Reviewer #3 (Comments to the Author):

Title: On the dynamics of ozone depletion events at Villum Research Station in the High Arctic

In this study, Pernov et al. investigate ozone and ozone depletion events (ODE) over a long time period (1996-2019) at the Villum Research Station located northeast of Greenland. A statistical analysis and machine learning (ML) approach is used to analyze the relation of ozone and meteorological variables as well as back-trajectories to study air mass history and surface properties. ODE frequency and duration were found to be highest in May, declining in April and March. Sunny and calm conditions connected with northerly winds seem to favor ODEs in Villum. The ML model revealed that radiation, time over sea ice, and temperature seem to be the most important variables for modeled ODEs during spring time.

To my knowledge, there has been no study which applied an ML approach to investigate ODEs at a specific location. This approach adds some further information regarding the interaction between variables and indicates threshold values for some variables that contribute to ODEs. However, since ML is still a fairly new method and probably relatively unknown to some in this community, I suggest, to include large parts of the ML description from the supplements into the main text, especially the explanation of SHAP values (see below in 'Specific comments).

> We originally moved the two pages of text describing the ML methods to the SI to reduce the overall length of the article. We admit this was not the correct decision in hindsight. Therefore, we have moved the entire description of the ML methods to the main text.

Further, I am uncertain about why one week was chosen for back-trajectories, as the ODEs at Villum are usually limited to a few hours, with only a few exceptional cases extending to several days. In addition, it was found that ODEs mainly occur under calm and stable meteorological conditions, which would suggest only minor transport of air masses during ODEs. This long time span could bias the analysis, particularly when examining the time above the mixing layer, which occurs towards the later part of the trajectory (see below in 'General comments' and 'Specific comments'). But overall, this paper is a pleasure to read, particularly the results and discussion parts are very well-executed. Therefore, I recommend publication in ACP with minor revision.

> We thank the reviewer for their comments and suggestions. We have addressed each comment below with review comments in black, author response in blue, and additions to the original text in red. We have indented the author's response for clarity. Lines numbers given in the author's response refer to lines in the revised manuscript.

General comments:

Why were 7-days backward trajectorys chosen? On page 27, lines 809-815, you already list the problems of these long backward trajectories (uncertainties, distortion due to the predominant time over the mixed layer further back along the trajectory). Furthermore, it is relatively unrealistic that air masses from 7 days before have a direct influence on ODEs in Villum, especially when they seem to occur mainly during calm and stable conditions and are therefore less affected by transport of air masses. Accordingly, it is also quite unlikely that the Chukchi Sea and the Beaufort Sea are relevant source regions for the "average" ODE in Villum. This could be the case in situations with a lot of transport (e.g. cyclone), but this seems to be the exception here.

> We selected a back trajectory length of one week to fully capture the air mass history of ODEs. The motivation for this selection was based on the longest ODE duration observed during the

study period (~6.5 days) and avoiding uncertainty associated with long trajectory calculations (~10 days) (Stohl, 1998). ODE air masses can extent over great distances in the Arctic (Halfacre et al., 2014) and satellite studies have shown that enhancements in reactive halogens (e.g., BrO) is a widespread phenomenon in the Arctic covering several million square kilometers including both sea-ice covered surfaces and continental regions (Bougoudis et al., 2020; Platt and Wagner, 1998; Richter et al., 1998; Schönhardt et al., 2012; Skov et al., 2004). Additionally, reactive halogens can be recycled on aerosol particles which can persist over large spatial scales (Peterson et al., 2017), therefore a longer trajectory time is warranted. Emission of reactive halogen species is ultimately a surface related process (snow on land, snow on sea ice, frost flowers, sea salt emission, refreezing leads, amongst others) and these halogen recycling can be sustained on aerosol particles aloft, selecting a longer trajectory time allows us to fully capture all of these processes. ODEs longer than one week have been observed at the Arctic land-based station, Alert, which observed an ODE of 9 days (Strong et al., 2002). Depletion of ozone has been shown to persist for several weeks in the central Arctic Ocean (Bottenheim et al., 2009). Other studies have used shorter (Bognar et al., 2020; Frieß et al., 2023) or longer (Begoin et al., 2010; Bottenheim and Chan, 2006; Simpson et al., 2018) trajectory lengths than one week, therefore a trajectory length of one week is a compromise between short and long trajectory lengths. We have added text to motivate our select of back trajectory length.

Lines 176-184: The trajectory length was chosen to avoid the uncertainty associated with extremely long trajectory calculations, while capturing the entire geographic extent of ODE air masses. This trajectory length of one week roughly corresponds to the longest observed ODE at Villum during the study period (~6.5 days, Sect. 3.1) and is shorter than the longest observed ODE at a land-based station (9 days at Alert by Strong et al. (2002)). Previous studies have shown that ODE air masses can extent over great distances in the Arctic (Halfacre et al., 2014; Peterson et al., 2017), therefore we selected a trajectory length of one week to fully investigate the air mass history of ODEs. Other studies have used shorter (Bognar et al., 2020; Frieß et al., 2023) or longer (Bottenheim and Chan, 2006; Begoin et al., 2010; Simpson et al., 2018) trajectory lengths than one week.

I would suggest to shorten the "Summary and Outlook" section, especially the last four paragraphs. In general, all of the topics mentioned in these four paragraphs are relevant and related to tropospheric ozone in the Arctic, but in some cases they are not directly related to what you did in your study (e.g. radiative forcing, AMDEs, cloud cover, etc.) so they come a bit out of the blue and lack context.

We purposefully called the last section “Summary and Outlook” over “Conclusions” so we could provide a detailed summary of our results and conclusions from this study as well as give an outlook on the effects of a changing climate on the occurrence of ODEs. Given there are significant changes in ODE frequency and ozone mixing ratios observed around the Arctic (Law et al., 2023; Tarasick and Bottenheim, 2002), a discussion about the possible environmental conditions that could affect these changes in future is highly relevant to the Arctic atmospheric chemistry community and ACP readers. The threshold ranges revealed by the ML model is highly pertinent to such a discussion as we identify thresholds for the influence of ambient meteorological parameters on the likelihood of an observation to be predicted as an ODE. This is especially pertinent given the rapid pace of change in meteorological conditions in the Arctic (Rantanen et al., 2022). Although these threshold ranges are site-specific to Villum, it is our hope that future studies will incorporate such a methodology to identify threshold range (or lack thereof) at other High Arctic sites with long-term in situ meteorological and ozone mixing ratio data.

While we aspired to investigate all aspects related to springtime ODEs, certain topics are inevitably outside the scope of the study such as radiative forcing although they remain important and relevant topics related to Arctic tropospheric ozone (as the reviewer noted). A discussion of these topics is pertinent for the larger Arctic atmospheric chemistry community, who would already be familiar with these concepts.

After review of the "Summary and Outlook" section, we have decided each paragraph adds value to the overall discussion and would foster future studies of Pan-Arctic ODEs and how climate change would affect them. Therefore, we opted not to remove them.

Specific comments:

Page 4 Line 132: It would be more coherent to use consistent units for the uncertainties, either % or ppbv.

We use common notation of uncertainty following EN norms. The uncertainty close to detection limit has to be indicated in absolute values as relative uncertainty (%) goes to infinity close to zero. At mixing ratios much greater than the detection limit and within the calibration range of the instrument the relative uncertainty is used. In this area the absolute uncertainty is a function of the mixing ratio. Thus, we keep the notation of uncertainty and which we have applied in earlier papers in e.g. Skov et al. (2020).

Page 6 Chapter 2.6: Include parts of the Supplement in here: missing data imputation, machine learning, model, ML explain ability approach

We have now moved the entire description of the ML methods from the SI to the main text.

Page 9 Lines 265-268: This sentence is very long and hard to read. I would suggest to split it up in several sentences.

We have split this sentence into two.

Page 10 Line 325 and following: Maybe include a 'snow on land' to every 'snow' in the text, to make clear that no snow on sea ice is analyzed.

We have made this change throughout the manuscript where appropriate.

Page 11 Figure 4: I suggest to only have 2 images per row, to make the individual plots bigger. Even when zooming in, the numbers on the bars are very hard to read.

We have rearranged the subpanels of Fig. 4 so that there are only two subpanels per row.

Page 13 Line 366: Is it really a ODE source region and not just a origin of the air masses? (see above General comments)

We have changed the text on these specific lines and throughout the manuscript to indicate we are referring to ODE air masses to be more precise.

Lines 475-477: During March, the main source regions for ODE air masses appear to be the Chukchi Sea while for Non-ODE air masses the main source region is Greenland with a minor contribution from the central Arctic Ocean (Fig. 6a and d).

Page 13, line 367: Perhaps it should be emphasized that Greenland plays a more important role for the Non-ODE source region (due to the higher trajectory frequencies) compared to the Arctic Ocean.

We have changed the text to indicate Greenland is the main source region and the central Arctic Ocean makes a minor contribution.

Lines 475-477: During March, the main source regions for ODE air masses appear to be the Chukchi Sea while for Non-ODE air masses the main source region is Greenland with a minor contribution from the central Arctic Ocean (Fig. 6a and d).

Page 15, Line 415: Maybe list different surface types here and mention already that land without snow does not play a role. This came as a bit of a surprise further down in the text.

We have added the surface types "(sea, sea ice, or snow on land)" to lines 528-529.

Page 16 Lines 428/429: Shouldn't it be '… start to descend earlier ...'?

This sentence refers to a comparison of Non-ODE air masses during May to those during March/April. During May, Non-ODE air masses begin their descent later along the trajectory (further back in time) compared to March/April. May Non-ODE air masses begin their descent on average at approx. 50 hours back along the trajectory while those during March/April begin their descent closer to the start of the trajectory. In other words, May Non-ODE air masses spend more time within the mixed layer compared to March/April Non-ODE air masses. To alleviate any confusion, we have added "compared to March and April" on line 541.

Page 17, Line 455: What is meant by 'model performance' here? I only see an increase in the 'Recall' variable from March to May.

We were referring to the Recall metric here, which gives the most informative measure of model performance although all three metrics are complementary. We admit the other two metrics do not increase from March to May and this was not explained in the text properly.

We have added the following lines to make this clearer in the text:

Lines 566-582: The evaluation metrics of the ML for all spring months combined and individual months are displayed in Table 1. We use three common metrics for evaluating a binary classification ML model: accuracy, recall, and AUC ROC (Area Under Curve Receiver Operating Characteristics). Briefly, accuracy is the fraction of correctly predicted observations regardless of label (ODE vs Non-ODE), recall is the fraction of ODEs correctly predicted and AUC ROC evaluates how well a model can discriminate between positive and negative labels across all decision thresholds for binary classification. In general, the ML model can accurately reproduce ODEs over all spring months combined as evidenced by how all three metrics are close to unity (their maximum value). However, when evaluating the results on an individual monthly basis, there is an increase in the recall metric and decrease in the accuracy and AUC ROC (see Sect. 2.6 for a detailed description of the evaluation metrics) from March to May (Table 1), which is likely connected to the increasing frequency of ODEs from March to May. With increased ODE occurrence, the recall metrics would increase as positive labels (ODEs) are more likely to be identified when they occur more often and the accuracy and AUC ROC metrics would decrease with the increased occurrence of positive labels due to a concurrent increase in number of incorrectly labeled ODEs. The ML model is also free from over-fitting given the close agreement between the train and test sets. Overall, this ML model is sufficiently accurate, robust, and suitable for the investigation of ODEs.

Caption of Table 1: The accuracy gives an overview of the model performance for both labels (ODEs vs Non-ODEs), recall gives the model performance only for positive labels (ODEs), and AUC ROC evaluates the model performance over different decision thresholds, together, these three metrics give a comprehensive view of the model's performance. The three metrics range from 0 (worst) to 1 (best).

Page 17, Line 458: The difference in the train and test data set and how the model is trained should be explained more detailed in Chapter 2.6.

We have added the following sentence in Sect. 2.6 about the differences between the train and test set.

Line 259-262: The purpose of the training set is for the model to learn how to model the data and the test set is used to evaluate the model's performance on unseen data.

We describe how the ML model learns (i.e., is trained on the data) in the previous paragraph.

Page 17 Line 466: Are the mean SHAP values meant by 'The mean ...'? Should be specified.

Yes, this is our intention here. We have added "SHAP values" to line 606.

Page 18 Line 485: The relationship between SHAP and ambient values and the information its results provide for this study should be explained more detailed (maybe with an example).

We have added the following text in Sect. 3.4 describing the SHAP methodology in plain terms.

Lines 594-605: The SHAP approach is designed to estimate the importance of each input variable to the model output based on coalitional game theory (Molnar, 2022) (see Sect. 2.6 for a more detailed description). SHAP values represent the marginal contribution of each input variable to the model output, or in other words: how important each variable is to the model for making a prediction. SHAP values can be positive or negative, with positive values indicating a variable is more likely to contribute to an observation being predicted as an ODE while negative values mean a variable is more likely to contribute to an observation being labeled as a Non-ODE. The SHAP methodology can produce both local and global explanations. The global importance gives an overview of the most important variables to the model output. The local importance of each observation can give information about the relationship between the SHAP and input values (positive or negative relationship, linear or non-linear), or in other words how does the model output vary over the range of input values.

Page 18 Line 497: Does 'negative effect on model prediction of ODEs' mean the model predicts ODEs wrong when RH is below average?

It does not, "negative effect on model prediction of ODEs" for below average RH values indicates that the model is more likely to predict a Non-ODE rather than an ODE. We have added text to indicate this.

Lines 638-639: (i.e., the model is more likely to predict a Non-ODE)

Page 19 Line 505: Maybe 'after this bin' should be replaced with 'towards lower temperatures'

We have made this change.

Page 20 Figure 10: I suggest to include a legend as it was done in Figure 4. An explanation of what the lines represent in the images should be included in the figure description.

We have changed the caption of Fig. 10 to indicate what the lines and bars represent and how the legend is the same for both.

Caption of Fig. 10: The relationships between SHAP and ambient values for (**a**) RH, (**b**) wind speed, (**c**), temperature, (**d**) radiation, (**e**) pressure, (**f**) wind direction, time air masses spent over (**g**) sea ice, (**h**) snow on land, and (**i**) time above the mixed layer for each month. Fifteen equally spaced bins were calculated for each variable, and the median of the SHAP values was computed for each bin, as represented by the colored lines. The value listed on the x-axis is the midpoint of each bin. The colored bars represent a histogram of the ambient values for each month. The relative frequency of each histogram bin for each variable is displayed on the right axis. The legend is the same for the colored lines and bars.

Page 22 Lines 598-600: Have there been any investigations into halogen release during ODEs in Villum (generally or specifically for this study)?

Measurements of halogen were not available for inclusion in this study so unfortunately there have not been.

Page 22 Lines 606-609: I suggest to make 2 sentences out of this long one.

We have split this sentence into two on lines 755-757.

Page 22 Line 616/617: Might it be possible to use the ERA 5 solar radiation to investigate solar radiation along the trajectory path?

The high resolution (0.25°) ERA5 data of surface solar radiation downwards was only extracted for the ERA5 grid cell containing the location of Villum Research Station and not for the entire Arctic region. This is due to the very large size of highly resolved gridded datasets for the entire Arctic over several decades. This reanalysis product is only representative of the surface and is not vertically resolved (similar to the solar radiation product from HYSPLIT and NCEP/NCAR (Kalnay et al., 1996)). ERA5 on pressure levels does not include solar radiation (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels?tab=overview). Therefore, we are unable to perform this analysis for this manuscript.

Page 23 Line 662: Maybe include a rough location of the buoys, so one can assume where northerly/easterly/westerly is located.

We have added the text "from the Beaufort Sea" to indicate the approx. location of the buoys on Line 815.

Page 24 Line 673: Which relationship is meant here?

We have added text to indicate the relationship we are referring to is for wind speeds and ozone mixing ratios and normalized ODE hours.

Lines 825-829: Our statistical analysis revealed no relationship between wind speeds and ozone mixing ratios/normalized ODE hours during March, a tendency for high normalized ODE hours with higher wind speeds during April (although little effect on ozone mixing ratios), and two modes during May (one at low and one at high wind speeds) (Fig. 4c).

Page 25 Line 734: What is meant by 'higher values' here?

We have changed the text to indicate that ODEs experience higher values of time over sea ice compared to Non-ODEs.

Lines 898-899: The amount of time spent over sea ice increases from early to late spring (Fig. S4f) and ODE air masses experience higher values of time over sea ice during each spring month compared to Non-ODEs (Fig. 5f).

Page 25, Lines 748/749: Are ODEs meant here and not air masses? If the air masses have low ozone levels, these can only be observed in Villum.

We are referring to the ODE air mass source regions of in these lines. We have changed the text to indicate this.

Lines 911-915: During April, ODE air mass source regions are located over the Beaufort and Chukchi Seas but also over the central Arctic Ocean, which represents a mix of FYI and multi-year sea ice (MYI). During May, ODE air mass source regions are in closer proximity to Villum, mainly arriving from the central Arctic Ocean, which contains the highest concentration of MYI.

We have changed the text throughout the manuscript to indicate we are referring to ODE air masses or ODE air mass source regions where appropriate.

Page 26, Lines 772-777: Split this into two sentences.

We have made this change.

Page 26, Lines 774/775 & 782: The acidity as an additional factor for ODEs comes a bit out of the blue here. I would suggest to include some sentences about acidity and its impact on halogens/ODEs in the Introduction or exclude the acidity part from the text.

We have added text in the introduction highlight the role of acidic surfaces in halogen propagation.

Lines 59-60: These reactions require the presence of a frozen, heterogenous surface aided by high acidity (Sander et al., 2006; Simpson et al., 2007, 2015).

Lines 93-95: This is likely connected to the need for an acidic, frozen heterogeneous surface (sea ice, snowpack, blowing snow, and aerosols) required for halogen propagation (Burd et al., 2017; Jeong et al., 2022), although other studies have not found such evidence (Halfacre et al., 2014; Jacobi et al., 2010).

We have added this reference in other locations throughout the manuscript where appropriate.

Page 27 Line 812: Have you tried what happens if you take shorter (e.g. 3 days) back-trajectories?

For this study, we have not extensively tested the sensitivity of the trajectory length, accuracy of the mixed layer height output from HYSPLIT, nor the starting altitude at the receptor location. Using a shorter back trajectory length would result in the source regions being closer to Villum due to the limited geographical extent of shorter trajectories. This would likely affect the results for March which had the furthest ODE air mass origins compared to April and May. The distribution of the air mass history variables would also change which can be discerned from Fig. 8, which shows the different surface types encountered by air masses as a function of time. This would likely result in the total percentage of time above the mixed layer being reduced. As mentioned in our reply in the general comments, we selected one week to fully capture the air mass history of ODEs. The accuracy of the mixed layer height is likely influenced by the presences of temperature inversions in the Arctic which are difficult to capture in meteorological reanalysis data (Gryning et al., 2023; Xi et al., 2024). The starting altitude, in combination with the accuracy of the mixed layer height, would affect the proportion of time air masses spent within or above the mixed layer, although this would have the largest influence closer to the measurement location. Starting trajectories at a shorter altitude could result in trajectories intercepting the surface would which affect their accuracy (Stohl, 1998) and starting them too high would result in trajectories not being representative of the measurement site. A previous study at Villum tested the effect of the starting altitude but on a short term campaign basis and found similar results for 20 vs 50 m although trajectories starting at 20 m often intercepted the surface (Pernov et al., 2021) thus a higher starting of 50 m was selected for that study and therefore this one. These topics are worth investigating, therefore, we have added text highlighting avenues for future studies.

Lines 985-988: Proper representation of air mass history therefore is an important aspect of evaluating ODEs and other atmospheric phenomena and future studies should evaluate this in more detail including the effects of varying trajectory lengths, the accuracy of the mixed layer height from HYSPLIT, and starting altitude at the receptor location.

Page 28 Line 835: 'high time' → long time?

We have made this change.

Page 28 Lines 840-842: see Page 13 Line 366

We have added text to make these two sections congruent.

Lines 1010-1011: During March, sea ice (likely FYI) in the Chukchi Sea is the main source region for ODE air masses.

Supplement:

S1 Machine learning modeling methodology

Second paragraph:

'We imputed missing data using the median value for the hour of the day for that day

of the year.' This sentence is very hard to follow, maybe describe it with an example.

We have added an example of this procedure.

Lines 234-235: For instance, if a value is missing for hour 12 on the 90^{th} day of the year then this value was imputed using the median of all values from hour 12 on the 90^{th} day of the year from the entire dataset.

Fourth paragraph:

'Tuning was performed for 1000 trials and the best parameters were selected.' Is parameters or hyperparameters meant here? If parameters, maybe shortly explain the difference.

We have changed "parameters" to "hyperparameters" on line 266.

Figure S3 description Lines 3/4: Which blue bars?

We have changed the text to show black bars represent not SS trends.

Caption of Fig. S3: The black bars represent trends that are not significant on the 95^{th} % confidence level.

Technical corrections:

Page 4 Line 120: please define i.d.

We have defined "i.d." as "inner diameter" on Line 124.

Page 6 Line 203: please define RH (first mention)

We have defined "RH" as "relative humidity" on Line 150.

Page 7 Line 234: please define CA (first mention)

We have written out the country name "Canada" throughout the text.

Page 8 Line 246: please define CL (first mention)

We have defined "CL" as "confidence level" on Line 136.

Page 8 Line 248: CL instead of Cl

"CI" on this line represents "confidence intervals" which we have defined on line 351 and in the caption of figure 3 on line 366.

Page 8 Line 258: (d) bold

We have made this change

Page 9 Line 291: ODE instead of ODEs

We have made this change

Page 23 Line 660: please define AK (first mention)

We have written out the state name "Alaska" throughout the text.

Supplement:

Figure S19 → Figure S9

We have made this change

Begoin, M., Richter, A., Weber, M., Kaleschke, L., Tian-Kunze, X., Stohl, A., Theys, N., and Burrows, J. P.: Satellite observations of long range transport of a large BrO plume in the Arctic, Atmospheric Chemistry and Physics, 10, 6515–6526, https://doi.org/10.5194/acp-10-6515-2010, 2010.

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