INTERANNUAL METHANE SOURCES AND SINKS INFERRED BY INVERSION OF ATMOSPHERIC TRANSPORT AND CHEMISTRY

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The results of an optimisation of inter-annual methane sources and sinks calculated by inversion of atmospheric observations are presented and analysed for the 1984-2003 period. We focus our presentation on sources trend and inter-annual variability. Comparisons with bottom-up estimates are presented for biomass burning and wetlands emissions (only in the poster).

Atmospheric methane observations reveal a large inter-annual variability as well as a general decline in the growth rate after 1991. The underlying processes are mostly identified but their timing and magnitude remain largely unknown. Chemical transport models (CTM) can be used in conjunction with atmospheric measurements, to analyse methane atmospheric signals in terms of surface fluxes or OH variations.

We developed a combined atmospheric inversion of methyl Chloroform (CH_3CCl_3) and methane (CH_4) atmospheric observations in order to optimize inter-annual OH concentrations and CH₄ surface sources and sinks, for the 1984-2003 period. The inverse procedure infers CH₄ monthly sources and sinks using INCA-LMDZ chemical transport model [Hauglustaine et al., 2004] nudged on analysed winds from ECMCF. LMDZ-INCA has been used in a forward mode to generate chemical response functions, that is the impact of OH sink for 4 three-dimensional latitudinal regions (30-90°N, 0-30°N, 0-30°S, 30-90°S) at measurement sites, and in a backward mode to calculate the source response functions, that is the impact of surface sources and sinks at measurement sites. The non-linear problem was linearized, which imposed to set up an iterative procedure described in details in Bousquet et al. [2005]. As a first step, OH variations and mean values were optimized using the iterative inverse procedure against CH₃CCl₃ measurements. In a second step, optimized OH fields are prescribed in the methane inversion in which surface sources only are optimized. Processes are solved separately over large regions based on TRANSCOM spatial partition [Gurney et al., 2002]. New¹³CH₄ time series have been included in the inversion to discriminate between methane emissions of different nature, mainly bacterial, biomass burning and fossil related sources. Additional constraints were added to the inverse procedure in order that month-to-month differences of methane sources that are known to vary slowly in time or to have only small seasonal variations (animal emissions, fossil related emissions) do not become unrealistic [Peylin et al., 2002].



Fig. 1: Global CH4 flux anomaly in TgCH4/yr as inferred by the inverse procedure. Red triangle figure out the results obtained by Dlugokencky et al. [2003] based on methane observations and a one-box model of the atmosphere. The inversion with varying OH (purple line) presents the largest IAV in methane sources.

 Y_{ear} At global scale, we find a mean source of 520±20 TgCH₄/yr for the period 1984-2003. Inter-annual variations of ±20 TgCH₄/yr are found, OH variations being the

main cause of differences at this scale (Fig. 1). The comparison with observation-based estimate by Dlugokencky is good, except when accounting for possible OH variability given by CH₃CCl₃ inversion. The 1991 rapid falling and the 1997-98 positive anomaly are clearly visible on global methane source (Fig. 1). Global source does not present significant trend over the long-term, hiding a compensatory effect between an increasing source in the tropics due to enhanced wetlands emissions and a decreasing source over North Asia and Europe, due to fossil-related sources and bogs emissions.

Adding ¹³CH₄ observation makes it possible to perform process-based inversions instead of only geographically based. Figure 2 plots the global biomass burning methane source, as inferred by the inverse procedure and calculated from satellite fire products by Van der Werf et al. [2003].



Fig.2. Global CH4 flux anomaly due to biomass burning in TgCH4/yr as inferred by the inverse procedure. Red triangle figure out the estimates of Van der Werf et al. (2003) based on satellite fire products. Blue triangle shows the variations of fire counts given by satellite. Two inversions are plotted, the reference case (black line, 1984-2003) and the inversion using ¹³CH₄ measurements (green line, 1994-2003).

The agreement between inversions and satellite-derived biomass burning flux estimates is good in phase. The three major fire anomalies detected by satellite (1991-92, 1994-95 and 1997-98) are caught by the inversion. Moreover, the use of ¹³CH₄ measurements increased the amplitude of the inverse retrieval of the 1997-98 anomaly and made it closer to Van der Werf estimate. The rest of the 1997-98 positive atmospheric anomaly is found to be due to wetlands emissions (not shown). After 2001, when half of available ¹³CH₄ stations appear, one can notice that the results of Van der Werf and this work get closer. These promising results will have to be confirmed when more longer ¹³CH₄ time series will be available. However, the uncertainties on 13C discriminations are still limiting the impact of ¹³CH₄ observations. We also found a good agreement between inferred wetlands IAV and an estimate based both on a simple model of wetland emissions [*Fung et al.*, 1991] and on satellite-based estimates of flooded areas [*Prigent et al.*, 2001 & pers. Com.].

REFERENCES

- Bousquet, P., et al. (2005), Two decades of OH variability as inferred by inversion of atmospheric transport and chemistry of Methyl-Chloroform, *ACPD*, *5*, 1679-1731.
- Dlugokencky, E. J., et al. (2003), Atmospheric methane levels off: Temporary pause or a new steady-state? *Geophys. Res. Lett.*, 30, 1992.
- Gurney, K. R., et al. (2002), Towards robust regional estimates of CO2 sources and sinks using atmospheric transport models, *Nature*, 415, 626-630.
- Hauglustaine, D. A., et al. (2004), Interactive chemistry in the Laboratoire de Meteorologie Dynamique general circulation model: Description and background tropospheric chemistry evaluation, *J. Geophys. Res.- atmos.*, 109, 3414.
- Peylin, P., et al. (2002), Influence of transport uncertainty on annual mean and seasonal inversions of atmospheric CO2 data, J. Geophys. Res.-atmos., 107

Prigent C. et al., Remote sensing of global wetland dynamics with multiple satellite data sets, GRL, 28, 2001.

Van der Werf, G. R., et al. (2003), Carbon emissions from fires in tropical and subtropical ecosystems, *Glob Change Biol*, 9, 547-562.