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Evaluating Effects of H_2O and Overhead O_3 on Global Mean Tropospheric OH Concentration

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1. Scientific motivation for studying tropospheric OH

Sensitivity of OH^{TROP} to climate change

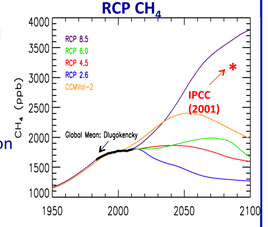
- IPCC (2001) predicted future OH would fall due to rising CH_4
- Best knowledge of global tropospheric OH (OH^{TROP}) comes from analysis of CH_3CCl_3 observations
- The CH_4 lifetime inferred from OH^{TROP} based on CH_3CCl_3 is 8.9 years; yet many sources use a CH_4 lifetime of ~12 years, which was found by modeling studies described in IPCC (2001)
- The 12 year CH_4 lifetime is central to subsequent IPCC reports and is called the "perturbation lifetime"
- Our work is motivated by understanding:
  - what factors other than rising CH_4 will affect OH^{TROP}
  - veracity of notion that future OH^{TROP} will decline

Importance of OH^{TROP}

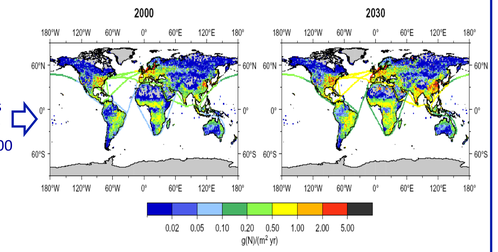
- OH serves as the main sink for many species: CO, CH_4, CH_3CCl_3, all HCFCs and HFCs, biogenic VOCs (isoprene and terpene), anthropogenic VOCs (formaldehyde, benzene, toluene, etc.)
- HO_x is integral to O_3 formation and destruction pathways; alone, HO_x depletes O_3, but with NO_x and VOCs, HO_x creates O_3

Methane Oxidation and OH

- Under low NO_x, the oxidation of CH_4 destroys HO_x
- Under high NO_x, the oxidation of CH_4 produces HO_x
- **Loss of methylperoxy radical CH_3O_2 with NO or HO_2 is critical**
- Future levels of CH_4 and NO_x are highly uncertain
  - CH_4 between now and 2100 varies dramatically among RCP scenarios
  - NO_x between now and 2100 will depend on whether the developing world implements selective catalytic reduction on coal power plants and catalytic converters on cars
- Future OH^{TROP} will also depend on overhead column O_3, local humidity, and biogenic emission of VOCs, etc.
- ***Given this complexity, we expect CCMs to project a wide range of values for OH^{TROP} during the rest of this century***



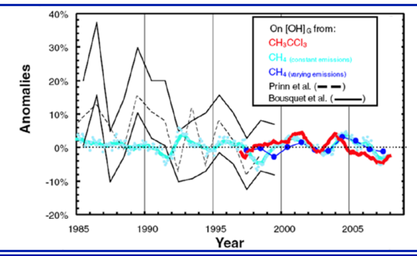
NO_x emissions from industry, power generation, traffic, domestic heating, and biomass burning used as input for prior CCM calculations, for years 2000 (38.0 Tg N / yr total) and 2030 (67.6 Tg N / yr total). From Eyring et al., *ACP*, 2007.



3. Preliminary results for expected changes in OH^{TROP}

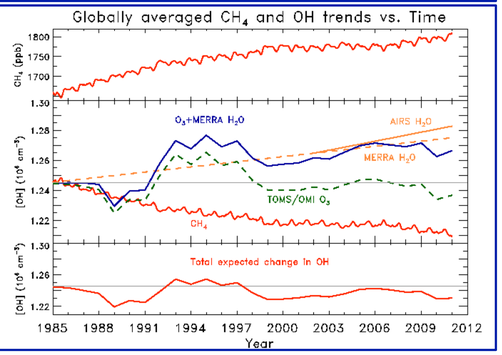
Previous Work on OH^{TROP} Trends

- Montzka *et al.,* 2011 found that OH^{TROP} does not vary interannually (from 1997 to present)
- Prior studies by Prinn *et al.*, 2001 and Bousquet *et al.*, 2005 suggest large interannual variability in OH^{TROP} (1985-2000)
- We suggest the OH^{TROP} behavior in all three studies may be physically possible, based on our preliminary results



Current Conclusions from OH^{TROP} Analysis

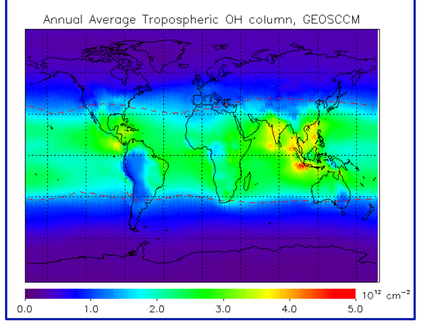
- Effect of CH_4 on OH^{TROP} is taken from IPCC (2001), Section 4.2.1.1, which states that "the feed-back of CH_4 on tropospheric OH" found using contemporary chemical transport models is −0.32% for every 1% increase in CH_4 (red line, middle panel of figure to right)
- Primary effect of overhead O_3 is rise in OH^{TROP} following the 1991 eruption of Mount Pinatubo (green dashed line) due to enhanced removal of stratospheric O_3 by volcanic aerosol
- Rising H_2O from MERRA and AIRS increases OH^{TROP} (orange lines) by an amount comparable to the decrease expected from rising CH_4
- Overall expected change in OH^{TROP} (bottom panel) shows higher level of interannual variability prior to ~1999 and lower variability thereafter

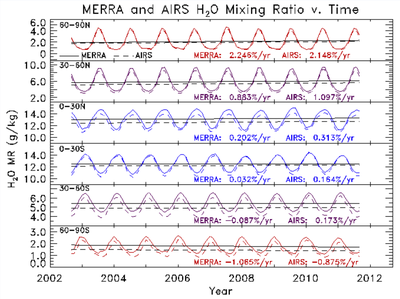


2. Methods for estimating changes in OH^{TROP}

Base Case OH Conditions

- Initial modern-day conditions for OH are taken from a time-slice run of GEOS CCM using 2005 emissions
- Monthly mean mixing ratios of OH and related species are provided on a 144 longitude, 91 latitude, 72 pressure level grid
- Calculated changes in OH due to H_2O and overhead O_3 are applied to initial OH field



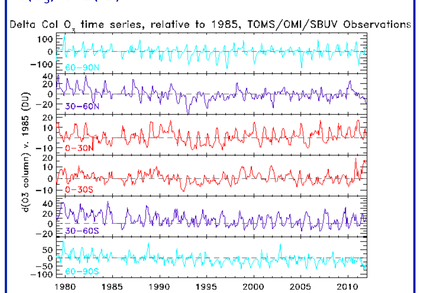


MERRA & AIRS H_2O

- Specific humidity files from the NASA Modern-Era Retrospective analysis for Research and Applications (MERRA) starting prior to 1985, and
- H_2O mixing ratio files from the NASA Atmospheric Infrared Sounder (AIRS) starting in 2002 were used to establish trends in H_2O by latitude
- OH^{TROP} is assumed to follow the square root of the change in H_2O using a steady-state assumption
- We plan to refine the effect of changing H_2O on OH^{TROP} by examining reaction rates from archived runs of GEOS CCM

Overhead O_3 Observations

- Total column O_3 trends were obtained from the NASA merged O_3 data set, consisting of measurements from SBUV, TOMS, and Aura OMI instruments
- We then use our photolysis code to estimate the impact on $J(O_3) \rightarrow O(^1D)$ of decreasing initial GEOS CCM overhead O_3 columns by amount suggested by the NASA product
- OH^{TROP} is assumed to change by the square root of $J(O_3) \rightarrow O(^1D)$



4. Future plans for refining estimates of ΔOH^{TROP}

Improve Estimate of $d(OH^{TROP}) / d(H_2O)$

- Reaction rates from recent runs of GEOS CCM are archived for reactions such as:

  $H_2O + O(^1D) \rightarrow 2OH$

- Using these reaction rates we will determine the proportion of OH that is produced via reaction with H_2O
- The determined scaling factor would be used to calculate a new ΔOH^{TROP} based on the H_2O trends
- Estimate time- and pressure-varying values of $d(OH^{TROP}) / d(H_2O)$
- Evaluate discrepancies between MERRA and AIRS H_2O trends

Evaluate CH_4/OH Feedback

- We will use a box model (details below) to probe relationship between CH_4 and OH^{TROP} and its dependence on NO_x

Box Model

We will use the GSFC Combined Stratosphere-Troposphere (COMBO) box model provided by Chang Lang (JHU):

- GMI chemical mechanism
- 118 species, 321 thermal reactions, and 81 photolysis reactions
- 5 modules:
  - –Aerosol optical depth & surface area
  - –Photolysis scheme
  - –Thermal reactions scheme
  - –Differential eqn solver
  - –Input-output
- Fast-JX photolysis & SMVGEAR II solver

Propagate Uncertainties

- Calculate uncertainties in AIRS and MERRA H_2O and NASA O_3 product
- Evaluate standard deviation in average fraction of OH production occurring via $H_2O + O(^1D)$
- Estimate uncertainty in the box model evaluation of $d(OH^{TROP}) / d(CH_4)$

Assessing OH^{TROP} in CCMs

- Through our involvement with the IGAC / SPARC Chemistry-Climate Model Initiative, we have requested:
  1. hourly, instantaneous output from participating CCMs 1 day/season, 1 year/decade
  2. archival of all species, reaction rates, J-values, and physical parameters relevant to OH chemistry
  3. this output for both the REF-C1 (hindcast) and REF-C2 (future) runs
- We plan to assess the causes of differences between OH in the CCMs
- Use of the box model enables us to distinguish between OH differences due to ***chemical mechanism*** and those due to ***differences in OH precursors***
- We can also use this output to predict future trends in OH^{TROP}, based on CH_4, H_2O, and overhead O_3 from the future CCM runs