

Improving the Representation of Secondary Organic Aerosol (SOA) in the MOZART-4 Global Chemical Transport Model

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INTRODUCTION/MOTIVATION

- ❑ Secondary organic aerosol (SOA) is formed through a series of oxidation reactions of VOCs followed by partitioning of products into particles.
- ❑ A major component of fine particles (PM_{2.5}) is organic aerosol (OA), a significant fraction of which is SOA.
- ❑ Airborne particles pose serious health risks and have climate consequences.
- ❑ Most global chemical transport models (CTMs) underestimate SOA compared to observation.
- ❑ Outputs from global models are frequently used as boundary conditions for regional CTMs.
- ❑ Improving representation of SOA in MOZART-4 will provide better estimates of SOA on global to regional scales.

MOZART-4 MODEL

- ❑ Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) is an offline global chemical transport model.
- ❑ There are 85 gas-phase species, 12 bulk aerosol compounds, 39 photolysis and 157 gas-phase reactions in MOZART-4.
- ❑ The aerosol component of the model includes calculations of sulfate, black carbon, primary organic and secondary organic (SOA), ammonium nitrate and sea salt.
- ❑ SOA formation is based on two product (2p) model

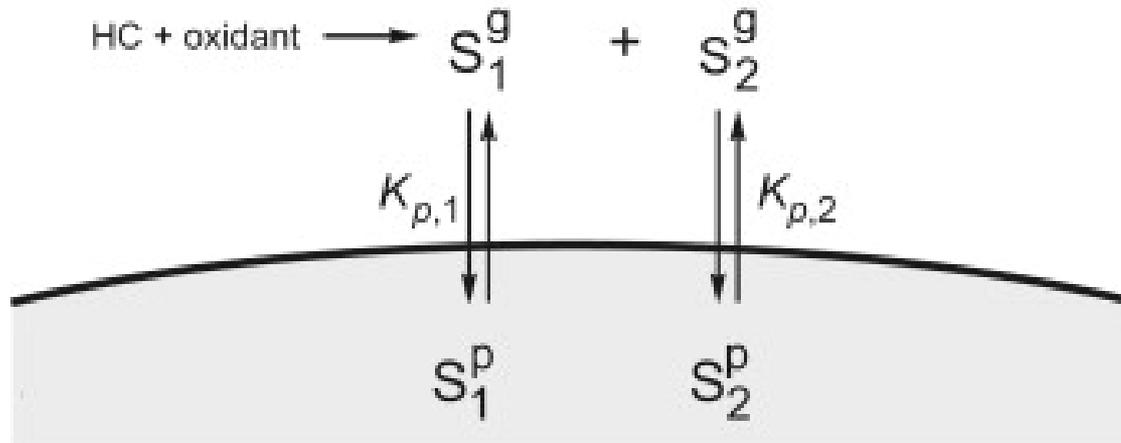


Fig. 1. Schematic of the two-product (2p) SOA model
(From Kroll and Seinfeld, 2008)

SOA yield calculation:
$$Y_p = M_o \sum_i \left(\frac{\alpha_i K_{om,i}}{1 + K_{om,i} M_o} \right)$$

Where α_i fractional yield of each S from reacted HC

K_{omi} partitioning coefficient

M_o total organic mass at a given time step

Steps to build and run MOZART-4

Step 1: Configuring system info
Gets system and directory info

Step 2: Preprocessing
Preprocesses MOZART chemistry input

Step 3: Building MOZART-4 executable
Extracts chemistry info, and compile to build
system specific executable

Step 4: Running simulation
Modify name list file as desired, and run the
model for different cases

Steps to update MOZART-4

Tagging appropriate parent VOC precursors in
the gas-phase reactions to treat them in SOA
calculations

Modifying mo_setsoa.F90 code to update two-
product (2p) parameters and add new parent
VOC species for SOA calculations

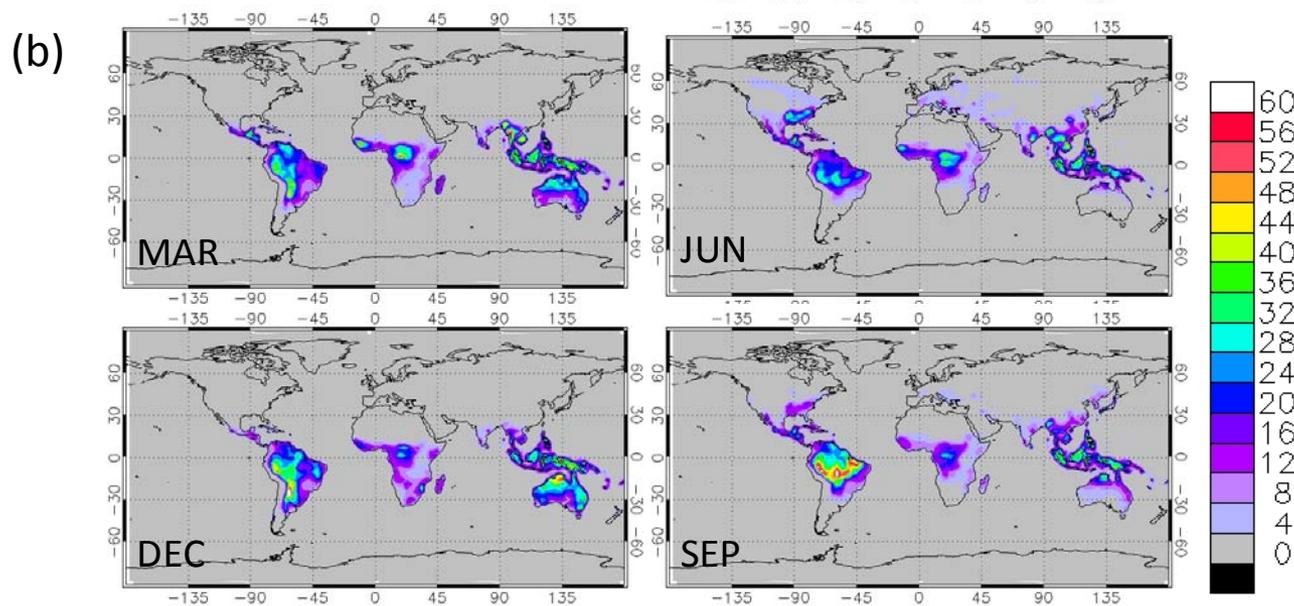
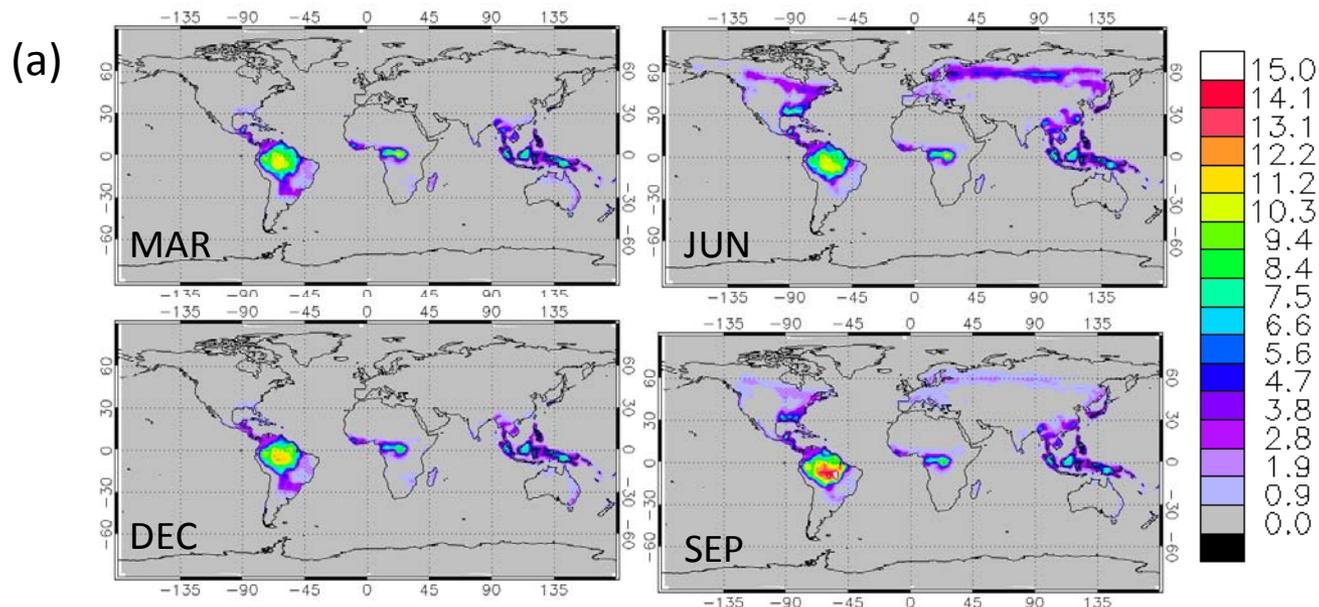
Fig. 2. Flow chart showing the steps in building and running MOZART-4

MODEL RUNS

- ❑ MOZART-4 was run for three cases: 1) Base-case: original model with normalized SOA parameters, 2) Case A: updated with two-product volatility basis set (2p-VBS) parameters, and 3) Case B: updated with 2p-VBS parameters and additional new parent VOCs
- ❑ Global domain with $2.8^{\circ} \times 2.8^{\circ}$ grid-cells, 28 layers extending up to ~ 2 hPa (dimension: 64 x 124 x 28)
- ❑ Global anthropogenic emissions from precursors of ozone and their effects in the troposphere (POET) emissions inventory, biogenic emissions from on-line calculations by MEGAN emissions model, fire emissions from global fire emissions database (GFED)
- ❑ Meteorology from the NCAR reanalysis of the NCEP forecast for the year 2006

Table 1. List of original and newly added parent VOCs contributing to SOA mass with updated 2p-VBS parameters used in MOZART-4

Status	Model parent VOC	SPRC 99	Oxidant	α_1	K_{om1}	α_2	K_{om2}
Original	C ₁₀ H ₁₆	TERP	O ₃ or OH	0.289	0.008	0.086	0.205
Original	TOLUENE	ARO1+ARO2	OH	0.325	0.008	0.124	0.146
Original	BIGALK	ALK3+ALK4+ALK5	OH	0.100	0.150	0.047	0.080
Added	ISOP	ISOPRENE	OH	0.178	0.011	0.022	2.106
Added	BIGENE	OLE2	OH	0.144	0.006	0.022	0.185
Added	C ₃ H ₆	OLE1	OH	0.078	0.005	0.006	0.167



Global emissions for 2006
 (i) Isoprene: 462 Tg
 (ii) Monoterpenes: 89 Tg

Fig. 3. Global distributions of monthly-average emission rates ($\text{mg m}^{-2} \text{day}^{-1}$) for (a) monoterpenes ($\text{C}_{10}\text{H}_{16}$), and (b) isoprene (C_5H_8).

RESULTS

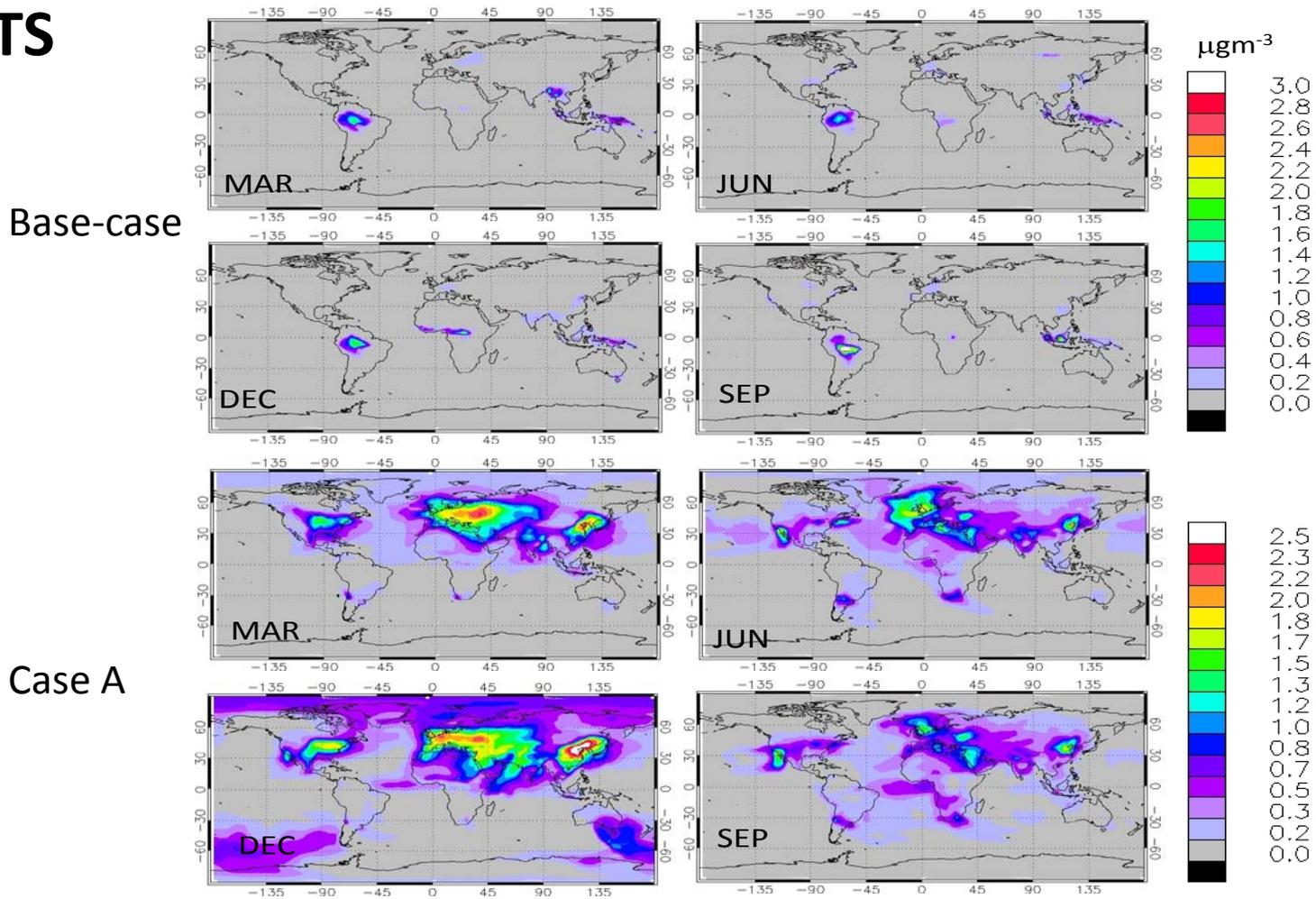
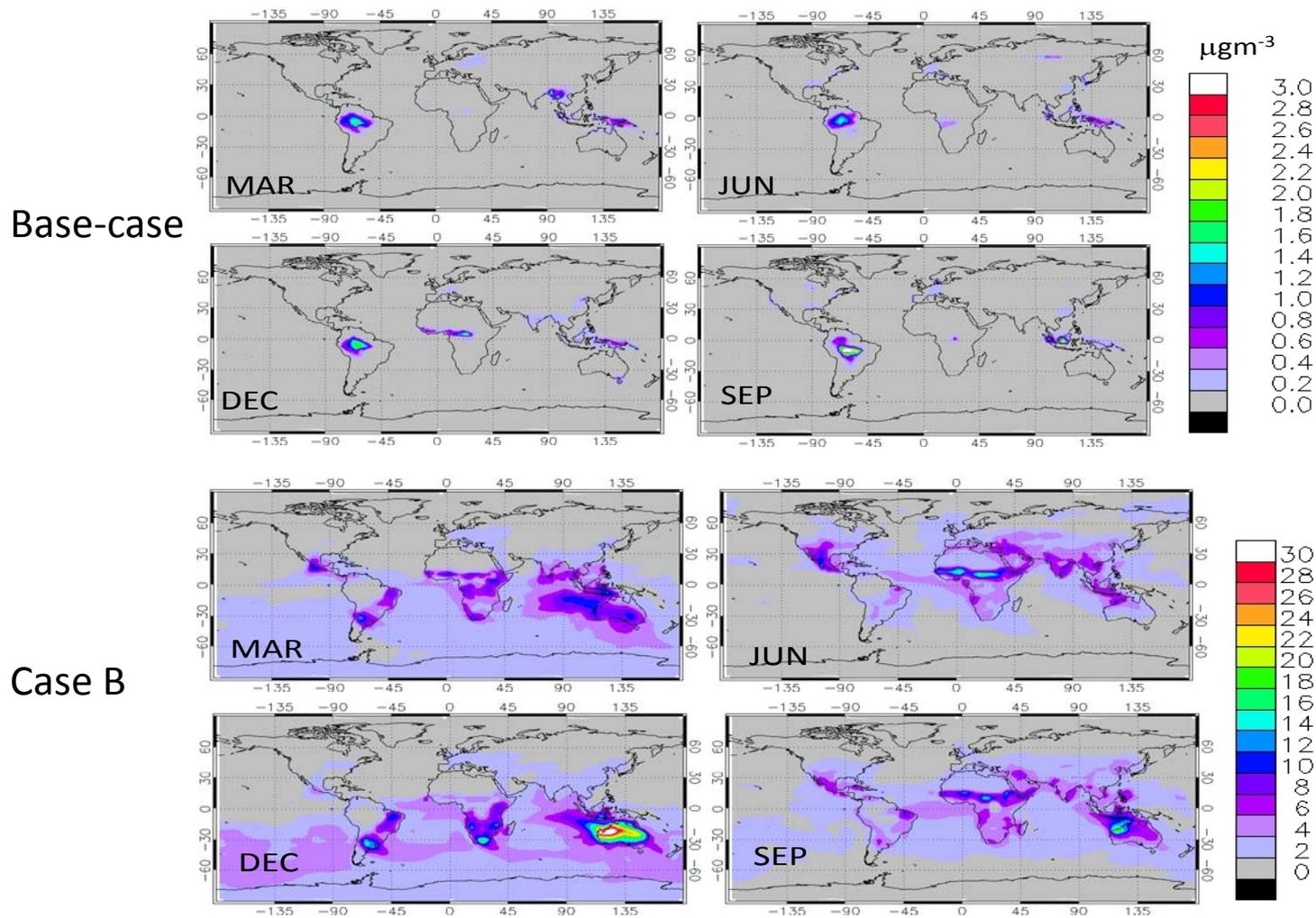


Fig. 4. Global distributions of simulated monthly-average SOA surface concentrations ($\mu\text{g m}^{-3}$) (top: Base-case) and relative increase from base-case (bottom: Case A) for March, June, September and December of 2006.



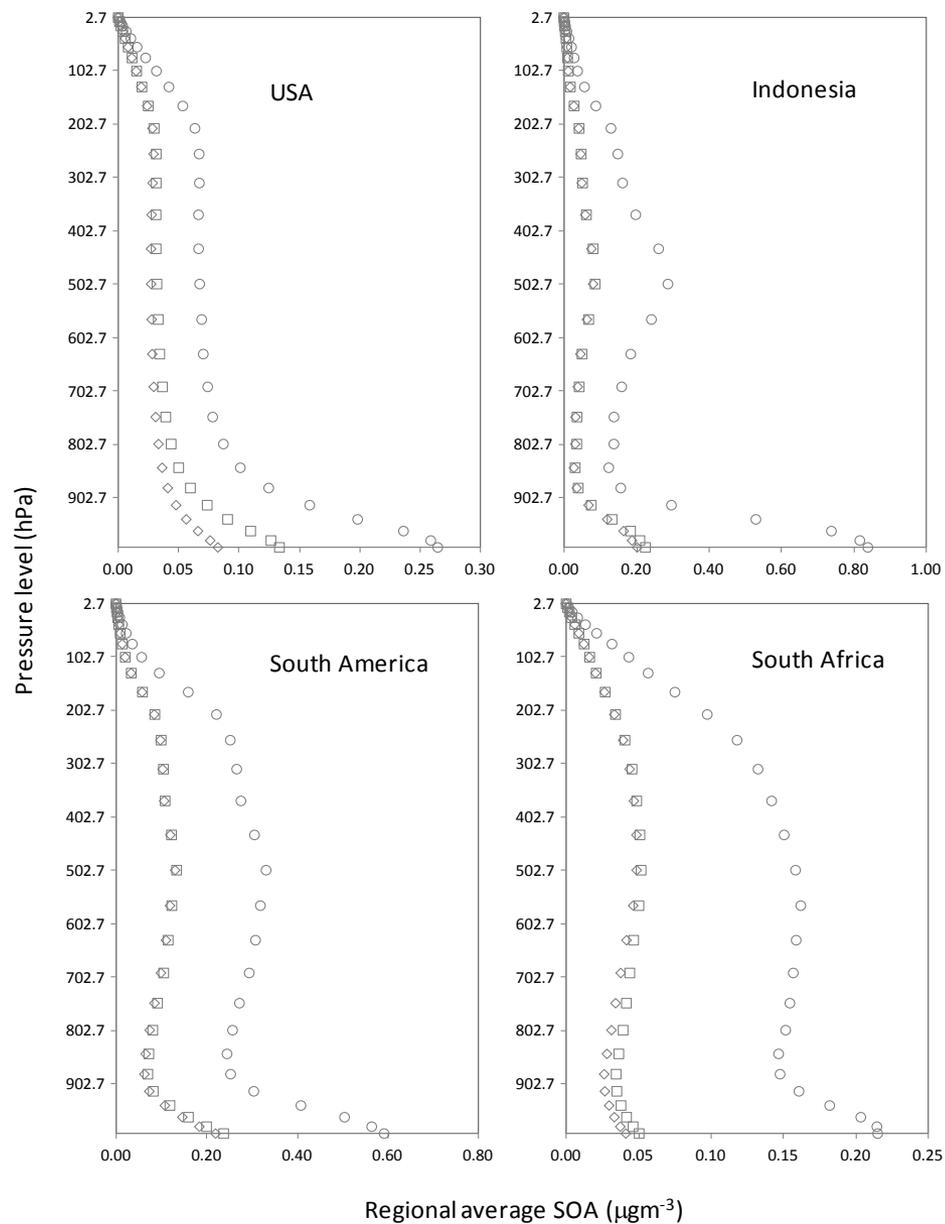


Fig. 6. Vertical distributions of annual-average SOA concentrations over different regions of the world. Open diamonds represent base-case, open squares represent Case A, and open circles represent Case B

Table 2. Regionally-averaged annual SOA concentrations at surface predicted for 2006

Region	Annual-average($\pm 1\sigma$) SOA (μgm^{-3}) at surface		
	Base-case	Case A	Case B
Canada (50-70N, 235-300E)	0.06 \pm 0.04	0.08 \pm 0.04	0.12 \pm 0.07
USA (25-50N, 235-300E)	0.08 \pm 0.05	0.13 \pm 0.10	0.27 \pm 0.19
Europe (30-70N, 10W-30E)	0.09 \pm 0.06	0.20 \pm 0.16	0.31 \pm 0.22
North Asia (45-70N, 60-150E)	0.05 \pm 0.04	0.07 \pm 0.05	0.12 \pm 0.07
Southeast Asia (10-45N, 60-125E)	0.09 \pm 0.11	0.16 \pm 0.18	0.42 \pm 0.45
Indonesia (10S-10N, 90-150E)	0.20 \pm 0.27	0.23 \pm 0.30	0.84 \pm 0.84
North Africa (0-30N, 20W-60E)	0.06 \pm 0.10	0.09 \pm 0.12	0.27 \pm 0.36
South Africa (40S-Eq, 0-60E)	0.04 \pm 0.06	0.05 \pm 0.08	0.22 \pm 0.27
South America (30S-Eq, 270-330E)	0.22 \pm 0.43	0.24 \pm 0.45	0.59 \pm 0.81
Australia (45-10S, 290-310E)	0.06 \pm 0.06	0.07 \pm 0.08	0.38 \pm 0.33

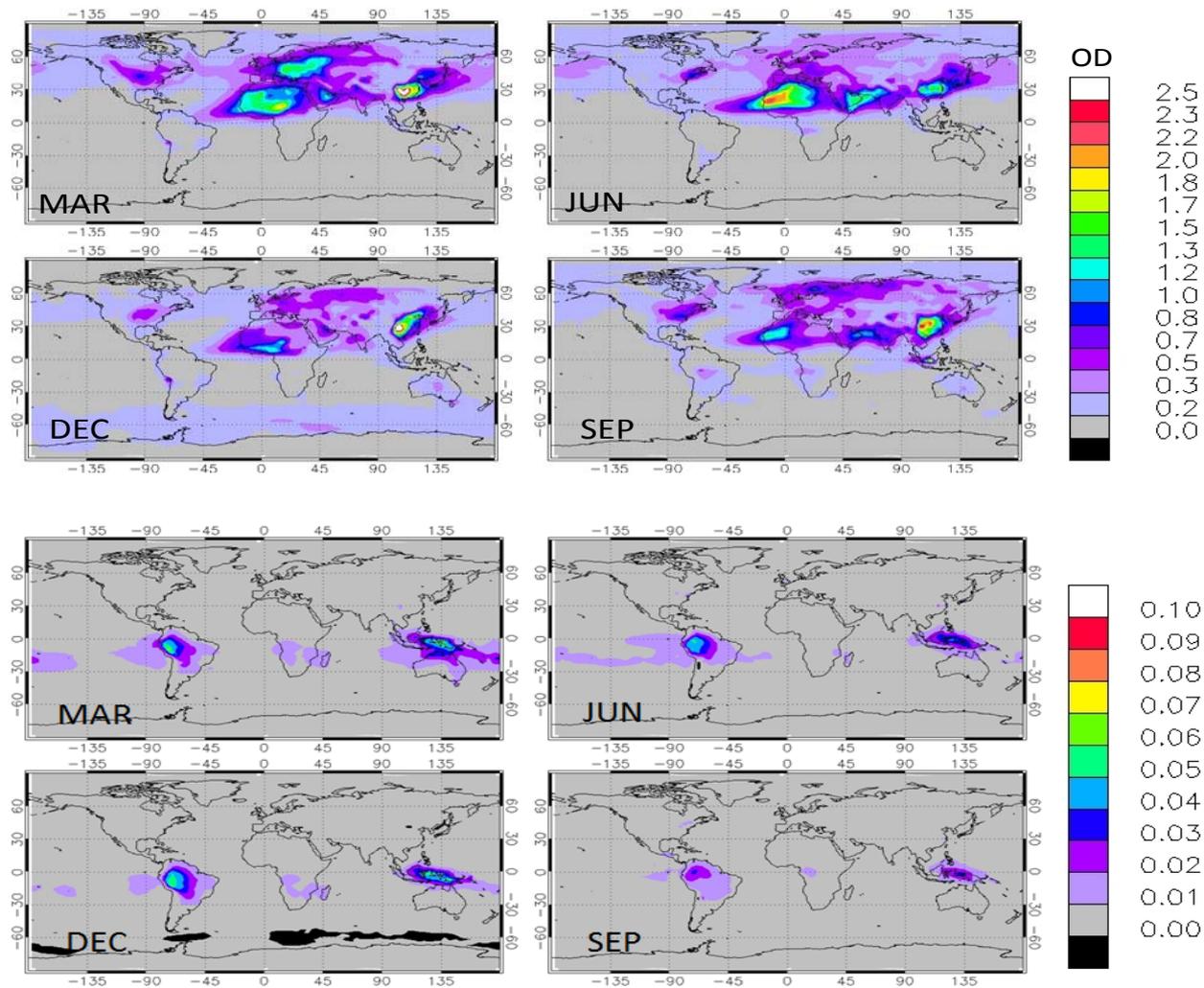


Fig. 7. Distributions of predicted aerosol optical depth (AOD) (top: base-case) and relative increase from base-case (bottom: Case B)

How does the updated MOZART-4 output compare to other models and observation?

- ❑ The amount of observational data for global SOA is essentially nonexistent, since SOA is largely not a measurable component of total aerosol. Most of the reported data have been calculated based on EC/OC and OC/OM ratios from measured fine particulate mass.
- ❑ Comparison with other model output is also difficult because of the differences in the types of chemical transport model, chemical mechanisms, treatment of parent VOCs contributing to SOA, estimation techniques for SOA, and sources of emissions inventories.
- ❑ Example comparison: over China, SOA concentrations of 0.05 – 1.0 $\mu\text{g m}^{-3}$ (this study) vs. 0.4 – 2.5 $\mu\text{g m}^{-3}$ (Jiang *et al.*, 2012).
- ❑ Current study predicted free tropospheric (2-6km) SOA max over the NW pacific of $\sim 0.08 \mu\text{g m}^{-3}$ vs $\sim 4.6 \mu\text{g m}^{-3}$ measured during the ACE-Asia study in 2001 (Heald *et al.* 2008).

SUMMARY

- ❑ The base-case simulation predicts annual average surface SOA of $0.22 \pm 0.43 \mu\text{g m}^{-3}$ in South America, $0.20 \pm 0.27 \text{ mg m}^{-3}$ in Indonesia, $0.08 \pm 0.05 \mu\text{g m}^{-3}$ in the USA, and $0.09 \pm 0.06 \mu\text{g m}^{-3}$ in Europe.
- ❑ Case A: updating the existing SOA parameters with 2p-VBS shows increases by $\sim 8\%$, $\sim 11\%$, $\sim 62\%$, and $\sim 110\%$ from the base-case in South America, Indonesia, USA, and Europe, respectively.
- ❑ Case B: updating the parameters and adding new VOC parent species shows even more dramatic increase in annual average SOA for above regions. SOA increases by $\sim 200\text{-}400\%$ from the base-case.
- ❑ Updating the existing MOZART-4 model increases SOA estimates from the base-case, but still under-estimates from observation
- ❑ On-going work includes update to the SOA module of CESM, which will replace the current SOA module in MOZART-4 in the future.

Additional Slides

Overview of Relevant Gas-Phase Chemistry

“Ox”
Day: OH, O₃ (HO₂, NO_x)

- Anthropogenic precursors: react w/OH
- Biogenic precursors
 - Isoprene reacts w/OH
 - Monoterpenes and sesquiterpenes react w/OH+O₃
- Then...branching based on NO_x concentrations

Night: O₃, NO₃

- Typically anthropogenic SOA precursors (benzene, xylene, toluene) react slowly with NO₃
- Biogenic precursors react w/NO₃
 - Isoprene, on order of reaction w/OH
 - Reactions w/α-,β-pinene particularly fast

