Atmospheric Methane Distribution, Trend, and Linkage with Surface Ozone

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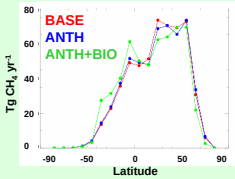
1. Introduction

- Methane (CH_4) emission controls can be a cost-effective strategy for abating both global surface ozone (O_3) and greenhouse warming [*West and Fiore*, 2005; see also poster by *West et al.*]
  → previous modeling studies used fixed CH_4 concentrations and globally uniform changes, but CH_4 is observed to vary spatially and temporally
- The major sink of CH_4 is reaction with tropospheric OH; emissions of CH_4 are shown in Section 2
- Surface CH_4 rose by ~5-6 ppb yr^{-1} from 1990-1999, then leveled off (Section 3), possibly reflecting:
  (1) source changes of CH_4 [*e.g. Langenfelds et al.*, 2002; *Wang et al.*, 2004] or other species that influence OH [*e.g. Karlsdóttir and Isaksen*, 2000]
  (2) meteorologically-driven changes in the CH_4 sink [*e.g. Warwick et al.*, 2002; *Dentener et al.*, 2003; *Wang et al.*, 2004]
  (3) an approach to steady-state with constant lifetime [*Dlugokencky et al.*, 2003]



Latitudinal distribution of 1990 CH_4 emissions for cases shown below

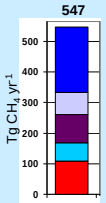
What is driving observed CH_4 trends? Does CH_4 source location influence the O_3 response?

2. Methane in the MOZART-2 CTM

Sensitivity simulations applying different CH_4 emission inventories:

- ~100 gas and aerosol species, ~200 reactions
- NCEP meteorology 1990-2004
- 1.9° latitude x 1.9° longitude x 64 vertical levels
- detailed description in *Horowitz et al.* [2003]

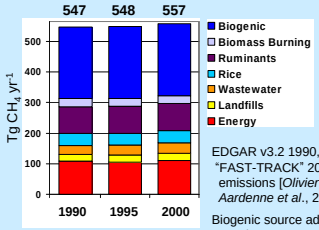
BASE
Constant emissions (1990)



Biogenic and biomass burning from *Horowitz et al.* [2003]

Anthropogenic (energy, rice, ruminants) from EDGAR 2.0 [*Olivier et al.*, 1999]

ANTH
Time-varying anthropogenic emissions

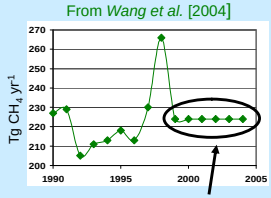


EDGAR v3.2 1990,1995 and "FAST-TRACK" 2000 anthrop. emissions [*Olivier*, 2002; *van Aardenne et al.*, 2005]

Biogenic source adjusted to match BASE 1990 total

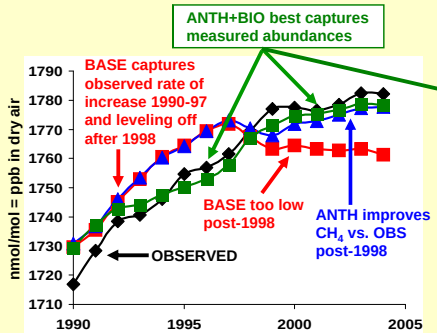
ANTH + BIO
Time-varying anthropogenic and wetland emissions

From *Wang et al.* [2004]



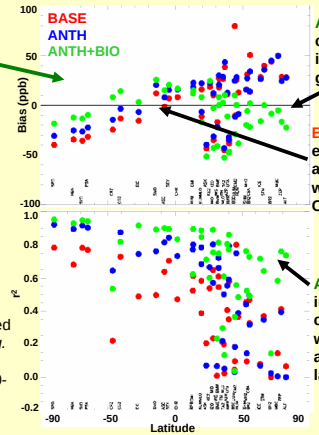
Apply climatological mean post-1998, scaled to equal biogenic total in ANTH (224 Tg yr^{-1})

3. Influence of Sources on Surface CH_4 Distribution and Trend



Global mean surface CH_4 concentrations as measured (or sampled in the model) at 42 Global Monitoring Division (GMD) stations [*e.g. Dlugokencky et al.*, 2005] with an 8-year minimum record. Values are area-weighted after averaging in latitudinal bands (60-90N, 30-60N, 0-30N, 0-30S, 30-90S).

Mean model bias and correlation with 1990-2004 monthly mean surface GMD observations

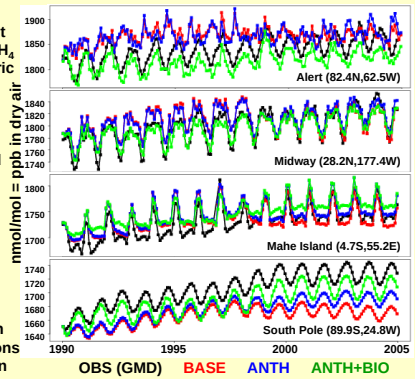


ANTH+BIO best captures the CH_4 interhemispheric gradient

BASE wetland emissions yield a closer match with observed CH_4 in tropics

ANTH+BIO improves the correlation with with observations at high northern latitudes

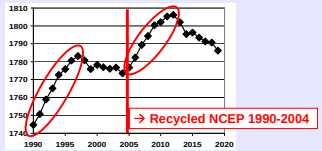
Surface CH_4 concentrations at selected GMD stations



ANTH+BIO improves:
(1) High N latitude seasonal cycle, (2) Trend,
(3) Low bias at S Pole, especially post-1998

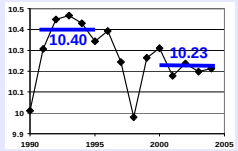
4. Meteorologically-driven Changes in the CH_4 Lifetime

Global mean surface CH_4 in BASE simulation (constant emissions)



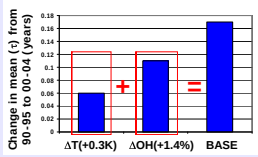
→ Meteorological drivers for trend
→ Not just an approach to steady-state

CH_4 Lifetime Against Tropospheric OH



→ Mean annual CH_4 lifetime shortens

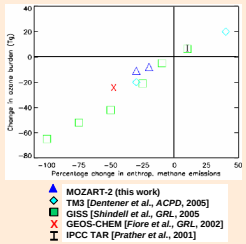
Deconstruct $\Delta\tau$ from 91-95 to 00-04 into individual contributions by varying T and OH separately



→ OH increases in the model by +1.4% due to a 0.3 Tg N yr^{-1} increase in lightning NO_x

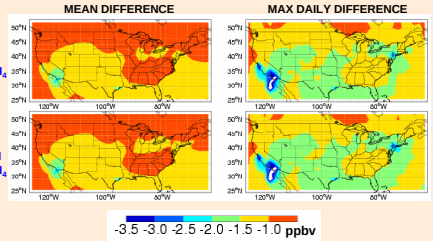
5. Ozone Response to CH_4 Emission Controls

Tropospheric O_3 response to anthropogenic CH_4 emission changes is approximately linear

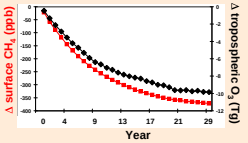


Simulations of anthropogenic CH_4 emission reductions (relative to BASE)

Change in summertime U.S. afternoon surface O_3



Change in CH_4 and O_3 approaching steady-state after 30 years



→ Stronger sensitivity in NO_x-saturated regions (Los Angeles), partially due to local O_3 production from CH_4
→ O_3 change independent of CH_4 source location except for <10% effects in the Asian source region

6. Conclusions

- **Ozone response is largely independent of CH_4 source location**
- **30% decrease in global anthropogenic CH_4 emissions reduces JJA U.S. surface afternoon O_3 by 1-4 ppbv**
- **BASE simulation (constant emissions) captures observed rate of CH_4 increase from 1990-1997, and leveling off post-1998**
- **ANTH emissions improve modeled CH_4 post-1998**
- **Wetland emissions in ANTH+BIO best match the observed CH_4 seasonality, interhemispheric gradient, and global mean trend**
- **τ_{CH4} decreases by ~2% from 91-95 to 00-04 due to warmer temperatures (35%) and higher OH (65%, resulting from a ~10% increase in lightning NO_x emissions)**

Future research should:

- consider climate-driven feedbacks from fire and biogenic emissions on τ_{CH4}
- develop more physically-based parameterizations of lightning NO_x emissions to determine whether higher emissions are a robust feature of a warmer climate

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