Atmospheric
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Supplement of

Assessing the relative impacts of satellite ozone and its precursor observations to improve global tropospheric ozone analysis using multiple chemical reanalysis systems

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S1 Validation using monthly ozonesonde observations

To complement Figure 5, we compares monthly mean partial ozone column with ozonesonde observations (Fig. S1). In the upper troposphere and lower stratosphere (UTLS), control runs fail to capture seasonal cycles in partial column with large positive and negative biases, especially for the southern high latitudes. In the middle and lower troposphere, control runs captures seasonal cycles well, but underestimated the observed partial ozone during summer of both hemispheres for many cases, except for the tropical lower troposphere. The MOCAGE model shows mostly constant partial ozone over the southern mid and high latitudes.

Data assimilation substantially improved model biases and seasonal cycles in parital ozone clumn in the UTLS for all the reanalysis products. In the middle troposphere, negative biases during summer of both hemispheres were improved for most cases, except for IASI-r over the southern high latitudes. In the lower troposphere, data assimilation reduced model biases for many cases, whereas their seasonal cycles were not clearly improved.

S2 Ozone analysis in GEOS-Chem adjoint with the OMI NO_2 DOMINO porduct

Figure S2 presents ozone concentrations obtained from the data assimilation analysis, the control run, and their differences in the GEOS-Chem adjoint calculation with OMI NO_2 DOMINO product. The GEOS-Chem using the DOMINO product-based NO_x emission shows the increases in ozone concentrations by up to 1% in the lower troposphere over the globe and in the middle troposphere over the mid and high latitudes of both hemisphere, compared to the control run. In contrast, the ozone concentration analysis was decreased by up to 5% in the tropical free troposphere.

Compared to the GEOS-Chem adjoint with the NASA standard product (Fig. 1), the assimilation of the DOMINO product show larger impacts of data assimilation on the ozone analysis. This difference can be attributed to the top-down estimates of surface NO_x emissions derived from different retrieval algorithms (Table S1).

## S3 Top-down NO_x emission comparison between TCR-2 and GEOS-Chem adjoint

Table S1 summarizes a priori and posteriori surface NO_x emissions derived from the TCR-2 and GEOS-Chem adjoint systems. Compared to TCR-2, GEOS-Chem adjoint used higher NO_x emissions as a priori, probably because different soil NO_x emissions obtained from the Global Emission Inventory Activity (GEIA) (Graedel et al., 1993) and calculated by the online scheme developed by Yienger and Levy II (1995) for TCR-2 and GEOS-Chem adjoint, respectively.

The data assimilation increment in global surface NO_x emissions was much smaller in GEOS-Chem adjoint with the NASA standard product (-0.34 Tg N yr^{-1}) than that in TCR-2 ($+8.21$ Tg N yr^{-1}) and GEOS-Chem adjoint with the DOMINO product ($+9.16$ Tg N yr^{-1}). This smaller NO_x increment leads to much less impacts of data assimilation on ozone analysis in GEOS-Chem adjoint with the NASA standard product. The difference between two GEOS-Chem adjoint calculations was attributed to systematic differences between the OMI NO_2 NASA standard and DOMINO products, especially for vertical

sensitivity (i.e., averaging kernels), by Qu et al. (2020). Differences in a prior emissions, assimilated satellite observations, and model processes can also affect the diverse impacts on ozone analysis in TCR-2 and GEOS-Chem adjoint.

Table S1. Global and annual total NO_x emissions in 2010 (Tg N yr^{-1}. The emissions were obtained from the TCR-2 and GEOS-Chem adjoint systems. "Prior" presents a priori emissions used in individual systems. "Posteriori" indicates top-down estimates of NO_x emissions. The satellite retrieval products were shown in the parentheses.

Reanalysis	NO_xemission [Tg N /yr]	
TCR-2	Prior	46.58
	Posteriori (QA4ECV)	54.69
GEOS-Chem adjoint	Prior	52.20
	Posteriori (NASA)	51.86
	Posteriori (DOMINO)	61.36
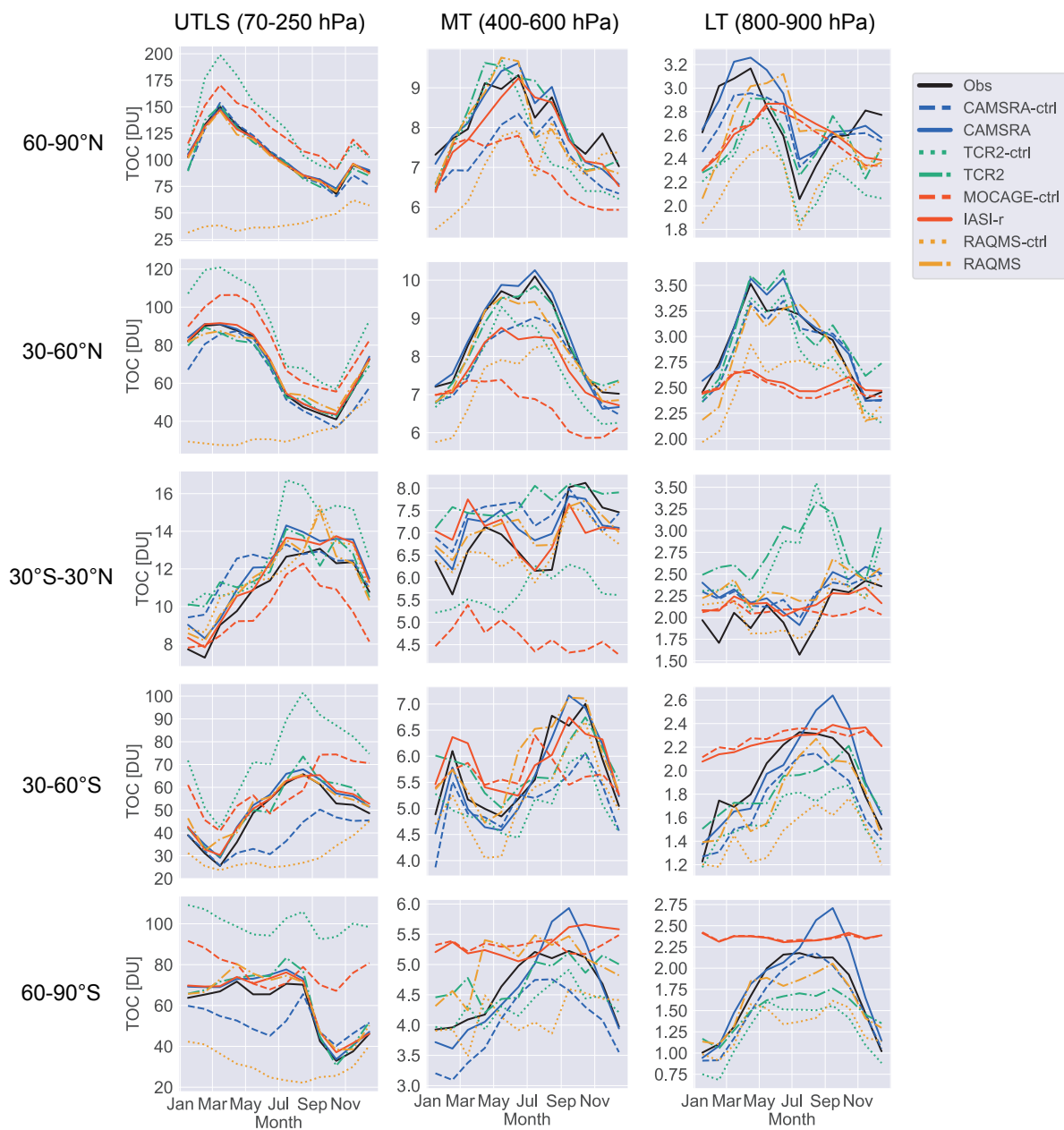
Figure S1. Monthly timeseries of partial ozone column obtained from the ozonesonde observations, the reanalysis products, and the control simulations in 2010. The first through fifth rows present the northern high latitudes (60–90°N), the northern mid-latitudes (30–60°N), the tropics (30°S–30°N), the southern mid-latitudes (30–60°S), and the southern high latitudes (60–90°S) The first through third columns are the upper troposphere and lower stratosphere (70–250 hPa), the middle troposphere (400–600 hPa), and the lower troposphere (800–900 hPa), respectively. The blue line indicates CAMSRA, the red line indicates IASI-r, the green line indicates TCR-2, the orange line indicates the RAQMS Aura reanalysis, and the black line indicates observations. The solid and dashed-dotted lines indicate the reanalysis products, while the dashed and dotted lines indicate the control simulations. The unit is Dobuson Unit (DU).

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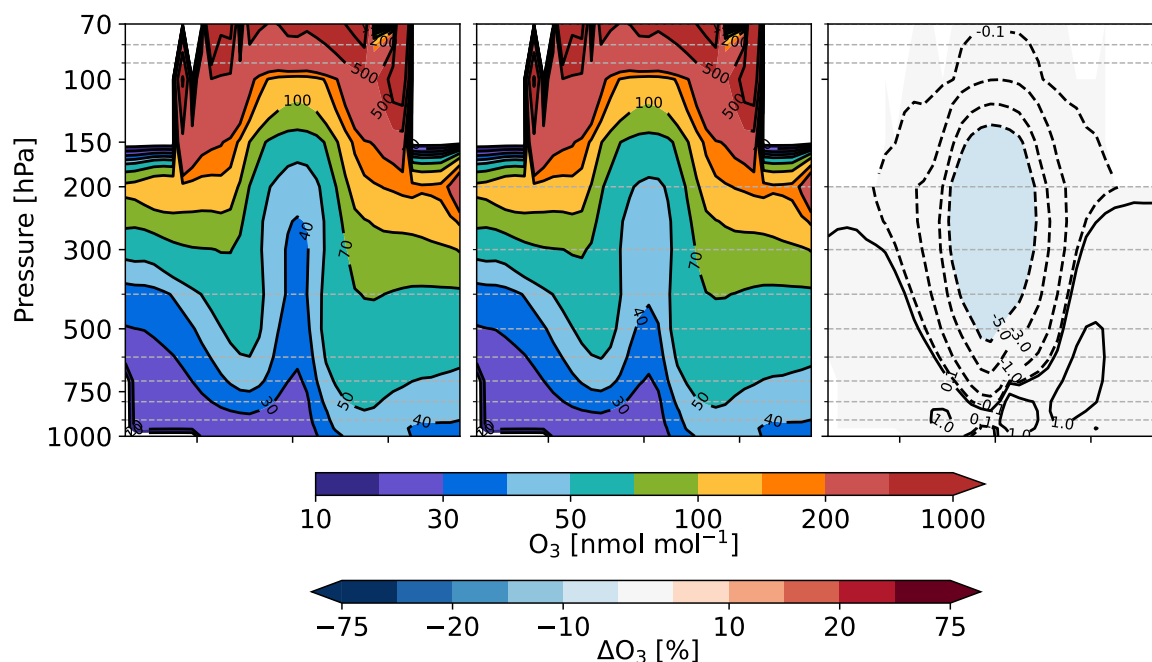
Figure S2. Latitude-pressure level cross sections of annual mean tropospheric ozone concentrations obtained from reanalysis products (left), control simulations (middle), and their differences (right) in 2010 obtained from the GEOS-Chem-adjoint calculation with the OMI NO_2 DOMINO product. The units of ozone concentrations and the differences between reanalysis products and control simulations are mole fraction in air (nmol mol^{-1}) and percentage (%), respectively.

References

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