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Quantifying chemical ozone loss in the Arctic stratosphere with GEOS-STRATCHEM Data Assimilation System

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SUMMARY

A faithful representation of polar stratospheric chemistry in models and its connection with dynamical variability is essential for our understanding of the evolution of the ozone layer in a changing climate and during the projected continuing decline of ozone depleting substances in the atmosphere. We use a new configuration of the Goddard Earth Observing System Data Assimilation System with a stratospheric chemistry model to study ozone depletion in the Arctic polar stratosphere during the exceptionally cold (in the stratosphere) winters 2015/2016 and 2010/2011.

BACKGROUND

Stratospheric ozone layer shields Earth's biosphere from harmful ultraviolet radiation and greatly impacts the thermal structure of the stratosphere, strongly affecting the climate. A growing number of recent studies suggest that the full inclusion of realistic ozone in atmospheric model simulations may help to improve both seasonal weather predictions and climate projections.

The best-known and most severe manifestation of the 20th century stratospheric ozone loss is the seasonal occurrence of ozone holes over Antarctica. Although to a lesser degree, the same chemical mechanism leads to springtime ozone depletion over the Arctic. The main pathway of ozone destruction in polar regions involves conversion of chlorine reservoir species, chlorine nitrate ($ClONO_2$) and hydrogen chloride (HCl), into active chlorine compounds, ClO_x, from which chlorine atoms are released in the presence of sunlight. Chlorine then catalytically destroys ozone. The conversion to ClO_x takes place on surfaces of small particles of polar stratospheric clouds (PSCs). This process depends on three conditions:

- The presence of chlorine compounds in the stratosphere
- Low temperature allowing the formation of PSCs
- Isolation of the air mass where the reactions take place

Stratospheric chlorine loading is still high, although it is declining following the implementation of the Montreal Protocol. Low temperatures and air isolation are attained within the stratospheric polar vortex, a large-scale cyclonic wind pattern that forms over the pole during wintertime and lasts through the first half of spring. When the vortex is weak, its temperature is elevated inhibiting PSC formation and strong mixing disperses ozone-depleting compounds at the same time replenishing the vortex ozone. On the other hand, a strong vortex acts in a manner similar to a laboratory beaker in which chlorine activation and ozone depletion take place with little mixing with the midlatitudinal ozone-rich air masses. The severity of polar stratospheric ozone loss strongly depends on the temperature and strength of the polar vortex, which exhibits strong interannual variability. The 2015-2016 winter was one of the coldest on record in the Arctic polar stratosphere. This led to high levels of chlorine activation and rapid ozone depletion until the first week of March, when the vortex broke down several weeks earlier than usually preventing further chemical destruction of ozone. The purpose of this work is twofold:

- To estimate the 2015/2016 Arctic ozone loss
- To test the Goddard Earth Observing System (GEOS) Data Assimilation System (DAS) with full stratospheric chemistry for future atmospheric reanalyses.

Experiment and Results

A new configuration of GEOS-DAS is used to study this ozone loss. Ozone evolution is computed with STRATCHEM, a stratospheric chemistry module that simulates over 160 chemical reactions among 34 transported and 17 derived species and contains a PSC scheme. STRATCHEM replaces the parameterized chemical module normally used in GEOS-DAS experiments, including multidecadal reanalyses. The observations assimilated by the system consist of radiance data from satellite-borne sensors, conventional measurements and ozone observations. The latter are obtained from two instruments: the Microwave Limb Sounder (MLS) and the Ozone Monitoring Instrument (OMI), both onboard NASA's EOS Aura satellite. Combining the real-world stratospheric meteorology constrained by observations with a chemistry model enables the production of a global three-dimensional representation of the atmospheric composition and its evolution during a specified period that can be readily evaluated against satellite data.

Figure 1 shows comparisons of the vortex-averaged GEOS ozone, HCl and chlorine monoxide (ClO) with data from the MLS instrument. The data shown are from the 480-K potential temperature surface, which corresponds to about 20 km above the ground. The excellent agreement between the modeled and MLS ozone is not surprising because MLS ozone data are assimilated in this experiment. But there is also a good qualitative agreement between the GEOS and MLS HCl, which is not assimilated. The slower decline of the model HCl concentrations likely resulted from deficiencies in the GEOS transport. This conclusion is supported by additional analysis of the GEOS nitrous oxide indicating that downward transport of air inside the vortex is underestimated in GEOS. Precise estimation of transport errors and strategies to improve the model performance are areas of active research in the GMAO. While transport issues affect the atmospheric composition and contribute to the estimated errors of the analysis, they also provide an insight into the performance of the model and the assimilation scheme and help to map out future improvements of both. Despite this shortcoming, the ClO concentrations from GEOS follow MLS very closely except in March when the model generates a high bias in ClO, indicative of a slightly delayed chlorine deactivation. This too is likely a consequence of model transport errors but can also be related to errors in the chemistry model that require further investigation. We note that the uncertainties in the ozone behavior in GEOS that result from these deficiencies have been estimated and included as error bars in the final analysis in Figure 3. Since ClO is directly responsible for ozone depletion, this close agreement during most of the winter provides confidence in the model performance with respect to ozone loss.

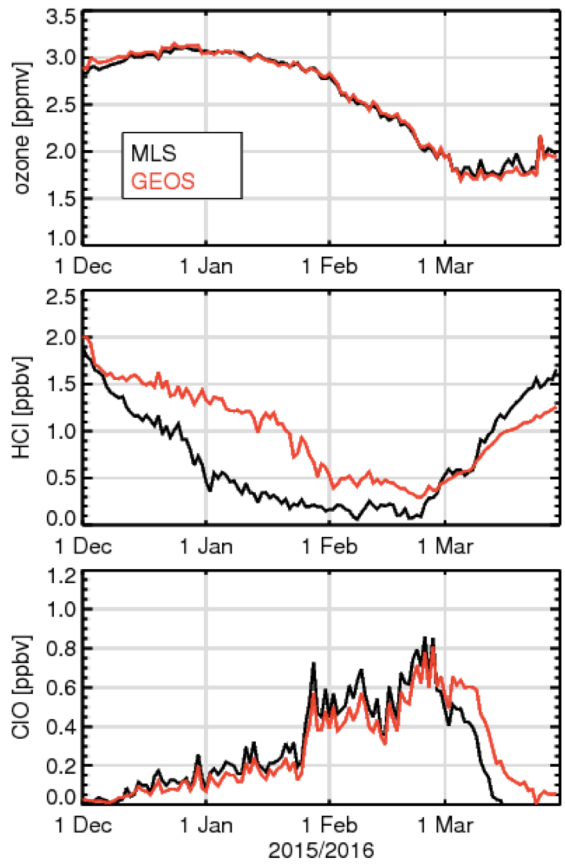

Figure 1. Time series of vortex-averaged ozone (top), HCl (middle), and ClO (bottom) at the 480-K potential temperature surface (~20 km) from MLS (black) and GEOS (red). Ozone is shown in parts per million by volume (ppmv) and the other species in parts per billion by volume (ppbv).

The GEOS model writes out chemical tendencies for ozone, i.e. ozone's rate of change due to chemistry alone, other processes excluded. This allows us to estimate the evolution of the ozone loss rate and the cumulative chemical ozone change during the winter-spring. The time series of the vortex-averaged chemical ozone tendencies in parts per billion by volume (ppbv) per day is shown in Figure 2 (bottom left panel) as a function of potential temperature, a convenient vertical coordinate for stratospheric studies. Here, the vertical range is 400 K to 800 K, corresponding to heights between about 16 km and 30 km. The top panel shows the pole temperature at ~20 km and the fraction of the polar vortex illuminated by sunlight. As the sunlit fraction reaches about 10% in mid-January, the rate of chemical ozone loss accelerates to about 20 ppbv/day and maximizes in the end of February at 40 ppbv/day. Three distinct maxima correspond to increases in the pole temperature in mid-January, the beginning and the end of February. These jumps indicate slight dynamical shifts of the position of the polar vortex that slides off the pole exposing larger portions of its air mass to sunlight, which accelerates photochemical

reactions that release atomic chlorine and strongly depend on solar illumination. The chlorine-catalyzed ozone depletion is limited to elevations below about 26 km (potential temperature of about 640 K). The ozone loss above that level is due to reactions with nitrogen oxides and is replaced by rapid photochemical production in March.

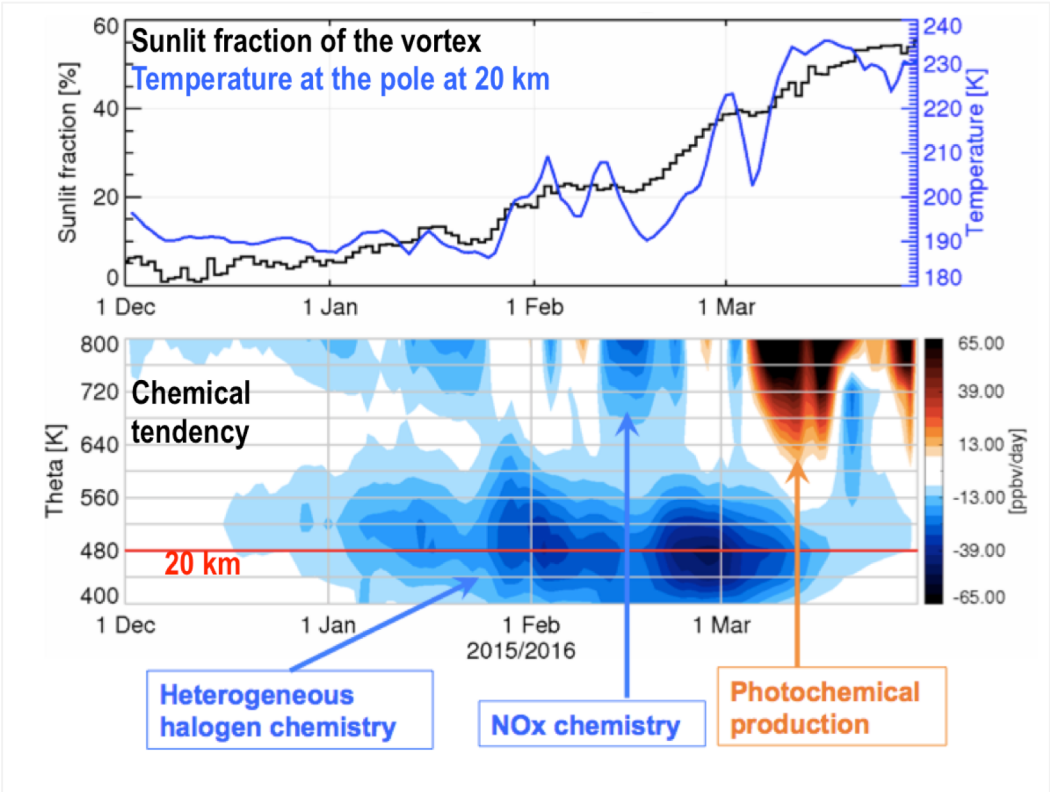

Figure 2. *(top)* The percentage of the 480-K polar vortex area illuminated by sunlight as a function of day (1 December 2015 to 30 March 2016) is shown in black. The 480-K temperature at the pole is plotted in blue. *(bottom)* The vortex-averaged daily ozone tendencies due to chemistry in ppbv per day.

Figure 3 shows the cumulative ozone loss due to chemistry between 1 December 2015 and 31 March 2016. The maximum loss of 2.1 ppmv occurred at 480 K (~20 km). This corresponds to 66 % of the vortex ozone on 1 January, a significant amount, compared to most Arctic winters. The same panel shows the cumulative loss during the record breaking winter 2010/2011. Note that while the maximum depletion then was 3.25 ppmv, the cumulative loss computed only up to 12 March 2011 (the approximate date when depletion was terminated in 2016) was about the same as 2016. This result indicates that the 2015/2016 winter was likely on track to match the extreme ozone loss

seen in 2011, an outcome precluded only by the unusually early breakup of the lower-stratospheric polar vortex around 12 March 2016. The error bars around the loss curve are estimated from the differences between six-hourly ozone forecasts and satellite data calculated within the data assimilation system.

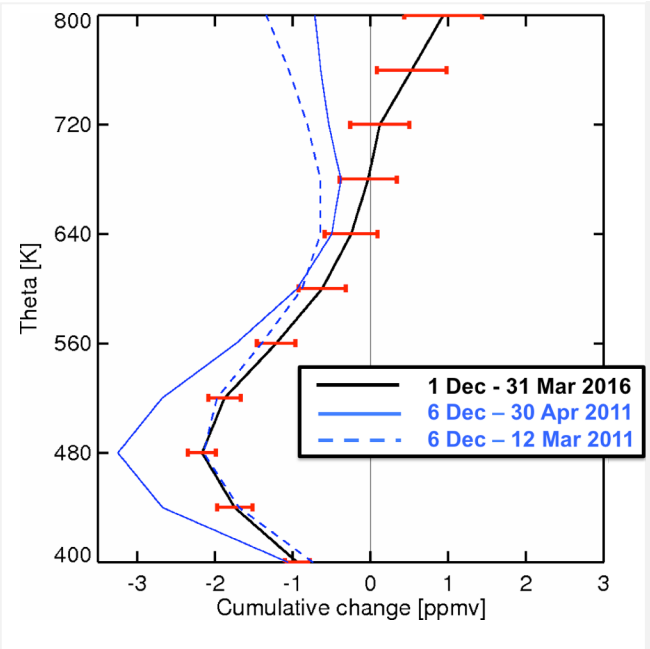

Figure 3. Time integrated (December-March) tendency from Figure 2 in ppmv (black), the same for 6 December 2010– 30 April 2011 (solid blue) and for 6 December 2010 – 12 March 2011 (dashed blue). The bars indicate an error envelope estimated from assimilation of ozone data.

We now turn to the spatial distribution of the chlorine compounds and ozone in the context of the stratospheric dynamics. Figure 4 shows maps of the GEOS chlorine reservoir species, active chlorine and ozone on the 480-K potential temperature surface (approximately 20 km above the Earth's surface) over the North Pole on 11 January at the beginning of ozone depletion and on 21 March 2016 after the breakup of the polar vortex. The maps are generated from the GEOS output. The top row shows the situation in the mid-winter: low concentrations of chlorine nitrate and HCl combined with high quantities of ClO_x within the polar vortex indicate chlorine activation but before significant ozone loss commences when a larger portion of the vortex becomes illuminated by sunlight. In the bottom row (over two months later) there is only a remnant of the polar vortex left, chlorine is deactivated ($ClONO_2$ has elevated concentrations, ClO_x is nearly gone) and very low ozone values, the result of earlier chemical loss, are seen inside the vortex remnant. Note the consistency of chemistry and dynamics in GEOS: sharp gradients of species'

concentrations are aligned with the edge of the polar vortex and the chemical composition inside the vortex is very different than in the rest of the hemisphere. An animated version of these, and some additional maps at the 500-K potential temperature level is available at https://gmao.gsfc.nasa.gov/animations/O3loss_arctic_strat.php.

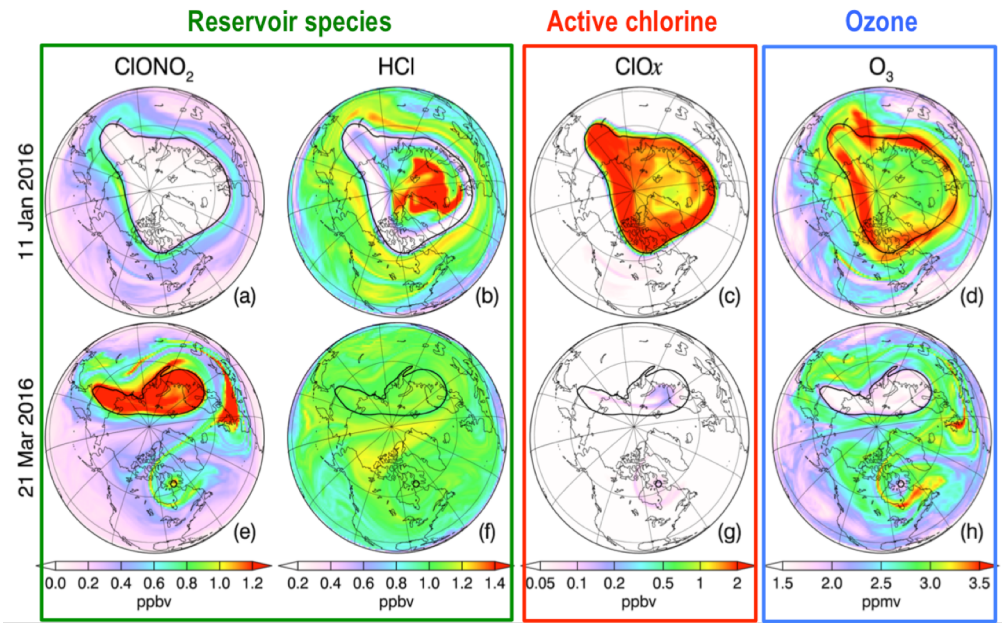

Figure 4. Maps of modeled $ClONO_2$ (a and e), HCl (b and f), ClO_x (c and g), and ozone (d and h) on the 480-K potential temperature surface on 11 January 2016 (a-d) and 21 March 2016 (e-h). The black contour is the edge of the polar vortex.

Finally, we compare the 2015/2016 and 2010/2011 ozone loss within a deep layer where most of the polar ozone resides. Figure 5 shows the cumulative change in the vortex ozone integrated between 200 hPa and 40 hPa (~12 km to 22 km). The results are shown in Dobson units (DU: 1 DU corresponds to 2.69×1020 ozone molecules per square meter within the specified layer). The cumulative loss on 10 March was about 80 DU in both winters. While the 2016 loss ceased quickly after the breakup of the polar vortex and did not exceed 90 DU, in 2011 the GEOS-estimated loss reached about 126 DU in April.

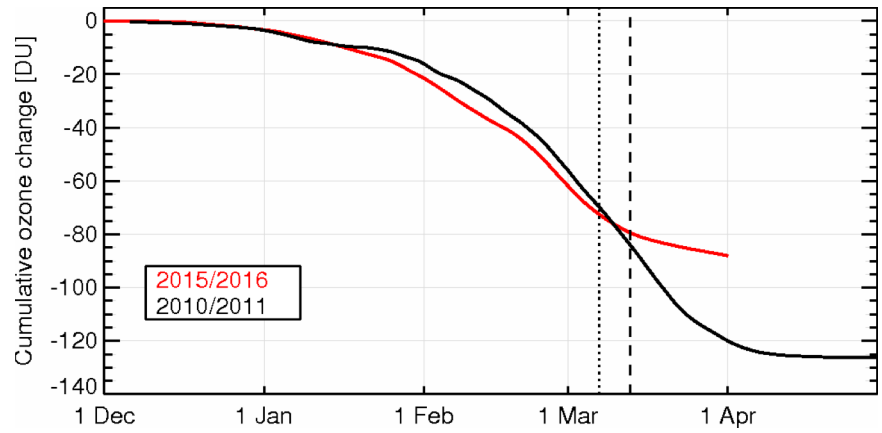

Figure 5. Cumulative vortex-averaged chemical ozone loss in the 200 hPa – 40 hPa column. Black: 2010/2011, red: 2015/2016. Ozone loss values are expressed in Dobson units. The dates of the 10 hPa zonal mean zonal wind reversal at 60°N (6 March) and the vortex breakup at 480 K (12 March) in 2016 are shown as a dotted and dashed line, respectively.

Conclusion

A new configuration of the GEOS Data Assimilation System with the STRATCHEM chemistry model allowed us to produce a detailed representation of polar stratospheric ozone chemistry during the exceptionally cold (in the stratosphere) 2015/2016 and 2010/2011 winters. The chemistry model simulates the evolution of a host of chlorine species consistent with the dynamics of the polar vortex. In particular, there is an excellent agreement between the vortex chlorine monoxide from GEOS and MLS data. In combination with data assimilation, the model allowed us to estimate the magnitude and temporal variations of chlorine-catalyzed ozone loss within the polar vortex and estimate its errors. We concluded that the maximum cumulative chemical ozone loss of 2.1 ppmv took place at about 20 km above the surface and was comparable to the record Arctic ozone depletion of 2011 up to the time of the early polar vortex breakup in early March. In 2011, the loss continued until the beginning of April.

These results underscore the value of including a full stratospheric chemistry model in future data assimilation-based atmospheric reanalyses but also help us identify deficiencies in transport of stratospheric constituents in GEOS, which will guide future investigations and model developments.

This work has been done in collaboration with our colleagues in the GMAO and other institutions and will be included in a peer-reviewed publication, Wargan et al. (2017) now in preparation

Reference

Wargan, K., J.E. Nielsen, G.L. Manney, M.L. Santee, N.J. Livesey, Z.D. Lawrence, S. Pawson and L. Coy, 2017: Chemical Ozone Loss During the 2015/2016 Arctic Winter in a Goddard Earth Observing System Assimilation Experiment with Stratospheric Chemistry. Submitted to *J. Geophys. Res*.