Review on Aerosol composition, air quality, and boundary layer dynamics in the urban background of Stuttgart in winter

The paper investigates the evolution of the planetary boundary layer (PBL) and associated aerosol concentrations for a field campaign and two case studies in winter 2018 in Stuttgart, Germany. The analysis uses a set of different in situ and remote sensing instruments to observe different characteristics of the PBL simultaneously. It further uses the PALM-4U model on a high spatial resolution (10m) to demonstrate the ability of such large eddy simulations to qualitatively reproduce the measured results. The study is in general well written and provide a meaningful contribution to the knowledge of inner-city PBL evolution and air quality.
Two aspects are my main concern. Firstly, it is not clear from the introduction to what extend the current publications differs from other investigations. E.g., the authors refer multiple times to Huang et al. (2019) but do not show the uniqueness of their own work. Secondly, the use of the PALM-4U model in this investigation is rather weak. There is a lot potential to use the model and validated conclusions made during the results section. It is rather added to this publication to show the overall agreement of the model with observations. I am certain that there are multiple publications that evaluate the PALM-4U model. Thus, I do not understand to what extent this paper differs from these previous investigations.
I recommend the author to carefully revise the manuscript and consider splitting it into two publications or extending the model evaluation to be more connected to the analysis made in the results section. Further details are given below.

General comments:

- The title suggests that the aerosol composition is investigated in the paper. However, the abstract does not account for this investigation. Please add this investigation to the abstract. Further, the abstract is short of quantitative results which can be of interest for other researchers.
- please provide a definition of “urban background in downtown Stuttgart”; As you mentioned in the text, the area of investigation in in-between a highly used road and the tracks heading to the main station. This does not sound like a “background” location.
- In many instances the paper refers to Huang et al. (2019). In the introduction there should be a paragraph that explains the differences to this study and how the current study complements the one by Huang et al.
- In the results section, the use of Figures is confusing and not consistent. I recommend to revise the paper and consider re-aranging the argumentation such that jumping back and forth between Figures will be avoided.
- The PALM-4U model plays a minor role in the analysis and does not contribute to the interpretation of the results. In fact, the observational data are used to validate, in a qualitative way, the PALM-4U simulation. Given the explanation in lines 461ff. is not sufficient for me. For a real investigation of PBL dynamics, a more thorough analysis would be required. I recommend to consider splitting the two parts (evaluation of PBL dynamics and air quality; verification of PALM-4U) to two publications or to use the model simulations to proof some of the results, e.g., the stability of the PBL in the case studies or regional transport effects. Further, I do not understand why the boundary conditions for the PALM-4U simulations are kept fixed. Please consider re-running the model with the improved boundary value handling to show also the effect of regional transport, vertical mixing of the aerosols on the different days etc.

Specific comments:
- line 1: Remove “the” in front of “cities”; further, add a comma “,” before “which”
- line 10: “temporal-spatial” → “spatio-temporal”
- line 39: add “the” in front of “boundary layer”
- line 49: what do you mean by “Most lidars overlap”? Please clarify.
- line 54: add “the” in front of “nocturnal boundary layer”
- line 61: It is not possible to “compare” the PBL. Only characteristics of the PBL (e.g., PBL height) can be compared. Please clarify.
- line 55 – 64: I feel that this is not directly related to the paper as it focuses on a joint investigation between observations and LES. As the introduction is already long, I recommend to remove this part.
- line 65: I’d refer LES to be a method rather than a model. Please change.
- line 71: “qualified” does not seem to be the right word. I recommend to use “attribute” instead
- line 78: “strong” → “strongly”; further, explain the chemical “eBC” (also for other chemicals on first appearance)
- line 95: Correct the citation “Samad and Vogt” (year is missing)
- line 96: remove “of a city” or “in Stuttgart”
- Figure S1: Please exchange “left” and “right” in the caption
- line 105: Both, downtown and urban background, is used to describe the area of the field campaign. Please be more concise on the description. From my experience I’d not refer to Stuttgart Neckertor as urban background.
- Figure 1: Please use only one shading /contour color for the plot. Overlaying two colors makes it hard to distinguish the two displayed fields. I recommend to remove the temperature from the plot.
- line 123: you state that the valley floor is at approx 300 m but the Rosensteinpark is at 247 m. Please be more concise and correct the valley floor height.
- line 132: “a LES utilizing PLAM-4U” → I don’t understand this phrase. Please clarify and correct the spelling of the model PALM-4U.
- line 150: description of variable “b” is missing. Please add. Also, w_f is described as W_f in the text. Please correct.
- line 188: please change “retrieved” to “retrieval”
- line 207: Please indicate the difference between PALM and PALM-4U. Please also consider restructuring the subsections. I recommend to rename subsection “Large eddy simulation” to “Modelling” or similar and but sections 2.4 an 2.5 as 2.3.1 and 2.3.2.
- line 216-218: This description of the initial profile that is taken as boundary profile is not clear to me. Please improve the description. Are you referring to the emission profile or the aerosol vertical profile?
- Figure 2: Change “Surfate” to “Sulfate” (Fig. 2b); The legend is not readable in Fig. 2e. In the caption, please correct the height of wind measurements (10m in the text, 2m in the caption).
- line 239: please provide the reason for the underestimation in the text.
- line 214/242: Your claim that the aerosol module of the model accounts for emissions, transport, deposition. From this, I derive that it does not account for chemical transformation. However, if a large fraction of aerosols is from ammonium nitrate (secondary aerosol), a discussion on the ability of the model to simulate the correct aerosol distribution is needed.
-line 258: This sentence needs more explanation. E.g., how can you derive stagnant dynamics from the shown profiles?
- line 262: If I interpret the scatter points correctly, two different regimes of PM10 concentrations are visible for PBL heights above 900m. Is this related to wind speed or transport effect from outside of the valley? Please give more details here
- Section 3.1: You use the term correlation throughout the section. However, this is rather empirical described by the plots. Please provide correlation values for all subplots of Fig. S5 to illustrate the strength of the correlation.

- line 305: please provide the dates for the two case studies.
- line 316: I find the reference to 1800 m confusing. What is the threshold of backscatter coefficients that lead to this conclusion? I'd rather claim that, in general, the aerosols are well mixed within the PBL (lidar) throughout the period
- line 317: at some point, the mentioned period should be explicitly dated.
- line223: "attitude" → "altitude"
- Figure S7: Please change the y-axis descriptions. In general, normalized values are unitless. Further, I do not understand the data you show. Shouldn't the different species concentrations add up to 100 % if you show normalized values? I assume you show staggered plots?
- line 332: please be more concise. Do you refer to Nitrate as "non-refractory particle mass" or where can I find this quantity in the plot?
- line 334-335: I would call a decrease of more than 50 % "slight". Please change.
- line 337-340: I am not sure if an increase of PBL height of 100 m can cause a reduction of eBC concentrations of more than 50 % but have no effect on other aerosols. I recommend to add some more details to this point.
- line 344: you mean Figure 4e? What do you mean by "and Figure 4 insert"?
- Caption Figure 5: "wavelength" -"wave"
- line 355: Add "UTC" after the time. Also for other places where the time is mentioned.
- line 379: "PLAM" → "PALM"
- line 380: Please rephrase and combine the two sentences to one.
- line 382-384: Do not go back to Figure 1 description. This is one example of confusing and non-consistent presentation of the results. I recommend to remove this statement as it is invaluable for the interpretation of the results.
- Figure 8: Why do you scale PM2.5 by a factor of 4 and not 4.5, which you have identified as bias between the model and observations?
- line 397: Again, you jump from Figure 8 to Figure 9 to Figure 8c within 3 sentences, without explicitly introducing Figure 9. Please restructure.
- Figure 9: What does the black line show? Is it arbitrary or does it reflect standard correlation values? Please provide explicit correlation values.
- line 404: Please remove as it is a repetition of results from above.
- line 414-415: please put this to the description of the PALM-4U model (around line 216, where I made another comment).
- line 416: Is there a proof for this statement, e.g., from the coarse resolution model runs?
- line 420: We saw in the Figures that nitrate is a significant contributor to local PM2.5 during this period. As nitrate is a secondary aerosol, how does this compares to your assumption, that secondary aerosols play a minor role?
- line 422: "cross" → "cross section"; "PLAM" → "PALM"
- line 453: Is it proofed that the local emissions are similar on the different days such that changes in emissions can be excluded as a possible source for the different aerosol concentrations? Please clarify.
- line 459: It is reasonable here to use the model to improve the understanding of the physical processes leading to the PBL evolution and to proof the findings from the analysis. See also my general comment on this aspect.