Authors response to Referee #1

We thank the Referee's helpful comments on our manuscript. Many sentences of the manuscript have been carefully rewritten and reorganized to make the statements clear. Authors' responses are highlighted in blue. The notation is as follows: P1 L12 means page 1 line 12

5 means page 1, line 12.

The paper by Galytska et al. analyses in details the influence of the 2010 eastern Europe wildfires on the atmosphere of Ukraine. It combines both retro-trajectories modelling and different sets of remote-sensing observations. This work is interesting and the analysis thorough but the manuscript in its present form need improvements.

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We thank the Referee for the positive comment and we agree that the manuscript needed improvements.

The paper is very long and should be shortened and better focused (see specific comments below).

15 We are following the Referee's suggestion and we have reworked and reorganized the manuscript to highlight its novelty; we have reduced its length by restructuring methodological paragraphs and better concentrating on main messages. We have optimized the titles of Section 2 and 3 to better address the sections subject.

For instance, the introduction written as it looks like a detailed summary of previous works on the subject rather than a presentation of scientific objectives. The authors should clearly state in the introduction the adding contribution of their study compared to previous works.

We agree to the Referee and we have reworked the Introduction entirely. The rewritten Introduction is structured as follows:

- We highlight that wildfires are an important global source of aerosol.
- 25 We explain importance of wildfires events during summer 2010. We address to previous studies (in chronological order) devoted to aerosol dynamics over the European territory of Russia (ETR) and Eastern Europe. Also, we highlight the papers of aerosol research performed for the territory of Ukraine.
 - We explain the contribution of our research in comparison with previously mentioned in the Introduction papers. We have made suggested changes on P2 L2-P4 L11.
- 30 Also, the analyse should include POLDER data that are well designed to track fine aerosols.

We thank the Referee for having drew our attention on this. This is a good point. Though, from our point of view, POLDER data are useful while estimating the content of fine aerosol, with particle size of approx. 0.3 µm. If the purpose of our manuscript would be estimating the physical and chemical properties of aerosol in the wildfires and its evolution during transport, or estimating the peculiarities of the formation of particle in the wildfires, then these data certainly would be involved. But the

- 35 estimating the peculiarities of the formation of particle in the wildfires, then these data certainly would be involved. But the purpose of our research is to analyze where, when and how the aerosol component of the wildfires was transported to the investigated region. In addition, inclusion of POLDER data would increase the size of the manuscript. Therefore, it seems that for such a synergetic data analysis from various satellite and terrestrial measurements it is necessary to perform a separate research.
- 40 Throughout the paper, the contribution of biomass burnings on the aerosol load over Ukraine is often discussed in a qualitative way. A more quantitative analysis could improve the quality of the paper : what is the ratio between transported biomass burning aerosols and locally-emitted particles during polluted days ? Is this ratio constant or variable according to meteorological situations ?
- 45 Thank you for having raised this issue. The ratio between transported biomass burning aerosols and locally-emitted particles depends significantly on various factors, in particular weather conditions. AERONET and satellite measurements do not enable

to identify locally-emitted aerosol in the atmospheric column. In situ measurements of aerosol properties, sources etc. could be used to resolve this issue. Although, we did not apply in situ data in our research. Thus, in our study we only estimated the impact of wildfires over Kyiv by comparing the aerosol properties for the dates when aerosol from wildfires was observed to those dates when aerosol from wildfires was absent. We address to the latter issue on P15 L11-17 as follows:

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'In our study we estimated the impact of wildfires over Kyiv by comparing the aerosol properties for the dates when aerosol from wildfires was observed to those dates when aerosol from wildfires was absent. We identified respective dates from the analysis of air masses transport to Kyiv, as described above. We compared aerosol properties as averaged values over different time periods: 1) when the number of fires and their brightness temperature were low, June 1-26 (40–65°N and 10–60°E, see

10 Fig.2), 2) when the numbers of fires significantly increased, July 18-August 14, and 3) when the number of fires and their brightness temperature remained high and the highest AOD values were observed, August 15-17'.

Specific comments and technical corrections :

15 P2: The largest numbers of these fires[...] and Kazakhstan : please add a reference

We have added the reference of Barnaba et al., 2011. Thank you.

P2 : The papers provided results ->the papers providing results

20

Thank you for the suggestion. Finally, we have withdrawn the entire sentence in the reworked version of Introduction.

P3 : in the UV band Holben et al. (1998) : add parenthesis to the citation

25 Added parenthesis (Holben et al., 1998). Thank you. P4 L24.

Section 2 : Please add a map highlighting the localization of Ukraine and the observational sites used in this study.

Thank you, we have inserted the map with the AERONET observational sites in Eastern Europe and Ukraine, which were used in our study. Please, see reworked manuscript Fig.1 on P5 or below.

P4 MODIS data : Why corrected MODIS AOD has much lower data than the collection 051 AOD for this specific region and period ?

35 Thank you for raising this issue. MODIS Scientific Data Set (SDS) orrected_Optical_Depth_Land contains AOD with the highest accuracy (Quality Assurance Confidence flag (QAC) = 3 over land). In our research we have used AOD with QAC = 2 and 1 to increase the amount of data. That is why we have further compared AOD between MODIS and AERONET.

Section 3.1 : This section should include meteorological data mentioned in section 2.4.

40

We have reworded the titles of Sect. 2 and 3.

Section 2 has been renamed from "Data sources" to "Methods and data sources". We aimed to collect all relevant information about methods used in our study in a respective section. Therein, Section 2.4 has also been renamed from "Meteorological data and the means of study of air masses transport in the investigated region" to "Weather conditions and transport of air masses"

45 now even more precisely states the purpose of the information provided there.We have renamed Section 3 from "Methodology and results" to "Results and Discussion". After careful consideration of your suggestion, we retained Section 2.4 in the same position.

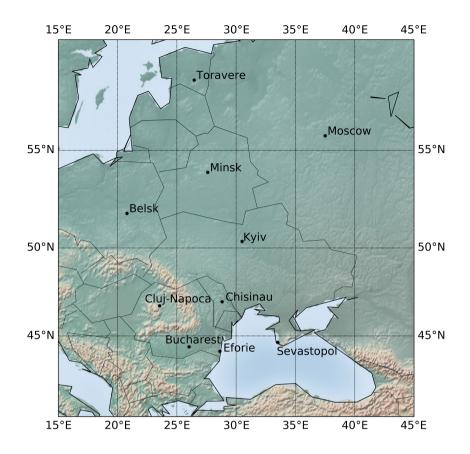


Figure 1. AERONET observational sites in Eastern Europe and Ukraine used in this study.

Table 2 : Milinevsky did not use AERONET data for the year 2014.

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We thank the Referee for having drew our attention on this issue. We have updated Table 2 with AOD values at 500 nm to make our analysis of AERONET data consistent and avoid switching between 440 nm and 500nm. In the reworked version of the manuscript we address to this issue as follows: 'In Table 2 we show a prolongation of the data record of Milinevsky et al. (2014, 2008-2013) for the Kyiv AERONET site by three more years up to the end of 2016. Even in this extended record the most significant aerosol pollution was observed in August 2010. This event is related, in particular to wildfires in the ETR and Eastern Europe'. Please, see P12 L15-18.

10 P7 : The highest content of aerosols over Kyiv was observed every year in spring (April-May) and late summer (July-August) : Could you explain why maximum pollution occur during these periods ? Is this the result of specific meteorological regimes ? Local emissions ? Absence of washout ?

We thank the Referee for his/her comment. We have addressed this issue on P12 L10-14 as follows: 'Between 2003 and 2014 ground-based and satellite observations showed the highest aerosol content over Kyiv every year in spring (April–May) and late summer (July–August; Bovchaliuk et al., 2013; Milinevsky et al., 2014). According to both studies, the observed spring peak in aerosol content is associated with transport of the Saharan dust across Eastern Europe, transport of sea salt aerosol

5 from the Black Sea and the Sea of Azov, and occasionally occurring agricultural fires. The summer peak results from wildfires, soil dust aerosol due to harvesting activity, and transport of Saharan dust'.

P8 : Coarse aerosol particles were mainly of local origin : which origin ? Please be more specific.

10 We have been more specific and added the information as follows: 'Coarse aerosol was mainly of local origin, such as city transport and heavy industry, while the fine mode particle likely was brought by air currents from Europe'. Also, we have moved this piece of text to the Supplement (P2) as suggested by other Referees.

Section 3.2: This section could be shortened. The main conclusions should be highlighted to improve the clarity of this section.

15

We thank the Referee for highlighting this issue. We reworked this section, shortened it by moving entire analysis of air transport during June 1-August 31 to the Supplement.

P11 : Could you explain the influence of the multiple scattering effect on the retrieval of the SSA. This part is not clear.

20

We thank the Referee for this comment. We have reconsidered our statement regarding the effect of multiple scattering on higher SSA values over Kyiv in comparison with Moscow. Also, we believe that this issue requires further thorough analysis, which can be in focus of further studies. In the rewritten version of our manuscript we have explained increased SSA values during August 15-17, 2010 by increased size of particles, which caused an increase of atmospheric column reflectance. We have addressed this issue on P15 L35-P16 L1 as follows: 'According to Eck et al. (2009), larger SSA values can be explained

by an increased particle size which increases the total reflectance of atmospheric column'.

P15 : AE is often larger than 1 for fine biomass burning aerosols so the AOD bias between MODIS and CALIOP is potentially larger than 2-4%.

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In our research we have compared AODs from CALIOP at 532 nm and MODIS/Aqua at 550 nm without corrections for the spectral differences. This could cause AOD bias between MODIS and CALIOP, larger than 2-4 %. We have addressed this issue in P8 L9-12 as follows: 'We did not apply any correction for potential spectral 10 differences while comparing CALIOP AOD 532 nm and MODIS/Aqua AOD 550 nm. It yields to an estimated systematic bias in our AOD comparison of approximately 2–6 % in the AE range between 0.5 to 1.8 (see Fig. 5b) and can be neglected in our cases, following Kittaka et al. (2011)'.

Figure 9 : Why 3 types of aerosol vertical profiles can be distinguished ? Is the result of different transport pathways ? Emissions processes ? Specific local meteorology ? Orography ?

40 We have analyzed 58 aerosol profiles for the period August 7-18, 2010. The main criterion for the definition of 3 types of profiles was the peculiarities of aerosol vertical distribution. We have addressed this issues on P19 L6-16 of reworked manuscript as follows:

'We also analyzed vertical distributions of aerosol extinction at 532 nm. We compiled analyzed profiles for the cases with high AOD 532 nm. This selection leads to 58 profiles for 11 tracks for the period August 7–18, 2010. The corresponding AOD

45 532 nm ranged from 0.44 (on August 13 11:00:06) to 2.93 (on August 18 00:08:26). Among selected profiles, 37 on 7 ground tracks were nocturnal, and 21 profiles on 4 tracks were measured during daytime. The profiles reveal that aerosol ranged from about 40 m to mostly 5 km altitude. The vertical distributions varied significantly during both day- and nighttime. According to the peculiarities of aerosol vertical distribution, we identified three types of profiles. 1) Type 1 consists of profiles showing at least a single aerosol layer of some hundred meters thickness, located at about 1 km altitude or higher. 2) Type 2 consists

of profiles showing a decrease of extinction coefficients with altitude, with a maximum extinction coefficient located near the surface. 3) Type 3 is characterized by relatively high extinction values over comparably large altitude ranges, spanning several km without showing distinct maxima. Fig. 9 depicts corresponding profiles, selected for those cases when the above mentioned features are well pronounced. All other cases are shown in the Supplement Fig. S37-S94'.

5

P19 : authors stated that satellite data has always lower accuracy : please explain

The satellite AOD is determined by inverse problem solution. As a rule it is ill-posed problem. That is why AOD from satellite measurements needs validation by ground-based measurements, mainly AERONET. These problems are widely discussed in scientific publications on the remote sensing techniques. Concerning MODIS see e.g. (Remer et al., 2005; Remer et al. 2008;

10 scientific publications on the remote sensing techniques. Concerning MODIS see e.g. (Remer et al., 2005; Remer et al. 2008; Levy et al., 2009, 2010, 2013), CALIOP see e.g. (Kittaka et al 2011) etc. More general discussion of the problem see e.g. in King et al. (1999).

P21 : Results on SSA presented in this study are contradictory to Chubarova et al. (2012) results. Please explain.

15

Observed SSA values in Kyiv during the period of active wildfires were higher in comparison with results, shown in Chubarova et al. We have explained the increased SSA by an increased size of particles, which caused an increase of the atmospheric column reflectance. We have addressed this issue on P15 L35-P16 L1 as follows: 'According to Eck et al. (2009), larger SSA values can be explained by an increased particle size which increases the total reflectance of atmospheric column'.

20

Authors response to Referee #2

We thank the Referee 2 for the detailed and constructive comments, valuable advice which we believe improved the quality and readability of our manuscript. We address the suggested improvements below in blue. The notation is as follows: P1 L12 means page 1 line 12

5 means page 1, line 12.

The manuscript by Galytska et al. attempts to describe the impact of res on the aerosol loads over Ukraine in summer 2010 using a combination of ground and space based remote sensing data. To my opinion the work would be more suitable for 'Atmospheric Chemistry and Physics' journal rather than for AMT. In fact, it analyses and discusses data, but does not provide

10 or use any new 'measurement technique'.That said, I also believe the manuscript is not mature enough for publication and needs major revisions.

We thank the Referee for these comments and we agree that we do not provide any new measurement technique in our research. Although, to our opinion, our research fulfills and covers the subject area of AMT, one of which is the intercomparison of mea-

- 15 surement instruments. Our research mostly comprises detailed description of applied methodology of data comparison between satellite and ground-based measurements and explanation of achieved results. Provided explanations of physical/chemical processes that occurred in the atmosphere during summer 2010 over Ukraine and Eastern Europe are present, but do not dominate in our research. Thus, we considered to submit this paper to AMT.
- 20 Main drawbacks are: 1) Lack of novelty, or at least lack in communicating the novel aspects of the study with respect to the abundant literature on similar aspects of the same fires episodes. 2) Length of the text and use of language both contributing making the reading difficult.

The authors need to make an effort to synthesize the information, focussing on the main aspects of novelty they believe this work contributes highlighting.

25

We completely agree to the Referee and reworked and reorganized the manuscript to highlight its novelty, reduced its length by restructuring methodological paragraphs, optimized the titles of Sections 2 and 3 to better address the sections subject. We have also shortened Sections 3 and 4 by generally more focussing on our major findings and their interpretations.

30 Additional General comments:

- Please use either PAST TENSE or PRESENT TENSE, do not mix the two.

We thank the Referee for having drew our attention on this. We have corrected the manuscript with past tense where appropri-35 ate.

- Please introduce the acronyms the first time you use them or insert a list at the end of the manuscript (e.g., ETR, AOD, AERONET, WMO, SSA...)

40 All acronyms have been spelled out the first time used. Thank you.

- Full sentences taken from other papers should be quoted.

Thank you. We quoted full sentences which were taken from other papers.

45

- I would suggest to use the term 'comparison' rather than 'validation' for the satellites vs AERONET cross analysis.

Thank you. We have replaced the term 'validation' with 'comparison' all over manuscript.

- List of references is often not given in the correct format.

We thank Referee 2 for the comment, although, we are not sure we recall this comment correctly. We revised the references, added missing parentheses in citations all over the manuscript. We also have revised bibtex citations. We hope, with this we could fulfill the request of Referee.

5

- As already mentioned, the language should be improved. Some examples are given in the list of specific comments below. However this should not be considered exhaustive.

We agree to the Referee. We have critically revised entire manuscript, carefully revised the grammar. We re-read the manuscript 10 after all modifications, checked the application of definite/indefinite articles with nouns, corrected manuscript with the past tense, did not pluralize nouns (e.g. aerosol). Also, English has been proofread.

Additional specic comments:

15

Abstract.

Line 5. Use 'plus MODIS ..' rather than 'and MODIS..'

We have corrected 'and MODIS' to 'plus MODIS' P1 L6. Thank you. 20

Line 8. The term 'air pollution' seems too generic here.

We have corrected 'the highest air pollution' to 'the highest aerosol content'. P1 L9.

25

Line 9. The term 'combustion center' is misleading, it could be rather replaced by 'fires' (the same all over the text).

We have replaced 'combustion centers' with 'fires' all over the manuscript. Thank you.

30 Line 11. 'Were' should be 'was'.

> We have rewritten the sentence from passive to active voice as follows: 'We analyzed aerosol spatio-temporal distribution over Ukraine using MODIS AOD 550 nm and further compared with the Kviv AERONET site supplotometer measurements:...'. In reworked manuscript it is P1 L13-14.

35

Line 16. Single scattering albedo is not a microphysical property.

We agree with the comment and rephrased the sentence as follows 'We estimated the influence of fires on the spectral SSA, size distribution, and complex refractive indices using Kyiv AERONET measurements, performed during summer 2010'. Please, check P1 L16-18.

40

Line 18. Please specify better what you mean with 'highest' when talking about 'aerosol pollution'.

We have improved the wording in this sentence and have replaced ' highest aerosols pollution' with 'maximum AOD' as fol-45 lows: 'In this study we showed that the maximum AOD in the atmosphere over Ukraine recorded in summer 2010 was caused by particle transport from the forest fires in Russia'. P1 L19-20.

Introduction

Introduction is too long and should be rewritten. Now it is rather a long list of short summaries from each single previous study on the same matter. From my point of view, the Introduction should be rewritten so to report a synthesis on what we do know and not know from previous studies, the latter (presumably) driving the motivation for the current work. Then the novelty of this work with respect to these previous studies should be clearly mentioned.

5

We agree to the Referee and have rewritten the Introduction entirely. Now it is structured as follows:

- Short introduction into importance of wildfires as a global source of aerosol.

- Description of importance of wildfires events during summer 2010. We have addressed here to previous studies (in chronological order) devoted to aerosol dynamics over the European territory of Russia (ETR) and Eastern Europe. Also, we have highlighted the papers of aerosol research performed for the territory of Ukraine.

- Explanation of the contribution and novelty of our research in comparison with other works, its driving motivation. We have made suggested changes on P2 L2-P4 L11 in the reworked manuscript.

Page 2 line 5. Insert references at the end of such a general statement.

15

10

We have completely withdrawn this sentence.

Page 2 line 9. Remove 'are' at the end of the line.

20 We have completely withdrawn this sentence.

Page 2-3. Description of the results by Konovalov et al. (2011) is too long (16 rows) and should be shortened focussing on the major findings.

- 25 We have rewritten and shortened the description of the results by Konovalov et al. (2011), focussing only on the major findings as follows : 'For example, Konovalov et al. (2011) analyzed the evolution of near-surface concentrations of carbonmonoxide, PM10 and ozone in Moscow region by comparing ground-based and satellite measurements with the modified version of the multi-scale chemistry-transport model for atmospheric composition analysis and forecast CHIMERE, http: //www.lmd.polytechnique.fr/chimere/. They used fire radiative power data retrieved from Moderate-Resolution Imaging Spec-
- 30 trometer (MODIS) on board National Aeronautics and Space Administration (NASA) Aqua and Terra satellites to study the spatio-temporal variability of the fires. They also used MODIS AOD 550 nm to correct a negative bias in fire radiative power measurements in case if fires obscured by heavy smoke. They found that "...extreme air pollution episodes in Moscow were mainly caused by fires taking place at relatively short range (less than 200 km) from Moscow; the transport of air pollution to Moscow from more distant fires was less significant. It was also found that a compensation of a possible negative bias in
- 35 the measured radiative power from fire obscured by heavy smoke is a crucial condition for a good performance of the model". Please, see P2 L14-23. Thank you.

A first Figure introducing the study area and all the main sites/regions referred to in the text should be inserted.

40 Thank you, we have inserted the map with the study area and used AERONET observational sites in Eastern Europe and Ukraine. Please, see reworked manuscript Fig.1 on P5 and figure below.

Section 2.1

45 Page 4 Lines 8-9. This sentence is questionable and should be rephrased. I do not think it is a matter of rating best and worse remote sensing techniques.

We agree with the Referee and completely removed the sentence.

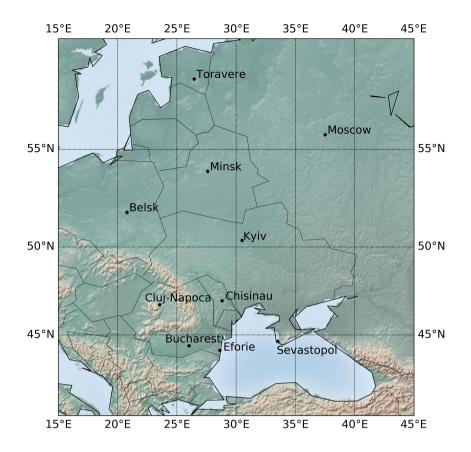


Figure 2. AERONET observational sites in Eastern Europe and Ukraine used in this study.

Page 4 Line 13. What do you mean by 'metrology' here?

We have withdrawn 'metrology' from the sentence. Reworked sentence looks as follows: "The basic principle of the network is to standardize the equipment, measurement techniques, and data processing, which are stored in a freely accessible centralized database'. P4 L16-17

Page 4 Line 20. Correct into (Holben et al., 1998)

Added parenthesis (Holben et al., 1998). P4 L24. Thank you.

10

5

Page 4 Line 21. Angstrom Exponent is used to describe the AOD spectral variability. It is not necessarily computed between 440 and 870 nm. Please clarify better.

We thank the Referee for raising this issue. It is recognized that there is often significant spectral variation of aerosol size distributions with an accumulation mode (Eck et al., 1999; O'Neill et al., 2000). In this paper we present only the 440-870 nm linear fit determination as a first-order parameter indicative of the general size distribution and the relative dominance of fine versus coarse mode particles (Holben et al., 2001). The 440–870 nm Angstrom is computed from linear regression of ln AOD versus ln λ scale at 440, 500, 675 and 870 nm (Eck et al., 2000).

5 versus ln λ scale at 440, 500, 675 and 870 nm (Eck, et al., 2009).

We have reworded and clarified this statements on P4 L24-28 as follows: 'The Angstrom exponent (AE) is also determined by the AERONET algorithm for the sunphotometer spectral range 340-870 nm from direct sun irradiance measurements. We used AE to interpolate the AOD on the required wavelength. We applied AE determined for 440-870 nm because it is suitable to

10 the aerosol size distribution during wildfires when relative dominance of the fine mode particles takes place (Eck et al., 1999; Holben et al., 2001)'.

Page 4 Line 22. What do you mean with 'altitude circle'? Do you mean 'principal plane' measurements? Please clarify. Also insert a comma after 'Sun'.

15

We have completely withdrawn this sentence in the reworked version of manuscript.

Page 4 Lines 29-30. Please list here which additional AERONET sites in Eastern Europe you used.

- 20 We listed AERONET sites of Eastern Europe on P4 L29-32 as follows: 'To assess the extent of the impact of wildfires in summer 2010 we used data from the following Eastern European AERONET sites (also shown in Fig.1): Minsk (Belarus), Moscow (Russian Federation), Toravere (Estonia), Belsk (Poland), Moldova (the official name of the site is Moldova, although it is located in Chisinau, Moldova), Cluj-Napoca, Bucharest, and Eforie (all Romania). We also used data from the only two Ukrainian sites that measured during summer 2010: Kyiv and Sevastopol'.
- 25

Section 2.2

Page 5 Lines 8-9. Please rephrase.

30 We have completely withdrawn this sentence.

Page 5 Line 11. Please rephrase.

We have rephrased the sentence as follows: 'Over land, aerosol properties are retrieved from spectral channels 0.47, 0.66, and
2.12 m. One of the primary aerosol products of the MODIS algorithms is the AOD at 550 nm in the atmosphere over land and ocean (Remer et al., 2005, 2008; Levy et al., 2007, 2010, 2013)'. P6 L7-9.

Page 5 Lines 14-16. The MODIS product you used is obtained from both land and ocean MODIS algorithms, not just the land one. The fact that you only use land retrievals given the region under examination is a different thing. Please rephrase.

40

We agree to the Referee and have rephrased the sentence as follows 'To estimate atmospheric pollution over Ukraine caused by aerosol from wildfires, we used AOD at 550 nm retrieved by the land algorithm and collected in the MODIS Aqua and Terra Level 2 Collection 005 and 051 Optical_Depth_Land_And_Ocean product file'. P6 L10-13.

45 Page 5 Line 27. This sentence should be rephrased as not clear in the current form.

We thank the referee for highlighting this issue. We have rephrased the sentence as follows: 'For comparably clean atmospheric conditions, when AOD is close to zero, AOD values in the range ± 0.05 are practically indistinguishable (Remer et al., 2005,

2008; Levy et al., 2009). Following Levy et al. (2009), we set corresponding data in this range to zero'. P6 L20-22.

Page 5 Lines 33-34. The given link should not include a specific period as it is now.

5 We have completely replaced the link 'http://rapidfire.sci.gsfc.nasa.gov/cgi-bin/imagery/firemaps.cgi?period= 2010221-2010230' by 'https://firms.modaps.eosdis.nasa.gov/map/' as the 10 day global fire maps are no longer being produced. P7 L2. Thank you.

Page 6 Line 5. Should be just 'vegetation fires'

10 We have replaced 'under conditions of forest fires and other vegetation or peat' with 'wildfires' to be consistent all over the manuscript. P7 L7.

Section 2.3

15 Page 6 Line 12. Should be 'at nadir'

Corrected to 'at nadir'. P7 L14. Thank you.

Page 6 Lines 15-16. Should be 'CALIOP measurements allow to derive aerosol and clouds vertical distribution...' or something 20 like that. Please rephrase. In the current form, the sentence is not fully correct.

We have rephrased current sentence in a more simple way: 'CALIOP measurements allow to derive the vertical distribution of aerosol and clouds'. Please, see P7 L18.

25 Page 6 Line 19. 'Were' should be 'was'.

We have rewritten the sentence from the passive to active voice as follows: 'For this study we used both parameters, vertical distribution of the extinction coefficient and AOD at 532 nm, defined along the path of the sub-satellite point'. P7 L20-21.

30 Section 2.4

Page 6 Line 27. 'on ' should be 'of'

We have corrected 'on' to 'of'. Thank you. P8 L15.

35

Page 7 Lines 1-2. Not clearly written. Please rephrase.

We have rephrased the sentence as follows: 'According Stohl (2002), the uncertainty of calculated HYSPLIT trajectories for a period longer than 24 hours is around 20 % in the horizontal direction in the free troposphere; after 120 hours the uncertainty
40 increases to about 400 km in the horizontal and about 1300 m in the vertical planes'. P8 L26-28. Thank you.

Section 3

Methodology and Results should appear in separate Sections.

45

We agree to the Referee and we have reworded the titles of Section 2 and 3.

Section 2 has been renamed from 'Data sources' to 'Methods and data sources'. We aim to collect all relevant information about methods used in our study in a respective section. Section 3 has been renamed from 'Methodology and results' to 'Re-

sults and Discussion'.

Page 7 Lines 4-5. This is a repetition. Please remove.

5 Thank you. Removed.

Page 7 Lines 11-18. Please make this part shorter focussing on the main points affecting the way you can make use of the CALIOP dataset. Better explain which is your 'collocation' criteria for CALIPSO given the limitations described.

10 We have restructured the manuscript, and the description of methodology, in particular CALIOP comparison with other instruments, is now presented in Section 2.3, P7 L23-P8 12.

Page 7 Lines 18-19. Not clear. Please rephrase.

15 We have removed the entire sentence. Although, as mentioned in the previous reply, we have restructured the paper and moved the description of methodology regarding CALIOP to the Section 2.3.

We address these changes on P7 L23-P8 L12 as follows:

'A comparison of CALIOP AOD with ground-based AERONET observations can be challenging because of different measurement characteristics of both instruments. The CALIOP lidar provides only fragmentary data on aerosol along CALIPSO

- 20 satellite's ground track, because of the small size of its light beam and cloudy conditions that frequently occur. Due to instrument's orbital period of 98 minutes, ground tracks of satellite consecutive passages at certain latitudes are shifted 24.5° to the west, making its spatio-temporal coverage rather sparse. Consequently, the probability of CALIOP to pass over the atmospheric column observed by the solar photometer AERONET is also rather limited (e.g. Redemann et al., 2012). During the three summer months of 2010 we found no coincident CALIOP-AERONET measurements over Kyiv, apart from the single collocation,
- 25 although not exactly matching the selection criteria according to Omar et al. (2013). In this case the closest CALIPSO ground track was found 60 km east from the Kyiv AERONET site.

Therefore, in this study we only compared CALIPSO/CALIOP CloudAerosol Layer Product AOD 532 nm (Winker et al., 2009, 2010) with MODIS/Aqua AOD 550 nm since the orbits of both satellites are in the A-Train afternoon constellation (http://atrain.nasa.gov/). Relative positions of CALIPSO and Aqua satellites in the A-Train provide great number of practically

- 30 simultaneous measurements with the time span of 2 minutes, while the spatial difference is only about 10 km. Each granule of MODIS data consists of consecutive scans across the satellite track. The footprint of CALIOP light beam on this granule looks like a sequence of points on the straight line, which are passing close to the center of a granule. Each of these points represent the center of the CALIOP measurement averaged over 5 km and once or twice matches with one of the pixels of a MODIS granule. To find these matches we calculated the distances and azimuth angles between the center of each CALIOP point and
- 35 the center of each pixel in MODIS granule in the same manner as for the MODIS-AERONET case, described in Section 2.2. We averaged MODIS data over areas 50 km × 50 km, while CALIOP data in the CALIPSO CloudAerosol Layer Product are averaged on the various distances along the satellite ground track, up to 80 km (see CALIPSO Quality Statements Lidar Level 2 Cloud and Aerosol Layer Products, Version Releases: 3.01, 3.02). We did not apply any correction for potential spectral differences while comparing CALIOP AOD 532 nm and MODIS/Aqua AOD 550 nm. This yields to an estimated systematic
- 40 bias in our AOD comparison of approximately 2–6 % in the AE range between 0.5 to 1.8 (see Fig. 5b) and can be neglected in our cases, following Kittaka et al. (2011)'

Section 3.1

45 I think a better definition of the term 'summer' used throughout the text is needed. Isn't summer defined as June-July-August in your work?

We agree with the Referee's comment and defined summer 2010 as June 1-August 31 in the Abstract and Introduction.

Why do you limit the fires analysis in Fig 1 to the period 1 July- August 20?

We have followed the Referee's comment and reworked the Figure. Now this Fiure includes fires location and brightness temperature (in K) of fires pixels in Eastern Europe and the ETR (40–65N and 10–60E) from June 1 to August 31. Please, note,

5 that in the reworked version of manuscript, this Figure corresponds to Fig. 2, not Fig. 1. Please, see updated Figure on P10 of reworked manuscript or below.

And why Fig 2 shows data from June 1 to August 24??

10 We have also reworked this Figure and its updated version covers total amount of the fire pixels for each day of summer 2010 from June 1 to August 31. Please, note, that in reworked version of manuscript, this Figure corresponds to Fig.3, not Fig.2. Please, see P11 or below.

Please clarify better the term 'summer 2010' since the beginning and try to be consistent with this in your Figures.

15

We agree with the Referee's comment and as mentioned before, we defined summer 2010 as June 1-August 31 in the Abstract and Introduction.

Please, show on Figure 1 the study area used in Figure 2.

20

We have followed the Referee's suggestion and in the reworked manuscript Fig.2 shows the study area used in Fig.3. Please note, that reworked figures differ from 'original' versions because we now used high-confidence data of brightness temperature of fire pixels. This means that the confidence level of calculated fire pixels is larger/equal 80%. In previous version of the manuscript we used all available data, which included low confidence data.

25

Page 8 Lines 1-2. Actually, given the large number of fires in Fig.1, it is not the best way to show 'fires concentration'. A metric like the number of fires per unit area should be rather used for this purpose. Either rephrase this sentence or modify Fig 1 accordingly.

30 We agree to the Referee and rephrased the sentence. We replaced "the highest fire concentrations" in the following way: 'Both the overall number and the brightness temperature of fires reached their maximum between July 26 and August 18'. P9 L8-9.

Page 8 Line 3. If I understand correctly, FRP stands for Fire Radiative Power (please, provide acronyms). However, note that 55 Fig 1 does not show FRP but T. Since these are different quantities, again either modify Fig. 1 or modify the sentence.

We have modified the the sentence and replaced "FRP" with 'brightness temperatures'. Thank you.

Table 1: From Table 1 (and relevant text) it is not clear to me which is the overall period considered to compute the values reported.

We have improved the caption of Table 1 (P11) to 'Level of air pollution caused by aerosol (AOD 500 nm) from June 1 to August 31, 2010 over the ETR and Eastern Europe according to AERONET' and improved the text on P9 L15-18 as follows 'We analyzed changes in AOD at 500 nm using all daily averaged measurements from June 1 to August 31 from the AERONET deteloge from Minel (Balene). Magazing (European (European) Terreger (Estenic), Balela (Balene).

45 the AERONET database from Minsk (Belarus), Moscow (Russian Federation), Toravere (Estonia), Belsk (Poland), Moldova (Chisinau/Moldova), Cluj-Napoca, Bucharest, Eforie (all Romania), Kyiv and Sevastopol (both Ukraine)'.

Page 9 Line 5. Replace with 'for dates of maximum AOD'.

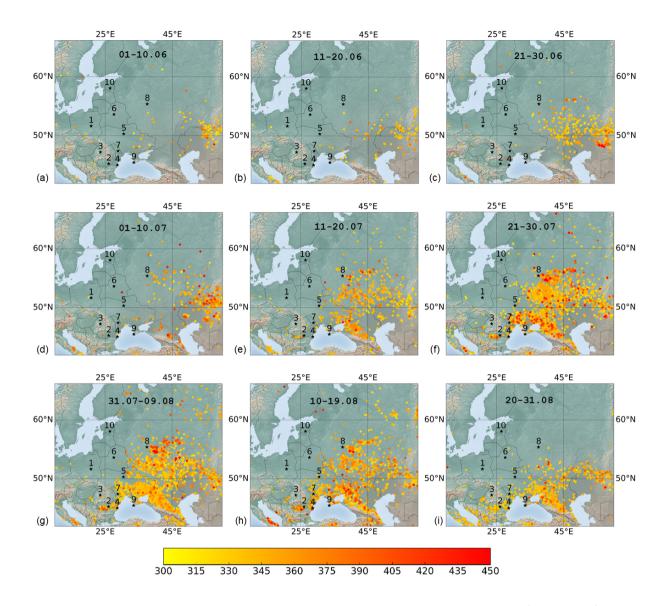


Figure 2. Fire locations and brightness temperature (in K) of fire pixels in the ETR and Eastern Europe (40–65°N and 10–60°E) detected by MODIS and accumulated over 10 day periods from June 1 to August 20: (a) June 1–10, (b) June 11–20, (c) June 21–30, (d) July 1-10, (e) July 11-20, (f) July 21-30, (g) July 31-August 9, (h) August 10-19, and a 12 day period (i) August 20-31. Black stars and numbers indicate the position of AERONET stations also shown in Fig.1: 1-Belsk, 2-Bucharest, 3-Cluj-Napoca, 4-Eforie, 5-Kyiv, 6-Minsk, 7-Moldova, 8-Moscow, 9-Sevastopol, 10-Toravere.

Change made. Thank you. P9 L23-24.

Page 9 Line 8- Page 10 Line 24. This part is too long and should be shortened.

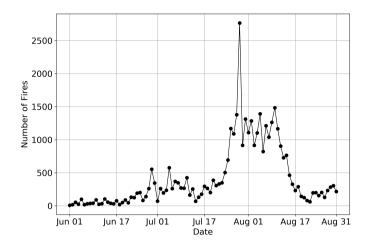


Figure 3. Total amount of fire pixels for each day of summer 2010 according to MODIS Aqua and Terra data over the area shown in Fig.1 (40–65°N and 10–60°E).

We agree to the Referee and have slightly shortened part of the text. However, the aim of this description is to show that the maximum AOD values over analyzed AERONET sites were formed under conditions of transport of aerosol from the areas of active wildfires. Moreover, such a detailed analysis of the transport of air masses to the analyzed AERONET sites has not been performed in earlier studies for the days of summer 2010, when maximum AOD was observed. Revised text looks as follows:

5

10

'Back trajectories for Moldova and Belsk, where the maximum AOD was observed the earliest within summer 2010 (July 13 and 16, respectively), are shown in Fig. 4a,b. The trajectories indicate that aerosol was transported to Moldova at altitudes from 0.5 to 1.5 km from the fires in the ETR and south-east of Ukraine (see also Fig. 2d,e). Into the region of Belsk, aerosol was transported across continental Europe (1.5–5 km) mostly from the Atlantic Ocean, but also from the Baltic across regions of active fires (Fig. 2) in the lower atmosphere (500 m).

Transport of aerosol to two Romanian sites (Cluj-Napoca and Eforie) with maximum AOD observed on August 1 (Fig. 4c,d) also occurred in the lowermost 1.5 km layer, originating from the southeast of Ukraine and Moldova (also the area with active fires). Back trajectories for Moscow and Toravere with a maximum AOD on August 7 are shown in Fig. 4e,f, respectively. Aerosol to Moscow was transported mostly from the surrounding regions with the most active fires. Air masses over Toravere

15 (Fig. 4f) originated from Asian regions and crossed areas of active fires in southeastern Ukraine in all analyzed altitudes (Fig.2g).

To Kyiv, where the AOD maximum was observed on August 15, aerosol was transported in the lower 4 km layer from the most active fires in the ETR, Ukraine, and Moldova (Fig. 4g, Fig. 2h). On August 16, the maximum was recorded in Sevastopol on the Black Sea coast, where air masses traveled in almost the entire range of analyzed heights (500 m–5 km) from the

- 20 ETR and Kazakhstan through the territory of active fires in the south-west of Russia (Fig. 4h). In Minsk and Bucharest the maximum AOD was observed one day later on August 17 (Table 1, Fig. 4i,j). Towards Minsk aerosol was transported at 3 km from Kazakhstan across the Caspian Sea, southern Russia, and Ukraine, where the active fires were observed; at 1.5 km from Ukraine, and at 500 m from the western regions of the ETR through Ukraine. To Bucharest aerosol from fires was transported at 500 m from the north-east, specifically through the ETR and the south-east of Ukraine.
- 25 According to the monthly weather reports of the Ukrainian Weather Center, a change in weather was observed on August 18-21. Atmospheric fronts of an active cyclone which moved from the southern Baltic region to Samara led to a significant change of weather pattern in Eastern Europe. This change caused a distinct decrease in fire activities and a wet deposition of

aerosol, lowering its content in the atmosphere above all investigated regions in the second half of August.'

Figure 3: Use of different dates does not help the understanding of the atmospheric circulation over the area in the investigated period. I suggest including a figure with wind patterns over the whole region at the different levels and dates, to be coupled to find info of Figure 1.

5 fires info of Figure 1.

We thank the Referee for having drew our attention to this issue. The aim of Fig. 3 (in reworked version of manuscript it is Fig.4) was to show that the maximum AOD values in the different AERONET stations were formed under conditions of transport of air masses from the areas of active wildfires. We did not include the map with the winds at the different levels and dates as we

- 10 are sure, that HYSPLIT model correctly represents the transport of air parcels. We achieved such conclusion after comparing HYSPLIT back trajectories with weather charts (not included in this paper) near the surface 1000 hPa, at 1.5 km 850 hPa, 3 km 700 hPa, and 5 km 500 hPa, provided by Ukrainian Weather Service, and also accessible at http://www.wetterzentrale.de/ and http://www.wetter3.de. Moreover, to our opinion, the coupling of wind maps into Fig.1 (now Fig.2) would not be possible as it accumulates fire pixels over the 10 day period (not the average number of fire pixels).
- 15

As a further suggestion, it would be more useful to plot back-trajectories together with the fires data to better understand if and how each site was (possibly) impacted by fires.

We thank the Referee for his/her suggestion. To further avoid additional Figures in the manuscript and save the space, we
have added the location of the AERONET sites to Fig. 2 in the reworked paper. Please, see Fig.2 above or on P10 of the reworked manuscript. We believe that this Figure addresses well location of the AERONET sites and spatial distribution of fires, accumulated over the 10 day period during summer 2010.

Section 3.2

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30

Section 3.2 is too long and confusing. It is not clear how much new information it contains with respect to Milinevsky et al. (2014). To my opinion, the authors should avoid describing what happens each couple of days and limit the analysis/discussion to the most important periods identified (e.g. those identified in Section 3.3), stressing on the results quantifying the fires impact over Ukraine in summer 2010 (as promised in the title). In doing so, it is important to clarify what was already known based on previous literature, and which are the new findings of this work.

We agree with the Referee and we have reworked this Section. We have revised the beginning of the section on P12 L10-18 as follows:

- 35 'Between 2003 and 2014 ground-based and satellite observations showed the highest aerosol content over Kyiv every year in spring (April–May) and late summer (July–August; Bovchaliuk et al., 2013; Milinevsky et al., 2014). According to both studies, the observed spring peak in aerosol content is associated with transport of the Saharan dust across Eastern Europe, transport of sea salt aerosol from the Black Sea and the Sea of Azov, and occasionally occurring agricultural fires. The summer peak results from wildfires, soil dust aerosol due to harvesting activity, and transport of Saharan dust. The lowest AOD was
- 40 observed in June and the middle of autumn. In Table 2 we show a prolongation of the data record of Milinevsky et al. (2014, 2008-2013) for the Kyiv AERONET site by three more years up to the end of 2016. Even in this extended record the most significant aerosol pollution was observed in August 2010. This event is related, in particular to wildfires in the ETR and Eastern Europe'.
- 45 We have reworked Table 2 (reworked manuscript P14) and recalculated AOD values for 500 nm to be consistent in our study. We do not use, show, or refer to AOD 440 nm in this manuscript any more. We also have extended Table 2 up to 2016 (all available level 2.0 data for the Kyiv AERONET site). Thank you.

We have also moved the description of air transport each couple days within June-August 2010 to the Supplement.

Figure 4b is redundant

We have removed Fig. 4b. Thank you. 5

Figure 5 should be removed or inserted as supplementary.

We moved Figure 5 to the Supplement. Thank you.

10

Page 16 Line 7-9. This last part of the Section is perhaps the most interesting one, in the sense that it adds information with respect to previous studies. However, these sentences should be rewritten to be more clear. For example avoid saying 'The fires impact on aerosol size from AERONET observations can be estimated by knowing XXX and size distribution of the aerosol particles XXX', as this is quite obvious. Again, the authors should try to focus on the most interesting results, without

unneeded redundancy. 15

> We agree to the Referee and revised the sentence on P15 L18-19 as follows: 'We estimated the impact of fires on the aerosol size from AERONET supphotometer observations by calculating correlation coefficients between the aerosol effective radius and AOD (Fig. 6a), following the approach of Chubarova et al. (2012)'.

We have also revised the paragraphs on P15 L19-P16 L5 20

Figure6c: How do you explain the low values of SSA all over the spectrum in Jul18-Aug 14 with respect to Aug 15-17?

The decrease of SSA during July 18-August 14 most probably was caused by increase of soot content in the air, which was transported from the fires. And increased absorption of aerosol in the long-wave part of spectra is explained by the slope of 25 SSA curve, which becomes steeper. Similar shape of the SSA curve is also observed during August 15-17, which agrees with increased absorption of aerosol and is similar to the period July 18-August 14. But larger SSA values during this 3 day period can be explained by increased size of particles, which caused an increase of atmospheric column reflectance. We have addressed this issue on P15 L35-P16 L1 as follows: 'According to Eck et al. (2009), larger SSA values can be explained by an

30 increased particle size which increases the total reflectance of atmospheric column'.

Table 3. As already commented, SSA is not a microphysical property. Please, reduce the number of significant digits. I also recommend moving it to the supplementary, as it does not add much to results in Fig. 6.

We have moved Table 3 to the Supplement(Table S35, P12), changed the caption of the table, reduced the number of significant 35 digits. Thank you.

Section 3.3

40 Page 17 Line 5. Again, caution using the term FRP as this was not introduced/shown in Section 2.

We agree with the Referee's comment and have avoided the application of term 'FRP'. Thank you.

Page 17 Lines 7-8. Unclear, please rephrase.

45

We have removed this paragraph as we do not divide summer 2010 in this Section into 3 periods in the reworked version of manuscript.

Section 3.3.1

This section is largely a description of the methodology to match AERONET and MODIS data. Page 19, Lines 3-25) could be better moved in a specific section within the Methods.

5

We agree with the Referee and we have moved the description of methodology into Section 2.2. Please, see P6 L23-30.

Figure 7 : I would use a different scale for the two panels. In the present form panel a) gives little information.

- 10 This is a good point. We agree that with the current scale Fig. 7a shows only relatively low AOD values over Ukrainian territory on July 17, which are homogeneously distributed. Fig.7a becomes more interesting in combination and comparison with Fig. 7b, which represent AOD values in August 15. Such a combination shows the major AOD increase in August within the whole summer 2010.
- 15 Page 21 Lines 9-17. Language should be improved.

We have improved the wording of this paragraph on P17 L16-29 as follows: 'The influence of aerosol pollution in Ukraine can be interpreted from the spatial distribution of MODIS AOD 550 nm. We analyzed data for 7 days with low AOD values, smaller than 0.5 over the Kyiv site: June 6 and 7 (MODIS/Terra), June 8

- 20 (MODIS/Aqua); July 14 and 17 (MODIS/Aqua), July 15 (MODIS/Terra); August 23 (MODIS/Terra). We also analyzed 3 days (August 15–17) with high AOD values, larger than 1.0. Figure 7 shows maps for the region 40-60°N and 22.5-40°E of MODIS/Aqua data for those two days when the aerosol load over the Kyiv AERONET site was low (a, July 17) and the highest (b, August 15). During the days with low aerosol content the AOD 550 nm was homogeneously distributed (e.g., Fig. 7a) over the whole territory. During the high pollution case, the spatial AOD distribution distinctly differed. The highest AODs were
- 25 observed over north-eastern and central regions of Ukraine, where AOD values reached and partly exceeded a value of 2 (Fig. 7b). This AOD distribution map (Fig. 7b) resembles our air mass back trajectory calculations to Kyiv in the altitude range of 0.5–3 km for August 15 fairly well (see Fig. 3, Sect 3.1). This indicates that the MODIS algorithm interprets aerosol over Kyiv in the same manner as AERONET. However, MODIS underestimates low AOD values and overestimates high AOD values in comparison with AERONET. Figure 7b also highlights the importance of the availability of satellite observations for estimating
- 30 air pollution over larger and remote regions, which cannot be deduced from a single site's ground-based measurements, as it is the case in Ukraine'. Thank you.

Section 3.3.2

- 35 Similarly to the previous case, this section is largely a description of methodology rather than of results. Please, try to separate methodology from results as this would help a lot the reading of the work. This section shows the same problem of others, i.e., it is structured as a list of dates with description of features, but lacks a synthesis of the main findings CALIPSO data tell us about the phenomenon under examination. It should be shortened and restructured so to highlight which is the important (possible quantitative) information CALIPSO adds to the overall picture.
- 40

We agree to the Referee and reworked the Section 3.3.2 .We have moved the description of the methodology to the Sect. 2.3. Please, see P7 L23 - P8 L12.

Page 22 Line 5. Specify where (in altitude) the laser beam has a 70 m diameter. Due to the beam divergence, this is not the same all over the laser path.

We moved this sentence to Section 2.3 and specified that the footprint of the laser beam is 70 m on the Earth's surface as follows: 'This results in the footprint on the Earth's surface, called in our paper ground track, of about 70 meters'. Please, see

P7 L16-17.

Page 22 Line 10. What do you mean by 'estimated CALIPSO AOD using MODIS AOD'? Please clarify better.

5 We have corrected the sentence as follows: 'Therefore, in this study we only compared CALIPSO/CALIOP CloudAerosol Layer Product AOD 532 nm (Winker et al., 2009, 2010) with MODIS/Aqua AOD 550 nm since the orbits of both satellites are in the A-Train afternoon constellation (http://atrain.nasa.gov/)' and moved the sentence to Section 2.3, P7 L32-34.

I do not see the reason for introducing the further term 'time-span' in this Section. Please identify specific periods of interest 10 for the study, give them names, and use these all through the text.

We agree with the Referee and have completely removed the term 'time span' together with Table 6 and Table 7.

Page 24 Lines 16-17. What do you mean by 'ground track'? Please specify.

15

20

We refer to the term 'ground track' to define the footprint of CALIOP lidar measurements on the Earth's surface. We have provided the explanation on P7 L14-17 as follows: 'The laser beam is directed almost at nadir with a slight forward tilt in the direction of motion of the satellite to avoid direct reflection of laser radiation from high reflectivity objects (surface water, snow, etc.). The divergence of the transmitted laser beam equals 100 rad. This results in the footprint on the Earth's surface, called in our paper ground track, of about 70 meters'.

Page 22 Lines 17-19. Unclear, please rephrase.

We have addressed this issue in Section 2.3, P8 L3-5 as follows: 'Each of these points represent the center of the CALIOP measurement averaged over 5 km which matches with one of the pixels of a MODIS granule'.

Figure 9. Please group profiles according to the three types you identify in the text. It is necessary to include labels a) to g) on a map (e.g. in Figure 8) to show the relevant location and better follow all the discussion in the text. Use the same X and Y-scale for all plots to simplify the comparison.

30

We agree to the Referee's suggestions and have provided several improvements:

- We have introduced one more aerosol profile For August 18 on Fig. 9 and substituted profile 'b'
- All profiles were set in chronological order
- We have included the term 'type of profile' to each (a-h) profile, shown in Fig. 9
 - We have reworked the caption of Fig. 9 as follows: 'Selected vertical profiles of aerosol extinction coefficient from CALIOP measurements over Ukraine during active fires period in summer 2010. The location of profiles is shown in Fig. 8b with corresponding a-h labels'
 - We have included the location of these profiles on Figure 8b with a-h labels.

40

Please, see reworked Figures 8 and 9 below and/or in the reworked manuscript P18, P19 correspondingly. Why not using the CALIPSO depolarization to further characterize aerosol layers?

We agree with the Referee that the application of CALIPSO depolarization data would provide us additional information on 45 aerosol properties, size, and shape. In our research we applied AERONET ground-based measurements to further analyze

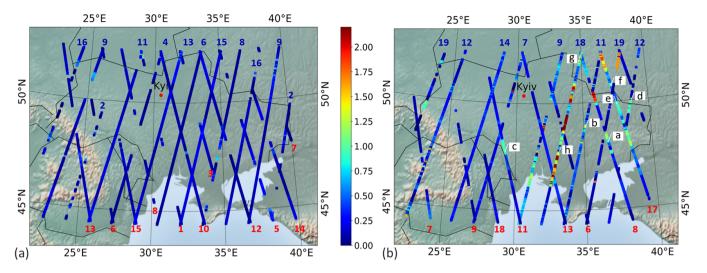


Figure 8. AOD 532 nm distribution over Ukraine from CALIOP measurements during 16 day period from June 1 to 16 (a), from August 4 to 19 (b), 2010. Red numbers at the bottom of the map indicate dates of each daytime track running to north-west and blue numbers at the top of the map indicate the date of each nocturnal track running to south-west. Labels a-h in Fig. 8b indicate the location of profiles, that are further analyzed and shown in Fig. 9.

aerosol spectral SSA and microphysical properties with the focus on the Kyiv site. The application of additional data set would at the first place extend the size of the paper, which we try to avoid. We also believe that this research should be performed additionally and published as kind of extension of our study with incorporating data from other years since 2010.

5 Section 4

Page 27 Lines 6-17. I would avoid the discussion on the reason for CALIPSO and MODIS AOD mismatch here. It is not very pertinent to this study and in the present form contains several questionable statements. In general, this section is also too long (3.5 pages). It has to be shortened once all the given suggestions have been addressed. It would be desirable to restructure this section in order to have a sort of 'main finding list' here.

10 section in order to have a sort of 'main finding list' here.

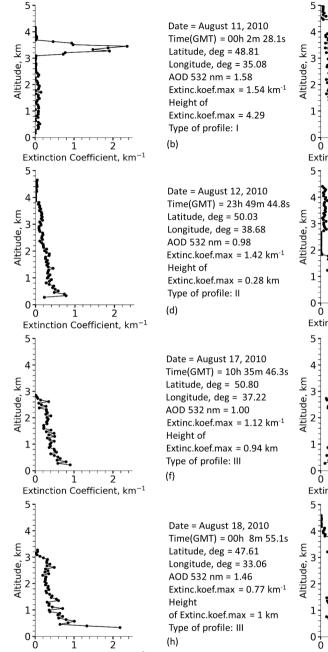
We agree to the Referee and we have entirely reworked this Section. Now it better highlights our findings, because we have avoided detailed comparison of our results with other authors as it was done in the previous version of the manuscript. Please, see the rewritten version of Conclusions on P22 L4-P23 L32.



Date = August 11, 2010 Time(GMT) = 11h 12m 05.0s Latitude, deg = 47.87 Longitude, deg = 29.15AOD 532 nm = 0.93 Extinc.koef.max = 0.77 km⁻¹ Height of Extinc.koef.max = 0.34 km Type of profile: II (c)

Date = August 17, 2010 Time(GMT) = 10h 35m 31.2s Latitude, deg = 49.92 Longitude, deg = 37.60AOD 532 nm = 0.92 Extinc.koef.max = 0.90 km⁻¹ Height of Extinc.koef.max = 0.22 km Type of profile: II (e)

Date = August 18, 2010 Time(GMT) = 00h 7m 36.2s Latitude, deg = 52.28 Longitude, deg = 35.03AOD 532 nm = 1.22 Extinc.koef.max = 2.18 km⁻¹ Height of Extinc.koef.max = 0.34 Type of profile: II (g)



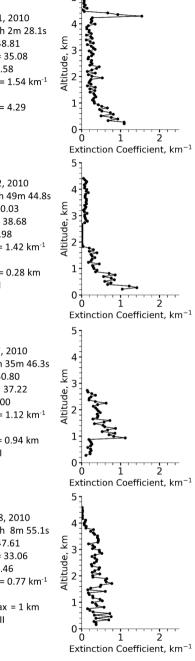


Figure 9. Selected vertical profiles of aerosol extinction coefficient from CALIOP measurements over Ukraine during active fires period in summer 2010. The location of profiles is shown in Fig. 8b with corresponding a-h labels.

(h)

2

Extinction Coefficient, km⁻¹

Authors response to Referee #3

The authors thank the Referee for his/her revision of our manuscript and helpful comments. We address the suggested improvements below in blue.

5 This paper analyses a number of sun-photometer and satellite observations, together with back-trajectory modelling, for a full analysis of the aerosol load over Ukraine and surrounding areas during the summer of 2010. It is shown that the high aerosol loads were generated by wild fires of exceptional intensity over central and western Russia.

This papers is very well presented. It is easy to read and the figures are of high quality. It goes onto the detail at each step. For instance, the authors use sun-photometer measurements to evaluate the satellite products, which is not the objective of the

10 paper. In addition, there are several statements that appear in the introduction, main body and conclusion of the paper. As a consequence, the paper is very long and not well focused.

Although I would have preferred a paper better focused on the analysis of the aerosol load and its origin, there is nothing wrong, and the paper offers a very thorough analysis of the atmospheric impact of the fires during the summer of 2010. It could be published as is.

- 15 We appreciate very much for the positive comments on our manuscript. We agree with the Referee that the content of the manuscript is too extensive in its original state. We have reworked and reorganized the manuscript to highlight its novelty, reduced its length by restructuring methodological paragraphs and better concentrating on main messages, and improved the language. Particularly, we have introduced the following changes:
 - We have shortened the Abstract.
- 20 All acronyms have been spelled out the first time used.
 - We rewrote the Introduction and inserted a figure showing a map with AERONET observational sites in Eastern Europe and Ukraine, which were used in our study (Fig.1 in the reworked manuscript). We have also added to both the Abstract and Introduction the description of the term "summer" 2010, which comprises the time period June to August 2010.
- We have optimised the titles of Section 2 and 3 to better address the sections subject. In detail, we have renamed Section 2 from "Data sources" to "Methods and data sources". We aim to collect all relevant information about methods used in our study. In the original manuscript, respective information was partly spread over the different sections describing results of our study. The current form of Section 2 allows the reader to better understand the core messages of our analysis and conclusions statements that are now not mixed with the technical information. Section 2.4 has been renamed from "Meteorological data and the means of study of air masses transport in the investigated region" to "Weather conditions and transport of air masses", now even more precisely state the purpose of the information provided there. In this respect, Section 3 has been renamed from "Methodology and results" to "Results and Discussion".
 - The majority of Figures and tables as well as their captions have also been reworked and improved (please, see the updated version of the manuscript).
 - We have shortened Section 3 and reworked Section 4 by generally more focussing on our major findings and their interpretations.
 - Language and Grammar have been proofread.

35

Authors response to Referee #4

We thank the Referee #4 for the time he/she spent on reading and revising the manuscript and raising a number of important points. We are following the Referee's suggestion and reworked the manuscript. Below we address all raised points one by one. All authors' responses are highlighted in blue. We address notation as follows: P1 L12 means page 1, line 12.

Review of Increased aerosols content in the atmosphere over Ukraine during summer 2010, Evgenia Galytska, Vassyl Danylevsky, René Hommel, and John P. Burrows.

10 This study re-visits the res of summer 2010 over western Russia and surrounding countries. The impact of aerosol burden and composition over Ukraine are evaluated. This study closes the loop on analyses using aerosol data from satellite and in-situ measurements to discuss that effect of the Russian res on neighboring countries; specically in this case, Ukraine.

The study is poorly written. There are numerous* issues with the grammar and sentence structure (I've included a few of the most egregious ones) that I strongly suggest having a colleague (fluent in English) vet the sentences for clarity and syntax. In particular what is missing is 'the' for proper nouns. For example in the text authors say 'we used data mostly from Kyiv site' when it should be written 'we used data from the Kyiv site'. Authors also pluralize words that should be singular.

Furthermore, authors do not stay on one tense. Pick one: past or present. The manuscript requires a re-read on this. Such error is too extensive to document.

We agree to the Referee #4 and we improved the grammar of the manuscript. We carefully re-read the manuscript after all modifications, checked the application of definite/indefinite articles with nouns, corrected manuscript with the past tense, did not pluralize nouns (e.g. aerosol). Also, English has been proofread.

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40

5

Having said that, the method and general results are not faulty. Ignoring the poor English, the study is good enough to be publish with major revisions, such as improving the writing and additional analysis to support many of the authors claims.

We appreciate very much the comment and we have reworked and reorganized the manuscript to highlight its novelty, reduced its length by restructuring methodological paragraphs. In particular, we have introduced the following changes:

- We have shortened the Abstract
- All acronyms have been spelled out the first time used
- We have rewritten the Introduction and inserted in Sect. 2 a figure showing a map with AERONET observational sites in Eastern Europe and Ukraine, which were used in our study (Fig.1 in the reworked manuscript)
- We have optimised the titles of Section 2 and 3 to better address the sections subject.

In detail, Section 2 has been renamed from "Data sources" to "Methods and data sources". We aim to collect there all relevant information about methods used in our study. In the original manuscript, respective information was partly spread over the different sections describing results of our study. The current form of Section 2 allows the reader to better understand the core messages of our analysis and conclusions - statements that are now not interrupted by technical information.

Section 2.4 has been renamed from "Meteorological data and the means of study of air masses transport in the investigated region" to "Weather conditions and transport of air masses", now even more precisely state the purpose of the information provided there.

In this respect, Section 3 has been renamed from "Methodology and results" to "Results and Discussion". Majority of Figures, Tables and their captions have also been reworked and improved (please, see the updated version of the manuscript). - We have shortened Sections 3 and 4 by generally more focussing on our major findings and their interpretations.

Authors write 'summer' 2010 in the introduction - define the months or was it one month, i.e. August, which you quote from previous studies? Be precise is it summer or merely August?

5 We make this point in re-written Abstract (P1 L2) and in Introduction (P2 L7), where we have defined the months in parenthesis, e.g. (June 1-August 31).

3rd paragraph in abstract - Remove 'apparently'. High pollution over Moscow that summer 'was' due to the fires. There's nothing apparent about it.

10

Thank you. We have rephrased the sentence and removed 'apparently' (P1 L9-10).

- I do not like the term 'combustion center'. Be precise it was fires.
- 15 We have corrected 'combustion centers' to 'fires' all over the manuscript. Thank you.
 - 'aerosol content' singular.

We have replaced plural 'aerosols' with singular 'aerosol' all over the manuscript. Thank you.

20

Last sentence in abstract: 'change of the particle microphysics'

We have corrected the sentence to "change of the particle microphysics' (P1 L22). Thank you.

25 Pg 2, Line 4: but a 'non' negligible

We have added the reference of Barnaba et al., 2011 to the particular sentence and retained the original usage of word "not".

Last sentence of the Introduction first paragraph needs to be re-written. The sentence is long for one.

30

Thank you. We have rewritten the sentence as follows "For several years great effort has been devoted to the study of the spatio-temporal distribution of aerosol in summer 2010 over the ETR and Eastern Europe." and now it is on P2 L13-14.

- Pg 2, Line 33: Remove indentation. Join paragraph to previous one or give the Konovalov et al study it's own paragraph.
- 35

We gave Konovalov et al. (2011) it's own paragraph, moreover, we analyzed all previous studies in chronological order.

Also, you define AERONET here but have already used it in the second paragraph.

40 Thank you. AERONET and other acronyms have been spelled out the first time used.

Define ETR

ETR and other acronyms have been spelled out the first time used.

45

Pg 3 line 6: 'aerosol'

Thank you. Change made.

Pg 3, line 10: Elaborate on the 'synopic situation' – I assume this is meteorology driven definition.

5 Thank you for raising this issue. We replaced 'synopic situation' with 'weather conditions' all over the manuscript.

Pg 3, line 34: 'in-detail studies' – poor English.

The text has been changed to 'Our research contributes significantly to the above mentioned studies of Bovchaliuk et al. (2013)
and Milinevsky et al. (2014), but unlike them, we focused on a comprehensive evaluation of the impact of the fires in summer 2010 on the tropospheric aerosol load with a major focus on Ukraine' and is now in P4 L1-3.

Pg 4, line 1: 'meteorological situation' – poor English - informal.

15 We have reworked the sentence and replaced 'meteorological situation' with 'weather conditions'. Updated sentence looks as follows: 'We reproduced the weather conditions with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model'. P4 L4-5

The Introduction needs work:

20 Authors need to do a better job expanding the first paragraph emphasizing the impact of Russian wildfires and making that distinction between fires and wildfires. Authors need to review and answer:

- What made the Russian 2010 fires exceptional? - How did it start? - How did it evolve? - Why is it important to study? (don't just state it and who is this international scientific community?) - Justify why after seven years, another study of the Russian wildfires is necessary?

25

30

We completely agree and support the suggestion of Referee. We have reworked the Introduction entirely. At the beginning rewritten Introduction addresses the importance of wildfires as a global source of aerosol. Then we provide the description of importance of wildfires events during summer 2010.

The second paragraph explains the main reasons of occurrence of fires (extreme heatwave on summer 2010, e.g. Barriopedro et al., 2011; Schevchenko et al., 2014) caused by high temperatures and low relative humidity, e.g. Witte et.al, 2011.

- Later, we address to previous studies (in chronological order) devoted to aerosol dynamics over the European territory of Russia (ETR) and Eastern Europe. We have also highlighted the papers of aerosol research performed for the territory of Ukraine. At last, we have explained the contribution and novelty of our research in comparison with other works, its driving motivation and significance/necessity. We have completely withdrawn the usage of term 'scientific community'.
- 35 We have made suggested changes on P2 L2-P4 L11

More in-depth review of the Russian fires is required because at this point I don't know what notable contribution this study makes or why it is important to continue studying this one-time event.

- 40 We have addressed this issue in the second paragraph of the Introduction as follows: 'Extensive wildfires during summer (June 1-August 31) 2010 over the European Territory of Russia (ETR) and partly Eastern Europe were caused by an extreme heatwave, that led to an all-time maximum temperature record over numerous locations (Barriopedro et al., 2011; Dole et al., 2011; Demirtas, 2017), including the territory of Ukraine (Shevchenko et al., 2014). High surface temperatures (35-41°C) and low relative humidity (9-28 %) over those regions (Witte et al., 2011) favored the occurrence and persistence of fires. In turn,
- 45 those fires caused significant air pollution in populated areas of Russia and combustion products (gases and aerosol) were spread over large areas of Eastern Europe.'

Although it is very worthwhile to review the seminal papers that covered the Russian wildfire the authors do not go about it sequentially. Review each study separately – give them their own paragaph if necessary, tie them together by theme (satellite,

model and in-situ or aerosol parameter or other), then introduce your study.

We followed the suggestion of Referee and reviewed each study sequentially and separately. Gave for each study its own paragraph.

5

Twice authors have introduced their study in different parts of the introduction.

We have removed repeating text and introduced the contribution and novelty of our research in the last paragraph of the Introduction. Please, see rewritten version of Introduction on P2 L2-P4 L11 in the reworked version of the manuscript.

10

Pg 4, lines 12-14: metrology is a noun, not a verb. That entire sentence needs to be revised.

We have withdrawn 'metrology' from the sentence. Reworked sentence looks as follows: 'The basic principle of the network is to standardize the equipment, measurement techniques, and data processing, which are stored in a freely accessible centralized database'. P4 L16-17

Pg 4, line 20: missing parenthesis in Holben et al

Added parenthesis (Holben et al., 1998). P4 L24. Thank you.

20

15

Pg 4, line 23: you mean inversion algorithm.

We have completely withdrawn this sentence in the new version of manuscript.

25 Section 2.1 - I would like to have a map of the sites listed in Table 1, in which you use data. Context is required.

Thank you, we have inserted the map with the AERONET observational sites in Eastern Europe and Ukraine, which were used in our study. Please, see below Fig. 1.

30 Pg 5, line 2 – This is a repeat sentence. Authors already mentioned using L2 data.

We have deleted the repetition, thank you.

Sentence in Pg 5, lines 10-13 needs to be re-written.

35

We agree with the Referee and have rewritten the sentence as follows: 'Over land, aerosol properties are retrieved from spectral channels 0.47, 0.66, and 2.12 m. One of the primary aerosol products of the MODIS algorithms is the AOD at 550 nm in the atmosphere over land and ocean (Remer et al., 2005, 2008; Levy et al., 2007, 2010, 2013)'. P6 L7-9

40 Pg 5, line 15: We 'use' – stay on one tense.

We thank the Referee for highlighting this issue. We kept 'we used' as we have corrected the manuscript with the Past tense.

Pg 5, lines 17: what are the 'favorable weather conditions' ?

45

We have completely withdrawn this sentence. Thank you.

The title of section 2.4 is poorly written. It needs to be re-written

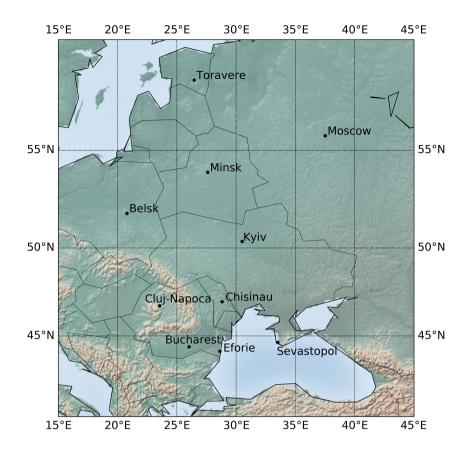


Figure 10. AERONET observational sites in Eastern Europe and Ukraine used in this study.

We agree with the Referee and have corrected the title to "Weather conditions and transport of air masses'. Thank you. P8 L13

Section 2.4 is the first time you mention the time period of your study. This needs to be mentioned in the introduction when you introduce your study.

5

We thank the Referee for this comment. We have added the time period (June 1-August 31, 2010) to the Abstract and Introduction.

Again, the English needs to be cleaned up in Section 3.

10

We agree with the Referee and improved the grammar and syntax in Section 3.

The following sentences are poorly written: "Significantly higher chance of AERONET site to be into the MODIS field of view and it provides opportunities for both instruments data comparison. In the following, first we directly compare MODIS AOD to sunphotometer AOD, followed by comparing the AOD of CALIOP to MODIS."

We have reworked Section 3 and moved description of methodology to corresponding subsections of Section 2. The sentences, mentioned by the Referee have been withdraw completely. Description of MODIS-AERONET comparison procedure is provided in Sect. 2.2 on P6 L 23.30. Description of CALLOR MODIS comparison procedure is provided in Sect. 2.3 on P7 L 22. P8

5 vided in Sect. 2.2 on P6 L23-30. Description of CALIOP-MODIS comparison procedure is provided in Sect. 2.3 on P7 L32-P8 L12.

Section 3: In the first paragraph you write 'We assess the influence of biomass burning during summer 2010 on aerosols over Ukraine and neighboring territories . . .' then in the last paragraph you again write 'To identify the impact of fires on air pollution by aerosols over Ukraine, including Kyiv . . .' . These two sound the same and repetitious.

We agree with the Referee that these repetitions are inappropriate and have withdrawn them completely. In the reworked version of manuscript the methodology is provided in Section 2.

15 The title of Section 3.1 is not precise. The Russian fires were a combination of forest, grass, and peat burning, as you previously mention. The title should either remove 'forest' or be precise and be 'forest, grass, and peat fires'.

We have made the change in title of Section 3.1 as follows 'Impact of wildfires and weather conditions during summer 2010 on aerosol air pollution in Eastern Europe'. Please, see P9 L1.

20

10

Again what is ETR?

The ETR has been spelled out the first time used. Thank you.

25 Again, I want to see a map of the locations of all cities in Table 1.

Thank you for the suggestion. We have inserted the map with the AERONET observational sites in Eastern Europe and Ukraine, which were used in our study. Please, see Fig.1 on P5.

30 Section 3.1: You refer to this 'synoptic process'. What is it? You mention meteorology quite a bit without actually explaining the weather conditions that set-up the ideal conditions for the wildfire to rage across the Eastern Europe.

Thank you for raising this issue. We replaced 'synopic situation', 'synoptic process' etc. with 'weather conditions' all over the manuscript.

35

In this section I would like authors to actually write about the meteorology instead of referring it in vague terms such as 'synoptic situation' (i.e. look at temperature, winds, pressure, RH or remove meteorology from the section since you don't talk about it).

40 That is the interesting idea. However, anomalies of surface T and RH for the territory of Western Russia (in our manuscript is the ETR) were shown by Witte et al., 2011. Moreover, Shevchenko et al., 2013 used maximum air T for 13 Ukrainian stations including summer 2010 to analyze heat waves events, which are defined as episodes with extremely high air temperature with duration of several days. That is why we have removed meteorology from the section and analyzed trajectories of air transport only.

45

Section 3.1 Last paragraph: Where do you see this change in synoptic processes on Aug 18-21?? You do not show a front or any changes in weather patterns. Are you quoting another study? These are grand conclusions without any data and results to back it up.

We agree that we have not been precise enough and we have improved the text as follows: 'According to the 5 monthly weather reports of the Ukrainian Weather Center, a change in weather was observed on August 18-21. Atmospheric fronts of an active cyclone which moved from the southern Baltic region to Samara led to a significant change of weather pattern in Eastern Europe. This change caused a distinct decrease in fire activities and a wet deposition of aerosol, lowering its content in the

5 atmosphere above all investigated regions in the second half of August'. P12 L5-8

Although, monthly weather reports of Ukrainian Weather Center are available only in Ukrainian and only after special request. Such a rapid change of weather is also seen in HYSPLIT back trajectories, calculated for the Kyiv site. We have added these trajectories and their description to the Supplement, Fig. S32-S34.

10

Table 1: What is the actual time period over which you calculate these values?

We have improved the caption of Table 1 to 'Level of air pollution caused by aerosol (AOD 500 nm) from June 1 to August 31, 2010 over the ETR and Eastern Europe according to AERONET' and improved the text on P9 L15-18 as follows 'We analyzed changes in AOD at 500 nm using all daily averaged measurements from June 1 to August 31 from the AERONET database 15 from Minsk (Belarus), Moscow (Russian Federation), Toravere (Estonia), Belsk (Poland), Moldova (Chisinau/Moldova), Cluj-Napoca, Bucharest, Eforie (all Romania), Kyiv and Sevastopol (both Ukraine)'.

Pg 9, line 2: The word 'apparently' should be removed. Many studies have already proven the fires caused the high air pollution 20 over Moscow – there's nothing apparent about it. Also replace 'combustion center' with what it actually is – fires. This term is not precise and can denote alternative combustion sources, such as anthropogenic.

We have removed 'apparently' and replaced 'combustion centers' with 'fires'. Thank you.

25 Pg 10, Back-trajectories show anti-cyclonic tendencies at most sites, other than Moscow and Toravere. The authors fail to notice the temporal and special extent of the stagnant anticyclone over Eastern Europe in their trajectory analysis.

We agree with the Referee and have addressed this issue on P9 L24-27 as follows: 'Our analysis of back trajectories revealed that air movements in the lower 5 km layer of the troposphere corresponded to anticyclonic circulation, which is seen in Fig.

30 4a-j at various altitudes as clockwise-shaped curves. The maximum AOD values from ten AERONET sites in the ETR and Eastern Europe were formed under conditions of air stagnation and accumulation of contaminants'.

The analysis in Section 3.1 is missing a large chuck of key information. I don't understand how the authors determine the height of the maximum AOD! Up to now, analysis has not been done to show where the peak in the aerosol profile is located to justify picking those heights. How do the authors determine these heights?

We thank for having drew our attention on this and agree that the reasoning was not precise enough. We make this point on P8 L24-25 as follows: 'We chose the lowermost 5 km tropospheric altitudes taking into account the analysis of the vertical distribution of aerosol according to CALIOP data (shown in Sect. 3.3.2).'

40

35

There are many places where it is written 'aerosols' when it should be singular. Double check.

Corrected to 'aerosol' all over the manuscript. Thank you.

45 Section 3.2

Pg 10, line 28: what is the 'period of observation'?

We have reworded the sentence as follows: 'In Table 2 we show a prolongation of the data record of Milinevsky et al. (2014, 2008-2013) for the Kyiv AERONET site by three more years up to the end of 2016'. P12 L15-16

Table 2: Now you are using a different AOD (440 nm) whereas before you were using 500 nm. What is the difference between using two different wavelengths? I think that needs to be explained. Are you using this wavelength to match with the

5 tween using two different wavelengths? I think that needs to be explained. Are you using this wavelength to match with the Milinevsky et al study?

We have reworked the Table 2 and recalculated AOD values for 500 nm to be consistent in our study. We do not use, show, or refer to AOD 440 nm in this manuscript any more. We also have extended Table 2 up to 2016 (all available level 2.0 data for the Kyiv AERONET site). Thank you.

Pg 11: Authors keep changing the description of the fires. Here is written 'active fires of vegetation'. Is it wildfires, forest fires, vegetation fires, combustion center, or fires of vegetation? Stick with one descriptor and be consistent. This is not an exercise in creative writing.

15

10

Thank you for the comment. We have corrected this issue and used 'wildfires' term throughout the entire manuscript.

Figure 12: Year is missing. Specific dates are mentioned in the text and it is hard to see on the x-axis. I would like authors to mark these dates referred to in the text.

20

We believe, Referee ment Fig. 4 on p.12. In the reworked version of manuscript this is Fig. 5. We have updated the capture of the Figure as follows: 'AOD 500 nm over Moscow and Kyiv (a), AE over Kyiv (b) during June 1-August 31, 2010'. We have also removed Fig. 5b as it was redundant. Thank you.

25 Pg 12, line 1: Use 'pronounced' rather than 'distinctive over Moscow', which was written incorrectly to begin with.

Corrected to 'pronounced'. P12 L19. Thank you.

Also, the next sentence needs to be re-written.

30

We have rephrased the sentence as follows: 'However, the aerosol pollution over Kyiv was also exceptional in comparison with the multi-annual average'. P12 L20.

Pg 13, sentence in lines 5-6: Are you referring only to June 1-2? Why bother? mid-June and the end of June show, by far, the lowest AE. You also show almost 40 observations for June 1 in Figure 5, so I don't know what authors are alluding to here.

We agree with the Referee's comment and we have moved the description of air transport each couple days within June-August 2010 to the Supplement. We believe that the precise analysis of transport of air masses within June -August 2010 is important, thus have not withdrawn it completely. By moving this piece of text to the Supplement we reduced the size of the manuscript but still kept the information available. We address to this issue on P12 L26-27 as follows: 'We provided detailed description

40 but still kept the information available. We address to this issue on P12 L26-27 as follows: 'We provided detailed description of AOD variations and the impact of air transport on those changes in the Supplement Fig. S1–S34'. We also have moved Fig. 5 to the Supplement.

Pg 13, first sentence in line 9: 'unstable weather, sometimes with clouds' – where are you getting this information? The language is also informal. For this paragraph what figures are you referring to that you're generating this analysis?? I shouldn't have to guess. I would like to see a graph of cloudiness and a precipitation index superimposed on the Figure 4c. The authors make numerous inferences to cloudiness with no supporting data.

We agree to the Referee that referring to clouds requires adding of supporting plots/data. Although, as we mentioned above, precise analysis of transport of air masses has been moved to the Supplement. As long as we do not mentioned these cloudy weather conditions in the reworked manuscript, we believe it is not necessary to add supporting data any more. Thank you.

5 Pg 14, line 3: Which figure are you getting the AOD at 500 nm value? What is the significance of switching between AOD's?

We have moved this analysis of transport of air masses to the Supplements. Also, we switched our analysis to AOD 500 nm completely. In the reworked manuscript Fig. 5a represents AOD 500 nm time-series over Kyiv and Moscow.

- 10 In Section 3.2 specific dates are being analyzed. I would like to (1) know which figures are being referred to and (2) a figure with the AOD (440 and 500) and the specific dates referred to in this section. Alternatively, a table will also work. This section is not easy to follow, particularly since the bulk of the analysis is located in the supplementary. Summarizing all this as a table should help.
- 15 We thank the Referee for his suggestion. We have moved the analysis of air transport during June-August 2010 to the Supplement. We have also switched our analysis of AOD to the wavelength of 500 nm and do not mix with 440 nm anymore.

Pg 14, sentence line 18-19: 'As seen from Fig 1a...' this sentence is imprecise and informal.

20 Thank you. We have moved this text to the Supplement and have corrected the sentence there as follows: "Fig. 1a shows that..."

Pg 14, line 33: '...different types of fires ...'. How do you know that?

We have moved this sentence to the Supplement and have substituted 'different types of fires' by fires'. Thank you.

25

Figure 6: you have the legend reversed: fine mode is the top curve/course mode is the bottom curve.

We have corrected the capture of Figure 6. Thank you.

30 What are the '3 periods' ? Are they written in (b) and (c). I shouldn't have to guess.

We have changed the caption of Fig. 6 on P16 as follows: 'Aerosol spectral SSA and microphysical properties from AERONET measurements at the Kyiv site during summer 2010: (a) – particle effective radius versus AOD for fine (left axis, top curves) and coarse (right axis, bottom curves) modes averaged for the entire summer 2010; (b) – particle size distribution, (c) – spectral

35 SSA, (d) – spectral RI, – real (right axis, top curves) and imaginary (left axis, bottom curves) parts. Representative periods are June 1-26 (triangles down), July 18-August 14 (triangles up), August 15-17 (circles), and August 18-31 (squares)'. Thank you.

Section 3.3

40 I would like to see a time series of AOD 500 and the periods of the pre-, active-, and post- fires that authors define.

Figure 5a provides AOD 500 nm time-series for the Moscow and Kyiv sites for the period June 1-August 31, 2010. We have decided to avoid the division of summer 2010 into pre-, active-, and post fires periods in order to simplify the understanding of the achieved results. Thank you.

45

How are these three fire periods different from Witte et al? Is there a time lag? How well-correlated is the onset, duration, and die-down of the fires over Russia compared to over Kyiv?

As we have mentioned in the comment above, we have avoided the division of summer 2010 into pre-, active-, and post- fires periods in order to simplify the understanding of our results.

Table 4: In the text you have resolutions of 10x10, 30x30, 50x50, however in the Table you have 10x10, 20x20, 30x30.

5

We thank the Referee for having drew our attention to this issue. In the reworked version of manuscript we have completely withdrawn Table 4 and avoided the analysis for different resolutions in order to simplify the understanding of the results.

I assume AODSph means the sub photometer - that should be clearly stated in the table and text.

10

We thank the Referee for having drew our attention on this. We have spelled out acronyms MODIS/Aqua AOD (AOD_{myd}), MODIS/Terra AOD (AOD_{mod}), and AERONET sunphotometer data (AOD_{Sph}) the first time used.

Table 7. Replace 'Time span X' in the first column with the actual time period.

15

We have completely withdrawn Tabe 7. Thank you.

Section 3.3.2

Pg 24, line 3-4: AOD > 0.5 are spotty – are they near urban areas? Is this due entirely to fires or a mix with urban signatures?

We thank the Referee for raising this issue. We have reworked Fig. 8. We have included CALIOP measurements during June 1-16 (Fig. 8a), when almost no fires burned as seen from Fig. 2a, b; and during August 4-19 (Fig. 8b), when the ETR and south-east of Ukraine were occupied by fires (Fig 2 g,h). In Fig. 8b we have also shown the location of aerosol profiles from Fig. 9

25 Fig. 9.

Figure 8a represents AOD spatio-temporal distribution in June 1-16, which was varying within 0-0.7. (Please, note that we have corrected from 0.5 to 0.7 in the reworked version of the manuscript). It resolves possible impact of local sources such as city transport, heavy industry, etc. Figure 8b shows high AOD values in south-east of Ukraine. We assume that during August 4-19 aerosol was dominating from the wildfires. Thus, we believe that the increase of AOD represents the impact of both local

30 sources and wildfires.

We have also shown in the reworked version of Supplement AOD 532 nm spatio-temporal variability from CALIOP in Fig. S36a-d: from June 17 to July 2 (a), from July 3 to 18 (b), from Jul 19 to August 3 (c), and during 12 day period from August 20 to 31 (d).

35

Pg 24, second paragraph: The June 1-July 18 CALIOP analysis – where are these data? I would like to see a similar Figure 8 but with this new period. Also, in this paragraph you say AOD is 1 however, in the previous paragraph for the same time period you say AOD > 0.5. There are two competing thoughts here.

40 As we have mentioned in the previous comment, we have reworked Fig. 8. We have included CALIOP measurements during June 1-16 (Fig. 8a), and during August 4-19 (Fig. 8b). The remaining CALIOP measurements are now part of the Supplement (Fig. S36a-c) in order to save the space of the manuscript.

We have reworked the sentence on P19 L2-3 as follows: 'In particular, during August 9-18 AOD values regionally exceeded 1 45 and reached a value of 2 at certain locations'.

Pg 25, line 8: 'particle concentration' – this is not the only place where authors erroneously use the plural.

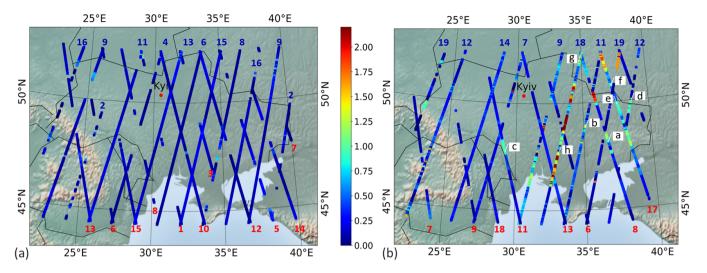


Figure 8. AOD 532 nm distribution over Ukraine from CALIOP measurements during 16 day period from June 1 to 16 (a), from August 4 to 19 (b), 2010. Red numbers at the bottom of the map indicate dates of each daytime track running to north-west and blue numbers at the top of the map indicate the date of each nocturnal track running to south-west. Labels a-h in Fig. 8b indicate the location of profiles, that are further analyzed and shown in Fig. 9.

We agree with the Referee and we have corrected to 'particle concentration' on P19 L17-18

Pg 25, line 21: Where is the Lugansk region in figure 8?

5 We have avoided the usage of city names in the reworked analysis to simplify the understanding of our results and avoid any confusions. We have corrected the sentence as follows: 'CALIOP's nocturnal track on August 12 ran over the east of Ukraine, where the highest AOD 532 nm of about 1.0 was found (location labeled 'd' in Fig. 8b)'. P21 L13-14

Pg 26, line 15: It is an exaggeration to allude to territories when authors really only did an in-depth analysis over Ukraine.

10

We agree to the Referee and have completely withdrawn the sentence.

Pg 26, sentence line 30-31: '... CALIOP measurements was rather challenging task due to fragmentation of data and their high variability.' Fragmentation of data implies it's faulty – what exactly do you mean? Also, why would highly variable data
be challenging. I think you mean simply is there isn't enough data comparable to AEROSOL and MODIS.

The Referee is completely right. We have withdrawn the entire sentence. Thank you for pointing this out.

In the discussion I would like to see a bullet form, or a concise summary list of the conclusions. I still don't know what they are or the value your study brings to this field. What are the key points of this study?

We have reworked this section with the main focus on our achievements. In the reworked section we highlight our main finding. Please, see rewritten version on P22 L4-P23 L32 and below:

²⁵ 'In this study we analyzed the influence of wildfires on aerosol dynamics over the ETR and Eastern Europe, in particular on air pollution conditions over Ukraine during an extreme heat wave event in summer 2010. Specific weather conditions with

high air temperature and low relative humidity (Witte et al., 2011) formed under anticyclonic circulation, which caused air stagnation and accumulation of contaminants. Moreover, those weather conditions were favorable for wildfires to evolve.

To reveal the connection between wildfires and aerosol properties over the ETR and Eastern Europe, we analyzed fire locations and their brightness temperature from MODIS measurements for the period June 1-August 31, 2010. We demonstrated

- that the fire activities increased from mid-July mostly over the ETR, Ukraine, and Moldova. The largest number and brightness 5 temperature of fires were observed during July 26-August 18. To consider the impact of those wildfires on aerosol dynamics over the ETR and Eastern Europe, we chose 10 AERONET sites in that region and computed HYSPLIT back trajectories to those sites. Our analysis of back trajectories showed that the observed AOD maximum over each of the considered sites was formed as a result of air transport from the areas of active wildfires. AOD maxima at the Belsk site (central Poland), Moldova
- 10 (Chisinau, Moldova), and Cluj-Napoca and Eforie (Romania) were caused mainly by fires in Ukraine and Moldova in July. AOD maxima over other AERONET sites were caused by aerosol from fires in the ETR. We also provided detailed analysis of aerosol dynamics over Ukraine. Despite the available studies of aerosol dynamics over Ukraine (Bovchaliuk et al., 2013; Danylevsky et al., 2011a, b; Milinevsky et al., 2014), we focused on the evaluation of the impact of the wildfires in summer 2010 on the tropospheric aerosol load, which has not been done before.
- 15 The ground-based AERONET measurements over the Kyiv site showed that for the entire observational period (from April 2008 to November 2016) the highest air pollution caused by aerosol was recorded in August 2010. The average AOD 500 nm in August 2010 exceeded multi-annual monthly mean (2008-2016, excluding 2010) by a factor of 2.2. We showed that during June 2010 the wildfires were not affecting AOD over the Kyiv site. Both, aerosol content and properties were determined mostly by local sources and air transport from Western Europe. In contrast, from July to mid-August, the AOD increase over the Kyiv site
- 20 was caused by air transport from the wildfire regions. The influence of fires resulted in an increased relative content of the fine mode in particles size distribution, accompanied by an increase of their effective radius (Fig. 6a,b). Occasionally the coarse mode also resulted in both an increase of AOD and a decrease of AE for days exhibiting a higher number of fires. We explained the predominant impact of fine mode aerosol on the AOD increase by its longer lifetime in comparison with the coarse mode.
- We also analyzed the impact of wildfires on aerosol spectral SSA at the Kyiv site during three different periods: 1) June 1-26, when the number of fires and their brightness temperature were low, 2) July 18-August 14, when the number of fires 25 significantly increased, and 3) August 15-17, when the highest AOD values were observed. Smaller SSA values during July 18-August 14 were likely caused by an increase of the soot content in the air, transported from the wildfires. SSA and RI spectral characteristics changed during that period, increasing the absorption capacity of aerosol, especially in longer wavelengths.
- During August 15-17 we observed relatively large SSA values. According to Eck et al. (2009), the observed increase of SSA 30 can explained by an increased particle size caused by wildfires, which in turn increased the total reflectance in the atmospheric column. Microphysical properties of aerosol over Kyiv under the influence of intense fires correspond well with general characteristics of biomass burning and polluted continental aerosol, as derived from AERONET sunphotometer measurements (Dubovik et al., 2002; Omar et al., 2005, 2009)...

Our comparison of AOD measurements from MODIS and AERONET showed strong functional relations between the

datasets with Pearson's correlation coefficients of 0.96 for MODIS/Aqua and 0.98 for MODIS/Terra. AOD measured by 35 MODIS is therefore well captured for the entire Ukraine. Both MODIS/Aqua and MODIS/Terra represent the aerosol content in the atmosphere over Ukraine for summer 2010 within measurement uncertainties of around 0.15 standard deviation of AOD. The spatial distribution of MODIS AOD revealed that the wildfires of summer 2010 significantly impacted the eastern, central, and southern parts of Ukraine. The AOD at 550 nm reached values of 2 (and more) at certain sites, especially in the 40 middle of August.

45

Our comparison of AOD between CALIOP and MODIS revealed that the correlation coefficient was not larger than 0.6 between datasets. Over Ukraine CALIOP mainly underestimated the AOD in comparison with MODIS for the entire summer 2010. This can be explained by findings of Kittaka et al. (2011), who showed that the CALIOP algorithm likely ignores tenuous aerosol, causing an underestimation of AOD in comparison with MODIS. They also found that the aerosol layer's base height can be detected at higher altitudes, leading to an AOD underestimation.

Despite the uncertainties and sparse availability of CALIOP measurements for the time and regions we analyzed, spatial distributions of AOD from CALIOP measurements over Ukraine corresponded well with those from MODIS, in accordance with Kittaka et al. (2011). Another advantage is that CALIOP also measures at nighttime. According to CALIOP observations,

the day- and nighttime AOD did not differ distinctly from each other during the analyzed period. This also corresponds to the global scale analysis of Kittaka et al. (2011).

We also analyzed aerosol profiles provided by CALIOP, which is the only source of aerosol vertical distribution for our study. We found that the aerosol was distributed at altitudes from about 40 m to 5 km and the extinction coefficient mostly ranged from some tenth to 1 km^{-1} , although sometimes it exceeded 8 km^{-1} in very dense plumes.

- 5 ranged from some tenth to 1 km⁻¹, although sometimes it exceeded 8 km⁻¹ in very dense plumes. Summarising, in this study we provided evidence of reasonable agreement between different types of aerosol measurements over Ukraine for the unique period in summer 2010. Further studies are needed to investigate the influence of the different fire regions on the air quality over Ukraine, which in our study could not be resolved well from the partly sparse coincidence of the datasets that are available until now. Not only other satellite instruments can be taken into account to further improve
- 10 the accuracy of pollution levels analysis. The expansion of the ground-based sunphotometer network and in particular the availability of in situ observations would help, for instance, to resolve the large spatial gradients of the pollution levels that have been found over relatively densely populated areas'.

Increased aerosols aerosol content in the atmosphere over Ukraine during summer 2010

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Abstract. In this paper we assessed the influence of one of the most important sources of aerosols aerosol in the atmosphere, biomass burning , during summer during forest fires throughout summer (June 1-August 31) 2010 on the its abundance, dynamics, and properties of aerosols over Ukraine, also considering over Ukraine. We also considered influences and effects over neighboring countries: ETREuropean Territory of Russia, Estonia, Belarus, Poland, Moldova, and Romania.

- 5 We used MODIS satellite instrument data were used to study fires distribution. Ground-based We also used ground-based remote measurements from the international sun photometers network AERONET and MODIS sunphotometer network AERONET plus MODIS, and CALIOP satellite instruments data were used to determine aerosols to determine the aerosol content and optical properties in the atmosphere over East Europe. HYSPLIT model was used to further Eastern Europe. We applied HYSPLIT model to investigate atmospheric dynamics and to simulate pathways of particles model pathways of particle transport.
- 10 We found that the highest air pollution aerosol content was observed over Moscow in the first half of August 2010, apparently due to the proximity of the most active combustion centers. Significant temporal dynamics of the aerosols fires. Large temporal variability of the aerosol content with pronounced pollution peaks during August 7–17 was observed at the Ukrainian (Kyiv, Sevastopol), Belarusian (Minsk), Estonian (Toravere), and Romanian (Bucharest) AERONET sites.

Acrosols We analyzed aerosol spatio-temporal distribution over Ukraine were constructed and analyzed using MODIS AOD 550 nm data validated by and further compared with the Kyiv AERONET site sunphotometer measurements, and ; we also compared CALIOP AOD 532 nm validated by with MODIS AOD data. Vertical distribution of aerosols We analyzed vertical distribution of aerosol extinction at 532 nm, retrieved from the measurements by CALIOP, were constructed CALIOP measurements, for the territory of Ukraine at locations where high AOD values were observed during intense wildfires. The influence of the fires. We estimated the influence of fires on the optical and microphysical properties of aerosol particles, such

20 as spectral SSA, size distribution, spectral single-scattering albedo and refractive indices, was analyzed and estimated and complex refractive indices using Kyiv AERONET measurements, performed during summer 2010.

In this study we show that the highest aerosols pollution showed that the maximum AOD in the atmosphere over Ukraine recorded on in summer 2010 was caused by particles transported particle transport from the forest fires in Russia. These Those fires caused the highest AOD 440 nm over 500 nm over the Kyiv site, which for in August 2010 exceeded the mean value for

25 the same month for the multi-annual monthly mean for the entire observational period by factor of two (2008-2016, excluding

2010) by a factor of 2.2. Also, the influence of fires resulted in a change of particles microphysical properties the particle microphysics in the regions , where the pollution was the highest.

1 Introduction

Biomass burning is well known to be a significant during wildfires is an important global source of trace gases and aerosols

- 5 aerosol in the atmosphere (Seiler and Crutzen, 1980; Crutzen and Andreae, 1990). For example, Barnaba et al. (2011) stated that wildfires (Wildfires as a mixture of peat burning and forest fires) significantly contribute contribute significantly to the fine mode (particles particle size < 1 µm) aerosols optical depth (AOD). The aerosol optical depth (AOD; Barnaba et al., 2011). According to Barnaba et al. (2011) "the largest numbers of these fires occur in Africa, Asia and South America, but a not negligible fraction also occurs in Eastern Europe and former USSR countries, particularly in the Russian Federation, Ukraine</p>
- 10 and Kazakhstan. In recent years, considerable attention by the international scientific community was paid to the impact on pollution of the aerosols from fires in central and western regions of Russia in summer 2010. Increased interest in these events is the result of the occurrence of the firesand.".

Extensive wildfires during summer (June 1-August 31) 2010 over the European Territory of Russia (ETR) and partly Eastern Europe were caused by an extreme heatwave, that led to an all-time maximum temperature record over numerous locations

- 15 (Barriopedro et al., 2011; Dole et al., 2011; Demirtaş, 2017), including the territory of Ukraine (Shevchenko et al., 2014). High surface temperatures (35-41° C) and low relative humidity (9-28 %) over those regions (Witte et al., 2011) favored the occurrence and persistence of fires. In turn, those fires caused significant air pollution in populated areas of Russia, and combustion products comprising gases and aerosols that are (gases and aerosol) were spread over large areas of Eastern Europe. Over the past few years, a significant amount of research has been done on the spatial and temporal distribution of aerosols.
- 20 For several years great effort has been devoted to the study of the spatio-temporal distribution of aerosol in summer 2010 over the ETR and Eastern Europe. The papers provided results on aerosol distribution and properties and their impact on air quality in the atmosphere over the area occupied by fires and neighboring territories, are

Konovalov et al. (2011); Witte et al. (2011); Péré et al. (2014); Chubarova et al. (2012).

- Chubarova et al. (2012) analyzed aerosols properties and radiative effects during 2010 fire events in Central Russia according to ground-based measurements AERONET in Moscow and Zvenigorod, as well as radiative measurements with a WMO calibrated pyranometers located in Moscow. An extremely high AOD was observed in Moscow and its suburbs on August 6-8, with an absolute maximum at 500 nm on August 7 reaching 6.4 at Moscow and 5.9 at Zvenigorod. Spatial distribution of AOD has been retrieved for these dates using MODIS satellite data. Chubarova et al. (2012) analyzed how smoke affects the key aerosols properties, such as particle size distribution, refractive index (RI), SSA, and the phase function asymmetry
- 30 factor. These quantities, measured during fire events, were compared 1) with those obtained earlier in-between the fires in same regions and 2) with analogous aerosol properties determined during wildfires but at other regions of the globe. They also assessed the impact of wildfire products on the incoming solar irradiance in different ranges of the optical spectrum, including UV-range, and estimated the radiative forcing (RF) at the top of the atmosphere (TOA). Chubarova et al. (2012) also explored

the impact of intense fires on the environment. Significant change in the atmospheric gas composition, aerosols concentration, and air temperature caused detrimental influence on human health. Particularly the mortality rate increased by 1.5-1.6 times in the central region of ETR during summer 2010 (Chubarova et al., 2012). For example, Konovalov et al. (2011) analyzed the evolution of near-surface concentrations of carbon-monoxide(CO), PM₁₀ and ozone (O₃)-in Moscow region during the

- 5 summer 2010 heat wave by comparing available by comparing ground-based and satellite measurements with mesoscale model simulations. They used a the modified version of the CHIMERE chemistry transport model (multi-scale chemistry-transport model for atmospheric composition analysis and forecast CHIMERE, http://www.lmd.polytechnique.fr/chimere/) to include wildfire emissions of primary pollutants and the shielding effect of smoke aerosols on photolysis. They used measurements, which were collected at automated monitoring stations with a nominal measurement frequency of three measurements per
- 10 hour (in Zvenigorod, Zelenograd, and Pavlovskii Posad). Fire radiative power (FRP) fire radiative power data retrieved from the measurements performed by Moderate-Resolution Imaging Spectrometer (MODIS) on board of National Aeronautics and Space Administration (NASA) Aqua and Terra satellites were used to study spatial and temporal variability of fire activities. AOD measured by MODIS at to study the spatio-temporal variability of the fires. They also used MODIS AOD 550 nm was also used to correct a negative bias in FRP-fire radiative power measurements in case of if fires obscured by heavy smoke.
- 15 Konovalov et al. (2011) also analyzed SSA retrieved from AErosol RObotic NETwork (AERONET) measurements performed in Moscow during the period of major fire events in summer 2002. It was found that the They found that "... extreme air pollution episodes in Moscow were mainly caused by fires taking place at relatively short distance range (less than 200 km) from the measurement siteMoscow; the transport of air pollution to Moscow from more distant fires was found rather in-significant. They also showed less significant. It was also found that a compensation of a possible negative bias in the measured FRP
- 20 radiative power from fire obscured by heavy smoke is a crucial procedure condition for a good performance of the model". These events Active fires during summer 2010 influenced the content and characteristics of aerosols properties of aerosol in the atmosphere not only above burning areas but also over most the most territory of Eastern Europe, where the fires were not as strong as in the ETR and eastern Ukraine. The tropospheric dynamics of aerosols pollution over these territories has been aerosol pollution over those territories was studied by Witte et al. (2011) by means of using ground-based and satellite
- 25 observations. They applied data from satellite instruments MODIS and OMI (Ozone Monitoring Instrument (OMI) in the area between 45–63° N and 23–63E (including territory over ° E (covering the territory of Ukraine). They studied the spatial and temporal spatio-temporal distribution and energy characteristics of burning cells using MODIS fire products and AOD at 550 nm, and OMI aerosol index (AI) at 354 nm, AOD , and SSA (and single scattering albedo (SSA), both at 388 nm). Their results correlate correlated well with the development of synoptic situation weather conditions determined by back-trajectory
- 30 simulations with the starting point in Moscow. Meteorological parameters, such as They determined vertical temperature profiles, pressure, humidity, and wind direction were determined directions according to radiosonde measurements over the ETR. Witte et al. (2011) showed that the aerosol pollution of the atmosphere over the Ukrainian territory was significantly lower in comparison with western and central regions of the ETR.

The aerosol shortwave direct RF Chubarova et al. (2012) analyzed aerosol properties and radiative effects during the 2010
fire events in central Russia according to ground-based measurements of AErosol RObotic NETwork (AERONET) in Moscow

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and Zvenigorod, as well as radiative measurements with World Meteorological Organization (WMO) calibrated pyranometers located in Moscow. They showed that an extremely high AOD was observed in Moscow and its suburbs on August 6-8, with an absolute maximum at 500 nm on August 7, reaching 6.4 at Moscow and 5.9 at Zvenigorod. They retrieved spatial distribution of AOD for those dates using MODIS satellite data. Chubarova et al. (2012) analyzed the way smoke affected aerosol particle

- 5 size distribution, refractive index (RI), SSA, and the phase function asymmetry factor. Those quantities, measured during fire events, were compared 1) with those obtained earlier in-between fire events in the same regions and 2) with analogous aerosol properties determined during wildfires but at other areas of the globe. They also explored the impact of intense fires on the environment. A significant change in the atmospheric gas composition, aerosol concentration, and air temperature caused detrimental influence on human health. Particularly the mortality rate increased by 1.5–1.6 times in the central region of the
- 10 ETR during summer 2010 (Chubarova et al., 2012).

Péré et al. (2014) described the aerosol shortwave direct radiative forcing during the peak of the 2010 Russian wildfires and their feedbacks impact on air temperature and atmospheric dynamicswas described by Péré et al. (2014), who used. They applied the CHIMERE offline coupled to the meso-scale WRF (mesoscale Weather Research and Forecasting) community model. This paper analyzes The authors analyzed the impact of fine aerosol particles (10 nm–5 µm) on shortwave (0.2–6.0 mm6

- 15 µm) solar radiation. Aerosol direct RF They simulated aerosol direct radiative forcing and feedbacks in the atmosphere were simulated for the period August 5–12, 2010 on the basis of from observational data obtained with ground-based AERONET supphotometers, and POLDER (POLarization and Directionality of the Earth's Reflectances) and CALIOP ((POLDER), as well as Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) satellite instruments. They analyzed AOD, SSA, and asymmetry factor of aerosolparticles were analyzed. Vertical distribution of aerosols was determined. They also determined the
- 20 vertical distribution of aerosol from the vertical profiles of CALIOP extinction coefficient measurements at 532 nm wavelength. Strong The authors detected strong perturbations of atmospheric composition over Russia were clearly detectable from these various from those data. Moscow was subjected to an important aerosol radiative effect, especially during the arrival of the aerosol plume on August 6–10.

The above-mentioned studies are focused in Moscow and other Russian areas. Satellite data and modeling studies hardly cover adjacent territories, including Ukraine. Only a few studies explored aerosol properties and aerosol impacts over Ukraine during severe wildfire events. Witte et al. (2011) provided AI, SSA, AOD 550 nm data averaged over the period of active fires (from July 22 to August <u>Despite previous studies focused on the summer</u> 2010), including the domain of latitudes 45–52N and longitudes 23–33E, where Ukraine is located. These data show significantly lower aerosol pollution of the atmosphere over Ukraine in comparison with western and central regions of ETR, although MODIS observed fire cells in

- 30 the eastern part of Ukraine during the same period. According to Péré et al. (2014), AOD 550 nm measured by POLDER and modeled for particular days of August 5–12, fires in the ETR and Eastern Europe, little attention was payed to aerosol impact over Ukraine. Which, as we show in this study, was also influenced by severe fires. Moreover, thorough studies on aerosol layer properties over Ukraine pending further improvement. To the authors best knowledge, very few publications can be found regarding this topic. For example, Danylevsky et al. (2011a), Danylevsky et al. (2011b) analyzed aerosol layer properties
- 35 from AERONET/PHOtométrie pour le Traitement Opérationnel de Normalisation Satellitaire (PHOTONS) ground-based

measurements over one of the assessments of RF and air pollution impact on the near-surface air temperature over Ukraine are much lower than over central regions of ETR. However, in other previously published studies by means of largest Ukrainian city, Kyiv during 2008–2009. Bovchaliuk et al. (2013) and Milinevsky et al. (2014) analyzed aerosol variability, its seasonal dynamics and the load of anthropogenic aerosol over the industrial areas over Ukraine using POLDER satellite and AERONET

- 5 ground-based measurements(Bovehaliuk et al., 2013; Milinevsky et al., 2014) was reported a marked increase of aerosol content in the atmosphere over Kyiv and some regions of Ukraine during summer 2010 in comparison with other years.
 Bovehaliuk et al. (2013) showed that the maximum measured AOD at 870 nm over the analyzed region (40–60° N and 20–50° E) during 2003-2011 was observed in summer 2010 due to the transport of aerosol from Russian wildfires.
 Milinevsky et al. (2014) analyzed seasonal variations of the aerosol load for the period 2008-2013 over eight Ukrainian cities
- 10 (including Kyiv) and Belarus (only Minsk). They also found a smoke particle induced increase of AOD 440 nm over the Kyiv site that could be traced back to biomass burning in the ETR in August 2010.

In this paper, we present results of in-detail studies of tropospheric aerosol pollution over Ukraineduring summer 2010. We use available data of Our research contributes significantly to the above mentioned studies of Bovchaliuk et al. (2013) and Milinevsky et al. (2014), but unlike them, we focused on a comprehensive evaluation of the impact of the fires in summer 2010

- 15 on the tropospheric aerosol load with a major focus on Ukraine. We used data from ground-based AERONET sunphotometer measurements , satellite data from MODIS and satellite measurements from MODIS (Aqua and Terra) and CALIOP instruments. The meteorological situation was reproduced from air mass back trajectory simulations with HYSPLIT We reproduced the weather conditions with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. We estimate computed trajectories of aerosol polluted air to the 10 AERONET sites in the ETR, Eastern Europe, and Ukraine in the lowest
- 20 5 km tropospheric layer. We also estimated the spatio-temporal influence of extensive biomass burning in wildfires in the ETR and Eastern Europe on air pollution caused by aerosols aerosol over Ukraine and neighboring areas, which was not the focus of above mentioned studies. We also evaluate available data of aerosols properties and content in the atmosphere, measured by the ground-based (AERONET) and satellite (MODIS , CALIOP) instruments. Combined measurements of those events of AERONET sunphotometer in Ukraine and the two satellite instruments MODIS and CALIOP have not been analyzed before.
- 25 Also, in contrast to earlier studies, we provided deeper insight into aerosol properties other than AOD and the vertical structure of the relevant tropospheric aerosol layers.

2 Data-Methods and data sources

2.1 AERONET data

Among all available methods of remote sensing of aerosols properties in the atmosphere, the most reliable data is derived
 from ground-based measurements of direct solar irradiation and sky radiation by automatic sunphotometers AERONET. This network The automatic sunphotometer network AERONET was founded by NASA and the PHOtométrie pour le Traitement Opérationnel de Normalisation Satellitaire (PHOTONS)PHOTONS; Lille1 University, National Centre for Space Studies CNES(CNES), and the National Center for Scientific Research of France CNRS (CNRS) and includes several hundreds

of AERONET sites over the world. The basic principle of the network is to standardize and metrology the equipment, the technique of measurement and processing of data measurement techniques, and data processing, which are stored in a freely accessible centralized database. Description The description of instruments and measurements procedures, data processing, calibration of supphotometers, the accuracy of measurements, and terminology are described in Holben et al. (1998) and in

- 5 the documentation from AERONET web page (http://aeronet.gsfc.nasa.gov/). According to the AERONET classification, all observed data are divided into 3 levels: Level 1.0 primary unscreened data, Level 1.5 cloud-screened data, Level 2.0 highest accuracy data, cloud-screenedand, quality-controlled and corrected for sunphotometers recalibration. We used Level 2.0 data in our research. Spectral The spectral AOD in the atmospheric column over the observational site is determined from direct solar irradiance measurements with errors of ±0.01 in the visible and near-infrared regions of the spectrum and with a
- 10 larger uncertainty (±0.02) in the UV band Holben et al. (1998)ultraviolet band (Holben et al., 1998). The Angstrom exponent (AE) is defined in the range of 440-870 nm , according to the AERONET algorithm. In case of successful measurements of the sky radiance along almucantar and altitude circle of the Sun the properties of aerosol particles such as size distribution , SSA, RI, and phase function are retrieved by inverse solution (Dubovik et al., 2000; Dubovik and King, 2000)also determined by the AERONET algorithm for the sunphotometer spectral range 340-870 nm from direct sun irradiance measurements.
- 15 We used AE to interpolate the AOD on the required wavelength. We applied AE determined for 440-870 nm because it is suitable to the aerosol size distribution during wildfires when relative dominance of the fine mode particles takes place (Eck et al., 1999; Holben et al., 2001).

There were only two AERONET sites in Ukraine in 2010: Kyiv (50N, 30E)and Sevastopol (44N, 33E). In this paper, we used data mostly from Kyiv site, because of predominant continental aerosol over most of Ukraine and Russia, while over

- 20 Sevastopol the essential component of aerosols is sea salt as To assess the extent of the impact of wildfires in summer 2010 we used data from the following Eastern European AERONET sites (also shown in Fig. 9): Minsk (Belarus), Moscow (Russian Federation), Toravere (Estonia), Belsk (Poland), Moldova (the official name of the site is Moldova, although it is located in the coastal zone of Crimea. To assess the extent of the impact of forest fires Chisinau, Moldova), Cluj-Napoca, Bucharest, and Eforie (all Romania). We also used data from the only two Ukrainian sites that measured during summer 2010: Kyiv and
- 25 Sevastopol.

To analyze aerosol dynamics over Ukraine during the Russian wildfires in summer 2010we also used data of neighboring Eastern European AERONET sites located in the countries, we used AERONET data mostly from the Kyiv site. Measurements of aerosol properties over Kyiv by AERONET sunphotometers started at the end of March 2008 (Danylevsky et al., 2011a, b)(Danylevsky et al., 2011a). The Kyiv PHOTONS/AERONET site is located at The the Main As-

- 30 tronomical Observatory of the National Academy of Sciences of Ukraine at Golosiiv forest situated at the south in the Golosiiv forest located in the southern part of Kyiv, at a distance of approximately 10 km from the city center. From 2008 until 2013, the site was equipped with the sun photometer CIMEL CE-318-2 polarized model with the filters of 440, 670, 870, 936, and 1020 nm. The surrounding landscape around the Kyiv site enables the sun photometer to scan celestial hemisphere completely sunphotometer to scan the entire celestial hemisphere. Data are sent to AERONET database twice a day and are
- 35 accessed and displayed on the Kyiv AERONET website website at the same day. Data observed during summer 2010 are



Figure 9. AERONET observational sites in Eastern Europe and Ukraine used in this study.

of Level 2.0 quality (cloud-screened and quality-controlled data). Up to 2013 the site was equipped with CIMEL CE-318-2 sunphotometer polarized model with filters 440, 670, 870, 936, and 1020 nm. We obtained AOD 500 nm data for the Kyiv site by data interpolation using Angstrom formula in the range of 440–870 nm. Similarly we obtained AOD 550 nm for the Kyiv site by data interpolation using Angstrom formula in the range of 440–675 nm. We calculated AOD values at different wavelengths to be further consistent with measurements, performed by different types of sunphotometer or satellite measurements.

2.2 MODIS data

5

Two nearly identical MODIS instruments are installed onboard the Terra (EOS AM-1) and Aqua (EOS PM-1) satellites. Terra (http://terra.nasa.gov/about/) flies in a sun-synchronous, near-polar, circular orbit with an inclination of 98.5° at an altitude of 705 km every 98 minutes (16 orbits per day). The local equatorial crossing time is approximately 10:30 a.m. in

10 a descending node. The Aqua satellite belongs to the afternoon A-Train constellation (http://atrain.gsfc.nasa.gov). The local equatorial crossing time is approximately 1:30 p.m. in an ascending node of a sun-synchronous, near-polar, circular orbit. MODIS data are widely used for researching the Earth's atmosphere, surface, and biosphere. To retrieve aerosol properties

over land measurements at the The MODIS instruments supply data used to study the Earth's surface and atmosphere from local to global scales. Over land, aerosol properties are retrieved from spectral channels 0.47, 0.66, and 2.12 µmare used. From these data the following parameters are calculated with the application of appropriate algorithms : AOD at spectral channels at 0.47, 0.55, and 0.66 µm wavelength, the relative amount of fine mode aerosol particles. One of the primary aerosol

5 products of the MODIS algorithms is the AOD at 550 nm , and the Angstrom component in the spectral range of 0.47–0.66 μm (Remer et al., 2005; Levy et al., 2007, 2013) in the atmosphere over land and ocean (Remer et al., 2005, 2008; Levy et al., 2007, 2010, 2013).

We used MODIS/Aqua and MODIS/To estimate atmospheric pollution over Ukraine caused by aerosol from wildfires, we used AOD 550 nm retrieved by the land algorithm and collected in the MODIS Aqua and Terra Level 2 Collection 005 and 051

10 Optical_Depth_Land_And_Ocean product retrieved by the C005 aerosol over-land algorithm, averaged over 10 km × 10 km cells in nadir and approximately 12 km × 12 km at the edge of the field of view. Due to the wide field of view, which equals to ±55(approximately 2300 km on the surface) cross track, MODIS can obtain observational data daily on each point of the surface in the mid-latitudes during favorable weather conditions.

We applied MODIS AOD data with 2 main purposes: 1)to compare with ground-based AERONET measurements over Kyiv

- 15 site, and 2) to estimate atmospheric pollution over Ukraine caused by aerosols from wildfires. Although file (see https:// modis-images.gsfc.nasa.gov/_docs/ATBD_MOD04_C005_rev2.pdf, https://modis-images.gsfc.nasa.gov/MOD04_L2/format. html). The documentation of the MODIS Level 2 Collection 051 Optical_Depth_Land_And_Ocean Scientific Data Set (SDS; Levy et al., 2010) dataset (Levy et al., 2009) does not recommend the application of these data for quantitative analysis, we applied them. Nevertheless we applied these data because the alternatively recommended SDS-dataset
- 20 Corrected_Optical_Depth_Land contains only a very small amount of data in the regions at time period we consider in our for the period we considered in this study. Earlier studies applying based on the Optical_Depth_Land_And_Ocean product already showed good agreement between AOD of MODIS (Quality Assurance Confidence flags 1, 2, 3; QAC) and AERONET on global scale (Levy et al., 2010; Remer et al., 2008; Bréon et al., 2011). Therefore we apply here (Remer et al., 2008; Levy et al., 2010). Local biases at special cases of aerosol pollution, such as wildfires, were reported by Levy et al. (2010). At this stage we
- 25 applied all data over the land with QAC 1, 2, 3 from this SDS dataset without any additional filter. We also following Levy et al. (2010) and all AOD ranged from -0.05 to filtering. For comparably clean atmospheric conditions, when AOD is close to zero, AOD values in the range ±0.05 consider equal to 0. are practically indistinguishable (Remer et al., 2005, 2008). Following Levy et al. (2009), we set corresponding data in this range to zero.

We compared MODIS Collection 5 data with AERONET data to further examine potential regional peculiarities in the
satellite measurements. To define spatial collocations we computed distances and azimuth angles between the centers of each
MODIS image pixel and the location of the Kyiv AERONET site. To compare AOD from MODIS and AERONET we averaged
the MODIS AOD over the pixels area centered on the Kyiv AERONET site. Although we did not calculate the spatial biases of
the AoD over this area. This simplification of the Ichoku et al. (2002) procedure is acceptable because MODIS images of land
do not exceed an area of 50 × 50 km, which is significantly less than the characteristic dimension of inhomogeneities in the

spatial distribution of aerosol in the atmosphere (Anderson et al., 2003). In addition, if several AERONET AOD measurements were available, we chose only one, performed at the closest time to the satellite measurement (not more than 30 minutes).

The MODIS mission also provides fire mapping on the a land surface, which we used to evaluate the activity of fires and their spatial distribution, similar similarly to Witte et al. (2011). For detecting fires and defining their characteristics, two

- 5 spectral channels (4 and 11 μm) are used by the MODIS data processing algorithm (Justice et al., 2002)uses two spectral channels (4 and 11 μm; Justice et al., 2002). Combustion temperature (in K) is defined from the measurements of the spectral brightness of the flamewith the application of the , applying the Stefan-Boltzmann law. A special An algorithm is applied to calculate the number of burning cells and the total area occupied by fires. Data of the spatio-temporal distribution of fires are freely available on the Internet () with the e.g. https://firms.modaps.eosdis.nasa.gov/map/) with support of NASA. To
- 10 visualize the <u>spatial</u> distribution of fires we used Fire Information for Resource Management System (FIRMS). With this tool, active fires are detected in each pixel with a size of approximately 1 km. For more information on algorithms on fire recognitionand evaluation of their intensity, the data supplied by the University of Maryland (ftp://fuoco.geog.umd.edu). We applied high-confidence (confidence level of calculated fire pixels is larger than 80 %) brightness temperature of fire pixels (at band 21). More information about the algorithms of fire recognition, principles of archiving and distributing data can be
- 15 found in Schroeder et al. (2008); Davies et al. (2009); Justice et al. (2011). In most cases, MODIS detects vegetation fires but sometimes also MODIS sometimes detects volcanic eruptions or flares of gas as well. Herewe interpret the results of observations under conditions of forest fires and other vegetation or peatin addition to vegetation fires. Here, we considered all observed signals as wildfires.

2.3 CALIOP data

- 20 CALIOP flies is a two-wavelength (532 and 1064 nm) polarization lidar providing high-resolution vertical profiles of aerosol and clouds. It is installed onboard the NASA Cloud-Aerosol Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite, which belongs to the afternoon A-Train constellation. CALIOP is a two-wavelength (532 and 1064 nm) polarization lidar and provides high-resolution vertical profiles of aerosols and clouds. A detailed description is provided on its official website (http://www-calipso.larc.nasa.gov/about/) and in Winker et al. (2003); Hunt et al. (2009)as well as
- 25 in Winker et al. (2003) and Hunt et al. (2009). CALIOP uses three receiver channels: one measures the 1064 nm backscatter intensity and two channels measure orthogonally polarized components of the 532 nm backscattered signal. The laser beam is directed almost in nadir, at nadir with a slight tilt forward forward tilt in the direction of motion of the satellite to avoid direct reflection of laser radiation from high reflectivity objects (surface water, snow, etc.). The divergence of the transmitted laser beam equals 100 µradand. This results in the footprint at on the Earth's surface, called in our paper ground track,
- 30 of about 70 meters. A pulse repetition rate of approximately 20 Hz provides vertical resolution of 15 m. The application of special algorithms to the CALIOP measurements of the vertical profile of the atmosphere backscatter coefficient enables to derive aerosol particles and clouds and to define vertical profiles of aerosol extinction for two wavelengths (532 and 1064 nm). Corresponding

CALIOP measurements allow to derive the vertical distribution of aerosol and clouds. The corresponding AOD is determined by extinction coefficient integration over altitude (Vaughan et al., 2009; Winker et al., 2009; Young et al., 2013; Omar et al., 2009). Vertical (Omar et al., 2009; Vaughan et al., 2009; Winker et al., 2009; Young et al., 2013). For this study we used both parameters, vertical distribution of the extinction coefficient and AOD at 532 nm, defined along the path of the sub-satellite pointwere used

5 for this study. We used . We applied Level 2 Cloud and Aerosol Layer and Profile products V 3.01 and 3.02 with a resolution of 5 km on the surface along the sub-satellite point, see (CALIPSO Quality Statements Lidar Level ...

A comparison of CALIOP AOD with ground-based AERONET observations can be challenging because of different measurement characteristics of both instruments. The CALIOP lidar provides only fragmentary data on aerosol along CALIPSO satellite's ground track due to the small size of its light beam and cloudy conditions that frequently occur. As long as the

- 10 instrument's orbital period lasts 98 minutes, ground tracks of satellite consecutive passages at certain latitudes are shifted 24.5° to the west, making its spatio-temporal coverage rather sparse. Consequently, the probability of CALIOP to pass over the atmospheric column observed by the solar photometer AERONET is rather limited (e.g. Redemann et al., 2012). During the three summer months of 2010 we found no coincident CALIOP-AERONET measurements over Kyiv, apart from the single collocation, although not exactly matching the selection criteria according to Omar et al. (2013). In this particular case the
- 15 closest CALIPSO ground track was found 60 km eastwards from the Kyiv AERONET site.

Therefore, in this study we only compared CALIPSO/CALIOP CloudAerosol Layer Product AOD 532 nm (Winker et al., 2009, 2010) with MODIS/Aqua AOD 550 nm since the orbits of both satellites are in the A-Train afternoon constellation (http://atrain.nasa.gov/). The relative positions of CALIPSO and Aqua satellites in the A-Train provide a large number of practically simultaneous measurements with the time span of 2 Cloud and Aerosol Layer Products, Version Releases:

- 20 3.01,3.02; minutes, while the spatial difference is only about 10 km. Each granule of MODIS data consists of consecutive scans across the satellite track. The footprint of CALIOP light beam on this granule looks like a sequence of points on the straight line, which are passing close to the center of a granule. Each of these points represent the center of the CALIOP measurement averaged over 5 km which matches with one of the pixels of a MODIS granule. To find these matches we calculated the distances and azimuth angles between the center of each CALIOP point and the center of each pixel in MODIS granule in
- 25 the same manner as for the MODIS-AERONET case, described in Section 2.2. We averaged MODIS data over areas 50 km × 50 km, while CALIOP data in the CALIPSO CloudAerosol Layer Product are averaged on various distances along the satellite ground track, up to 80 km (see CALIPSO Quality Statements +Lidar Level 2 Cloud and Aerosol Profile ProductsLayer Products, Version Releases: 3.01, 3.02). We did not apply any correction for potential spectral differences while comparing CALIOP AOD 532 nm and MODIS/Aqua AOD 550 nm. It yields to an estimated systematic bias in our AOD comparison
- 30 of approximately 2–6 % in the AE range between 0.5 to 1.8 (see Fig. 5b) and can be neglected in our cases, following Kittaka et al. (2011).

2.4 Meteorological data Weather conditions and the means transport of study of air massestransport in the investigated region

To analyze the impact of weather conditions on the distribution of aerosols aerosol in the atmosphere during June-August summer 2010we used synoptic maps from, we used weather charts from the Ukrainian Weather Service on of isobaric surfaces

- 5 representing altitudes of 2 m, 1.5, 3, and 5 km - Synoptic maps of international research centers were also used, namely maps of (not shown in this paper). We also considered weather charts namely at 500hPa/surface pressure maps from 1015 hPa of the Global Forecast System (GFS,) and; http://www.wetterzentrale.de) from the National Centers for Environmental Prediction (NCEP) and merged Global and European model (GME; http://www.wetter3.de) from the German Weather Service().
- To confirm the analysis of synoptic charts we also calculated trajectories of air masses using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT-4HYSPLIT; http://ready.arl.noaa.gov/HYSPLIT.php), developed 10 by the National Oceanic and Atmospheric Administration's (NOAA) Air Resources Laboratory (ARL: Draxler and Hess, 1998; Bowman et al., 2013). HYSPLIT is freely available software (). According to (Draxler and Hess, 1998; Bowman et al., 2013). Furthermore, to study the aerosol inflow towards AERONET sites shown in Fig. 1, we computed back trajectories of air masses at different heights for dates of maximum measured AOD at each of
- those sites. Back trajectories were simulated for 168 hours (7 days) at altitudes of 500 m, 1.5, 3, 4, and 5 km at 12:00 GMT 15 starting time. We chose the lowermost 5 km tropospheric altitudes taking into account the analysis of the vertical distribution of aerosol according to CALIOP data (shown in Sect. 3.3.2). According Stohl (2002), the uncertainty of calculated trajectories is \pm HYSPLIT trajectories for a period longer than 24 hours is around 20 % in the horizontal direction for the time period more than 24 hours in the free troposphere, and; after 120 hours the uncertainty increases to about 400 km in the horizontal and 20
- about 1300 m in the vertical planes.

3 Methodology Results and results Discussion

We assess the influence of biomass burning during summer 2010 on aerosols over Ukraine and neighboring territories according to ground-based and satellite measurements and analyze the synoptic-scale situation by modeling air mass trajectories. To estimate the impact of biomass burning, we analyze data from several AERONET sites located in Eastern Europe. From all

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available Ukrainian AERONET stations, continental aerosols properties were measured only in Kyiv in 2010. To analyze aerosols properties over other regions of Ukraine we used MODIS data for assessing column AOD and CALIOP data for assessing column AOD and vertical distributions of aerosol extinction.

To prove our results, we also validate satellite data by comparing with ground-based AERONET measurements. The best option is when all instruments would measure optical properties of the same air mass at the same time. Since the track of

CALIOP laser beam runs across the field of view of MODIS/Agua, both devices are scanning the same air mass. However, 30 because of differences in the fields of view of satellite instruments, in particular the small field of view of CALIOP, not many collocations are found for our comparison. Because of the Earth's rotation the trajectory of the sub-satellite point on the Earth's surface shifts to the west approximately by 24.5(e.g. about 1880 km for the latitude 50) during CALIPSO orbit period. Since the satellite track runs every 16 days (233 satellite rotations) along the same route the probability of the CALIOP to pass over the atmosphere column observed by the solar photometer AERONET is very low (the column radius on the earth's surface is about 10-15 km depending on the effective height of the aerosol layer and the season). Significantly higher chance of AERONET site to be into the MODIS field of view and it provides opportunities for both instruments data comparison. In the following, first we directly compare MODIS AOD to sunphotometer AOD, followed by comparing the AOD of CALIOP to MODIS.

5

To identify the impact of fires on air pollution by aerosols over Ukraine, including Kyiv, we analyzed the transport of air masses entering the Kyiv region in summer 2010 by calculating back trajectories of air masses during 7 days at different altitudes. To identify aerosols sources and contributions from dynamics in the atmosphere, we analyzed aerosols profiles at observational sites along the air mass trajectories. Freely available CALIOP observations are the only source of such

information for areas under consideration. Since only a few profiles match collocation criteria, we examined vertical aerosols 10 profiles and associated characteristics for the whole summer in 2010.

3.1 Impact of forest fires wildfires and weather conditions during summer 2010 on aerosols aerosol air pollution in **Eastern Europe**

To analyze the impact of forest fires wildfires on air pollution caused by aerosols in Fig. 1 we plotted fire distributions and total

- radiation intensities over Eastern Europe aerosol, we plotted distributions and the ETR brightness temperature of the fire pixels 15 over the ETR and Eastern Europe (40–65°N and 10–60°E) in summer 2010 as observed by MODIS - Each of map contains 10-days sum of the number of burning cells from July (Fig. 2). Each map accumulates fire pixels over 10 day periods from June 1 to August 20. The corresponding time-series of the 31, except Fig. 2i, which covers a 12 day period during August 20-31. Black stars and numbers in Fig. 2 indicate the position of the AERONET stations. Figure 3 shows time series of the
- total number of burning cells in the studied area (40–65N and 10–60E) is shown fire pixels per day in the same area as in Fig. 2. 20 From Fig. 1 its obvious that the burning of forests, grass and peat Figure 3 indicates that the fire activities increased from mid-July on occurred throughout Europe, but the highest fire concentrations were throughout Eastern Europe. The largest number of fires was observed in the ETR, Ukraine, and Moldova. Both the overall number and intensity of fires in this regions reached a the brightness temperature of fires reached their maximum between July 26 and August 18. The highest count of fires and
- also largest FRP were observed in-largest number of fires, as well as the largest brightness temperature were observed on July 25 29 (Fig. 12c, d-f and Fig. 23). Such large fire areas, intensities and durations high brightness temperature, and long duration were caused by peculiar specific weather conditions over the ETR and Eastern Europe during the second half of summer 2010, which led to accumulation of aerosols 2010. This favoured the accumulation of aerosol in the atmosphere over these regions (Witte et al., 2011; Chubarova et al., 2012; Péré et al., 2014).
- Ground-based AERONET observations showed an increased aerosols content over Eastern Europe aerosol content over 30 the Eastern European sites during July and August 2010. We analyzed changes in AOD at 500 nm using all measurements on daily average, Level 2.0, from daily averaged measurements from June 1 to August 31 from the AERONET database for the sites from Minsk (Belarus), Moscow (Russian Federation), Toravere (Estonia), Belsk (Poland), Chisinau (Moldova (Chisinau/Moldova), Cluj-Napoca, Bucharest, and Eforie (all Romania), Kyiv and Sevastopol (both Ukraine). AOD 500 nm

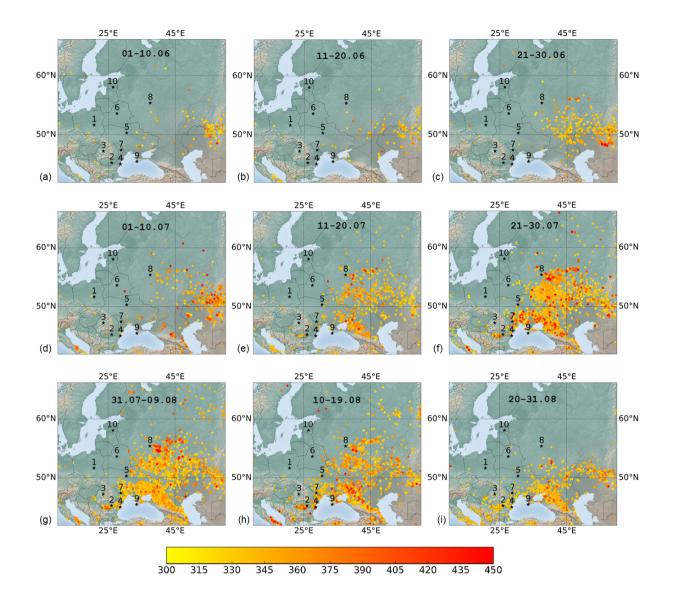


Figure 2. Localization Fire locations and intensity-brightness temperature (in K) of fires fire pixels in the ETR and Eastern Europe (40–65°N and ETR during summer 2010 according to 10–60°E) detected by MODIS data represented as a sum of burning cells and FRP every accumulated over 10 days day periods from July June 1 to August 20: (a) July June 1–10, (b) July June 11–20, (c) July June 21–3130, (d) August 1–10July 1-10, (e) July 11-20, (f) July 21-30, (g) July 31-August 9, (h) August 11–20, 10-19, and a 12 day period (i) August 20-31. Black stars and numbers indicate the position of AERONET stations also shown in Fig.1: 1-Belsk, 2-Bucharest, 3-Cluj-Napoca, 4-Eforie, 5-Kyiv, 6-Minsk, 7-Moldova, 8-Moscow, 9-Sevastopol, 10-Toravere.

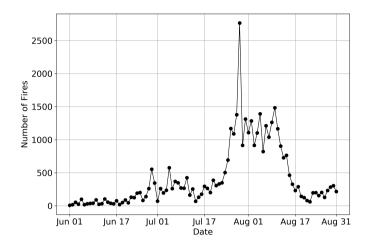


Figure 3. Average number Total amount of burning cells during fire pixels for each day of summer 2010 according to MODIS Aqua and Terra data over territory of the area shown in Fig.1 (40–65°N, -10-60 and 10-60°E).

data for the Kyiv site was obtained by data interpolation using Angstrom formula in the range of 440–870 nm. The analysis revealed a significant temporal dynamics of the aerosols aerosol content with pronounced peaks during August 15–17 in Kyiv, Sevastopol, Minsk, and Bucharest (see . Table 11). This indicates the accumulation of aerosols from one source of pollution within a single synoptic process. Also, highest AOD Table 1). Also, the highest AOD values were observed in Romania (Cluj-

5 Napoca and Eforie sites) on August 1, in Moscow and Toravere on August 7, in Moldova and Belsk in July (observations from Moldova from July 25 until October are were not available). The highest air pollution eaused by aerosols over all analyzed territorics was observed over among all analyzed sites caused by aerosol was detected in Moscow in the first half of August apparently due to the proximity of the most active combustion centersfires.

To study in-detail the aerosols inflow to the measurement sites, as given in Table 11, we computed back trajectories of air

10 masses for each of the corresponding dates at different heights analyze the aerosol inflow towards AERONET sites from Table 1, we calculated HYSPLIT back trajectories for dates of maximum AOD (Fig. 34). Trajectories were simulated for 168 hours (7 days) from the respective sites at altitudes of 500 m, 1.5, 3, 4, and Our analysis of back trajectories revealed that air movements in the lower 5 km at 12:00 GMT starting time. km layer of the troposphere corresponded to anticyclonic circulation, which is seen in Fig. 4a-j at various altitudes as clockwise-shaped curves. The maximum AOD values from the ten AERONET sites in

the ETR and Eastern Europe were formed under conditions of air stagnation and accumulation of contaminants.

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Back trajectories for locations where high aerosols loads were observed first, i.e. Moldova and Belsk, where the maximum AOD was observed the earliest within summer 2010 (July 13 and 16, respectively), are shown in Fig. 34a,b. Trajectories indicate , aerosol were The trajectories indicate that aerosol was transported to Moldova from Europe (at the altitude of 3–4 km), from the ETR (500 m) and the Black Sea (1.5 km)throughout Ukraineat altitudes from 0.5 to 1.5 km from the fires in the

 Table 1. Level of air pollution caused by aerosols_aerosol (AOD 500 nm) in summer_from June 1 to August 31, 2010 over the ETR and

 Eastern Europe according to AERONET.

No	Site	Number of meas.	Mean AOD	Std.Dev	Min AOD	Max AOD	Date of Max AOD
1	Belsk	1938	0.18	0.11	0.05	0.74	16-07-10
2	Bucharest	2381	0.30	0.16	0.06	0.97	17-08-10
3	Cluj-Napoca	1220	0.25	0.13	0.04	0.62	01-08-10
4	Eforie	1855	0.24	0.10	0.06	0.59	01-08-10
5	Kyiv	2732	0.30	0.20	0.05	1.26	15-08-10
6	Minsk	1368	0.25	0.20	0.04	1.27	17-08-10
7	Moldova	1343	0.22	0.11	0.05	0.62	13-07-10
8	Moscow	1573	0.36	0.46	0.05	4.62	07-08-10
2	Sevastopol	3564	0.23	0.12	0.04	0.93	16-08-10
10	Toravere	1296	0.20	0.19	0.03	1.23	07-08-10

ETR and south-east of Ukraine (see also Fig. 2d,e). Into the region of Belsk, aerosols were transported throughout aerosol was transported across continental Europe (1,51,5-5 km), mostly from the Atlantic Ocean, but also from the Baltic, across regions of quite active fires by that time active fires (Fig. 12) in the lower atmosphere (500 m). Transport-

Transport of aerosol to two Romanian sites (Cluj-Napoca and Eforie) - with maximum AOD observed on August 1 (Fig. 34c,d) - also occurred in the lower layerof the atmosphere in an altitude of 1, 5 km, lowermost 1.5 km layer, originating from the southeast of Ukraine and Moldova (also the area with active fires); at 3-4 km altitude – from the Atlantic through Europe, the Mediterranean and the Balkans; and in the highest layers (5-6 km) to Eforie from the Middle East through the Mediterranean and the Balkans.

. Back trajectories for Moscow (Fig. 3e) and Toravere (Fig. 3f), with and Toravere with a maximum AOD on August

10 7 , correspond to anticyclonic circulation: according to back-trajectories, aerosols to Moscow were are shown in Fig. 4e.f. respectively. Aerosol to Moscow was transported mostly from the surrounding regions with the most active fires. Air masses entering Toravere over Toravere (Fig. 4f) originated from Asian regions and crossed areas of active fires in southeastern Ukraine in all analyzed altitudes . For Kyiv (Ukraine), (Fig. 2g).

To Kyiv, where the AOD maximum was observed on August 15:-, aerosol was transported in the lower 4-km atmospheric

- 15 layer, the air movement corresponded to anticyclonic circulation, which caused the air stagnation and contaminants accumulation over the ETRand Eastern Europe; at the altitude of 5 km air was transported across the Europe from the Atlantics 4 km layer from the most active fires in the ETR, Ukraine, and Moldova (Fig. 34g, Fig. 2h). On August 16, the maximum was recorded in Sevastopol on the Black Sea coast, where air masses travelled traveled in almost the entire range of analyzed heights (500 m–5 km) from the ETR and Kazakhstan through the territory of active fires in the south-west of Russia (Fig. 34h).
- In Minsk and Bucharest the maximum was observed a AOD was observed one day later on August 17 (Table 111, Fig. 34i,j). Towards Minsk , aerosols were transported from different regions at different altitudes. At 5 km, air masses came from

the Mediterranean Sea through the Balkans, Romania and the western part of Ukraine; at 4 km they originate from the Alps and travelled throughout Central Europe and Poland; at aerosol was transported at 3 km from Kazakhstan across the Caspian Sea, south of southern Russia, and Ukraine, where the active fires were observed; at 1,5 km -1.5 km from Ukraine, and at 500 m – from the western regions of the ETR through Ukraine. To Bucharest air aerosol from fires was transported at 500 m

5 was transported from the north-east, specifically through the ETR and the south-east of Ukraine. At altitudes of 1.5 km and 5 km, air of Atlantic origin moved throughout Europe and entered Bucharest region. The air trajectory at 4 km is similar to the previous ones just the air originates from Iberian Peninsula, and at 3 km the air originates from Africa.

On August 18-21, According to the monthly weather reports of the Ukrainian Weather Center, a change in synoptic processes was observed : atmospheric weather was observed on August 18-21. Atmospheric fronts of an active cyclone - which moved

10 from the south of southern Baltic region to Samara , led to a significant change of weather pattern in Eastern Europe. This change caused a distinct decrease in fires activities , fire activities and a wet deposition of aerosols, thus their lower aerosol, lowering its content in the atmosphere above all investigated regions in the second half of August.

3.2 Aerosols Aerosol dynamics over Kyiv according to AERONET measurements and analysis of back trajectories

Milinevsky et al. (2014) analyzed aerosols pollution over Ukraine for the years 2008-2013 from AERONET sunphotometer
 observations, also noting a significant higher AOD over Kyiv in July–August 2010. Table 2 2shows our analysis of the AERONET measured aerosols content over Kyiv and its variability during the period of observation, also including-

Between 2003 and 2014 for comparing with Milinevsky et al. (2014). The highest mean and median values for the year were observed in 2010 mainly due to impact of wildfires in July and August. The highest content of aerosols over Kyiv was observed ground-based and satellite observations showed the highest aerosol content over Kyiv every year in spring

- 20 (April–May) and late summer (July–August). The lowest AOD was observed in June and in the middle of autumn, in agreement with Milinevsky et al. (2014). However, the least variable AOD during the warm period of the year was observed in June, while in June 2010 it was also one of (July–August; Bovchaliuk et al., 2013; Milinevsky et al., 2014). According to both studies, the observed spring peak in aerosol content is associated with transport of the Saharan dust across Eastern Europe, transport of sea salt aerosol from the Black Sea and the Sea of Azov, and occasionally occurring agricultural fires. The summer peak results
- 25 from wildfires, soil dust aerosol due to harvesting activity, and transport of Saharan dust. The lowest AOD was observed in June and the middle of autumn. In Table 2 we show a prolongation of the data record of Milinevsky et al. (2014, 2008-2013) for the largest for this month. The most significant air pollution caused by aerosols for the entire period of AERONET observations in Kyiv was recorded in August Kyiv AERONET site by three more years up to the end of 2016. Even in this extended record the most significant aerosol pollution was observed in August 2010. This event related to the active fires of vegetation in the ETR
- 30 and in other parts of Eastern Europe.

The impact of fires on the AOD over Kyiv during summer 2010 was less distincts as over Moscow (Fig. 4). However, relative to multi-annual average conditions, also the aerosol pollution over Kyiv was exceptional. The average AOD 440 nm for August 2010 exceeds the mean value for the same month for the entire observation period (excluding 2010) by about factor two (see Table 22). In addition, the mean 500 nm AOD for June 2010 over Kyiv was approximately 0.20, while its daily average on

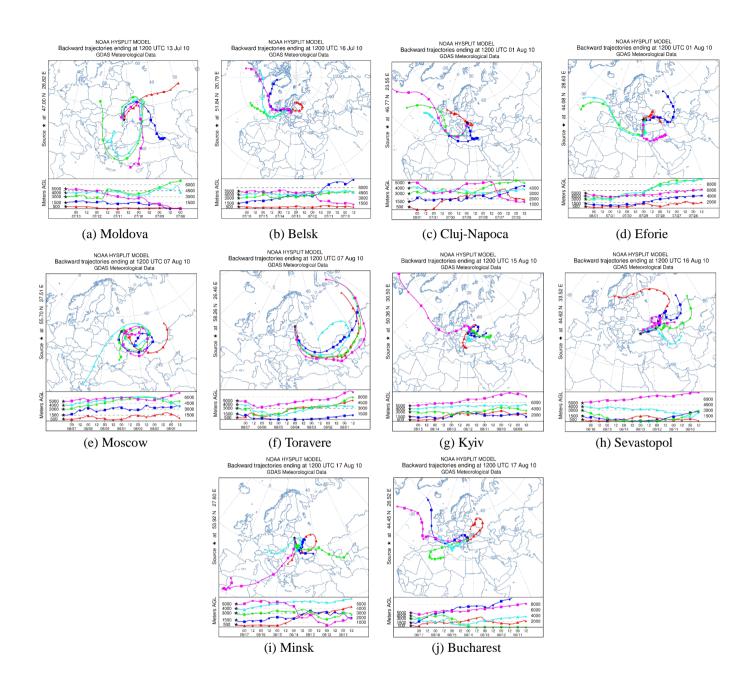


Figure 4. Back trajectories of air masses to the AERONET sites on the for dates with the highest of maximum AOD during summer 2010.

August 15 was approximately 1.09. That is 5.5 times higher than the average AOD 500 nm for the August of the same year was 0.44. During the summer period, from early June until approximately August 18, the aerosols content over Kyiv gradually

Table 2. Annual changes of monthly averaged AOD at 440 500 nm over Kyiv during the warm season for the available observational period from AERONET observations.

	AOD 500 nm							
Year	April	May	June	July	August	September	October	
2008	0.181-0.15	0.253-0.21	0.239-0.20	0.178-0.14	0.338 0.27	0.310 0.25	0.222-0.18	
2009	0.384-0.31	0.255-0.21	0.236-0.20	0.239 0.20	0.225 0.19	0.278-0.23	0.191-0.16	
2010	0.322- 0.27	0.200-0.17	0.240-0.20	0.322- 0.27	0.520 0.44	0.144-0.12	0.135-0.12	
2011	0.386-0.32	0.239 0.20	0.232-0.20	0.328-0.27	0.253 0.21	0.325-0.19	0.309-0.19	
2012	0.275-0.23	0.241-0.20	0.212- 0.17	0.205-0.21	0.248 0.20	0.150 0.13	0.157-0.13	
2013	0.268 0.22	0.192-0.17	0.242 0.20	0.252-0.14	0.206 0.18	0.163-0.14	0.214 0.17	
2014	0.253-0.18	0.239-0.17	0.152-0.13	0.214 0.17	0.270 0.24	0.197-0.15	0.187-0.13	
2015	0.17	0.17	0.14	0.21	0.17	0.24	<u>0.10</u>	
2016	0.21	0.17	0.24	0.29	0.12	0.15	0.15	
Mean	0.296 0.23	0.231 0.19	0.222 0.19	0.248 0.21	0.294 0.22	0.224 0.18	0.202 0.15	
					0.2570.20 (without 2010)			

increased by more than an order of magnitude (Fig. 4a). This event is related, in particular to wildfires in the ETR and Eastern Europe.

Computing the back trajectories of air parcels that were transported to Kyiv at altitudes of 500 m, 1.5, 3, 4 and 5 km, we analyzed the possible sources of aerosols from various regions during this period (see Supplementary material, Fig. S01–S30).

5 AOD changes in June were defined by both local sources and the transport with air masses mainly from the western areas.

-AOD 500 nm over Moscow and Kyiv (a), AOD 440 nm over Kyiv (b), AE over Kyiv (c).-

According to synoptic analysis, at the beginning of June the air in the lower 5-km of the atmosphere was transported to Ukraine from the south, mainly from North Africa and the Mediterranean Sea through the Balkans and the Black Sea. Therefore, the relative maximum AOD during June 1–2 (Fig. S01-S02) was caused by local sources and, in addition, by

10 particles transported from southern regions, as mixtures of continental soil dust and urban-industrial aerosols with sea salt and possibly dust from Sahara. This is testified by both relatively low values of AE. The impact of the wildfires on the AOD over Kyiv during summer 2010 was less pronounced as over Moscow (Fig. 45c), and prevailing coarse mode AOD over fine mode defined by spectral deconvolution algorithm (SDA) (O'Neill et al., 2003)a). However, it should be noted that during these days AOD measurements were performed in partly cloudy conditions and an observational selection was considered. Thus, for June

15 1 we had only 3 measurements in the evening, but for June 2 there were more data (Fig. 5).

Increased AOD values during June 8–14 also were observed in the conditions of unstable weather, sometimes with clouds, however the series enable to assess the behavior of AOD for every day. Air masses in the altitudes the aerosol pollution over Kyiv was also exceptional in comparison with the multi-annual average. In August 2010, the average AOD 500 m-nm exceeded

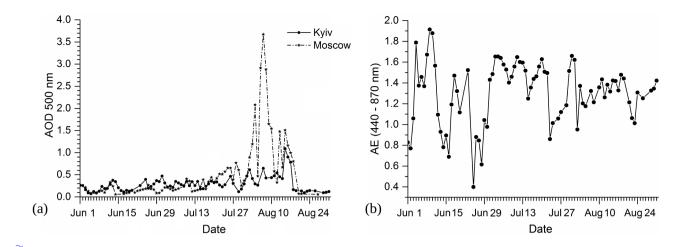


Figure 5. AOD 500 nm over Moscow and Kyiv (a), AE over Kyiv (b) during June 1-August 31, 2010.

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the mean value for the same month over the entire observation period (excluding 2010) by a factor of 2.2 (see Table 2). The mean AOD at 500 nm for June 2010 over Kyiv was 0.20, while its daily average on August 15, 2010 was more than 5 km were transported to Kyiv from the Atlantic across Seandinavia, the Baltic Sea, circulating over the Western, Central and Eastern Europe (Fig. S03-S09). The increase of AOD was caused by aerosols of different types: according to AE and SDA, on June 8–11 fine mode of aerosols prevailed, and further increase of AOD during June 12–13 was caused by the arrival of coarse mode aerosols, which explains the rapid decrease of AE (Fig. 4e). Coarse aerosol particles were mainly of local origin, while the fine

- mode particles likely were brought by air currents from Europe. times higher, 1.09. During June 23–26 AOD was measured in cloudy conditions, so the fact of its increase-
- During the period from June 1 to August 18, 2010 the aerosol content over Kyiv gradually increased by more than an order of magnitude (Fig. 45a,b)cannot be considered as reliable because of the small amount of data (9 measurements in the evening of June 23). By computing back trajectories to Kyiv at altitudes of 500 m, 1.5, 7 measurements on June 25 3, 4, and 3 measurements on June 26, Fig. 5). Herewith, the dynamics of airflow during these days was more diverse (Fig. S10-S12). At 500 m air was transported to Kyiv mainly from Kazakhstan through the steppe zone of ETR and eastern and south-eastern regions of Ukraine; at the altitudes of 1.5km, we analyzed possible sources of aerosol from various regions. We provided
- 15 detailed description of AOD variations and the impact of air transport on those changes in the Supplement Fig. S1-5 km mainly from Turkey and the Caucasus through the Black and Azov seas, often changing direction and heights. In general, the accuracy of derived AOD values, obtained from a rather small number of individual measurements through short-term gaps in the clouds, can not be considered as sufficient. Because the algorithm of detection and elimination of the influence of clouds on measured AOD is not able to distinguish a thin uniform layer consisting of cloud water droplets from aerosols in the
- 20 sunphotometer field of view (Smirnov et al., 2000). Therefore, we do not interpret relatively high AOD values for June 27–29 as a significant aerosols load over Kyiv during these days. On June 30 the average AOD at 500 nm was equal approximately 0.35.

It was computed for almost continuous dataset observed that day. This means that the aerosols content on June 30 increased comparing to the June 16–20.

During June 27–29 the dynamics of tropospheric circulation and the transport of air masses through Kyiv significantly changed (Fig. S13–S15). Preceding this period, since the beginning of June, air masses predominantly travelled to Kyiv mainly

- 5 from the Atlantic throughout the European continent. However, during June 27–29 the air mass transport path changed as seen in our back trajectory calculations that moved first southward and finally eastward. So that finally from June 29 on , air in above-mentioned altitudes was transported to Kyiv from Kazakhstan and Central Asia, across the Caspian Sea, the steppe zone of the ETR, and eastern regions of Ukraine. In addition, the 500 m back trajectory indicated a rapid change of the air motion from the north-west. On 30 June air motion remained unchanged (Fig. S16) S34.
- 10 According to our analysis of air mass transportand, spatio-temporal distribution of the intensities of fires in Eastern Europe, it was seen that vegetation fires wildfires and their brightness temperature, the wildfires were not the main source of increased concentrations of aerosols AOD over Kyiv in June. The complex impact of continental particles and aerosols of marine origin was dominating. The relatively high AOD value on July 1 (see Fig. 5) was observed based on less than 10 measurements during that day. There were also no more measurements the day after, on July 2. During both days, wind directions at different
- 15 altitudes varied much. Air entered Kyiv on July 1 at lower altitudes (500 m-1.5 km) passing central regions of the ETR on June 28–30. At 3–5 km altitude air came from Both local sources (city transport, heavy industry, etc.) and air transport from Western Europe determined the aerosol content and properties, hence continental and marine aerosol was dominating. In the steppe area between the Caspian Sea and the eastern border of Ukraine. As seen from Fig. 1a, a large number of fires burned there during those days. Thus, combustion products in the form of aerosols likely influenced the atmosphere over Kyiv and increased the 20 AOD.

In the morning of July 2 air masses entered Kyiv from the North Atlantic at altitudes of 500 m–3 km; at the end of day also at altitude of 5 km (Fig. S17). In the followingthree days, July 3–5, when washout by rain cleaned the atmosphere in the evening of July 2ndfollowing, AERONET-measured AODs were temporarily low, with values below 0.3 at 440 nm. On these days cloudiness decreased significantly and more measurements could be taken into account, thus, decreasing the uncertainty

25 of the derived time-series. Between July 3we used corresponding AODs as reference values for the further estimation of the aerosol content and its properties during July and August 2010, when the wildfires took place.

From the end of June 2010 the transport of air masses to Kyiv changed significantly as shown in back trajectory simulations (Supplement Fig. S15–5 air masses entered Kyiv from the North Atlantic and Arctic through various parts of Europe, particularly at lower altitudes (500 mS16). The air masses reached Kyiv at different altitudes after crossing the regions with the wildfires.

- 30 Additionally, the number and the brightness temperature of wildfires were gradually increasing during July-1.5 km) through the ETR (Fig. S18-S20). During July 6–7 the synoptic situation changed (Fig. S21-S22). Air masses now travelled westward at altitudes 0.5–4 km, originating mainly from central Kazakhstan and the ETR. Those air masses crossed the steppe zone of ETR and south-east Ukraine, where active fires were observed (Fig. 1a,b) and caused the relatively high AODs over Kyiv (Fig. 4a). Another rapid change in the synoptic situation and transport pathways occurred again on July 8–9: air masses came from
- 35 the Atlantic coast of Western Europe, travelled eastward causing an AOD decrease over Kyiv-August (Fig.4b, S23-S24).

The aerosols load over Kyiv during July 10–27 (Fig.4a,b)was changing under the influence of air masses mainly coming from the Atlantic and the Arctic Ocean through the ETR, where different types of fires burned (Fig. S25). These air masses crossed Kazakhstan and steppe zones of the ETR and through south-eastern Ukraine. Air masses were also circulating over the Central and Western regions of the ETR, Belarus and Ukraine and other neighboring countries, where at that time vegetation and

- 5 peat fire were observed (Fig. S26). The number of fires and their intensity were gradually increasing towards end of July(Fig. 1b,e2 and 3). Consequently, the increase of AOD over Kyiv during those days this period was caused by the accumulation of aerosol particles from the regions of active fires or transported from the wildfire regions or was formed under the influence of combustion products. Intermediate AOD decreases are caused by below cloud scavenging from occasional rains over Kyiv. Rainy weather on July 27–28 over Kyiv site and a change in the transport pathways caused a significant decrease in
- 10 AODObserved intermediate decreases of AOD, e.g. during July 29–30. Air masses entered Kyiv in the altitude range 500 m–5 km, coming from North Atlantic, passing Western Europe, the Mediterranean, the Balkans, Romania, Moldova, and western regions of Ukraine (Fig. S27).-

During July 31–30, were mostly caused by deposition from occasional rains over Kyiv. From the beginning of August 1 an increase of AOD over Kyivwas observed (Fig. 4a,b), with air masses originating over the Atlantic and traveling throughout

- 15 Scandinavia, Western and Southern Europe, the Mediterranean, the Balkans, later circulating over Ukraine, the Baltics, Poland, Romania and Moldova. Likely the increase of AOD was observed due to intense biomass burning in Ukraine and neighbouring territories, as shown by Fig. 1c,d. On August 2, air masses in the altitude range 500 mtill August 18 the weather conditions were stable and corresponded to anticyclonic circulation (see Sect. 3.1, Supplement Fig. S29-S32). Those conditions caused an accumulation of aerosol from wildfires over Kyiv and high AOD values, in particular on August 15–3 km were transported
- 20 to Kyiv from western and central regions of the ETR, from Kazakhstan throughout North Caucasus and the steppe zone of the ETR, as well as throughout south-eastern Ukraine (Fig. S28). In those regions intense forest, peat and vegetation fires were detected at that time (Fig. 1). Accordingly, these events were the reason of aerosols pollution over this region and Ukraine in particular till August 18.17.

Between August 2–18 AOD over Kyiv were increasing and reached maximum valueson 15 August with about 1.5 at 440 nm

25 and 1.26 at 500 nm, respectively. This is more than factor 2 larger than during the fire period before August 2. For the following 2 days, August 16–17, the AOD level remained high. It increased so much because air masses circulated over the entire region, from Ural and Kazakhstan to Central Europe and the Baltic According to our analysis of AE values (Fig. S29-S30). During August 18–21 atmospheric fronts of an active cyclone, which moved from the south of the Baltics to Samara region, caused significant weather change with rains in Eastern Europe. Aerosol washout cleaned the atmosphere over investigated sites and

30 caused the sharp AOD decrease seen in the time-series of Fig.45a,b. Until the end of analysing period, i.e. end of August 2010, the average AOD level over Kyiv remained as low as during less polluted days in June 2010.

According to SDA analysis by O'Neill et al. (2003), fine mode aerosols dominated the b) and results received with the application of spectral deconvolution algorithm (SDA; O'Neill et al., 2003), fine mode aerosol predominately contributed to the observed AOD increase over Kyiv in the periods 6–7 and 10–27 July, as well as on 31 July2010. The 500 nm AOD reached

35 values up to 0.1, except on July 26–27. On those two days just a few measurements (Fig. 5) were available due to cloudiness.

Thus, derived AOD values are more uncertain. A similar situation was observed later in August , when relatively high AODs were found, but only due to fine mode aerosols. On August 2 and 12, the coarse mode had August 2010. Occasionally, coarse mode aerosol had a significant impact on the observed AOD increase and the decrease of AE (Fig. 4corresponding AE decrease during the time when more fires burned (e.g. July 23 and August 2; Fig. 2f-g, Fig. 5e). b). This decrease could occur under the

5 influence of local pollution in combination with weather conditions.

Kyiv AERONET observations can also be used We also applied AERONET observations from Kyiv to analyze the impact of vegetation fires products on microphysical properties of aerosol particles in the wildfires on the aerosol spectral SSA and microphysical properties during summer 2010. Key aerosol properties, indicating microphysical processes in an aerosols population, were retrieved by inverse solution from AERONET sun photometer measurements along the almucantar of the

- 10 Sun (Dubovik et al., 2000; Dubovik and King, 2000). These parameters are effective Effective radius, size distribution (i.e. bimodal log-normal particles particle volume distribution), spectral SSA, and RI (real and imaginary parts). The impact of fires on these parameters can be estimated using the information of air masses transport by identifying the date when observed over Kyiv aerosols were transported from active fires regions. And then by comparing these data with observations when aerosols had different origin. In the following, we compare those properties as averages over periods characterizing
- 15 the fires strength and their influence on the time evolution of the Kyiv-site AOD were retrieved by Dubovik et al. (2000) and Dubovik and King (2000) by inverse solution from AERONET sunphotometer measurements along the almucantar of the Sun. The influence of the wildfires on the aerosol properties over Moscow and surrounding regions in summer 2010 was estimated by Witte et al. (2011); Chubarova et al. (2012), and Péré et al. (2014) by comparing them with multi-annual average. In our study we estimated the impact of wildfires over Kyiv by comparing the aerosol properties for the dates when aerosol from
- 20 wildfires was observed to those dates when aerosol from wildfires was absent. We identified respective dates from the analysis of air masses transport to Kyiv, as described above. During the first period between June-We compared aerosol properties as averaged values over different time periods: 1) when the number of fires and their brightness temperature were low, June 1-26, the number of active fires in the considered area (40–65°N and 10–60°E, see Fig. 2), 2) remained low. During the second period from July 18 to August) when the numbers of fires significantly increased, July 18-August 14the fire count increased largely.
- 25 The third period comprise the following three days when, and 3) when the number of fires and their brightness temperature remained high and the highest AOD values were observed(August 15–17). Partly data are also shown averaged over August 18–September 7, a period when the fire count significantly decreased., August 15-17.

The fires impact on aerosols-

30

We estimated the impact of fires on the aerosol size from AERONET sun photometer observations can be estimated by knowing the correlation between the aerosols sunphotometer observations by calculating correlation coefficients between the

- aerosol effective radius and AOD (Fig. 6a)and size distribution of the aerosol particles in the total atmospheric column (Fig. 66b). a), following the approach of Chubarova et al. (2012). The correlation coefficient for the fine mode is about 0.65, for the coarse mode it is only about 0.14. This indicates that the majority of the observed AOD increase was caused by fine mode aerosol. Changes in the aerosol size distribution in the total atmospheric column are shown in Fig. 6b. Bimodal volume distri-
- 35 butions, inferred according to Dubovik and King (2000) and averaged over the four aforementioned periods, clearly show that

the fine mode fraction of aerosols is more sensitive to the fires strength when transported via similar pathwaysaforementioned periods confirm that the main contributor to the aerosol content over Kyiv was the fine mode from wildfires. This corresponds to longer atmospheric lifetime of the fine mode aerosol in comparison with the coarse mode aerosol (Seinfeld and Pandis, 2006). In the period when most fires burned (July 18–August 14), the volume distribution of both modes increased by about 30-40 %,

- 5 due to the observed AOD increase over Kyiv. In the following three days (August 15–17), when maximum AODs were AOD was observed (Fig. 4a,b), only the fine mode increased further by more than factor a factor of 2. This is an overall increase, eompared Relative to the earlier period with less fires activity (June 1–26), of almost about factor fire activities (June 1-26), the overall increase was about a factor of 4. Consistently, after August 18, when the atmosphere over Kyiv was cleaned again, aerosols sizes the aerosol content in both modes were even smaller decreased, in turn, was even lower than during earlier
- 10 periods, indicating to be returned similar to the summer-time natural background level.

The dependence of <u>SSA in the</u> visible and near-infrared on the fires intensity <u>SSA from the fire activities</u> and aerosol load over Kyiv is obvious evident from Fig. 6c by reduced values during the more intense fire period between July 18 and August 14. Most probably, these changes of <u>SSA</u> were caused by <u>increase of an increase of the</u> soot content in the air, which was transported from the fires. <u>Increased absorption of aerosol particles A corresponding increase of aerosol absorption in the long-</u>

- 15 wave part of spectra is seen in the slope of the SSA spectral dependence, which becomes steeper. This holds true also during the three days when the highest AODs have been observed on It is similar for August 15–17, although multiple light scattering effects in the dense aerosol layers lead to generally larger SSA values , on about were observed, which were the same level as during the period when less fires burned in June 2010 (multiple light scattering is not fully taken into account in the AERONET inversion algorithm). Because SSA of particles 2010, According to Eck et al. (2009), larger SSA values can be explained by
- 20 an increased particle size which increases the total reflectance of atmospheric column. Since aerosol SSA is defined by their its complex RI, it explains similarities in spectral changes of SSA and real RI and also spectral changes of both real and imaginary RI (Fig. 6d, which describes absorption properties of particles. The RI spectral dependence, however, is less pronounced than for SSA, but multiple light-scattering likely affects the imaginary part only. More detailed values of SSA and complex RI that were defined by measurements at Kyiv AERONET site during summer 2010 are given in Table 3, showing We additionally
- 25 provided the daily averaged SSA as well as real and imaginary RI for 440 and 870 nm, Level 2.0 from AERONET database for the specific dates during summer 2010 at the Kyiv AERONET site (Table S35).

3.3 Using satellite data for estimating wildfires' influence on aerosol aerosol pollution over Ukraine

3.3.1 Using Estimations of aerosol pollution using MODIS data.

In order to estimate air pollution caused by wildfire aerosols we apply AOD parameter from the satellite instruments MODIS

30 (on board Terra and Aqua) and CALIOP (on board CALIPSO). In Section 3.3.1 we compare the AOD derived from MODIS with collocated AERONET measurements performed over Kyiv site (as described in Sec. 3.2) and analyze the aerosols effect on air pollution over Ukraine. In Section 3.3.2 we compare AOD between MODIS and CALIOP for regions outside Kyiv and

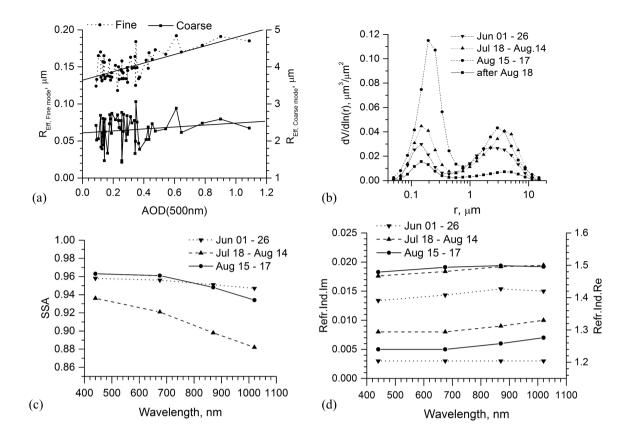


Figure 6. Aerosol particles spectral SSA and microphysical properties from AERONET measurements at the Kyiv site during summer 2010: (a) – particles particle effective radius versus AOD for fine (left axis, bottom top curves) and coarse (right axis, top bottom curves) modes averaged during 3 periodsfor the entire summer 2010; (b) – particles particle size distributionaveraged during 4 periods; (c) – spectral SSA;-, (d) – spectral refractive indexRI, – real (right axis, top curves) and imaginary (left axis, bottom curves) partsaveraged during 3. Representative periods are June 1-26 (triangles down), July 18-August 14 (triangles up), August 15-17 (circles), and August 18-31 (squares).

provide detailed analysis of CALIOP aerosols profiles over Ukraine within summer 2010. Comparison between CALIOP and AERONET was not performed because of lack of available collocations.

To link atmospheric pollution caused by biomass burning aerosols with fires activity, AOD variations and changes of weather conditions we divided summer 2010 into 3 main periods: pre-fires, active fires, and post-fires. These periods were

distinguished according to characteristics of the spatio-temporal evolution of MODIS FRP (Sec. 2) and AOD 500 nm over the
 Kyiv AERONET site, together with an analysis of air mass transport to Kyiv from the HYSPLIT model.

Although a steady transport of air masses from regions of ETR, where fires burned, to Eastern Europe, including Ukraine, established only from approximately July 10 and both number and FRP during the following week were still relatively low. Consequently, the AOD over Kyiv did not increase significantly and remained on about the same level as during June and the

10 beginning of July. Both fires activity over Eastern Europe and averaged AOD over Kyiv started to increase after reaching an

 Table 3. Aerosol particles microphysical properties from AERONET measurements at Kyiv site during summer of 2010 (SSA – Single Scattetring Albedo; Re, Im – real and imaginary parts of refractive index).

	440nm-	870nm -	Re440nm-	Im440nm-	Re870nm	Im870nm-	
+	2	3	4	5	6	7	8 -
09.06.2010	0.952 -	0.922 -	1.379 -	0.0059 -	1.383 -	0.0054-	+
12.06.2010	0.947 -	0.928 -	1.347 -	0.0029-	1.414	0.0043-	2
13.06.2010	0.969 -	0.974	1.356-	0.0020-	1.414	0.0015 -	3
14.06.2010	0.965 -	0.978 -	1.483 -	0.0022-	1.497 -	0.0015 -	+
30.06.2010	0.916 -	0.908 -	1.498	0.0082-	1.521	0.0066-	+
10.07.2010	0.941 -	0.891 -	1.367-	0.0077 -	1.388	0.0097 -	+
19.07.2010	0.933-	0.867 -	1.427 -	0.0081 -	1.460	0.0112	4
20.07.2010	0.947 -	0.900-	1.398	0.0061 -	1.437-	0.0075 -	2
31.07.2010	0.943 -	0.892 -	1.409 -	0.0073 -	1.430-	0.0086-	3
02.08.2010	0.941 -	0.945 -	1.521 -	0.0064 -	1.552-	0.0051 -	4
05.08.2010	0.939 -	0.917 -	1.490-	0.0060-	1.532-	0.0058 -	+
07.08.2010	0.940 -	0.916 -	1.505 -	0.0082-	1.532	0.0089 -	8
08.08.2010	0.916	0.886 -	1.514	0.0091-	1.556-	0.0092-	4
10.08.2010	0.928 -	0.869 -	1.475 -	0.0105-	1.500-	0.0156 -	3
11.08.2010	0.942	0.897 -	1.464-	0.0082-	1.485-	0.0114 -	8
12.08.2010	0.943 -	0.905 -	1.498	0.0076-	1.516	0.0096-	3
13.08.2010	0.932	0.884	1.478	0.0087-	1.511-	0.0116	10
14.08.2010	0.939 -	0.894	1.491 -	0.0080-	1.524	0.0113-	3
15.08.2010	0.964	0.953 -	1.517-	0.0056-	1.527-	0.0059-	8
16.08.2010	0.965	0.946	1.493 -	0.0056-	1.508	0.0071-	10

intermediate minimum on July 15–16. On July 18, AOD 500 nm was approximately similar to its average over three previous weeks. Therefore, we define this time span from June 1 to July 18 as the pre-fires period.

For the following four weeks, the averaged AOD as well as the number and intensity of fires increased continuously over investigated region. Thus, the time span from July 19 to August 18 is considered as active fires period. On August 18 weather conditions started to change rapidly, leading to a significant AOD decrease over Kyiv. These conditions define post-fires period



from August 19–31. Widely used procedure of MODIS AOD data validation by AERONET sunphotometers technique is stated in many publications, such as (Ichoku et al., 2002; Remer et al., 2005, 2008; Levy et al., 2010). In our analysis MODIS Collection 5 data are examined

for potential regional peculiarities by means of comparison with relevant AERONET data and comparison slightly differs from

the procedure used by Ichoku et al. (2002). We used MODIS image pixels containing Kyiv AERONET site computing only distances and azimuths angles between centers of each pixel of image's granule and AERONET site using coordinates of these points and determining the minimum of the distance set. MODIS AOD averaged over the pixels area centered on the Kyiv AERONET site are used to compare with AERONET AOD. This simplification of Ichoku et al. (2002) procedure is acceptable

- 5 because MODIS images of land areas do not exceed an area of 50x50 km, which is significantly less than characteristic dimension of inhomogeneities in the spatial distribution of aerosols in the atmosphere (Anderson et al., 2003). In addition, we chose only one AERONET AOD measurement performed at time closest to the satellite measurement (±30 min). AERONET AOD 550 nm were interpolated by Angstrom equation using AE determined by AERONET algorithm at spectral interval 440–675 nm.
- 10 However, to estimate effects of possible irregularities in aerosol spatial distribution MODIS AOD 550 nm have been determined by three algorithms. 1) Choosing only one pixel closest to the AERONET site, with the distance between pixel center and AERONET station not exceeding 10-12 km. On average those pixels size is approximately 10 km × 10 km. It ensures that we compare AOD obtained at the same air mass. During June –August During June 1-August 31, 2010 such events were found : 29 for Aqua and 36 for Terra. And the time span between sunphotometer and we found 39 events for MODIS/Aqua
- 15 and 40 events for MODIS/Terra measurements exceeded which satisfied the collocation criteria with the AERONET Kyiv site, described in Sect. 2.2. The time span exceeded 10 min at 4 cases whereas for MODIS/Aqua it happened one time only. 2) 3x3 pixels were selected from the MODIS dataset, with Kyiv AERONET site in the central pixel. The area averaging in this case is equal to approximately 30 km × 30 km. During three summer months of 2010 we found 38 events for Aqua and 40 for Terra, the time span minutes between sunphotometer and MODIS/Terra measurements exceeded 10 min in 6 casesand
- 20 Aqua measurements in eight cases, for MODIS/Aqua in 7 ones. 3) 5x5 pixels were selected from MODIS dataset with Kyiv AERONET site in the central pixel, the area averaging was equal to approximately 50km × 50 km. This case corresponds to area size analysed by Ichoku et al. (2002). During the three summer months 2010 we found 39 events for Aqua and 40 for Terra (the same practically as in previous case), the time span between sunphotometer and Terra - in seven cases. We approximated the comparison of MODIS AOD (AOD_{mud} for MODIS/Aqua measurements exceeded 10 min in 8 cases and and AOD_{mud}
- 25 for MODIS/Terrain 7 cases. Unfortunately, we did not find MODIS data suitable for comparison with AERONET during post-fires period (i.e. after August 18). Comparisons were approximated by) with AERONET supplotometer data (AOD_{Sph}) with the straight-line equation with the coefficients determined by the least-square technique. It was assumed that random AOD measured We assumed that the randomly measured AOD by ground-based and satellite instruments are comparable and normally distributed. Linear regression coefficients, correlation coefficients and standard deviations of the points from regression
- 30 line were computed for three cases: 1) all AOD 550 nm data for all three summer months; 2) for pre-fires period only; 3) for active fires period only. Results of this analysis is presented in Table 4. We found that the differences AOD_MODIS AOD_Sph As a result, we derived the following linear equations. For MODIS/Aqua $AOD_{mud} = -(0.09 \pm 0.02) + (1.20 \pm 0.06) \times AOD_{Sph}$ with a Pearson's correlation coefficient R = 0.96 and standard deviation SD = 0.07. For MODIS/Terra $AOD_{mod} = -(0.11 \pm 0.01) + (1.26 \pm 0.04) \times AOD_{Sph}$, R = 0.98, SD = 0.05. Only 18 % of AOD differences between MODIS/Aqua and
- 35 the sunphotometer and 22.5 % between MODIS/Terra and the sunphotometer are out of the range $\pm (0.05+0.15 \times \text{AOD}_{Sph})$

Table 4. Coefficients of Linear regression equation connecting AOD 550 nm determined for each of three spatial scales from MODIS and ground-based sunphotometers measurements: $AOD_{MODIS} = a + b \times AOD_{Sph}$. Other parameters: R – Pearson's correlation coefficient; SD – standard deviation of the points from regression line; N – number of data used.

Area/-						
Period	$\frac{10 \times 10 \ km^2}{10 \times 10^2}$	20x20 km²-	30x30 km²	$\frac{10 \times 10 \ km^2}{10 \times 10 \ km^2}$	20x20 km²-	30x30 km² -
All	a=-0.05.02 -	a=-0.06.02 -	a=-0.09.02 -	a=-0.08.02	a=-0.08.01 -	a=-0.11.01
Months-	b=1.19.07 -	b=1.19.06	b=1.20.06 -	b=1.27.05 -	b=1.25.04	b=1.26.04
	R= 0.96 -	R=0.95 -	R=0.96	R=0.97-	R=0.98	R=0.98
	SD=0.07 -	SD=0.08	SD=0.07 -	SD=0.06 -	SD=0.04 -	SD=0.05 -
	N=29 -	N=38 -	N=39 -	N=36 -	N=40 -	N=40
Pre-Fires	a=0.002.004 -	a=0.03.04 -	a=0.004.03 -	a=-0.09.04	a=-0.07.03	a=-0.08.03 -
	b=0.86.20	b=0.70.17 -	b=0.70.15	b=1.35.20	b=1.20.15	b=1.14.13
	R=0.81 -	R=0.73	R=0.78	R=0.89-	R=0.91 -	R=0.92
	SD=0.07 -	SD=0.07 -	SD=0.06 -	SD=0.05 -	SD=0.05 -	SD=0.04 -
	N=12-	N=16-	N=16	N=14	N=16-	N=16-
Fires-	a=-0.07.04	a=-0.09.04	a=-0.125.035	a=-0.10.03	a = -0.11.02	a = -0.15.03
	b=1.24.10	b=1.26.09	b=1.28.075	b=1.31.07	=1.30.05	b=1.33.06 -
	R=0.96	R=0.96	R=0.97	R=0.97	R=0.99	R=0.98
	SD=0.08 -	SD=0.08	SD=0.07 -	SD=0.07 -	SD=0.04 -	SD=0.06
	N=16	N=20-	N=21 -	N=19	N=21 -	N=21

determined over land on, which was determined by Remer et al. (2008) and Levy et al. (2010) over land ina global scale for MODIS Collection 5 data, confirming Remer et al. (2008) and Levy et al. (2010). A statistical analysis for pre- and active fires periods is presented in Table 5.

- 5 Pearson's correlation coefficients (R) in Table 4 The correlation coefficients indicate functional relations between the AOD from AERONET and MODIS in all both cases. Also, R for Terra data are higher in all cases than Aqua. Lower R during pre-fires period are explained by larger uncertainties of AOD estimated from both obtained regression equations showed that MODIS and AERONET , when aerosols load is low. Generally, our regression analysis is in good agreement with results of global analysis by Remer et al. (2008); Levy et al. (2010). AODs match well within 0.40 < AOD < 0.45 and differ by no more</p>
- 10 than $0.10 \le AOD \le 0.15$ at 550 nm for Kyiv measurements during summer 2010. This indicates that the MODIS algorithm interprets aerosol over Kyiv in the same manner as AERONET. Thus, we assume assumed that both MODIS/Aqua

 Table 5. Statistics of matching between MODIS and sunphotometer AOD 550 nm in specified bounds during summer 2010 over Kyiv

 AERONET site: Abs(AOD_{MODIS} - AOD_{Sph}

Area/						
Period	10x10km²-	20x20km²-	30x30km²-	10x10km²	20x20km² 30x30km²	
Pre-fires	75% / 12-	81% / 16-	81% / 16-	71% / 14 -	81% / 16-	69% / 16-
Fires-	81%/16	90% / 20	81%/21	79%/19	86% / 21 -	81%/21

and <u>Terra correctly MODIS/Terra</u> represent the content of <u>aerosols aerosol</u> in the atmosphere over Ukraine for <u>the</u> summer 2010 within measurements uncertainties.

Estimations of aerosols pollution over Ukraine using MODIS.

From the The influence of aerosol pollution in Ukraine can be interpreted from the distribution of MODIS AOD 550
nmdistributions obtained from the MODIS Aqua. We analyzed data for 7 days with low AOD values, smaller than 0.5 over the Kyiv site: June 6 and Terra Level 2 Collection 051 Optical Depth Land And Ocean product, the influence of aerosol pollution in Ukraine becomes obvious. For example, Fig. 7 shows 7 (MODIS/Terra), June 8 (MODIS/Aqua); July 14 and 17 (MODIS/Aqua), July 15 (MODIS/Terra); August 23 (MODIS/Terra). We also analyzed 3 days (August 15–17) with high AOD values, larger than 1.0. Figure 7 shows the maps for the region 40-60°N and 22.5-40°E of MODIS/Aqua data for those two

- 10 days when the aerosols aerosol load over the Kyiv AERONET site was lowest low (a, July 17) and the highest (b, August 15). Analysis of data for 7 days (June 6 and 7 from MODIS/Terra and June 8 from Aqua; July 14 and 17 from Aqua, July 15 from Terra; August 23 from Terra) with low AOD over Kyiv site and for 3 days with maximum AOD (August 15–17) showed the following: On low pollution days the During the days with low aerosol content the AOD 550 nm is homogeneously distributed in the analyzed region, with values not exceeding 0.5. In the was homogeneously distributed (e.g., Fig. 7a) over
- 15 the whole territory. During the high pollution case, the spatial AOD distribution distinctly differs. Highest pollution is found differed. The highest AODs were observed over north-eastern and central regions of Ukraine, values reached where AOD values reached and partly exceeded a level of 2 and higher. (Fig. 7b). This AOD distribution map (Fig. 7b) resembles fairly well our air mass back trajectory calculations to Kyiv in the altitude range of 0.5–3 km for August 15 fairly well (see Fig. 3, Sect 3.1). Fig. 7b This indicates that the MODIS algorithm interprets aerosol over Kyiv in the same manner as AERONET.
- 20 Though, MODIS underestimates low AOD values and overestimates high AOD values in comparison with AERONET. Figure 7b also highlights the importance on of the availability of satellite observations for estimating air pollution over larger and remote regions, that elearly cannot which can not be deduced from a single site's ground-based measurements, as it is the case in Ukraineso far/right now.

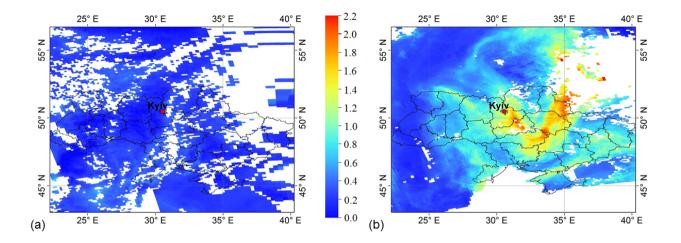


Figure 7. AOD 550 nm distribution from MODIS/Aqua measurements on July 17 (a) and August 15 (b)

3.3.2 Using CALIOP dataEstimations of aerosol pollution using CALIOP data.

Using CALIOP AOD to study spatio-temporal distribution of aerosols and its validation by ground-based AERONET observations is challenging due to different measurement characteristics of both instruments. CALIOP lidar provides only fragmentary data on aerosols along CALIPSO satellite's ground track, because of the small size of light beam (with diameter approximately 70

- 5 m) and cloudy conditions that frequently occurred. Due to orbital period of 98 minutes, ground tracks of satellite consecutive passages of certain latitude are shifted 24.5to the west, thus its spatio-temporal coverage is rather sparse. During three summer months in 2010 we did not find CALIOP AERONET coincident measurements over Kyiv, but one: the closest CALIPSO ground track was 60 km east from Kyiv AERONET site, hence, not matching the selection criteria (40 km) according to (Omar et al., 2013) for CALIOP-AERONET comparisons.
- 10 Therefore, in this study we estimated CALIOP AOD using MODIS /Aqua AOD since both satellites orbits are in A-Train afternoon constellation (). Relative positions of CALIPSO and Aqua satellites in the A-Train provide great number of practically simultaneous measurements with the time span of 2 minutes, while the spatial difference is only about 10 km. We applied MODIS Aqua Level 2 Collection 051 Optical_Depth_Land_And_Ocean product AOD We calculated linear regression parameters of AOD measurements from MODIS (550 nmand CALIPSO CloudAerosol Layer Product- AOD-) and CALIOP (532 nm;
- 15 together with Aerosols Profile Product Level 2.0 Version 3.01 and 3.02 (Winker et al., 2009, 2010).

Each granule of MODIS data consists of consecutive scans across the satellite track. The footprint of CALIOP light beam on this granule looks like a sequence of points on the straight line passing close to the center of a granule. Each point represents the center of CALIOP dataset, averaged over 5-km and once or twice matches with one of the pixels of MODIS granule. To find these matches we calculated the distances and azimuth angles between the center of each 5-km segments of the CALIOP

20 track and the center of each pixel in MODIS granule in the same manner as for MODIS-AERONET case (see above).

Table 6. Coefficients of linear regression equation connecting AOD 550 nm determined for each of two spatial scales from MODIS and CALIOP (532 nm) measurements: $AOD_{MODIS} = a + b \times AOD_{CAL}$.

Area/ Period	10x10<i>km</i>²	30x30km²	50x50km²
Pre-fires	a=0.09.004	a=0.1.003 -	a=0.1.004
	b=0.19.02	b=0.17.02	b=0.15.02 -
	R=0.25	R=0.20	R=0.16
	SD=0.09 -	SD=0.10	SD=0.10
	N=1106	N=1425 -	N=1491 -
Fires-	a=0.05.009 -	a=0.06.008-	a=0.04.008 -
	b=0.72.025 -	b=0.72.025 -	b=0.81.026-
	R=0.62	R=0.58	R=0.61
	SD=0.19	SD=0.20	SD=0.19
	N=1325 -	N=1625 -	N=1675-

While comparing CALIOP AOD 532 nm and MODIS) for more than 3690 points of collocated measurements during June 1-August 31, 2010. We derived linear equation between CALIOP AOD_{cal} and MODIS/Aqua AOD 550 nm, no corrections for potential spectral differences were applied. This yields to an estimated systematic bias in our AOD comparison of approximately 2–4% in the AE range between 0.5 to 1.0, thus, may be neglected in our cases, following Kittaka et al. (2011). The two

5 instruments comparison was performed in the same manner as our MODIS-AERONET comparison (see Section 3.3.1), in similar temporal and spatial scales.

Linear regression parameters (Table 6) indicate a weak correlation between AOD_{myd} as follows: $AOD_{cal} = (0.06 \pm 0.003)$ + $(0.68 \pm 0.015) \times AOD_{myd}$. A Pearson's correlation coefficient of R = 0.59 indicated rather close relations between AOD from CALIOP and MODIS AOD during pre-fires period (i. e. low AOD), with $0.16 \le R \le 0.25$, and stronger correlation

- 10 during period of active fires with R ≈ 0,6. A direct comparison of the two AOD datasets is given in Table 7, averaged over pre-fires period (time span 1, June 5–July 17), active fires period (time span 2, July 19–August 13), and for a single day, when one of the highest AOD was measured over Kyiv AERONET site (time span 3, August 17). CALIOP and MODIS AODs are very similar during time span 1. During time span 2, when aerosols from active fires influenced the atmosphere over Ukraine , the MODIS AOD exceeded those measured by CALIOP by 7–16%, depending on the spatial scale of the MODIS average.
- 15 The smallest difference is found in 50 km × 50 km average, and the largest in 10 km × 10 km average. But in case of largest aerosols loading during time span 3, AOD CALIOP exceeds that of MODIS by 7–12% and the differences are maximum at smallest averaging area and vice versa.

Such differences measurements. CALIOP and MODIS measured similarly at AOD = 0.2, although CALIOP overestimated lower and underestimated higher AOD values relative to MODIS. Thus, CALIOP underestimated AOD for the majority of the

Period	Averaging area-	AOD CALIOP	AOD MODIS-
		(532 nm)	(550 nm)
Time span 1-	10x10km²	0.115 -	0.127-
	30x30km²	0.121 -	0.125 -
	50x50km²	0.122-	0.118
Time span 2	10x10<i>km</i>²	0.205 -	0.240-
	30x30km²	0.208-	0.233 -
	50x50km²	0.206-	0.220 -
Time span 3	10x10km²	0.723 -	0.671 -
	30x30km²	0.715 -	0.642 -
	50x50km²	0.715	0.631-

compared data over Ukraine in summer 2010. Also, the standard deviation from the regression line (SD = 0.15) pointed to considerable discrepancies between the two satellites datasets. This means that the application of MODIS and AERONET data for the territory of Ukraine within summer 2010 is likely more reasonable to apply. Observed discrepancies between MODIS and CALIOP data can be explained by different independent measurements techniques, algorithms, and by uncertainties of

5 CALIOP daytime measurements. In this paper we do not analyze peculiarities of MODIS and CALIOP data and reasons of their discrepancies, as more detailed analysis can be found in Kittaka et al. (2011); Redemann et al. (2012).

AOD 532 nm distribution over Ukraine from CALIOP measurements during the 16-day period from August 4 to 19, 2010. Red numbers at the bottom of the map indicate dates of each daytime track running to north-west and blue numbers at the top of the map indicate the date of each nocturnal track running to south-west.

- Taking also into account uncertainties of both day- and nighttime CALIOP measurements, itthe instrument's AOD reasonably captures air pollution from wildfire aerosolsaerosol. Most CALIOP data were obtained during night and have a higher fidelity than the daytime measurements (Omar et al., 2013). Up to now CALIOP is the only instrument that provides data on nocturnal pollution of the atmosphere by aerosolsaerosol. Due to its sparse coverage over investigated regions, the data product can be used only to roughly estimate severeness of pollution over longer periods. This is well illustrated in Fig. 8, exemplary
- 15 showing all day- and nighttime AOD measurements during a 16-days period from August 4 to 1916 day period from June 1-16

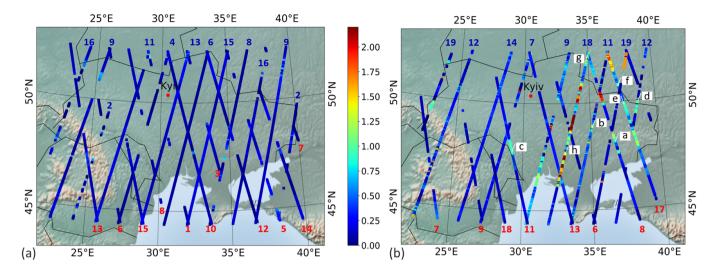


Figure 8. AOD 532 nm distribution over Ukraine from CALIOP measurements during 16 day period from June 1 to 16 (a), from August 4 to 19 (b), 2010. Red numbers at the bottom of the map indicate dates of each daytime track running to north-west and blue numbers at the top of the map indicate the date of each nocturnal track running to south-west. Labels a-h in Fig. 8b indicate the location of profiles, that are further analyzed and shown in Fig. 9.

(a) and August 4-19 (b), 2010 that corresponds to one the repeating cycle of the instrument. The tracks, that are Ground tracks, oriented from northeast to southwest correspond to nocturnal measurements and those – oriented from southeast to northwest correspond to daytime measurements. Fig.8 8shows distinct pattern of aerosols pollution, except over western Ukraine, mostly during 11-18 August with AOD values exceeding 0.5. This is in good agreement with MODIS observations on August 15 as shown in Fig. 77h. During these days weether conditions were stable and did not showed much. For both instruments higher

5 shown in Fig. 77b. During these days weather conditions were stable and did not changed much. For both instruments higher values of 1–1.2 are seen over central and eastern Ukraine.

The CALIOP measured AOD 532 nm over Ukraine and surrounding territories ranged approximately from 0 to 0.5 during 0.7 (Fig. 8a, Supplement Fig. S36a,b) during the first part of the summer 2010 (June 1 – July 1-July 18), when number and intensity brightness temperature of fires were still low - (Fig. 2a-e). CALIOP confirms that with on July 19 aerosols the aerosol content

- 10 in the atmosphere significantly-increased, corresponding to fires activity. During August 4–17 over certain regions of Ukraine, mainly east and south, observed AOD 532 nm was around 1. After August 17 number and intensity of fires, thus, aerosols content decreased dramatically and AOD distribution returned to pre-fires values. In general, these results are in agreement with our findings from MODIS . the wildfire activities (Supplement Fig. S36c). During August 4-19 (Fig. 8b) a distinct pattern of aerosol pollution was observed, except western part of Ukraine. In particular, during August 9-18 AOD values regionally
- 15 exceeded 1 and reached a value of 2 at certain locations. This is in good agreement with MODIS observations on August 15 as shown in Fig. 7b. During those days weather conditions were stable and did not change much. For both instruments high AOD values (greater than 1.2) are seen over central and eastern Ukraine.

CALIOP measurements also provide information about the <u>We also analyzed</u> vertical distributions of aerosols aerosol extinction at 532 nmfrom the Cloud&Aerosol Profile Product Level 2.0 Version 3.01 and 3.02 (Young et al., 2013; Omar et al., 2013). The aerosol extinction is proportional to concentration of the aerosol particles along the light beam and extinction vertical profiles reveal vertical distribution of aerosols in the atmosphere column. Here, we analyzed profiles compiled for those

- 5 sections of CALIOP ground tracks where the highest. We compiled analyzed profiles for the cases with high AOD 532 nmwas measured. This selection leads to 58 profiles for 11 tracks we analysed for the period August 7–18, 2010. The corresponding AOD 532 nm ranged from 0.44 (on August 13 11:00:06) to 2.93 (on August 18 00:08:26). Among selected profiles, 37 on 7 ground tracks were nocturnal, and 21 profiles on 4 tracks were measured during daytime. The profiles reveal that aerosols aerosol ranged from about 40 m to 6-mostly 5 km altitude. The vertical distributions varied significantly both during during both
- 10 day- and nighttimeand. According to the peculiarities of aerosol vertical distribution, we identified three types of profileswere identified. 1) The first type Type 1 consists of profiles showing at least a single aerosol layer of some hundred meters thickness, located at about 1km-1 km altitude or higher. 2) The second type Type 2 consists of profiles showing decreasing a decrease of extinction coefficients with increasing altitude altitude, with a maximum extinction coefficient located near the surface. 3) The third type Type 3 is characterized by relatively high extinction values over comparably large altitude ranges,
- 15 spanning several kilometers, km without showing distinct maxima. Fig. 9 depicts corresponding profiles, exemplarily selected for those cases when the above mentioned features are well pronounced. All other cases are shown in the Supplementary material (Fig. S31-S88)Supplement Fig. S37-S94.

Aerosol The aerosol extinction profile shown on in Fig. 9a reveals (location labeled 'a' in Fig. 8b) represents the vertical distribution of particles concentration at two points on daytime CALIOP trace of August 08 particle concentration at the daytime

- 20 CALIOP track in August 8, 2010running through, which crossed the south-east of Ukraine (see Fig. 8). Distance between the points with coordinates 47.67N, 36.96E and 48.29N, 36.71E is approximately 70 km, b). AOD 532 nm at these points was 1.00 and that point was 0.84respectively. At these points almost all aerosols concentrate. At that location almost all aerosol concentrated in a narrow layer at altitudes between 3.1 and 3.8 km (type an altitude of around 3.5 km (Type 1 profile). Extinction profileshown on 9Shown in the Supplement Fig. S40 reveals a similar aerosol profile, which was measured few seconds earlier in a distance of 70 km from the site labeled 'a' in Fig. 8b.
 - An extinction profile of Type 1 shown in Fig. 9b was observed at some points on 320 km segment of a segment of a CALIOP midnight track on August 11 between 48.81N, 35.08E (AOD = 1.58) and 46.11N, 34.04E (AOD = 0.99). Aerosols at these points located mainly in three thin layers with maximums at 0.3, 1.3, and 4.2 km. Considered track segment lies on distance 11. Aerosol at this location labeled 'b' in Fig. 8b was distributed at different altitudes from the surface to approximately 5 km.
- 30 There was only a thin layer showing a maximum extinction at 4.3 km altitude. In addition, a considerable amount of aerosol was observed near the surface. The chosen track segment was located about 210–250 km westward from the daytime track segment of August 8.

Our back trajectory suggests CALIOP observed the same air masses on both days at different measurement times. Daytime CALIOP The daytime CALIOP track on August 11 runs over west regions crossed the western region of Ukraine. Peak The

35 peak aerosol load in terms of AOD 532 nm is found in south-west-was observed in the southwest region of Ukraine, with values

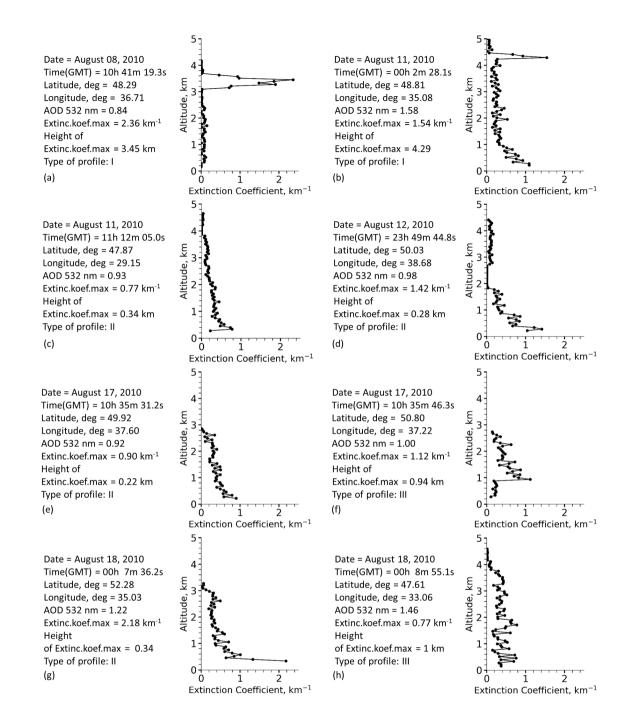


Figure 9. <u>Typical Selected</u> vertical profiles of <u>the aerosols aerosol</u> extinction coefficient from CALIOP measurements over Ukraine during active fires period in summer 2010. The location of profiles is shown in Fig. 8b with corresponding a-h labels.

around 0.9 (<u>location labeled</u> 'c' in Fig. 8). Here, the measured aerosols b). The measured aerosol extinction profile is of type 2, showing a maximum at 300–350 m altitude and gradually decreases above, up to 5 km altitude (Fig. 9c).

CALIOP's nocturnal track on August 12 also runs over ran over the east of Ukraine, where the highest aerosols AOD 532 nm of about 1.0 is found approximately over Lugansk region (Fig was found (location labeled 'd' in Fig. 8b). Over a track segment

5 of about 80 km length, most of aerosol particles the aerosol concentrated below 2 km altitude, with a maximum extinction coefficient at approximately 280 m. Another ,-less dense aerosol layer was observed also above between 3 to 4.5 km altitude (Fig.99d)...9d). The profile can also be identified as Type 2.

Five days later, on August 17, the instrument measured was measuring over the same region during daytime (Fig.8locations 'e' and 'f', Fig. 8). But here, aerosols conditions were b). On this day, the aerosol distribution was much more variable than

- 10 on during days before. In Fig. 9eand f we Figures 8e,f show two consecutively measured profiles with 15 sec time difference that corresponded to approximately 100 km distance between appropriate points. The maximum extinction coefficient of the profile, measured first (Fig. 9e), first profile (Type 2, Fig.9e) was located at about 220 m and above gradually decreases was gradually decreasing with altitude. On the second profile, other profile (Type 3, Fig. 9f) the maximum extinction coefficient was found significantly higher, at approximately 1 km altitude(Fig. 99f), Below, extinction values indicate, Below.
- 15 the extinction coefficient profile indicates practically clean air with values not exceeding $0.3 \text{ km-1}\text{km}^{-1}$. In the two measurements, the vertical extent of aerosol is about the same (approximately similar (at about 3 km). The Also, the AOD of both profiles do not differ much and range from 0.92 to 1.0.

On August 18, the day when the weather situation significantly changed, the extinction coefficient profile shows profiles of extinction coefficient at midnight showed a much higher variability (Fig. 9g,h, locations labeled 'g' and 'h' in Fig. 8b). The

- 20 layer extents up to about 4 km. The analysed track was located westwards relative to the daytime track on August 17. The analyzed segment of CALIOP's nocturnal track had a length of about 360 km and is located between 52.28N, 35.04E and 49.33N, 33.75E. The track was westward oriented, relative to the daytime track on August 17. Relatively 250 km and was located south-west from the site labeled 'g' in Fig. 8b (see the Supplement Fig. S85-S89). The aerosol layer at location 'g' in Fig. 8b extended up to about 3.5 km (profile Type 2, Fig. 9g). Further along the track up to the coast of the Black Sea aerosol
- 25 vertical distribution changed into profile Type 3 (Fig. 8b, label 'h' and Fig. 9h). At certain locations relatively high extinction coefficients are-were found from near the surface up to about 2km. Although being highly variable, the vertical structure does not decline much until 3.8 km altitude, in contrast to other profiles discussed before. The AOD 532 nm was significantly larger than measured before, equal to 1.46. As shown in the Supplementary, AOD values up to 2.93 were measured on this day (Fig. S844.5 km (Fig. 9h), but at few other locations aerosol concentrated at several relatively thin layers at different altitudes
- 30 (Supplement Fig. S90–S94).

Finally wide range of the extinction coefficient variations has to be noted in the maximums of profiles. It ranges In conclusion, our CALIOP profile measurements are well reflecting the large diversity of aerosol layers in the region and period under investigation. Although the maximum extinction coefficient was approximately 1 km^{-1} in most analyzed profiles, we noted a large spread from some tenth to 8.5 km⁻¹ (Supplement Fig. S86) in certain very dense plumes but the most frequent occurring

35 value was approximately 1 km^{-1} .

Our CALIOP profile measurements reflect well the large diversity of aerosol layers in the region and period under review. Those structures, as lofting of highest pollution regions above ground, multiple layers or zones of relatively clean air, cannot be determined. The advantage of analyzing aerosol vertical distributions allows a better evaluation of air pollution cases, for example by determining altitudes of highest aerosol content - information that cannot be concluded from vertically integrated

5

observational parameter, like AOD, although other instrument's spatial sampling allows investigation on more coherent spatial structures aerosol properties, like the AOD.

4 Discussions and conclusions Conclusions

Analysis of AOD measured by AERONET, MODIS and CALIOP, together with transport of air masses by application of back trajectories simulations, allowed estimating air pollution over Ukraine caused by exceptional wildfires in Eastern Europe during

- 10 In this study we analyzed the influence of wildfires on aerosol dynamics over the ETR and Eastern Europe, in particular on air pollution conditions over Ukraine during an extreme heat wave event in summer 2010. These fires were the biggest by area burned, the longest by duration and the most intense for the whole period of AERONET aerosols observations in Ukraine. On Kyiv AERONET site, these fires caused the highest AOD measured so far. However, analysis of the spatio-temporal variability of atmospheric pollution caused by aerosols from fires over above-mentioned territories was not done before.
- 15 In this research we used ground-based measurements from AERONET sites and data from satellite instruments MODIS (Aqua and Terra) and CALIOP (CALIPSO). The highest accuracy in such remote measurements is achieved by measuring spectral AOD with the AERONET sun-photometers and by solving inverse problem, which retrieves microphysical properties of aerosol particles (see section 2.1). These data were used as the standard for validation of satellite measurements that always have lower accuracy Specific weather conditions with high air temperature and low relative humidity (Witte et al., 2011) formed
- 20 under anticyclonic circulation, which caused air stagnation and accumulation of contaminants. Moreover, those weather conditions were favorable for wildfires to evolve.

Since there were very few AERONET sites over investigated territories in summer 2010, we filled spatio-temporal gaps in the ground-based observations by applying satellite data. For this purpose we used measurements from MODIS on board the Terra and Aqua satellites because the wide field of view of this instrument (over 2 000 km) scans each location at least twice a day.

- 25 Thus, we validated MODIS data by comparing with collocated AERONET measurements (section 3.3.1). Also, the algorithms of determination of aerosols optical properties from MODIS measurements are well developed and tested over ocean and land for different natural zones, also data quality is known (Remer et al., 2008; Levy et al., 2010). Our comparison over Kyiv in summer 2010 is in agreement with other studies, see Remer et al. (2008); Levy et al. (2010). The functional relation between the AOD measured by AERONET and MODIS over Kyiv (section 3.3.1, Table 4) suggests that the MODIS data product is
- 30 reasonable for researched territory.

Analysis of spatio-temporal evolution of AOD obtained from CALIOP measurements was rather challenging task due to fragmentation of data and their high variability. Although AOD measurements from CALIOP have higher uncertainties in comparison with MODIS, the clear advantage is that they are also performed at night-time. Despite the sparse availability of

CALIOP To reveal the connection between wildfires and aerosol properties over the ETR and Eastern Europe, we analyzed fire locations and their brightness temperature from MODIS measurements for the time and regions we analysed, it was the only source of aerosols vertical distribution.

Spatial distribution of AOD from CALIOP measurements over Ukraine corresponds well with MODIS, with the correlation

- 5 coefficient R = 0.6, see section 3.3.2, Table 6, 7. This is in agreement with Kittaka et al. (2011), who compared CALIOP Version 2 Aerosol Layer Product with MODIS Collection 5 and found only a small global mean bias in the CALIOP daytime AOD, relative to MODIS. On regional scale, biases of both signs were larger and varied with seasons. Additionally, over land this regional-seasonal bias was not larger than MODIS expected uncertainty. However, data presented in Table 7 show systematical differences between MODIS and CALIOP. Interestingly, in periods with moderate pollution the AOD from
- 10 MODIS is larger than from CALIOP. But during the short period of highest pollution, i.e. August 15-17, 2010 the CALIOP AOD is larger. These peculiarities were also noticed in other studies. E.g. Ma et al. (2013) compared monthly mean gridded data and noted systematically higher CALIPSO AOD over other major biomass burning regions (such as South Africa) during fire seasons. One reason explaining these peculiarities may be found in the CALIOP Version 2 retrieval algorithm itself, because it retrieves aerosols extinction that is converted into AOD. According Kittaka et al. (2011), the CALIOP algorithm
- 15 likely ignores tenuous aerosols, causing an underestimation of AOD compared to MODIS. Also, the height of aerosols layer's base may be detected at higher altitudes by CALIOP, leading to AOD underestimation (Kittaka et al., 2011). Main factors that determine data accuracy are uncertainties of instrument's calibration and the low signal-to-noise (STN) ratio for daytime measurements. That is why an algorithm is used to estimate the "lidar ratio" (the ratio of particle extinction to 180-degree backscatter) from the 532 nm backscatter and it is also the significant source of uncertainties in defining aerosols parameters

20 by the algorithms of the corresponding inverse solution (Omar et al., 2009; Winker et al., 2009). Taking into account atmospheric dynamics by applying back trajectories calculated with HYSPLIT for the range of altitudes up to 5 km allowed to relate the increase of observed aerosols abundance over AERONET sites in Eastern Europe with biomass burning in the particular territories of investigated region. To estimate the spatial extent of aerosols from wildfires in central parts of ETR and other regions of period June 1-August 31, 2010. We demonstrated that the fire activities increased from

- 25 mid-July mostly over the ETR, Ukraine, and Moldova. The largest number and brightness temperature of fires were observed during July 26-August 18. To consider the impact of those wildfires on aerosol dynamics over the ETR and Eastern Europe, we analyzed data from AERONET sites and MODIS measurements in Eastern Europe during summer 2010. Observed increase of AOD at chose 10 AERONET sites in that region and computed HYSPLIT back trajectories to those sites. Our analysis of back trajectories showed that the observed AOD maximum over each of the 10 AERONET sites considered in this paper was caused
- 30 by wildfiresevents. As discussed in section 3.1, by analysing back trajectories for the days with maximum AOD values, the air masses, which were transported to considered sites was formed as a result of air transport from the areas of active wildfires. AOD maxima at the Belsk site (central Poland), Chisinau (Moldova (Chisinau, Moldova), and Romania sites (Bucharest, Cluj-Napoca, Eforie) in the lower atmospheric layers up to 1.5 km, were passing areas with active fires, mainly throughout and Eforie (Romania) were caused mainly by fires in Ukraine and Moldova Maximum AODs in July. AOD maxima over other

AERONET sites were caused by aerosols, that were transported with air masses along the anticyclone periphery over Russian Federation regions, where also intense forest fires burned.

Analysis of spatio-temporal distributions of fires number and intensities over Eastern Europe as well as our air mass back trajectory calculations led to the conclusion that most aerosols from forest fires in Russia were transported to Ukraine, and

5 later to Belarus and Baltic countries, from around July 18 until August 18–19. This was the time of most intense fires in this region, which lasted from July 22 to August 18–19, aerosol from fires in the ETR. We also provided detailed analysis of aerosol dynamics over Ukraine. Despite the available studies of aerosol dynamics over Ukraine (Bovchaliuk et al., 2013; Danylevsky et al., 2011a, b; Milinevsky et al., 2014), we focused on the evaluation of the impact of

the wildfires in summer 2010, corresponding to Witte et al. (2011). From analyzing spatio-temporal AOD distributions from

10 MODIS and CALIOP we conclude that the fires had significant impact on the air quality above the whole territory of Ukraine. Most affected regions were the eastern, central and southern parts of the countryon the tropospheric aerosol load, which has not been done before.

CALIOP shows approximately the same aerosols content for day- and nighttime measurements (no distinct diurnal cycle) for all investigated periods over Ukraine, and is therefore in agreement with global analysis by Kittaka et al. (2011). In section

- 15 3.3.2 we showed vertical distributions of aerosols extinction measured by CALIOP for the territory of Ukraine, where high AOD values were observed during intense wildfires. We did not validate CALIOP data with The ground-based measurements due to lack of collocations. In order to estimate the reliability of CALIOP data we used results of other authors obtained for similar situation. Chazette et al. (2010) performed a validation of CALIOP Version 2 daytime aerosol extinction product under low content of aerosols AOD 532 nm < 0.3 during a field experiment on April 28, 2006 over France with the ground-based</p>
- 20 and airborne lidars, also employing observations from the CIMEL sunphotometer and the satellite instruments MODIS and SEVIRI. In their works measurement sites were mainly located in urban areas where rather complex aerosol compounds are expected. Chazette et al. (2010) showed that CALIOP provides high quality information on the structure of aerosol layers with AOD 532 nm < 0.3.</p>

Kacenelenbogen et al. (2011) performed a validation of the CALIOP Version 2-AERONET measurements over the Kyiv site

- 25 showed that for the entire observational period (from April 2008 to November 2016) the highest air pollution caused by aerosol was recorded in August 2010. The average AOD 500 nm in August 2010 exceeded multi-annual monthly mean (2008-2016, excluding 2010) by a factor of 2.2. We showed that during June 2010 the wildfires were not affecting AOD over the Kyiv site. Both, aerosol content and 3 daytime aerosol extinction product under conditions of high aerosols pollution (AERONET AOD 532 nm \approx 0.71). They analyzed measurements of August 4, 2006 over territory of USA by applying data from lidar installed
- 30 on aircraft, AERONET site, and from the MODIS, and POLDER instruments. Vertical profiles of aerosols extinction retrieved from CALIOP agree well with ground-based instruments, with significantly better agreement of CALIOP Version 3 data. The main differences are caused by the CALIOP extinction retrieval, which is insensitive to aerosols outside relatively thick layers (tenuous aerosols),in turn leading to an properties were determined mostly by local sources and air transport from Western Europe. In contrast, from July to mid-August, the AOD underestimation. In summary, studies of increase over the Kyiv site was
- 35 caused by air transport from the structure of aerosol layers on the atmosphere pollution over great urban areas using CALIOP

observations appear feasible when significant pollution occurs. In our study, we analyzed CALIOP data for events of high aerosols content only, and we assume that results presented in section 3.3.2 correctly represent vertical distributions of aerosols over Ukraine during active fires and when aerosols were transported in the lower 5 km of the atmosphere wildfire regions. The influence of fires resulted in an increased relative content of the fine mode in particles size distribution, accompanied by an

5 increase of their effective radius (Fig. 6a,b). Occasionally the coarse mode also resulted in both an increase of AOD and a decrease of AE for days exhibiting a higher number of fires. We explained the predominant impact of fine mode aerosol on the AOD increase by its longer lifetime in comparison with the coarse mode.

In section 3.2 we provided evidence of the fires impact on microphysical characteristics of aerosol particles according to measurements of Kyiv AERONET site . Similar conclusions were also provided by other studies

- 10 (Chubarova et al., 2012; Witte et al., 2011; Péré et al., 2014), who analyzed the variability of microphysical aerosols properties, like SSA and RI, under the impact of wildfires during summer 2010 over Eastern Europe. Chubarova et al., (2012) analyzed those effects for Moscow and suburbs from AERONET measurements. Witte et al. (2011) provides averaged estimations of SSA according to satellite (OMI) and ground-based (AERONET) measurements during intense fires (July 22-August 18, 2010) for all Eastern Europe including Ukraine. Péré et al. (2014) analyzed We also analyzed the impact of wildfires on aerosol
- 15 spectral SSA at the Kyiv site during three different periods: 1) June 1-26, when the number of fires and their brightness temperature were low, 2) July 18-August 14, when the number of fires significantly increased, and 3) August 15-17, when the highest AOD values were observed. Smaller SSA values during July 18-August 14 were likely caused by an increase of the soot content in the air, transported from the wildfires. SSA and RI , received from POLDER and AERONET measurements, and compared to chemical-transport model CHIMERE data, mainly for Moscow region.
- 20 In this study, we analyze in some detail microphysical properties retrieved from measurements at Kyiv AERONET site only. The influence of fires resulted in an increase of the relative content of the fine mode in particles size distribution, accompanied with a decrease of both their effective radius and SSA. Therefore, aerosols absorptive capacity increased, and changes of spectral properties of SSA and RI real and imaginary parts were observed (Fig. 6). spectral characteristics changed during that period, increasing the absorption capacity of aerosol, especially in longer wavelengths. During August 15-17 we observed
- 25 relatively large SSA values. According to Eck et al. (2009), the observed increase of SSA can explained by an increased particle size caused by wildfires, which in turn increased the total reflectance in the atmospheric column. Microphysical properties of aerosol particles over Kyiv under the influence of intense fires correspond well with general characteristics of biomass burning and polluted continental aerosols aerosol, as derived from AERONET sun-photometers sunphotometer measurements (Dubovik et al., 2002; Omar et al., 2005, 2009). Both aerosol types are mainly spherical, their size distributions are dominated by the
- 30 fine mode, and they are both moderately absorbing. In particular these types of aerosols are not separated in the algorithms of satellite measurements. Particularly the CALIOP algorithm exploits the probability that smoke aerosols are likely to be lofted, leaving unresolved cases where the smoke is in the boundary layer. Because the main objective of the algorithm is to select an appropriate lidar ratio, a potential misinterpretation of these two types has no effect on the retrieved extinction because both types are assigned the same aerosol extinction-to-backscatter ratio of about 70 sr (Omar et al., 2009).

Witte et al. (2011) showed AOD 550 nm measured by MODIS and SSA (388 nm) measured by OMI, averaged over the active fire period from July 22 to August 18, 2010 for domains covering territory of Ukraine and neighboring areas (domain 9: 45–52N and 23–33; domain 10: 45–52N and 33–43E). The SSA is retrieved on assumption that black carbon and organic components dominate in aerosols, thus SSA can be overestimated for urban areas, and the vertical aerosols distribution has the

- 5 shape of Gaussian with the maximum at the altitude of 3 km. Our analysis of vertical profiles of extinction coefficient for the cases of significant aerosols pollution indicates that this is assumption is unrealistic. Witte et al. (2011) found SSA = 0.98±0.02 (domain 9) that are much higher than our data received over Kyiv for the same time period (Fig. 6c). Nevertheless WitteOur comparison of AOD measurements from MODIS and AERONET showed strong functional relations between the datasets with Pearson's conclusion "… Decreased OMI SSA (increased aerosolabsorption) is consistent with increased smoke", agrees
- 10 well with our results. And Witte's AOD s correlation coefficients of 0.96 for MODIS/Aqua and 0.98 for MODIS/Terra. AOD measured by MODIS is therefore well captured for the entire Ukraine. Both MODIS/Aqua and MODIS/Terra represent the aerosol content in the atmosphere over Ukraine for summer 2010 within measurement uncertainties of around 0.15 standard deviation of AOD. The spatial distribution of MODIS AOD revealed that the wildfires of summer 2010 significantly impacted the eastern, central, and southern parts of Ukraine. The AOD at 550 nm of 0.34±nm reached values of 2 (and more) at certain
- 15 sites, especially in the middle of August.

Our comparison of AOD between CALIOP and MODIS revealed that the correlation coefficient was not larger than 0.6 is representative for the investigated area, but obviously too low between datasets. Over Ukraine CALIOP mainly underestimated the AOD in comparison with MODIS for the entire summer 2010. This can be explained by findings of Kittaka et al. (2011), who showed that the CALIOP algorithm likely ignores tenuous aerosol, causing an underestimation of AOD in comparison

- 20 with mean value for Kyiv (0.45). For eastern Ukraine (domain 10), Witte et al. (2011) shows SSA = 0.97±0.02 and AOD 550 nm = 0.44±0.26, that are matching well with our conclusion that fires caused the largest and most significant impact on this part of Ukraine. For comparison: average values at Kyiv AERONET site for SSA (440 nm) in pre-fires period were equal to 0.96 and 0.94 during active fires; for SSA (870 nm) were 0.95 and 0.91 respectively (Fig. 6c). MODIS. They also found that the aerosol layer's base height can be detected at higher altitudes, leading to an AOD underestimation.
- 25 Chubarova et al. (2012) estimated the impact of forest fires during summer 2010 on aerosols properties over Moscow according to AERONET measurements by comparing averages over periods of active fires and for the decade 2001–2010. During fire periods both the relative content of fine mode particles and the geometric mean radius of their size distributions significantly increased. This is in agreement with our results and conclusions of the other authors for different geographic regions (Dubovik et al., 2002) and (Eck et al., 2009). Also, Chubarova et al. (2012) found an increase in the real part of the refractive
- 30 indices and a decrease of imaginary parts. This results in larger SSA values in the visible and infrared spectrum. This is contradictory to our results (Fig. 6c, d), but agrees with Eck et al. (2009), where this relationship is explained by increase of particles size, that results in increasing the total reflectance of particles in the atmospheric column.

Finally, as the measurements in Despite the uncertainties and sparse availability of CALIOP measurements for the two above-mentioned studies were performed close to time and regions we analyzed, spatial distributions of AOD from CALIOP

35 measurements over Ukraine corresponded well with those from MODIS, in accordance with Kittaka et al. (2011). Another

advantage is that CALIOP also measures at nighttime. According to CALIOP observations, the sources of combustion, the transport of air from day- and nighttime AOD did not differ distinctly from each other during the analyzed period. This also corresponds to the global scale analysis of Kittaka et al. (2011).

We also analyzed aerosol profiles provided by CALIOP, which is the only source of aerosol vertical distribution for our study.

5 We found that the burning cells to Kyiv which took a significant amount of time, resulted in the transformation of aerosol was distributed at altitudes from about 40 m to 5 km and the extinction coefficient mostly ranged from some tenth to 1 km⁻¹, although sometimes it exceeded 8 km⁻¹ in very dense plumes.

Summarising, in this study we provided evidence of reasonable agreement between different types of aerosol measurements over Ukraine for the unique period in summer 2010. Further studies are needed to investigate the influence of the aerosols

- 10 properties and different fire regions on the air quality over Ukraine, which in our study could not be resolved well from the partly sparse coincidence of the composition of particles/datasets that are available until now. Not only other satellite instruments can be taken into account to further improve the accuracy of pollution levels analysis. The expansion of the ground-based sunphotometer network and in particular the availability of in situ observations would help, for instance, to resolve the large spatial gradients of the pollution levels that have been found over relatively densely populated areas.
- 15 Data availability. The AERONET dataset of AOD, AE, aerosol size distribution, SSA, and RI are available at https://aeronet.gsfc.nasa.gov. MODIS (Terra and Aqua) AOD dataset is available at https://ladsweb.modaps.eosdis.nasa.gov/search/. Dataset of spatio-temporal fires distribution is available at ftp://fuoco.geog.umd.edu/. Username:fire, password:burnt CALIOP dataset of Level 2 Cloud and Aerosol Layer and Profile products V 3.01 and 3.02 are available at https://eosweb.larc.nasa.gov/ project/calipso/calipso_table.
- 20 HYSPLIT model is available at https://ready.arl.noaa.gov/HYSPLIT.php; results of HYSPLIT simulations presented in this manuscript are available upon request from the authors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This publication includes results of the research that is performed in Astronomical observatory of Taras Shevchenko National university of Kyiv, project 160023-01Ukraine, project 160023-01, funded by the Ministry of Education and Science of Ukraine.

25 Parts of the research has have been funded by the University and State of Bremen.

We thank the (PI investigators) We thank PI investigators and their staff for establishing and maintaining the Minsk, Moscow, Toravere, Belsk, Chisinau, Cluj-Napoca, Bucharest, Eforie, Kyiy, and Sevastopol AERONET sites used in this investigation.

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