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Comment on egusphere-2022-370

Anonymous Referee #2

Referee comment on "A three-dimensional simulation and process analysis of tropospheric Ozone Depletion Events (ODEs) during the springtime of Arctic using CMAQ" by Simeng Li et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-370-RC2, 2022

Review for article egusphere-2022-370 titled “A three-dimensional simulation and process analysis of tropospheric Ozone Depletion Events (ODEs) during the springtime of Arctic

using CMAQ” by Li et al.

In this manuscript, the authors have presented a modeling case study of ozone depletion events at Barrow, Alaska in spring 2019. They make use of the CAMQ model to show that the ODEs can be explained mainly by bromine chemistry and advection of ozone poor airmasses. The model is compared with ozone observations, along with some meteorological parameters for validation.

Unfortunately, the paper falls short on several aspects. The main criticism of the manuscript is that it does not convince the reader about the applicability of the model for reproducing ozone depletion considering the specific tuning of the model to get the ozone right. This tuning suggests that the emissions and chemistry are not correctly described in the model even if it gets the meteorology right. Considering this, I cannot recommend that this paper be published in its current form. I would prefer to see a more detailed section highlighting the uncertainties, with some quantification of the uncertainties using sensitivity analysis. The value of the paper would come from understanding what needs to be improved rather from the using of tuning to reproduce two ODEs. From the results, the model does well at getting the meteorology correct, so the issue should be with the emissions and chemistry. How would this paper help in understanding which is the main issue and what needs to be improved? More detailed comments are given below:

Major comments:

- The increase in k_{R3} by one order of magnitude is justified by saying that it helps explain the ODE – no comparison with bromine observations is offered. This begs the question, is the model getting the ODE right for the wrong reasons? No uncertainty evaluation has been done to find the sensitivity of the model to k_{R3} or if any other factors can help in explaining the ODEs. The authors mention that '*ozone and other species in the computational domain can be greatly affected by the implemented boundary conditions'* – so how much confidence do they have in the model simulations and which is the main driver of the uncertainty?
- Boundary conditions for ozone – the authors have applied their boundary conditions for ozone using arbitrary values without further explanation. They have changed the CAM-Chem outputs to '*For the ozone in the boundary layer, if it is on the surface of sea ice, ozone was set to 3 ppbv; if it is over the sea, ozone was set to 10 ppbv; if it is at a coastal area, ozone was set to 15 ppbv. Furthermore, because the free atmospheric ozone can also be remarkably affected by the bromine explosion and the ODEs, ozone in the free troposphere in the implemented boundary condition was also reduced to half of the original value to consider the influence of the bromine explosion'*, mentioning that further information is in the code availability section. However, this section just lists the input data with no explanation for why these values are used and what observations they base these values on. One gets the impression that the model has been tuned to simulate the observed ozone depletion, hence quantification of the effect of chemistry and dynamics is inherently biased.
- This issue is also acknowledged by the authors on lines 190-193 – '*It can be seen that without this adjustment, the simulated ozone on March 29th would be largely different from the observations*'. Hence, the skill of the model to simulate ozone is doubtful.
- Maximum concentrations of BrO, peaking at above 100 pptv are much higher than reported anywhere in the Arctic. This suggests that the model is not able to simulate the bromine well and/or suffers from the lack of other sinks for ozone. Recently it has been shown that iodine also plays an important role in the magnitude of ODEs in the Arctic (Benavent et al., 2022). The overestimation of BrO could be a result of missing chemistry.
- The authors could also compare the tropospheric bromine column from the model to satellite observations. This would offer some validation of bromine chemistry.
- Considering that the process analysis would be biased by the above assumptions. How valid are the numbers in figure 12 and what is the uncertainty in the ozone loss numbers? The paper would benefit from a detailed discussion on the uncertainties in the model for future improvement considering the large sensitivity of the model to the boundary conditions, initial ozone concentrations, and uncertainties in the assumptions for the emissions.

Minor comments:

Line 16: 'In the Arctic, due to the lack of human activities, the tropospheric ozone remains at background level' – this statement is not strictly true since ozone concentrations in the Arctic have increased due to long-range transport. (Sharma et al., 2019)

Line 19 – define ppbv

Line 19 – …. commonly called ozone depletion events.

The reaction cycle (I) needs to be stoichiometrically balanced.

Line 28 - the total amount of Br and BrO keeps constant – rephrase to say that the total amount of bromine stays constant. 'bromine species play as catalysts' is not correct, both the species are not catalysts but rather bromine is.

Line 43: major oxidant in the atmosphere shifts from ozone to – ozone is not the major oxidant, but OH is.

Line 47: … the internal relationship…

Line 57 - … and a high BrO …

Line 75 and 110: …which considers the halogen chemistry.

Line 110: originally instrumented used/implemented in CMAQ

Line 114: Please give the webpage as a citation or include the list of reactions in the supplementary text.

Line 125 - For the ozone in the boundary layer

Line 150 – The authors used ppb and ppbv interchangeably – please use only one form.

The language throughout the paper needs to be improved for grammatical errors. Several sections are missing or using too many articles and/or singular words have been used instead of plural words. My suggestion is that either the journal helps with the language editing, or the authors use one of the several available services to improve the writing.

Line 186: 'It enables us for the subsequent analysis' – please rephrase.

Line 187- '…possibly due to the uncertainty in boundary conditions'. The mismatch could also be due to poor simulation of bromine chemistry or errors in reaction rates assumed for k_{R3}.

Please include the loss of bromine to deposition in figure 13.

References:

Benavent, N., Mahajan, A.S., Li, Q., Cuevas, C.A., Schmale, J., Angot, H., Jokinen, T., Quéléver, L.L.J., Blechschmidt, A., Zilker, B., Richter, A., Serna, J.A., Garcia-nieto, D., Fernandez, R.P., Skov, H., Dumitrascu, A., Pereira, P.S., Abrahamsson, K., Bucci, S., Duetsch, M., Stohl, A., Beck, I., Laurila, T., Blomquist, B., Howard, D., Archer, S.D., Bariteau, L., Helmig, D., Hueber, J., Jacobi, H.-W., Posman, K., Dada, L., Daellenbach, K.R., Saiz-Lopez, A., 2022. Substantial contribution of iodine to Arctic ozone destruction. Nat. Geosci. https://doi.org/10.1038/s41561-022-01018-w

Sharma, S., Barrie, L.A., Magnusson, E., Brattström, G., Leaitch, W.R., Steffen, A., Landsberger, S., 2019. A Factor and Trends Analysis of Multidecadal Lower Tropospheric Observations of Arctic Aerosol Composition, Black Carbon, Ozone, and Mercury at Alert, Canada. J. Geophys. Res. Atmos. 124, 14133–14161. https://doi.org/10.1029/2019JD030844