Response to the second reviewer comment's:

The authors thank the second reviewer for the fruitful discussions through all the three rounds of comments and responses. The manuscript ends up with a much more collective point of view taking into account not only the aerosol research domain but also that of atmospheric dynamics. The co-editor and the reviewer will find thereafter the response to the last comments. It has to be noted that the page and line numbers relate to the third version submitted to ACP that was sent to the co-editor and editorial board on the 19th of July 2018.

The authors also add at the end of the response to the second reviewer comment's the response to the co-editor comment's sent on the 19th of July 2018.

After reading through the authors' replies and the careful comments by the editor I only have a few additional remarks. Most of the replies and corrections help to clarify the paper but I think with a little additional work some more meaningful results could have been obtained. But I will leave this for future work.

Some remaining concerns:

- DBinv: Since the authors are really so insisting on keeping this parameter in their analysis I suggest at least to change the name of it throughout the manuscript. Instead of 'drainage basin for thermally lifted pollutants', which I am still not convinced it represents, the authors should just stay with 'inverse drainage basin' and describe it as potentially reflecting the probability for thermally lifted pollutants to reach the site. Seems to be the same thing, but it really makes a difference if a confusing name is given or a topographic parameter is just tested. However, then in the conclusion it should also be made clear that the parameter was not a good predictor and, hence, the suggested analogy of inverse drainage flow and flow barriers did not work so well.
  The authors do agree to change the name for DBinv. The following modifications were done:
  - *p.2 line 29 : "To construct the ABL-TopoIndex, we rely on the criteria that the ABL influence will be low if the station is one of the highest points in the mountainous massif, if there is a large altitude difference between the station and the valleys or high plains, if the slopes around the station are steep, and finally if the inverse drainage basin potentially reflecting the source area for thermally lifted pollutants to reach the site is small."*
  - *P. 6 line 19: "4) the inverse drainage basin, which potentially reflects the source area for thermally lifted pollutants, is small. "*
  - *P. 7 line 33:" **Parameter 5 – DBinv:** Since the air masses have to be thermally lifted from the valleys and plains towards the summit to influence the station measurements, the size of the inverse drainage basin (DBinv) can be calculated with standard hydrology tools using an inverse topography, where the altitude Z is changed to –Z allowing the summit to become a hole. It potentially represents the region from which pollutants such as aerosols can be thermally lifted without crossing any topographical barrier. DBinv is related to criterion 4 for a large spatial scale (500 km x 500 km). Figures 4d and 5d are examples of the DBinv calculation for BEO and PYR. The ABL influence should increase with increasing size of the inverse drainage basin."*

- *P. 9 line 23: "Figure 4d shows that when the inverse drainage basin is calculated with the inverse topography, BEO is in the center of a large inverse drainage basin that covers most of the plotted domain."*
- *P.21 ine21:" The ABL-TopoIndex is a topographical index based on the hypsometric curve, the slope of the terrain around the station and the inverse drainage basin that potentially reflects the source area for thermally lifted pollutants"*
- *P. 38 line 4: "Figure 4: a) Topography on a 750x750 km^2 domain around BEO (Moussala, white dot) in Bulgaria. The main hydrologic flow paths from the station grid cell are given by the cyan lines. The color scale on the left only applies to Fig. 4a. b) hydrographical network, c) hydrologic drainage basins calculated from the real topography, the different drainage basins are defined by various colors and d) "inverse drainage basin" calculated from the inverse topography (DBinv)."*
- *P. 39 line 3:" Figure 6: a) ABL-TopoIndex, b) inverse drainage basin, c) hypsometric percentage of the station elevation, d) hypsometric percentage of the station elevation minus the 50% hypsometry, e) local slope in a circle of 10 km radius centered on the station, f) gradient in elevation as a function of the domain size for some European high altitude stations."*

To clearly indicate that DBinv is not a good predictor, the following sentence was added to the conclusion (p. 22 line 5):" The inverse drainage basin seems to be the least explanatory parameter in terms of ABL influence and this large scale parameter should either be further evaluated or be combined with a source inventory to increase its relevance for identifying boundary layer influence."

- Latitude and thermally induced transport: Sure latitude is a good predictor. The differential heating that is necessary to produce thermal lifting obviously depends, among other factors, on latitude. Why else are the authors focusing on summer for mid-latitude sites. The absence of strong differential heating is another factor why the results of ZEP are so poor. There simply is no thermal lifting.

  The authors do agree with this statement and add this information in p. 13 line 6:" ZEP, situated at very low altitude (475 m) and very high latitude (78.9°), also has a very high ABL-TopoIndex value. It was also not included in the correlation analysis since its seasonal and diurnal cycles exhibit different features than the high altitude or middle latitude stations (see Sect. 4.1)."

- Regression model: I was thinking about generalised additive models (GAM, Wood et al., 2006) and parameter selection like in Jackson et al. (2009). Certainly there is a way to deal with non-normal distributions if necessary. There are even more advanced methods coming up in the age of machine learning (random forests, etc.). But I can see that this goes beyond the scope of the current paper but it could have significantly improved the paper and removed some of the speculative aspects.

  The authors thank the reviewer for these very interesting references. They think however that the context between the cited study and the manuscript presents some substantial differences. The parameters selection for GAM in Jackson et al. (2009) is based on (to a large extent) already known chemical reactions with a good overall agreement achieved between

modelled and measured chemical species concentration. Our study is definitely a first step trying to understand the role of the topography in the ABL-influence at high altitude sites and the aerosol parameters used to validate the ABL-TopoIndex do not constitute the sole explanatory variables. Even though the authors do not have any experience with GAM, they expect at least two potential sources of problems:

> *1) an exponential family distribution has to be specified for the univariate response variable along with a link function (see https://en.wikipedia.org/wiki/Generalized_additive_model). As already specified in the second response to the referee comments, the only distribution describing the aerosol parameters is the Johnson distribution, which is not part of the exponential family distribution.*
>
> *2) the number of aerosol time series available for high altitude stations remains sparse (between 15 and 23 for the analyzed parameters) and could be a clear source of large uncertainties.*

Nonetheless,the approach is very interesting and could be included in future development of the ABL-TopoIndex.

References

Jackson, L. S., Carslaw, N., Carslaw, D. C., and Emmerson, K. M.: Modelling trends in OH radical concentrations using generalized generalized additive models, Atmos. Chem. Phys., 9, 2021–2033, doi:10.5194/acp-9-2021-2009, 2009.

Wood, S. N.: Generalized Additive Models: An Introduction with R. Chapman & Hall/CRC. Boca Raton, USA, ISBN 9781584884743, 2006.

Answer to the co-editor comment's:

The authors thank the co-editor for his comments; these are addressed in what follows:

- We are now at the stage that the one of the reviewers is satisfied with your responses and the revisions that have been made in the manuscripts in some iterations. However, the second reviewer still had some significant criticism and comments regarding your last revision and the response to the provided reviews. I have been going through all the files including the last review and your last response and the revised document. Based on this I invited the reviewer to indicate if this last round of revisions and your response has properly addressed the concerns/issues of the earlier version of the manuscript. In addition, reading over again the ms I still came across quite some statements, sentences that I had to read over again also not always being convinced that these were correct regarding grammar but also not always optimally expressing that what you would like to express. Below, you can find these points that came across and that also need to be further addressed in another

revision. I recommend you to ask one of the native English speaking co-authors to carefully check once more again the whole documents for the text to remove these flaws.

Elisabeth Andrews is the second author of this paper. She is a native English speaker with more than 60 publications in peer-reviewed journals. She has reviewed each version of the manuscript as well as all the responses to the referees' comments. She has also helped in addressing the language usage comments of the co-editor noted below. As usual, she will check the final version of the manuscript.

Major comments:

- Title; you have changed the title also considering the changes introduced in the ms based on the reviewers comments. However, this changed title is also according to me still not optimally covering the actual contents of the paper; especially the use of the term aerosol layer is not reflecting the contents. The suggested change in title by one of the reviewers seems to cover much better the actual content. Your study focuses on identifying the influence of topography on aerosol measurements at high-altitude stations.

  The authors agree to change the title to: "Identification of topographic features influencing aerosol observations at high altitude stations".

- In addition, it seems that have not tackled the issue of the reviewer on the use of global modelling products to also assess the role of topography in aerosol properties at higher elevations. I am myself not so convinced that the resolution is already sufficiently high to indeed use model products for this, at least not from global models. There are though meso-scale models that can resolve some of the fine-scale meteorological features at resolutions down to some km scale (e.g., WRF). My point is that you should at least address this remark by the reviewer even if at the end you decide not to include this aspect in your paper.

  The authors do agree that the modelling approach could provide additional information and would be a useful direction for future studies. While the second referee clearly would like to see a modelling component added to this current investigation, even s/he acknowledges in his/her second review that this may be beyond the scope of this paper saying:
  "I still think it would be possible to create a meteorological criterion that would indicate situations with likely thermally induced flow from existing global scale model products. Don't forget that the latter have resolution down to 0.1 degree by now. Also it has been done successfully before with observational data, so why not check with model data. ***However, I see that this may go well beyond the scope of the current analysis.****" Below we've expanded our response to this comment.*

  Most global models are typically not of sufficiently high resolution to simulate complex terrain (e.g., Benedetti et al., 2018). A preliminary study at MeteoSwiss (trainee work, personal communication) with the COSMO model shows that a higher degree of detail in the topography and surface fields leads to a better estimation of the convergence/divergence of the mesoscale flow over the Alps during the day/night (Alpine pumping). However, this was for limited locations in the Swiss Alps and for limited meteorological conditions (i.e., 14 clear-sky summer days). In contrast, the analysis presented in the manuscript aims to define an index that can be applied to all high altitude stations around the world. A study by Wang et al (2016) shows that, in addition to model resolution, the spatial resolution and distribution of emissions sources is also an important factor in how well models are able to represent observations of aerosol. We expect that complex topography would make high spatial

resolution of emissions even more critical. We feel that it is clearly beyond the scope of this paper to utilize output from one or more high resolution models (regional or global) to evaluate the aerosol observations presented here. Doing so would involve evaluating the model assumptions and parameterizations of aerosol and aerosol precursor emission distributions, aerosol transformation and removal processes as well as the details about how the meteorology is modelled for each high altitude station. The effects of mountain topography is complex to model and a topic of ongoing research (see for example, Serafin et al., 2018, Arnold et al., 2012) while models also struggle with simulating observations of aerosol particles - even in less complex terrain (see for example, Mann et al., 2014; Tsigaridis et al., 2014; Eskes et al 2018.). In the future we hope to partner with members of the modelling community to further explore our initial findings on how topography influences aerosol particle observations at high elevation sites.

Arnold, D., et al.:" Issues in high-resolution atmospheric modeling in complex topography – the HiRCot Workshop ", Croatian Meteorological Journal, 47, 3–11, 2012.

Eskes, H.J. et al.:" Validation report of the CAMS near-real time global atmospheric composition service, December 2017 - February 2018", Copernicus atmosphere monitoring service (CAMS) report, CAMS84_2015SC3_D84.1.1.11_2018DJF_v1.pdf, 2018. Available at HTTP://ATMOSPHERE.COPERNICUS.EU/QUARTERLY_VALIDATION_REPORTS

Benedetti, A. et al., "Status and future of Numerical Atmospheric Aerosol Prediction with a focus on data requirements," Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-42. 2018.

Mann, G. et al., "Intercomparison and evaluation of global aerosol microphysical properties among AeroCom models of a range of complexity," Atmos. Chem. Phys., 14, 4679–4713, 2014.

Serafin, S., et al. :"Exchange Processes in the Atmospheric Boundary layer Over Mountainous Terrain", Atmosphere, 9, 1–32. https://doi.org/10.3390/atmos9030102, 2018.

Tsigaridis, K. et al. "The AeroCom evaluation and intercomparison of organic aerosol in global models," Atmos. Chem. Phys., 14, 10845–10895, 2014.

Wang, R. et al., "Estimation of global black carbon direct radiative forcing and its uncertainty constrained by observations." J. Geophys. Res.: Atmospheres, 121, 5948–5971, 2016.

Minor comments:

- Page 6, line "non-GIS environment(Schwanghart and Scherler, 2014)" put space there.
  Done

- Page 6; line 20: "Based on these criteria, the red station on Fig. 2 will be less influenced by the ABL…", alternative; "Based on these criteria it can be inferred from Figure 2 that the "red" station will be less influenced by the ABL….
  The sentence was corrected as proposed by the co-editor.

- Page 7, line 2: "have hypso% *values* larger than 50%."
  The sentence was corrected as proposed by the co-editor

- Page 7, you refer in line 16 for example to small spatial scale and in the definition of the previous criteria to large spatial scale; here it is essential to indicate (again) what you deem being a small and a large spatial scale.
  The size of the spatial scale is now mentioned explicitly for each parameter constituting the ABL-TopoIndex.

- Page 7; line 23, what do you mean with "and there are some steps for CHC and BEO", what steps ? and steps of what size?
  The nature and significance of the "steps" is now detailed in the text:" For example, there is a rapid decrease of the altitude difference with increasing distance that gradually levels off for radius larger than 7 km for JFJ and for radius larger than 4 km for MBO; there is a continuous decrease of the altitude difference for PYR and ASK up to radius larger than 30 km; and there are sites for which the altitude difference stays constant for a portion of the domain radius (see for example CHC and BEO) indicating the presence of flat terrain.."

- Page 9: line 26: "PYR (5079 m) is the second highest station considered here, but..", reading this section and statement this line was confusing since the term "here" suggests that this is referring to the second highest station of this case study. You mean here that PYR is the second highest stations of all stations considered in the study and would then also state it this way for clarity.
  The sentence was modified as proposed by the co-editor:" PYR (5079 m) is the second highest station of all stations considered in this study, but the station is located at the foot of Mount Everest (8848 m) at a confluence point of several valleys (Fig. 5a and b)."

- Page 10, line 2: "The ABL-TopoIndex depends on the size of the chosen domain (Fig. 6a) so that the various algorithms were tested to several domain sizes ranging from 50 to 1000 km2. The gradient G8 and the local slope LocSlope are calculated on small fixed horizontal scales (0.5-1 and 10 km, respectively)". I had to read these two sentences a couple of times also not being convinced that this is the most optimal way to express what you intent to say. There are actually more of these sentences in the ms and would anyhow propose to still have once a native speaking English co-author (I guess there is one given the large list of co-authors) to critically check the ms for such potential flaws. Alternative: "Since the ABL-TopoIndex depends on the size of the chosen domain (Fig. 6a) we have conducted an evaluation of the sensitivity of the various algorithms to the domain size using a range from 50 to 1000 km2. The gradient criterion G8 and the local slope criterion LocSlope are calculated on small fixed horizontal scales (0.5-1 and 10 km, respectively)"
  The sentence was replaced by the one proposed by the co-editor. The whole manuscript has been checked by a native English speaker and the last version of the manuscript will be once again carefully gone over.

- Page 10: line 9: "the concentration of thermally lifted pollutants"
  The sentence was corrected as proposed by the co-editor.

- Page 10, line 9/10: "The hypso% decreases continuously for stations situated in a dominant position in their mountainous massif such as JFJ, SBO or BEO (Fig. 6c)". This is another example of a sentence that should be read carefully and revised. "continuously" here expresses something like over time whereas you want to express here that this parameter increases with an increase in the domain size.
  The sentence was modified: "The hypso% decreases continuously with domain size for stations situated in a dominant position in their mountainous massif such as JFJ, SBO or BEO (Fig. 6c)"

- Line 15: “with domain size”, change to “with an increase in domain size”
  The sentence was corrected as proposed by the co-editor. The sentence on Line 16 was similarly modified.

- Page 10; line 23-24 “To compare these two parameters, we show in Fig. 7 the ABL-TopoIndex as a function of the altitude for all grid cells in a 5km x 5km domain around a selection of stations”
  *The sentence was modified: To compare these two parameters, we show in Fig. 7 the ABL-TopoIndex as a function of the altitude for all grid cells in a 5km x 5km domain *for a subset* of stations.*

- Page 10; line 26: “..very steep and ASK a very flatt ABL-TopoIndex decrease with altitude”; first of all “flat”, then secondly, what is a flat decrease?? I guess you would like to say that there is a strong increase and a small decrease or?
  The sentence was modified as proposed by the co-editor:” Fig. 7 shows that the OMP and PYR regions have a very large ABL-TopoIndex decrease with altitude while ASK exhibits a very small ABL-TopoIndex decrease with altitude.”

- Line 33: “were constructed”, alternative “are located”
  The sentence was corrected as proposed by the co-editor.

- Page 11: line 6: “are grouped on Fig. 8”: are grouped as shown in Fig. 8 (change all this consistently in the text, e.g. “on Fig. 9”
  This was corrected in the whole text.

- Page 12, line 10: “their proximity *to* other massifs such as the Alps”
  The sentence was corrected as proposed by the co-editor.

- Page 14, line 14-15: “…ABL influence, in case of lifting processes without precipitation, is found for the ABL-TopoIndex….”
  The sentence was changed: “…ABL influence in the case of lifting processes without precipitation…”

- Page 14; line 18-19: “(mean the altitude over the 9 grid cells, similarly to the ABL-TopoIndex calculation)”
  The sentence was rearranged and changed to“The Spearman’s rank correlation coefficients of the 5th, 50th and 95th percentiles of the measured aerosol parameters with site altitude latitude, ABL-TopoIndex as well as all the individual parameters constituting the ABL-TopoIndex are presented in Fig. 9. (Similar to the ABL-TopoIndex calculation, the mean of the altitudes of the grid cell containing the station and its eight neighboring cells was used.)” ”

- Page 15, line 12, “lowest and the greatest monthly amplitudes”, should be according to me “smallest and largest monthly amplitudes”, also check further the ms for this: e.g. “the greatest ABL influence” should be “the largest ABL influence” (possibly a matter of taste). You use many times the term greater it should be larger/largest or higher/highest, e.g. “higher correlations”
  The whole paper was checked and the antonyms low/high, big/small, least/greatest are now used. Moreover the “greatest ABL influence” was always replaced by “the largest ABL influence” and “greatest correlation” with “highest correlation”.

- Page 15, line 21: “diurnal cycle minimal and maximal strengths of the absorption coefficient”. This expression reads also not well. You figure caption text seems to express it in the proper way, modify this text.
  The sentence was modified:” The ABL-TopoIndex is s.s. correlated with the minimum and the maximum of the monthly diurnal cycles of the absorption coefficient.”

- Page 16, line 6: “First the possible species and phenomena enabling the estimation of the ABL influence”, what do you mean here with species?? Do you refer here to compounds. I also think you generally want to refer here more generally to “parameters”
  The word “species” was changed to “parameters.

- Page 16, line 27, “NOy” y lowercase
  Done

- Page 16, line 28: “should be in most cases”; “could be in most cases”
  The sentence was corrected as proposed by the co-editor.

- Page 16, line 29 “hence involving aerosol washout”
  The sentence was modified:” However, one has to remain conscious that many lifting processes co-occur with precipitation and, hence, potential aerosol washout.”.

- Page 17: line 4-5: Your statement about using a model to further assess how pollutants can be used as a proxy for BL influence reads weird: what do you mean with a thermodynamic model? “…bounded to a 3D thermodynamic model adapted to complex topographies would be required before using absolute pollutant concentrations as indicators of ABL influence at high altitude sites”, rather “..constraining simulations with meteorological models able to explicitly resolve the role of fine resolution orography would be ….” (see also my previous comment on the title/introduction and first major comment of the reviewer).
  The sentence proposed by the co-editor was inserted in the ms.

- In addition, this text is part of your modification of the ms responding another major comment provided by one of the reviewers. There is another part of this modification that raises quite some questions: “ A further use of DBinv to restrict the area of potential pollution sources could also be envisaged since this parameter describes the domain from which pollutants can reach the high altitude station by convection and without crossing topographical barriers. This delicate issue can however be avoided by instead considering dynamical parameters such as the various temporal cycles”. What is delicate here? What are the dynamical parameters? You mention here the temporal cycles (diurnal and seasonal?) but in what parameters. I wonder what the reviewer will express about this modification. I am myself not very convinced and consequently suggest you to check this once more again carefully
  The expression “delicate issue” was replaced by the following sentence: ”The identification of pollution source areas potentially affecting the high altitude stations can be avoided by instead considering dynamic parameters such as the temporal cycles of various pollutants”.

- Page 17: line 14-15: “ Usually the spring leads to higher aerosol loading than the autumn probably related to higher ABL height in the spring”; do you refer here to a higher aerosol loading/concentrations at the high-altitude stations?? I don’t see how a deeper ABL would result in a higher aerosol loading in the ABL, actually the opposite would be expected not having any changes in the sources (more dilution)

The ambiguity was removed by changing the sentence:" Usually, at high altitude stations, the spring leads to higher aerosol loading than the autumn; this is probably related to higher ABL height in the spring"

- Page 18: line 7: "has similar dependency as the ABL as a function of latitude", dependency on what? I know that in the following sentences you give examples but rephrase this sentence.
  The sentence was modified:" Further, the RL has a similar dependency as the ABL on latitude, i.e., the RL's maximum height also depends on the duration of the incoming radiation."

- Page 18, line 16-17: "modify the theoretical cycles and lead to a broadening of the time of the extrema. These difficulties make obtaining clear statistical cycles another reason contributing to the observed low correlations", this text is another example of statements that need definitely to be rephrased. What are theoretical cycles: cycles that you would anticipate based on basic theory? What is meant with a broadening of the time? It would be broadening the time frame or increasing the duration, and which extrema?? And the second sentence needs complete revision.
  Both sentences were modified:" However, the statistical determination of the diurnal and seasonal cycle amplitudes suffer from several difficulties: 1) the low aerosol concentrations observed at high altitude often result in measurements near the detection limit leading to large uncertainties, 2) the high hourly autocorrelation of the data requires a pre-whitening procedure (see supplement) in order to be able to detect the diurnal and seasonal cycle, 3) meteorological conditions (e.g., cloud coverage, precipitation, seasonal fluctuations, etc.) modify the clear-sky diurnal cycles. These factors constraining the observation of clear statistical temporal cycles in the measurement data also contribute to the observed low correlations between the diurnal and seasonal cycles of the aerosol parameters and the ABL-TopoIndex."

- Page 18, line 26- 27: Correlation between topography and aerosol parameters and "in Sect. 3.5"
  Done

- Page 19, line 22-23: "Globally, NPF is the reason why the greatest correlations are found with the 50 percentile of the number concentration, instead of with the 5 percentile found for the absorption and scattering coefficients", also this sentence needs to be rewritten: correlations with?
  The sentence was deleted.

- Page 20, line 7,when you mention these terms "the Efremov-Krcho classification, the hypsometric curve" you should shortly explain them but also indicate why you considered to include these terms in the analysis. On the other hand, the discussion is already now (way too) long.
  The authors agree that description in the manuscript is very short. On the request of one referee, a complete description of these parameters were added in the supplement (see Table 2). In order to keep the discussion as short as possible, only the basic domains from which these eliminated parameters were taken are now added:" Several other parameters taken from the topography, morphology or hydrology fields such as the topographical wetness index, the upstream catchment area, the Efremov-Krcho landform classification, the integral and index of the hypsometric curve, and the topographic prominence were tested but eliminated as being not relevant for various reasons (Table S2)."

- Page 20, line 28: "It is …"
  Done

- References to “de Wekker” should be listed in the references under the “D” and not the “W” references.
  Done

- Figure 1: the station names with the different colors come out sometimes quite poorly, like the stations in the US. Use only or white or black characters?
  The authors propose to use the same color code for figures 1 and 8 and to add a grey scale topography as a background.