

Reviewer #1:

This study (egusphere-2026-2494) investigates how irradiance and mixing layer height (MLH) affect the vertical distribution of trace substances in rural areas of Germany, using data from two summer drone measurement campaigns conducted up to 500 m above ground, along with data from ground-based stations. The findings suggest that irradiance is the primary factor influencing diurnal mixing mechanisms. Furthermore, the research highlights that ground-based measurements do not always accurately represent the conditions throughout the entire mixed layer. However, several concerns need to be addressed.

Dear reviewer #1,

Thank you for your detailed feedback on our manuscript. We have to admit that we missed the publication of Tan et al. (2025), which provides valuable input to our analysis and, in addition, helps to understand our observations. We included the categorization of different types of PBLs introduced by Tan et al. (2025) in the introduction/motivation, analysis and discussion sections. Individual comments to your feedback are given after the respective bullet points.

1. In this study, the sample size for nighttime analysis is quite small, relying on data from only one night (June 20–21, 2024). Although the data is of high resolution, its statistical representativeness is limited, so caution should be taken when drawing conclusions. It is advisable to include additional nighttime profiles or to clearly define the study as a case study.

You are completely right that the statistical representativity of this study is limited. However, this section does not aim on providing the general fine-structure for a specific location, but aims on showing the sensitivity and necessity of measuring a variety of variables in highly resolved vertical profiles. To make this clearer, we now have inserted “case study” into the sections title (Sect. 3) to highlight the limited statistical significance due to the limited sample size:

“Case study: Height criteria and fine-scale structure of the nocturnal boundary layer”

We also specified this in the last section of the introduction by adding the information that the insights result from a single nighttime event:

“a [...] using gradient-based methods (up to 120 m altitude with 2 m vertical resolution) during a single nighttime event, featuring a fog event and a low-level jet”

2. In section 3, the authors conduct a multi-variable analysis of MLH using various meteorological variables, including potential temperature, temperature, humidity, wind speed, and the bulk Richardson number, as well as profiles of trace substances such as O₃, CO₂, and particle number and mass concentrations. While this comparative approach offers potential value, the manuscript currently treats the resulting height estimates primarily as alternative indicators of a single “MLH.” Conceptually, boundary-layer height can be classified into three distinct physical categories (e.g., Tan et al. (2025)):

Thermal PBL height defined by static stability, e.g., potential-temperature or virtual-potential-temperature gradients/parcel method.

Dynamic PBL height defined by dynamic stability, e.g., bulk Richardson number or wind-shear criteria

Material PBL height, defined by the vertical distribution of atmospheric constituents, e.g., aerosol or trace gas gradients.

The three heights mentioned are not interchangeable; they correspond to distinct physical processes and often differ, particularly when there is weak dynamical forcing or when residual-layer chemistry and surface sources or sinks are present. The current manuscript fails to adequately recognize or discuss this important distinction. The authors should explicitly define and consistently distinguish thermal, dynamic, and material boundary-layer heights throughout the text, figures, and captions. The authors should also clarify why differences among the three methods are inevitable, focusing on the physical mechanisms involved instead of suggesting that one of the measured MLH values is a more accurate or comprehensive representation of MLH.

We totally agree on the fundamental issue that the distribution of different atmospheric variables is caused by various different atmospheric processes, and hence result in the detection of different mixing layer heights in the atmosphere. Therefore, the differentiation between thermal, dynamical and material PBL is useful.

Firstly, we explain explicitly why we use the term “mixing layer” in the introduction instead the term “PBL”, because the mixing layer is not always identical to the PBL:

“The mixing layer height (MLH) describes the upper boundary of a well-mixed lowermost layer, where chemically or dynamically different air masses adjoin. Reliably distinguishing the mixing layer from the adjacent layers is essential for the interpretation of microphysical and dynamical processes. The most-studied stratification in the lowermost troposphere is the PBL and free troposphere, at which boundary chemical, and thermodynamical different layers interface. We use the term “mixing layer” explicitly to describe a distinct atmospheric layer, independent on its location and time, i.e., a mixing layer can be the PBL or any shallow layer near ground.”

Secondly, we want to make use of your feedback and refer to dynamical, thermal and material MLHs, which itself can easily be assigned by our set of measured variables. We introduced the classification of Tan et al. (2025) in our introduction in L49 and in the first paragraph in Section 3/L140:

“Tan et al. (2025) characterized different PBLHs simultaneously and distinguished between a thermal, dynamical and material PBLH and attributed them to different atmospheric processes, which explains why different PBLHs coexist.”

“Remote sensing of various variables, revealed the simultaneous existence of different types of MLHs based on the variables referring to statical or dynamical stability and the vertical distribution of atmospheric constituents that identified stratification in a complex terrain with mountainous, urban and waterbody influences. Respective variables led to the definition of a thermal, dynamical and material boundary layer, respectively (Tan et al., 2025). This differentiation between those categories helps to explain atmospheric processes.”

The core logic in section 3 is flawed. The author tries to pinpoint a so-called "ground truth" that would perfectly represent the average state of the mixing layer. However, as previously mentioned, the three types of PBLH have different implications. The thermal, dynamic, and material boundary

layer heights each reflect distinct physical properties, and the differences among them highlight the complexity of atmospheric processes rather than being attributed to measurement errors.

We understand your concern and fully agree that different boundary layer heights can be deduced from different atmospheric processes. In section 3, we want to highlight the advantage of using a multi-variable in-situ measurement approach to identify the fine-structure of the lowermost atmosphere during night. We made this clearer now by the following statement:

“Deriving the fine-structure of the NBL from different types of MLHs allows explaining atmospheric processes that cannot be identified otherwise.”

In the whole text of section 3, we now explicitly name the kind of boundary layer based on the variables they were observed by. Now, we also added a discussion about the coupling of thermal and dynamical boundary layer heights to the material boundary layer height during night:

“These analyses of the NBL fine-structure using hourly vertical profiles of multiple variables show that there is a strong difference between the power and potential of different variables to detect details in atmospheric stratification. We could show that in the early night, a dynamical MLH goes along with a material MLH, due to long-range transport. In contrast a developing fog event was identified using a thermal MLH even before it was observable using a material MLH, while a dynamical MLH was absent in a similar altitude range. A material MLH still indicated the slow recovering of the fog’s aftermaths. The absence of a dynamical and thermal MLH at this time in this altitude range highlights the low dynamics due to a lack of energy, which could be converted by latent heat that hinders convective mixing.”

and the daytime (see response in next comment).

3. In section 4, the discussion focuses on the representativeness of near-surface measurements for column-averaged concentrations of CO₂, O₃, and particle number density within the lowest 500 m. This topic is closely related to the material boundary layer, particularly concerning the actual vertical distribution and mixing of atmospheric constituents. However, the entire analysis framework in Section 4 relies exclusively on planetary boundary layer heights derived from the ICON-D2 model, which is defined only by thermal PBL height. Consequently, the manuscript equates the thermal mixed-layer height with the height that governs the vertical distribution of the very tracers being assessed for representativeness. This substitution lacks justification. The authors' own nocturnal results in Section 3 have already demonstrated that thermal and material layer heights often differ. This discrepancy also applies to the daytime conditions discussed in Section 4. Relying solely on thermal PBL height as a reference for a material boundary layer issue introduces a conceptual inconsistency. Such flawed reasoning undermines the physical interpretation of the reported gradients and the R metric. The authors should either (i) derive and use material layer heights based on their own in-situ tracer profiles for the daytime analysis, or (ii) provide a rigorous justification, supported by direct comparisons, for why the ICON-D2 thermal PBL height can be deemed a reliable proxy for the material mixed layer in this context.

Thank you for pointing out that the combination of the ICON-D2 obtained PBL height and the UAS measurements might result in inconsistencies and that a more detailed explanation and justification

of this combination is missing. From our point of view, this combination is reasonable due to following aspects that were integrated into the manuscript, if not already included:

- a) The ICON PBLH is estimated on “ [...] Brunt–Väisälä frequency $> 5 \times 10^{-5} \text{ s}^{-1}$, vertical gradient of potential virtual temperature $> 0.3 \text{ K km}^{-1}$, and vertical gradient of absolute humidity must be curved” (Section 2.3). While the Brunt-Väisälä frequency and the potential virtual temperature are variables defining the thermal PBLH, the criterion of using the gradient of the absolute humidity refers to the material PBLH as defined by Tan et al. (2025).
- b) Xin et al. (2026) shows that in summer time under convective conditions different types of PBLH (categorized in Tan et al., 2025) are “tightly coupled”. This implies that markers which indicate the dynamical/thermal PBLH can also be used for a rough estimation of the material PBLH. This is probably even more the case for our rural remote measurement site, compared to a measurement in a megacity as in Xin et al. (2026), where an inhomogeneous system of convective and pollution sources complicates the analysis of PBLHs even more. Furthermore, a differentiation between nighttime and daytime BL regimes is recommended in Xin et al. (2026), when a tight coupling is not necessarily given. This is considered in our study by using solely daytime periods in section 4. The modeled PBLH was not used in section 3.
- c) The analysis of representativity and gradients in section 4 is solely applied for periods when the PBLH exceeded the probed vertical range, i.e., 500 m. Assuming a scenario, in which the thermal, dynamical and material PBLH differ by 10% would flatten the gradients in Figs. 2 and 4, accordingly. However, the interpretation would not change, since the relative uncertainty is larger anyway. Surface chemistry or diffusional losses would be affected negligibly; hence R values are even more robust.

To address your concerns and a) and b) of our response, we added the following statement at the end of section 2.3:

“The obtained model-based PBLH qualifies for comparison with trace substance and meteorological variables, as a) dynamical, thermal and material PBLHs show a tight coupling in summer daytime under strongly convective conditions (Xin et al., 2026) and b) thermal and material criteria were used in the model for the estimation of a single combined PBLH.”

and this statement in the beginning of section 4:

“Recent studies differentiate between a thermal, material and dynamical PBLH that can originate from different atmospheric processes as already shown in section 3 (Tan et al., 2025). In summer time, especially during daytime, these PBLHs are tightly coupled, hence a further differentiation is not required here (Xin et al., 2026).”

and c) in the discussion of the gradients and the representativity R in section 4.3.3:

“As the PBLH varies depending on its definition, for example, whether it is identified as thermal, dynamical or material PBLH, a 10% variability between different PBLHs in summer would result in a 10% change in the gradients of the variables in Fig. 4. However, the interpretation would not substantially change under such conditions since the relative uncertainty is larger anyway. R values

are even more robust since surface chemistry and diffusional losses occur in the lowest layer and would be negligibly affected by a slightly different PBLH.”

Reviewer #2:

The authors presented a very interesting vertical profiling study of meteorological parameters and air pollutants in Germany using an uncrewed aerial system (UAS). Detailed analysis on mixing layer and boundary layer heights were performed using various criteria, and justification given. It also provided interpretations on how the dynamics of planetary boundary layer (PBL) affects vertical distributions of air pollutants, as well as how representative ground-based measurements are to cover the lower part of the PBL. Recommendations are given for accurate identification of the mixing layer and boundary layer heights in both nighttime and daytime scenarios, as well as to reconcile the ground-based and vertical measurements to better represent the relationships between emission and PBL dynamics that shape air pollutant distributions. The study can be published after some minor revisions, and my comments are given as below.

Thank you for your positive review. We incorporated your feedback and/or commented on them in blue.

- Although the authors justified that an extensive comparison is out of the scope of the current study (P13/L357), brief mentioning on recent studies of vertical profiling using UAS in some proper locations in Introduction and Discussion sections might help readers to appreciate the uniqueness of the current study.

Indeed, the integration of our work into previous research was not clear. We added a paragraph in the introduction:

“Recent studies used multi-platform approaches to improve UAS-based characterization of PBLs. UAS fleets have been applied to monitor atmospheric transport in a horizontal plane (Wetz et al., 2023), or vertical profiles to rule out temporal changes in the probed air masses (Borchers et al., 2025). Ground-based measurement stations have demonstrated to be not always useful as a reference (Barbieri et al., 2019), but also for spatial and temporal correction of UAS-based measurements for in-flight characterization of measurement instruments (Greene and Salesky, 2023; Moormann et al., 2025). For boundary layer studies, such multi-platform studies focused on, e.g., wind dynamics (Greene et al., 2022), altitude- and daytime-dependent aerosol composition within the PBL (Borchers et al., 2025), reconstruction of PBLs after the passage of rain fronts (Moormann et al., 2026), or the air pollutant distribution near civilization (Pohorsky et al., 2025). Furthermore, UAS can detect shallow layers near the ground by variables indicating material, dynamical and thermal mixing layer heights (Greene et al., 2022; Chilinski et al., 2024; Moormann et al., 2026).

Two fundamental assumptions for such studies are the representativity of UAS measurements within the PBL in general and the comparability of variables between UAS- and ground-based measurements. Greene et al. (2025) addressed the first assumption along with recommendations for sampling strategies, while this study focuses on the second one, using parallel UAS-based and ground-based in-situ measurements at two similar hilly rural sites in Central and Southern Germany during summer to evaluate [...]”

And in the summary/discussion section:

“Night-time vertical profiles of temperature, humidity, wind speed, and trace matter (CO₂, O₃, particle concentrations) in the lowermost 120 m were used to detect fine-scale nocturnal

stratification as an extended multi-variable approach, in contrast to studies relying on either only a few pollutants (Chilinski et al., 2024; Pohorsky et al., 2025), or solely dynamical tracers (Jozef et al., 2022). To detect shallow mixing layer heights (MLH) on an hourly time scale, we applied the criterion "strong gradient change" to each variable and minimized methodical errors due to temporal air masses changes throughout a flight by a ground-based measurement station as recommended by Greene and Salesky (2023)."

and

"Based on gradient-based similarity relationships (Greene et al., 2022; Sorbjan, 2010), we found that solar irradiance should be used as an additional parameter, in addition to PBLH, to describe vertical gradients, [...]"

- There are a few places that measured data can be provided or referred to, so as to make the points more clearly.

1) P6/L185: I thought latent heat flux was measured in the ground-based MoLa (P4/L103). Please show data to support the statement here in P6/L185.

Thank you for this comment. Highly time-resolved data of temperature and 3D wind have been measured with a flux tower 25 m distant to MoLa (Annex A1). A time series is now included, which shows an increase of the latent heat flux after the fog event in agreement with the statement in the text, while the turbulent kinetic energy remains an order of magnitude lower compared to the campaign's morning mean, reflecting reduced mixing of the lowermost atmosphere. We now have included a comment and reference to the new Fig. S5 in the manuscript:

"Apparently, the preceding fog event initiated the formation of a cold pool during night (Chachere and Pu, 2016), which converts turbulent energy into an upwards directed latent heat flux and may have formed a shallow inversion layer in (45 ± 5) m above the ground (Wilson and Fovell, 2016). Fig. S5 shows that despite irradiance levels during this morning are similar to the campaign's morning mean, turbulent kinetic energy is a magnitude lower than usual at this time of the day, while latent heat flux remains unaffected and still increases until the air is dried to 75% RH."

Figure S5 is presented here:

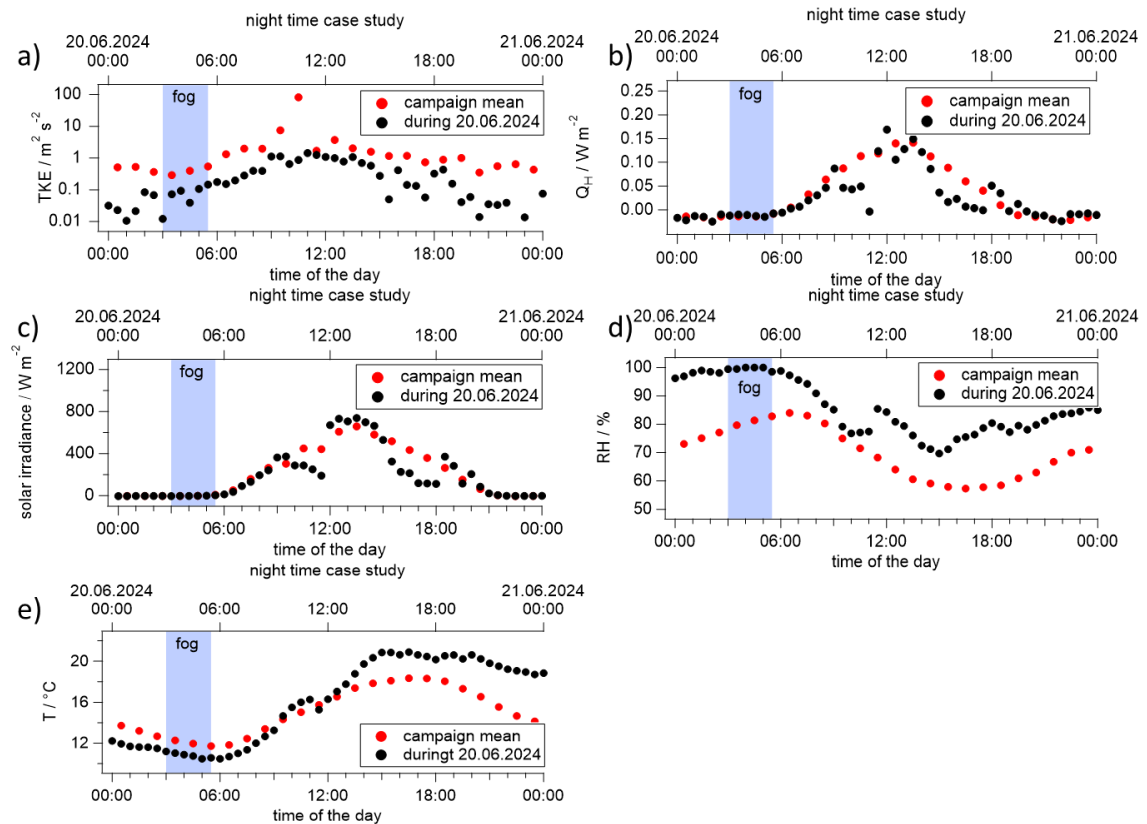


Figure S5: The comparison of the thermodynamical variables turbulent kinetic energy (a), latent heat flux (b), solar irradiance (c), relative humidity (d) and the temperature (e) between the night of the case study and the diurnal mean of the campaign shows similar irradiance and latent heat flux levels, while the turbulent kinetic energy is decreased in the case study night. Available energy is consumed to dry the air rather than to enhance convective mixing, which postpones the expansion of the lowermost atmospheric layer.

2) P9/L280: please show or refer to the data of correlation between PBLH and irradiance.

A reference to Fig. S7 can already be found at the end of the paragraph.

- There are a few places that the authors used O₃ titration by NO to interpret steep slopes of O₃ profiles in early morning and/or foggy conditions. This chemistry is for sure real and may be responsible. But can the author provide a brief comparison of time scales of passive diffusion, active convection, and O₃ + NO reaction to further support that it is the case?

You are correct that there is more than one way to lose O₃, however chemical loss is the most relevant process:

We included this text in L365:

“For a general understanding, a rough comparison of the efficiency of physical and chemical loss mechanisms might be useful to demonstrate the relevance of different loss pathways. The chemical loss of O₃ by reaction with NO can be estimated by using the rate constant $k_{\text{NO}+\text{O}_3} = 1.9 \times 10^{14} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ and a reasonable NO concentration $[\text{NO}] \approx 0.1 \text{ ppbv}$ during night and $[\text{NO}] \approx 0.5$

– 1.0 ppbv during daytime (Dewald et al., 2022; Iupac, 2025), resulting in lifetimes $\tau_{chem} = \frac{1}{k_{NO+O_3}[NO]} \approx 30 - 60 \text{ min}$ during daytime and up to 10 h in the night. Physical loss due to diffusion can be estimated by the related lifetime $\tau_{diff} = \frac{L^2}{K_z}$ with the vertical length L and the eddy diffusivity K_z . Assuming $L=25 \text{ m}$ for the vertical range that is relevant for chemical processes, and $K_z \leq 0.1 \text{ m}^2 \text{ s}^{-1}$ for moderately stable conditions as in the morning and during night time (diffusion dominated) and $K_z \approx 1 \text{ m}^2 \text{ s}^{-1}$ for convective conditions as during the day (turbulence dominated) above grassland (Stull, 1988). We can now estimate a diffusion-related lifetime $\tau_{diff} = 2 \text{ h}$ for stable and a convection-related lifetime $\tau_{conv} = 10 \text{ min}$ for convective conditions. The lifetimes τ_{chem} and τ_{diff} are in a similar range, while convective conditions reduce the lifetime. This shows that chemical and physical loss are both relevant depletion pathways and mainly depend on the convective conditions and prevalent NO levels.”

- For PNC, the authors assumed that they are mainly from direct anthropogenic emission and disperse by convection. Was there any new particle formation (NPF) and growth events captured in the campaigns (say in more sophisticated size distribution measurements in MoLa) that might affect this assumption?

Thank you for this comment. On average, the majority of the particles originates from anthropogenic emissions, e.g., air masses from the Frankfurt area (Fig. S2), however NPF took place frequently in the early afternoon as recorded by MoLa (Fig. S9). Therefore, during these times, NPF might be the dominating source over advected anthropogenic air masses. Gradients in the morning are likely due to regional emissions near the ground as the mixing in the morning of the lowermost layers is not sufficiently strong to homogenize the pollutant distribution in the lowest 500 m. We have changed the text in Section 4.2.2 accordingly:

“PNC_{>10 nm} reflects processes in the particle phase and is strongly influenced by anthropogenic emission sources at ground-level, noting that particle formation also happens naturally under radiative influenced conditions also at higher altitudes (see. Fig. S9).”

to:

“PNC_{>10 nm} reflects processes in the particle phase and can be influenced by anthropogenic emission sources at ground level or by new particle formation (NPF). Figure S2 shows that, although polluted air doubles the particle number concentration, anthropogenic air masses advected from the Frankfurt area are already well-mixed at the measurement site and do not show a gradient in PNC_{>10 nm}. Continuous size-resolved particle size distribution measurements (>7 nm) by MoLa at ground level indicate NPF starting around noon on most days, when NPF-precursors are well-mixed in the lowermost layers (Fig. S9). Therefore, NPFs happens along the whole vertical range. Conclusively, the gradient of particles, observed in the morning, is likely due to regional emissions near the ground, when convection has not homogenized the material PBL, yet.”

- P10/L312: I thought the FLab measurements were up to 500 m only. Where did the data at 1300 m come from? Remote sensing from MoLa? Please clarify.

Indeed, Flab measurements were limited to 500 m above ground level, however, the planetary boundary layer height (PBLH) was estimated by model data, which covers the altitude range from

0 m to several thousand meters above ground level. This was clarified in the sections' first paragraph: *The PBLH was derived from the ICON-D2 model as described in Sect. 2.2.*

- P16/L411: it is not clear whether the 0-25 m results were from ground-based measurement only, or the average of the bins below 25 m from the UAS measurements. Please specify.

In Sect. 4.3.1 we now specify that we have only used UAS measurements for this part of the study:

“R is the ratio of the mean vertical column value of the lowermost altitude bin (0 - 25 m a.g.l., for 25 m vertical resolution, from drone measurements only) for a certain variable to the respective mean column value over the lowermost 500 m range and [...].”

- P20/L509: the underestimation of O3 is much larger than 5% shown in Figure 6, and the statement here is inconsistent with the statement in P23/L613. Am I missing anything?

You are right, including the largest uncertainty bar we end up with an $R = 0.94$, i.e., an underestimation of 6% (right y-axis in Fig. 6). This was corrected. The statement in P23/L613 refers to the morning and evening hours when several atmospheric layers were present in the lowermost 500 m. However, the values given in P20/L509 refer to stratification-free conditions, as now implemented in the text.

“Mixing ratios of trace gas variables as CO2 and O3 in the stratification-free lowermost 500 m can be measured at ground with a slight underestimation by 0-2% and 0-6%, respectively.”

- Section 4.4: the first point of the recommendation is about estimation of PBLH, which makes this section not just about the representativity of ground-based measurements. Better to merge it into Section 5.

Here, we argue from the point that knowing the PBLH helps to estimate the pollutant distribution, see the whole section 4.2.1. With more accurate (more or less frequent) experimental estimations of the PBLH, model estimations are expected to be improved on the long term. In the summary, we refer to this in P25/L630:

“Further we propose that a vertical characterization should be conducted for every ground-based measurement site to estimate site-specific effects and validate the use of model-derived PBLHs to estimate the range of representative ground-based measurements.”

- Figure 2: should the schematic sketch of $dPNC/dz$ vs. $\ln(z)$ (panel a) have a positive slope as what is shown in cloudy days (blue) data points in panel c?

Thank you for pointing this out. The “ $dPNC/dz$ ” in the right bottom graphic in Fig. 2a) on the y-axis was supposed to show the absolute value of the gradient. For clarity, the blue dotted line was flipped and now shows the value itself, i.e. an increase from a negative value at very small $\ln(PBLH)$ to larger (negative) values at larger $\ln(PBLH)$. Additionally, we added the linear approximation of $PNC(z)$ in panel a) to clarify that $dPNC/dz$ was determined from this linear approximation and not from the exponential functions which are theoretically expected. This was added to the legend. z_1 was changed to $PBLH_i$ as we describe the gradient-dependency with respect to the PBLH and not to an arbitrary altitude. The new Fig. 2 is shown here:

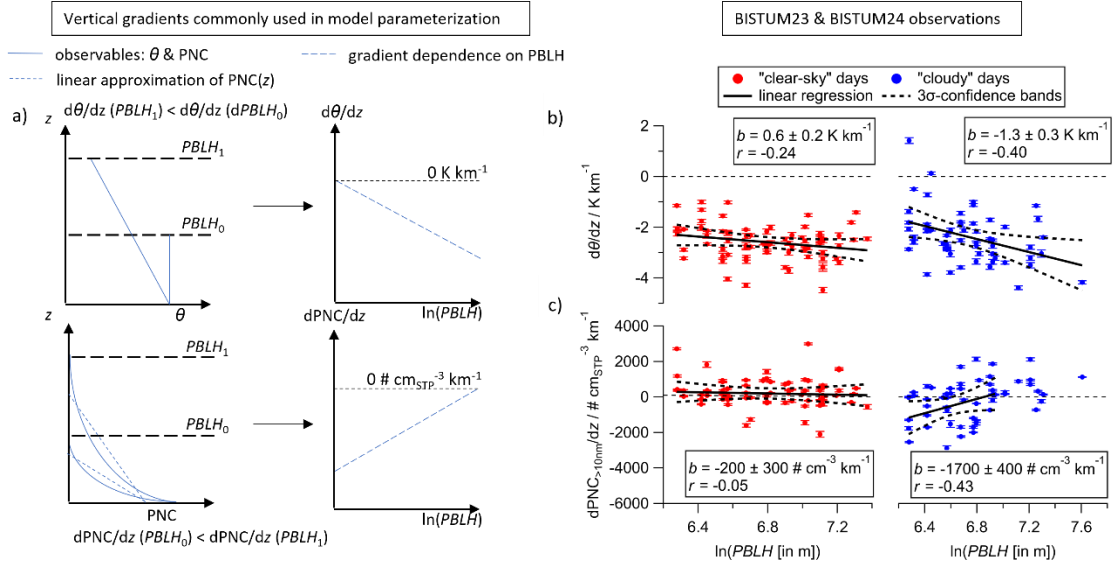


Figure 2: (a) Schematic description of the dependency of atmospheric stability (reflected by $\theta(z)$, with the altitude z) and a pollutant with source at ground level ($PNC(z)$) on altitude for the well-mixed planetary boundary layer with a height $PBLH_i$ as commonly used in model parameterizations (a, left). Deriving the dependence of $d\theta/dz$ and $dPNC/dz$ from PBLH results in a linear-logarithmic dependency of respective gradients on the boundary layer height (a, right). Experimental data of gradients of the potential temperature θ (b) and the particle number concentration $PNC_{>10\text{ nm}}$ (c) in the lowermost 500 m in a mixed PBL from both campaigns are plotted versus associated logarithms of respective planetary boundary layer heights ($PBLH$, in m). Linear regression with the slope b and its 3σ-confidence bands are depicted in solid and dotted lines.

Figures 4 and 5: the legend labeling is a bit misleading. Should it be (closed blue circle): “cloudy” days for 0 – 500 m, (open blue circle): “cloudy” days for 0 m to $PBLH$, (closed red circle): “clear-sky” days for 0 – 500 m, and (open red circle): “clear-sky” days for 0 m to $PBLH$?

Thank you for this comment. We changed the legend according to your suggestion to make the graphic easier to read.

- Figure 6: better to rearrange parameters in the x axis according to the discussion, i.e., air pollutants first, and then meteorological parameters, as two clearly separated categories.

We have already tried to separate the parameter type according to the discussion (from left to right: pollutants, meteorological parameters on each x-axis). This was done using two x-axes, so that the representativity R can be scaled differently to better display the uncertainties from the analysis. The left y-axis scaling covers $R = 0.5 - 1.8$ and the right y-axis scaling included only $R = 0.9 - 1.15$. We prefer not to change the order to keep the scaling easy to read.

- P8/L242: a typo for $PBLH$.

This was corrected.

- P13/L352: “permanent” to “static”?

This sentence deals with temporal variability, hence “not permanent” seems to be the better choice to us.

- P14/L384: “stronger” to “more strongly”.

This was changed.

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