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Development of the adjoint of the unified tropospheric–stratospheric chemistry extension (UCX) in GEOS-Chem adjoint v36

Irene C. Dedoussi et al.

Correspondence to: Irene C. Dedoussi (icd23@cam.ac.uk)

S1 Development further specifics

Table S1 list the 37 active chemical tracers that have been added to the UCX adjoint model. Table S2 describes the updates to processes (modules) as well as structural changes implemented from the GEOS-Chem UCX standalone forward model.

Table S1 List of the 37 tracers added to the adjoint model, where CFCX is CFC113, CFC114, and CFC115, HCFCX is HCFC123, HCFC141b, and HCFC142b, and SO4g is gaseous H_2SO_4.

Br2	CHBr3	BrCl	HCFCX	H2402	ClOO
Br	CH2Br2	HCl	CFC11	Cl	OClO
BrO	CH3Br	CCl4	CFC12	ClO	Cl2
HOBr	N2O	CH3Cl	HCFC22	HOCl	Cl2O2
HBr	OCS	CH3CCl3	H1211	ClNO3	H2O
BrNO2	CH4	CFCX	H1301	ClNO2	SO4g
BrNO3					

Table S2: UCX additions descriptions

Module	Description
chemgrid	Chemgrid introduces structural changes to the code, by changing the vertical definition of the domain of the chemical solver, thus adding flexibility to the so far troposphere-focused GEOS-Chem model. The previously defined top of the chemistry grid as the top of the tropopause is now updated in all model modules to the top of the chemgrid. The full resolution (72 layer) vertical global grid is now used, as opposed to the default, reduced vertical resolution (47 layer) that the model is set up with.
Additional species	Thirty-seven new tracers, needed in the stratospheric chemistry calculations, have been added to the GEOS-Chem adjoint model. In order to produce an adequate and representative initial condition for the concentration of these tracers, values from the forward-model GEOS-Chem UCX model were used. A complete list of the added species is included in Table S1.
Long-lived species	This module contains routines and variables used to apply surface mixing ratio boundary conditions to the newly added long-lived species, and is needed for the UCX calculations.
Fast-JX	The photolysis scheme has been updated to Fast-JX which expands the spectrum analyzed to 18 wavelength bins covering 177-850 nm, and extends the upper altitude limit to approximately 60 km (Eastham et al., 2014). These are needed for stratospheric processes and are particularly relevant for the ozone chemistry in the stratosphere.

Unified Chemistry Extension	This module introduces the routines and variables associated with the full stratospheric chemistry, and uses subroutines from the aforementioned modules. The majority of the existing chemical mechanism is kept, with 217 kinetic and 43 photolytic reactions being added to reflect the stratospheric chemistry capabilities. The full list of added re- actions is provided in Table B of Eastham et al., 2014, matching those from Rotman et al., 2001 with updated rates from Sander et al., 2011. The Kinetic PreProcessor is used to automatically generate the chemical mechanism (Daescu et al., 2003; Damian et al., 2002; Sandu et al., 2003). In addition to the 'forward model' code, KPP also automatically differentiates the mechanism and generates the corresponding code for the adjoint part of the model (Sandu et al., 2003).

S2 Evaluation further specifics

Figure S1 presents the 2010 annually averaged NO_x mixing ratios in the standalone forward GEOS-Chem UCX model, the pre-UCX GEOS-Chem adjoint base model, and the base (forward) model of the GEOS-Chem UCX adjoint.

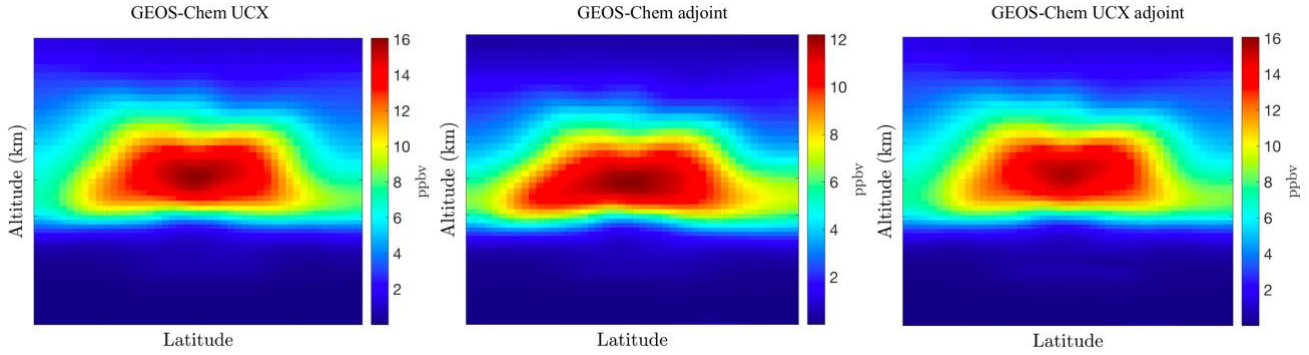

Figure S1 Annually averaged zonal NO_x mixing ratios (in ppbv) for 2010 in the standalone forward GEOS-Chem UCX model (left), the pre-UCX GEOS-Chem adjoint base (forward component) model (middle), and GEOS-Chem UCX adjoint the base (forward component) model (right).

S3 Altitude variation of sensitivity of O_x to NO_x

0-10 km 10-20 km 20-30 km

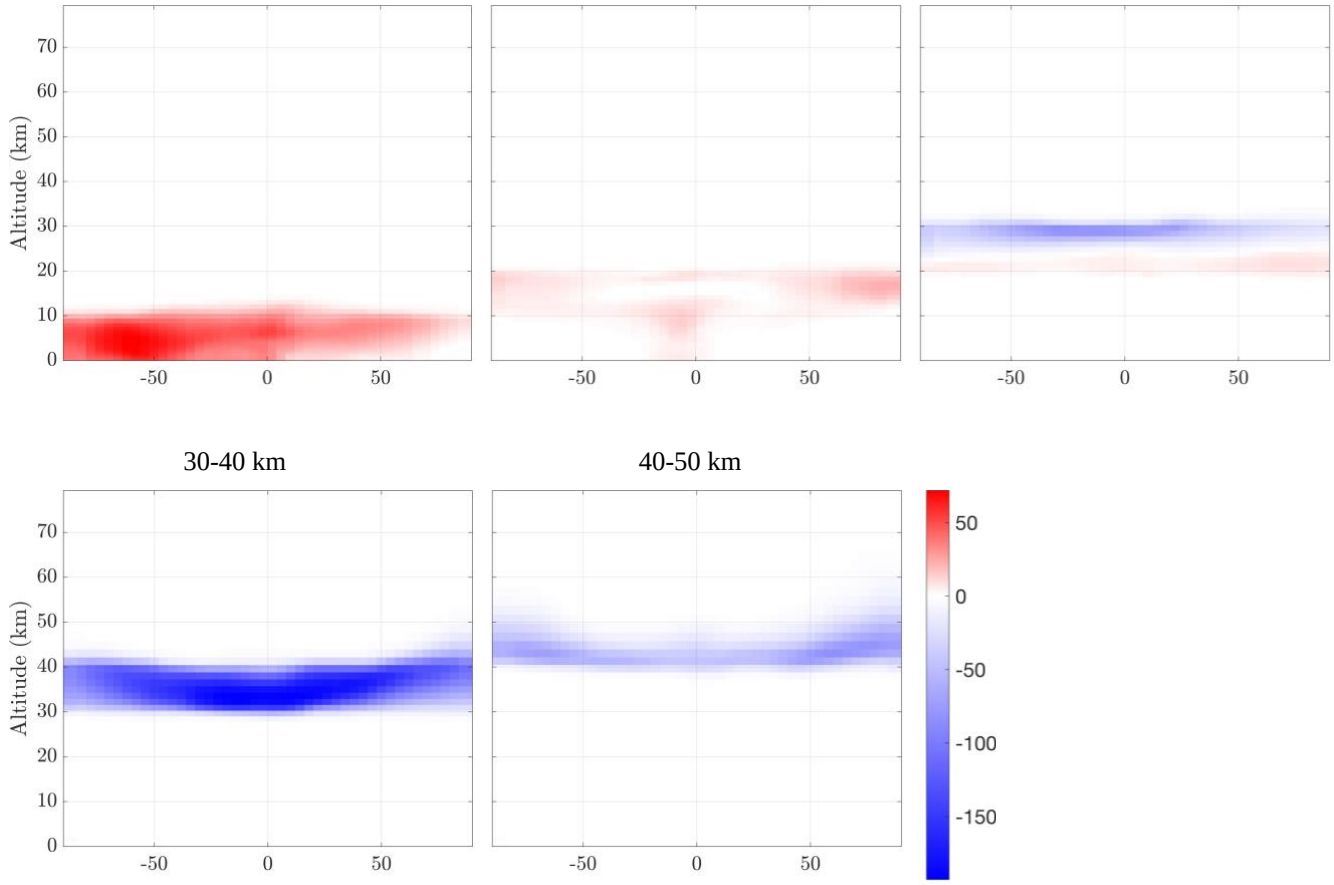

Figure S2: Zonal sensitivities of aggregate O_x at five stratospheric altitude bands with respect to perturbations in NO_x mass in all domain altitudes, in kg/kg for a two-week simulation in March. Altitudes 10-20 km and 20-30 km are presented in the main manuscript (with a different colorbar).

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