

Answer to Reviewer 1:

The authors wrote a case-study based paper about how the dynamics and chemical composition of the boundary-layer changes after frontal passages. They evaluate a warm front and cold front using observations (e.g. surface-based and drone) and numerical weather prediction models. The paper itself was clearly written, and the topic is relevant as I am not aware of many studies that look to the ABL during periods of precipitation. I appreciate how the authors incorporated both observation and land model data. That being said, I have a number of suggestions and comments that I think should be addressed before publication. Most importantly, I think the evidence that supports important conclusions from this manuscript are not clearly evident to me. It is likely a combination of the figures selected and the order of the text.

Dear Reviewer 1, thank you very much for the review and feedback on our manuscript. To clarify the argumentation and structure of the manuscript, the overview figures Fig. 4 and 6 and the related text are now introduced in the beginning of the respective case study section, which enhances the understanding of the context and the more detailed results in this study.

Major Comments:

1. There is too much material in the supplemental material. In my opinion, some of figures that are in the supplemental material are essential for supporting the conclusions that are drawn from the paper. The manuscript relies heavily on them. Those figures either should be included in the paper, or the results section should be rewritten so that the supplemental figures are not relied on as heavily as figures in the paper.

Thank you for noting this. We have decided to include Fig. S6 in the main text to support argumentation of Sect. 3.1, same for Fig. S9 and S10 and Sect. 4.1. As argumentation in Sect. 4.5 relies heavily on Fig. S14, it was included in the main manuscript as well.

2. ABL Terminology and Methodology.
 - a. Lines 130-135: how do you calculate ABL height from the observations and the model? Are they the same method? Explain this more, and discuss (somewhere) the effect of the definition of ABL height on your results. For example, ABL height very much depends on the method (e.g., Seidel et al, 2010).
 - i. There are some citations (or justification) missing here. Where do these thresholds come from? Why would Brunt-Vaisala frequency be used for mixing layer height?
 - b. How do you compute it from observations?
 - i. From reading the manuscript, I get the impression that the terms NBL, MHL and PBL are used somewhat interchangeably. They are all different concepts, and in the case of MHL and NBL, they cannot co-exist. A mixing layer implies a convective ABL and a nocturnal ABL typically wouldn't have a mixed layer to my knowledge. Please revise the terminology accordingly.

Thank you for pointing possible misunderstanding out. We have revised the usage of the different descriptions of ABLs. The abbreviation ABL and MHL were not used, but MLH (mixing layer height), which is a metric describing the upper boundary of an atmospheric layer (which is assumed to be mixed within itself). The MLH is not supposed to describe a mixing state of the respective layer like CBL (convective boundary layer) and is made clear in the beginning of Sect. 4.4:

“Until this point of the study the MLH is used as a synonym for the height of the planetary boundary layer (PBLH), which is considered the most relevant measure separating the free troposphere and the ground-influenced layer with different dynamical characteristics and potential impacts on the chemical composition.”

The calculation of the MLH from the model has been adapted from (Wang and Wang, 2014). In addition to the direct calculation using this approach, also the profiles for relative and absolute humidity and θ , as well as the Brunt-Vaisälä frequency (as a measure of the change of the stability of the vertical profile at the upper edge of the PBL (e.g., Hyun et al., 2005)) are analyzed and checked for consistency. The critical threshold values have been determined from a statistical analysis of all simulated profiles at the measurement site by a subjective measure which also fit well with observations from the ceilometer data.

The last statement was included in the end of Sect. 2.3 after explaining MLH criteria:

“The critical threshold values have been determined from a statistical analysis of all simulated profiles at the measurement site by a subjective measure which also fit well with observations from the ceilometer data.”

In the original version of the manuscript, “NBL” was used widely for the lowermost layer identified in the morning, which is a remnant of the NBL (although its already daytime, see answer to your comment 3b). The term “NBL” was changed to “NBL remnant” to avoid confusion and highlight, that this is an atmospheric layer residual from nighttime.

3. In the results, the evidence for some of the claims/conclusions is not clear. In particular, with regards to the presence of a cold pool in Case II and the presence of an NBL.
 - a. Where do you see evidence of a cold pool? If it is from ICON, I suggest including a figure that shows it. Right now, it reads as a cold pool might be there because of surface evaporation (which wasn't measured I think?) would increase after a rain event. If all the evidence is from the ceilometer (Fig. S10) that should be moved into the main body.

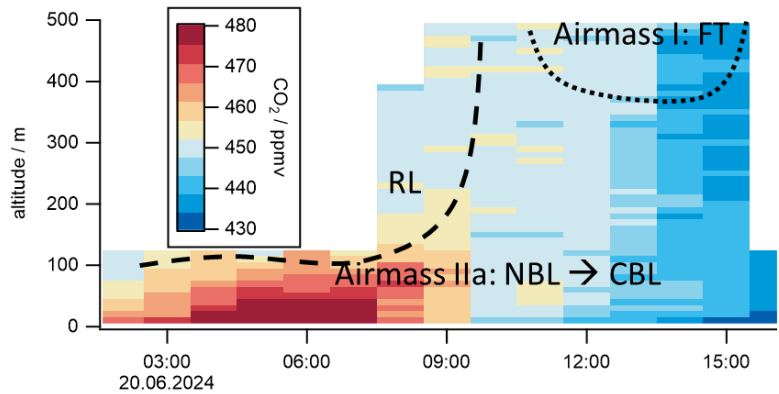
The evidence for a cold pool originates from the sum of indicators from vertical profiles measured with the UAV and ground-based observations:

- i. In Fig. 7c, for $d\theta/dz$, the lowermost bins at 50 m to 150 m a.g.l. between 14:00 and 16:00 show a strongly stabilized shallow layer, while conditions aloft are more unstable.
- ii. In Fig. 7d, the gradient of the absolute humidity is strongly negative for the lowest two height bins, and decreases between 16:00 and 19:00. This indicates evaporation.
- iii. Regarding the probing height in 7a and e, the O_3 mixing ratio stays low at the 50 m bin indicating a clear separation from the air masses included in the 150 m bin (100 m to 200 m a.g.l.), while the Richardson number highlights a clear separation of different mixing layers between the 50 m and 150 m mark. The gradient dissipates for well mixed conditions from 17:30 on.
- iv. “The increase of equivalent potential temperature after the last rain at ground level by 6 K and by 2.1 K for the mean vertical profile between 0 and 100 m, indicates a cold pool (Tompkins, 2001)”.

The argumentation in Sect. 4.2 was rephrased to include the four indicators:
“In our case, following the rain events at 13:30 and 14:45, we observe a cold pool from ground and FLab measurements, identified based on four indicators: (i) stratification between the lowermost 100 m (cold pool) and aloft is visible for the (thermo-)dynamical markers $d\theta/dz$ and Ri (Fig. 5a, c) as well as for the O_3 mixing ratio (ii), which is ~ 10 pptv (20%) lower in the lowermost layer (Fig. 5e, presumably due to depletion in the wet cold pool); (iii) the increased negative gradient and variance of the absolute humidity (Fig. 5d) and (iv) an increase of the equivalent potential temperature by ~ 6 K at ground and by 2.1 K for the whole 500 m range (Fig. 5b) show the evaporation and conditional enhanced latent heat flux typical for cold pools (Tompkins, 2001; Engerer et al., 2008).”

- b. From your results, it is not clear to me why you’re considering the ABL before the rain an NBL. For example, in Case I, you have positive sensible heat flux, relatively strong TKE (Fig. 1) and negative Ri and negative slope in $d\theta/dz$ (Fig. 2). Same with Case II, it looks like a typical morning ABL growth (e.g., 50 m is unstable before the rain).

Thank you for pointing out that the term “NBL” for airmass IIa in Case I is used wrongly. The airmass IIa in Case I should best be described as a suppressed convectively boundary layer (CBL) originating from NBL dissipation. At this time of the day, the NBL has already dissipated into a CBL as the distribution of CO_2 (an inert marker for the NBL) indicates in the following figure, where the dotted lines highlight different MLHs:



For Case I, consequentially, we have changed the description of a “NBL” to “(suppressed) CBL” to highlight the internal convective mixing of the layer IIa. The strong TKE at ground, a negative Ri and a negative slope in $d\theta_{eq}/dz$ in the vertical profiles indeed indicate turbulent motion in the CBL. However, this turbulent motion is apparently not strong enough to allow mixing lowermost layer IIa with the free-tropospheric airmass I aloft as demonstrated by the clear chemical separation (free tropospheric air (O_3 -rich & particle-poor) that is layered aloft NBL-originating-air (O_3 -poor & particle-rich)).

For Case II we disagree and would stick to the term “NBL”. It was assigned to the respective airmass IIa, which dissipates in the early morning from 08:00 on and does not exceed the 250 m mark until 10:00.

- c. I like Figs. 4 and 6 for making the concept easy to understand, but I don’t think the data that underlies them is clear in the manuscript.

We now introduce Figs. 4 and 6 earlier at the beginning of the respective section, when the individual case is presented the first time. The additional information and interpretation of the NBL/CBL/FT- separation and the cold pool identification were added in the following subsections. Conclusions are hopefully more reasonable with this new structure and information.

- 4. It is quite a board paper. Topic-wise, you cover ABL dynamics and chemical composition and you use many different methods. In the abstract, introduction, and summary, clarify the intent and significance of the paper. For example, what can we apply to other sites? Is this a validation of ICON?

The observations are brought into context with the larger scale by using the ICON data. The consistency between the observations and the simulation results depicts the validity of the observations not to be only local phenomena of the measurement site and allows for process-oriented analysis, e.g., how the BLH is changing in the event of the warm front, whereas in situations with strong rainfall local small-scale cold pool development can deteriorate the PBL structure. Consequently, intense cold fronts or intense precipitation from convergence lines lead to a stronger modification of the BL characteristics including the height estimates. The physicochemical processes inside the boundary layer are assumed to be representative for similar topography (rural hilly regions and plain fields).

Now, we imply the intent and significance of the paper in the second last paragraph of the introduction and in the beginning of the abstract, explain the significance at the end of the abstract and in the second last paragraph of the summary.

Technical points:

1. Line 45 – It is a bit confusing why MOST is brought up here in the context of observational limitations.

The introduction of MOST was removed from the introduction to reduce the load of material, which is not crucial to understand the manuscript.

2. It would help if you included the equations for equivalent potential temperature and vertical potential temperature as they are derived variables.

Both equations are now included in Sect. 2.2.

3. Line 87 – I am not familiar with “aspiration” in this context, and I don’t understand the meaning here.

The term “aspiration” was changed to “air mass inflow” for better clarity.

4. The order of the supplemental material is out of order compared to how it is referenced in the paper.

The order and references of supplemental material was corrected.

5. The order of referencing figures in the paper is also out of order (e.g. Fig. 4 is referenced in text before Fig. 3).

We revised the order of referenced figures completely as Fig. 4 and Fig 6 were shifted closer to the beginning. Now, the order and figure references are all correct.

6. Some citations are the wrong type (e.g. lines 118, 119, 120, 124 and other places). If using latex, change `\citep` to `\cite` for the proper citation type.

Using Microsoft Word for writing the draft, we do not see an inconsistency how the citations were included. We are confident that any changes in type will be removed during the typesetting process.

7. Line 132 – you haven’t defined the MLH yet.

The definition of a MLH was added in the beginning of Sect. 2.3 where it appears the first time.

8. Fig. 1. Check the units of Q_H are identified correctly. Is it truly $< 2 \text{ W m}^{-2}$ all day? And what is the meaning of $Q_H * 10$ in the legend.

The units of Q_H are correct. The high values of up to “ 1.5 W m^{-2} ” are due to the artificial factor of 10 on Q_H to display TKE and Q_H on the same axis. This is now clarified in the figure caption.

9. Line 214 – rephrase the “like, e.g.” combination.

“,e.g.” was removed to reduce the duplicity of this adverb.

10. Line 271 – I’m not familiar with “lability” in this context.

We think that the term “lability” can be used as a synonym for low tropospheric stability, e.g., $d\theta/dz < 7.5 \text{ K km}^{-1}$, as used in typical synoptical analysis, i.e., that the air mass is unstable against moist adiabatic vertical movement.

11. Line 426 – don’t use “significant” if you didn’t do a significance test.

The term “significant” was removed.

Answer to Reviewer 2:

This study aims to understand chemical and meteorological processes occurring in the boundary layer during summer rain events, by using a combination of drone-based measurements, ground-based observations, and ICON forecast model data. The aim of the work is novel and interesting, yet the structure and the methodology suffer from a number of drawbacks which limit the impact and the full potential of exploitation for the work. Below I report my major comments based on my revision of the paper.

Dear Reviewer 2, thank you for your comprehensive feedback and critics. We revised the manuscript and have changed several sections to lower concerns and improve the understanding of the concept and the results.

General comments

The structure of the Introduction section is chaotic and messy. It starts from understanding atmospheric processes during different weather events, then begins to talk about fronts, then of models and measurements, then again of atmospheric dynamics, Monin-Obukhov theory (MOST), drone-based observations, fronts, ... Therefore, I suggest improving the logic of the Introduction removing multiple jumps from one topic to the other, and more clearly demonstrating the gaps present in the literature and the key research questions addressed in the current work.

Thank you for letting us know that the introduction appears to be not as clear as desired. An improved version of the introduction does not contain MOST and first focuses on boundary layer dynamics and the difficulties to characterize them before moving towards measurement approaches.

Additionally, we added the major gaps in the literature to motivate the research goal of investigating the influence of rain events on the chemical composition of the PBL:

“While model data lack the high spatial resolution in the planetary boundary layer (PBL, Lochbihler et al., 2021), in-situ measurements are usually limited to specific meteorological variables, while chemical markers and frontal influence on the air mass’ chemical composition is neglected. Large data sets of local data cover underlying dynamics of different rain fronts, however, they often do not cover a sufficient vertical range (Bopape et al., 2021; Helbig et al., 2021; Wang et al., 2021) or focus on different ecosystems such as tropical rainforest (Machado et al., 2024b; Machado et al., 2024a). The influence of entrainment at the PBL top on the chemical composition has been examined (Pohorsky et al., 2025; Platis et al., 2016), but, to our knowledge, detailed analyses that couple PBL dynamics with precipitating frontal passages under midlatitude continental-plain conditions are scarce.”

Although not directly described in the Abstract, the methodology seems to be based not only on the ICON model, but also on HySplit back-trajectories driven by 0.25° GFS analysis data for calculation of boundary layer height. The use of back-trajectories in such complex terrain and driven by such coarse resolution meteorology is substantially problematic, as it is extremely difficult for such a meteorological model to successfully catch the micrometeorological and mesoscale processes occurring in the study site. In addition, it is not clear why you need this estimation when you have ceilometer data and higher resolution models. Last but not least, it is not clear why you restrict the focus to 24 hours only trajectories.

In addition to the HySplit trajectories we also calculated trajectories from ICON-D2 high resolution meteorological fields. However, given the vicinity of the measurement site to the borders of the model domain of ICON-D2, those back trajectories often did not provide information beyond a few hours before arrival at the measurement site. Nevertheless, a high degree of consistency between the ICON-D2 and the HySplit trajectories have been found for the overlapping domain.

The following statement was added:

*“Besides the HySplit trajectories driven by 0.25°-GFS analysis data, ICON-D2 analysis and hourly forecast data for the gaps between the analysis time events (3 hourly) were utilised to calculate corresponding MLHs, backward trajectories within the German domain **and to validate the coarser HySplit trajectories** (Figs. S3 and S4).”*

I tend to agree with the other reviewer highlighting the relevant number of figures in the Supplementary, which also seem to be rather fundamental as per their discussion in the manuscript (in addition, with an odd and not consequential numbering). Figures in the Supplementary Material can support the findings, but should not contain relevant information that is fundamental to the discussion of the results.

Thank you for noting this, too. We have reconsidered the number and selection of figures in the manuscript and supplement to include the figures which are most crucial into the manuscript, whereas the supplementary material mainly contains figures that rather serve as a reference or do not contribute that much.

We merged the previous figures 1 and S6, S9 and S10, and included S14 in the main text as several sections in the manuscript refer to them.

Section 5 only contains a summary of the findings, while an inclusion of the conclusions, limitations and next steps or outlook could be rather beneficial for improving the impact of the work over the long-time horizon.

The major limitation was the low number of two analyzed cases, the absence of in-rain flights and the resolution of hourly profiles that could be improved in future studies. This is now mentioned in the manuscript in the second last paragraph of the summary:

*“**Despite being limited to two cases with hourly resolution and no in-rain flights**, the study shows that combined model- and ground-based analysis can conceptually identify complex boundary layer phenomena like pollutant sinks and local stratification.”*

We included more detailed suggestions for future studies and implications in the final paragraph of the study:

“Findings of this study are only based on two in-detail analyzed case studies, depicting different types of fronts. Further field studies with similar vertical resolution at different locations with varying surfaces like in flatland, alpine mountains or cities could help to better characterize frontal dynamics. Reanalysis of MLHs with ICON for comparable cold pool scenarios as in Sect. 4 could allow finding a parameterization to identify and predict cold pools solely on a model basis, which is important as the occurrence of cold pools is expected to increase with climate warming in the mid-latitudes (Borquez et al., 2020). This could also help to estimate the MLH more precisely under strongly unstable conditions.

Recorded BVOC data from field campaigns should be reanalyzed regarding O₃ depletion after rain above grassland, where vertical O₃ measurements were available.”

Specific comments

Lines 9-11: I doubt there is such a clear division between research in polluted area and at continental rural mid-latitude sites. In addition, this is for sure out of scope for the beginning of the abstract.

We wanted to emphasize the setting of previous studies regarding its geographical region and as it is expected to influence boundary layer dynamics differently. Lines 9-11 were rephrased to motivate the research gap differently in a short sentence.

“Understanding atmospheric processes is essential for improving weather forecasts and models, but in continental regions the effects of dynamical mixing and frontal events on the chemical composition of the planetary boundary layer remain poorly characterized.”

Lines 20-22: What does this mean more in general for boundary layer dynamics?

Local small scale cold pool formation is an uncertainty for the model simulations even though the general feature can be reasonably represented by the ICON-D2 model. However, the extent and intensity of these cold pools, especially after heavy rainfalls, and their effects provide opportunities for improved PBL parameterization schemes to simulate the MLH accurately.

In the last paragraph of the abstract the following general implication of the analysis of such unstable conditions was included:

“Analysis of these events help to reduce uncertainty of model simulations and enhance the PBL parameterization scheme.”

Lines 90-139: Given the high number of data sources, perhaps having a Table with all data sources and key information extracted, as well as resolution, ... could be helpful for the reader.

Thank you for pointing this out. The information for experimental and model data was included into Table A1 and A2 in the Appendix A.

Lines 150-152: this note is not in the appropriate location.

This note was shifted to the beginning of section 3.

Lines 158-160: Not clear what is the assumption if this is something you are observing in your results. Maybe it is a speculative result?

You are right, the message was not as clear as desired. We rewrote the sentence to clearly distinguish between observation, expectation and result:

“A strong upper free tropospheric layer (I) that suppresses a trajectory uplift is shown by the local scale observation of an immediate but short-lived dynamical stabilization above the lowermost layer and its hardly changed conditions below. Despite the missing updraft transport, the rain events can indeed be described as a warm front (see Sections 3.2 and 3.3.1).”

Lines 163-167: The caption contains material that is rather for the main text.

This figure was extended with additional graphs from the Supporting Information. The interpretation

of the data given in the figure caption of the previous version was now moved to the main text and the figure caption was shortened to avoid redundancy.

Lines 173-175: Rather, the high correlation indicates the presence of the same temporal pattern, which is quite obvious and expected, but does not demonstrate the existence of a cause-effect relationship.

You are right, that the high correlation between irradiance and the sensible heat flux is expected, however, to us the irradiance-influenced sensible heat flux is plausible. We have therefore opted for a more restrained wording:

*“The strong correlation between irradiance and sensible heat flux throughout the day (Pearson correlation coefficient $r = 0.87$) **suggests** that sensible heat flux is generated mainly by direct irradiance and **probably** not driven by stored energy from the ground (Fig 1).”*

Line 222: Change “Contrary” to “Conversely”

Done.

Lines 229-232 and lines 309-311: Suggest rephrasing this sentence to highlight the findings and the eventual agreement with previous studies. The current structure does not reflect what is original in the observations, while it highlights more the previous studies.

For 229-232: *Lambe et al. (2011)* was included to demonstrate that an increasing O/C-ratio in the aerosol composition can eventually originate from an O₃ increase. The finding that delayed BL break-up alters photochemical aging is given right afterwards as consequence in context with the study:

“In addition to the photochemical production of O₃, mixing with free tropospheric air also causes a rapid increase of O₃ at ground level (Neuman et al., 2012), potentially causing photochemical aging of the aerosol (Lambe et al., 2011), as suggested by the strong correlation ($r = 0.94$) that we found for the time series of O₃ levels and the O/C ratio of organic particulate aerosol measured with the ground-based MoLa (Fig. S8). This strong correlation of O₃ with aerosol aging and the observed stratification pattern show that a delayed breakup of the NBL can also lead to a delay in the onset of diurnal chemistry. An inflow of already-aged aerosols as a reason for enhanced O/C ratios is unlikely due to the stability of the PM₁ concentration measured at ground.”

For 309-311: Two further studies, which suggest cold pool dynamics (*Lochbihler et al., 2021; Haerter and Schlemmer, 2018*), were cited and the fact that vertical profiles prove their assumptions and compliment the (Tompkins, 2001) study is emphasized:

“Simulations of cold-pool dynamics indicate that, under continental conditions in the mid latitude, strong latent heat flux can increase convective mass flux (Lochbihler et al., 2021). Consistent with this, (Tompkins, 2001) reported increases in equivalent potential temperature along recovered cold pools from ground measurements. In our case (Figs. 5b, S14), following the rain events at 13:30 and 14:45, we observe increases in equivalent potential temperature of ~6 K within an hour at the surface for each event, and aloft by 2.1 K and >3 K, respectively, at the sampled altitude levels up to 500 m. These high-vertical-resolution observations between 13:30 and 14:45 support the model-based hypothesis by Lochbihler et al. (2021) and Haerter and Schlemmer (2018) that strong latent heat flux

and associated moist updrafts can promote the formation of new convective cells like the upcoming rain front for our case study.”

Lines 262-264: This sentence needs to be rephrased as it is not clear.

Thank you for this hint. The sentence was rephrased for clarification:

“The influence of thunderstorms on the pollutant distribution and air mass dynamics in the lower troposphere was investigated by vertical profiling. Throughout a day when a thunderstorm occurred at the measurement site eleven FLab-based measurements were conducted hourly, as long as weather conditions allowed for safe operation.”

Line 275: What do you mean by “aspiration of air masses”? (this term is also used at line 232 and 87, but it is not very technical).

The term “aspiration” was changed to “inflow” or “air mass inflow” for better clarity.

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