Authors' response to reviewer #3

We thank reviewer # 3 for carefully reading our manuscript and the provision of many comments and suggestions. They gave as useful hints where improvements of the paper were necessary to better understand our methodology and conclusions. However, some of the raised questions have already been discussed in the submitted paper at different places, and some are clearly beyond the scope of the paper and/or cannot be resolved with the available data sets. Nevertheless we have considered all comments of reviewer #3. Our replies are given in italics.

The changes (revised version vs. AMTD-paper) are highlighted as displayed by latexdiff ("diff.pdf", maybe renamed when uploaded as a supplement). For the sake of clarity only small changes are explicitly mentioned in our point by point replies, otherwise we refer to the corresponding parts of "diff.pdf" (in blue). Note, that some of our responses interact with comments of the other reviewers, so sometimes it is difficult to refer a change to one specific reviewer's comment.

Point by point replies

General comment

This is an interesting manuscript as it discusses the relationship of the mixing layer height (MLH) and near surface pollutant concentrations. The authors perform correlations of the MLH and PM10, NOx, and O3, and found varying results. The authors believe that the effects of the heterogeneity of the emission sources, chemical processing and mixing during transport exceed the differences due to different MLH retrievals. With regard to the use of the different MLH retrieval methods (Vaisala proprietary software, COBOLT), which are solely based on aerosol backscatter signal, I was wondering, if radiosondes have been used for a conclusive validation during the BAERLIN campaign.

→ *Intercomparisons between aerosol-based MLH retrievals (lidars, ceilometers) and retrievals based on temperature-, wind- or water vapor profiles (e.g. from radio sondes) have been carried out in several studies; some papers have been cited in the manuscript. COBOLT has been developed using ceilometer measurements in Munich and compared with radio sondes data of Oberschleißheim (distance 8 km only). So we don't feel that it is necessary to demonstrate this again in this*

paper. Moreover, it was not a goal of BAERLIN2014, and the closed radio sonde station is in Lindenberg, almost 60 km away from the ceilometer site!

Also, I was wondering why other methods such as the Haar wavelet method or a cluster method have not been considered/discussed.

→ *The Haar wavelet method is one component of COBOLT when the Sobel operator is applied (see new citation of Comeron et al., 2013). This is mentioned in the revised version of the manuscript when we provide a much more detailed description of COBOLT (according to a suggestion of reviewer #1, pages 11–14 of diff.pdf). Moreover, we had already included three citations in the original manuscript (Cohn and Angevine, 2000, Brooks, 2003, Baars et al., 2008) that use this wavelet covariance transform. Caicedo et al. (2017) who applied the cluster method are cited as well (see response to reviewer #1).*

With regard to the relation of the MLH with air quality, it is well known that the local change of any given pollutant is not only controlled by the MLH, but by a combination of emission, chemical transformation, removal, advection, convection and turbulent mixing. Also, it is known that at the microscale level urban structures cause flow disturbations and thus deviations from the mean air quality of a larger, representative fetch in an urban area. An example is the well-known wind rotor system in street canyons. Thus the relationship of the MLH with surface concentration critically depends on the fetch area representative for a given measurement site. These well-known processes are not properly addressed in the paper.

→ *We agree with the reviewer that surface concentrations of pollutants do not only depend on the MLH and that our paper is not the first that points out these facts. Accordingly we have mentioned these processes e.g. in Sect. 5.1 (p 20, l27 ff. of the AMTD-version, p 20, l32 ff. including citations) and in the conclusions (p 25, l29 of the AMTD-version). As a consequence of the comments of reviewer #3 we have extended this discussion. Moreover, we have added a paragraph to the introduction where we describe the objectives of our study more clearly (see page 3 of diff.pdf). This was indeed not clear enough in the submitted manuscript: we want to focus on the ceilometer retrieval and the potential over-interpretation of correlations. These aspects have not yet been covered in the literature.*

The reviewer's statement on the influence of "microscale level urban structures" certainly points out a very important aspect, which in sum would however have resulted in exploding project costs. Some aspects e.g. of chemical transformation and deposition can be reasonably well while not perfectly described by a chemical boxmodel. However, the vertical mixing aspect in such a model, determined by the MLH cannot be reproduced acceptably well without observations. The information provided by BAERLIN2014 supplies the effect of the MLH and therefore vertical exchange to the change of pollutants, i.e. the fraction of change that can be explained by meteorology.

Due to the rather flat larger area of Berlin, it can be expected that transport processes may play a dominant role in the distribution of pollutants, both at the mesoscale and microscale level. I am surprised to see that the authors did not consider any of the findings associated with the BERLIOZ experiment in 1998 (mostly published in Journal of Atmospheric Chemistry 42, 2002, but also others), which focused on the upwind-downwind conditions found for the Berlin case, as well as the pollutant concentrations within the boundary layer and aloft in the same area and the impact of long-range transport.

→ *The BERLIOZ campaign (Berlin Ozone Experiment) was a huge campaign focussed on the impact of Berlin on the surroundings. It never investigated Berlin itself but a northwest-southeast transect through Berlin approximately 50 km on either side in Brandenburg, such as e.g. Pabsthum. In contrast the focus of BAERLIN2014 was the metropolitan area of Berlin and Potsdam and the influence of vegetation inside this area in detail. Thus, one could use references and results of BERLIOZ for broader discussions only.*

Reviewer #3 seems to focus rather on large scale effects than on small scale mixing. Berlin is not affected by the surrounding countryside, somewhat more the opposite. This actually caused the BERLIOZ project nearly to fail, because the anticipated effects were hardly found (e.g. huge ozone plumes downwind, large PM_{10} clouds etc.). As stated earlier the idea of the reviewer to conduct investigations for Berlin and Brandenburg including (very) detailed experiments and modelling approaches is far off realistic financial and personnel limits.

In conclusion, we don't feel it necessary to include any outcome from BERLIOZ as the scientific objectives of that experiment were quite different from our study. Neither the derivation of the MLH from

ceilometer measurements was part of BERLIOZ nor the distribution of pollutants inside the city.

I would not expect an unambiguous relationship between the MLH and surface concentrations at any given location and under any given meteorological situation in the Berlin area. Rather, I would only expect a dominant role of the MLH on surface concentration, when advection is at a minimum, i.e. under stagnant wind conditions. In its current form this paper neglects the discussion of the MLH with regard to different wind regimes, both with regard to wind speed as wind direction. It should also be mentioned that not only pollutants can be transported, but also physical properties of the boundary layer including the MLH depending on the history of air masses. This extended in-depth analysis is a crucial requirement for a potential publication in AMT.

→ *We agree with the reviewer that advection plays a relevant role for the correlation between MLH and concentrations. This was briefly mentioned in the manuscript (see answer above). We have also elaborated this aspect in more detail in the revised version (pages 24–26 of diff.pdf) taking into account wind measurements of the German Weather Service at three sites in Berlin. We use these additional data set to select days when the wind was "predominantly stagnant". However, we want to emphasize that our mean diurnal cycles (MLH, concentrations) are averages over two months. So, the assessment of the contribution of a single process to the correlation between MLH and surface concentrations is hardly possible.*

More specific, mostly minor issues

- Page 2, L25-28: The paper by Czader et al. (2013) should be added as it is one of the earlier examples to use ceilometer derived MLHs for validation in conjunction with comprehensive air quality modeling.

  → *In Czader et al. (2013) we only find the reference to "remote sensing techniques" providing MLH at one site (Moody tower). Details were however found in Haman et al., 2012: here, CL31 measurements of almost two years have been evaluated for the diurnal cycle of the MLH in Houston, Texas. They use proprietary software of Vaisala. We have added both citations (pages 3 and 6).*

- Page 5, L18-19: I think both terms MLH and Hml mean the same. I suggest to use one term throughout the entire text.

  → *Our idea was to use MLH as "word" in the text, and H_{ml}, $H_{ml,v}$ etc. for mathematical expressions. We have checked this for consistency and changed it whenever necessary.*

- Page 5, L22: Please define what "width" would mean exactly: horizontal or vertical?

  → *Width is related to the MLH as derived from ceilometer measurements. As this could be misunderstood we changed the sentence to ...into intervals of 200 m. (see page 6 of diff.pdf).*

- Page 6, L9: Please define what is meant exactly by "secondary material"?

  → *We changed "material" to "secondary aerosol compounds", see page 6 of diff.pdf*

- Page 7, L1-3: "These data....in whole Germany". Is this statement important in understanding the contents of the paper? I suggest to remove it.

  → *We removed it as it is indeed not essential for the understanding. Anyway, for me it was an interesting information showing the extent of automatic air quality stations currently operational in Germany (see page 7 of diff.pdf).*

- Page 8, L4: What "information" is exactly meant?

  → *We have clarified this sentence: ...the option to combine in-situ measurements at the surface with data concerning the vertical direction (see page 9 of diff.pdf). The combination with aerosol optical depth would be another example. MLH is also useful to constrain model calculations as mentioned (see page 2 of diff.pdf).*

- Page 8, L6: Suggest to remove ", which is one hour different to UTC.", as UTC is not being used in the paper.

→ *This was included only as a explanation for readers who are more familiar with GMT. But we agree that it can be removed (see page 9 of diff.pdf).*

- Page 10, L16: Please explain what is actually meant by "cross-platform" here, and why it would be helpful?

  → *We wanted to emphasize that the code can be run on Windows and Linux platforms, so it is potentially useful for a large community. Moreover, Phyton is free of charge in contrast to e.g. MatLab. We have extended the whole section in accordance with the comments of reviewer #1; in this context we have also considered the comments of reviewer #3 (pages 11–14 of diff.pdf).*

- Page 15, L26: "Concentration measurements" of what?

  → *This could be "everything". In our study concentrations (PM_{10}, NO_x, O_3) are discussed but – if the corresponding data sets are available – the statement is also true for any other trace gas or e.g. $PM_{2.5}$. The sentence should only emphasize that problems may occur if data sets with low temporal resolution are considered during the rapid growth of the ML. To make this clearer we have substituted one word (page 18, line 21 on diff.pdf).*

- Page 15, L30: "The latter...(Pappalardo et al., 2014)". Please explain the schedule of EARLINET and explain whether the BAERLIN approach was important for the EARLINET approach or the other way round (which is more likely).

  → *The EARLINET schedule was defined in the year 2000. On the one hand it considers the diurnal cycle of the ML (measurements when the vertical extent is approximately constant for several hours) and on the other hand the performance of Raman lidars (they perform better during night). This was not influenced by our study, and our study is independent of the EARLINET schedule as we determine the full diurnal cycle. We only mentioned this because our (and similar) results confirmed that the selection made by EARLINET was reasonable (see COBOLT-retrieval shown in Fig. 5). For further illustration we have included Fig. 6. It shows the differences of the afternoon values of MLH when different MLH-retrievals are applied.*

To make this clearer we have modified the corresponding sentence as follows: The latter has been the reason for including a measurement around two hours after local noon in the regular EARLINET schedule (page 18 of diff.pdf).

- Page 15, L32-33: Please mention that these specific COBOLT results refer to the entire campaign period.

  → *We added ...for the whole period of 67 days (page 18 of diff.pdf).*

- Page 17, L11-12: What is exactly meant by "All measurements are performed under ambient conditions"? They way it is written it would mean that the air quality station was not air-conditioned.

  → *According to a comment of reviewer #2 we completely rephrased and re-arranged this paragraph. In this context we have also considered the comments of reviewer #3 and removed things that were not relevant in the context of our study (see page 7 of diff.pdf and answer to "Page 17, L15-16" below.*

- Page 17, L18: I think this "significant horizontal heterogeneity" refers to surface measurements here. Please clarify.

  → *Yes. Most of the measurements were made from bicycles. We have clarified this: Episodic mobile (bicycle) measurements from BAERLIN2014...(page 21 of diff.pdf)*

- Page 17, L15-16: What is exactly meant by "inorganic species": gas-phase, particle bound or both?

  → *Inorganic species refer to gaseous compounds like CO, NO and NO_2. The whole paragraph has however been rephrased and reorganized according to a suggestion of reviewer #2. Main parts have been moved to Sect. 3 (from page 21 of diff.pdf to page 7 of diff.pdf), and unnecessary information was deleted.*

- Page 17, L17: The reference "von Schneidemesser et al., 2017" is still in preparation and therefore not citable.

  → *We removed this citation and the text (lines before this citation) that was related to this paper which is currently still under preparation.*

- Page 17, L17-18: "Here we do not discuss these topics...". In this case please remove the preceding L13-17 as they are not within the scope of this paper.

  → *See our above response to the comment on "Page 17, L11-12"; the sentence was removed.*

- Page 18, L26-28: What is the justification for using these different correlations? The statistically most reliable quantity would be the median anyway, as it minimizes the impact of outliers. This is in particular true for such a quantity as PM10, which is mostly primarily emitted.

  → *We agree with the reviewer: that was the reason why we use the median in Figs. 7, 9 and 10 in the AMTD-manuscript. The same argumentation as the comment of the reviewer was given in the original manuscript (page 20, lines 6 ff). The different combinations of averages and medians as defined on page 18, lines 26 ff were only introduced to demonstrate the consequences on the correlations in the subsequent discussion. See also the new Fig. 7 (page 20 of diff.pdf).*

- Page 20, L23: "...with a lot of vegetation, a high density of buildings...". This sounds like a contradiction: where there is high density of buildings how can there be lot of vegetation at the same time?

  → *This description is made from the perspective of a German citizen. A "high density of buildings" does not mean that there is no space left for trees, bushes etc., often arranged as small "parks" of some tens of meters in length and width, or buildings organized as squares with trees inside a yard, to increase the quality of living. For example, southeast of the ceilometer is an area of approximately 100 × 70 m with "a lot of vegetation" whereas buildings dominate elsewhere. To avoid misunderstands we replace "high density of buildings" by "in a typical residential neighborhood in the inner part of a big German city; see page 24 of diff.pdf". A similar expression has also been used in Sect. 3.*

- Page 20, L17-18: The authors mention aerosol formation. Would PM10 data provide any indication for aerosol formation? If so, please explain.

→ *When we summarized our conclusions from Fig. 9, we mentioned different aspects that are responsible that no unique correlation coefficient (MLH vs. PM_{10}) has been found for entire Berlin. In this context only the absolute value of PM_{10} is relevant.*

- Page 20, L17: The authors mention that relative humidity may have an impact on PM10. Would PM10 concentration decrease or increase with relative humidity?

  → *The whole paragraph was significantly extended (see also replies to Reviewers #1 and #2) by including more investigations on correlations under special meteorological conditions (see pages 24–26 of diff.pdf). In this context the statement on the relative humidity became unnecessary (one can assume a small increase due to uptake of HNO_3).*

- Page 21, L1-2: What classes in addition would the authors recommend?

  → *We do not necessarily need more classes but the attribution might be reviewed. However we don't have any influence on this classification and the criteria for this classification. The same is true for the selection of the locations of the air quality stations. It is not unlikely that political reasons have a certain influence as well. We have added a short remark at the end of Sect. 5.1 (page 26 of diff.pdf).*

- Page 21, L2-4: This statement is obvious and has been considered in many urban air quality networks over many decades.

  → *This conclusion is indeed not unexpected. Nevertheless many publications do not clearly describe the conditions under which their correlation has been calculated or use only one site in a metropolitan area and leave it open how representative their conclusions are. So there remains room for misunderstandings, and we feel that it is justified/necessary to emphasize this statement (again). Accordingly we have expressed this objective in the updated introduction (see page 3 of diff.pdf).*

- Page 21, L31 - Page 23, L5: It is well-known that O3 can be mixed from the residual layer into the convective layer, also for the case of

Berlin (e.g. see BERLIOZ special issue in the Journal of Atmospheric Chemistry 42, 2002). The excellent correlation of the MLH with ozone in urban areas may not be surprising at all, as both processes are ultimately driven by incoming solar radiation provided there are sufficient precursors for O3 formation available. In other words the relation between the MLH and O3 is apparent, but not causally determined. This should be mentioned.

→ *We agree with the reviewer: we have used the same argumentation in that paragraph of the AMTD-version of the manuscript including the citation of a paper by Fallmann et al.; so it has already been mentioned. To make this clearer we slightly rephrased this paragraph (see page 27 bottom of diff.pdf).*

- Page 24, L7: "Whether ...studied". I suggest to remove this sentence. It is obvious that the potential impact of the MLH on ambient concentrations decreases with decreasing distance to the corresponding emission source.

  → *We have removed this sentence.*

- Page 24, L13: As I remember Xu et al. (2011) do not report MLH observations and thus no correlation with primary or secondary pollutants.

  → *Xu et al. (2011) discussed the influence of the MLH on surface concentrations of several trace gases in a general way. However, they did not use own measurements of the MLH. As a consequence we agree with the reviewer that this citation is not really relevant and dropped it.*

- Page 25, L10-12: I guess it is well-known that there is not one only parameter which controls surface concentrations.

  → *Again we agree with the reviewer, but our motivation was to investigate the role of the MLH-retrieval for correlation studies in view of its uncertainty and the inhomogeneity of urban air quality. This message was obviously not as clear as it should have been (see corresponding comments of all reviewers). As a consequence we have added a clear statement on our objectives to the introduction (see page 3 of diff.pdf).*

- Page 25, L27-28: In their paper the authors have tried to argue that MLH is not the only parameter which controls surface pollutant concentrations. Why then would it be of interest to perform a winter study in Berlin and why is it of importance that PM10 concentrations are 50% higher in winter compared to summer in Berlin. If there is no consistent correlation of MLH with PM10 in summertime why should it be different in wintertime?

  → *Reviewers #1 and #2 regret that only data from two months were available. This cannot be changed for obvious reasons. However, we believe (together with the reviewers) that a longer observation time would provide additional insight: in winter the MLH is expected to be shallower, the concentration of PM_{10} is larger, and the meteorological conditions (including atmospheric chemistry) are different. We do not expect that in winter the MLH is the only parameter that controls the concentration of pollutants, but it is not clear if the variability of R is more or less pronounced. It is scientific tradition to investigate any problem under different conditions if possible (see our comments on available resources). To point this out we have added an additional explanation: We do not expect that in winter the MLH is the only controlling parameter, but it is not clear if the correlation (and its variability) is more or less pronounced (see page 31, lines 17 ff of diff.pdf).*

- Page 26, L4-6: The authors state that MLH data is beneficial for box-model calculations and validation of chemistry transport models. While I would agree on the authors statement in this sentence I do not completely understand what the authors justification would be for this, since according to their paper the authors largely argue that there is no consistent correlation of the MLH with air pollutants. This should be clarified.

  → *In any case there are multiple counteracting processes merging in our findings as has been mentioned in the paper. As a consequence interpretation is much more complex than simply getting $R \approx \pm$ 1 for all times, but this does not reduce the usefulness of a reliable determination of the MLH. This was our statement in the last paragraph of our conclusions. We have clarified this by extending the conclusions on validation and combination of models and measurements (see page 31, bottom, of diff.pdf). It would,*

e.g., be nice to tackle the question of the homogeneity of the MLH over a city like Berlin by models and compare the results with distributed ceilometer measurements (see our corresponding replies to similar questions of all reviewers), maybe possible in future. Moreover, model calculations can help to understand the interaction and the relevance of different meteorological and chemical processes; in this context it could be useful to have independent measurements to validate at least parts of the model output (again a question of resources to set up a adequate field campaign).

Additional references (for more see also reply to reviewer #1):

- Czader, B. H., Li, X., and Rappenglueck, B.: CMAQ modeling and analysis of radicals, radical precursors and chemical transformations, J. Geophys. Res., 118, 11,376–11,387, doi: 10.1002/jgrd.50807, 2013.
- Haman, C. L., Lefer, B., and Morris, G. A.: Seasonal Variability in the Diurnal Evolution of the Boundary Layer in a Near-Coastal Urban Environment, J. Atmos. Oceanic Technol., 29, 697710, 2012.