The Role of Acetone on Global Atmospheric Composition

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Abstract

Acetone is an abundant volatile organic compound with important influence on ozone and atmospheric self-cleaning processes. The budget of acetone is influenced by various sources and sinks. Direct sources include anthropogenic, terrestrial vegetation, oceanic, and biomass burning emissions, while chemistry forms acetone from other compounds. Sinks include deposition onto the land and ocean surfaces, as well as chemical loss. The GISS Earth System Model, ModelE, is capable of simulating a variety of Earth system interactions. Previously, acetone had a very simplistic representation in the ModelE chemical scheme. This study assesses a greatly improved acetone tracer scheme, in which acetone's sources, sinks and atmospheric transport are now tracked in 3 dimensions. Extensive research was conducted to assess how well past literature supported the new global acetone budget. Anthropogenic, vegetation, biomass burning, and deposition schemes fit well with previous studies. While their net fluxes were well-supported, source and sink terms for chemistry and the ocean were overestimated and underestimated, respectively. In iterations of the chemistry scheme, it was found that the production of acetone from hydrocarbon oxidation is a strong leverage to the overall chemical source. Spatial distributions reveal that ocean uptake of acetone dominates northern latitudes, while production is mainly in mid-southern latitudes. Ocean surface conditions influence ocean-acetone interactions and will be considered when modifying the ocean scheme in future work. The seasonality of acetone-related processes was also studied in conjunction with field measurements around the world. These comparisons show promising results, but have shortcomings at urban locations, since the model's resolution is too coarse to capture high-emission areas. Overall, an analysis of the acetone budget aids the development of the tracer in the GISS ModelE, a crucial step to parameterizing the role of acetone in the atmosphere.

The Role of Acetone on Global Atmospheric Composition

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1. Motivation

Acetone (C_3H_6O) is an abundant volatile organic compound with important influence on ozone and atmospheric self-cleaning processes. The budget of acetone is influenced by various sources and sinks. Direct sources include anthropogenic, natural vegetation, oceanic, and biomass burning emissions, while chemistry forms acetone from other compounds. Sinks include deposition onto the land and ocean surfaces, as well as chemical loss. The GISS Earth System Model, ModelE, is capable of simulating a variety of Earth system interactions. Previously, acetone had a very simplistic representation in the ModelE chemical scheme. This study assesses a greatly improved acetone tracer scheme, in which acetone's sources, sinks and atmospheric transport are now tracked in 3 dimensions.

2. Introduction

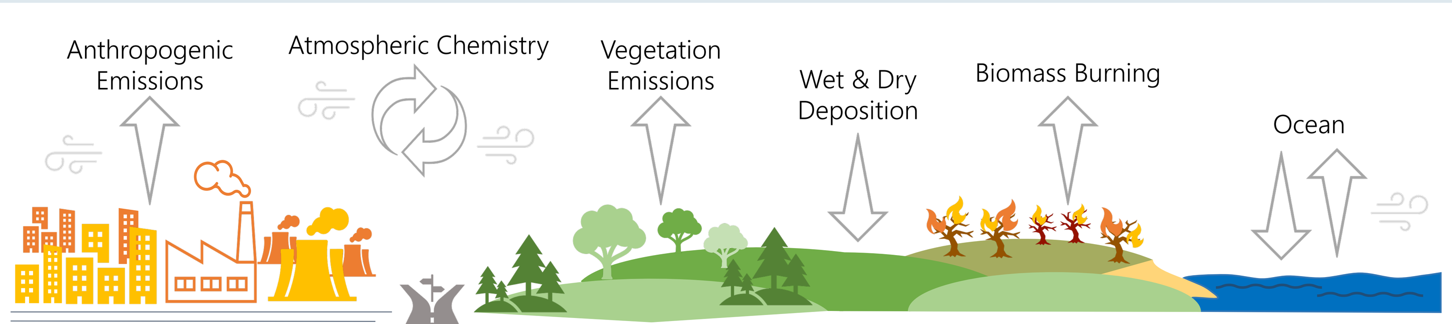


Fig 1. Sources and Sinks of Acetone in the Atmosphere

The new GISS acetone tracer scheme parametrizes the sources and sinks of acetone.

- **Sources:** Anthropogenic and biomass burning sources are given by standard CEDS emissions (Hosely et al., 2018; van Marle et al., 2012). Vegetation emissions utilize MEGANv2.1 (Guenther et al., 2012).
- **Sinks:** GISS ModelE wet and dry deposition schemes are used (Shindell et al., 2001).
- **Bidirectional Fluxes:** Chemical production comes from paraffin and terpenes, and destruction comes from oxidation by OH, reaction with Cl, and photolysis (Shindell et al., 2003). The ocean uses a two-film model (Liss & Slater, 1974).

3. Global Acetone Budget

Table 1. Global Acetone Budget Table

	This Study – Baseline [2021]	*Wang et al.* [2020] [a]	*Wang et al.* [2020] [b]	*Brewer et al.* [2017]	*Fischer et al.* [2012]	*Elias et al.* [2011]	*Jacob et al.* [2002]	Other Estimates [2000-2016] [e]
Burden (Tg)	2.93	3.5	3.80	5.57	5.60	7.20	3.80	3.50 – 4.20
Global Deposition (Tg/yr)	-22.2	-25.2	-12.4	-12.4	-12.0	-19.0	-9.0	-26.0 – -6.0
Biomass Burning (Tg/yr)	1.59	4.0	2.40	2.60	2.80	2.40	4.50	3.22 – 9.0
Anthro Emissions (Tg/yr)	1.00	0.50	3.40	3.60	0.73	1.60	1.10	1.02 – 2.0
Vegetation Emissions (Tg/yr)	36.1	39.8	32.2	37.1	32.0	76.0	35.0	15 – 56
Net Ocean (Tg/yr)	3.94	-8.10	1.30	-7.50	-2.0	-8.0	13.0	4.0
Ocean Source (Tg/yr)	15.2	33.4	45.7	51.8	80.0	20.0	27.0	20.0
Ocean Sink (Tg/yr)	-11.3	-41.5	-44.4	-59.2	-82.0	-28.0	-14.0	-62.0
Net Chemistry (Tg/yr)	-20.5	-11.1	-26.1	-22.5	-21.0	-53.0	-45.0	-33.0 – -5.50
Chem Source (Tg/yr)	33.3	38.5	26.1	24.1	31.0	27.0	28.0	15.5 – 55.6
Chem Sink (Tg/yr)	-53.8	-49.6	-52.2	-46.6	-52.0	-80.0	-73.0	-61.1 – -33.4
Chemical Lifetime (days) [c]	19.9	25.8	26.6	43.6	39.3	32.9	19.0	20.9 – 35.6
Lifetime (days) [d]	12.3	11.0	12.7	17.2	14.0	21.0	14.5	12.8 – 35

[a] CAM-Chem Model (Wang et al., 2020)
[b] GEOS-Chem Model (Wang et al., 2020)
[c] Chemical Lifetime = Burden/Chemical Sink
[d] Total Atmospheric Lifetime = Burden/Total Sink
[e] *Singh et al.* [2000, 2004], *Arnold et al.* [2005], *Folberth et al.* [2006], *Marandino et al.* [2006], *Guenther et al.* [2012], *Beale et al.* [2013], *Khan et al.* [2015], *Dufour et al.* [2016].

The GISS acetone tracer is compared to previous model budgets (Table 1). Box and whisker plots are given, and the values from GISS ModelE are overlaid in blue circles (Figure 2).

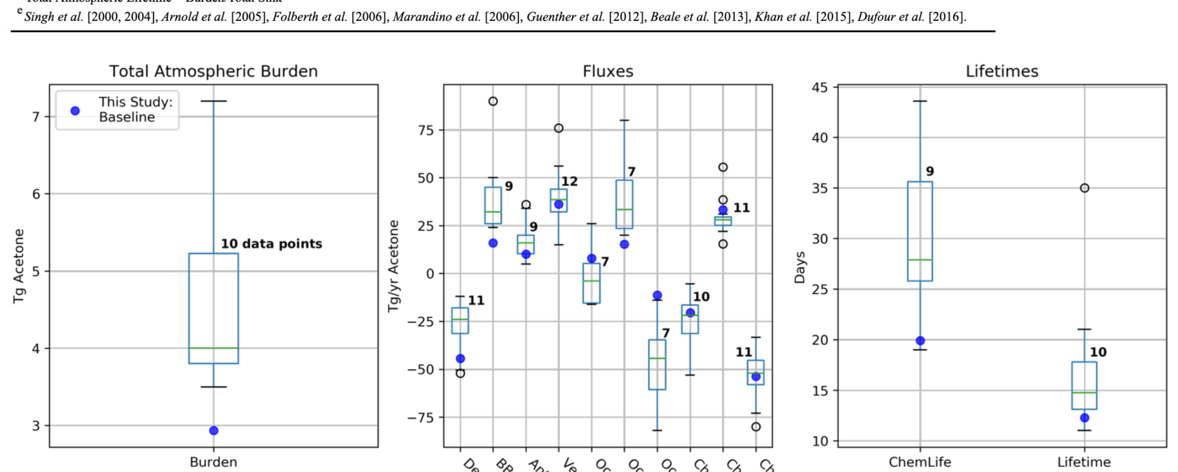


Fig 2. Global Acetone Budget Boxplots

4. Model Evaluation

Spatial Distributions of Acetone

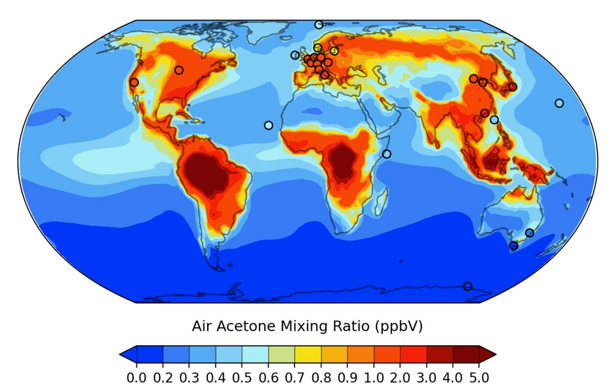


Fig 3. Global Acetone Mixing Ratios

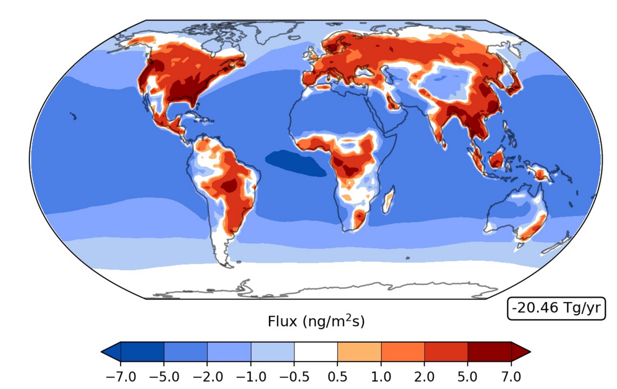


Fig 4. Global Chemistry Fluxes

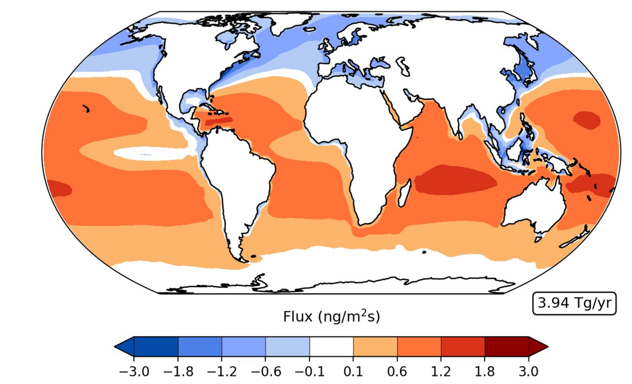


Fig 5. Global Ocean Fluxes

- Acetone mixing ratios are most concentrated over the continents due to anthropogenic, vegetation, and other terrestrial emissions. This compares well with field measurements (Figure 3).
- Chemical destruction is concentrated over continents, and destruction is primarily over tropical oceans (Figure 4).
- The ocean is a sink in the northern latitudes, a source in the tropics, and near equilibrium at southern latitudes. This corroborates well with previous studies (Fischer et al., 2012; Wang et al., 2020) (Figure 5).

Measurements taken from: de Gouw et al., 2004; Dolgorouky et al., 2012; Galbally et al., 2007; Guérette et al., 2019; Hu et al., 2013b; Huang et al., 2020; Langford et al., 2010; Lewis et al., 2005; Li et al., 2019; Read et al., 2012; Schade & Goldstein, 2006; H. B. Singh et al., 2003; Solberg et al., 1996; Warneke & de Gouw, 2001; Yoshino et al., 2012; Yuan et al., 2013.

Seasonality of Acetone

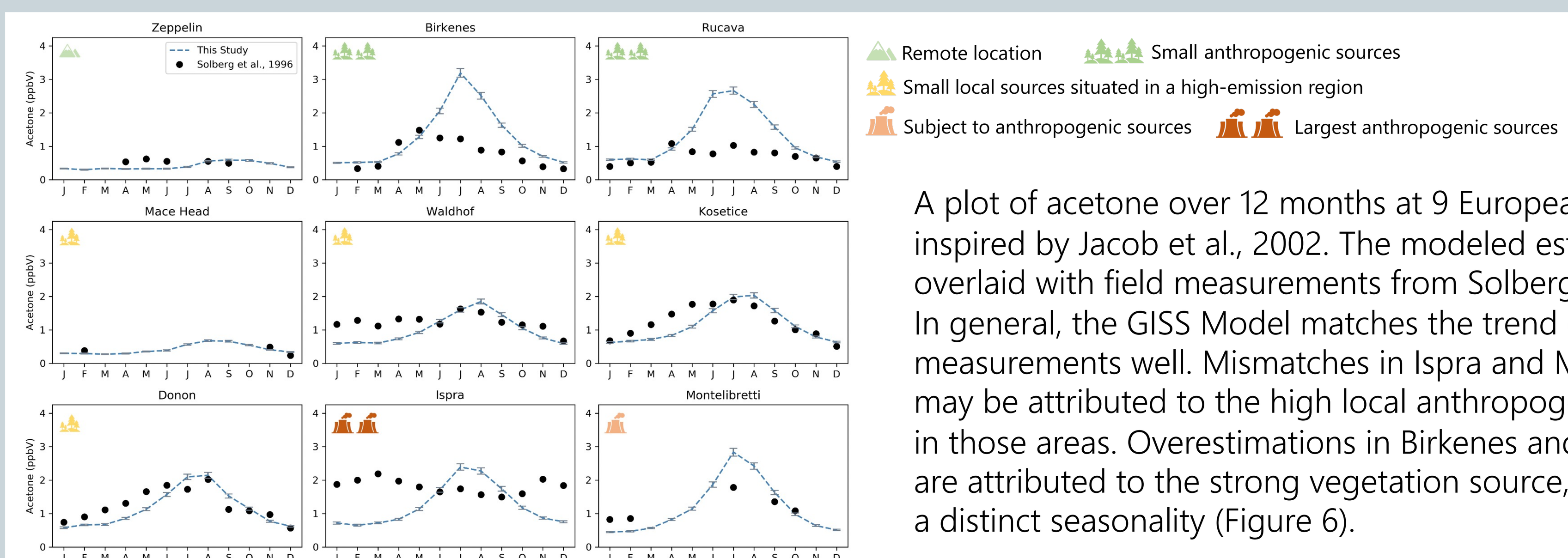


Fig 6. Monthly Acetone at European Sites

A plot of acetone over 12 months at 9 European sites was inspired by Jacob et al., 2002. The modeled estimates are overlaid with field measurements from Solberg et al., 1996. In general, the GISS Model matches the trend of the measurements well. Mismatches in Ispra and Montelibretti may be attributed to the high local anthropogenic sources in those areas. Overestimations in Birkenes and Rucava are attributed to the strong vegetation source, which has a distinct seasonality (Figure 6).

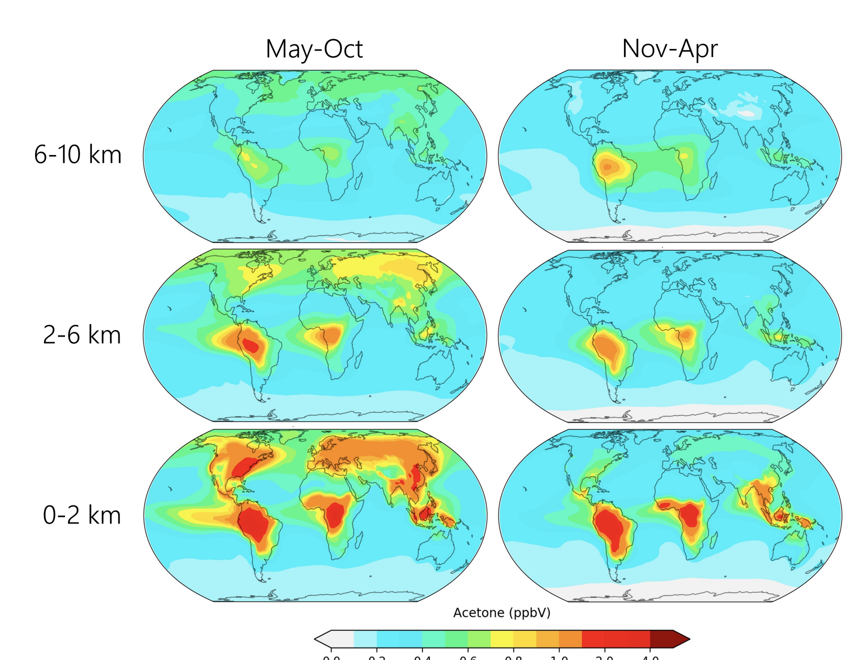


Fig 7. Vertical Distribution of Acetone Over Long Seasons

A plot of acetone mixing ratios in the atmosphere was inspired by Fischer et al., 2012. Acetone mixing ratios are higher in May-Oct than Nov-Apr, and this relationship is stronger in the lower atmosphere (Figure 7).

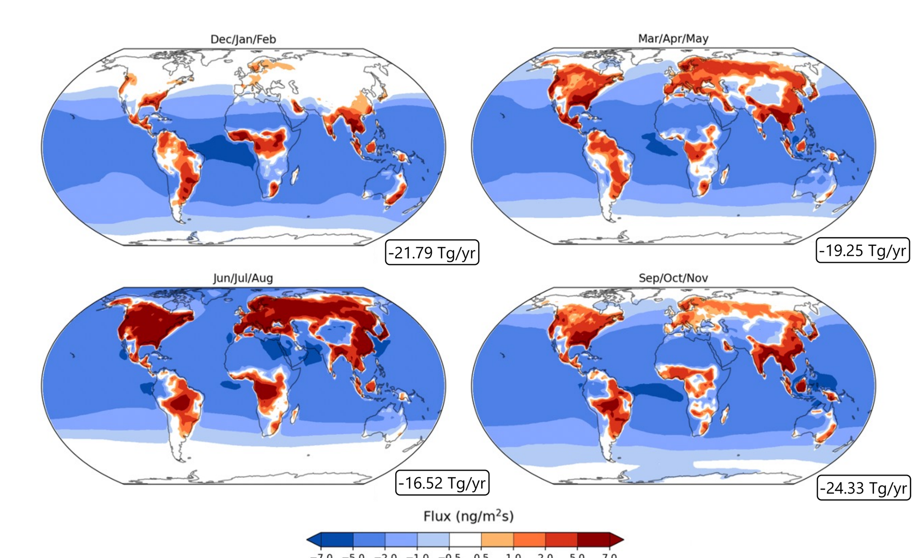


Fig 8 Chemistry Flux Over Four Seasons

Chemical production is strongest in Jun/Jul/Aug and weakest in Sep/Oct/Nov. Chemical destruction is strongest in Jun/Jul/Aug. Net values for each month's bidirectional chemical flux are shown in boxes on the lower left (Figure 8).

Ocean-acetone interaction also reveals seasonality trends. Production is strongest in Dec-May and weakest in Jun-Nov.

5. Sensitivity Simulations

Table 2. Sensitivity Studies

GISS ModelE Sensitivity Simulation	Sensitivity Parameter	Description
Chem_Cl0	Chemistry Source	Acetone + Chlorine reaction rate = 0
Chem_Terp0	Chemistry Source	No reaction for production of acetone from terpenes
Chem_Par0.5	Chemistry Source	Half the yield of acetone from paraffin (17.5%)
Chem_Par2.0	Chemistry Source	Double the yield of acetone from paraffin (70%)
Veg_0.7	Vegetation	0.7 factor of acetone from MEGAN
Ocn_2.0	Ocean	Ocean acetone concentration from 15nM to 30nM
Dep_f0	Deposition	f_0 changed from 0.1 to 0
BB_2.0	Biomass Burning	Double biomass burning emissions

We explored sensitivities to the baseline in both directions. Chemistry simulations altered the strengths of sources. Terrestrial simulations altered strengths of vegetation and biomass burning, and changed parameters for the ocean and deposition schemes.

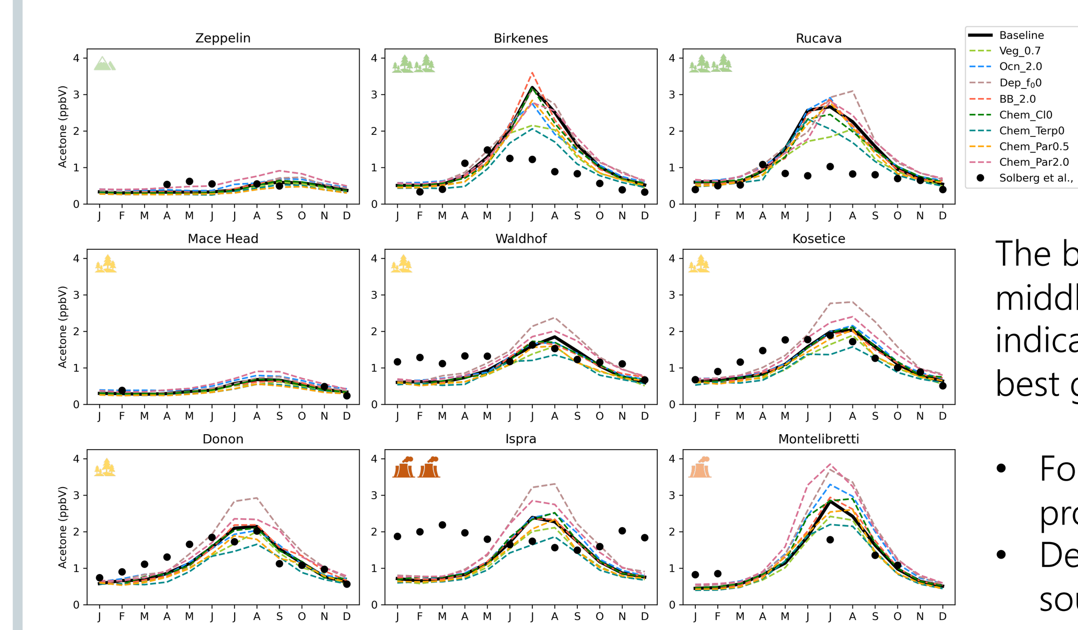


Fig 9. Monthly Acetone at European Sites – Sensitivity Studies

The baseline model lies in the middle of the uncertainties, indicating that it is an acceptable best guess (Figure 9).

- Found nonlinearities in acetone production from paraffin.
- Deposition f_0 and vegetation sources are critical.

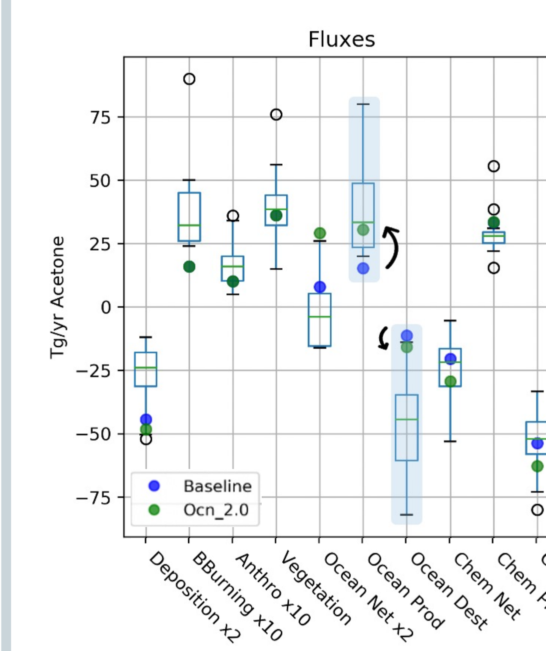
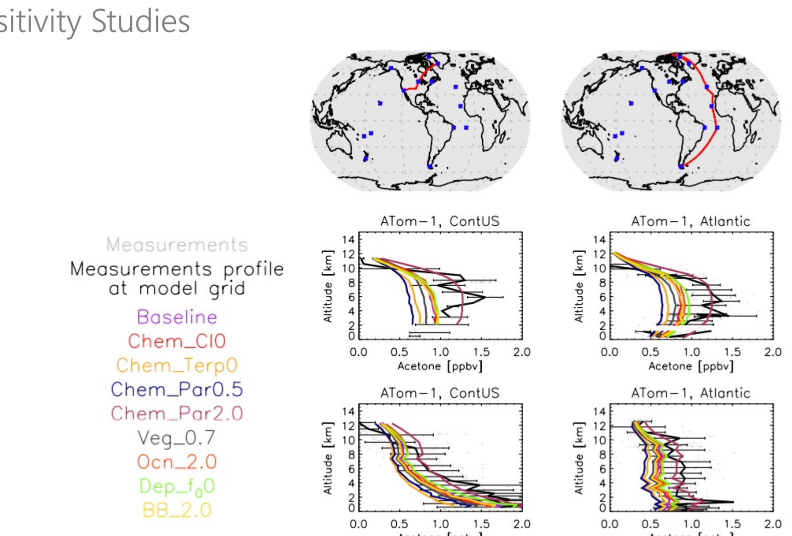
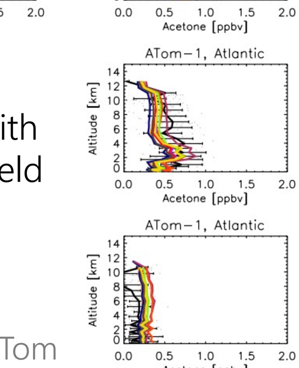


Fig 10. Baseline vs Ocean Sensitivity Fluxes

Production depends more on ocean acetone concentration than destruction (Figure 10)





The baseline simulation and sensitivities are compared with ATom remote atmosphere field measurements (Figure 11).

Fig 11. Sensitivities and ATom

6. Conclusions and Future Work

Extensive research was conducted to assess the simulated global acetone budget in context with past modeling literature. The model agrees well with vertical profiles (ATom) and surface field measurements. The chemical formation of acetone from precursor compounds was found to be an uncertain yet impactful factor. Vegetation was observed as the dominant acetone source with high seasonality, and the ocean acetone concentration was found to have nonuniform impacts on the budget. A limitation of the model is that its resolution may be too coarse to capture high-emission urban areas.

A scientific paper on this project is in progress. Future work involves using the same methodology for improving other trace gases in the model, as well as assessing potential feedbacks between acetone and the rest of the chemistry. Additionally, a non-uniform ocean acetone concentration will be tested. Overall, an analysis of the acetone budget aids the development of the tracer in the GISS ModelE, a crucial step to parameterizing the role of acetone in the atmosphere.

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