

Dear Prof. Giorio

Please find below our response to the three most recent reviews of our manuscript. We would like to thank the reviewers for their effort in reading our manuscript and giving us feedback.

As mentioned in a previous email, we suspect that the review by Reviewer #3 / Report #2 was written by a large language model / AI system. As requested, we have addressed these comments to the best of our capabilities.

In our point-by-point responses below the reviewers' original comments are in black, while our responses are in blue.

Reviewer #2, Report #1

(reviewer recommended publish as is without any specific comments)

We would like to thank reviewer #2 for supporting the publication of our manuscript as is. We are pleased that the reviewer confirms that all the comments brought forward in the first round of reviews have been adequately addressed.

Reviewer #3, Report #2

The use of a multi-path LP-DOAS system to achieve continuous vertical profiling of trace gases at 20–200 m represents a significant methodological improvement over conventional balloon-borne snapshot soundings or single-level fixed-point monitoring. The long-term time-series profiles of trace gases within the Fairbanks winter persistent stable boundary layer (PSL) help fill an important observational gap in the vertical distribution of pollutants under stable stratification. I therefore recommend the manuscript for minor revision. However, I have several specific points that require clarification or further discussion:

We acknowledge the submission of these comments on our manuscript. However, we want to note that we believe that these comments were either written by a large language model (LLM/AI) or that the reviewer has not actually read our manuscript. Below we will provide reasons supporting this assessment in the responses requested by ACP.

1. Signal intensities and SNR considerations (Figure 1): What are the actual signal intensities at the 3963 m and 1154 m paths, respectively? Given that the optical path

lengths range from 1154 m to 3963 m, the instrument signal-to-noise ratio (SNR) likely varies substantially among the different distances. How is the reliability of the retrieved data assessed across these varying SNR conditions, and what criteria were used to ensure the robustness of the derived products?

Since we are using Differential Optical Absorption Spectroscopy (Platt and Stutz, 2008)) the intensity of the absorption measurements has minor relevance to the retrieved concentrations or the scientific content of the manuscript. It is well known that intensity is, in most cases, not the determining factor in the errors of the retrieved concentrations. The methodology on how to calculate uncertainties in LP-DOAS measurements is described at length in many publications and summarized in the book by Platt and Stutz (2008) cited in the manuscript. This methodology specifically considers photon noise, which varies with pathlength. Reiterating this methodology is beyond the scope of the manuscript, and we believe that the following sentence in the Supplement is sufficient: *“Data is displayed as mixing ratio at ambient temperature together with the measurement errors that were determined by each fitting procedure (Platt and Stutz, 2008; Stutz and Platt, 1997).”*

All uncertainties of the reported LP-DOAS mixing ratios were propagated based on the established spectral retrieval of the measured absorption spectra and thus include noise variation due to different path lengths. All DOAS data is displayed with error bars and reported with errors respective to their specific absorption path in the manuscript and considered in our scientific results.

2. Interpretation of O₃ variations (Figure 2): Can the observed ozone changes in Figure 2 be unambiguously attributed to stratospheric intrusion events, or are there other plausible boundary-layer processes that might explain the variability?

We do not understand this question. At no point in the manuscript do we discuss stratospheric intrusions as they are not relevant to our case. The entire manuscript is dedicated to explaining that ozone variation stems from the emission of NO at the surface followed by the NO + O₃ reaction and vertical mixing.

3. Near-surface enhancement of HCHO and HONO (Figure 2): The concentrations of HCHO and HONO in the lowest layer (12–17 m) are markedly higher than those at all other heights. Could the authors elaborate on the underlying mechanisms responsible for this pronounced near-surface enhancement?

This was explained in the manuscript at lines 203-206:

“HONO mixing ratios and their vertical profile show a similar, but not identical, behavior to NO₂ and SO₂. While it is beyond the scope of this study to determine the source of HONO, we can speculate that HONO is directly emitted, but we cannot rule out a contribution from secondary chemical formation. HCHO mixing ratios seem to follow those of the other trace gases, but have a larger variability.”

Reiterating our previous point, the entire manuscript is about the accumulation of primary pollutants in the polluted surface layer and we believe, since HONO and HCHO are not the focus of the manuscript, further explanations would distract the reader from our main scientific questions.

4. Role of molecular mass in vertical dispersion (Figure 4): The discussion on wind speed effects in Figure 4 is interesting. I would like to ask whether the vertical dispersion range of gases is expected to be dependent on molecular mass, and if so, whether such a mass-dependent effect might influence the observed vertical gradients.

The troposphere is turbulent and gases are well mixed. They do not separate due to their molecular weight. Because this is common knowledge to the reader of ACP, we decided against addressing this question in the manuscript.

We would like to add here that trained atmospheric scientists would not ask this question, which is a clear indication that this review was written by an LLM/AI.

5. Horizontal representativeness of the vertical profile (Figure 6): The measurements in Figure 6 span heights from 12 to 191 m and are reported as column concentrations. Where is the horizontal location of the retrieved profile relative to the light paths? More specifically, how is a given vertical level linked to its corresponding horizontal position along the multi-path configuration?

Figure 6 shows retrieved mixing ratio profiles based on observations and the corresponding model results, not “column concentrations”. We assume horizontal homogeneity, as explained in lines 170 -181, and argue that the vertical profiles are representative of the downtown Fairbanks area. This topic was thus already addressed in our manuscript.

6. Selective presentation of time-series (Figure 9): Time-series plots are shown for O₃ and HONO in Figure 9, but NO₂ is not included. Is there a specific reason for omitting the NO₂ time series? Including it, or explaining its absence, would improve the completeness of the

analysis.

Figure 9 does not include HONO and Figures 2, 3, 6, 7, 8, 10, S1, S2, S4, and S5 include NO₂ mixing ratios. The unjustified claim that data is selectively presented can only stem from the fact that an LLM/AI system is unable to fully read our figures.

7. Uncertainty in vertical profile retrieval: The LP-DOAS system uses only four light paths, yet the manuscript presents vertical profile results with relatively high spatial resolution. A more detailed quantitative assessment of the retrieval uncertainty—particularly regarding the spatial resolution and precision achievable with such a limited number of optical paths—is essential. If such an assessment is not feasible, the authors should at least explicitly acknowledge the limitations in terms of vertical resolution and accuracy in the conclusions section.

This is extensively discussed in the second paragraph of Section 2.3 and the retrieval uncertainties are shown explicitly in the example in Figure 8 (gray shading representing errors in Figure 8). In addition, the supplement includes Figures S4 and S5, which show how well the retrieval captures the actual measurements.

Reviewer #4, Report #3

This manuscript describes the long-path DOAS measurements taken during the ALPACA campaign in Fairbanks, Alaska in the winter of 2022, when shallow polluted boundary layers often persist for several days due to stagnant air conditions from temperature inversions. Using a new fitting algorithm, they used LP-DOAS measurements at 4 heights to construct a vertical profile of NO₂, O₃, SO₂, and HCHO within the polluted layer, and then constructed a PACT-1D transport scheme to characterize the movement of pollutants throughout the layer.

It's a good study, well defined and described, and combines observations with theoretical modeling to understand the vertical structure of polluted layers in Fairbanks, which can be one of the most polluted places in the US in winter. The analysis of residence times within the PSL is particularly relevant. I would recommend publication after the authors address a few issues, listed below.

We would like to thank the reviewer for their encouragement and helpful comments. We have addressed all the comments and provide a response and the changes made to the manuscript below.

Figure 1: It isn't clear where the CTC is located. Is it at the orange triangle? How far is that from the parking garage? Can you put this information in the caption?

Thanks for pointing out that this information was missing. We added the following text to the caption of Figure 1: *"In-situ measurements were located at the CTC building marked by the yellow triangle. The distance between the LP-DOAS and the CTC building is approximately 500~m."*

Line 179: Do the 3m and 158m in situ O₃ measurements agree with each other or with the retrieved values? I would expect the latter, but the phrasing of this sentence makes it unclear.

We agree the sentence was unclear we added "with the retrieval results" so that the sentence is now clearer:

"In particular, the agreement between the 3~m surface mixing ratios and the 158~m Birch Hill ozone in situ measurements with the retrieval results show that our retrievals are not noticeably impacted by horizontal inhomogeneities and that we can use the retrieved profiles to analyze the vertical structure of the Fairbanks atmosphere."

Line 194: "Further formation of NO₂ from continued emissions of NO, which can lead to large spikes in NO_x, does not contribute to NO₂ due to a lack of ozone" is a confusing sentence. Do you mean that continued emissions of NO stop forming NO₂ at some point once the ozone is titrated? Maybe rephrase.

We agree that this sentence was not very clear and revised it as:

"Further formation of NO₂ from continued emissions of NO is inhibited by the lack of ozone. NO emissions under these conditions can lead to large spikes in NO_x that are not reflected in NO₂."

Figure 3: It's hard to see which day is which when the ticks only mark every 5 days. Can you include more ticks, or put a box around the time periods that you discuss in further detail in the text?

That's a good point. We added minor ticks for every day on every x-axis in the plot. Boxes would make this plot even more complicated, especially with the color panel on the top, so we decided against them.

Line 229: It seems odd to include the no-PSL case in the lowest dT bin. Doesn't this throw off the statistics of the plot? Could you only include PSL cases in this statistical analysis? Likewise with the wind plots

The current analysis has a 200m cut-off due to the height of our retrieval domain. Because the transition from a PSL to a more mixed atmosphere is continuous, i.e. we go from a box-like profile to a less well-defined profile/PSL (Figure 8a,f,k vs. Figure 8 c,h,m), any cut-off would be arbitrary. Excluding no-PSL cases could thus lead to an erroneous representation, and we believe that including all heights we retrieve and showing the spread of the data is the best approach.

Line 266: What is the ADEC?

Thanks for catching this. It is the "*Alaska Department of Environmental Conservation*". We have written out the name at the first mention of ADEC.

Line 273: What is the correction suggested by Brett et al 2025? How close is it to the 2.3 and 3 factors that you used? How sensitive are your results to this selection of the scaling factor?

Brett et al. (2025) used a log-linear function from a NO_x enhancement factor of 1.5 to a factor of 10 for daily average temperatures between 0 and -40 °C. At -20 °C, the typical temperature in the period we analyzed, their enhancement factor is ~3. We modified our text to more clearly state this: "*..aligns well with the suggested correction of approximately 3 for total NO_x for -20°C by Brett et al. (2025)...*"

As discussed in Supplement Section S3 and shown in Figure S2, the results are quite sensitive to the NO and NO₂ emissions. The factors we used for our final model runs are those that lead to the closest match between model and observations. Because the purpose of our study was not to improve emission inventories (this was already done in

much more detail by Brett et al. 2015), we think the discussion in Supplement Section S3 is sufficient to give the reader an impression of the sensitivity.

Section 5.1: Although a good illustrative case study, I'm not sure this section is adding anything to your analysis and could be removed or moved to the SI. Figure 9 is also quite hard to read due to the thin line traces.

We are glad that the reviewer has been convinced by the residence time analysis in our paper. Unfortunately, not all colleagues have an easy time accepting our calculations, as it goes somewhat against the idea that stable boundary layers are stagnant. The example in Figure 9 serves as direct observational evidence on how fast the PSL fills up with pollution and that PSL mixing ratios reach a pseudo-steady-state within 2-3 hours that can only be explained by mixing of pollutants out of the PSL. It is thus an important piece of information supporting the main conclusions of our manuscript. For those reasons we very much prefer to leave it in the main part of the manuscript.