# Reply To Referee #2

We would like to thank Referee #2 for the valuable comments on our manuscript, which greatly improved our manuscript. We have revised our manuscript according to the suggestions. In the following, we made the point-to-point reply to the comments raised by Referee #2.

**Major comments:**

**Q2.1 The discussion is a bit superficial. Especially in the subsequent impact of different criteria on ODEs event identification (i.e., health impact, climate change, etc.). Besides, as mentioned in the final "Conclusions and Future Work", authors should involve more observation data (such as BrO) to further evidence the assumption or the attribution in the discussion.**

**A2.1** Thanks a lot for your insightful comments on our manuscript.

First, about the discussion on the subsequent impact of using different criteria to identify ODEs, in this study, we found that when using a criterion based on a constant threshold (10 ppbv or 5 ppbv) or thresholds considering the monthly averaged ozone and the standard deviation, an overall decline in the occurrence frequency of ODEs at Utqiagvik, Arctic from the year 2000 to 2023 can be revealed. Our findings are also in good agreement with those of Law et al. (2023), who reported a notable increase in observed surface ozone levels in springtime especially in April of BRW from 1993 to 2019. Burd et al. (2017), in their study on the Arctic BrO season, found a decrease in BrO concentrations in springtime and an earlier ending of the BrO season at BRW from 2012 to 2016, which may also indicate a reduction in the occurrence of ODEs at BRW, aligning with our findings. This declining trend of ODEs can lead to a weakening of the deposition of active mercury (Hg(II)), which is highly toxic and can pose serious health risks to humans. Therefore, the decline in ODE frequency could lead to a reduction in the health hazards associated with mercury deposition in mid-latitude regions. However, when a different criterion is used, a more significant declining trend of ODEs or no declining trend may be discovered, which may alter the conclusions. This is also the reason why we tested different criteria of ODEs in this study and compared the conclusions achieved. We have expanded the discussion in the revised manuscript to address these issues such as the potential health benefits resulting from the reduced frequency of ODEs. Please see lines 337-343, 422-424 and 426-428.

Regarding the reviewer's suggestion to involve more observational data such as BrO to support our conclusions, we fully agree with the reviewer that additional observational data, particularly BrO measurements, would greatly enhance our analysis and provide more robust evidence to support our conclusions. Unfortunately, we currently do not have access to such data. At present, we are actively seeking opportunities to collaborate with other research groups and institutions to obtain a broader range of observational data in the future. In the meantime, we will clearly acknowledge the limitations of our current dataset in the manuscript and emphasize the importance of obtaining additional data to further support our findings and assumptions. Please see lines 451-454 in the revised manuscript.

**Q2.2 The trend discussed in Section 3.1 and Section 3.3 are opposite, the discussion**

**throughout the paper should be more rigorous and unified.**

**A2.2** Thank you very much for the comments. We carefully checked the discussions in Section 3.1 and Section 3.3, and found the statements in Section 3.1 inaccurate which may have led to some confusion of the reviewer.

In Section 3.1 we described the interannual variability of ODE hours generally, because the focus of this section is the comparison of results between different criteria. In this section, we mentioned that there seems to have a *weak* upward trend in ODE hours between 2000 and 2012 if the year 2000 is excluded, suggesting a *slight* increase in ODE hours from 2000 to 2012. However, after the year 2012 (from 2012 to 2022), a *significant* decrease in ODE hours is indicated. In Section 3.1 of the original manuscript, we did not mention the general trend of ODE hours between the year 2000 and 2022, which may confuse the reviewer. Actually, a general decline in ODE occurrence frequency over this 23-year period can be observed from the results shown in Fig. 3(a) in the manuscript.

In Section 3.3, we provided a more quantitative analysis of the temporal behavior of ODEs, focusing on the monthly and yearly variability of ODE hours at BRW. We found April as the predominant month for ODEs and revealed a significant decrease in ODE hours over these 23 years, especially when using more stringent or relative thresholds. This section complements the broader interannual trends discussed in Section 3.1 by providing a finer temporal resolution and a more detailed examination for specific months and criteria.

In order to clarify the confusion of the reviewer, we have added more explanations in Section 3.1. Please see lines 183-184. We hope the confusion of the reviewer can be clarified after making these modifications.

**Q2.3 In section 3.2, the authors should add all the other tests' results in SI, other than the TM1, TM4, VM, and IF presented in main text.**

**A2.3** Thanks for the comment. According to the suggestion, we added the results of all other tests (TM1-5 ppbv, TM1-4 ppbv, TM2, TM3, and TM5) to the Supplementary Information. Please see Figs. S2 and S3 in the revised Supplementary Information. We hope it can provide a more comprehensive view of various criteria we used to identify ODE hours and their respective outcomes.

**Q2.4 A map of station location is necessary, especially when the author discuss the relationship between meteorological condition (such as wind directory) and ODEs event.**

**A2.4** Thank you very much for the suggestion. We added a detailed map of the station location into the revised version of the manuscript (see Fig. 1 in the manuscript). This map clearly shows the location of the BRW station at Utqiaġvik in the Arctic and includes key geographical features such as the Arctic Circle and major continents.

**Q2.5 The font in all figures should be of same size, there are some figures having too-small font, such as Figure 3, 5 and 6 etc... I suggest authors to replot all figures, all of which are not clear enough, and the styles are more like a report, not paper.**

**A2.5** Thank you for the suggestions on the font size and the clarity of figures in our manuscript. We have thoroughly revised all the figures to ensure consistency and clarity. We have also enlarged the fonts in Figures 3, 5, and 6 to make them more legible. Please see all the new figures in the revised manuscript. If the reviewer have any further recommendations or additional feedback on the improvements of the figures, we will continue to improve them until they are clear enough. Thanks again for the comment.

**Specific comments:**

**Q2.6 Line 20, the reason why the ODE only happen in spring should be explained in the Introduction part.**

**A2.6** Thanks. We have included an explanation for why ODEs predominantly occur in the spring in the introduction section of the revised manuscript. The revised text now explains that a unique combination of meteorological and chemical conditions in spring, including the presence of sunlight, strong temperature inversion, and the availability of halogen ions from ice/snow surfaces, creates the ideal environment for ODEs to occur. In contrast, these conditions are not simultaneously met in other seasons, which explains the seasonal occurrence of ODEs.

We added the following text into the Introduction section (lines 54-61 in the revised manuscript):

"The occurrence of ODEs is predominantly confined to the spring season due to the unique combination of meteorological and chemical conditions (Lehrer et al, 2004). First, the sunlight during spring is essential for photochemical reactions to take place, which is crucial for converting inert halogen ions into reactive halogens. Second, the strong temperature inversion that forms in the spring effectively isolates the boundary layer air from the free troposphere, preventing the downward mixing of ozone-rich air from aloft, allowing the reactive halogens to efficiently deplete the local ozone. Additionally, the snowpack above the sea ice in springtime acts as a significant source of halogen ions, as the brine layer on the sea ice is enriched with halogens that can be released into the atmosphere through photochemical processes. These factors collectively create the ideal conditions for ODEs to occur in the spring, while such conditions are not simultaneously met in other seasons."

**Q2.7 I suggest the author to move the Figure 1 to SI, and present the original O3 concentration trend since 2010 instead.**

**A2.7** Thanks for the suggestion. We have moved the original Fig. 1 to the Supplementary Information as Fig. S1. In place of the original Fig. 1, we present the figure showing the original $O_3$ concentrations in different months from the year 2000 to 2022 (see Fig. A1 in this rebuttal). This figure provides a direct view of the ozone changes over the past decade, which we believe will be more informative for the readers.

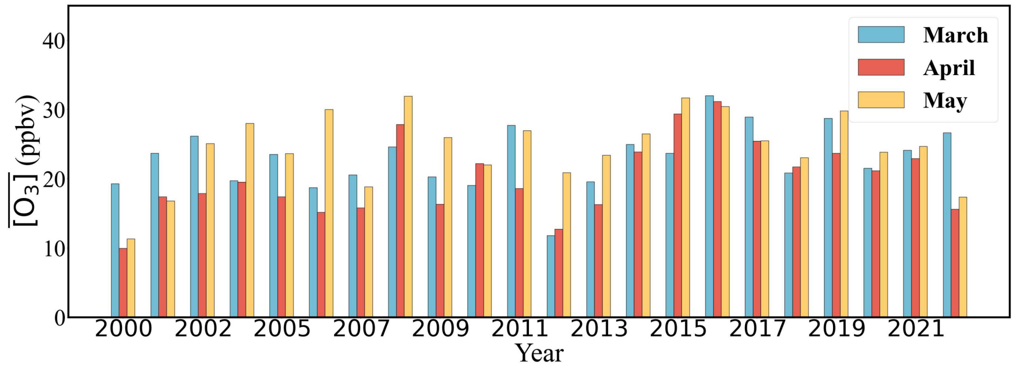


Figure A1 Monthly averaged ozone mixing ratios at the BRW Station for spring months (March, April, and May) from the year 2000 to 2022

**Q2.8 The Lines in Figure 2 is not easy to distinguish, I suggest authors to classified them into two panels, such as traditional results in one, and the rest methods in another one. The color of lines should reflect the method clusters. The current color setting is too hard to follow.**

**A2.8** Thanks a lot for the suggestion. We modified Figure 2 according to the reviewer's suggestion (see Fig. A2 in this rebuttal). In the present version of the manuscript, the figure is divided into two subplots. The upper subplot presents the results of the TM series methods, while the lower subplot shows the results of the VM and IF methods. In addition, we have re-ordered the curves in each subplot according to the mean values of the criteria. Moreover, we have adjusted the colors of the lines to better reflect the method clusters, ensuring that the current color setting is more intuitive and easier to follow. We hope these changes will address your concerns and enhance the clarity of the figure.

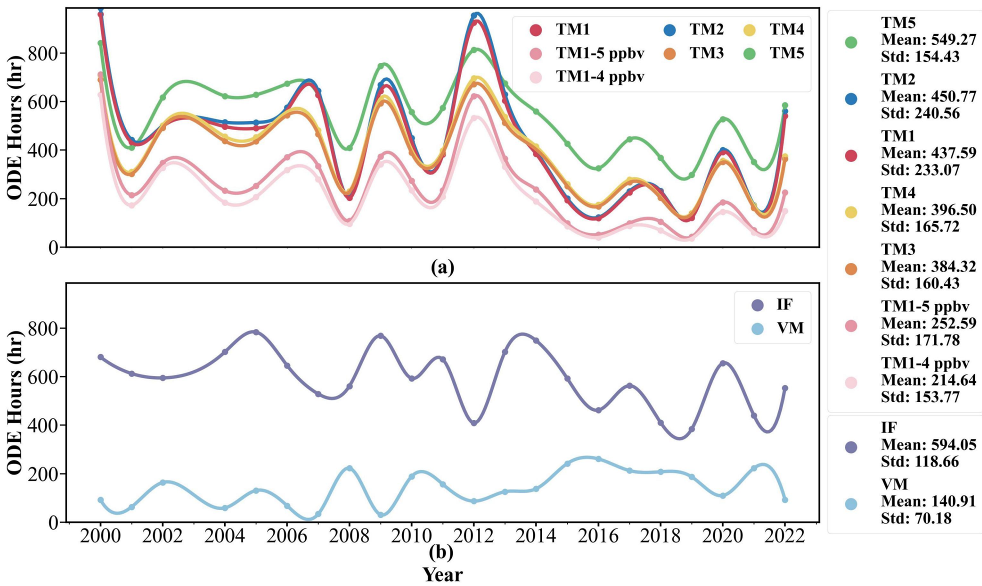


Figure A2 Number of ODE hours identified by each criterion from 2000 to 2022

**Q2.9 Line 200, the IF curve and VM curve are not alike at all in Figure 2. How do the authors have this conclusion: "it is seen that after the year 2014, the IF curve behaves**

**similarly to those of the TM methods, while before 2014, the IF curve's trend is more like the VM method's trend"?**

**A2.9** Thank you for the comment. Upon re-examining Figure 2, we agree that our initial conclusion regarding the similarity between the IF curve and the VM curve was not that accurate. We appreciate the reviewer for pointing it out.

We have modified the text to more accurately reflect the observed trends in the data. Specifically, we changed the number "2014" to "2008", as the IF curve's trend before 2008 is more similar to the VM method's trend. The updated text now clearly states these observations without any misleading statements. Thanks again for pointing this issue out.

"With respect to the Isolation Forest method (see the IF curve in Fig. 3b), generally, the number of ODE hours screened by this method is comparable to those picked out using TM methods. Interestingly, it is seen that after the year 2014, the IF curve behaves similarly to those of the TM methods, while before 2008, the IF curve's trend is more like the VM method's trend, although the values are significantly higher, indicating a possible consideration of the standard deviation in the IF method. Because of the black box characteristics of machine learning models (Hassija et al., 2024), it is difficult for us to further explore the reasons and principles behind the screening results of this method. Further interpretability of this machine learning method is also one of the areas we aim to investigate in the future."

**Q2.10 Line 235. Add results in 2013 and 2022 in SI.**

**A2.10** We have added the results for 2013 and 2022 in the Supplementary Information (SI) in the revised version of the manuscript. Please see Figs. S4(a) and (b) in the revised Supplementary Information. Thanks.

**Q2.11 Line 265, the conclusion of "the machine learning approach exhibits a limitation in accurately identifying ODE hours in years characterized by a high frequency of ODE occurrences" is too arbitrary, since the authors only tried one ML method.**

**A2.11** Thanks for the suggestion. The reviewer is correct that our conclusion was overly broad, as we only tested one machine learning method, the Isolation Forest (IF). We have changed the text as follows to be more precise.

"Thus, the IF method exhibits a limitation in accurately identifying ODE hours in years characterized by a high frequency of ODE occurrences."

Please also see line 295 in the revised manuscript.

**Q2.12 How is regression done in Figure 5?**

**A2.12** The reviewer doubted about the linear regression analysis shown in Figure 5. The specific steps and details of the regression analysis are as follows:

To assess the trend of ozone concentration over time, we employed linear regression analysis. Linear regression is a classical statistical method used to establish a linear relationship between a dependent variable (such as ODE hours) and one or more independent variables (such as the year). Through the method of least squares, the linear regression model can find the best-fit line to quantify the trend of the dependent variable with respect to the independent variable (Draper & Smith, 1998; Weisberg, 2005; Montgomery et al., 2021).

The general form of the linear regression model is:

$$Y=aX+b,$$

where Y is the dependent variable (ODE hours in this study). X is the independent variable (year in this study). The slope a is the regression coefficient, representing the rate of change of Y with X. The intercept b represents the value of Y when X=0.

For computational convenience, we transformed the variable X (i.e., year) into its difference from the year 2000 (i.e., X-2000). Thus, the model can be written as:

$$Y=a(X-2000)+b,$$

where the slope a and the intercept b are determined by data fitting.

To evaluate the significance of the regression model, we also calculated the p-value. The p-value is usually used to judge whether the regression is significantly or not, with $p<0.05$ typically indicating that the regression is statistically significant.

$$a=\frac{\sum_{i-1}^{n}(X_i-\bar{X})(Y_i-\bar{Y})}{\sum_{i-1}^{n}(X_i-\bar{X})^2}$$

$$b=\bar{Y}-a\bar{X}$$

$$SE(a)=\frac{\sum_{i-1}^{n}\frac{(Y_i-\bar{Y})^2}{n-2}}{\sum_{i-1}^{n}(X_i-\bar{X})^2}$$

$$t=\frac{a}{SE(a)}$$

$$\text{p-value}=2\times P(T>|t|)$$

Where $\bar{X}$ and $\bar{Y}$ are the means of X and Y, respectively. Where a is the regression coefficient and SE(a) is its standard error. Where T is the t-distribution random variable with $df=n-2$ degrees of freedom, and P represents the probability. The results shown in the present manuscript indicate that the trend of ozone concentration over time is statistically significant ($p<0.05$) or highly significant ($p<0.01$).

We added a sub-section describing the linear regression used in the revised manuscript. Please see the Section 2.3 (lines 161-174) in the revised manuscript .

**References**

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