Response to Reviewer 1 of "Vertical distribution of pollution trapped by wintertime surface inversions in Fairbanks, Alaska, during the ALPACA experiment" by Stutz et al.

We thank the reviewer for the review and the valuable comments. We have carefully considered the various comments in the review and changed the manuscript accordingly. Our detailed responses to the questions/comments (black) are provided in blue. Additions to the manuscript and supplement are in italics:

Specific comment

Chapter 2

The retrieval assumes that concentration varies only with altitude, but the upper light paths are long slant paths extending roughly 4 km toward across non-uniform terrain and likely non-uniform source intensity. I think how horizontal gradients might alias into the retrieved vertical gradients should be discussed. A simple 2-D synthetic test or at least a representativeness discussion would greatly strengthen the interpretation. Additionally, the four reflector arrays are observed sequentially rather than simultaneously, therefore the rapid transition may induce apparent vertical structure or timing lags. How can these uncertainties be evaluated?

Thanks for pointing out that we omitted an explanation that we temporally interpolated the measurements of the upper three paths to the measurement time of the lowest path. Our experience has shown that path-averaged data does not vary rapidly, as rapid/small-scale variations in trace gas concentrations are averaged/smoothed out. This temporal interpolation is thus sufficient within the uncertainty of the observations and retrievals. We would like to note that the measurement errors were propagated though the interpolation, which were then used in the retrieval. We recognized that this was not explained in the manuscript and have added the following sentences:

"*To derive the four parameters in Equation 1, we first interpolated the path-averaged LP-DOAS data from the upper three light paths onto the measurement time of the lowest light path, while propagating the measurement error to the interpolated data. Because path-averaged data varies slowly, a linear interpolation was chosen. Similarly, we averaged the in-situ data over the time interval of the lowest light path measurements. A non-linear Levenberg-Marquardt fit then optimized the model function describing the altitude averaged, time interpolated, LP-DOAS data and the concentration of in-situ observations.*"

We agree with the reviewer that a discussion of the impact of potential inhomogeneity would be useful for the reader. We can discuss two unique cases: In the case of a stable polluted surface layer the air aloft predominately originates from the unpopulated areas outside of Fairbanks and we therefore expect that it is vertically and horizontally fairly uniform, i.e. our assumption of horizontal homogeneity is likely quite good. The good agreement between the retrieved ozone levels at 158m with in-situ measurements at the same altitude confirms this conclusion. In the second case, the atmosphere is well mixed, which should generally reduce inhomogeneities. However, in cases in between these extremes, horizontal inhomogeneities could occur.

The presence of substantial horizontal inhomogeneities would lead to complex vertical profiles that do not follow our sigmoidal model function. Consequently, the retrieval would not reproduce the observations. Thus, we can argue that good agreement between retrieved and observed mixing ratios can serve as a measure for the accuracy of the retrievals. We therefore included two additional figures in the Supplement (S4 and S5) to show the performance of the retrieval for NO_2 and ozone and the following text to the manuscript.

"Our retrieval assumes horizontal homogeneity within the extent of our light paths. The very good agreement of NO_2 and SO_2 on the lowest light path with the in-situ observations (Figure 2) confirms that this path is representative of surface mixing ratios in downtown Fairbanks. In the presence of a PSL, the volume probed by the upper three light paths is dominated by background air, which is typically well mixed vertically and horizontally. Our retrievals confirm that in most cases air above a well-developed PSL does not show a vertical profile within the uncertainties (see below). In the case of a well-mixed atmosphere, horizontal inhomogeneities are also reduced. In cases of intermediate mixing this assumption may, however, not strictly hold. Because horizontal inhomogeneities would distort vertical trace gas profiles, we can use the agreement of the retrieved point and altitude-averaged mixing ratios with the observations to assess whether horizontal inhomogeneities would prevent interpretation of our vertical profiles. Figures S4 and S5 show that the agreement of the retrieved and observed mixing ratios for O_3 and NO_2 is excellent. In particular, the agreement between the 3 m surface mixing ratios and the 158 m Birch Hill ozone in situ measurements 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."

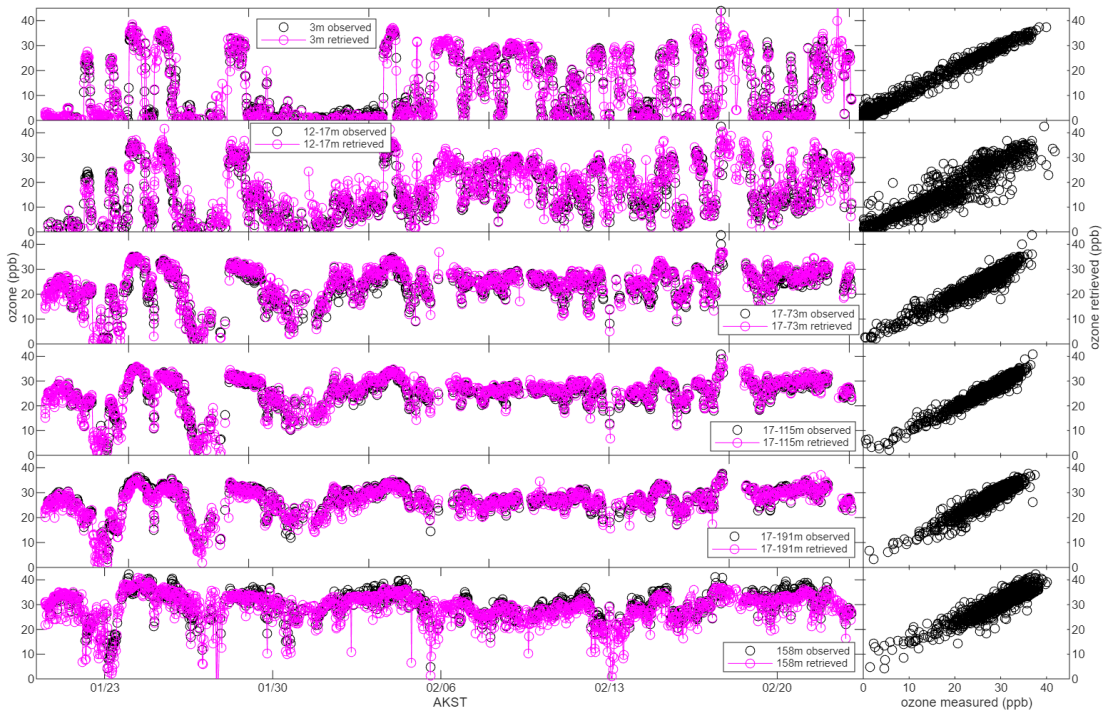

Figure S4: Comparison of retrieved (magenta) and observed (black) in-situ and path-averaged ozone mixing ratios. The panels on the right show the close correlation between retrieved and observed values.

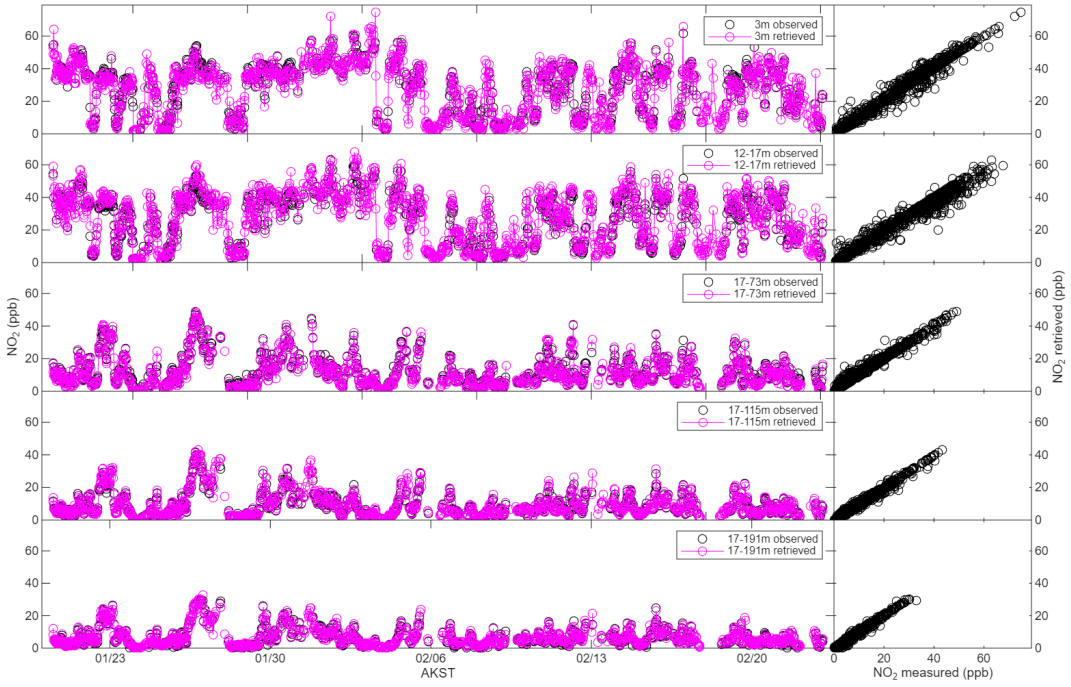

Figure S5: Comparison of retrieved (magenta) and observed (black) in-situ and path-averaged NO2 mixing ratios. The panels on the right show the close correlation between retrieved and observed values.

For NOx measurement, chemiluminescence analyzers commonly use molybdenum converter, which can convert other NOz species (e.g. HONO, HNO3, ...); Especially in high NOx region with the existence of oxidant, its influence can be enlarged that be expected. I think the model can represent this measurement uncertainties and its impact on analysis if there is a proper chemical mechanisms.

Thanks for pointing out that this deserves an additional explanation. Because emissions are fresh, residence times are short, and oxidant concentrations smaller than at lower latitude the levels of NO_z species are typically much lower than NOx concentrations. Consequently, NO_z interferences from heated molybdenum converters do not affect our results and conclusions. We have added the following text to the Supplement to explain this in more detail. The figure from Roberts et al. (2025), which we refer to in this text, was added for clarity to this response but not to the manuscript itself:

"*NO_2 was measured by converting it to NO via a heated molybdenum converter. Molybdenum converters are known to partially convert $NO_z = NO_y - NO_x$ to NO, thus potentially introducing artifacts in measured NO_2 mixing ratios. However, in Fairbanks winter, freshly emitted NO and NO_2, as well as NO_2 from the rapid reaction of NO with ozone, are the dominant part of NO_y. Potential interfering species, such as HONO and nitric acid, contribute at most a few percent to NO_y. This understanding of NO_x /NO_y chemistry in Fairbanks has been confirmed by a direct comparison of NO_x and NO_y by Roberts et al. (2025) (Figure S1 in Roberts et al. (2025)), which shows a close correlation of R^2=0.99 and a slope near unity. Landis and Edgerton (2024) also compared a heated molybdenum converter instrument under high NO_x conditions in northeastern Alberta, Canada, with a spectroscopic and chemiluminescence with photolytic converter NO_2 instruments. They found agreement of NO_2 of better than 7% over NO and NO_2 mixing ratio ranges similar to the one we report here.*

Both studies thus confirm that under conditions of fresh emissions, as found at the surface in wintertime Fairbanks, NO_2 observations using a heated molybdenum converter are not noticeably impacted by potential NO_z interferences. It should be noted that these considerations do not contradict prior findings of interferences introduced by heated molybdenum converters; rather, they show that conditions in Fairbanks make these interferences too small to affect the conclusions of our study. "

The following figure is from the electronic supplement of Roberts et al., 2025 and shows the close correlation between NO_x and NO_y.

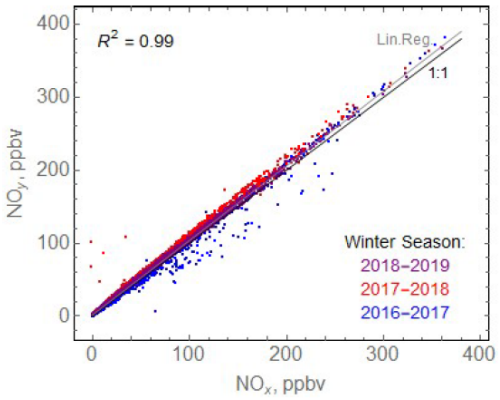

Figure S1: Scatter plot of NO_y versus NO_x measured by ADEC analysers at the NCORE site in Fairbanks during the winter seasons (November to February) of 2016-2017 (blue), 2017-2018 (red) and 2018-2019 (purple).

Roberts, T. J., Cesler-Maloney, M., and Simpson, W. R.: Low-cost electrochemical gas sensing of vertical differences in wintertime air composition (CO, NO, NO_2, O_3) in Fairbanks, Alaska, Faraday Discussions, 258, 307–327, doi: 10.1039/D4FD00177J, 2025.

Chapter 3

Could the authors explain why NO2 was selected as the primary tracer for identifying PSL structure, rather than SO2 or a combined multi-species framework? Because NO2 is not significantly stable in aspects of chemical cycle.

While this was already briefly explained in the manuscript (lines 193-194), we acknowledge that our explanation may have been too short. We have therefore expanded this discussion and now state:

"*NO_2 was selected due to its small measurement error and because it has the largest dynamic range, i.e. the difference between mixing ratios at the surface and aloft, of all the observed trace gases. In addition, the 3 m in-situ observations as well as the lower light path mixing ratios have a smoother temporal variability than SO_2 (see Figure 2), making the profile retrievals, and thus the interpretation of the PSL height, more reliable*."

Chapter 4

Lots of descriptions are related to the model setup. I think 4.1-4.3 should be moved to supplementary information. Or I recommend constructing methodology chapter for the basic information about observations and models before result and discussion.

We understand the reviewers' concern. However, the model, and thus its setup, is a crucial component of our interpretation strategy. In particular, how we implemented our conceptual understanding of the PSL behavior in the model is important to fully comprehend our main conclusions. We thus believe these sections should be part of the main text.

We initially thought about combining observation and model methods in one chapter, but found that it was difficult to read, as the model setup is informed by the field data and its interpretation, especially regarding the surface layer height behavior (section 3.3). The structure of first discussing observations (Chapters 2 and 3) followed by the model description and results (Section 4) will make it easier for the reader to follow our arguments and help understand why we set up the model in the way described in section 4.1 – 4.3.

Also considering the observed condition (~200 ppbv of Nox), scaling NO emissions by 2.3 and NO2 emissions by 3 is a major adjustment. Please show unscaled vs scaled simulations to clarify whether the main conclusions are robust to the exact scaling factors. Especially the difference of scailing in NO and NO2 can change the NO/NO2 ratio, which strongly impact to O3-Nox cycling.

As stated in lines 251 – 255 of the original manuscript, the fact that the current ADEC NO_x emissions are too low was already discussed by Brett et al. (2025). Following the reviewer's suggestion, we added a section to the supplement comparing model results with and without the emission scaling factor with the surface observations.

We added the following statement at the end of Section 4.2 in the main text:
"… , as discussed in more detail in the supplement (Section S3)."

And the following section to the supplement:
"***S3 Scaling of NO and NO_2 emissions***
As mentioned in the main text, Brett et al. (2025) reported that the current ADEC emission inventory substantially underestimated NO_x, most likely due to uncertainties in the emission factors of combustion sources at the cold temperatures in Fairbanks. We confirmed this result by comparing a run of our model with the original ADEC emission data (yellow line, Figure S2) with a run where NO emissions were increased by a factor of 2.3 and NO_2 emissions were increased by a factor of 3 (red line, Figure S2). With the original ADEC emission data, modeled NO mixing ratios are considerably lower than the observations. Modeled NO_2 mixing ratios are slightly smaller than the observations, while modeled ozone mixing ratios are slightly larger. The smaller model-observation difference of NO_2 and ozone

can be explained by the efficient conversion of NO to NO_2 by the $O_3 + NO \rightarrow NO_2 + O_2$ reaction. The run with the increased NO and NO_2 emissions leads to much better agreement with the observations for all three trace gases (red line, Figure S2). This independently confirms the findings by Brett et al. (2025) that ADEC NO_x emissions for Fairbanks are too low."

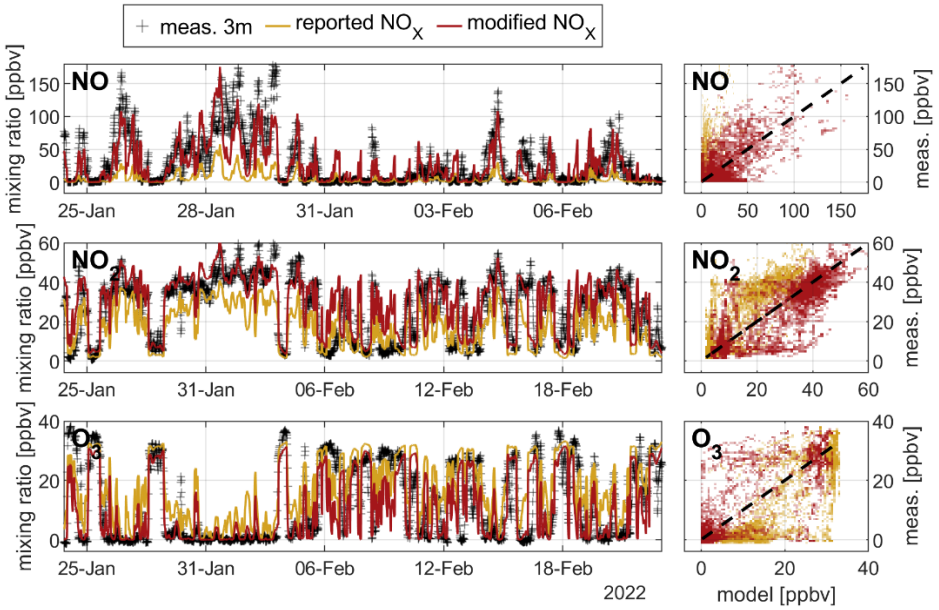

Figure S2: Comparison of model runs using the original and scaled ADEC NO and NO_2 emissions with 3 m in situ observations.

Chapter 5

The residence-time analysis is interesting. I recommend additional sensitivity tests to another condition (e.g. deposition). These points matter because the residence-time estimate is one of the paper's most policy-relevant conclusions.

Our model and the residence time calculation include deposition for reactive trace gases. The latter was mentioned in line 325 of the original manuscript. The turbulence-advection scheme is based on measured wind speeds and the surface layer heights. The variability of the residence time throughout the studied period reflects its sensitivity to windspeed and surface layer height.

We re-analyzed our model results and even during the most stable polluted scenarios the loss through deposition is at most 10% of the total pollutant loss (Kuhn et al., 2026). It will thus affect the residence time by a small amount, which is already included in our analysis. Because varying the deposition, for example in a sensitivity study, would modify the overall

residence time by an even smaller amount, we have focused our additions to the text explaining the role of deposition on the trace gas budget.

We have added the following sentences to the manuscript to clarify that deposition is included in the model and that its contribution to the residence time is small.

In section 4.1 we added a new sentence:

"*The model considers molecular uptake of trace gases on the ground, which, in combination with the transport scheme, leads to deposition (see Tuite et al. (2021) for details).*"

We also modified a sentence in Section 5.4 which now reads:

"*An analysis of the SO_2 budget reveals that turbulent mixing and advection are the dominant processes controlling SO_2 (and other trace gas) concentrations, while chemical loss and deposition of SO_2 account for less than 10% of the total budget in our case and thus of lesser significance (Kuhn et al., 2026).*"

Supplementary

I'm not sure about the temperature dependency of absorption cross section. The in-situ condition is not similar to room temperature, which can induce strong uncertainties in HONO quantification. Please clarify about this limitation.

The temperature dependence limitation for HONO was specifically discussed in the supplement (lines 9 – 10), where we stated: "The temperature dependence of HONO absorptions is currently unknown, but it is likely in a similar range (< ~10%) than NO_2."

We do not believe that the temperature effect on HONO concentrations would be strong, but without a low-temperature HONO cross section measurement, which is outside the scope of our study, the best we can do is state explicitly which cross section was used. We also note that the temperature dependence of the HONO absorption cross section has no impact on the main conclusions of our manuscript.