We would like to sincerely thank both reviewers and the editor for their thorough work and the helpful comments on our manuscript. The feedback helped us to improve the readability, clarify the proceeding of the analysis described and significantly improve the discussion of our manuscript.
In this document, we reply to the reviewers comments as follows: all remarks by the reviewers are listed in black text, and our corresponding replies are given in blue text. In addition to the referee comments, we also edited the copyright statement of Figure 1 directly in the revised manuscript, as proposed by the editorial support.

Responses to Anonymous Referee#4

The authors present an interesting and novel study outlining the use of drone-based vertical profile measurements of $O_3$and NO (collected over a 2-day field campaign) in a 4D-Var data assimilation system coupled to the EURAD-IM CTM to optimize the vertical and spatial distribution several pollutants, as well as the related emission fields. These drones may serve to fill a gap in our observations, particularly within the PBL where few vertically-resolved measurements exist. In general, the manuscript is well written and easy to follow, although I did find there to be quite a lot of minor formatting and grammatical errors throughout.
While the authors do present some reasonable evidence to support the conclusions that the drone observations lead to an improvement in the simulation of these trace-gases and that optimized emissions can be obtained, my largest concern is with the robustness of the results. In particular, I am not entirely convinced with the optimization of the emission fields, given the fact that the assimilation was only performed over a very short time-period and with generally few observations.

We would like to thank Referee#4 very much for seeing the value of our manuscript and for identifying the weaker points of our analysis. We concur that there are a few limitations in the evaluation mainly due to the limited availability of observations. However, we intended to pursue the analysis taking into account the limiting factors and discussing these accordingly. We think that with the analysis, we are still able to highlight the potential of drone observations for atmospheric chemical data assimilation.

In this iteration of the review, we have addressed the remaining critical points raised by both reviewers. We focused on discussing more clearly the limitations imposed by the experimental design and how they should be considered when interpreting the results.

Major Comments:

- P6, L150-151–The authors mention they only used the ascent profiles from the drones in the assimilation exercise due to the “higher accuracy”. What is exactly meant by this – why were the descent profiles less accurate? Shouldn’t it be the same if the ascent/descent rate is fixed at 1m/s.

  Similarly, how much does the result change when assimilating both the ascent and descent profiles in the 4D-var system? This could give some indication of how sensitive the assimilation system is to additional measurements (particularly those that might be more uncertain). Due to the overall low number of observations being assimilated over a short window of less than 12 hours on each day, I suspect that this could have a notable influence on the results and is important to investigate.

  The authors even highlight this themselves later on when discussing the optimized emissions; P12, L231-233 “However, their generalization and significance should be rated carefully, mainly because of the limited number of drone profiles being available, the short assimilation windows selected, and the deficiency to perform a long-term statistical analysis”.

We understand the intent of the referee to take the descending profiles into account to include a larger number of profiles. The choice to keep only the ascending profiles is however justified by the design of the drone and the instrumental setup. Since the propellers of the drone are located below the instrumental load, the turbulence created during the descent can disturb the sensor detection. This is in particular evident for the electrochemical sensors even if the ascent/descent rate is constant. By focusing on the ascending profiles only, this interference is minimized due to laminar flow around the sensor surface, such that the data obtained is probing the atmospheric composition more accurately.
Certainly, the number of observations generally impacts assimilation results. However, the measurement error is also a critical factor influencing the analysis. In the case of the drone flight operations only up to an altitude of 350 m, the ascending and descending profiles were taken within a very short temporal window. For most of the flights, the pairs of ascending and descending profiles were thus detected during nearly the same model time step. Thus, assimilating two observations – one more accurate with a smaller observation error, one less accurate with a large observation error – during the same or a subsequent time step would not provide major additional information and not change the results much. Our priority was to retain the most reliable observations to avoid overfitting of the model.
For a better analysis and optimization of emissions, a longer assimilation window with more temporally even distributed observations would be beneficial. For example, additional observations in the afternoon and evening would have provided valuable information about the temporal evolution of pollutants in the lower atmosphere and thus indirectly also information about the occurring emissions.
To clarify the text passages discussed by the referee, we have rephrased them and added the corresponding explanations in the manuscript.

- From Figure 3, there appears to be a very significant bias in the modeled vs. measured NO profiles on the order of ˜30 ppbv, and if this difference was placed in relative terms (in %) it would appear even more extreme. This is only very briefly addressed in Section 4.1, L215: "On both days, the reference simulations underestimate the NO vertical distribution at all heights, with the strongest discrepancies at ground level.". I believe a greater investigation of the source of the bias should be performed or at least explained here, as it may indicate a broader issue in the model (e.g., incorrect NO_x chemistry or partitioning between NO and NO_2). I think this is particularly crucial if you are going to assume that the partitioning and chemistry of NOx is correct in the model for the purposes of estimating the NO_2 emission corrections based solely on the optimized NO fields.

We would like to express our gratitude for this discussion item. One reason for the discrepancy between the modelled and the observed profiles occurs due to the nature of the model, which relies on relatively static model input parameters (in terms of emissions e.g. not taking into account exceptional emission patterns as for example traffic jams or bypasses due to road closures) that are not designed to reproduce a single "point" observation. This is a well-documented challenge associated with regional chemistry transport models. Local sub-grid emissions, which are not accurately represented in the model, appear to be underestimated in the simulation results as revealed by our analysis.
A more detailed examination of the origin of the discrepancy is beyond the remit of this manuscript, which explores the potential of drone data to be used for data assimilation. However, we are aware of an imbalance of ozone and nitrogen oxides at night. The O_3 minimum is overestimated and NO_2 is underestimated (e.g. Lange et al. (2023); Gauss et al. (2024)). This issue is currently under investigation and will be discussed in more detail in a forthcoming manuscript.

- P13, L263-264, the authors write "However, it is unfortunately not possible to directly obtain information about the NO_2 emissions from the TROPOMI data.". While this is somewhat true, in theory information on the NO_2 emissions could be derived from assimilating the S5P observations in the 4D-var system. I'm not necessarily saying this must be done in this paper as it may fall slightly outside of the scope, but it could provide an interesting comparison with those optimized emissions obtained solely by assimilating the drone observations of NO. I think it would also provide increased confidence in the results if separately assimilating both datasets provided a similar result in terms of the optimized emission fields (at least for NO_2) for these days.

We agree with the referee that assimilating TROPOMI NO_2 column data would indeed provide an opportunity to evaluate the analysed emission corrections of our analysis. However, such an additional analysis would be beyond the scope of our manuscript and would introduce a completely different focus as it would include an additional, completely different set of uncertainties. For future 4D-var analyses assimilating drone observations, we can imagine including a comparison with data assimilation analyses assimilating data from ground-based monitoring stations and/or satellite data.

Minor Comments:

- P1, L6: "4D-var takes advantage of the inverse technique...", I think it would be better here to say something along the lines of "4D-var is an inverse modelling technique". There is no single inverse technique, but rather a slew of inverse modelling approaches and methods.

  Thank you for bringing this to our attention. We have made the suggested change in the abstract: "4D-var is an inverse modelling technique that allows for simultaneous adjustments of initial values and emissions rates."

- P2, L25-29, the authors state that very few ground-based monitoring networks exist that provide vertically resolved measurements of these pollutant species. They mention LIDAR and sonde networks, but fail to mention ground-based spectrometer networks such as the Network for the Detection of Atmospheric Composition Change (NDACC) or the Pandonia Global Network (PGN; global network of Pandora UV-Vis spectrometers). These networks provide vertically resolved measurements of O_3 and NO_2. I suggest that the authors revise this section of the text to include mention of these other networks as well.

  We appreciate the referee's comment. Indeed, we overlooked the necessity of mentioning these observation networks. To complete the discussion of vertically resolving networks, the relevant paragraph has been revised and additional information has been incorporated:
  "Similarly, ground-based Fourier Transform InfraRed (FTIR) spectrometers, which from part of the Network for the Detection of Atmospheric Composition Change (NDACC), are capable of retrieving vertically resolved mixing ratios of a range of atmospheric constituents. However, the vertical resolution of these profiles is constrained by their dependence on a priori information, and the network's spatial coverage remains sparse (De Mazière et al., 2018; García et al., 2021). Multi-axis differential optical absorption spectroscopy (MAX-DOAS) is also capable of retrieving trace gas and aerosol vertical profiles (Tirpitz et al., 2021)."

- P3, L69: "The aim is to investigate the ability of the 4D-var to adjust". Minor grammatical comment, but I suggest inserting "system" here so it reads "The aim is to investigate the ability of the 4D-var system to adjust".

  We are grateful to the referee for identifying this error. The recommended correction has been implemented.

- General comment on the text on P10 and Table 3 – In the text here, you provide some absolute differences (in ppbv) while later on you start providing relative differences (in %) for the biases, however in Table 3 you only provide absolute biases (in ppbv). I find this makes it harder to follow. For clarity, I suggest either providing both simultaneously in the text, or choose one and be consistent.

We thank the referee for pointing out this important remark. To ensure clarity, we have now incorporated both the percentage and the absolute values into the text.

- P12, L226: "The 4d-var data assimilation..." the 'D' should be capitalized here.

  Affirmative. This typo has been corrected.

- P13, L267-268: "...especially for emissions that are emitted at high altitudes, such as power plants and industries.", I am not really sure the top of a smoke-stack is considered "high altitude" as implied here. Maybe change the text to "higher altitudes", or something similar.

  Thank you for the recommendation. In accordance with your suggestion, the text has been revised to read 'higher altitudes'.

- P14, Figure 4 – In the titles of the left column figures "Nox" should be changed to "NO_x". Also, "Septembre" → "September" in the top titles of the middle and right-most column of figures. I also suggest using a consistent range for the color-bars at least for the middle and right-most column of figures. As it currently stands, upon a quick look it appears as though the road-transport sector has the largest difference, but this is only because the color bar scale is much lower.

  We would like to thank the referee this pertinent observation. The proposed amendments have been incorporated into the revised Figure 4.

- P15, Figure 5 – In the color-bar label, "molec cm-2"should be written as "molec. cm^{-2}".

  We have corrected the label to "molec. cm^{-2}" as suggested.

- P15, L285-286, the improvement in the Pearson correlation coefficient of 0.15 here does not coincide with the values listed in Table 4. Only a difference of 0.04 is listed in the table.

  The difference of 0.04 reflects the improvement in the Pearson correlation coefficient during the data assimilation period. In this context, we provide the statistics for the 24-hour forecast, which are listed in parentheses in the Table 3. Thanks to this and a later comment, we have amended Table 4's caption to provide greater clarity on this matter.

- P15, L288-291; the authors state here that a "remarkable improvement" in the O_3 concentrations is seen for the beginning of the day on September 23, but neglect to discuss the fact that the optimized simulation agrees more poorly with the observations for the remainder of the day, particularly at the end of the day. I think this should be highlighted more in the text here, as the overall simulation has not necessarily been "improved" with respect to the ground-based observations, only for a short period in which there was originally a large discrepancy. This is discussed to some extent later on P20, but should probably be mentioned here first.

  We agree with the referee's remark. We have amended the wording in aforementioned line to read : "A remarkable improvement in the O_3 concentration is noticed within the initial seven hours of the day, while a deterioration is observed between 16:00 and 24:00." As we provide a more detailed explanation later on, we believe this adequately addresses the referee's request for clarification.

- P16, Table 4; I think it should be made clearer (in the table caption), what values are being provided in the brackets. It does not seem to be mentioned anywhere, are these standard deviations/standard errors?

  Thank you for pointing out the lack of clarity in this matter. The values in parentheses represent the statistical data pertaining to the 24-hour forecast. The caption has been revised in order to provide greater clarity. Please find the updated version here: "Statistical comparison of ground-based observations and model outputs (REF: reference run, DA: assimilation run) for O_3, NO, and NO_2 during the assimilation window and, in parentheses, the 24-hour forecast on 22-23 September 2021. The Bias and RMSE are in μgm^{-3}."

- General comment on P15-16; similar to my earlier comment about the absolute/relative differences when discussing the comparisons, in the text here on these two pages the authors make statements such as "the assimilation of drone observations results in a strong reduction of the bias by 87% and the RMSE by 20% with an amelioration in the Pearson correlation of 0.15". I believe important context is lost by the authors only providing the differences, but not mentioning in the text what the initial and final values were. For example, if the original bias was 2 ug m^{-3} and the new bias is 1 ug m^{-3}, then you can say there was a 50% decrease in the bias, but this would not be as significant as it sounds. I think the text should be amended here to include this.

  We thank the referee for these pertinent remarks, which have been addressed in the revised manuscript. In the revised version, we now provide both percentages the corresponding absolute values.

- P17, Figure 6 – O3 and NO2 in the subplot titles should be subscripted as O_3 and NO_2.

  O_3 and NO_2 are now correctly subscripted in the Figure 6.

- P19, Figure 7 – This figure looks a bit too simplistic and like it was made hastily, and I do not think is of publication quality, particularly in relation to the other figures in the manuscript. "O3"in the title should also be subscripted, this also applies to Figure A2, A3, and A4.

  Indeed. The figures have been revised in order to enhance their visual quality and ensure alignment with the established publication standards. All notation errors have now been corrected across Figures 7, A2, A3, and A4.

- P20, L338-339; the text currently reads "Moreover, the assimilation process allows to obtain optimised emissions rates", this should read "allows *one* to obtain..."or "the assimilation process provides optimized emission rates".

  We thank the referee for pointing out this wording mistake. The phrase has been corrected as follows: "Moreover, the assimilation process provides optimised emissions rates for each day."

- P20, L348-350 "This is supported by the findings of Wu et al. (2022), affirming that observation at high altitudes can be advantageous for optimising emissions under suitable wind 350 conditions", what exactly is supported by the findings of Wu et al. (2022) here? The authors need to elaborate on this point.

  Thanks for identifying the lack of information. For better clarity, this sentence has been edited as follows: "In a recent study by Wu et al. (2022), it was demonstrated that for high-altitude observations, the efficiency of emission rate optimization is conditioned by favorable wind conditions and strong vertical diffusion."

- P20, L361: "Secondly, Some...", "some" should not be capitalized here.

  This typo has been corrected in the revised manuscript.

- P20, L362: "(Fig.6, Fig.A3, and Fig.A4)" should have spaces between "Fig." and the figure numbers.

  Affirmative. This mistake has been corrected in the revised manuscript.

- P21, Figure 8: "NO2" should be subscripted in the y-axis label of the bottom left figure panel.

  Sure. This has been corrected in the revised manuscript.

- P22: The section heading for 'conclusion' should be capitalized.

  Thanks. This typo has been corrected in the revised manuscript.

- P22, L395; Minor comment but Observing System Simulation Experiments (OSSEs) is a more commonly used term for this, not OSE. I am also not quite sure what the authors mean here by "...to assess the advantages and limitations of integrating drone observations into CTMs through the application of a variational data assimilation technique", is that not exactly what was done in this work?

  We thank the referee for highlighting this important point. The sentence was indeed poorly formulated. Our future work aims to conduct OSSEs to assess the added value of assimilating drone observations into operational systems, alongside conventional observations type. The primary objective is to determine whether integrating drone data with conventional observations can further improve the assessment of local emissions and refine the vertical distribution of pollutants in the model. This was not feasible in the current work due to limited observation data from the campaign. Positive results from OSSEs might advocate for a wider integration of drone observations in operational forecasting systems.
  The term OSSEs has been corrected and the related phrase has been modified as follows: "From a future perspective, a valuable extension of this work will be to conduct Observing System Simulation Experiments (OSSEs) to evaluate the added value of integrating drone-based observations into the air quality forecasting system, compared to conventional observations such as ground-based measurements and satellite data."

Responses to Anonymous Referee#3

This manuscript describes a modeling study in which the emissions of mainly nitrogen oxides are adjusted by assimilating drone-based observations of ozone and nitrogen monoxide. This is a interesting topic, since understanding how to use drone observations in chemical data assimilation could open up new pathways to quantify local emission sources. Overall, I find the study technically solid, and as it stands, I have no strong objections to its publication. However, the paper would become more interesting if a more stronger analysis of the mechanisms taking place in the inversion could be included. The current discussion is detailed enough, but not very conclusive.

We appreciate the review comments Referee#3 and their recognition of the significance of our study. During this review process, we have incorporated several amendments to the manuscript with the objective of strengthening the discussion on the potential of drone observations for data assimilation in the context of trace gas forecasts. Please find our responses to the more detailed points below.

For example, why did the model fail to capture the O_3 levels on the night of 23rd September, why did the assimilation system resolve the discrepancy by a large emission adjustment (as opposed to initial values), and to what extent are the optimized emissions supported or explained by independent data? At minimum, it could be useful to examine the spatial scale of the patterns that the assimilation aims to capture. Figs. A3 and A4 show quite high heterogeneity in both O_3 and NO_2 on the night of 23rd, which suggests that the assimilation result might be sensitive to model resolution. On the other hand, if the patterns are region-wide, they might be rather driven by a model rather than emission inventory bias.

We agree with the referee that these are valid and important questions given the direct comparison of model results and observations. Regarding the first question, we agree that the model does not well predict the O_3 and NO_2 concentrations during the night of 23rd September. These results can be attributed to both the model and the emissions. On the model side, one potential factor we suspect is the representation of the planetary boundary layer height, which could significantly affect nighttime predictions. The issue is known and currently under investigation. Since it is independent of the assimilation process, we considered a comprehensive evaluation beyond the scope of our manuscript. However we approach the emissions that are one of the largest sources of uncertainty in model predictions. The objective of utilising the 4D-Var method is to improve the representation of emissions, and thereby enhancing the overall model forecast. In our results, we observed a limited and minor improvement during the night, which we attribute to the fixed temporal profile used for the emissions optimisation. This topic has been discussed in detail in section 4.3.1. Another reason can be the lack of observability – preferable information content – provided by the limited number of drone profiles. In response to the second question, the greater impact of optimizing emissions compared to initial values is primarily due to the inversion process being more effective under favorable/higher wind conditions. Finally, it is challenging to directly validate the optimized emissions with observational data. However, the validation of the analysis against ground-based observations suggests that the optimized emissions lead to a superior analysis, showing that the optimization is moving in the desired direction. This indicates that the uncertainty in the a priori emission values has been reduced, despite the limitation imposed by the short data assimilation window.

Related to the model resolution, it might be worth noting that the representation errors of a single drone profile might not be independent of each other. This would violate the assumption of a diagonal R matrix and effectively result in overstating the accuracy of the assimilated measurements.

We thank the referee for pointing out this important remark. We agree that both measurement and representation errors in the observations are likely correlated. However, the unavailability of detailed error estimates makes it challenging to accurately quantify these correlations. Therefore, in our simulations, we assume that the errors are uncorrelated. To compensate for the absence of correlation, we have increased the error variances to their maximum reasonable values, thereby ensuring that the observations are given appropriate weight in the assimilation.
The topic of the representation error is of significant interest and worthy of discussion. The work of Janjić et al. (2018) provides a detailed examination of the various definitions of this error and current research is exploring an methodology for assessing this error in EURAD-IM, with the view to conducting similar analysis.

Minor Comments:

- L120: Does the block-wise structure of K relate to chemical or spatial correlations, or both?

  The matrix K accounts for both chemical and spatial correlations. To enhance the clarity of the sentence, we have modified it in the manuscript as follows: "The matrix **K** is defined as block diagonal, with non-zero entries for correlations between species and near-by emissions. The variance and correlation values are provided in Paschalidi (2015)."

- Section 3.1: What was the vertical resolution of the profiles? How many data points did the assimilated profiles consist of?

  The vertical resolution of these profiles is approximately 10 meters, with in total 254 data points assimilated on 22 September 2021 and 257 on 23 September 2021 for both O_3 and NO. We added this information to the manuscript.

- L190: Would the discussion of background error correlation lengths fit better to Section 2.2?

  We see the referee's point of discussing the background error correlation length in the section, where we introduce the data assimilation method. However, since we treat the correlation length differently for different analysis setups, we decided and now reassure that it is better to be discussed in the simulations setup section, as it is for example model grid resolution dependent. Therefore, we prefer to leave the statement where it is placed now.

- L265: How much is 16 Mgd^{-1} relative to the total daily emission over the region in Fig. 1?

  The emission of 16 Mgd^{-1} represents approximately 3.46% of the total daily NO_x emissions in the analyzed region, where the total daily NO_x emission is about 462 Mgd^{-1}.. This information has been added into the manuscript.

- L363: "constrains the optimisation to more flexible adjustments..." - not sure if I understand.

  Thank you for pointing out this confusing statement. We rephrased it to: "It is assumed that the temporal emission profile is more certain than the emission strength. Deriving e.g. hourly emission factors instead would allow for more flexible adjustments of the emissions, which would be beneficial for the nowadays strongly regulated emission sources, such as the power production (dependent on the availability of renewable energy)."

- Fig. 1: N and E seem to be swapped in the tick labels

  We appreciate the attention of the referee. This mistake has been corrected in the revised manuscript.

- Fig. 8: The vertically oriented row labels (DA_23SEP – REF_23SEP etc.) are small and difficult to read; please consider showing at least the chemical species more clearly.

  We thank the referee for this recommendation. The figure has been improved in the revised manuscript.

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