

Evaluating the Potential Importance of Monoterpene Degradation for Global Acetone Production



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Summary

- Acetone is one of the most abundant volatile organic compounds (VOCs) in the atmosphere, but estimates of the global source of acetone vary widely.
- A better understanding of acetone is essential because acetone serves as a source of HO_x in the upper troposphere and as a photochemical precursor to peroxyacetyl nitrate (PAN).
- Although there are primary anthropogenic and pyrogenic sources of acetone, the dominant sources are thought to be from direct biogenic emissions and photochemical production.
- Recent work suggests that photochemical degradation from monoterpenes may also represent a significant contribution to global acetone production^[1].
- We evaluate the importance of monoterpene degradation to global acetone production by adjusting the acetone yield derived from monoterpenes for a range of case studies in GEOS-Chem.
- When simulating an upper bound on yields, the model-to-measurement comparison improves for North America at both the surface and in the upper troposphere.
- The inclusion of acetone production from monoterpene degradation also improves the ability of the model to reproduce observations of acetone in East Asian outflow.
- In general the addition of monoterpenes degrades the model comparison for the tropical Southern Hemisphere.

Model Configuration and Evaluation

GEOS-Chem Global Simulations

- GEOS-Chem v10-01i
- GEOS-5 Meteorology
- 4x5 resolution
- 2005 spinup/2006 output
- MEGAN v2.1 Biogenic Emissions
- NASA Aircraft Data as in Fischer et al. (2012)^[2]

Monoterpene Speciation

- Speciation involved 8 representative monoterpenes including α -pinene, β -pinene, limonene, Δ 3-carene, myrcene, sabinene, trans- β -ocimene, and an 'other monoterpenes' parameter which includes 34 trace species delineated by Guenther et al. (2012)^[3].
- Acetone yields from monoterpenes were determined from a variety of species-specific chamber studies.

Model Cases and Acetone Chamber Yields from Terpenes

Monoterpene Species	Fischer et al. (2002)	Mean	Min	Max	Lower Bound	Upper Bound
α -Pinene	0.116 ^[2]	0.11 ^[4]	0.02 ^[5]	0.20 ^[6]	Min*(1/3)*	Max*(3)*
β -Pinene	0.116 ^[2]	0.09 ^[5]	0.009 ^[5]	0.15 ^[9]	Min*(1/3)*	Max*(3)*
Limonene	0.116 ^[2]	0.004 ^[6]	0.004 ^[6]	0.20*	Min*(1/3)*	Max*(3)*
Δ 3-Carene	0.116 ^[2]	0.14 ^[4]	0.09 ^[6]	0.18 ^[5]	Min*(1/3)*	Max*(3)*
Myrcene	0.116 ^[2]	0.37 ^[4]	0.22 ^[6]	0.51 ^[4]	Min*(1/3)*	Max*(3)*
Sabinene	0.116 ^[2]	0.20 ^[4]	0.16 ^[4]	0.30 ^[10]	Min*(1/3)*	Max*(3)*
Trans- β -Ocimene	0.116 ^[2]	0.19 ^[4]	0.18 ^[7]	0.20 ^[7]	Min*(1/3)*	Max*(3)*
Other Monoterpenes	0.116 ^[2]	0.11*	0.02*	0.50 ^[4] *	Min*(1/3)*	Max*(3)*

* : α -Pinene Analog ° : Terpinolene Yield * : Uncertainty of 3.00 in MEGAN2.1 Emissions

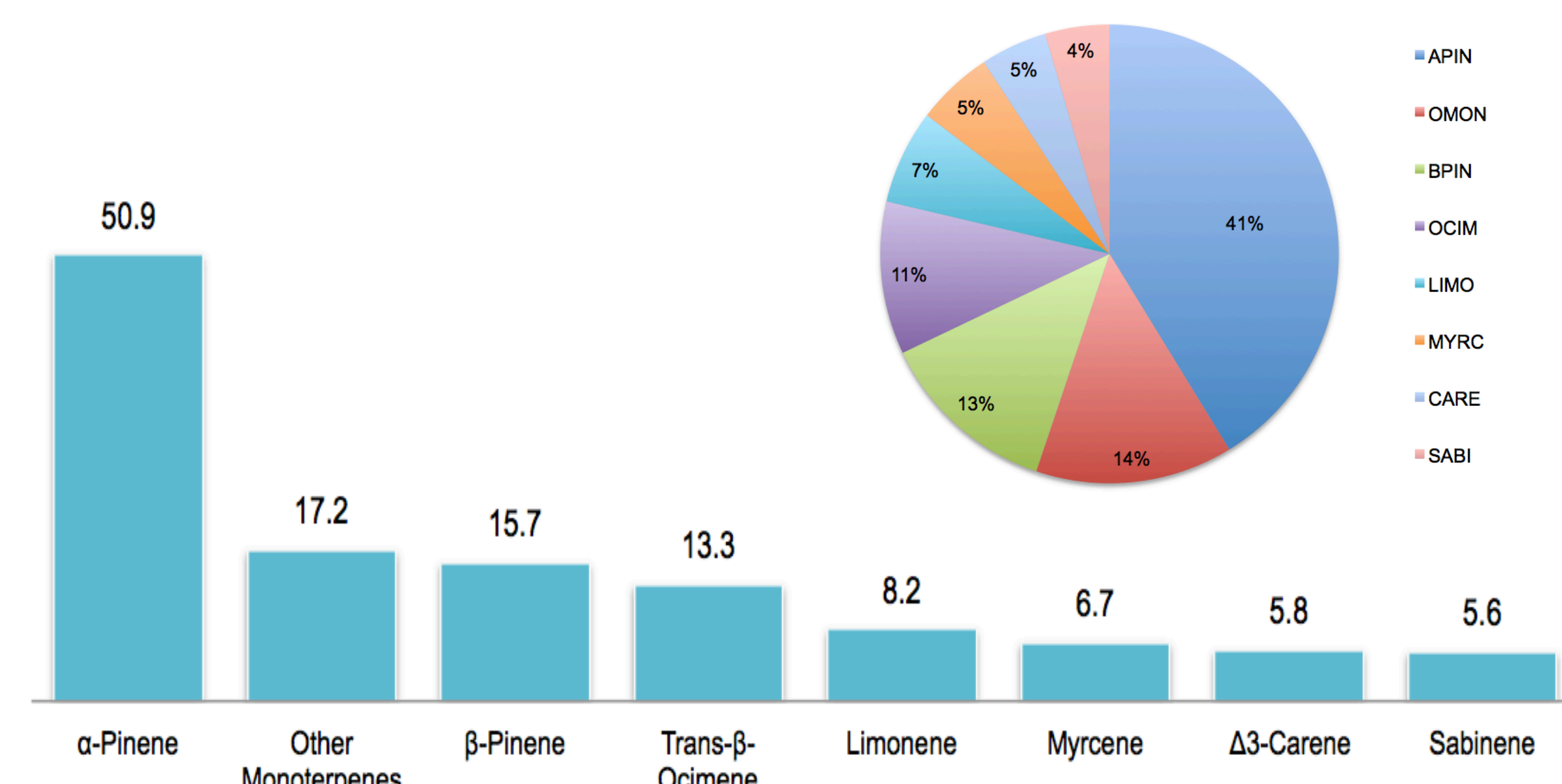


Figure 1. MEGAN global annual monoterpene emissions (Tg) and relative percent contribution

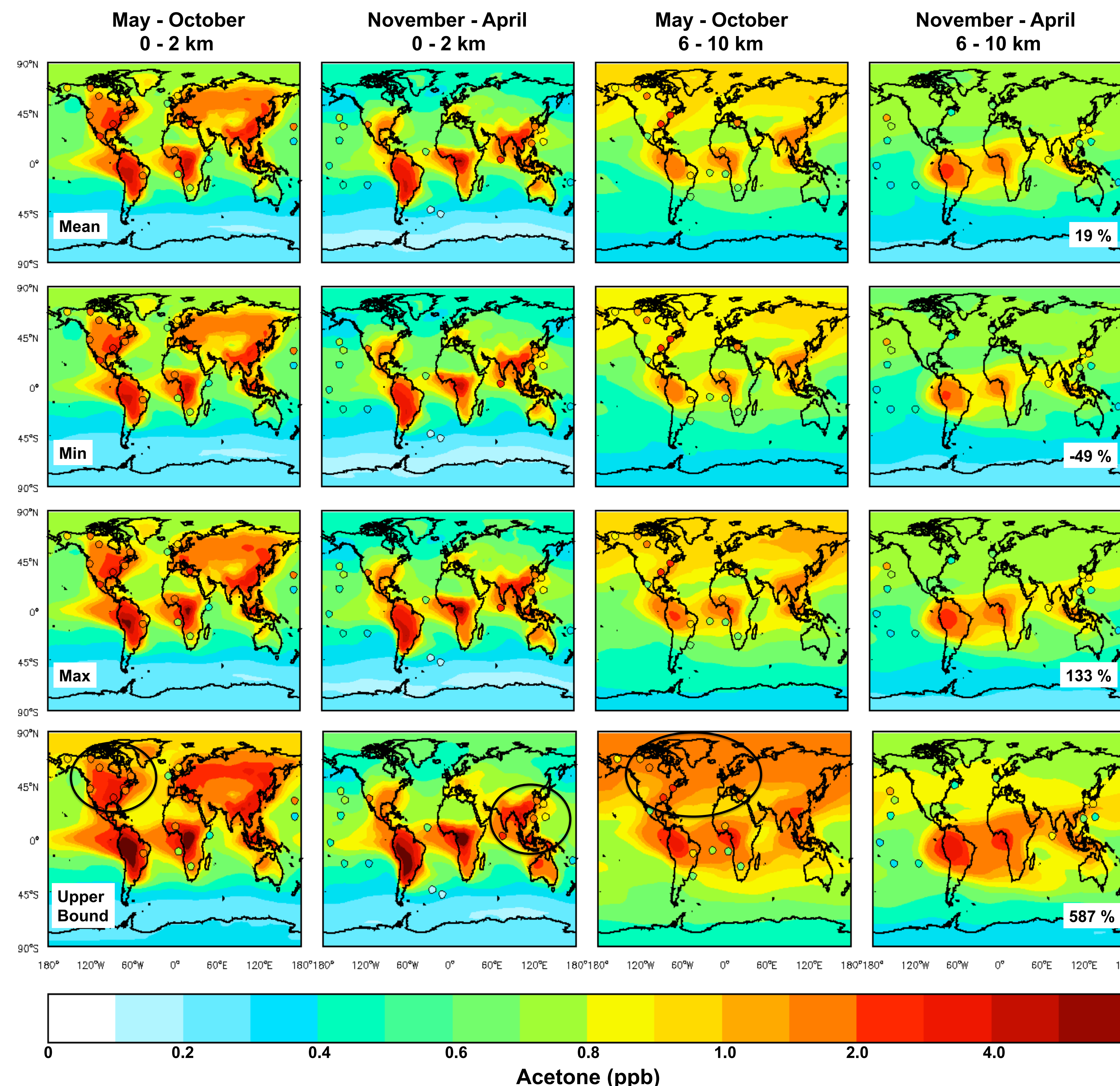


Figure 2. Simulated and observed global distribution of acetone for 2006 are shown as solid contours for four potential scenarios. For all model simulations there was an annual anthropogenic source of 3.58 Tg, biomass burning source of 2.54 Tg, terrestrial biosphere source of 37.1 Tg, and the oxidation of iso-alkanes contributed 18.4 Tg. The filled points on the maps are vertically binned aircraft campaign, ship cruise, and surface site data that have been averaged over regionally coherent regions^[2]. Black circles indicate model improvements with respect to observations. The percent change in the total annual global source of acetone from the oxidation of biogenic VOCs as compared to Fischer et al. (2012) is shown in the bottom right corner of the figure.

Conclusions

- Acetone concentrations display a strong seasonal cycle.
- The upper bound case, which incorporates the inherent uncertainty in MEGAN emissions, reveals a much larger yearly acetone source from the degradation of monoterpenes (587 % increase) compared to the Fischer et al. (2012) emissions.
- As a result of this dramatic biogenic increase, model-to-measurement comparisons improves for the boundary layer (0 - 2 km) and the upper troposphere (6 - 10 km) over North America.
- Model-to-measurement improvements occur over East and South Asia in the boundary layer (0 - 2 km).
- The observational data do not support increased acetone emissions in the tropical Southern Hemisphere.

Future Work

- Explore the causes of the underprediction in the model.
- Analyze additional aircraft and satellite observations for model-to-measurement comparisons.
- Investigate if the recent boom of natural gas production significantly affects model simulations.
- Explore the effects of SOA chemistry on monoterpene-derived acetone yield as MEGAN only deals with emissions and not chemical mechanisms.

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