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Supplement of

Impact of biogenic very short-lived bromine on the Antarctic ozone hole during the 21st century

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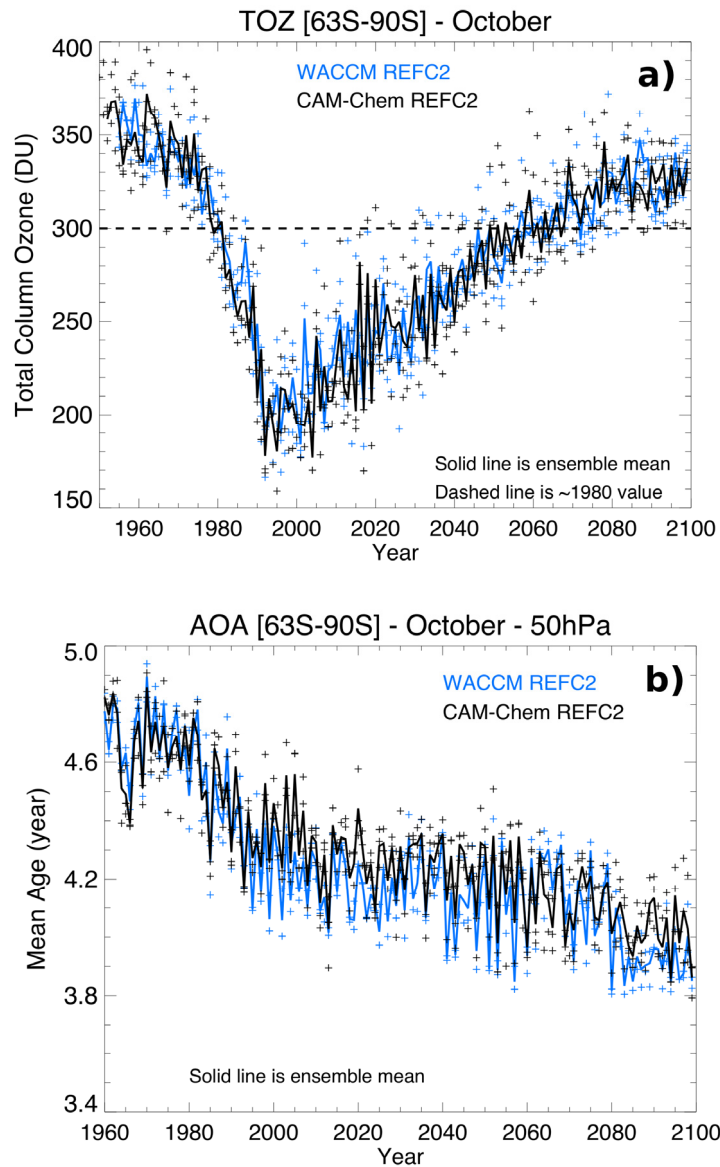
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2 **1 Validation of CAM-Chem in the stratosphere**

3 CAM-Chem, as well as WACCM, were part of CCMVal-2 and so were included in many of
4 the papers comparing the evolution of stratospheric ozone (Eyring et al., 2010a) as well as the
5 model sensitivity to different greenhouse scenarios (Eyring et al., 2010b). More recently, both
6 CAM-Chem and WACCM participated in the CMIP5 inter-comparison project, computing
7 stratospheric ozone interactively (Eyring et al., 2013a). Note that for those studies an identical
8 geographical and altitude configuration as the one described here was used, and CAM-Chem
9 return dates estimations is behaving very much in the middle of the simulated return periods
10 of the multi-model range (see Fig.1 in Eyring et al., (2010a)).

11 Lamarque et al. (2008) showed that even when CAM has a relatively low model top (~40
12 km), the model shows good ability at reproducing a variety of large- scale changes in climate
13 and chemical composition in the stratosphere when forced with the observed sea-surface
14 temperatures and surface concentrations of long-lived trace gases and ozone-depleting
15 substances. The model upward propagation of gravity waves (GW) due to the existence of a
16 positive anomaly in the zonal wind distribution at mid-latitudes has the effect of increasing
17 the momentum deposition associated with the GW, and indicates the presence of enhanced
18 residual circulation (see Fig. 18 in Lamarque et al., (2008)). Additionally, (Lamarque and
19 Solomon, 2010) analysed the role of long-term increases in CO₂, SST and halocarbons in
20 explaining the observed trend of ozone in the tropical lower stratosphere using CAM-Chem
21 (v3), and compared the model performance against WACCM (see their Fig. 1, vertical
22 distributions of the tropical vertical velocity).



1

2 **Figure S1: Comparison between CAM-Chem (black) and WACCM (light-blue) performance in**
 3 **the stratosphere for REFC2-CCMI simulations including the ~5 pptv additional VSL^{Br}**
 4 **contribution: A) Total ozone column averaged within the southern polar cap (TOZ^{SP}) during**
 5 **October; B). Mean Age of Air (AOA) at 50 hPa during October. CAM-Chem output correspond**
 6 **to the ensemble mean of three independent realizations (*sim*⁰⁰⁴, *sim*⁰⁰⁵ and *sim*⁰⁰⁶), while**
 7 **WACCM results correspond to a unique simulation. Note that the expected return date to 1980**
 8 **Ozone levels is approximately the same for the two models.**

9 CAM-Chem updates since WMO-2010 helped to improve the model performance. The
 10 implementation of a non-orographic gravity wave scheme for convection and fronts
 11 (originally developed for WACCM), as well as an inertia-gravity wave (IGW)
 12 parameterization, reduced stratospheric polar temperatures (which were biased warm) and
 13 increased chlorine activation and vortex size. As the limited vertical resolution (compared to

1 WACCM) does not allow the internal computation of the quasi-biennial oscillation (QBO),
2 the QBO is imposed by relaxing equatorial zonal winds to the observed inter-annual
3 variability. Additionally, stratospheric aerosol and surface area density data has been updated
4 to the common observation-derived dataset for the CCMI project (Eyring et al., 2013b;
5 Hegglin et al., 2014). A complete validation of current CAM-Chem version, focused on
6 tropospheric issues but including total ozone column as well as stratospheric dynamics, is
7 given in (Tilmes et al., 2016; see Figs. 2, 5 and 8).

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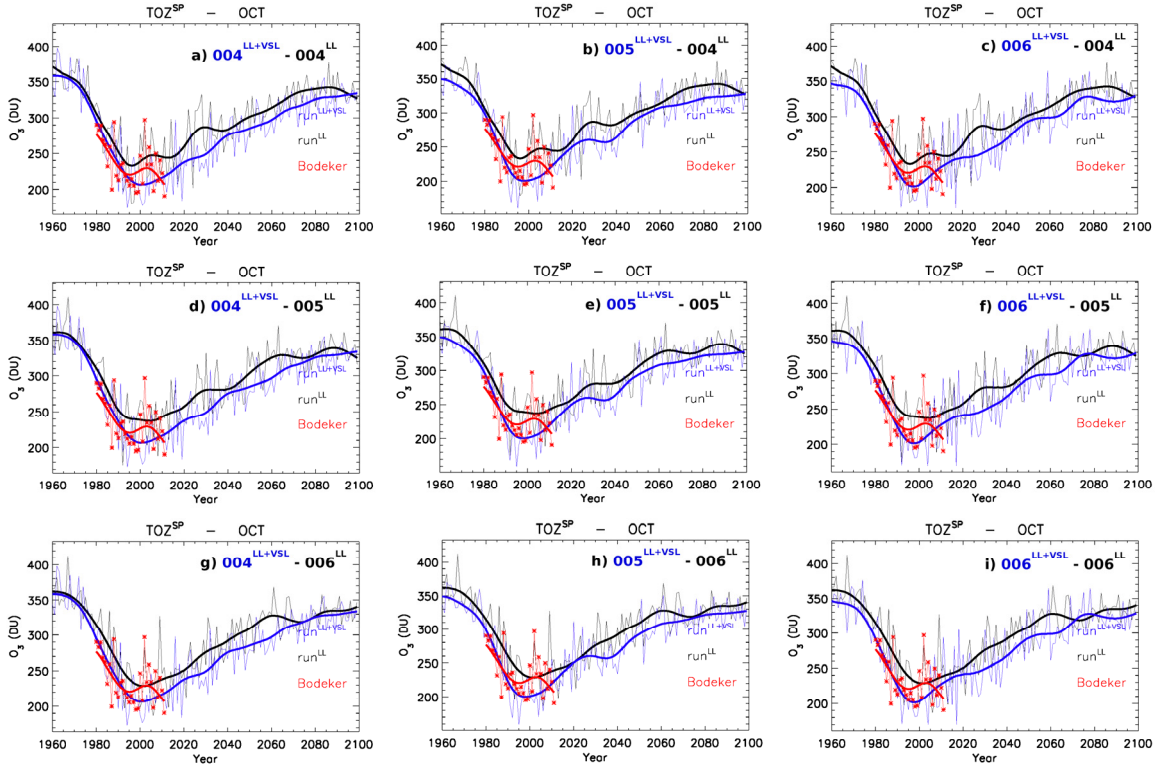
7 **2 Results for individual ensemble members**

8 Figures S2-S3 show equivalent plots to Fig.2A and 2B in the main text, but considering each
9 individual ensemble member instead of the ensemble mean. As all setups are independent one
10 of the other, we present a 9-panel figure where each of the run^{LL} realizations (004^{LL} , 005^{LL}
11 and 006^{LL}) is compared against all of the run^{LL+VSL} simulations (004^{LL+VSL} , 005^{LL+VSL} and
12 006^{LL+VSL}), and viceversa. In all cases, the monthly-mean output for October, as well as a
13 smoothed curve considering an 11-year hamming window is shown. Return dates values
14 shown in Table 1 of the main text were extracted from these panels.

15 Figure S4 show equivalent plots to Fig. 5 in the main text, but includes the non-smoothed
16 data. In this case, the daily output of the Total Ozone Colum was used to compute the Ozone
17 Hole Area (OHA) and Ozone Mass Deficit (OMD) for each simulation, and the monthly
18 mean for October was computed from the daily data. Only 3 of the 9 possible comparisons of
19 independent ensemble members are shown.

20 The smoothed TOZ^{SP} and OHA timeseries present a large-scale oscillation that appears
21 randomly for each independent simulation, which introduces local maxima and/or minima at
22 different periods of time. Even when the oscillations are reduced when the ensemble mean is
23 computed, they still appear when the differences between sim^{LL+VSL} and sim^{LL} are computed
24 (as well as when any couple of independent simulations are considered). We've performed
25 different types of smoothing (moving average, hamming filter, etc.) and/or variable window
26 widths (between 5 and 20 years) to perform the fit, and found no dependence on the filter nor
27 the smoothing window used. We've assigned these random oscillations to the intrinsic free-
28 running model variability between the individual ensemble members, and suggested
29 increasing the number of realizations and/or using other chemistry-climate models in order to
30 reduce the uncertainties.

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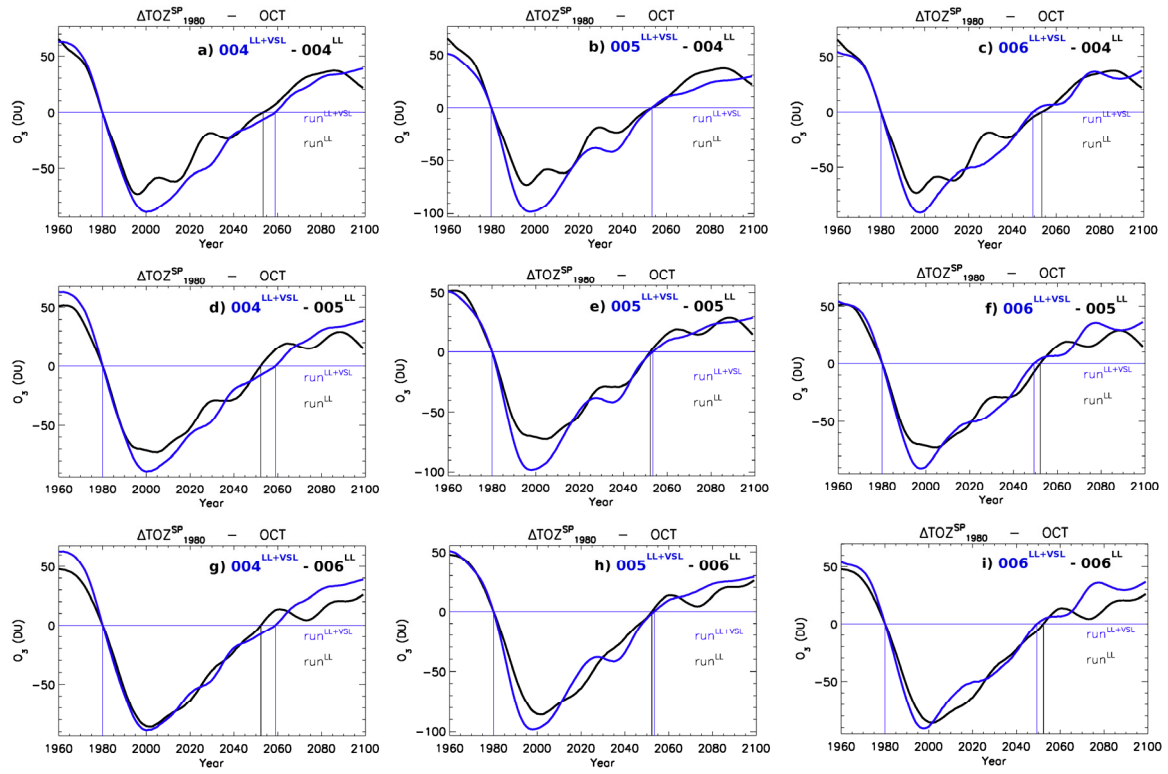


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Figure S2: Temporal evolution of the absolute total ozone column averaged within the southern polar cap (TOZ^{SP}) during October. The monthly TOZ^{SP} mean for each independent ensemble (thin lines) as well as the 11-years smooth timeseries (thick lines) is shown in blue for run^{LL+VSL} and black for run^{LL} . Red lines and symbols show merged satellite and ground base measurements from the Bodeker database averaged OCT within the same spatial and temporal mask as the model output. Equivalent results for the model ensemble mean are shown in Fig. 2A of the main text.

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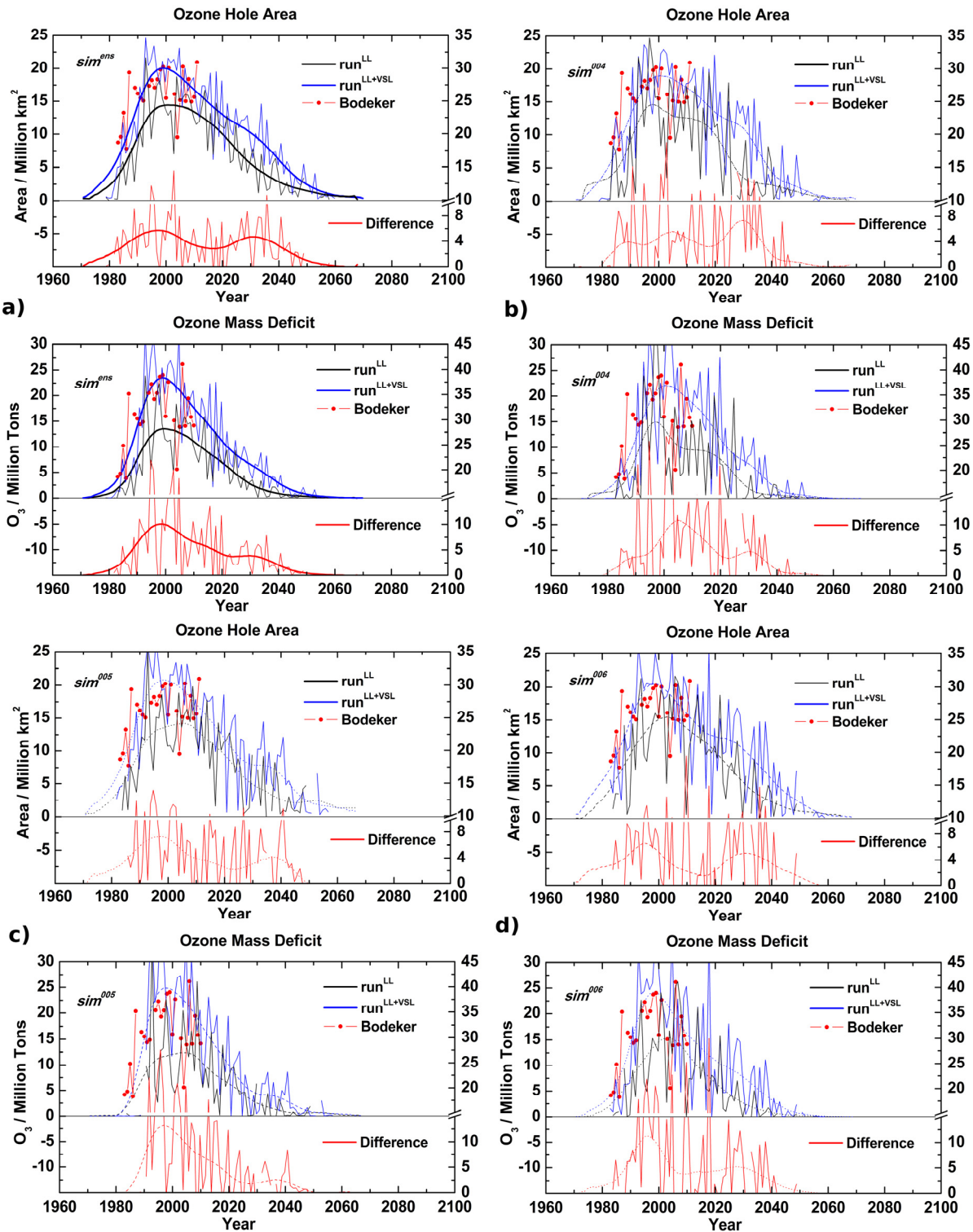


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Figure S3: Temporal evolution of the total ozone column relative to October 1980 ($\Delta\text{TOZ}^{\text{SP}}_{1980} = \text{TOZ}^{\text{SP}}_{\text{year}} - \text{TOZ}^{\text{SP}}_{1980}$). The zero horizontal line indicates the October $\Delta\text{TOZ}^{\text{SP}}_{1980}$ column for each experiment, while their respective return dates to 1980 are shown by the vertical lines. Equivalent results for the model ensemble mean are shown in Fig. 2B in the main text.

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2 **Figure S4: Temporal evolution of the ozone hole area (top) and ozone mass deficit (bottom) for**
 3 ***run^{LL}* (black) and *run^{LL+VSL}* (blue) simulations (left axis), as well as the difference between runs**
 4 **(red, right axis). Thin lines show the October monthly mean value for each independent**
 5 **simulation, while the thick/dotted/dashed lines show the smoothed curve considering an 11-year**
 6 **Hamming window for a) *sim^{ens}*; b) *sim⁰⁰⁴*; c) *sim⁰⁰⁵*; and d) *sim⁰⁰⁶*.**

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