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Atmospheric Measurement Techniques Discussions

AMTD

2, C1415–C1431, 2010

Interactive Comment

Interactive comment on "Optical properties of different aerosol types: seven years of combined Raman- elastic backscatter lidar measurements in Thessaloniki, Greece" *by* E. Giannakaki et al.

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I am not quite sure if AMT is the right journal for this contribution since no new technique is presented. Why didn't you choose a journal that is more related to the presentation of measurement results and new findings?

Reviewer's opinion regarding AMT is consistent with the AMT home page statement (Aims and Scope section), reading: "The main subject areas comprise the development, intercomparison and validation of measurement instruments and techniques of data processing…". However, the same statement follows: "…and information retrieval

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Interactive Discussion

Discussion Paper

for gases, aerosols, and clouds". Following the introductory AMT paragraph ("Atmospheric Measurement Techniques (AMT) is an international scientific journal dedicated to the publication and discussion of advances in remote sensing, in-situ and laboratory measurement techniques for the constituents and properties of the Earth's atmosphere"), we believe that AMT is a new journal that should include research articles on advanced lidar techniques for aerosol characterization, therefore we believe that is an appropriate journal for the current contribution.

Before accepting the paper I would recommend major revisions concerning the methodology applied in the study. It seems like no attention has been paid to a seasonal dependence of the observed parameters except for the optical depth. The assumption of a well-mixed boundary layer up to a height of 1.5 km in winter for the classification of the kind or origin of the present aerosol species seems airy. Furthermore, column mean values are used when the properties of certain kinds of aerosols are discussed. Thus, the height resolved lidar observations are sold under value since such a study could easily be performed with a sunphotometer. The advantages of lidar measurements are not fully utilized in this study. I suggest that the authors focus on the analysis of free-tropospheric aerosol layers since anthropogenic aerosol is omnipresent in the boundary layer at such an urban site. Regarding the style of the manuscript, I suggest a careful cleaning from redundant content. Papers are cited with little system and sometimes without relation just to fill the list of references. The phrasing needs to be simplified to form shorter and more precise sentences instead of constructions that expand over 3-4 lines.

Reviewer's comments raised in this introductory paragraph are repeated under the General Remarks section and are analytically addressed by the authors in the following.

General remarks:

Sentences are phrased too detailed and often too long, especially in the Abstract and

AMTD

2, C1415–C1431, 2010

Interactive Comment

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Interactive Discussion

Discussion Paper

the Introduction. Please use short and plain sentences. Redundant phrasing should be avoided to keep the paper clear and readable. Please screen the paper for needless text. Many sentences could be shortened.

Many sentences have now been shortened. Abstract and introduction have been rephrased. A lot of sentences have been deleted and instead of the removed text, appropriate references are given.

The use of quotation marks is disturbing since it implies (at least to me) that something else is meant.

The quotation marks were deleted.

Citations are unstructured and sometimes inappropriate.

Citations have been updated.

Some aspects of the study reduce the height resolved lidar measurements to column mean values not different from what is derived by applying a sunphotometer. Why is the additional information so carelessly wasted? A discussion of the general vertical extend of the aerosol layer over the measurement site and its dependence on the time of the year would have been a valueable information.

The scope of this paper is to present the variability of the lidar-derived optical parameters over Thessaloniki, focusing on the grouping of the measurements in relation with different air flows possibly related with specific aerosol sources. The effect of the different aerosol loads due to advection from distinct sources is then described. The philosophy of our approach is based on the statistical description of the aerosol load over our site. A study focusing on the vertical extend of the aerosol layers in dependence with time is a future work already pronounced in the current paper.

Maybe you should focus on free-tropospheric layers only to screen your profiles from local influences and achieve a more suitable aerosol type classification. Furthermore, you would avoid uncertainties introduced by an incomplete overlap. Your mean OD of 0.52 for clean conditions suggests a huge contribution from the planetary boundary layer that might influence all your profiles.

A detailed analysis of free-tropopsheric layers regarding the optical and geometrical properties is the scope of a future study already announced in this paper. Uncertainties of the incomplete overlap are mainly affect backscatter coefficient at 532 nm and thus Ångström exponent values. Following the suggestion of referee #1 we have revised the cases with large Ångström exponents. We have excluded these cases were overlap seems to affect the measurement in heights above 1.5 km. The statement that optical depth of 0.52 stands for clean conditions is not right and indeed suggests a huge contribution from the planetary boundary layer that influence all the profile. Although the fact that extinction coefficient values do not differ a lot between continental polluted and continental clean classes, the correlation plot between lidar ratio and Ångström exponent values is characteristic for the second class. This is the reason that we want to keep these cases in a different class as they associated with certain meteorological conditions. However, we now call this class as continental NW with less free-tropospheric contribution.

Specific remarks: 3029, line 8-10: This is hard to believe if you consider industrial regions in China and India, biomass burning in the Amazon or Central Africa, and the regions of strong influence of mineral dust like Western Africa and the Arabian Peninsula. Is there also newer literature to proof this statement?

We have deleted the sentence: "Aerosol optical depth derived from the Advanced Very High Resolution Radiometer (AVHRR) classified the Mediterranean Sea as one of the areas with the highest aerosol optical depths on the world (Husar et al., 1997)". Literature for the geographic area of our interest is given.

3029, line 17: Rather use vertical than spatial to state the advantages of lidar measurements. The whole sentence needs to be simplified.

We have rephrased the whole sentence as: "High vertical and temporal resolution of lidar measurements give detail information on the aerosol structures"

3029, line 24: Wrong citation: it is Ansmann 1992 as is correctly stated in the References section.

The reference has been corrected

3029, line 24: What about particle shape?

We have rephrased the sentence as: "The lidar ratio contains information on the aerosol type, since it depends on the index of refraction and on the size and shape of particles"

3029, line 26: Skip Ansmann 1992 and Müller 2002, 2005 (the latter deal with microphysical properties!) and rather cite Müller 2007: Aerosol-type dependent lidar ratios!

We have deleted the references of Ansmann et al., 1992 and Müller et al., 2002, 2003, 2005 and we have included the reference of Müller et al. 2007.

3029, line 28 and later: Skip the vertical in front of profiles. The reader should be aware of this by now. The same holds for the repeated reference to your measurement site Thessaloniki, Greece.

When we are referring to profiles the word vertical is omitted from this point and later on.The measurement site of Thessaloniki, Greece has been deleted when is not needed.

3030, line 20: Is it possible to keep the description of the system as short as possible and refer to a published paper?

The paragraph describing the LAP system has been shortened. For the description of the system the reader can be refer to the paper of Amiridis et al. 2005.

3031, line 12: Why is the Angstöm exponent estimated and not simply calculated?

The sentence was rephrased as: "The backscatter related Ångstöm exponent can be calculated..."

3031, line 19: All profiles in Figure 2 go down to 500 m. How can that be if the overlap correction only allows for a retrieval down to 1000m?

The overlap correction has been applied following the method proposed by Wandinger and Ansmann, 2000. Overlap corrected profiles are kept for overlap factors greater than 0.7. In some cases, overlap corrections allow retrievals down to 500 m, even if these cases are rare. Mean profiles reported down to 500 m, were calculated by some of the above mentioned cases. Following the reviewer's suggestions, we present now in Figure 2 profiles down to 1 km only. However, we should notice that the mean profile down to 1 km has been retrieved by individual measurements that may or may not have information down to 1 km.

3031, line 21: This paragraph is confusing! Do the errors described in this paragraph originate from the retrieval intercomparison performed in the framework of EARLINET (citation of Matthias 2004a is inappropriate!) or are these estimations of your real measurement errors? If these are estimations of errors of your system, why is the error for Raman backscatter larger than the one for the Klett solution and why is the error for backscatter larger than the one for extinction? If you want to discuss the errors of your retrieved products, the reference to the EARLINET retrieval intercomparison seems to be redundant.

Considering the errors in lidar algorithms, what we mention in the text is not the actual error in the estimation of the backscatter coefficient with Klett method but the deviation of our estimates from the solution in the frame of the intercomparison exercises that took place within EARLINET (see cited papers). Following the reviewer's suggestion, the following information has been added: "Typical statistical errors due to the signals detection are below 10% in the PBL for backscatter and extinction coefficients at 355 nm. In the free troposphere, typical backscatter coefficient errors at 355 nm are below 30% for values higher than 0.25 Mm-1sr-1. For extinction coefficient at 355 nm, errors are below 30% in the free troposphere for values higher than about 8 Mm-1. For backscatter coefficient at 532 nm additional uncertainties are being introduced by the fact that an unknown lidar ratio has to be assumed. Both the system and algorithms used have been quality checked within the EARLINET”

3032, line 10: How do you account for the change of PBL top height with season of year? Does the PBL top in Thessaloniki reach 1500m in winter (and in the evening)? A modulation of the arrival height of your trajectory with season of year seems more appropriate. Did you check individual measurement for this assumption? If so, mention it in the text.

The PBL over Thessaloniki experiences large variations due to season of the year as it is pointed by the reviewer (approximately between 200 m/winter to 2500 m/summer). Our classification is based on the fact that we have observed the maximum of the mean backscatter profile (from all years and seasons) at 1500m. Accordingly, we chose to classify our measurements by clustering 4-days back-trajectories arriving over Thessaloniki at the same height, of 1500m. However, in the second classification followed in our paper, the 4-days back-trajectories have been calculated at 6 different arrival heights and the classification has been based on that height were each aerosol layer is observed. The six different arrival height approach is now mentioned in the text in section 2.2.

3032, line 17: This whole paragraph is redundant and can be shortened or dropped. A simple reference to the DREAM model is sufficient.

The paragraph was deleted and we just give the reference of Nickovic et al., 2001.

3033, line 1: Just the facts! Keep this paragraph short and do not describe pictures that are not shown.

The paragraph was deleted and we only mention the web-site from where someone can download the data.

3033, line 13: What about the Monday noon measurement?

We have corrected the text, describing EARLINET routine measurements that performed three times per week. However, our intention is to focus only on the nighttime Raman measurements.

3033, line 22/ Figure 1: This seems a bit too general. How are these mean values calculated? Do you only consider regions of vertically constant aerosol burden or do you integrate the profile and divide it by the aerosol layer depth? How representative is this mean value if separate aerosol layers occur?

The mean extinction coefficient values reported in the text are the averaged values of the extinction for each day, from the height of 1000m up to the reference height. In this point we only want to present our measurements in a statistical base. As already mentioned, our intention (in the near future) is to perform a statistical analysis on free tropospheric layers (both for geometrical features and optical properties).

3034, line 8: an unknown backscatter coefficient

The word known was replaced by the word unknown

3035, line 13: If you calculate a vertical mean lidar ratio for each measurement (and thus simplify your temporally and vertically highly resolved measurements to a single value) you loose all the structures that might show up in the lidar profiles. That does not seem to be desirable. I am not sure if you really can extract any information of a height average of a mean profile representing a time range of seven years! This might include too many different weather conditions. Does the standard deviation of the lidar ratio of 22sr originate in a strong vertical (for each individual measurement) or temporal (different means of individual measurements) variation of your measured parameters? The lidar ratio should by highly variable for individual measurements at your site. How trustworthy are these mean values if the lowermost 1000m of the column above the measurement site are not covered?

The purpose of section 3.1 is to present our measurement in terms of timeseries and frequency distribution plots. In the last paragraph of this section, vertical mean values of lidar ratio are presented, along with standard deviation. In our opinion, these values give to the reader a quick impression on the range of optical parameters detected in our site. Considering the lidar ratio variability of 22 sr, this could be considered as an indicator of the presence of different aerosol types (as already mentioned in the text) and a further presentation (section 3.2) and analysis (sections 3.3 – 3.4) of different aerosol types is performed later in the study to address the points raised by the reviewer.

3036, line 4: Parts of this paragraph were mentioned early and might not need to be repeated. See notes 3032, line 10. The part describing the different clusters could be shortened, especially since it is mentioned later that this method is not suitable for aerosol discrimination.

We have removed all the repeated information. However we have kept the text which describes the main directions of air masses over Thessaloniki with basic statistical information of optical properties for each cluster. Cluster analysis is in fact inadequate for aerosol discrimination, but on the other hand is a tool that (in conjunction with the sources) can help characterizing the aerosol loads over a region.

3036, line 26: Why do the mean values for the different clusters show such a large standard variation? I would expect more homogeneous conditions for properly selected clusters. Maybe the selection of distinct clusters not automatically implies a selection of different aerosol types. As you mention in the next paragraph, all this seems pretty vague. Do not waste to much time on something you later assess to be obviously insufficient.

Cluster analysis in general, describes the major air flows that could potentially advect particles over a site. The clusters computed are strictly related with the meteorology of the site. Unfortunately for our station, the surrounding (geographically known) aerosol sources are not distributed according to the meteorology, and this is the reason why some of the sources affect more than one cluster. Therefore, mean values for the different clusters show large variations, since different aerosol types from a variety of sources are included in each cluster. We think that the cluster analysis is the first step to associate the behavior of our measurements with meteorology. Then, an attempt is been taken to describe the variability of measured parameters on different aerosol loads and types, related with the potential sources that each cluster may include. We believe that the methodology followed to analyze our data in terms of meteorological grouping and association with potential aerosol contribution from distinct sources should be presented and is useful for an unfamiliar with our site reader. We agree with the reviewer that additional tools are needed for the attribution of our measurements to specific aerosol types. However, the statistical information presented here is also descriptive for our site, presenting 7-years lidar climatology. It is also true that for a site like Thessaloniki with mixed aerosol pollution the discrimination of different aerosol types is extremely difficult. However, lidar measurements are a powerful tool for such an attempt, and this is the scope of the pronounced future work, where we will try to fulfill this task.

3038, line 5: You observe higher wind speed under the influence of high pressure systems? And what is meant with katabatic vertical motion? Subsiding cold air under high pressure? Katabatic wind usually is associated with downslope motion of cold air at mountain ranges.

Westerly and Northwesterly flows were characterized by high wind speeds and showed a katabatic motion in the vertical (indicated by the trajectory vertical paths). The katabatic vertical motion of these air masses over Thessaloniki indicated that these were not modified significantly by anthropogenic activities while traveling across Europe, since they (most probably) did not reach the boundary layer over the continent to be affected by such pollution.

3038, line 6: Why is it clean continental and continental polluted? Is there also clean maritime or should it be continental clean?

Maritime aerosol type is out of our conversation, following reviewer's next comment of the reviewer. The continental clean and polluted cases, are distinguished obviously due to the differences on the lidar ratio and Angstrom exponent mean profiles for those categories. Cleaner air masses are assumed when free tropospheric contribution is lower.

3038, line 9: The data could easily be screened for the influence of maritime aerosols if you neglect the lowermost 1.0-1.5 km of the aerosol column (being your range of incomplete overlap).

This is true, we have neglect the first kilometer, so the dataset is not contaminated by marine aerosols. The argument has been removed.

3038, line 16: In the remaining part of the paragraph you state that an observation of distinct aerosol types is hardly possible at your site since usually different types of aerosols are present. However, you aim at obtaining the lidar ratio of these key aerosol types that cannot be distinguished clearly. Focussing on specific events with clear aerosol conditions seems to be more promising. The announced analysis of individual aerosol layers would be more appropriate.

This is exactly the scope of our future study announced in the present one. However, using cluster analysis and knowing the main aerosol sources for our site, we present in this paper the contribution of different aerosol types on the measured optical properties. See also answer to the comment pg 3036, line 26

3039, line 5: How can the mean profiles in Figure 2 reach down to 500 m when your overlap effect only permits reliable values down to 1000 m (see instrumental section of your paper)?

Figure 2 gives the profiles of the optical properties down to the 1st km (now). However, we should notice that overlap corrections allow us to retrieve reliable profiles also down to 500m (following the approach of Wandinger & Ansmann, 2000). These profiles have been cut off to 1000m for homogeneity, following reviewer's suggestion. (see also answer to comment pg 3031, line 19)

3039, line 23: How can you separate the particle size of different aerosol types when you mention earlier that a clear classification of individual profiles to one distinct aerosol type is hardly possible? The Angström exponents of Saharan dust should be well below unity. The values you show in Figure 2 represent rather mixed conditions.

Our concept is to present our lidar measurements in a way that these measurements would be representative of the air flows over our site. These measurements represent in fact climatological values for certain meteorological conditions over our site, indicating additionally the possible contributions of different aerosol types from sources related with the air flows. The complexity of the aerosol load over our site makes it hard to distinguish different aerosol types, however is possible to distinguish different signatures, as we mention in our paper. In our opinion, reported values for distinct aerosol type (e.g. mineral aerosol) should be trusted if the measurements are performed in the Sahara desert. However, these particles are travelling in long distances, they are mixed with other aerosol types etc., and the mixed conditions are described in this paper, taking advantage of the complexity of our site.

3040, line 7: OD 0.60-0.75 is moderate? This is heavily polluted in most places! Your statement might become clearer if you discuss it with respect to the annual mean OD observed in Thessaloniki.

“Moderate” has been removed following the reviewer's suggestion, since these values are greater than the annual AOD mean values observed in Thessaloniki.

3040, line 13: OD 0.52 is usually not clean. Local sources seem to influence your measurement. Did you compute trajectories for different height levels or just at 1500 m as stated earlier? I would suggest that you focus on free-tropospheric layers only (see general remarks).

AMTD

2, C1415–C1431, 2010

The statement that optical depth of 0.52 stands for clean conditions is not right and indeed suggests a huge contribution from the planetary boundary layer that influence all the profile. Although the fact that extinction coefficient values do not differ a lot between continental polluted and continental clean classes, the correlation plot between lidar ratio and Ångström exponent values is characteristic for the second class. This is the reason that we want to keep these cases in different classes. The measurements that belong to the second class are associated with certain meteorological conditions (the back-trajectories show that air masses originate in high altitudes from West and Northwest directions and descend in lower height as approaching our place). To avoid confusion, we now call this class as continental NW with less free-tropospheric contribution. The classification of measurements with cluster analysis algorithm has been done only for the height of 1500m. However, in the second classification the 4-days back-trajectories have been calculated at 6 different arrival heights and the classification has been based on that height were the aerosol layer is observed. The following statements have been added to the text. “In addition, the atmospheric trajectories are calculated at six arrival heights of 0.5, 1.5, 2.5, 3.5, 5 and 7 km and for arrival time of 19:00 UTC, which corresponds approximately to the time of the the routine measurement.” (section 2.2). “In that point we also checked trajectories in several arrival heights and the classification of each measurement has been based on that height were aerosol layers are observed.” (section 3.2).

3040, line 18: Is this the same data as presented in Figure 2 or do you now show vertical mean values of individual measurements? In the first case your data look wrong. However, your data contain the information on boundary layer aerosol. You should screen this influence by only considering free-tropospheric aerosols.

In Figure 3 (concerning the comment pg 3040 line 18) we present vertical mean values of individual measurements averaged in height ranges above 1.5km (figure 3 does not change significantly if we consider heights above 2 km). Our averaging is consistent with reviewer’s suggestion. In figure 2 we present mean vertical profiles for each category (that is averaging all the profiles that belong in this category for each height interval).

3041, line 6: Are such large Angström exponents for Saharan dust reported in the literature?

As we already mentioned in previous comments, the Saharan dust layers may be contaminated with other aerosol types as well. However, we believe that the great Angstroms reported here are indicative for this mixing. Mineral particles can co-exist with locally emitted aerosols, and these cases are also reported in our plots.

3041, line 8: These are bad comparisons! You basically compare your findings to the possible range of lidar ratios.

As we mentioned previously, the Saharan dust cases reported here, are associated also with other aerosol types. Our results indicate that such mixtures show lidar ration in the range of (indeed) possible lidar ratios for all the aerosols probed. Similar results have been presented by Mona et al., (2006) and Balis et al., (2004) for Mediterranean lidar stations.

3041, line 25: That is a vague statement. Do your 21 biomass burning cases show vertical variability of your parameters or is it always spread across the whole column?

As we mention in the text, for smoke advection from the North East, we observe homogeneous distributions of the lidar ratio and Angstroem exponents across the whole column, and this is exactly what we are saying in line 25 (lack of significant vertical variability).

3042, line 4: Isn't gas-to-particle conversion associated with new particle formation? 3042, line 5: What is meant with "condensation of large organic particles from their gas phase. . ."? 3042, line 8 to the end of the paragraph is confusingly written. Considering this paragraph, we just reporting the possible physical and chemical processes that could be responsible for the increase of the smoke particle size due to aging pro-

AMTD

2, C1415–C1431, 2010

Interactive Comment

cesses. Specifically for the statements: "gas-to-particle conversion": this is a possible physicochemical process that can lead to new particle formation from gasses released by fires. "condensation of large organic particles", we do not state that at any point of the text, rather than "condensation of large organic molecules". Related references describing the above processes are given here and to Amiridis et al., 2009.

3042, line 10: Are backscatter-related Angström exponents compared?

We are not comparing absolute Angstrom exponents retrieved by our measurements with other studies, since the particle size and correspondingly Angstroms can differ between smoke masses from different fires. We compare however our findings considering the fact that Angstrom exponents show a decrease with age of the plumes.

3042, line 24: It is hard to draw any conclusion if you always consider the whole column and drop the advantages of lidar measurements. A statement like this can also be drawn from combined sunphotometer/backscatter lidar measurements.

This is a study that is concentrated on Raman lidar measurements focusing on the maximum of information can be retrieved from such a lidar. The advantages of Raman lidar technique are well known and will be used in our future work. However, in our opinion, is not a disadvantage of this paper the fact that "statements like this can also be drawn from combined sunphotometer/backscatter lidar measurements". Then, our results are correctly compared with sunphotometric retrievals from Catrall et al., (2005).

3043, line 3: What is meant by significantly modified? Somehow pollution (OD=0.5!) must be injected into these air masses. It is hard to see a difference in the extinction values (Figure 2) of continental clean and continental polluted.

We believe that the pollution is of local origin, since the free tropospheric contribution seems to be minimum for these cases (continental 1 and 2). The continental cases have been distinguished not due to (indeed) minor differences on the extinction profiles but due to greater differences in the backscatter profiles and significant differences in

AMTD
2, C1415–C1431, 2010

the lidar ratio and Angstrom mean profiles. This is evident on Figure 2. We now change the name of continental clean class.

3043, line 8: Please shorten your conclusion section.

The conclusion has been shortened

3044, line 12: That is no finding. It is known that the lidar ratio is size dependent.

The statement has been removed.

3044, line 17: You say smoke particles are larger than dust particles? The lidar ratio increases with decreasing particle size. What about the influence of absorption?

Basically, we say that larger particles are found for Saharan dust (pg3044, line 1). However, the sentence has been rephrased in order to be clear: For smoke, the large values of lidar ratios are associated with larger particles possible due to aging of smoke along their trajectory from the source region to Thessaloniki

3044, line 26: CALIPSO operates at 532 nm, your system at 355 nm!

This is true. The statement was removed. However, our statement was referring to the fact that such methodologies presenting possible lidar ratios in relation with angstrom exponents, can be valuable for CALIPSO (of course for 532nm).

3044, line27: You mention that a clear separation of aerosol species is hardly possible at your site. So how can you generalize your findings for distinct aerosol types?

The reviewer's argument has been already answered in previous comments (e.g. comment 3039, line 23).

3045, line 1: In this paragraph you state that you cannot discern between different aerosol types for the whole column since you always observe anthropogenic aerosol in the boundary layer. The announced detailed analysis sounds much more promising than this paper.

AMTD

2, C1415–C1431, 2010

The text from pg 3044, line 23 to pg 3045, line 6 has been deleted. In this way we shorten our conclusion and we state only in our findings.

References: Please shorten all author lists of more than 10 contributors to XXXXX et al. Carefully screen your list for unused references.

We have screened our reference list for unused references. Also, for references with more than 10 contributors we refer to this as XXX et al.

3047, line 19: Is this paper cited in the paper?

No, so we have deleted from the reference list.

Figure 1, upper left picture: adjust the unit of the mean extinction coefficient. The unit of mean extinction coefficient has been corrected

Figure 2 State the number of cases in the graphs. Why do the profiles reach down to 500 m? It is sufficient if you show profiles up to a height of 5 km. I would also adjust the axes of the backscatter coefficient plots to 20 and 6 Mmˆ-1 srˆ-1, respectively. The lidar ratio plot should only reach to 100 sr. AE from 0 to 4. The Angström exponent for Saharan dust seems too large.

The units were adjusted as proposed by the referee and the numbers of cases were also stated. The profiles now reach 1000m.

Figure 3 State the number of cases in the graphs. Why don't you use height-resolved correlation plots? The vertical mean includes the boundary layer that might be purely anthropogenic at your site.

We state now the number of the cases in the graphs. These graphs are derived from vertical averaged values from 1.5 km up to the reference heights. If we consider mean values from 2km up to the reference height the figure doesn't change significantly.

Interactive comment on Atmos. Meas. Tech. Discuss., 2, 3027, 2009.