Adjoint inversion of Chinese non-methane volatile organic compounds sources using space-based observations of formaldehyde and glyoxal

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Abstract

We used the GEOS-Chem model and its adjoint to quantify Chinese non-methane volatile organic compound (NMVOC) emissions for the year 2007,constrained by spaced-based monthly mean columns of formaldehyde and glyoxal. The use of glyoxal observations in addition to formaldehyde observations helped differentiate NMVOC species and was essential in constraining aromatic emissions. Our best estimate for Chinese annual total NMVOC emission was 28.1 (20.9-32.7) Tg C y^{-1}, including 16.7 (13.2-19.6) Tg C y^{-1} from anthropogenic sources, 10.5 (6.9-12.7) Tg C y^{-1} from biogenic sources, and 0.9 (0.8-1.1) Tg C y^{-1} from biomass burning. Our top-down estimate for the Chinese annual emission of anthropogenic aromatics was 6.0 (5.0-7.3) Tg C y^{-1}, 26% higher than the estimate in the MIX-Asia inventory (4.7 Tg C y^{-1}). As a result of our updated NMVOC emission estimates, the simulated afternoon surface ozone concentrations over China increased by 11 ppbv in summer.

Methods: model and satellite data

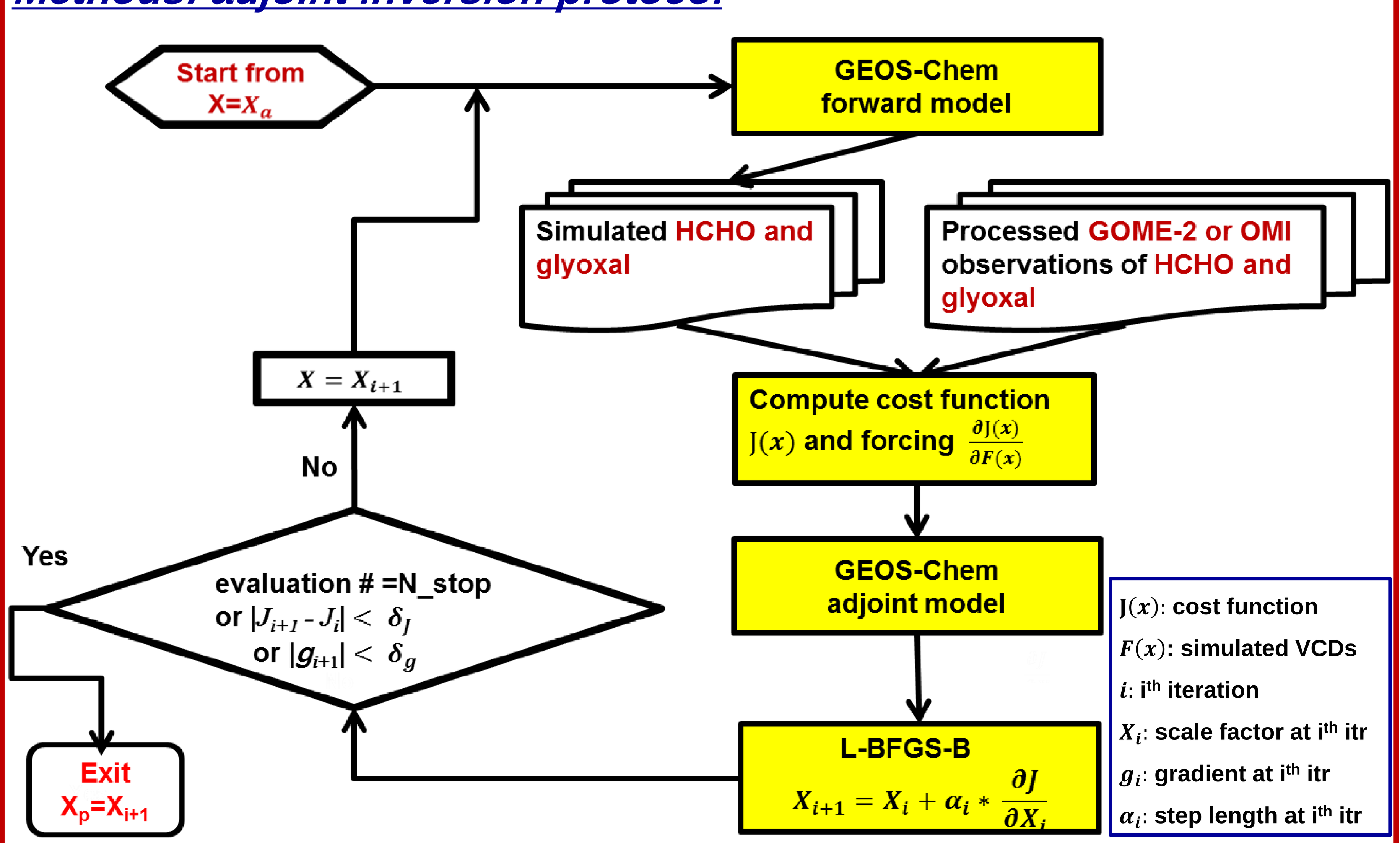
- We developed the dicarbonyl simulation adjoint for the GEOS-Chem chemical transport model (Henze et al., 2007; Fu et al., 2008)
- GOME-2 HCHO (De Smedt et al., 2012) and CHOCHO (Lerot et al., 2010) vertical columns in 2007
- OMI HCHO (Gonzalez Abad et al., 2015) and CHOCHO (Miller et al., 2015) vertical columns in 2007

Methods: inversion experiments

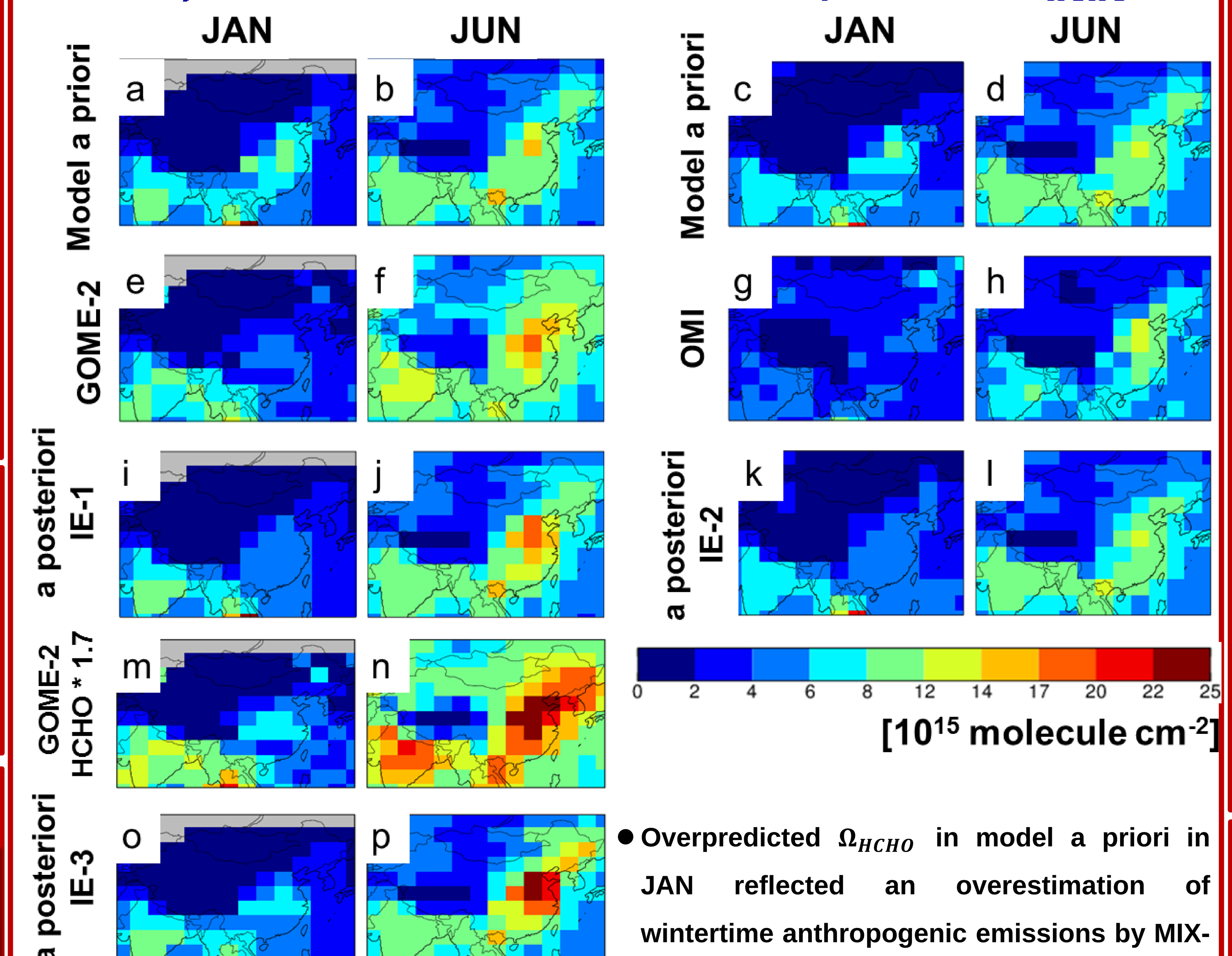
Inverse experiment	Satellite constraints and corresponding uncertainties	Anthropogenic[b]	Biogenic[c]	Biomass burning[d]	Total (Tg C y^{-1})
		A priori emission			
		15 (±200%) (4.7 for aromatics)	8.9 (±55%) (5.1 for isoprene)	0.8 (±360%)	24.4
		A posteriori emission (this work)			
IE-1	GOME-2 (±90% for HCHO, ±150% for glyoxal)	15.1 (5.7 for aromatics)	10.4 (6.8 for isoprene)	0.8	26.5
IE-2	OMI (±90% for HCHO, ±150% for glyoxal)	13.2 (5.0 for aromatics)	6.9 (3.4 for isoprene)	0.8	20.9
IE-3	GOME-2 HCHO * 1.7 (±90% for HCHO)	18.9 (6.0 for aromatics)	12.7 (8.8 for isoprene)	1.1	32.7
IE-4	OMI glyoxal (±150% for glyoxal)	19.6 (7.3 for aromatics)	11.8 (8.0 for isoprene)	1.0	32.4
Best estimate[a]		16.7 (13.2 ~19.6) 6.0 (5.0~7.3)	10.4 (6.9~12.7) 6.8 (3.4~8.8)	0.9 (0.8~1.1)	28.1 (20.9~32.7)

[a] Best estimate: average of 4 inversion experiments (IE-1, IE-2, IE-3, and IE-4). [b] The *a priori* anthropogenic emissions: MIX-Asia (Li et al., 2014). [c] The *a priori* biogenic emissions: MEGAN 2.1 (Guenther et al., 2006). [d] The *a priori* biomass burning emissions: GFED3 (van der Werf et al., 2010) plus doubled crop fire VOCs emissions from Huang et al. (2012).

Methods: adjoint inversion protocol

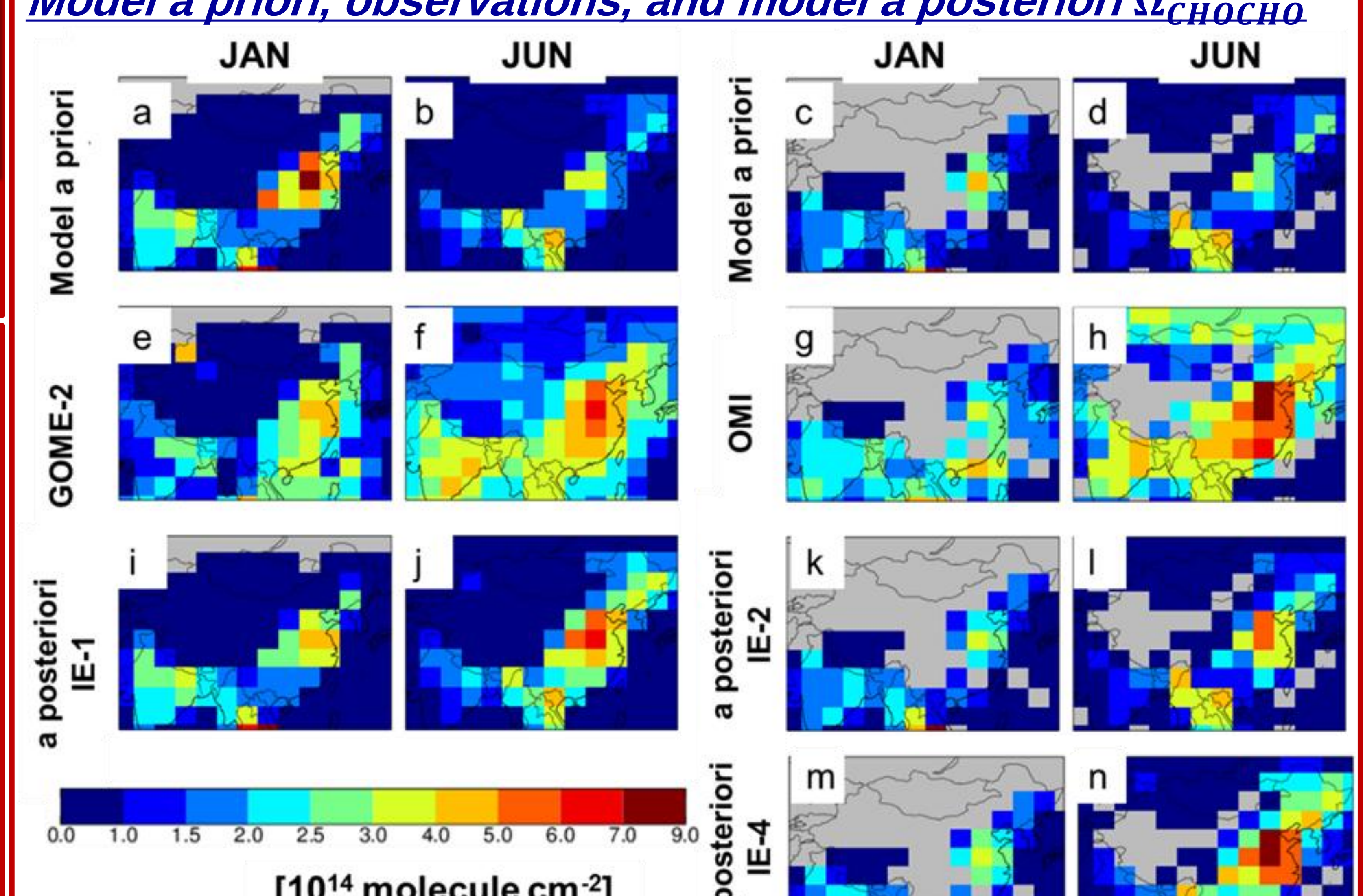


Model a priori, observations, and model a posteriori Ω_{HCHO}



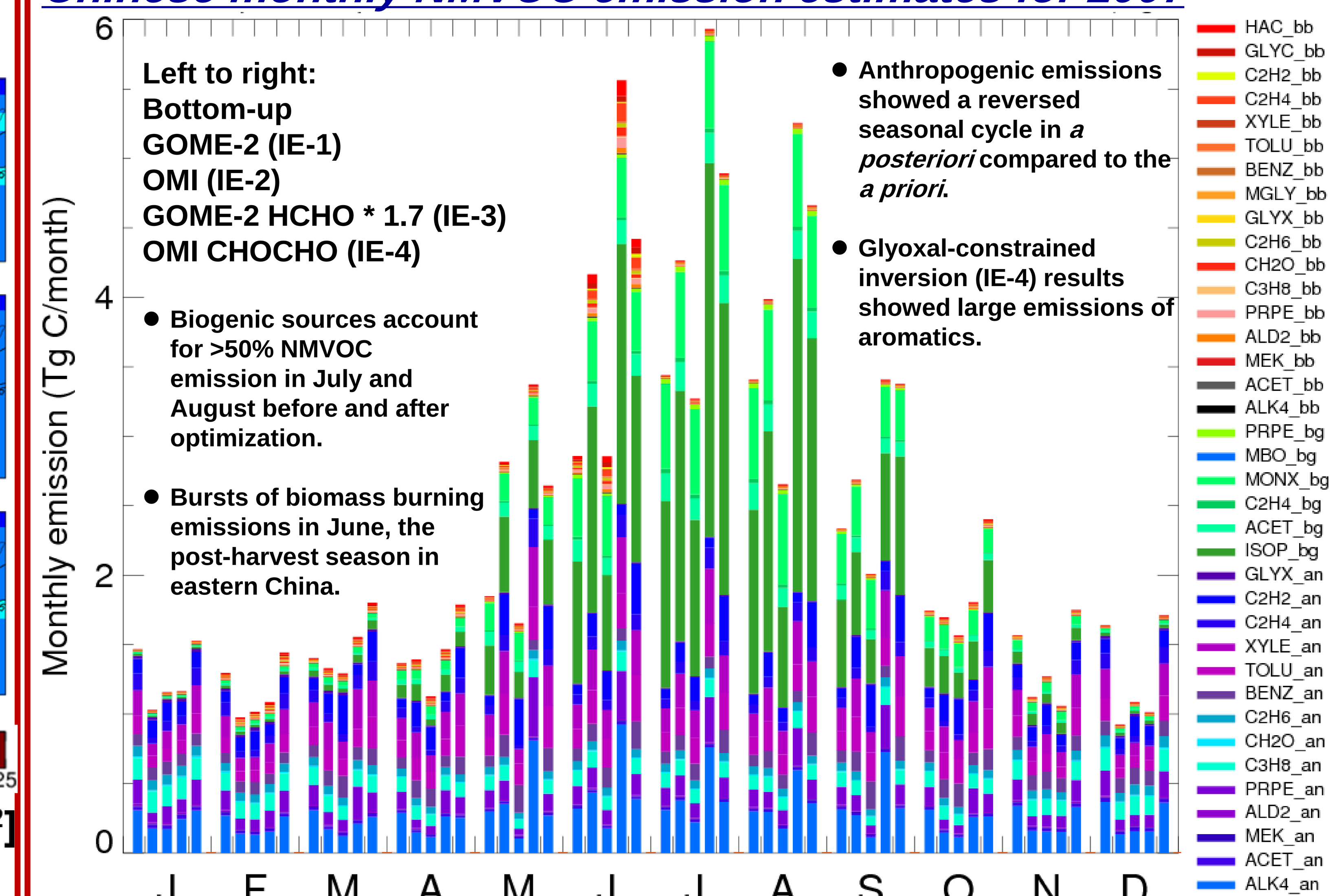
- Prevalently higher Ω_{HCHO} and stronger summer/winter contrast observed by GOME-2 than by OMI.
- IE-3, constrained by scaled-up GOME-2 Ω_{HCHO} only, was used to explore the upper limit of isoprene emission.
- Overpredicted Ω_{HCHO} in model a priori in JAN reflected an overestimation of wintertime anthropogenic emissions by MIX-Asia, while model-observation gap in JUN involved anthropogenic, biogenic, and crop fire emissions due to Chinese overlapped emission sources in summer.
- Model-observation gap was largely reduced in model a posteriori.

Model a priori, observations, and model a posteriori Ω_{CHOCHO}

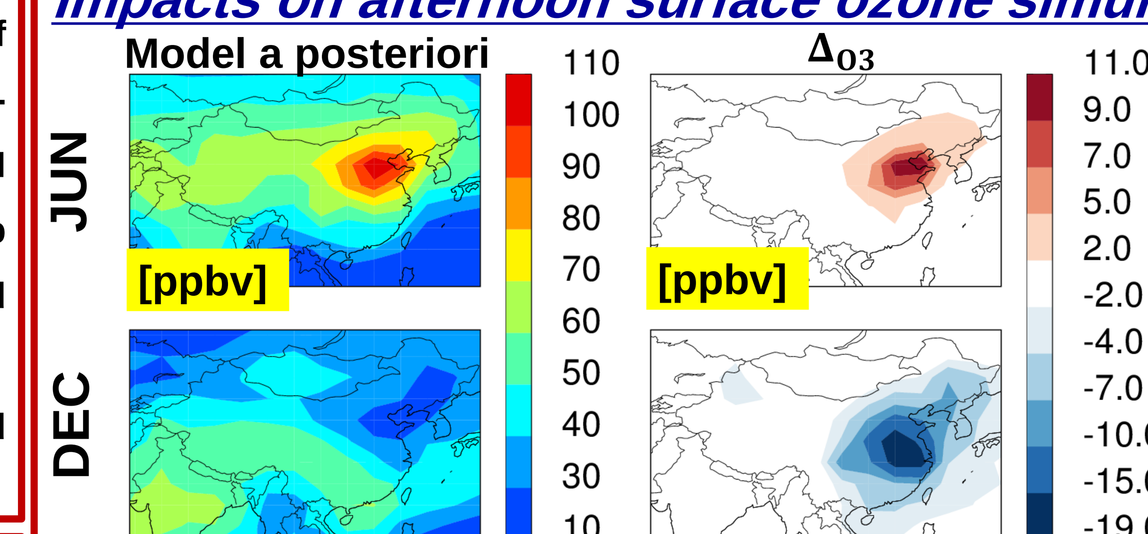


- Compared to satellite observations, simulated Ω_{CHOCHO} in model a priori had a reversed seasonal cycle, which was corrected in model a posteriori.
- IE-4, constrained by OMI glyoxal only, was used to explore the upper limit of Chinese aromatics emissions
- Compared to HCHO observations, glyoxal observations had a stronger west-to-east gradient pattern, suggesting that glyoxal is more related to anthropogenic sources and can help to differentiate between VOCs sources together with HCHO constraints.

Chinese monthly NMVOC emission estimates for 2007

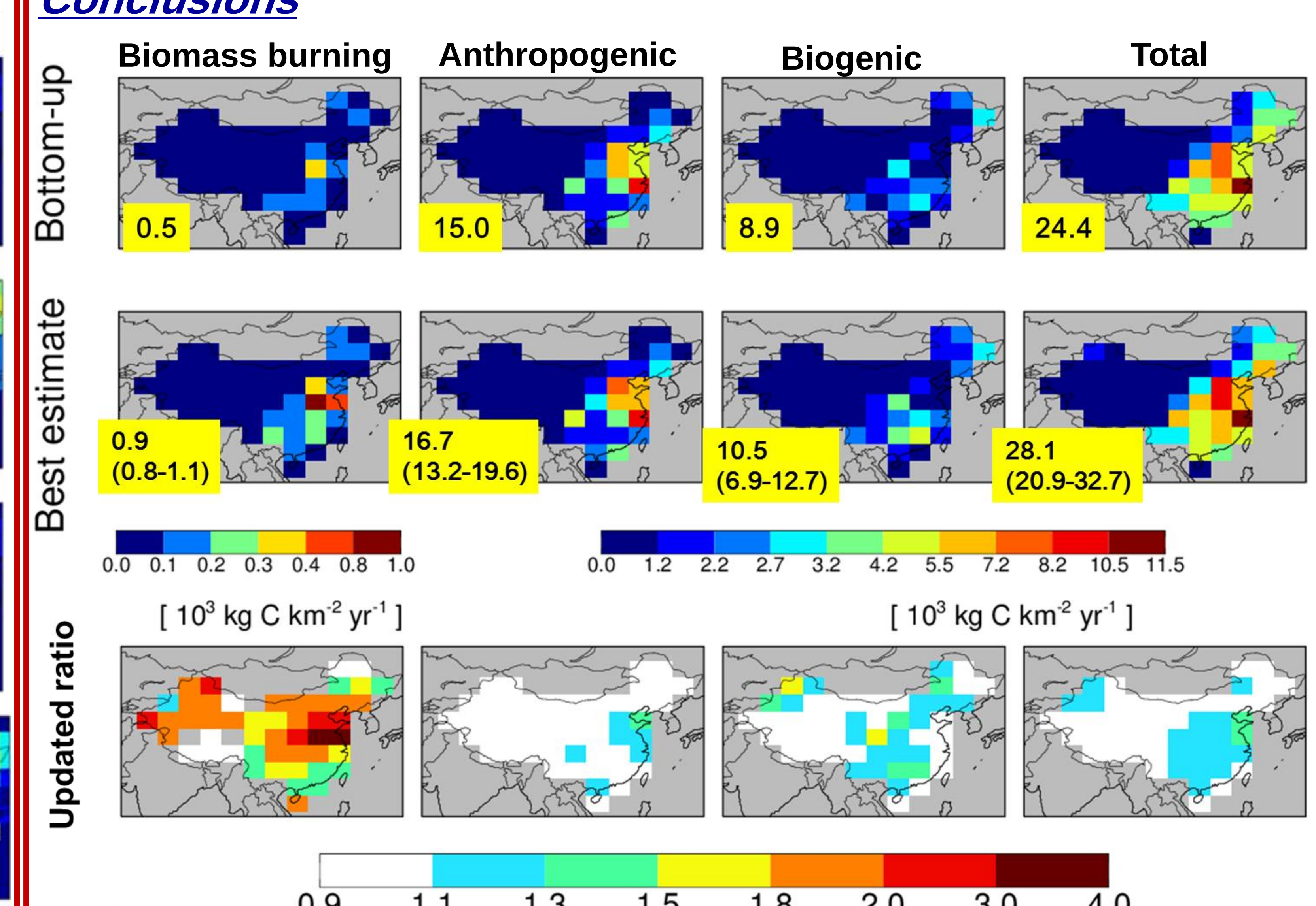


Impacts on afternoon surface ozone simulation in 2007



- Δ_{O3} = Model a posteriori – Bottom-up
- GOME-2-inferred NMVOC emission (IE-1) change enhanced seasonal variation of simulated monthly mean afternoon surface ozone across Eastern China

Conclusions



- Optimized Chinese anthropogenic NMVOC emission was 16.7 (13.2 – 19.6) Tg C y^{-1}, 11% higher than MIX-Asia estimate and with a different seasonal pattern and species profile
- Optimized Chinese biogenic NMVOC emission was 10.5 (6.9 – 12.7) Tg C y^{-1}, 18% higher than the estimate by MEGAN v2.1; biomass burning 0.9 (0.8-1.1) Tg C y^{-1}
- The change of simulated monthly mean afternoon surface O3 due to GOME-2-inferred VOCs emissions updates ranged from -19 to 11 ppbv.