence of fine mode aerosol might not be the urban effect. The authors should make the analysis in the non-urban conditions in the region for this statement as well.

GM. We re-arranged all figures and provide them in a correct sequence form with text corrections. We have removed Fig. 4 and correspondent sentences from the text.

17. Page 10739, line 13. You mentioned prevailing the coarse mode particles from January to May 2012. Please, analyze this effect in more details, use the vertical profiles and any additional information (for example, available synoptic meteorological data to remove possible cirrus contamination) to prove your statement.

GM. We correct that part of the text and include the POLDER data analysis. Unfortunately the vertical profile data are not available in our region. The first lidar campaign for these measurements is planned this summer.

18. Page 10739 line 22. Please, put this part in a separate section with more details on the meteorological conditions with the description of the pollution sources, etc. Please, make a comparison with the other results. I would recommend to add much more material and discussions in this section.

GM. We have removed the text related Kyiv/Kyiv-AO analysis due to insufficient amount of measurements in the AERONET Kyiv-AO site to make strict conclusions. We will continue this analysis together with Microtops II sun photometer measurements after their processing in the separate paper.

19. Page 10741, line 1. There is no strong seasonal dependence in real part of refractive index. Please check.

GM. We examine the seasonal dependence of refractive index using the results presented in new Fig. 6N, see below.

20. Page 10741, line 4. Please, make a table with the statistics of cases and mean, median, st deviation, etc. AOT values for different sites.

GM. We have changed this part of the paper and consider seasonal behavior of SSA and complex refractive index using data of all four spectral channels for Kyiv site only. The results are illustrated in Fig. 6N.

21. Page 10741, line 8. It is strange to see biomass burning as a source of aerosol in winter. Please clarify.

GM. We revise the text.

22. Page 10741. Section 5 “Comparison: : : Please combine this part with the previous seasonal changes analysis.

GM. We combine this part of text with the seasonal changes analysis in the Section 4 (Discussion).

23. Page 10743. Section 6. I would recommend to make the analysis only on POLDER data over the whole territory with adding winter data from AERONET. Otherwise the title of the section is not right and there is too small material. At least the data on many Ukranian and Belarus urban sites should be added. Please, re-arrange the structure.

GM. We re-arrange the structure of the text according your advice. The structure of the paper is given in our answer on Referee #2 Major remark 1.

24. Page 10744, line 9. That should be studied in more details using the profile information or other data.

GM. We expand the discussion of the Saharan dust effect relying on our results of aerosol transfer study in the East European region using the back-trajectory and cluster analysis methods. We consider also additional results published on that issue (e.g. Pavese et al., 2012).

New References

- Alexandrov, M. D., Lacis, A. A., Carlson, B. E., and Cairns, B.: Remote Sensing of Atmospheric Aerosols and Trace Gases by Means of Multifilter Rotating Shadowband Radiometer. Part I: Retrieval Algorithm, *Journal of the Atmospheric Sciences*, 59, 524–543, 2002a.
- Alexandrov, M. D., Lacis, A. A., Carlson, B. E., Cairns, B.: Remote Sensing of Atmospheric Aerosols and Trace Gases by Means of Multifilter Rotating Shadowband Radiometer. Part II: Climatological Applications, *Journal of the Atmospheric Sciences*, 59, 544–566, 2002b.
- Amiridis, V., Giannakaki, E., Balis, D. S., Gerasopoulos, E., Pytharoulis, I., Zanis, P., Kazadzis, S., Melas, D., and Zerefos, C.: Smoke injection heights from agricultural burning in Eastern Europe as seen by CALIPSO, *Atmos. Chem. Phys.*, 10, 11567–11576, doi:10.5194/acp-10-11567-2010, 2010.
- Boersma, K. F., Eskes, H. J., and Brinksma E. J.: Error analysis for tropospheric NO₂ retrieval from space, *J. Geophys. Res.*, 109, D04311, doi:10.1029/2003JD003962, 2004.
- Boersma, K. F., Eskes, H. J., Dirksen, R. J., Vander, A. R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitao, J., Richter, A., Zhou Y., and Brunner D.: An improved tropospheric NO₂ column retrieval algorithm for the Ozone Monitoring Instrument, *Atmos. Meas. Tech.*, 4, 1905–1928, 2011.

- Bréon, F.-M., Colzy, S.: Cloud Detection from the Spaceborne POLDER Instrument and Validation against Surface Synoptic Observations, *J. Appl. Meteor.*, 38, 777–785, doi:10.1175/1520-0450, 1999.
- Bréon, F.-M., Vermeulen, A., and Descloitres, J.: An evaluation of satellite aerosol products against sunphotometer measurements, *Remote Sens. Environ.*, 115, 3102–3111, doi:10.1016/j.rse.2011.06.017, 2011.
- Burrows, J. P., Dehn, A., Deters, B., Himmelmann, S., Richter, A., Voigt, S., and Orphal, J.: Atmospheric remote-sensing reference data from GOME: Part 1. Temperature-dependent absorption cross-sections of NO₂ in the 231 – 794 nm range, *J. Quant. Spectrosc. Radiat. Transfer*, 60, 1025 – 1031, 1998.
- Ehhalt, D., Prather, M., Dentener, F., Derwent, R., Dlugokencky, E., Holland, E., Isaksen, I., Katima, J., Kirchhoff, V., Matson, P., Midgley, P., and Wang, M., In: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change* [Houghton, J. T., Ding, Y., Griggs, D.J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson C.A. (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881 pp.
- Goloub, P., Tanré, D., Deuzé, J. L., Herman, M., Marchand, A., Bréon, F.-M.: Validation of the first algorithm applied for deriving the aerosol properties over the ocean using the POLDER/ADEOS measurements, *IEEE Trans. Geosci. Remote Sens.*, 37, 1586–1596, doi:10.1109/36.763270, 1999.
- Herman, M., Deuzé, J. L., Marchand, A., Roger, B., and Lallart, P.: Aerosol remote sensing from POLDER/ADEOS over the ocean: Improved retrieval using a nonspherical particle model, *J. Geophys. Res.*, 110, D10S02, doi:10.1029/2004JD004798, 2005.
- Pavese, G., Calvello, M., Esposito, F., Leone, L., and Restieri, R.: Effects of Saharan Dust Advection on Atmospheric Aerosol Properties in the West-Mediterranean Area, *Advances in Meteorology*, 2012, Article ID 730579, doi:10.1155/2012/730579, 2012.
- Rublev, A. N., Chubarova, N. E., Trotsenko, A. N., Gorchakov, G. I.: Determination of NO₂ Column Amounts from AERONET Data, *Izvestiya, Atmospheric and Oceanic Physics*, 40, 54–67, 2004.
- Uscka-Kowalkowska, J.: An analysis of the extinction of direct solar radiation on Mt. Kasprowy Wierch, Poland, *Atmos. Res.*, 134, 175–185, 2013.
- Zawadzka, O., Markowicz, K.M., Pietruczuk, A., Zielinski, T., Jaroslowski, J.: Impact of urban pollution emitted in Warsaw on aerosol properties, *Atmos. Environ.*, 69, 15–28, <http://dx.doi.org/10.1016/j.atmosenv.2012.11.065>, 2013.

Figures added, corrected:

To Referee #2 Major remark 2.

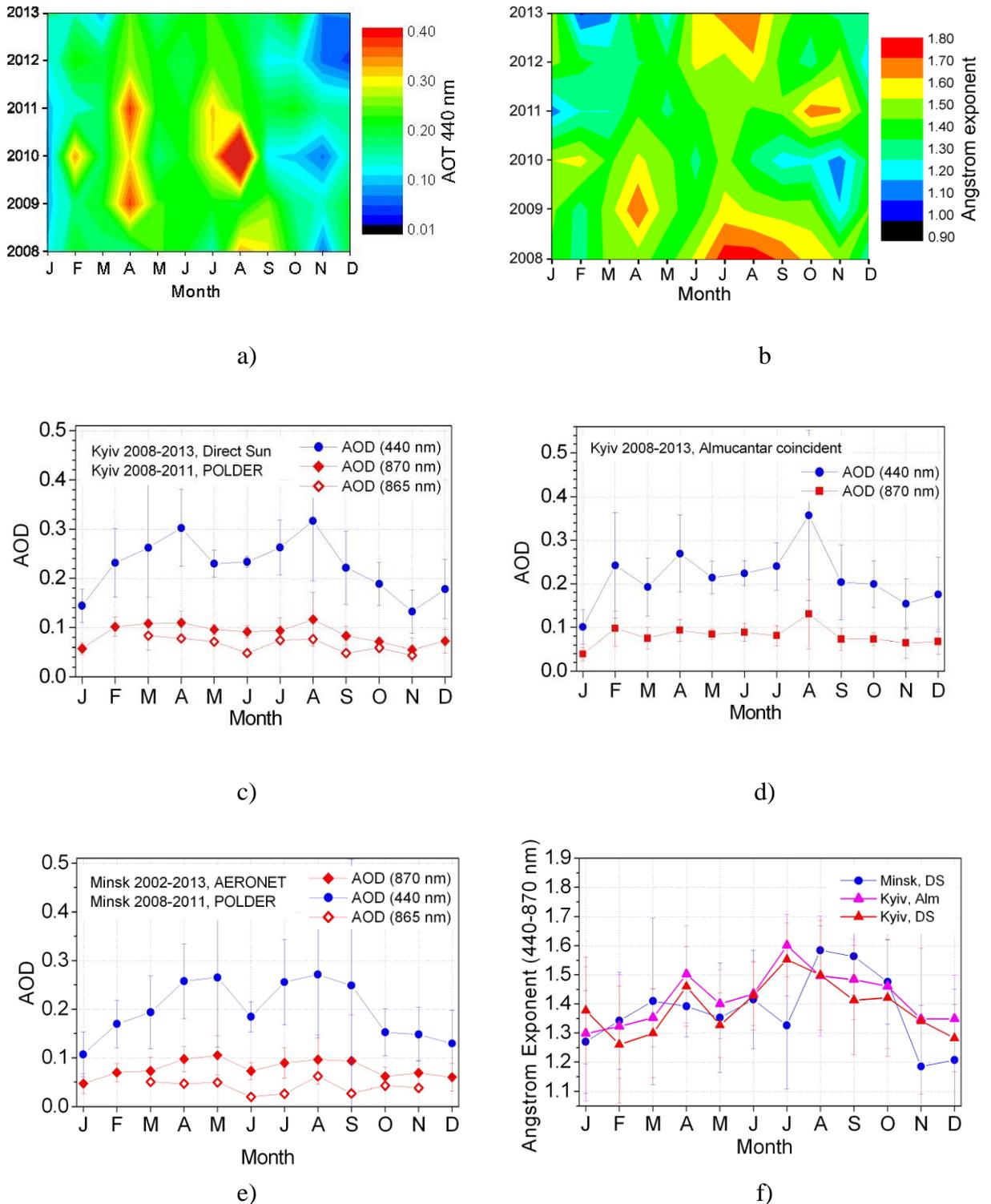


Fig. 2N. (a), (b) The features of AOD (440 nm) and Angstrom exponent (440–870 nm) season variations for Kyiv site from year to year, (c)–(e) mean values seasonal changes of AOD (440 nm) and AOD (870 nm) by AERONET data and AOD (865 nm) by POLDER data for Kyiv and Minsk, (f) the set of Angstrom exponent (440–870 nm) values. For all data STD is shown except POLDER data to avoid overloading. Months are shown by their first letters in the order January–December.

To Referee #2 Major remark 2.

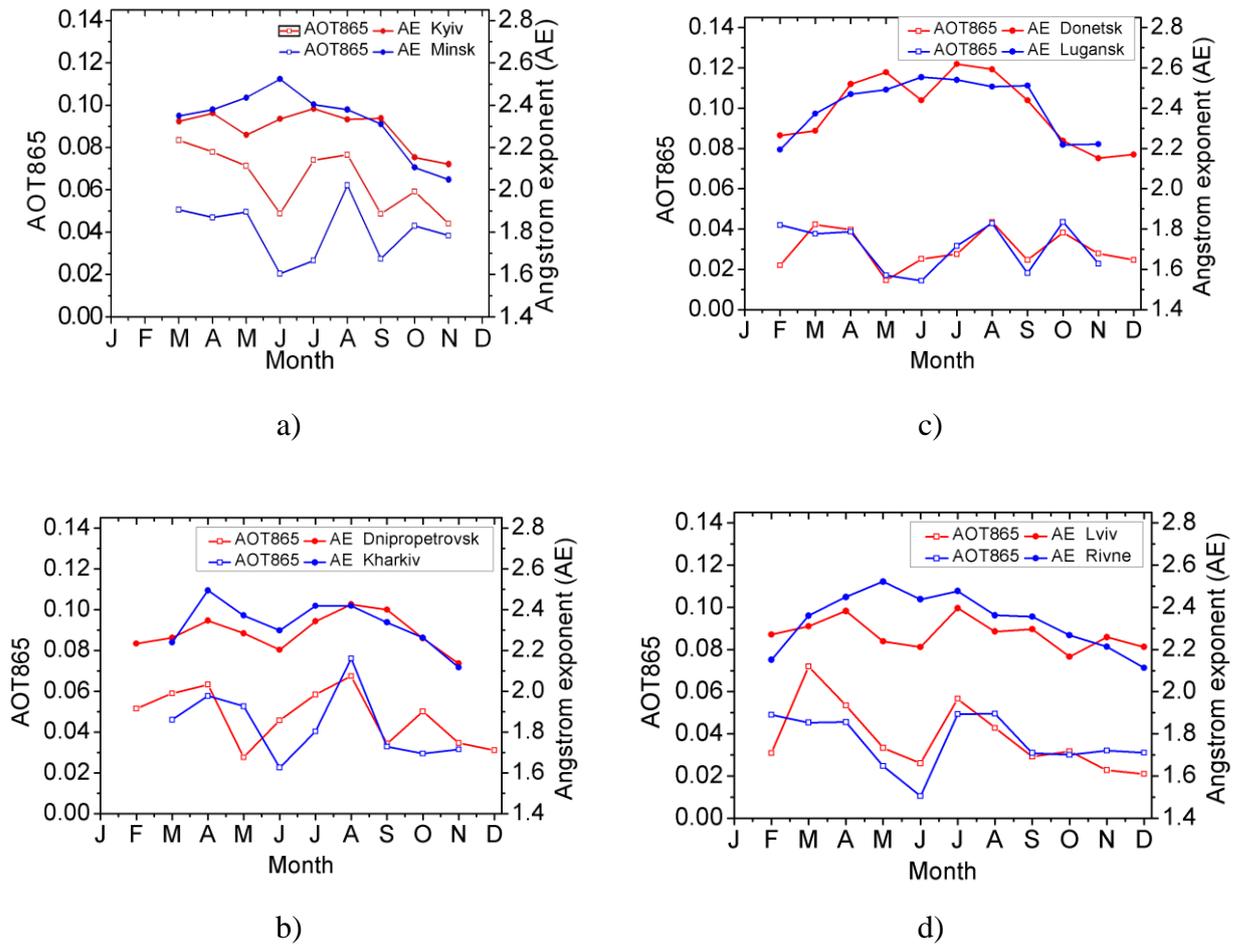
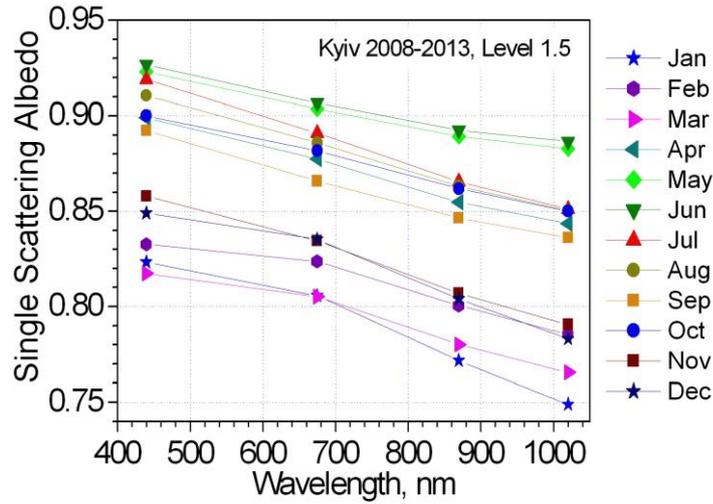
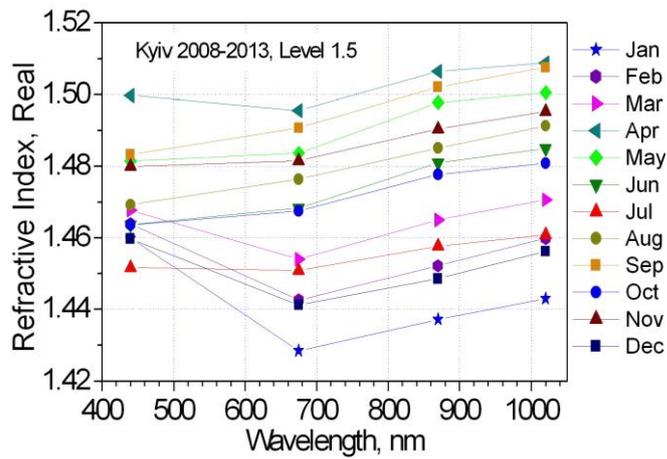


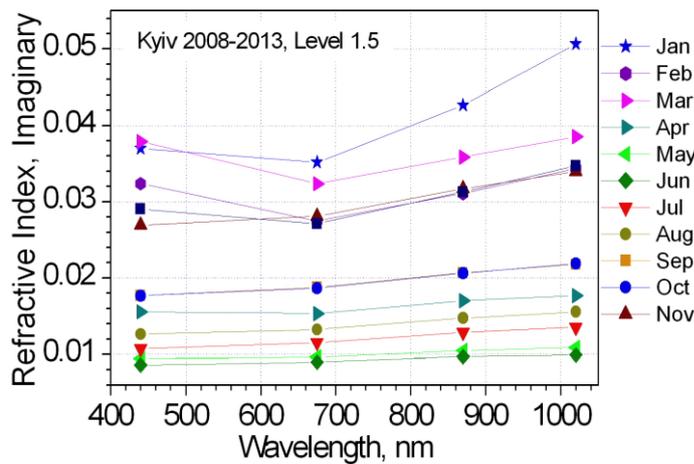
Fig. 4N. Seasonal variations of AOD (865 nm) values and Angstrom exponent (670–875 nm) values averaged over 2008–2011 in the atmosphere over the Kyiv, Minsk, Donetsk, Lugansk, Dnipropetrovsk, Kharkov, Lviv and Rivne urban/industrial areas based on POLDER data. Months are shown by their first letters in the order January–December.



a)



b)



c)

Fig. 6N. (a) Single scattering albedo and (b), (c) refractive index spectral dependence by the Kyiv AERONET site data.