Isotopic constraints on the decadal trends of global methane emissions favor increasing fossil fuel emissions over recent decades

C.L. Butenhoff (cbuten@pdx.edu), A.L. Rice, F.H. Röger, D.G. Teama, M.A.K. Khalil, R. Rasmussen

Dept. of Physics, Portland State University, Portland, OR, 97207

BACKGROUND

Despite concerted effort in recent years to understand the changing budget of atmospheric methane (CH₄), there remains considerable uncertainty on the trends and magnitudes of individual methane sources over decadal scales. This uncertainty reduces confidence in our ability to forecast future changes and impedes efforts to mitigate climate change. Of particular interest to two recent studies have linked a decrease in methane mixing ratios to a decrease in fugitive fossil fuel CH₄ emissions. This top-down result however disagrees with bottom-up inventories that show flat or increasing fossil fuel CH₄ emissions. Recently archived air sampled at a marine Northern Hemisphere mid-latitude site (Cape Meares, Oregon, 45°N, 124°W) spanning the years 1978 to 1999 was analyzed for methane isotope abundance. The isotopes of methane (δ¹³C and δD) place important constraints on the emission history of methane over the past few decades. These new measurements, available at roughly monthly resolution, extend the existing isotope record and provide new insight into the behavior of atmospheric CH₄. We present a time-dependent synthesis inversion of the global 3D Chemistry Transport Model GEOS-Chem to determine source histories consistent with measured CH₄ and δ¹³C-CH₄. Our main objectives are (1) to use Bayesian inversion to estimate source trends from new and existing CH₄ and δ¹³C-CH₄ data; (2) determine if δ¹³C-optimized emissions are consistent with new and existing δD-CH₄ data; and (3) determine if recent fugitive fossil fuel methane emission scenarios are consistent with the observed CH₄ and isotope record.

A detailed description of the Cape Meares air archive analysis can be found in Teama (2013). Here we describe the time-dependent retrieval of CH₄ fluxes using the GEOS-Chem CTM (v9-01-03) with the following setup:

- Ten methane source categories based on their carbon isotopic signature and geographical distribution
- Optimization of fluxes was performed using a fixed-lag (11 months) Kalman smoother
- A priori emissions of most sources taken from Edgar2.1
- Biomass burning taken from GFED3.2
- Wetland emissions by a scheme described by Pickett-Heaps et al.
- Base inversion uses static monthly varying 3D OH fields with global annual mean of 10.1 ppm
- CH₄ observations from NOAA ESRL GLOBALVIEW-CH₄ product
- ¹³CH₄ data from current work, SIL INSTAAR, Tyler et al.3, Quay4, Francey et al.5. The total record includes 17 sites
- Systematic errors were estimated by 42 sensitivity tests (inversions)

METHODS

A time-dependent retrieval of methane fluxes spanning nearly twenty-five years has been performed using new measurements of atmospheric methane isotopes from an archive of stored air taken at Cape Meares OR.

The inversion was able to reproduce CH₄ and δ¹³C successfully at nearly every site. δD data was well-simulated by δ¹³C-optimized emissions up until year 2000, after which the simulated δD significantly exceed observed data.

The inversion estimates a ~30 Tg CH₄ increase in fugitive fossil fuel emissions since 1985 with the highest growth rate occurring after year 2000. This result is consistent with some bottom-up estimates but is not consistent with recent estimates based on atmospheric methane and other inverse studies. The model also estimates an overall decrease in biomass burning emissions since 1985 with most decrease attributed to C3 vegetation.

If the inversion is forced using a fugitive fossil fuel scenario consistent with recent ethane measurements, emissions from waste (e.g. landfills) greatly exceed estimates from bottom-up inventories suggesting this scenario is not consistent with methane isotope data.

SUMMARY

Fig. 2 Comparisons between measured (blue) and simulated (a) monthly-mean CH₄ and (b) δ¹³C-CH₄ at a range of latitudes show good agreement. Simulations based on a posteriori emissions are shown with the black line, while simulations based on the prior emissions are shown with the red line and indicate the improvement made by the inversion. Blue shadows represent uncertainty ranges of the measurements.

Fig. 3 Desasonalized emission anomalies from (a) aggregated fossil fuel, biomass burning, wetland, and biogenic source categories, and (b) from the ten separate source categories. Colored lines in (a) and dark blue lines in (b) indicate the inversion results. Red lines in both indicate anomalies in the prior emissions. Shaded areas represent the full range (light) and 90% (dark) of the inversion scenarios. Note long term upward trend in the fossil fuel emissions and downward trend in biomass burning and wetland categories.

MODEL-DATA COMPARISON

Fig. 1 List of the different inversion scenarios performed to test sensitivity of inversion results

FOSSIL FUEL EMISSION SCENARIOS

Fig. 5 Testing fugitive fossil fuel scenarios Recent studies suggest fugitive fossil fuel emissions were flat or decreased from 1985-2000 with an uptick after 2005. We reran the inversion with various imposed fossil fuel emission scenarios to test the effect on the other sources (1-6). Red lines are the S1 scenario from Kirschke et al.11 assigning all changes to gas-soil (solid) and split between gas-oil and coal-biofuel (dotted). A constant fossil fuel source is shown by the green line, and the Edgard2 fossil fuel scenario is shown by the blue line. The base inversion emissions are shown by the black line. The table summarizes changes in emissions for each scenario based on linear regressions from 1985-2000 and 2000-2009. Major deviations are highlighted by the red box.

Fig. 4 Mean methane source strengths. (a) Comparison of a priori and posteriori source strengths over the time period 1985-2008. The results from the S1 inversion scenario are taken as posterior values. The errors on the posterior emissions are the emission-calculated standard deviations (i.e. internal errors), while the ensemble spread is the difference between the largest and smallest estimate of all inversion scenarios. Values in parentheses are the corresponding interquartile range containing 50% of all inversions. (b) Box plots for the ensemble of inversions. Boxes contain the interquartile range of the data, inside line is the median, whiskers extend to the lower of max/min or 1.5*IQR. Outliers are identified by open circles. The red x’s are the priors and the black lines indicate the range of emission estimates from IPCC AR4. Purple circles are estimates from inversion scenario S20.

REFERENCES


ACKNOWLEDGEMENTS

We are grateful to the GEOS-Chem support team in providing and maintaining GEOS-Chem source code. A special thanks goes to Kevin Wecht at Harvard for assistance with GEOS-Chem CH₄ simulation. We acknowledge NOAA-ESRL for use of the GLOBALVIEW-CH₄ and INSTAAR Isotope data, and the Miller Foundation for funds to support computing infrastructure at PSU. This research was supported in part by the National Science Foundation – Atmospheric and Geospace Sciences award 0952097.