

*We thank the reviewers for agreeing to review our work and for providing many valuable comments, suggestions and corrections, helping to improve our manuscript.*

*The responses are organized as follows: the reviewer's comments are in black, our answers are in italics and blue, and the changes proposed for the revised manuscript are in italics and red.*

## **REFeree #1**

### **General comments**

As from the title, the paper aims to investigate a long-term time series of ozone data measured in the 2012-2023 period at the OPE station in France, shedding light on the causes of the signals detected. To this aim, the ozone dataset is integrated with data on pollutants, methane, meteorological variables, and radiotracers. Although the topic is surely interesting, the work remains superficial and lacks some logic even in the structure but also deeply reflected in the methodological approach.

*We thoroughly revised the manuscript to improve the presentation and clarity of the approach we followed.*

Starting with the introduction, the authors present a long list of papers and processes responsible of different things (ozone variability, radiotracers, ...) but at the end the reader remains unsatisfied with the missing gaps and main motivation of the research paper. The presentation and discussion of results present significant drawbacks, which can be summarized as follows:

The results are often compared with other papers, but the results are seldomly discussed in view of what they could imply in terms of the processes responsible or what is already known on the topic; *Further discussions of the results have been added to the results section. Spectral Analysis were also added.*

STT events and their connections with both ozone and radiotracers variability at midlatitude high-altitude stations have been the subject of several research studies, which are never cited here; *Several citations were added:*

*Bonasoni, P., Evangelisti, F., Bonafe, U., Ravegnani, F., Calzolari, F., Stohl, A., Tositti, L., Tubertini, O., and Colombo, T.: Stratospheric ozone intrusion episodes recorded at Mt. Cimone during the VOTALP project: case studies, Atmos. Environ., 34, 1355–1365, doi:10.1016/S1352-2310(99)00280-0, 2000.*

*Cristofanelli, P., P. Bonasoni, L. Tositti, U. Bonafè, F. Calzolari, F. Evangelisti, S. Sandrini, and A. Stohl: A 6-year analysis of stratospheric intrusions and their influence on ozone at Mt. Cimone (2165 m above sea level). J. Geophys. Res., 111, D03306, doi:10.1029/2005JD006553, 2006.*

*Gaffney JS, Marley NA, Cunningham MM, Kotamarthi VR.: Beryllium-7 measurements in the Houston and Phoenix urban areas: an estimation of upper atmospheric ozone contributions. J Air Waste Manag Assoc. 55(8):1228-35. doi: 10.1080/10473289.2005.10464707. PMID: 16187592. 2005.*

*Kalabokas, P., Jensen, N. R., Roveri, M., Hjorth, J., Eremenko, M., Cuesta, J., Dufour, G., Foret, G., and Beekmann, M.: A study of the influence of tropospheric subsidence on spring and summer surface ozone concentrations at the JRC Ispra station in northern Italy, Atmos. Chem. Phys., 20, 1861–1885, <https://doi.org/10.5194/acp-20-1861-2020>, 2020.*

*Lee H.N., L. Tositti, X. D. Zheng and P. Bonasoni: Analyses and comparisons of <sup>7</sup>Be, <sup>210</sup>Pb and activity ratio <sup>7</sup>Be/<sup>210</sup>Pb with ozone observations at two GAW stations from high mountains, J. Geophys. Res, 112, D05303, doi:10.1029/2006JD007421, 2007.*

*Liu, H., D. J. Jacob, J. E. Dibb, A. M. Fiore, and R. M. Yantosca: Constraints on the sources of tropospheric ozone from  $^{210}\text{Pb}$ - $^7\text{Be}$ - $\text{O}_3$  correlations, J. Geophys. Res., 109, D07306, doi:10.1029/2003JD003988, 2004.*

*Trickl, T., Couret, C., Ries, L., and Vogelmann, H.: Zugspitze ozone 1970–2020: the role of stratosphere–troposphere transport, Atmos. Chem. Phys., 23, 8403–8427, <https://doi.org/10.5194/acp-23-8403-2023>, 2023.*

Circulation at both local and synoptic scales, and not only vertical transport, may have significant implications on ozone variability: not only this is not studied here, but also this is never cited as a potential additional process responsible for at least some of the observed variabilities;

*These processes have been discussed in the revised manuscript.*

I have not seen any results (e.g., spectral analysis) clearly pointing out the main periods appearing in the different time series;

*Spectral analysis has been added to the revised manuscript.*

At the same time, the correlation analysis presented remains superficial and does not consider the presence of (potential) spurious correlations (the presence of correlation in principle only highlights the presence of similar time patterns in the time series);

*Correlation analysis has its own pros and cons. The revised manuscript uses Correlation analysis carefully along with other analysis tools (such as wavelet coherence, linear regression, classification) to build confidence in the associations between parameters.*

There is no effort in highlighting limitations (e.g., the use of very coarse resolution data for boundary layer height) and in drawing general conclusions from the study.

*The manuscript has been revised to highlight limitations of the study and clarify the main conclusions.*

In particular, while reading the results section, I had the feeling that the authors were trying to put together all the analyses done, but the logic behind the structure and the connection between the different analyses remain very unclear and for sure not deep. To conclude, and summarizing, the results section seems an extensive list of different results without any efforts to put them together or investigate in depth what they are telling us.

*The manuscript has been revised to improve the presentation of our approach and to clarify our main results.*

## **Specific comments**

### **Page 1**

Lines 16-25: It is not clear how meteorological parameters were used: does transport (not only in the vertical, but for instance, mountain-valley breeze regimes) play a role in the  $\text{O}_3$  patterns? The relation with temperature and radiation seems straightforward and not particularly new. It is also not clear if the diurnal and seasonal pattern was studied (e.g.,  $\text{O}_3$  higher in the afternoon,  $\text{O}_3$  higher in spring, ...). Finally, there is no quantification apart from the overall trend of the baseline, while the influence of specific processes remains unquantified at least in the abstract.

*The abstract has been rewritten.*

Lines 19-21: Are anomalies and peaks utilized as synonyms? Anomalies in principle can be positive or negative, but here it seems implicit that all are positive.

*The abstract has been rewritten.*

Lines 18-19: not clear why afternoon and nighttime, and not diurnal and nighttime.

*The abstract has been rewritten.*

Line 29: Ozone in the stratosphere is beneficial because its absorption of UV radiation (and which type?) shields life on Earth's SURFACE from it. This has also a climate (radiative forcing) effect. Please explain better.

*The first paragraph of the introduction has been rewritten to account for the referee's remarks.*

*"In the stratosphere, ozone is formed by recombination of di-oxygen and an oxygen atom, resulting from the O<sub>2</sub> photolysis. Stratospheric O<sub>3</sub> absorbs UV-B and UV-C radiation, warms the surrounding air and protects life on the Earth's surface. Conversely, it is a burden in the lower troposphere where its oxidative properties have significant health (e.g., Fleming et al., 2018; Achebak et al., 2024) and ecosystem impacts (e.g., Krupa and Manning, 1988; Lefohn et al., 1988; Cheesman et al., 2024). Tropospheric ozone also contributes about 10-15% to the climate forcing induced by greenhouse gases (e.g., IPCC, 2021)."*

Lines 35-38: Please explain better that only with NO<sub>x</sub> the net cycle would be null (production = destruction).

*To answer the referee's remark we included a description of the NO<sub>x</sub> limited and VOC limited regimes in the text.*

Lines 40-41: In principle, this is very rare as normally the two layers are essentially isolated because of stability conditions. Downward transport from the stratosphere require specific conditions which are not mentioned here. Please explain better (with references).

*A paragraph and references have been added to the processes leading to stratospheric air incursion into the troposphere.*

*"The transport of air masses from the stratosphere to the troposphere (STT) is also a significant contributor to surface ozone concentration as the stratosphere is the main ozone production zone (e.g., Stohl et al., 2003; Cristofanelli et al., 2006; Archibald et al., 2020). This intermittent transport occurs during specific atmospheric synoptic situations such as tropopause folding behind cold fronts and gravity wave breaking favoring stratospheric air intrusions (e.g., Holton et al., 1995; Stohl et al., 2003)."*

## **Page 2**

Lines 15-17: Please try to describe these.

*This part has been significantly modified and descriptions have been added.*

## **Page 3**

Lines 35: Which type of aerosols, and in particular, of which dimensions? This has important implications.

*We have now indicated that these radionuclides are mainly attached to submicron size particles*

## **Page 4**

Line 14: What do you mean by "sometimes"?

*This part of the text has been moved and "are sometimes revealed" changed in "can be revealed by"*

Line 38: I assume "temperature" and "pressure" are atmospheric temperature and barometric pressure.

*We have changed "temperature" in "atmospheric temperature" and "pressure" in "barometric pressure" as requested*

## **Page 5**

Lines 3-5: not clear if these results are from elsewhere (in which case, a reference is missing), or if they are from this specific paper, and in this case what conclusion the reader should draw (e.g., type of climate? Type of circulation?). A reference to the mean wind intensity is also missing.

*The text indicates that the average values reported are the result of our processing of meteorological data for the study period. We have also indicated that the "climatological" averages are typical of a*

*continental climate in Western Europe. We do not provide average wind intensities, as such averages have no particular meaning.*

Lines 5-7: The use of such coarse resolution data may represent a clear drawback: are you sure that the reanalysis with its coarse resolution can catch the complex terrain features in the study area, clearly impacting on the boundary layer height variability?

*The terrain is not particularly complex: it is relatively flat with smooth hills with altitudes ranging between 200 m and 400 m over a radius of at least 100 km. However, we have added a paragraph describing the reliability and limitation of the estimation of the BLH by ERA 5 data.*

*We modified section 2 accordingly.*

Lines 10-12: The change of instrumentation may have several drawbacks or at least effects on the signal, with discontinuities and inhomogeneity appearing. Please discuss.

*A discussion on this topic was added in section 2.3.*

Lines 15-17: Not clear how the different instruments are used. If utilized as for NO<sub>x</sub>, meaning changing instruments, this may have implications as discussed above.

*A discussion on this topic was added in section 2.3.*

Lines 19-23: not clear, please explain better.

*A better explanation was given at the end of section 2.3.*

Lines 25-26: This can be omitted as already described thoroughly in the Introduction.

*This sentence has been suppressed.*

Lines 25-32: This is not really relevant, the important thing is to describe how they are measured in your case/dataset.

*We maintain this statement because it explains why a weekly resolution is necessary to be able to measure the radionuclides.*

## **Page 6**

Line 5: It is not only the height of the mixing layer that is relevant, but the type (convective, residual, ...) that matters.

*We have now stated that the nighttime mixing layer is mainly a residual mixing layer and afternoon mixing layer is mainly a convective mixing layer.*

Lines 13-14: What do you mean by “standard parameters”? Maybe they result from some spectral analysis and not extendable to other datasets where the time variability is different.

*The parameters set are the parameters used in the fitting model CCGCRV from NOAA. The parameters are described three lines after. They are well defined for the analysis of multi annual time series of atmospheric compounds mixing ratio measurements (<https://gml.noaa.gov/ccgg/mb/rl/rlfit/rlfit.html>)*

Lines 25-27: I think it should be mentioned and described better that the purpose is to investigate the processes behind O<sub>3</sub> anomalies, using proxies. And then describe the types of proxies used and the processes they are representative of.

*This has been done: “Because we have a special interest in identifying the processes behind surface O<sub>3</sub> anomalies, spectral analyzes were performed as well on the residuals to check for their respective properties in the frequency domain and to look for their coherence level.”*

*And*

*“We use NO<sub>x</sub>, CO and CH<sub>4</sub> as indicators of chemical precursors, various meteorological parameters (solar radiation, temperature, precipitation rate and duration, relative and specific humidity, BLH) as indicator of the weather/climate conditions and <sup>7</sup>Be and <sup>22</sup>Na as indicators of STT contributions.”*

Lines 31-40 and page 7, lines 1-2: I do not understand the reason to compare with other stations and in particular for different time periods. Please explain better.

*The comparison has been recentered on rural background European stations. This is mentioned to underline the OPE coherence with other rural background European environment.*

#### Page 7

Figure 1, caption: Could you please explain the meaning of the shaded area?

*The figure has been redrawn, and the legend has been completed.*

Lines 7-16: Could you then discuss how these results help in discovering processes related to O<sub>3</sub> diurnal variability in the different seasons?

*This was mentioned p 8 lines 15-19: “The observed broad spring/summer maximum is commonly interpreted as a combination of contributions from large scale transport of enhanced background with the addition of large photochemical production from regional emissions.”*

*This has been slightly modified to underline that this statement applies to our data set.*

#### Page 8

Lines 4-18: I am not sure where these results come from (i.e., to which figure can they be related?).

*These results come from our data analysis. We have rewritten the sentence to make it clearer.*

*“At the OPE station, our data show that the mean surface ozone concentration was  $63.6 \mu\text{g.m}^{-3}$  (31.9 ppbv) over the 2012-2023 period. Ozone concentrations at OPE are in line with those observed at low-altitude European stations: ...”*

*We do not produce figures for this, as we only indicate mean, minimum/maximum concentrations for different months.*

#### Page 9

Lines 9-10; What do you mean by “more important”?

*It means “higher”. The sentence has been rewritten: “The afternoon O<sub>3</sub> concentrations were higher than the nighttime concentrations, with an average difference of  $20 \mu\text{g.m}^{-3}$ . In general, maximum surface O<sub>3</sub> concentrations are observed when the boundary layer is developed (during the afternoon and in summer) suggesting that the contribution of the photo-chemical production of O<sub>3</sub> dominates the dilution due to the increase of the boundary layer height and the vertical turbulent mixing”.*

Lines 12-24: Can you please discuss which are the processes responsible for such seasonal patterns?

*A discussion was added in the revised version:*

*“At background locations such as OPE, the late winter maxima of CO and NO<sub>x</sub> are mostly explained by the long-range transport of enriched air masses coming from urban and industrial areas with large anthropogenic emissions in winter combined with reduced mixing and longer compounds lifetimes (due to reduced destruction of CO by OH radicals and reduced photo-chemical reactions of NO<sub>x</sub>) leading to greater accumulation. The summer CO and NO<sub>x</sub> minima are mostly related to a combination of the reduction of the emissions in upstream regions with an increased mixing associated with deeper boundary layer and shorter compounds lifetimes (associated with the maximum occurrence of OH radicals and enhanced photo-chemical reactions of NO<sub>x</sub>) (Novelli et al., 1998; Gilge et al., 2010; Cristofanelli et al., 2021).”*

Lines 30-32: There are other studies presenting seasonal patterns of <sup>7</sup>Be at midlatitude, and also high-altitude stations. Please revise.

*We have revised accordingly and added two new references: Gerasopoulos et al., 2001 and Brattich et al., 2017.*

Figure 2, 3 and 4: the figures present not only average values but also uncertainty bars which are not discussed.

*These are not uncertainty bars but the 95% confidence interval (figure 2) and the standard deviation (figure 3) of the monthly means. This is now clearly indicated in the captions of these figures.*

#### **Page 11**

Line 1: I cannot see this ratio plotted in Figure 3.

*The  $^7\text{Be}/^{22}\text{Na}$  ratio was removed in this revised version because we only had a limited use of this parameter in our main analysis.*

Lines 3-12: The seasonal variability of STT processes have been already thoroughly analysed at midlatitude high-altitude stations in several papers, also, but not restricted to, applications of cosmogenic radiotracers. Please discuss your findings in view of literature.

*We have further discussed the seasonal variations of  $^7\text{Be}$  and  $^{40}\text{K}$  but the discussion about the  $^7\text{Be}/^{22}\text{Na}$  ratio was removed.*

Lines 11-12: Not clear, please revise.

*The sentence has been removed.*

Lines 18-19 and page 12, lines 1-2: How does this can be related to other results? If it cannot be linked to other results, then it is only another part of the study site characterization in Section 2.

*This paragraph has been modified according to the comment.*

#### **Page 12**

Lines 4-8: It is not clear how this relates to Figures and results presented in other sections.

*The number of the relevant figures has been added.*

Line 10: Where are these thresholds coming from?

*We have modified the text to explain the thresholds:*

*“These STT events thresholds were defined as 70th percentile of the weekly  $^7\text{Be}$  and  $^{22}\text{Na}$  activity concentration. The ozone peak events were defined using the WMO air quality objectives of  $100\ \mu\text{g}\cdot\text{m}^{-3}$  and the long-term EU objective for ozone of  $120\ \mu\text{g}\cdot\text{m}^{-3}$  which is also the 95<sup>th</sup> percentile of the afternoon concentration in our dataset.”*

Lines 4-17: The discussion remains quite superficial and subjective, without any references to previous supporting literature.

*The discussion aims to describe the close association between the seasonal cycle of the STT proxies' events and ozone events.*

#### **Page 13**

Line 7 and 9: How was significance analysed? What do you mean by marginal significance?

*The significance was analyzed using the p-value of the estimates. We added a sentence in section 2 with a reference describing the method : “The Theil-Sen estimator, significance test and associated confidence levels are described in Carslaw and Ropkins (2012).”*

*Table S2 in SI shows the linear trend slope estimates with their upper/lower interval and confidence levels and significance. The pstar displayed the significance level according to the p value  $p < 0.001 = ***$ ,  $p < 0.01 = **$ ,  $p < 0.05 = *$  and  $p < 0.1 = +$ .*

*We mentioned a marginal significance because the significance levels were between 95% and 99% and not above 99%.*

Could you discuss what is the process related with such increases?

*We discussed further the trends in the revised section 3.3.*

Lines 27-29: They have been also associated with changing circulation patterns in some studies.



*The sentence has been modified and a reference has been added. “Recently, similar decreasing trends were observed in Spain and Italy (Cristofanelli et al., 2021, Adame et al., 2022) and were related to the reduction of anthropogenic emissions over Western Europe following the implementation of stronger environmental protection policies such as the EU clean air directive. Changes in the atmospheric circulation may have also played a role as was mentioned by Crespo-Miguel et al. (2024).”*

#### **Page 14**

Lines 12-16: Either you present and discuss the results, either you omit them directly, this halfway of presenting the results without discussing and after that switch to another topic saying that these results are not meaningful for the purposes of this work is not understandable and leave the reader with many doubts.

*We have put efforts into improving the presentation and discussion and to explain our approach to smooth the reading.*

#### **Page 15**

Lines 19-20: The impact of a particular transport pattern comes here out of the blue.

*The sentence has been removed.*

#### **Page 16**

Lines 11-18: How are the classes and their link with quantiles defined?

*The text explained that the seven classes have been selected to cover a wide range from large negative anomalies to large positive anomalies using a progressive increase in quantiles (10, 25, 40, 60, 75, 90 %).*

Figure 10, caption: Please explain or remind to the reader what is the meaning of AMJJAS and NDJF, which is a quite unusual separation (normally, the seasons are studies)

*This is now explained and justified on page 9 and detailed in the caption*

#### **Page 22**

Line 41: Saharan dust air masses have a particular link with O<sub>3</sub>, in particular they have been connected with O<sub>3</sub> reduction in some studies.

*We have removed the part concerning the June 2019 event.*

#### **Page 23**

Lines 11-22: Not clear how this connects to the rest of the paper.

*We have removed the part concerning the June 2019 event.*

#### **REFeree #2**

The introduction is very comprehensive, including highly up-to-date references. Some information has been included as supplementary material that could be interesting to incorporate into the main body of the manuscript, such as the seasonal evolution of CO, CH<sub>4</sub>, and the mixing layer height. The seasonal cycles of O<sub>3</sub>, NO<sub>x</sub>, CO, CH<sub>4</sub>, <sup>7</sup>Be, etc., are presented; however, these are actually monthly evolutions based on weekly averages, and in the case of ozone and NO<sub>x</sub>, it corresponds to the 0–24 h seasonal evolution.

Many databases, different statistical techniques, and various time periods are used, but the main objective of the study is not clearly defined. The last part of the results section (Section 3.3) was difficult to understand, and perhaps some effort should be made to clarify it. Some methodologies and time periods used are not clearly mentioned in Section 2. This work includes 15 figures, which I believe is an excessive number. The authors should reduce both the amount of information presented in the paper and the number of figures, and they should clearly define the study's objective and scientific contribution. Although the database used has significant scientific value, the study should be

more focused and not cover many different topics. Therefore, I encourage the authors to undertake a thorough revision.

*We have revised the manuscript to address the different referees' general and specific comments and particularly to improve the presentation of our approach and to clarify our main results. The number of figures has been reduced from 15 to 12, and the last part has been removed.*

Specific comments.

Page 1. Lines 30-31. "...where it has significant health...". As a suggestion, it would be helpful to indicate the cause of this impact. Maybe it is simply an oxidant agent that is considered an air pollutant at certain levels.

*This is now specified according to the referee's suggestion.*

Page 3. Lines 1-2. "...contrasted with Asia and China...". As China is in Asia, maybe the authors mean Southeast Asia.

*OK. This has been corrected according to the referee's suggestion.*

Page 3, Lines 12-13. "...is largest in spring and weakest in summer...". But isn't this statement dependent on the region?

*We have added "in Europe"*

Page 3, Lines 32-35. "Indeed,  $^7\text{Be}$  is produced...". This information about stratospheric tracers,  $^7\text{Be}$  and  $^{22}\text{Na}$ ; is it really necessary for this work?

*Because we use  $^7\text{Be}$  and  $^{22}\text{Na}$  as tracers, it is necessary to be clear on their respective origin. And it takes few lines.*

Page 4, Line 18. "...regional background station, OPE, ...". Although it has already been mentioned in the Abstract, this is the first time it appears in the manuscript. As a suggestion, I would write its full name here.

*Done*

Page 4, Line 16. "...This work contributes...". Have there been previous studies related to surface ozone measurements at this observatory, or is this the first study on surface ozone at OPE? Perhaps they are referenced later in the manuscript.

*We have rewritten this part. This is the first paper analyzing the different scales of temporal variability of surface ozone at OPE.*

Page 4, Line 36. "...a ground-based weather station operated...". Can we assume that the wind sensor is positioned at 10 m above ground level, and that temperature, relative humidity, and pressure sensors are at approximately 1–1.5 m? Is this correct?

*This is correct. The text was modified accordingly*

Page 5, Line 6. "...ERA5 reanalysis at the grid point...". What is the spatial resolution of this grid? I assume it is  $0.25^\circ \times 0.25^\circ$ . Perhaps providing more information about ERA5 would be useful.

*ERA5 reanalysis Data is available on a  $0.25^\circ \times 0.25^\circ$  grid. We added more information in section 2.2*

Page 5, Line 8. The database used for  $\text{O}_3$ ,  $\text{NO}_x$ , etc., contains hourly data.  $\text{O}_3$  is mentioned in  $\mu\text{g m}^{-3}$ , CO in ppm, and  $\text{CH}_4$  in ppb. What units are ultimately used in this work? Additionally, although CO and  $\text{CH}_4$  measurements are available at 10 m and 100 m, were both levels used in the analysis?

*We chose to keep the unit used in the available database ( $\mu\text{g.m}^{-3}$ , in the case of European Environmental Agency and ppbv in the case of Integrated Carbon Observation System).*

*A sentence was added: "We used hourly data reported in  $\mu\text{g.m}^{-3}$  following the recommendation of the European Environmental Agency."*



*We also specify: “Hourly concentrations are reported on the WMO-CO-2014 scale in dry mole fraction ppbv for CO and on the WMOX2004A in dry mole fraction ppbv for CH<sub>4</sub>, as available on the ICOS Carbon portal (<https://www.icos-cp.eu/observations/carbon-portal>).”*

Page 6, Line 13. “We used the CCGCRV...”. As a suggestion, could the authors clarify the meaning of CCGCRV?

*This has been done*

Page 6, Line 15. “...the trend component using the TheilSen and STL methods...”. If the authors are referring to the Theil-Sen estimator, perhaps it should be written as Theil–Sen. Also, could the meaning of STL methods be explained?

*Regarding the comment about “TheilSen”, we have modified the refence to Theil–Sen as correctly pointed out.*

*Regarding STL, we added its full name “Seasonal-Trend decomposition using LOESS (STL)”*

Page 6, Line 29. After reading section 3.1, a more appropriate title might be “Monthly variations from weekly averages for ozone, ....”.

*The seasonal cycles are computed using weekly data and are not aggregated on a monthly timescale. The data shown on figures 2 and 3 have a weekly resolution so the mean annual cycle has 52 points.*

Page 7, Line 5. Figure 1 appears before it is mentioned in the text.

*The figure has been moved after the text.*

What method was applied to normalize the seasonal daily cycles? Is it relative to the daily mean? Since several approaches can be used, how do the authors justify using relative values for comparison with other stations (Page 7, lines 11–14)? Absolute values are more commonly used in such comparisons.

*The figure has been modified to present the original data (i.e., not normalized).*

Page 7, Line 6. “...spring to winter seasons”. Perhaps in Section 2, the months corresponding to each season could be specified.

*The months are indicated on the figure.*

Page 8, Line 19. “Figure 2 shows...”. Again, Figure 2 is included before it is cited in the text.

*The figure has been moved after the text.*

My suggestion to the authors is to use the same scale on the Y-axis for all four graphs in Figure 4. This would allow for better comparison.

*We think that the referee discussed figure 2.*

How were the percentiles for each month calculated, from hourly or monthly averages?

*The weekly parameters were derived from the original hourly data and they describe different parts of the data distribution (baseline is the 5th percentile and peak is the 95th percentile). For a given week of 7 days, we have 168 hourly data points, and we take the 5th and 95th percentiles from these 168 values. The mean afternoon and mean nighttime are the average of the hourly data points restricted to afternoon and nighttime. A visual illustration of this process of aggregating hourly data onto weekly data is given in figure S2 in the Supplementary Information. We added a description of the process in the SI.*

Page 8, Line 8. “...when the boundary layer is larger...”. In reality, there is no strict linear relationship, since beyond a certain height, if the boundary layer is too high, precursors disperse vertically and do not form ozone. In some cases, lower heights are more efficient for ozone production.

*We modified the sentence to make it clearer: “In general, maximum surface O<sub>3</sub> concentrations are observed when the boundary layer is developed (during the afternoon and in summer) suggesting that the contribution of the photo-chemical production of O<sub>3</sub> dominates the dilution due to the increase of the boundary layer height and the vertical turbulent mixing.”*

Page 9, Line 11. NO<sub>x</sub> and O<sub>3</sub> are expressed in  $\mu\text{g m}^{-3}$ , while CO and CH<sub>4</sub> are expressed in ppb, meaning concentrations and mixing ratios are being used simultaneously. Is it possible to standardize these units?

*We chose to keep the units used in the available database ( $\mu\text{g.m}^{-3}$ , in the case of European Environmental Agency and ppb in the case of Integrated Carbon Observation System).*

Page 9, Line 25 (Section 3.1.3). If the information in Sections 3.1.2 and 3.1.3 is presented in the same Figure 3, why are these two subsections not combined?

*The two subsections are now combined.*

Page 10, Line 5. “Figure 3: Averaged...”. To help identify the graphs in this figure, why haven’t the authors used labels such as (a), (b), (c), etc? That might make it clearer. Additionally, the labels in the graphs do not match those mentioned in the text. For example, is the median CH<sub>4</sub> labeled as “ch410q50” or is the median CO labeled as “co\_10q50”? I suggest the authors clarify this for better readability.

*The graphs are now labelled using the parameter displayed (NO<sub>x</sub> CH<sub>4</sub>, CO, <sup>40</sup>K, <sup>7</sup>Be and <sup>22</sup>Na) ... and the labels on axis has been removed.*

Page 11. Figure 4. I suggest improving and making the labels in the graphs more concise. For example, in “...weekly mean relative humidity (top left panel), temperature...”, the reader understands that “RH %” refers to “Relative Humidity (%)” and “TM oC” to “Temperature (oC),” but the labels could be improved by adding spaces and placing units in parentheses for better clarity.

*The figure and the caption have been improved.*

Page 12. Line 4. I understand that the aim of Section 3.14 is to identify or associate the occurrence of ozone peaks with STT. Is this correct? This point is not entirely clear.

*The title of point 3.1.4 is “Peak ozone and STT events”. This is what is discussed in this paragraph.*

*The title has been modified to “Links between STT events and O<sub>3</sub> positive peaks”*

Page 12. Line 9. “...afternoon O<sub>3</sub> peak events...”. I understand that the authors define two types of ozone events, those exceeding 50 and 65 ppb. What criteria were applied to define these values? Perhaps the definition of events should be included in Section 2.

*We now specify “The ozone peak events were defined using the WMO air quality objectives of 100  $\mu\text{g.m}^{-3}$  and the long-term EU objective for ozone of 120  $\mu\text{g.m}^{-3}$  which is also the 95<sup>th</sup> percentile of the afternoon concentration in our dataset.”*

Page 12. Line 20. Figure 5. The graph should include a label explaining the meaning of each coloured line, not just in the caption.

*The figure has been modified accordingly.*

Page 13. Line 20. Figure 6. I suggest that the authors make the Y-axis labels more explicit. Labels should be added to indicate the meaning of each coloured line. These figures are not easy to understand.

*The figures have been modified to improve clarity.*

Page 14. Line 10. Figure 7. The same comments apply as for Figure 6. What does “RR” stand for Rainfall? The reader should not have to assume meanings to understand the work. As a suggestion, Figures 6 and 7 could perhaps be combined.

*This figure has been modified and moved to the Supplement Information.*

Page 14. Lines 27-30. These results were expected.

*This part has been removed.*

Page 15. Lines 18-19. "...the cold period (October to March) is longer than the warm period (April to September)". I think the definition of the cold and warm periods should have been presented earlier in the manuscript.

*This is now defined in 3.1.1. when discussing the O<sub>3</sub> seasonal cycle.*

Page 16. Line 8. "...Having identified two different periods using the correlation analysis,...". But which periods are the cold and warm periods?

*The text is now "Having identified two contrasting periods using the seasonal cycle, the wavelet and the correlation analysis, we then classified the ozone anomalies into seven classes ..."*

Page 16. Line 9. "Using the quantiles 10%, 25%, 40%, 60%, 75%, and 90%. These seven classes...". Why were these quantiles selected? What are these seven classes? But there are only six quantiles. It's a bit confusing.

*The text explained that the seven classes have been selected to range from large negative anomalies to large positive anomalies using a progressive increase in quantiles (10, 25, 40, 60, 75, 90 %). This corresponds to seven classes: 0-10; 10-25; 25-40; 40-60; 60-75; 75-90; 90-100.*

Page 22. Section 3.5. Is this paper also devoted to extreme events of surface ozone?

*Following this comment, we have removed the part concerning the June 2019 event.*

Page 22. Line 3. "...200 µg m<sup>-3</sup> ever recorded at the OPE regional station". This exceeds the threshold defined in the European Directives; perhaps this could be mentioned.

*We have removed the part concerning the June 2019 event.*

Page 22. Lines 13-16. But the factors that trigger an ozone episode in the Barcelona area may not be the same as in OPE. For example, "...the foehn effect induced by a local mountain wind regime".

*We have removed the part concerning the June 2019 event.*

Page 23. Line 12. "...Donon (Figure 2, Figure 15)...". Please, correct it. The use of the ozone data recorded in these background stations was mentioned in Section 2? Are the stations part of an air quality network?

*We have removed the part concerning the June 2019 event.*

### REFeree #3

The paper presents a large and comprehensive data set regarding the concentration of ozone in surface air. The introduction is detailed. Perhaps a figure showing the interplay of all the parameters might be helpful for readers with less background on this topic (or a reference to a review paper with such a figure).

*A reference to (Lu et al., 2019) has been made. This review paper includes a figure synthesizing the processes involved in tropospheric ozone chemistry.*

Much emphasis is given in the paper to the role of STT using <sup>7</sup>Be and <sup>22</sup>Na as proxies. This certainly is a valid and interesting approach. However, the interpretation of the cosmic radionuclides can be rather complex. Before averaging different years the <sup>7</sup>Be and <sup>22</sup>Na data should ideally be corrected for the changing intensity of cosmic rays. This could be done based on annual means of cosmic-ray neutron flux which is measured at various locations. (Perhaps such a correction would not change the seasonal cycles shown in Fig. 3 fundamentally, but in any case it should be mentioned that the correction is not applied.) Moreover, the <sup>7</sup>Be/<sup>22</sup>Na ratio is dependent on the residence time, since <sup>7</sup>Be decays much faster than <sup>22</sup>Na. This may be important with atmospheric residence times longer than one week.

*The  $^7\text{Be}$  and  $^{22}\text{Na}$  weekly time series are influenced by the 11-year solar cycle. The residuals computed by removing the low frequency variations and seasonal cycles have been corrected for this effect. We have also addressed this point in the specific comments below.*

With respect to the role of STT I would be interested in a quantitative estimate of its contribution to the surface  $\text{O}_3$  concentrations, especially for the June 2019 event case study. Should such more quantitative conclusions not be possible, the authors should explain why and perhaps say which additional information might enable one to do so. It seems that the correlation of the cosmogenic radionuclides and  $\text{O}_3$  in summer is also due to solar radiation (i.e. sunnier and warmer weather) which enhances both vertical advection (i.e. higher cosmogenic RN) as well as photochemical  $\text{O}_3$  production. Therefore, a stratospheric contribution of  $\text{O}_3$  may be difficult to be discerned. Is it possible to estimate the fraction of stratospheric ozone, e.g. based on typical  $\text{O}_3/^{22}\text{Na}$  ratios in the troposphere or upper stratosphere?

*In the revised manuscript we removed the June 2019 event. We also further discussed the limitations in the use of  $^7\text{Be}$  and  $^{22}\text{Na}$  to quantify the stratospheric contribution to surface ozone.*

In this context, it would be helpful to give some quantitative information of the vertical profiles of  $\text{O}_3$  from the stratosphere to the surface and to compare these with  $^7\text{Be}$  and  $^{22}\text{Na}$ . As the authors mention, it is not straight forward to distinguish stratospheric from upper tropospheric inputs based on  $^7\text{Be}$  and  $^{22}\text{Na}$ .

*Adding quantitative information of the vertical profiles of  $\text{O}_3$  from the stratosphere to the surface would require a significant addition to the paper including ozone probes data from neighboring sites or modelling results. This suggestion will be kept for additional work.*

Figures: There are probably too many figures in the paper. Some of the sub plots in Figs. 2-4 could go to the SI. The boxplots in Figs. 10-12 are not ideal to represent the data. The colors do not add any information. I think scatter plots with the indication of the R-value like in Figs. 13 and 14 are more informative. And again, some of these plots might go to the SI.

*The number of figures has been reduced from 15 to 12, and the last part has been removed. Some figures were moved to the SI.*

Overall, in my opinion, the paper could be improved by addressing the key processes contributing to surface ozone in a more focused and quantitative way.

*We have revised the manuscript to address the different referees' general and specific comments and particularly to improve the presentation of our approach and to clarify our main results. The revised version now is more focused and includes more quantitative results.*

Some more specific remarks and minor points:

page 5, line 40: "solar contribution" sounds like the particles from the sun would contribute to the production of Be-7 and Na-22. "solar modulation" would be a more appropriate term.

*The sentence has been rewritten as "In addition, the sunspot number index was used as a proxy of solar activity to evaluate its contribution to the modulation of  $^7\text{Be}$  and  $^{22}\text{Na}$  stratospheric production."*

page 6, line 12: "We fit the weekly ..." this sentence is not clear.

*The sentence has been rewritten "The mean seasonal cycle was estimated for each parameter and a trend analysis using several complementary tools was performed to isolate low-frequency interannual variations"*

first paragraph of 3.1.1: MHD is cited twice.

*This part was removed.*

Figure 1: How are the values normalized? To annual average and standard deviation?

*The figure has been modified to present the full data and not the normalized daily cycles.*

page 9, line 31: replace "that" by "than".

*Done.*

In the same sentence it is not clear which stations are compared.

*The sentence has been modified to: "However, during the warm months (with high activity levels extending from May to August), the OPE  $^7\text{Be}$  maximum is much more extended than the ones observed at northern European stations or at Monte Cimone in Italy (Brattich et al., 2017; Leppänen and Poluianov, 2022)."*

page 11, top paragraph: "stratosphere-troposphere transport is larger between March and August ..."  
How do the  $^7\text{Be}$  and  $^7\text{Be}/^{22}\text{Na}$  data include March and August into a period of larger stratosphere-troposphere transport?

*This paragraph has been modified to clarify.*

page 12, line 10: the threshold value for  $^7\text{Be}$  should be variable from year to year in order to reflect the varying source term (solar modulation of cosmic rays).

*Following this comment, we computed the events number index using the raw  $^7\text{Be}$  data and using a  $^7\text{Be}$  data normalized by its annual mean (to take into account the solar modulation). This does not have any impact on the seasonal cycle shown on figure 4. It has an impact on the interannual variations of the events numbers, but we are not interested in this feature in the present work. We kept the initial index for clarity using the raw  $^7\text{Be}$  data (avoiding another normalization processing step).*

Figure 5: add a legend

*This has been done.*

page 13, line 31: what about boreal wetlands?

*We have added "boreal" in the sentence: "The causes of these increases, which are still stiffly debated, may include an increase of the anthropogenic emissions, as well as the emissions from tropical and boreal wetlands which also contributed to high growth rates between 2020 and 2022 (Lin et al., 2024)."*

page 15, line 7: the cited figures are not correct

*The cited figures are now correct.*

page 15, line 10: March is not really transitional but quite well in line with AMJJAS in Figs. S11 and S12

*The figures S11 and S12 show monthly correlations. But later, we used weekly data. March is transitional in the sense that the shift from "cold" to "warm" regime may not be fixed in time and changes from year to year. To avoid such shifting weeks, we excluded the "transition months" of March and October in our weekly based analysis. We modified the manuscript to add:*

*"March and October were not further considered as they are transitional periods between the two contrasted seasonal regimes. As we use weekly data, the shift from "cold" to "warm" regime (in spring) and from "warm" to "cold" (in fall) may not be fixed in time and changes from year to year. To avoid such shifting weeks, being some years in the warm regime and some others in the cold regime, we excluded the "transition months" of March and October in our weekly based analysis."*

page 24, line 3: I would suggest to replace "sun radiation activity intensity changes" by "solar modulation of cosmic rays intensity".

*The sentence has been changed to "On the other hand, the interannual variations of  $^{22}\text{Na}$  and  $^7\text{Be}$  mirrored the solar modulation of cosmic-rays intensity, dominated by the so called 11-year solar cycle."*

Moreover, this is not really an important conclusion in the context of surface ozone concentrations.

*Maybe, but this has contributed to assessing the variability of radionuclides and thus to identify the causes of the surface ozone variability.*

page 24, line 14: "These weather conditions and synoptic situations also favor more active photochemical processes, suggesting that situations with a significant ozone component from stratospheric origin could frequently be associated with a larger ozone regional near surface photochemical production." Do the authors imply any cause-and-effect relationship here, or is that simply the co-incidence that warmer, sunnier weather affects both, vertical advection and photochemical ozone production?

*The sentence preceding the paragraph mentioned by the referee was, in the submitted manuscript, "They were associated with drier conditions and a solar radiation increase". And we mention that these synoptic situations also favor more active photochemical processes. So there is both coincidence and cause.*

*Nevertheless, the conclusion has been thoroughly revised to summarize the main results and improve its clarity.*