To Referee #1

We are grateful to you for your interest and comments on this manuscript.

Please find the point-by-point answers to your comments in green.

Please note that the present version of this manuscript reports mixing ratios in ppt and no longer in ppb to show the significant decimal digits.

Please note that, following the referee's comments, we have reformulated/refined the statistical analysis described in Section 2.3, updated the results and discussion accordingly, and have finally merged Section 3.1 with Section 3.2 (now Section 3.1).

The manuscript "Long-term analysis of atmospheric propane over Southern Europe based on observations conducted at the WMO-GAW station of Monte Cimone" presents a 13-year time series of propane, measured at Monte Cimone, Italy. Notably, all the data are available and can be immediately accessed, and I applaud the authors for keeping the monitoring station active and making the data openly accessible.

I really enjoyed reading this manuscript and consider it a nice piece of work. While some readers might find the detailed analysis tedious, I believe this work is essential for detailed investigations of our atmosphere.

Nevertheless, I have a few minor comments for the authors to consider.

Major comment:

This preprint is under review for Atmospheric Chemistry and Physics (ACP). According to the journal's scope, "Articles should have important and clearly argued implications for our understanding of the state and behavior of the atmosphere [...]". I consider this work valuable and important, as it confirms much of the previous work on C3H8. However, not all readers may share this view. Please, consider presenting this manuscript as a "Measurement Report".

We agree about this point with the reviewer#1 since the paper mostly focuses on discussing new observations rather than delving into understanding the behaviour of the atmosphere. We will reconsider the manuscript as “Measurement Report”.

Minor comments:

Line 6): JFJ is used but has not been defined. Please check the consistency with the following sentence.

Thank you for pointing this out. We revised the sentence as follows: “..Jungfraujoch (JFJ, Switzerland)..”

Line 132): I suggest removing "slight" as the trend is anyhow statistically significant.

Following your suggestion, the sentence was rephrased as follows:

"Between 2011 and 2023, C_3H_8 mixing ratios exhibited a significant decrease of .."

Line 290): It would be beneficial to briefly compare other emissions databases (e.g., CAMS or CEDS) for the three countries investigated. Similarly, it would be helpful to again cite a few articles mentioned in the introduction that show the underestimation of emissions has been known since 2018 (Dalsoren et al., 2018).

Following your suggestions, we added the following to Section 3.6 Inversion estimates:

"Comparisons of our estimates of propane emissions with the estimates from the Copernicus Atmosphere Monitoring Service (CAMS) confirm increases of 141 and 113% over the CAMS global anthropogenic emissions inventory (Soulie et al. 2024) for Italy and France, respectively."

"Previous studies related the observed low estimations of propane emissions at the global (Etiope and Ciccioli, 2009) and regional (Bourtsoukidis et al., 2020) scales to missing geologic sources, and fossil fuel emissions in the USA (Tzompa-Sosa et al., 2019) or the northern hemisphere (Dalsøren et al. 2018).In this context, previous studies (Dalsøren et al., 2018; Tzompa-Sosa et al., 2019; Rowlinson et al., 2024; Ge et al., 2024) concluded that current emission inventories underestimate anthropogenic fossil fuel emissions of C_3H_8."

Line 295): There is a missing period (.) before "Similar".

Thank you for pointing this out. We revised this typo accordingly.

Figure 10): I recommend using "CH" for Switzerland (instead of "CHE").

Figure 10 denotes the countries with the three-letter country codes (e.g., CHE for Switzerland, ITA for Italy, and FRA for France)) defined in ISO 3166-1 alpha-3 by the International Organization for Standardization (ISO). We preferred the three-letter country codes to the two-letter country codes (e.g., CH for Switzerland, IT for Italy, and FR for France) because the ISO 3166-1 alpha-3 allows an easier association between the codes and the country names than the two-letter country codes.

References

Etiope, G. and Ciccioli, P.: Earth’s degassing: a missing ethane and propane source, Science, 323, 478–478, 200

Bourtsoukidis, E., Pozzer, A., Sattler, T., Matthaios, V. N., Ernle, L., Edtbauer, A., Fischer, H., Könemann, T., Osipov, S., Paris, J.-D., et al.: The Red Sea Deep Water is a potent source of atmospheric ethane and propane, Nature Communications, 11, 447, 2020.

Dalsøren, S. B., Myhre, G., Hodnebrog, Ø., Myhre, C. L., Stohl, A., Pisso, I., Schwietzke, S., Höglund-Isaksson, L., Helmig, D., Reimann, S., et al.: Discrepancy between simulated and observed ethane and propane levels explained by underestimated fossil emissions, Nature Geoscience, 11, 178–184, 2018.

Ge, Y., Solberg, S., Heal, M. R., Reimann, S., van Caspel, W., Hellack, B., Salameh, T., and Simpson, D.: Evaluation of modelled versus observed non-methane volatile organic compounds at European Monitoring and Evaluation Programme sites in Europe, Atmospheric Chemistry and Physics, 24, 7699–7729, 2024.

Tzompa-Sosa, Z. A., Henderson, B., Keller, C. A., Travis, K., Mahieu, E., Franco, B., Estes, M., Helmig, D., Fried, A., Richter, D., et al.: Atmospheric implications of large C2-C5 alkane emissions from the US Oil and Gas Industry, Journal of Geophysical Research: Atmospheres, 124, 1148–1169, 2019.

Rowlinson, M. J., Evans, M. J., Carpenter, L. J., Read, K. A., Punjabi, S., Adedeji, A., Fakes, L., Lewis, A., Richmond, B., Passant, N., Murrells, T., Henderson, B., Bates, K. H., and Helmig, D.: Revising VOC emissions speciation improves the simulation of global background ethane and propane, Atmospheric Chemistry and Physics, 24, 8317–8342, https://doi.org/10.5194/acp-24-8317-2024, 2024.

Soulie, A., Granier, C., Darras, S., Zilbermann, N., Doumbia, T. et al., Global anthropogenic emissions (CAMS-GLOB-ANT) for the Copernicus Atmosphere Monitoring Service simulations of air quality forecasts and reanalyses, EARTH SYSTEM SCIENCE DATA, 16, 5, 2024, p. 2261-2279, COPERNICUS GESELLSCHAFT MBH, https://data.europa.eu/doi/10.5194/essd-16-2261-2024, JRC133834.

To Referee #2

We are grateful to you for your interest and comments on this manuscript.

Please find the point-by-point answers to your comments in green.

Please note that the present version of this manuscript reports mixing ratios in ppt and no longer in ppb to show the significant decimal digits.

Please note that, following the referee's comments, we have reformulated/refined the statistical analysis described in Section 2.3, updated the results and discussion accordingly, and have finally merged Section 3.1 with Section 3.2 (now Section 3.1).

General Comments:

The manuscript presents a comprehensive long-term analysis of atmospheric propane at the Monte Cimone WMO-GAW station, based on an impressive 2011–2023 dataset that is also publicly available. The authors deserve congratulations for their sustained monitoring efforts and for making the data accessible to the scientific community. However, I agree with the second reviewer that the manuscript sometimes reads like a "measurements report" rather than a cohesive scientific analysis. The links between sections are often missing, which gives the impression of a series of independent analyses rather than a unified study. Below, I suggest ways to strengthen the narrative and improve the scientific rigor of the paper.

Major Comments:

1. Linking sections 3.1 and 3.2: seasonal trends and long-term trends

The authors mention an increase in the amplitude of the seasonal cycle (Section 3.2). To deepen the analysis, it would be valuable to disaggregate the long-term trend (Section 3.1) by season. Is the increase in amplitude due to higher winter concentrations or lower summer concentrations?

Suggestion: Perform a seasonal trend analysis and discuss how it relates to the findings in Section 3.2. This would provide insight into whether the observed changes are driven by emission variations, meteorological factors, or chemical processes (e.g., OH variability).

Following your suggestion and the questions raised by reviewer#3, we refined the statistical analysis to derive the trend parameters by using daily means mixing ratios instead of raw data to better match the skills of the CCGCRV model developed by NOAA. Moreover, we conducted a sensitivity test running a Monte-Carlo bootstrap data selection. The description of methods was updated in Section 2.3. Finally, the description and the discussion of the results have been merged in a single section (i.e., Section 3.1, present version of the manuscript). Figure 4 was revised by showing trend lines of C_3H_8 seasonal amplitudes and yearly peaks and troughs.

2. Contextualizing trends (2018–2020 and 2021–2023) with extreme values (Section 3.5)

The trends discussed in Section 3.1 (2018–2020 and 2021–2023) should be analyzed in the context of the numerous extreme values reported in 2018 and 2023 (Section 3.5). Could these extreme values be skewing the trends? A discussion on the robustness of the trends in light of these extremes would strengthen the analysis.

Thank you for pointing this out.

As a result of the revised statistical analysis, the two trends previously identified are now less “evident”, leading to a slightly different discussion of results. We removed the comments on short term variation of C3H8 on the trend curve between 2018 and 2023.

To better explain the statistical analysis performed, we added to Section 2.3 the following: “The seasonal cycle and trend calculations were done on bootstrap-resampled subsets consisting of 90% of the dataset for 50 iterations.”

“The long term trend, the trends on the long-term trend curve (Section 3.1), and trend of seasonal amplitudes (Fig. 4) are estimated with the Theil-sein regression (scikit-learn Python package (Pedregosa et al., 2011)) and Mann-Kendall (pyMannKendall Python package (Hussain and Mahmud, 2019)) testing for significance.”

3. COVID-19 impact analysis: accounting for meteorological variability

The analysis of COVID-19 impacts lacks a discussion of meteorological influences (e.g., Petit et al., 2021). Changes in air mass origin or meteorological conditions (e.g., PBL height, wind patterns) could have influenced propane concentrations independently of emission changes.
Suggestion: Include a comparative analysis of meteorological conditions across years (e.g., using ERA5 reanalysis data or backward trajectories for multiple years). The 2022 trajectory analysis is interesting but insufficient for drawing conclusions about interannual variability.

Thank you for pointing this out.

This topic was deeply investigated in our previous paper on anomaly during COVID period, with the conclusion that “The analysis of local wind speed patterns, specific humidity and O_3 monthly diel cycles did not indicate substantial changes in the vertical transport associated with the thermal circulation system in the area. The low O_3 values that characterized MAM and JJA 2020 cannot be attributed to differences in the synoptic-scale circulation compared with the previous five years. “

We added the following to Section 3.4:

"Cristofanelli et al. 2021 observed no substantial variations in the synoptic-scale circulation and vertical transport related to the thermal circulation system at CMN in summer 2020 compared to the previous five years."

4. Potential OH Bias in the Inversion (Section 3.6)

The inversion results (Section 3.6) may be sensitive to the OH concentrations used. Could a potential overestimation of OH lead to an underestimation of propane emissions?

Suggestion: Discuss the uncertainties in OH concentrations and whether there is a seasonal bias in the OH fields used. If possible, perform a sensitivity analysis to assess how variations in OH impact the inversion results.

Thank you for your suggestions on the OH bias.

We performed a sensitivity inversion excluding the OH loss term completely. Posterior emissions were reduced to less than half of the reference values (Figure D1 included in the appendix), demonstrating the significant influence of OH chemical loss on the emission estimates. Regarding the concern about OH overestimation leading to emission underestimation, we would like to note that the opposite relationship holds: an overestimation of OH would require the inversion to increase emissions to compensate for excessive simulated loss. The sensitivity analysis confirms that our inversion framework responds appropriately to changes in the chemical loss term. While OH fields are subject to uncertainties of approximately 10–20% (Naik et al., 2013; Wolfe et al., 2019) in current chemical transport models (GEOS-CHEM considered in our study), any systematic bias in OH would propagate linearly into the emission estimates.

5. Methods section: clarifications needed

- Quality Control (QC) Procedures

The QC procedure for propane measurements is not detailed. To ensure confidence in the trend analysis, the authors should provide:

- A table summarizing blank and standard measurements over time.
- Relative standard deviations for standards.
- Any observed instrumental drift and how it was corrected.

Details on the analytical protocol are illustrated in LoVullo et al. 2016. We rephrased and expanded the text in Section 2.1 to include the relevant information:

"The time frame of this study covered the period from January 2011 to December 2023. In situ online measurements were performed at the World Meteorological Organization's

Global Atmosphere Watch (WMO/GAW) global station on CMN (44°12′N, 10°42′ E, 2165 m above sea level). Monitoring of non-methane volatile organic compounds (NMVOC) performed at CMN station is generally representative of emissions occurring in the European continent as reported by Lo Vullo et al. (2016). Measurements of C3H8 were performed with a gas chromatograph-mass spectrometer (GC–MS Agilent 6820 + Agilent 5975C) operating in Selected Ion Monitoring (SIM) mode, preceded by an online sample enrichment using a preconcentration system Unity-2-AirServer-2 (Markes International), following a method described in Maione et al. (2013) and Lo Vullo et al. (2016), according to ACTRIS standard operating procedure (SOP) (https://www.actris.eu/sites/default/files/Documents/ACTRIS-2/Deliverables/WP3_D3.17_M42.pdf) and audited under the GAW programme of the WMO by the World Calibration Center for volatile organic compounds in 2018. Real ambient air samples are collected every second hour, alternated with whole-air calibration mixture (working standard) to correct for any short term instrumental drift, resulting in 12 real air measurements per day. Each month the working standard is calibrated against a certified "30-compounds Ozone precursor mixture" at 500 ppt level in nitrogen from the National Physical Laboratory (NPL-UK). System blanks are evaluated on a weekly basis, with concentrations changing over time but limited well below 15 ppt, with results adjusted accordingly. Total uncertainty for each measurement is calculated as the error propagation of i) the reproducibility of the repeated working standard runs on the same day, ii) the detection limit, and iii) the scale propagation error (derived by the regular NPL/quaternary standard check, additional details in appendix F1). Final QC of the dataset is checked yearly by an external reviewer as part of the ACTRIS-EBAS SOP procedures before submission for data release to the EBAS repository. In addition, C3H8 observations for the year 2022 from JFJ WMO-GAW global station have been used for atmospheric inversion modeling as described in Section 2.5. C3H8 measurements at JFJ are carried out within the framework of ACTRIS activities, following the same analytical protocol for CMN but with different instrumentation (i.e. the Medusa-AGAGE setup (Miller et al., 2008; Prinn et al., 2018)). Measurements of CO and CH4 were performed at CMN according to the methods described in Section A."

In Appendix A, we added the following:

"Carbon monoxide, and methane datasets:

From 2011 to 2015 CH4 and CO observations were carried out using a GC-FID system, designed according to AGAGE protocol for the GC-MD setup. The calibration of the GC-FID system was performed with a set of 6 NOAA calibration mixtures with concentrations spanning the range of the ambient air values. Quality assurance/quality control procedures were done regularly in compliance with AGAGE protocols by means of GCWerks (SIO) software. From 2015 (for CH4) and from 2018 (for CO), observations have been carried out by using Cavity Ring Down Spectroscopy (CRDS) instruments. From 2018, CO and CH4 observations have been performed at CMN in the framework of ICOS. Within ICOS, observations are carried out in a standardized way for measurement set-up, used materials, quality assurance strategy and data creation workflow. Hazan et al. (2016) and Yver-Kwok et al. (2021) provided a detailed description of the quality assurance programme for ICOS measurements. CH4 observations from 2015 to 2017 have been carried out by CAMM - Italian Air Force in the GAW/WMO framework. The quality assurance programme has been designed based on the recommendation provided by GAW/WMO (https://library.wmo.int/idurl/4/69756). In particular, a multipoint calibration is performed every three months against three laboratory standards provided by NOAA whose mole fractions exceed the range for ambient air. Calibration data are post-processed and calibration coefficients are derived through linear regression and used to correct the in situ air measurements. A specific water vapour correction determined during a system and performance audit by the WMO/GAW "World Calibration Center for Surface Ozone, Carbon Monoxide, Methane, Carbon Dioxide and Nitrous Oxide" was applied to the data (Zellweger et al., 2018). CAMM operators manually screened the data to remove anomalous events related to instrumental/sampling issues or local emissions."

In Appendix F, we added a table reporting the precision of the calibration tank used and a plot with the time dependent variation of the daily working standard precision used to evaluate precision of each measurement of propane.

- JFJ Dataset: The Jungfraujoch (JFJ) dataset is used in the inversion but is not described. Clarify the QC procedures, measurement frequency, and any averaging methods applied to this dataset.

The two stations follow the same ACTRIS standard operating procedures and calibration scheme, even though JFJ was -and still is- using a different instrumentation, for which proper citation is reported.

Following your suggestion, in Section 2.1 the text has been modified as follows:

"In addition, C3H8 observations from Jungfraujock (JFJ, Switzerland) WMO-GAW global station have been used for atmospheric inversion modeling as described in Section 2.5. C3H8 measurements are carried out within the framework of ACTRIS activities, following the same analytical protocol of CMN but with different instrumentation (i.e. the Medusa-AGAGE setup (Miller et al., 2008; Prinn et al., 2018)). "

.

- Averaging 2-Hourly C_3H_8 Data to 3-Hourly Bins

The authors mention that 2-hourly C_3H_8 measurements were averaged to 3-hourly resolution for the inversion. However, the methodology is unclear:

- How were measurements assigned to 3-hourly bins (e.g., 00:00–03:00, 03:00–06:00)?

- How were bins with only one measurement (e.g., 04:00 in the 03:00–06:00 window) handled?

Suggestion: Clarify the averaging method and justify its suitability for the inversion.

Following your suggestion, we added the following description in Section 2.5:

“For the inversion, observations from CMN and JFJ were aggregated to a 3-hourly temporal resolution. Fixed 3-hour time bins (e.g., 00:00–03:00, 03:00–06:00) were defined, and all observations within each bin were averaged and assigned to the bin start time. If a bin contained only a single observation, that observation was assigned to the corresponding bin start time.”

Minor Comments:

Abstract: Define "JFJ" (Jungfraujoch) at first mention.

Thank you for pointing this out. We revised the sentence as follows: “..Jungfraujoch (JFJ, Switzerland)..”

Line 143: Define "BB" (biomass burning).

Following your suggestions, we revised the typos related to JFJ and BB.

Figure 2:

- Clarify the names and locations of the sites (ALT, BRW, KUM, etc.) in the caption.
- Explain the two y-axes (the red axis for LEF is confusing as LEF is shown in a different color).
- Clarify what is meant by "CMN trend curve (2 years) or (1 year)".

Following your suggestions, caption of Figure 2 was revised as follows:

” ….. Shaded lines show 95% confidence interval for CMN trend curves with a long-term cutoff value of 1 or 2 years. ALT – Alert (Nunavut, Canada); BRW – Utqiaġvik, formerly Barrow (Alaska, USA); KUM – Cape Kumukahi (Hawaii, USA); SUM – Summit (Greenland, Denmark); MHD – Mace Head (Ireland); LEF – Park Falls (Wisconsin, USA).”

Moreover, shaded lines were added for showing the 95% confidence interval for CMN trend curves calculated on randomly sampled subsets consisting of 90% of the dataset for 50 iterations.

Figure 5:

- Define the seasonal periods (e.g., winter = December–February) in the text.

- Specify whether UTC or local time is used (in other figures as well).

  Annual and seasonal diurnal variations of propane mixing ratio measured at Monte Cimone (Italy) between 2011 and 2023.

Following your suggestions:

- caption of Figure 5 was revised as follows: "….Spring – from March to May; autumn – from September to November; winter – from December to February; summer – from June to August."

- UTC was added to the x-axis title of figures 1, 2, 5, and C2-C5.

Figure 9: Remove the connecting lines during data gaps to avoid misleading interpretations.

Following your suggestions, connecting lines were removed in data gaps.

Conclusion:

This study provides a valuable long-term dataset and important insights into atmospheric propane trends. By strengthening the links between sections, addressing potential biases, and providing more methodological details, the authors can significantly enhance the impact and clarity of their work. I look forward to seeing a revised version that incorporates these suggestions.

References:

Petit, J.-E., Dupont, J.-C., Favez, O., Gros, V., Zhang, Y., Sciare, J., Simon, L., Truong, F., Bonnaire, N., Amodeo, T., Vautard, R., and Haeffelin, M.: Response of atmospheric composition to COVID-19 lockdown measures during spring in the Paris region (France), Atmospheric Chem. Phys., 21, 17167–17183, https://doi.org/10.5194/acp-21-17167-2021, 2021.

References

Cristofanelli, P., Arduni, J., Serva, F., Calzolari, F., Bonasoni, P., Busetto, M., ... & Putero, D. (2021). Negative ozone anomalies at a high mountain site in northern Italy during 2020: a possible role of COVID-19 lockdowns?. *Environmental Research Letters*, *16*(7), 074029. DOI 10.1088/1748-9326/ac0b6a

Hazan, L., Tarniewicz, J., Ramonet, M., Laurent, O., and Abbaris, A.: Automatic processing of atmospheric CO_2 and CH_4 mole fractions at the ICOS Atmosphere Thematic Centre, Atmos. Meas. Tech., 9, 4719–4736, https://doi.org/10.5194/amt-9-4719-2016, 2016.

Hussain, M. and Mahmud, I.: pyMannKendall: a Python package for nonparametric Mann-Kendall family of trend tests, Journal of Open Source Software, 4, 1556, https://doi.org/10.21105/joss.01556, 2019.

Miller, B. R., Weiss, R. F., Salameh, P. K., Tanhua, T., Greally, B. R., Mühle, J., and Simmonds, P. G.: Medusa: A sample preconcentration and GC/MS detector system for in situ measurements of atmospheric trace halocarbons, hydrocarbons, and sulfur compounds, 80, 1536–1545,505 https://doi.org/10.1021/ac702084k, 2008.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P., Bergmann, D., Cameron-Smith, P., et al.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry andClimate Model Intercomparison Project (ACCMIP), Atmospheric Chemistry and Physics, 13, 5277–5298, 2013.

Pedregosa, F., Varoquaux, G., Gramfort, A., Michel, V., Thirion, B., Grisel, O., Blondel, M., Prettenhofer, P., Weiss, R., Dubourg, V., Vanderplas, J., Passos, A., Cournapeau, D., Brucher, M., Perrot, M., and Duchesnay, ˙E.: Scikit-learn: Machine learning in Python, Journal of Machine Learning Research, 12, 2825–2830, 2011

Prinn, R. G., Weiss, R. F., Arduini, J., Arnold, T., DeWitt, H. L., Fraser, P. J., Ganesan, A. L., Gasore, J., Harth, C. M., Hermansen, O., Kim, J., Krummel, P. B., Li, S., Loh, Z. M., Lunder, C. R., Maione, M., Manning, A. J., Miller, B. R., Mitrevski, B., Mühle, J., O'Doherty, S., Park, S., Reimann, S., Rigby, M., Saito, T., Salameh, P. K., Schmidt, R., Simmonds, P. G., Steele, L. P., Vollmer, M. K., Wang, R. H., Yao, B., Yokouchi, Y., Young, D., and Zhou, L.: History of chemically and radiatively important atmospheric gases from the Advanced Global Atmospheric Gases Experiment (AGAGE), Earth Syst. Sci. Data, 10, 985–1018, https://doi.org/10.5194/essd-10-985-2018, 2018

Wolfe, G. M., Nicely, J. M., St. Clair, J. M., Hanisco, T. F., Liao, J., Oman, L. D., Brune, W. B., Miller, D., Thames, A., González Abad, G., et al.: Mapping hydroxyl variability throughout the global remote troposphere via synthesis of airborne and satellite formaldehyde observations, Proceedings of the National Academy of Sciences, 116, 11 171–11 180, 2019.

Yver-Kwok, C., Philippon, C., Bergamaschi, P., Biermann, T., Calzolari, F., Chen, H., Conil, S., Cristofanelli, P., Delmotte, M., Hatakka, J., Heliasz, M., Hermansen, O., Komínková, K., Kubistin, D., Kumps, N., Laurent, O., Laurila, T., Lehner, I., Levula, J., Lindauer, M., Lopez, M., Mammarella, I., Manca, G., Marklund, P., Metzger, J.-M., Mölder, M., Platt, S. M., Ramonet, M., Rivier, L., Scheeren, B., Sha, M. K., Smith, P., Steinbacher, M., Vítková, G., and Wyss, S.: Evaluation and optimization of ICOS atmosphere station data as part of the labeling process, Atmos. Meas. Tech., 14, 89–116, https://doi.org/10.5194/amt-14-89-2021, 2021.

Zellweger, C., Steinbacher, M., and Buchmann, B.: System and Performance Audit of Surface Ozone, Carbon Monoxide,Methane and Carbon Dioxide at the Global GAW Monte Cimone, Italy, June 2018, WCC-Empa Report 18/1, Dübendorf, Switzerland, 2018.

To Referee #3

We are grateful to you for your interest and comments on this manuscript.

Please find the point-by-point answers to your comments in green.

Please note that the present version of this manuscript reports mixing ratios in ppt and no longer in ppb to show the significant decimal digits.

Please note that, following the referee's comments, we have reformulated/refined the statistical analysis described in Section 2.3, updated the results and discussion accordingly, and have finally merged Section 3.1 with Section 3.2 (now Section 3.1).

(https://doi.org/10.5194/egusphere-2025-5098-RC3).

This manuscript presents an analysis of long-term atmospheric propane observations over Southern Europe, based on measurements conducted at the Monte Cimone GAW station (Italy). Long-term, high-quality VOC datasets are relatively scarce, and the availability of such measurements is valuable and worthy of publication. The manuscript focuses exclusively on propane measurements from 2011 to 2023.

That said, most of the analyses presented here are not particularly novel. Similar datasets, methodologies, and interpretations have been reported in numerous previous studies. While the data themselves are of interest, the scientific advancement beyond the existing literature is limited.

I am surprised that the authors did not incorporate measurements of other VOCs from the same instrument at Monte Cimone into the analysis. Including additional compounds would have enabled a more comprehensive and nuanced investigation of propane atmospheric behavior, sources, and chemical processing.

In my view, the comparison between bottom-up emission inventories and top-down estimates (Figure 11) represents the most valuable contribution of this study. I strongly encourage the authors to expand this part of the manuscript, providing a clearer explanation of the methodology and how the results were derived. This should also be accompanied by a more critical discussion of the uncertainties associated with the inversion results. While I am not a specialist in statistical methods, it seems that additional uncertainty analysis—potentially including a Monte Carlo evaluation—would substantially strengthen this section. Furthermore, the apparent underestimation of propane emissions in southern Italy by the inventory warrants deeper investigation and discussion of potential sources. And again, incorporating other VOCs as emission tracers might provide insight into emission categories.

The manuscript summarizes - a recap- the state of investigation of this specific compound in response to some recent papers (Helmig et al. 2016, Angot et al. 2021) presenting anomalies on propane and ethane at the global scale. The aim of the present manuscript is to contextualise the discussion to the European domain based on the long-term dataset available from our station. The suggestions from the reviewer will be taken into account for future research based on additional VOCs measurements at the CMN station.

We thank the reviewer for the valuable suggestions on strengthening the inversion analysis. Following these recommendations, we performed an additional 20-member Monte Carlo ensemble simulation to robustly quantify posterior uncertainties, now reported as the 90% confidence interval. We have also expanded the methodological description to include details on the cost function, solver, and uncertainty estimation approach. Furthermore, we conducted a sensitivity analysis excluding the OH loss term to evaluate the influence of OH-related biases on the emission estimates, and added a discussion on potential sources contributing to the underestimation of propane emissions in the prior inventory.

Specific Comments

- **Line 4:** The term "vary" is likely inappropriate in this context. It would be more accurate to state that atmospheric propane mole fractions did not exhibit a statistically significant trend over the observational period.

  To avoid confusion, we revised this sentence as follows:

  " Over the study period, C_3H_8 background mixing ratios exhibited a significant decrease of -3.8 [-5; -2.3; 95% confidence interval] ppt per year."

- **Line 5:** I recommend reporting changes in seasonal amplitude in absolute units (ppb) rather than percent, as the reference value for the percentage change is unclear.

  Following your suggestions, we revised the sentence by reporting changes in seasonal amplitude in ppt per year.

- **Line 6:** Jungfraujoch (JFJ) is mentioned without prior introduction. Please introduce the site and its relevance before referencing it.

  Thank you for pointing this out. We revised the sentence as follows: "..Jungfraujoch (JFJ, Switzerland).."

- **Lines 15–16:** The meaning of the reported 99.3% value is unclear. Please clarify what this percentage represents.

This sentence was rephrased as follows:

"According to Hodnebrog et al. (2018), the specific radiative forcing for the indirect effects of C3H8 is about 99% of its total specific radiative forcing through the interactions with O3 formation and CH4 removal."

- **Line 57:** Please specify the calibration frequency in hours or days; the term "regularly" is too vague. In addition, further details on propane quantification are needed. Was propane quantified using selected ion integration? Was the mass spectrometer operated in scan or SIM mode? How were blanks determined and treated? Was a single-point calibration used, or were dilution curves and system linearity assessed? How was instrumental drift corrected? Given that multiple VOCs were likely quantified during each run, please explain why the analysis focuses exclusively on propane. Were the propane measurements audited by the World Calibration Center for VOCs?

  Details on the analytical protocol are illustrated in LoVullo et al. 2016. We rephrased /expanded the text in Section 2.1 to include the relevant information:

"The time frame of this study covered the period from January 2011 to December 2023. In situ online measurements were performed at the World Meteorological Organization's Global Atmosphere Watch (WMO/GAW) global station on CMN (44°12′N, 10°42′ E, 2165 m above sea level). Monitoring of non-methane volatile organic compounds (NMVOC) performed at CMN station is generally representative of emissions occurring in the European continent as reported by Lo Vullo et al. (2016). Measurements of C3H8 were performed with a gas chromatograph-mass spectrometer (GC–MS Agilent 6820 + Agilent 5975C) operating in Selected Ion Monitoring (SIM) mode, preceded by an online sample enrichment using a preconcentration system Unity-2-AirServer-2 (Markes International), following a method described in Maione et al. (2013) and Lo Vullo et al. (2016), according to ACTRIS standard operating procedure (SOP) (https://www.actris.eu/sites/default/files/Documents/ACTRIS-2/Deliverables/WP3_D3.17_M42.pdf) and audited under the GAW programme of the WMO by the World Calibration Center for volatile organic compounds in 2018. Real ambient air samples are collected every second hour, intertwined by whole-air calibration mixture (working standard) to correct for any short term instrumental drift, resulting in 12 real air measurements per day. Each month the working standard is calibrated against a certified "30-compounds Ozone precursor mixture" at 500 ppt level in nitrogen from the National Physical Laboratory (NPL-UK). System blanks are evaluated on a weekly basis, with concentrations changing over time but limited well below 15 ppt, with results adjusted accordingly. Total uncertainty for each measurement is calculated as the error propagation of i) the reproducibility of the repeated working standard runs on the same day, ii) the detection limit, and iii) the scale propagation error (derived by the regular NPL/quaternary standard check). Final QC of the dataset is checked yearly by an external reviewer as part of the ACTRIS-EBAS SOP procedures before submission for data release to the EBAS repository. In addition, C3H8 observations for the year 2022 from JFJ WMO-GAW global station have been used for atmospheric inversion modeling as described in Section 2.5. C3H8 measurements at JFJ are carried out within the framework of ACTRIS activities, following the same analytical protocol for CMN but with different instrumentation (i.e. the Medusa-AGAGE setup (Miller et al., 2008; Prinn et al., 2018)). Measurements of CO and CH4 were performed at CMN according to the methods described in Section A."

- **Line 105:** If JFJ data are included in the analysis, the measurement techniques and quality assurance procedures for that site should be described with the same level of detail as those for Monte Cimone

  The two stations follow the same ACTRIS standard operating procedures and calibration scheme, even though JFJ was -and still is- using a different instrumentation, for which proper citation is reported.

  Following your suggestion, in Section 2.1, the text has been modified as follows:

  "In addition, C3H8 observations from Jungfraujoch (JFJ, Switzerland) WMO-GAW global station have been used for atmospheric inversion modeling as described in Section 2.5. C3H8 measurements are carried out within the framework of ACTRIS activities, following the same analytical protocol of CMN but with different instrumentation (i.e. the Medusa-AGAGE setup (Miller et al., 2008; Prinn et al., 2018)). "

- **Line 133:** The text first states that no clear trend is present but then claims a statistically significant decrease at the 95% confidence level. Please reconcile this apparent contradiction and use appropriate statistical terminology. It is also unclear how the reported slope values were derived. The curve shown in Figure 1 does not appear to represent a linear trend. Please explain how these slope values were calculated and how they represent changes over the full data period.

  A significant decrease of -3.8 [-5; -2.3; 95% confidence interval] ppt per year refers to the mean decrease [95% confidence interval] per year calculated with Theil-Sein regression and Mann-Kendall testing for significance on the trend curves with bootstrap sampled subsets consisting of 90% of the dataset for 50 iterations.

  We added a description of the method followed for calculating the slope and significance in Section 2.3:

  "The seasonal cycle and trend calculations were done on bootstrap sampled subsets consisting of 90% of the dataset for 50 iterations…..

  The long term trend, the trends on the long-term trend curve (Section 3.1), and trend of seasonal amplitude (Fig. 4) are estimated with the Theil-Sein regression (scikit-learn Python package \citep{pedregosa2011}) and Mann-Kendall (pyMannKendall Python package \citep{hussain2019}) testing for significance."

In Section 3.1, the sentence was rephrased as follows:

"Between 2011 and 2023, C_3H_8 mixing ratios exhibited a significant decrease of -3.8 [-5; -2.3; 95% confidence interval] ppt per year (solid green line in Fig. 1, and Fig. 2). "

- **Line 134:** The unit should be ppb yr^{-1}, not ppb.

  Thank you for pointing this out. The unit is now ppt per year.

- **Lines 134–146:** Differences observed over relatively short time windows (e.g., 2–3 years) may be strongly influenced by interannual meteorological variability. Such variations should not be readily interpreted as emission changes without longer observational periods (at least five years) or appropriate meteorological normalization.

  We agree with the reviewer about the importance of assessing the potential influence of meteorological variability when discussing short-term changes in mixing ratios. As a result of the revised statistical analysis, the two trends previously identified are now less "evident", leading to a slightly different discussion of results. Therefore, we removed the comments on short term variation of C_3H_8 on the trend curve between 2018 and 2023.

- **Lines 148–166:** The seasonal behavior of propane has been well documented and explained in the existing literature, including studies cited here. This section offers limited new interpretation and could be substantially shortened.

  Following your suggestion, this section was shortened from 18 to 11 lines.

- **Line 167:** Meteorological influences should be considered more explicitly in this context.

  We agree that year-to-year variation in transport may influence the variability in the concentration of atmospheric tracers. However, no evidence of trends or changes in transport patterns was reported by previous studies on the characterization of long term measurements of O_3 (Cristofanelli et al. 2021a), NO_x (Cristofanelli et al. 2021b), and mineral dust (Vogel et al. 2025) at CMN.

  In Section 3.1, we added the following:

"Previous studies (Cristofanelli et al. 2021a; Cristofanelli et al. 2021b; Vogel et al. 2025) about long term measurements of atmospheric components at CMN did not observe any trends or changes in the transport patterns."

- **Line 171:** Please provide additional detail on how seasonal minima and maxima were determined, as this procedure is more complex than implied.

  Is the reported change in seasonal amplitude statistically significant? An annual change of 0.16% over 12 years amounts to less than 2% in total, which is relatively small. It is unclear whether this signal exceeds measurement noise or reflects meaningful changes in chemistry or emissions.

  Thank you for this comment. Following your comment, we realized that the CCGCRV curve-fitting method developed at NOAA for discrete and discontinuous flask samples performs better on daily mean mixing ratios rather than hourly measurements. Therefore, the present version of the manuscript presents the curve fitting as performed on the daily mean mixing ratios, filtered for days with at least eight out of 12 measurements.

  Moreover, calculations were done on bootstrap-sampled subsets consisting of 90% of the daily mean values instead of 80% of the observations because the CCGCRV curve-fitting method is not meant to handle abundant data gaps. The new analysis resulted in a more robust description of the long-term decreasing trend and a substantial steady, slightly decreasing trend of the seasonal amplitude,

  We revised the 2.3 Statistical analysis section by adding the following description of the method performed for evaluating seasonal minima and maxima:

  "...The smoothed curve is obtained by combining the harmonic components and the residuals from the filter with a short-term cut-off value of 60 days. Following previous studies (Angot et al., 2021; Dlugokencky et al., 1997), for each year, the amplitude (peak-to-trough) of the C3H8 seasonal cycle was calculated as the difference between the maximum and minimum values of the smoothed curve (red line in Fig. 2) according to the analysis of the first derivative of the smooth curve. The seasonal cycle and trend calculations were done on bootstrap-resampled subsets consisting of 90% of the daily mean values for 50 iterations."

- **Line 181:** According to the recent literature, NO_x emissions in the Northern Hemisphere have generally been decreasing over the past decade rather than increasing. Please revisit this statement.

  We agree with the referee about this point. We reorganized the discussion about the seasonal amplitude trend and removed the sentence with a mention of NO_x

emissions that was wrongly related to the Northern Hemisphere instead of East-Asia (China).

- **Line 203:** Please clearly define how the seasons are classified.

  Following your suggestion, we revised the sentence as follows: "... in winter (i.e., December, January, and February) and at 15:00 UTC in the other seasons (i.e., Spring- March, April, and May; Summer- June, July, and August; Autumn- September, October, and November).

- **Line 230:** Several studies report increased ozone levels during the COVID-19 pandemic. Please discuss your findings in the context of this broader literature.

  We agree with the reviewer about the fact that the COVID lockdown was often associated with increases in O3 ambient levels.

  In this context, several studies (e.g., Sicard et al. 2020, Collivignarelli et al. 2020, Grange et al. 2021) about changes in air quality in European cities during the COVID lockdown explained the increases in ozone ambient levels with decreases in NOx emissions.

  On the contrary, several studies (e.g., Bouarar et al. 2021, Steinbrecht et al. 2021, Chang et al. 2022) observed negative O_3 anomalies in the free troposphere.

  Putero et al. (2023) observed persistent negative anomalies for O_3mixing ratios in four European high-elevation sites in 2020 in both spring (March–May) and summer (June–August), except for April.

  High-elevation monitoring sites such as Monte Cimone are representative of boundary layer or background atmospheric conditions depending on whether the measurements are performed below or above the boundary layer height.

  Therefore, different changes in ozone ambient levels occurred during the COVID lockdown depending on the area (e.g., urban or remote sites) and the elevation (high elevation or not) of the measurement stations.

- **Lines 230–243:** This section appears highly speculative and inconclusive. I recommend removing it or substantially shortening it.

  Following your suggestion, lines 229-243 were rephrased as follows:

  "With regard to the lower values in summer 2020, our results align with previous findings that suggested a link between lower emissions of O3 precursors and decreases in O3 mixing ratios measured at CMN (Cristofanelli et al., 2021a) and several high-elevation sites in Europe (Putero et al., 2023). The main drivers of variations in atmospheric C3H8 mixing ratios are changes in emission sources, atmospheric chemistry, and transport. Cristofanelli et al. (2021a) observed no substantial variations in the synoptic-scale circulation and vertical transport related to the thermal circulation system at CMN in summer 2020 compared to the previous five years. In this context, the CAMS (Soulie et al., 2023) and EDGARv8.1 (Crippa et al., 2024) inventories estimated that the 2020 anthropogenic emissions of propane from Europe were 91% and 99.3%, respectively, of the emissions averaged over the period 2011-2019. In addition, comparisons of aircraft campaign measurements across Europe showed lower OH mixing ratios in the free troposphere during the COVID-19 lockdown compared to previous campaigns (Nussbaumer et al., 2022). The expected longer residence time due to the lower reaction rate between C3H8 and OH is therefore at odds with the lower C3H8 concentrations recorded at CMN in 2020, which is likely attributable to reduced emissions."

- **Line 248:** Please be more precise in this description. If a 96% cutoff is applied, it is unclear how 11–15% high-occurrence values can simultaneously be reported. These metrics appear to use different reference definitions, which need to be clearly explained.

  The events with high C3H8 mixing ratios were calculated based on a threshold of 96% of the seasonal daily mean mixing ratios. Between 11 and 15% of the events with high C3H8 mixing ratios occurred in 2013, 2015, 2018, and 2023.

- **Figure 6:** I recommend adding bold numerical labels for the median values, as these are used for the statistical comparisons.

  Following your suggestion, we added bold numerical labels for the median values.

- **Line 295:** A period is missing after the word "located".

  Thank you, this typo was revised.

- **Line 310:** Deriving a linear trend over a period that includes a pronounced anomalous minimum during the COVID-19 pandemic is questionable. This limitation should be explicitly acknowledged and discussed.

  Thank you for pointing this out.

  Following your suggestion, the trend analysis was also performed on two sub-datasets (i.e., pre-COVID from 2011 to 2019, and post-COVID from 2022 to 2023), to avoid any influences on the trend analysis resulting from the drastic changes in activities and emissions during the COVID-19 pandemic, the associated lockdowns and recovery phase between 2020 to 2021.

  The trend in pre-COVID time (2011-2019) was comparable to the long-term trend of the full study period, with a significant decrease of -2.6 [- 4.7; -0.6] ppt per year.

Post-COVID (2022-2023) showed a significant decrease of -10 [-14.1; -6.7] ppt per year.

However, the relatively short time span of two years of the post-COVID time does not assure the fact that the observed trend is the result of local fluctuations. Therefore, there is the need for analysis of a longer propane dataset to provide a more robust evaluation of the trend following the COVID-19 pandemic.

We added the following to the Section 3.1:

"The trend analysis was also performed on two sub-datasets (i.e., pre-COVID from 2011 to 2019, and post-COVID from 2022 to 2023), to avoid any influences on the trend analysis resulting from the drastic changes in activities and emissions during the COVID-19 pandemic, the associated lockdowns and recovery phase between 2020 and 2021. Both sub-datasets confirmed a significant decreasing trend. Specifically, the pre-COVID time trend of -2.6 [- 4.7; -0.6] ppt per year was comparable to the long-term trend of the full study period."

References

Angot, H., Davel, C., Wiedinmyer, C., Pétron, G., Chopra, J., Hueber, J., Blanchard, B., Bourgeois, I., Vimont, I., Montzka, S. A., et al.: Temporary pause in the growth of atmospheric ethane and propane in 2015-2018, Atmospheric Chemistry and Physics, 21, 15 153–15 170, 2021. https://doi.org/10.5194/acp-21-15153-2021

Bouarar, I., Gaubert, B., Brasseur, G. P., et al.. (2021). Ozone anomalies in the free troposphere during the COVID-19 pandemic. *Geophysical Research Letters*, *48*(16), e2021GL094204. https://doi.org/10.1029/2021GL094204

Chang, K.-L., Cooper, O. R., Gaudel, A., Allaart, M., Ancellet, G., Clark, H., Godin-Beekmann, S., Leblanc, T., Van Malderen, R., Nédélec, P., Petropavlovskikh, I., Steinbrecht, W., Stübi, R., Tarasick, D. W., and Torres, C.: Impact of the COVID-19 economic downturn on tropospheric ozone trends: an uncertainty weighted data synthesis for quantifying regional anomalies above Western North America and Europe, AGU Advances, 3, e2021AV000542, https://doi.org/10.1029/2021AV000542, 2022.

Collivignarelli, M. C., Abbà, A., Bertanza, G., Pedrazzani, R., Ricciardi, P., & Miino, M. C. (2020). Lockdown for CoViD-2019 in Milan: What are the effects on air quality?. *Science of the total environment*, *732*, 139280. https://doi.org/10.1016/j.scitotenv.2020.139280

Cristofanelli, P., Arduni, J., Serva, F., Calzolari, F., Bonasoni, P., Busetto, M., Maione, M., Sprenger, M., Trisolino, P., and Putero, D.: Negative ozone anomalies at a high mountain site in northern Italy during 2020: a possible role of COVID-19 lockdowns?, Environmental Research Letters, 16, 074 029, 2021a. DOI 10.1088/1748-9326/ac0b6a

Cristofanelli, P., Gutiérrez, I., Adame, J., Bonasoni, P., Busetto, M., Calzolari, F., Putero, D., and Roccato, F.: Interannual and seasonal variability of NOx observed at the Mt. Cimone GAW/WMO global station (2165 m asl, Italy), Atmospheric Environment, 249, 118 245, 2021b https://doi.org/10.1016/j.atmosenv.2021.118245

Grange, S. K., Lee, J. D., Drysdale, W. S., Lewis, A. C., Hueglin, C., Emmenegger, L., and Carslaw, D. C.: COVID-19 lockdowns highlight a risk of increasing ozone pollution in European urban areas, Atmos. Chem. Phys., 21, 4169–4185, https://doi.org/10.5194/acp-21-4169-2021, 2021

Helmig, D., Rossabi, S., Hueber, J., Tans, P., Montzka, S. A., Masarie, K., Thoning, K., Plass-Duelmer, C., Claude, A., Carpenter, L. J., et al.: Reversal of global atmospheric ethane and propane trends largely due to US oil and natural gas production, Nature Geoscience, 9,445 490–495, 2016. https://doi.org/10.1038/ngeo2721

Hussain et al., (2019). pyMannKendall: a python package for non parametric Mann Kendall family of trend tests.. Journal of Open Source Software, 4(39), 1556. https://doi.org/10.21105/joss.01556

Lo Vullo, E., Furlani, F., Arduini, J., Giostra, U., Graziosi, F., Cristofanelli, P., Williams, M. L., and Maione, M.: Anthropogenic non-methane volatile hydrocarbons at Mt. Cimone (2165 m asl, Italy): Impact of sources and transport on atmospheric composition, Atmospheric Environment, 140, 395–403, 2016. https://doi.org/10.1016/j.atmosenv.2016.05.060

Liu, G., Shen, L., Ciais, P., Lin, X., Hauglustaine, D., Lan, X., Turner, A. J., Xi, Y., Zhu, Y., and Peng, S.: Trends in the Seasonal Amplitude of Atmospheric Methane, Nature, 641, 660–665, https://doi.org/10.1038/s41586-025-08900-8, 2025

Miller, B. R., Weiss, R. F., Salameh, P. K., Tanhua, T., Greally, B. R., Mühle, J., and Simmonds, P. G.: Medusa: A sample preconcentration and GC/MS detector system for in situ measurements of atmospheric trace halocarbons, hydrocarbons, and sulfur compounds, Anal.Chem., 80, 1536–1545, https://doi.org/10.1021/ac702084k, 2008.

Nussbaumer, C. M., Pozzer, A., Tadic, I., Röder, L., Obersteiner, F., Harder, H., Lelieveld, J., and Fischer, H.: Tropospheric ozone production and chemical regime analysis during the COVID-19 lockdown over Europe, Atmospheric Chemistry and Physics, 22, 6151–6165, 2022. https://doi.org/10.5194/acp-22-6151-2022

Pedregosa, F., Varoquaux, G., Gramfort, A., Michel, V., Thirion, B., Grisel, O., ... & Duchesnay, É. (2011). Scikit-learn: Machine learning in Python. *the Journal of machine Learning research*, *12*, 2825-2830.

Prinn, R. G., Weiss, R. F., Arduini, J., Arnold, T., DeWitt, H. L., Fraser, P. J., Ganesan, A. L., Gasore, J., Harth, C. M., Hermansen, O., Kim, J., Krummel, P. B., Li, S., Loh, Z. M., Lunder, C. R., Maione, M., Manning, A. J., Miller, B. R., Mitrevski, B., Mühle, J., O'Doherty, S.,Park, S., Reimann, S., Rigby, M., Saito, T., Salameh, P. K., Schmidt, R., Simmonds, P. G., Steele, L. P., Vollmer, M. K., Wang, R. H., Yao, B., Yokouchi, Y., Young, D., and Zhou, L.: History of chemically and radiatively important atmospheric gases from the Advanced Global Atmospheric Gases Experiment (AGAGE), Earth Syst. Sci. Data, 10, 985–1018, https://doi.org/10.5194/essd-10-985-2018, 201

Putero, D., Cristofanelli, P., Chang, K.-L., Dufour, G., Beachley, G., Couret, C., Effertz, P., Jaffe, D. A., Kubistin, D., Lynch, J., Petropavlovskikh, I., Puchalski, M., Sharac, T., Sive, B. C., Steinbacher, M., Torres, C., and Cooper, O. R.: Fingerprints of the COVID-19 economic downturn and recovery on ozone anomalies at high-elevation sites in North America and western Europe, Atmos. Chem. Phys., 23, 15693–15709, https://doi.org/10.5194/acp-23-15693-2023, 2023. https://doi.org/10.5194/acp-23-15693-2023

Sicard, P., De Marco, A., Agathokleous, E., Feng, Z., Xu, X., et al. (2020). Amplified ozone pollution in cities during the COVID-19 lockdown. *Science of the Total Environment*, *735*, 139542. https://doi.org/10.1016/j.scitotenv.2020.139542

Steinbrecht, W., Kubistin, D., Plass-Dülmer, C., Davies, J., et al. COVID-19 crisis reduces free tropospheric ozone across the Northern Hemisphere, Geophys. Res. Lett., 48, e2020GL091987, https://doi.org/10.1029/2020GL091987, 2021

Vogel, F., Putero, D., Bonasoni, P., Cristofanelli, P., Zanatta, M., and Marinoni, A.: Saharan dust transport event characterization in the Mediterranean atmosphere using 21 years of in-situ observations, Atmos. Chem. Phys., 25, 15453–15468, https://doi.org/10.5194/acp-25-15453-2025, 2025.