

**Source attribution of
the changes in
atmospheric methane
for 2006–2008**

P. Bousquet et al.

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Source attribution of the changes in atmospheric methane for 2006–2008

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Abstract

The recent increase of atmospheric methane is investigated by using two atmospheric inversions to quantify the global distribution of sources and sinks for the 2006–2008 period, and a process-based model of CH₄ emissions by natural wetland ecosystems. Global emissions derived from inversions are found to have increased by 19 Tg on average in 2007 (16 to 21 Tg) and by 13 Tg in 2008 (6 to 20 Tg), as compared to the 1999–2006 period. A positive anomaly of tropical emissions is found to be the main contributor to the global emission anomaly of 2007 (~60–75%), with a dominant share attributed to natural wetlands (~66%). Abnormally high wetlands emissions from high latitudes are also detected by both inversions in 2007, contributing 15–30% of the global anomaly. Good agreement is found between the results of the wetland ecosystem model and the inversions for 2007. The inferred distribution of the source anomaly in 2007 is shown to be consistent with the observation of a more pronounced increase in near surface methane atmospheric growth rate at high latitudes, because the dilution of surface fluxes by convection is strong in the tropics and weak at high latitudes. The source anomaly in 2008 is found to be much larger in the wetland ecosystem model than in the inversions, suggesting a too strong sensitivity of bottom-up modeled emissions to precipitation. Changes in OH radicals during 2006–2008 are found to be less than 1% in inversions, with only a small impact on the inferred methane emissions.

1 Introduction

Atmospheric methane (CH₄) is a major greenhouse gas and plays a key role in the production of tropospheric ozone (IPCC, 2007). After a decade of near stable concentrations, the growth rate of atmospheric methane has started to increase again, with changes of 8.0±0.6 ppb in 2007 and 6.3±0.6 ppb in 2008 (update from Dlugokencky et al., 2009). The main sources of CH₄ are natural wetlands, anthropogenic activities (livestock production; rice cultivation; production, storage, transmission, and distribu-

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tion of fossil fuels; waste waters and landfills), and biomass burning, both natural and human-induced. Global emissions are between 500 and 600 TgCH₄ yr⁻¹, with 60–70% being of anthropogenic origin (IPCC, 2007). The destruction of CH₄ by OH in the troposphere represents about 90% of CH₄ loss in the atmosphere, making the atmospheric CH₄ budget very sensitive to OH changes. The rest of the sink is due to an uptake of CH₄ by soils, reaction with Cl in the marine boundary layer, and to destruction in the stratosphere by reactions with OH, Cl, and O(¹D) (IPCC, 2007).

Because anthropogenic CH₄ emissions only change gradually with time, the year-to-year variability of CH₄ emissions is controlled by wetland emissions (Bousquet et al., 2006; Chen and Prinn, 2006) and by biomass burning emissions, the latter being estimated to play a significant (Bousquet et al., 2006) or a dominant role (Langenfelds et al., 2002) in particular during climate events such as the 1997–1998 El Niño or the 2002–2003 dry period over the northern mid-latitudes (Simmonds et al., 2005). The role of the OH sink in atmospheric CH₄ variations may be significant (Bousquet et al., 2005; Prinn et al., 2005; Rigby et al., 2008) but it is still controversial, given discrepancies in the magnitude of OH interannual variations calculated by atmospheric chemistry models (~1–3%; Dentener et al., 2003; van Weele et al., 2009) or estimated by atmospheric inversions based on 1,1,1-trichloroethane (~4–10%, Bousquet et al., 2005; Krol et Lelieveld, 2003; Prinn et al., 2001).

Several studies have addressed the question of the apparent stabilization of atmospheric methane during 1990s. The collapse of the former USSR economy led to a decrease of CH₄ emissions in the 1990s (Dlugokencky et al., 2003). Indeed, the EDGAR4 inventory of anthropogenic emissions (EDGAR4 database: European Commission, 2009) do show a decrease in CH₄ emissions from continental Europe (including western Russia) between 1990 and 2005, and a stagnation in North America, but a significant increase from emerging countries, especially China, contributing to an increase in tropical and East Asian CH₄ emissions. Overall, global anthropogenic CH₄ emissions are estimated to have increased by 32 Tg since 1990 in the EDGAR4 inventory, especially after 1999. In the global inversion of Bousquet et al. (2006), the low

growth rates of the late 1990s and early 2000s, is attributed to competing influences of increasing anthropogenic emissions and decreasing natural wetland emissions, consistent with the EDGAR4 inventory trends and with drier conditions encountered in various regions of the Northern Hemisphere in the late 1990s/early 2000s (Hoerling and Kumar, 2003).

Various lines of evidence point out to natural wetlands playing a dominant role in the recent increase of CH₄ atmospheric growth rate since 2007. Surface temperature and precipitation anomalies during years 2007 and 2008 were large and positive over the main wetland regions emitting CH₄. A very high annual mean temperature was recorded over Siberia in 2007 mainly in autumn (+4 °C compared to 1961–1990, National Climatic Data Center, 2008), a region with large wetland areas. Tropical areas, strong contributor of wetlands area at global scale, experienced the 3rd-largest (2007) and the largest (2008) positive precipitation anomalies from 1986 to 2008 (Dlugokencky et al., 2009; Schneider et al., 2008). In the tropics, processes relative to hydrology appear to be the dominant driver of wetland CH₄ emissions, whereas temperature leads at high latitudes (Walter et al., 2001; Ringeval et al., 2010c). An analysis of CO observations, a tracer used as a proxy for biomass burning emissions, further indicates that the 2007 positive CH₄ growth rate anomaly is not related to biomass burning in northern latitudes (Dlugokencky et al., 2009). Biomass burning in the Tropics could have contributed to the 2007–2008 CH₄ growth rate anomaly, but not as a dominant factor (Dlugokencky et al., 2009; Rigby et al., 2008). Based on analysis of observations of CH₃CCl₃, Dlugokencky et al., (2009) suggest no significant contribution to the CH₄ anomaly in 2007 from decreasing [OH], contrary to Rigby et al. (2008) who inferred a –4% decrease of OH from 2006 to 2007 but with a large uncertainty (±14%) that make the two estimates statistically compatible.

Atmospheric inversion is a powerful tool to infer the time-varying distribution of regional sources and sinks of CH₄ by assimilating atmospheric observations in a model of atmospheric chemistry-transport using prior information of the surface CH₄ fluxes (Bousquet et al., 2006; Chen and Prinn, 2006; Bergamaschi et al., 2005; Hein et al.,

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1997; Houweling et al., 1999). However, as with most top-down approaches using atmospheric observations, atmospheric inversions can hardly provide insights on the underlying processes causing the emissions. On the other hand, ecosystem models calculating wetland or fire emissions incorporate knowledge of local processes, but often need additional constraints to up-scale their local estimates to regional and global scales for producing CH₄ large-scale emissions that are compatible with the global atmospheric signals (Ringeval et al., 2010a).

In this paper, we investigate of the changes in atmospheric CH₄ for 2006–2008 using the results of two atmospheric inversion models (Bousquet et al., 2006; Pison et al., 2009) and of a new ecosystem model for CH₄ wetland emissions (Ringeval et al., 2010b).

2 Methods

2.1 Inversion models.

We use two different inversion models, both based on the Bayesian formalism. CH₄ observations are assimilated into an atmospheric chemical-transport model together with prior information on the spatio-temporal distribution and uncertainties of CH₄ sources and sinks to estimate the magnitude and the uncertainties of optimized surface emissions.

The first inversion model (hereafter referred as INV1) is an analytical inversion that has been used to infer the sources and sinks of CO₂ and CH₄ (Bousquet et al., 2000; Peylin and Bousquet, 2005; Bousquet et al., 2005, 2006) and recently H₂ (Bousquet et al., 2010). Briefly, it solves for monthly surface CH₄ emissions for the different processes source types and for 11 large regions (10 land regions + 1 ocean), using monthly mean observations at up to 68 surface stations from the NOAA/ESRL, CSIRO and IPSL/LSCE surface monitoring networks. The offline version LMDZt version 3 of the LMDZ-GCM, nudged on analysed winds (Uppala, 2005), is used to model atmo-

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spheric transport (Hourdin and Talagrand, 2006; Hourdin et al., 2002). Prior emissions are taken from inventories (Matthews and Fung, 1987; Olivier and Berdowski, 2001; van der Werf et al., 2006). The OH tree-dimensional fields are pre-optimized by an inversion of CH_3CCl_3 (MCF) observations as described in Bousquet et al. (2005).

5 Monthly uncertainties are prescribed for prior CH_4 emissions of $\pm 150\%$ for each region, and for CH_4 observations (from ± 5 ppb to ± 50 ppb, with a median of ± 10 ppb), with no error correlations. A simple filter is also added in the time domain: changes of the inferred fluxes from one month to the next are limited to $\pm 150\%$ (sources with a seasonal cycle in the prior) or to $\pm 10\%$ (sources with no seasonal cycle in the prior)
10 of the prior month-to-month differences, according to Peylin et al. (2002). This noise filter avoids the creation of unrealistic large month-to-month flux differences. A more complete description of the method can be found in Bousquet et al., (2005). We define a reference inversion scenario (S_0) based on these assumptions, complemented by 10 additional scenarios using: climatological instead of interannually varying OH (S_1),
15 prior natural wetland emissions based on (Kaplan, 2002) (S_2), only observations from the NOAA/ESRL surface network, representing roughly 50 sites (S_3), only sites with valid observations along the 1984-2008 period (S_4), all sites with at least 5 years of measurements (S_5), uncertainties on observations divided by two (S_6) or multiplied by two (S_7), uncertainties on prior fluxes divided by two (S_8) or multiplied by two (S_9), and
20 no filtering to limit the month-to-month noise in inferred fluxes (S_{10} , see Peylin et al., 2002). This ensemble of 11 inversions provides monthly optimized CH_4 emissions per region and per emitting process, and the residual uncertainties.

The second inversion model (here after referred to as INV2) is a variational inversion based on (Chevallier et al., 2005) coupled with the Simplified Assimilation Chemical System (SACS) developed by (Pison et al., 2009) and the LMDZt transport model
25 version4, which has an improved parametrization of mixing in the planetary boundary layer (Hourdin and Talagrand, 2006) as compared to version3 used in INV1. The SACS assimilation package represents a simplified methane oxidation chain, keeping only the main reactions linking CH_4 to CO and H_2 , through reactions with hydroxyl radi-

cals (OH) and formaldehyde (HCHO). The reaction between OH and methyl-chloroform (CH_3CCl_3) is also represented within the SACS as a constraint on OH concentrations. Only the total CH_4 flux is inferred in INV2, and not the individual source types as in INV1. The prior variance of fluxes in each grid cell are set to $\pm 100\%$ of the maximum flux over the eight neighboring grid cells and the current grid cell each month (Pison et al., 2009). The error correlations of the CH_4 fluxes are modeled using correlation lengths of 500 km on land and 1000 km on oceans, without time correlations (Chevallier et al., 2005). Daily mean CH_4 observations at continuous measurement stations and individual flask observations at flask stations are assimilated for the same stations as in INV1 to estimate weekly CH_4 emissions at the model resolution. The relevant cost function and the norm of its gradient computed by the adjoint of LMDZt and SACS are minimized with the algorithm M1QN3 (Gilbert and Lemaréchal, 1989). The inversion results consist in eight-day maps (7081 cells) of CH_4 emission fluxes and of four correction coefficients for the OH column abundances of four latitudinal bands of the same surface ($90^\circ\text{S}/30^\circ\text{S}$, $30^\circ\text{S}-0^\circ$, $0^\circ-30^\circ\text{N}$, $30^\circ\text{N}-90^\circ\text{N}$). INV2 was run from up to mid 2009, to avoid border effects, with 43 iterations and more than 99% reduction of the norm of the gradient of the cost function. A complete description on INV2 can be found in Pison et al., (2009). For comparison, global flux from INV2 was scaled to have the same mean value as in the INV1 inversion for the 1999–2006 period ($515\text{ TgCH}_4\cdot\text{yr}^{-1}$).

The main advantage of INV1 is the low computing cost because of the large-region approach and of the pre-calculation of transport and chemistry, that allows many sensitivity tests to be performed. Also, in INV1, we separate the different source types. The main advantage of INV2 is the estimation of CH_4 emissions on a fine grid, the same than the transport model, which avoids aggregation errors in the flux domain (Kaminski et al., 2001), and the assimilation of observations at the time of the measurements (and not as monthly means), which limits the aggregation error in the time domain.

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2.2 Model of natural wetlands emissions

Wetland CH₄ emissions were computed using the global vegetation model ORCHIDEE, which simulates land energy budgets, hydrology and carbon cycling (Krinner et al., 2005), and which was further developed to calculate CH₄ emissions from natural wetlands (Ringeval et al., 2010a, b). CH₄ emissions are calculated monthly for each 1° × 1° model grid cell as the product of an emitting water saturated area by a flux density for the period 1990-2008 (see Appendix A). The saturated areas within each grid cell are computed by the subgrid hydrology model TOPMODEL (Beven and Kirkby, 1979; Ringeval et al., 2010c) and scaled globally to the inundated areas derived from a suite of satellite observations from multiple sensors of (Prigent et al., 2001, 2007). The wetland CH₄ flux density is computed in each grid point using an update of Ringeval et al. (2010a) model, which was adapted from the Walter et al. (2001) model. Three pathways of transport (diffusion, plant-mediated transport and ebullition) and oxidation are included. Two different climate forcing datasets are used to drive the wetland emission model, one based on the CRUNSEP dataset developed by Viovy and Ciais (2009), and one with ECMWF precipitation replacing NCEP precipitation (see Appendix A). For comparison, global wetland emissions from ORCHIDEE were scaled to have the same mean value as in the INV1 inversion for the 1999–2006 period (165 TgCH₄ yr⁻¹).

3 Results

We use the 8-year period 1999–2006 during which atmospheric CH₄ abundance was rather stable (Table 1), as a reference period to calculate annual emission anomalies each year between 2006 and 2008 (Fig. 1, Table 2) for natural wetland CH₄ emissions (ORCHIDEE and INV1) and for total CH₄ emissions (INV1 and INV2). As INV2 do not separate source types, only the total CH₄ flux can be compared with INV1 total CH₄ flux.

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3.1 Global scale

At the global scale, a positive anomaly of CH₄ emissions in 2007 (resp. 2008) is inferred by both inversions, of 16±4 (resp. 6±4) Tg for INV1, and of 21 (resp. 20) Tg for INV2 (Fig. 1, table 2). The makes an average emission anomaly of +19 Tg in 2007 and of +13 Tg in 2008, as seen by the two inversions. These anomalies are statistically significant considering the estimated uncertainties. In INV1, the global source variations are dominated by natural wetlands in 2006 and 2007, and by a mix of non-wetland sources in 2008. The emission anomaly from natural wetlands represents ~75% of the 2007 global flux anomaly in INV1. This result is robust across the 11 inversions of the INV1 ensemble. Only the inversion (S2 in INV1) using another wetland prior distribution and prior values of the wetland fluxes produces a dominant positive anomaly for wetlands in 2008, illustrating the importance of the choice of the prior estimates and distributions (Fig. 1). A possible underestimation of the 2008 global emission anomaly in INV1 may be due to an end effect in INV1, with no observations assimilated after 2008. Inversion INV2, which assimilates observations until March 2009, enables to better constrain fluxes towards the end of 2008 than inversion INV1, and produces a positive anomaly in 2008 (+20 Tg) comparable to the 2007 anomaly (+21 Tg).

The optimized OH fields are lower by 1% and 0.5% in INV1 and INV2 respectively in 2007 and 2008, as compared to 2006. Therefore less CH₄ emissions are required to match the global growth rate constraint than when accounting for OH changes. This is qualitatively, but not quantitatively, in agreement with the results of Rigby et al. (2008), as they find a much larger decrease in OH in 2007 (-4%), implying a smaller increase in methane emissions in 2007 than in our study. Neither the simultaneous MCF inversion performed in INV2, nor the offline MCF inversion performed in INV1, supports large variations of OH from one year to the next as in Rigby et al. (2008). If OH radicals are maintained constant from one year to the next (sensitivity inversion S1 of INV1), we do find a larger positive anomaly for INV1 in 2007 (+27 Tg) and 2008 (+12 Tg) than with

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varying OH.

ORCHIDEE model provides a global positive flux anomaly for the three years 2006–2008 (as compared to the reference period 1999–2006). There is a good agreement of ORCHIDEE with the INV1 wetland flux anomaly in 2007. In 2008 the wetland flux positive anomaly given by the ecoystsem model (+38 Tg) is much larger than the global anomaly of INV1 (+6Tg) and of INV2 (+21 Tg), even when considering the estimated uncertainties. In 2006, ORCHIDEE does not agree on the sign of the anomaly with both INVERSIONS (see the analysis next section). Over 2006–2008, the ORCHIDEE run using ECMWF precipitation data is in better agreement with inversions, with significantly less anomaly than the one with NCEP precipitation, indicating a large sensitivity of ORCHIDEE to precipitation.

Finally, in INV1, other sources than natural wetlands contributing to the 2007 flux anomaly are biomass burning (+2±5 Tg, mostly in South America) and anthropogenic sources (+2±6 Tg). Contributions from landfills, mostly in Asia, dominate the anthropogenic flux anomaly in 2007 (not shown). In 2008, the anthropogenic flux anomaly (+8±6 Tg) explains the positive flux anomaly in INV1, as natural wetlands contribute a negative flux anomaly (−3±4 Tg, Table 2), with contributions from landfills, fossil fuels, and rice in Asia. The rest of the 2008 anomaly is due to biomass burning (+1±5 Tg). The partition between these sources may be uncertain because of the negative correlations of error existing between them. Error correlations can be calculated using the posterior covariance matrix calculated during the inverse procedure in INV1. If one assumes that errors are proportional to fluxes, as error correlations between two source types/regions get closer to −1, these two source types/regions are less and less well separated by the atmospheric observations. We find rather small error correlations between individual source types at global scale, as they range from 0 to −0.55. This result, although only qualitative, indicates a rather good capacity of INV1 to separate the different methane source types at global scale.

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3.2 Tropics

Tropical emissions are the largest contributor to the global anomalies of methane emissions for 2006-2008 for all models with one exception for INV2 in 2008. In 2007, higher emissions from tropical regions explain between 60% (INV2) and 75% (INV1) of the global flux anomaly. Natural wetlands dominate the tropical flux anomalies. INV1 attributes 67% of the Tropical flux anomaly and 50% of the global flux anomaly in 2007 to natural tropical wetlands (Table 2). In 2008, INV1 (+5±7 Tg) and INV2 (+3 Tg) agree on a smaller tropical anomaly, which is, in INV1, not attributed to wetlands but is spread over different type of sources.

The two INV1 scenarios with an alternative distribution of wetlands (S2) and with constant OH (S1) show the best agreement with the ORCHIDEE simulation driven by ECMWF precipitation in 2007 (Fig. 1). The small contribution of tropical wetlands in 2008 found in INV1 differs from the large positive anomaly calculated by ORCHIDEE (+32 Tg), which is directly linked to higher precipitation over tropical lands (+20% between 2005 and 2008) in both NCEP and ECMWF forcing datasets, although slightly smaller in ECMWF. The large increase of wetland emissions from 2006 to 2008 calculated by ORCHIDEE is not consistent with the smaller observed growth rate in 2008, unless another source is reduced or OH is increased, but neither possibility is supported by INV1 or INV2. ORCHIDEE may overestimate CH₄ emission changes from three main reasons. First, tropical floodplains are not explicitly represented in the model. Second, wetland extent can increase drastically in response to a positive anomaly of precipitation while “in reality”, this expansion does not necessary lead to a raise in methane emissions, for instance if inundation happens over a non-rich carbon soil. Third, larger precipitations may lead to an increase of CH₄ oxidation in the water above the ground, thus limiting methane emissions to the atmosphere. This effect is not included in Walter et al. (2001) model and thus in ORCHIDEE yet.

Another striking result for the tropics is the large negative anomaly of tropical wetland (and consequently of global) emissions inferred in 2006 by INV1 (−20 Tg for wetlands,

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–17 Tg for total emissions). INV2 also provides a negative flux anomaly, but smaller than in INV1 (–2 Tg) whereas ORCHIDEE provides a positive anomaly (+10 Tg). In INV1, the negative flux anomaly is mainly found over South America and Africa. Little surface observations are available to constrain the inversion over these two regions, making the inversion estimate more uncertain than for other regions. In this context, the space-based instrument SCIAMACHY on ENVISAT can provide independent information on the variability of CH₄ atmospheric column-averaged mixing ratios particularly over tropical regions (Frankenberg et al., 2008). The SCIAMACHY retrievals from Frankenberg et al. (2008) show a negative anomaly over South America and Africa of about 5–6 ppb in 2006 (as compared to 2003–2009 average), which is qualitatively consistent with the negative emission anomaly inferred by INV1 and INV2 for these tropical regions. We further compared the SCIAMACHY CH₄ column anomaly over South America with the CH₄ column anomaly calculated by LMDZt using optimized fluxes from INV1 and INV2 (Fig. 2) for the period 2006–2008. Both inversions show a minimum in tropical CH₄ column-averaged mixing ratio in mid-2006, which is consistent with the SCIAMACHY data, albeit less pronounced and shifted by 2–3 months. After 2006, the two inversions and SCIAMACHY retrievals agree very well on the relative magnitude of the methane increase in the atmosphere for South America (Fig. 2) and also for other tropical regions (not shown). This shows a consistency between tropical CH₄ flux changes inferred from surface-based inversions and from independent satellite data of column-averaged mixing ratios. However, this agreement also reveals that we cannot really discriminate a large negative anomaly (INV1) from a small negative anomaly (INV2) over South America in 2006 using SCIAMACHY data.

3.3 Mid latitudes

At mid latitudes (30–50° N), the CH₄ anomalies are not statistically different from zero for the different models. This is interesting because the mid-latitudes contain 30% of the global source, but this source seems to vary little from one year to the next. One exception is inversion INV2 in 2008 (+20 Tg). This large anomaly is hard to explain

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because there is less wetland area present at mid latitudes (only 14% of the global wetland source) and anthropogenic emissions are unlikely to vary that much from one year to the next. INV2 may produce more variability at mid latitudes because of the assimilation of daily data from 5 continuous stations at these latitudes. These continuous data increase the relative weight of mid latitudes in the cost function from less than 25% in INV1 to more than 33% in INV2, as compared to other latitudes. When tightening the prior uncertainties on these five sites in INV2 from ± 15 ppb (reference case) down to ± 3 ppb, the variability at mid-latitudes increases even more (not shown). As we assimilate an increasing number of types of observation in atmospheric inversions (flasks, continuous, aircraft, and satellites), the relative weights among these data appears to be an issue that will have to be further investigated as initiated in Bergamaschi et al., (2009).

3.4 High latitudes

At high latitudes ($>50^\circ$ N), a consistent positive wetland emission anomaly of $+5\pm 5$ Tg (INV1) and $+8$ Tg (ORCHIDEE) is found in 2007, explaining the total boreal positive anomaly of INV1 ($+3\pm 4$ Tg) and possibly explaining the one of INV2 ($+5$ Tg). In inversions, the high latitude anomaly represents 15-30% of the global anomaly in 2007. In 2008, no significant anomaly is found at high latitudes (considering estimated uncertainties). In ORCHIDEE, the 2007 positive anomaly is due to higher temperatures impacting both methane flux densities and wetland extent, especially during summer and autumn. In INV1, boreal America and boreal Eurasia are found to have contributed almost equally to the positive flux anomaly. This may reflect a difficulty for the inversion to partition emissions between these two regions with regional error correlations reaching $-0.3/-0.4$ in INV1. In 2007, wetland or total CH_4 flux anomalies are smaller at high latitudes than in the tropics.

For INV1, the high latitudes wetland flux anomaly is about half of the tropical anomaly (37% for ORCHIDEE). Further, in INV1, the total flux anomaly at high latitudes is less than three times the tropical one (half in INV2). These results may appear contradic-

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tory with the larger anomaly in atmospheric surface growth rate found by Dlugokencky et al. (2009) at high latitudes as compared to the tropics. To investigate this issue, we have performed a test with LMDZt model. A pulse of 1 Tg of CH₄ was emitted over one month for each of the two regions: Boreal Eurasia (13 million km²) and tropical Asia (6 million km²). After the month of emission, the flux pulses were transported for 11 months, with no chemistry applied. The resulting mean atmospheric surface CH₄ mixing ratios induced by each pulse are very different (Fig. 3a, b), because atmospheric transport is horizontally efficient at high latitudes but vertically efficient in the tropics to disperse CH₄. (Fig. 3c, black and red lines). Note that, for this analysis, the calculated surface CH₄ mixing ratios were rescaled on a grid with cells of equal surface. As a consequence, the maximum impact of a 1 Tg pulse at neighboring surface stations is found to be 2 to 3 times larger at high latitudes as compared to the tropics (Fig. 3c). Inversions account for these regional differences in vertical mixing, and place accordingly the inferred wetland anomaly more in the tropics than at high latitudes. In fine, the inferred bottom-up and top-down flux anomalies inferred in 2007 are consistent with the larger atmospheric surface growth rate observed at high latitudes. This shows that flux anomalies cannot be just deduced from inspection of surface atmospheric data, but that atmospheric transport must be explicitly and properly modelled.

4 Conclusions

We have analyzed recent changes in the CH₄ budget from two atmospheric inversions and the wetland ecosystems emission model ORCHIDEE over the period 2006–2008:

- a consistent picture between inversions was obtained for 2007 with a positive flux anomaly of 16–21 Tg, characterized by a dominant tropical contribution (60–75%) and a significant contribution from high latitudes (~15–30%).
- natural wetlands are found to explain ~2/3 of the 2007 global anomaly, in good agreement with the ORCHIDEE model that explains the positive anomaly of nat-

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ural wetlands by wetter than normal conditions in the tropics and by unusually warmer temperatures at high latitudes in 2007.

– In 2008, both inversions produce a positive anomaly in total CH₄ emissions, but of different magnitude, giving a large range of 6–21 Tg. On the other hand, ORCHIDEE gives a large positive flux anomaly in 2008 even higher than in 2007, due to increasing tropical precipitation. Additional constraints both on inversions (more atmospheric observations assimilated in 2009) and on ORCHIDEE (to better quantify the sensitivity to precipitation) are required to reduce the uncertainties on the 2008 flux anomaly.

– The atmospheric CH₄ variations calculated with the optimized emissions of the two inversions for 2006–2008 are shown to be consistent with independent CH₄ column-averaged mixing ratio retrieved by SCIAMACHY satellite instrument. The observed drop of atmospheric CH₄ columns in 2006, associated to a tropical flux negative anomaly in inversions, and the increase in 2007–2008 are reproduced consistently by both inversions.

– Finally, OH changes remain small (<1%) in both inversions and only slightly modulate methane fluxes.

Natural wetlands can largely modulate the atmospheric growth rate of methane over a few years, or even a decade, depending on regional temperature and precipitation trends, and on natural climate oscillations. Nevertheless, as long as global methane anthropogenic emissions increase, as estimated by bottom-up inventories such as EDGAR4 (+32 Tg globally from 1990 to 2005), one should not forget that, even without counting possible future releases from marine hydrates or permafrost, methane should continue to increase in the atmosphere on decadal time scales. In 2009, atmospheric methane increased by ~5 ppb, slight lower than the increases of 8.0 ppb in 2007 and 6.3 ppb in 2008 based on updated NOAA global averages (1σ uncertainties are ±0.6 ppb). It is too soon to tell if the 2007 to 2008 period was a temporary positive

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anomaly such as 1997-1998 or 2002-2003, but, at this stage, this recent anomaly is the largest and most persistent since systematic observations began.

Appendix A

5 ORCHIDEE ecosystem model for methane wetland emissions

CH₄ emissions are computed monthly (mass per month) for each model grid cell as the product of an emitting area (surface per month) and of a temperature- and precipitation dependent emission factor (mass per month and per surface) for the period 1990–2008. Wetland area dynamics are computed by the inclusion of TOPMODEL (Beven and Kirkby, 1979) with bias correction of Saulnier and Datin (2004) into ORCHIDEE. For each grid cell, using both topographic heterogeneities and soil moisture computed by ORCHIDEE, a sub-grid saturated fraction (i.e. water table at the soil surface) is computed, as well as fractions with water table at various depths. The simulated space-time distribution of saturated soils is evaluated globally (Ringeval et al., 2010c) against inundated area derived from a suite of satellite observations from multiple sensors of Prigent et al. (2001, 2007). As done in Ringeval et al., (2010b), we use Prigent et al. (2001) satellite data to represent wetland areas and calculate anomalies from the saturated area given by TOPMODEL, relatively to the 1993-2000 climatology of the satellite data. Moreover, in the present work, for boreal ecosystems, resulting wetlands are further filtered using soil organic carbon data as done by Wania et al. (2009) to diagnose the presence of peatlands. In boreal regions, wetlands are assimilated to peatlands with a large soil carbon content. Soil carbon accumulates under anaerobic conditions is necessary as a substrate for methanogenic microbes. Thus not only saturated conditions but also soil carbon is required to have CH₄ emissions at high latitudes. Because ORCHIDEE is not able to produce peat accumulation yet (see Koven et al., 2009), resulting wetlands fractions are further multiplied, for boreal ecosystems, by a map giving fractional peatland cover per grid-cell. This map is obtained using soil

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organic carbon data from IGBP DIS as done by Wania et al. (2009). Hypothesis behind multiplication of the two products is that the inundated fraction is the same for an entire grid-cell as for a sub-grid peatland into this grid-cell.

CH_4 fluxes are computed using an update of the process-based model of Walter et al. (2001) for each sub-grid water-table class given by TOPMODEL. The model simulates CH_4 production, three pathways of transport (diffusion, plant-mediated transport and ebullition) and oxidation. Contrary to initial version of Walter et al. (2001), the substrate for methanogenesis is computed from active soil organic carbon calculated by ORCHIDEE (see (Ringeval et al., 2010a) for more details). Identification of each grid-cell to a wetland type is based on preponderant vegetation type.

After a spin-up using pre-industrial conditions and transient simulations over 1860–1990 (Ringeval et al., 2010b), ORCHIDEE is run over the 1990–2008 period to analyse recent year-to-year variability. Over this last period, two sets of meteorological forcings are used. First, ORCHIDEE is forced by the monthly value of Climatic Research Unit from University of East Anglia before 2003, and NCEP analysis anomalies relative to 2002 CRU data after 2003 (Viovy and Ciais, 2009], together with a weather generator (Krinner et al., 2005). Relative humidity comes from the NCEP analysis (Sheffield et al., 2003). In the second set of forcing conditions, a sensitivity test is performed by replacing the precipitation fields from NCEP by the ones from the ERA40 reanalysis (Uppala et al., 2005).

Global mean wetland CH_4 emissions simulated by ORCHIDEE are $\sim 205 \text{ TgCH}_4 \text{ yr}^{-1}$ over the 1999–2006 period. This is at the upper end of the IPCC range (IPCC, 2007). The distribution over latitude bands is 36, 31, and $138 \text{ TgCH}_4 \text{ yr}^{-1}$ for boreal ($>50^\circ \text{ N}$), temperate ($30^\circ \text{ N}–50^\circ \text{ N}$) and tropical wetlands ($30^\circ \text{ S}–30^\circ \text{ N}$), respectively.

For comparison, the global emissions of ORCHIDEE ($\sim 205 \text{ TgCH}_4 \text{ yr}^{-1}$) are scaled each year according to the global emission from natural wetlands estimated by INV1 for the reference period 1999–2006 ($\sim 165 \text{ TgCH}_4 \text{ yr}^{-1}$). A scaling ratio of 0.8 is therefore applied for the years 2005 to 2008 to ORCHIDEE wetland emissions.

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References

- Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Villani, M. G., Houweling, S., Dentener, F., Dlugokencky, E. J., Miller, J. B., Gatti, L. V., Engel, A., and Levin, I.: Inverse modeling of global and regional CH₄ emissions using SCIAMACHY satellite retrievals, *J. Geophys. Res.-atmos.*, 114, D22301, doi:10.1029/2009JD012287, 2009.
- Bergamaschi, P., Krol, M., Dentener, F., Vermeulen, A., Meinhardt, F., Graul, R., Ramonet, M., Peters, W., and Dlugokencky, E. J.: Inverse modelling of national and European CH₄ emissions using the atmospheric zoom model TM5, *Atmos. Chem. Phys.*, 5, 2431–2460, doi:10.5194/acp-5-2431-2005, 2005.
- Beven, K. and Kirkby, M.: A physically based, variable contributing area model of basin hydrology, *Hydrol. Sci. J.*, 24, 43–69, 1979.

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10, 27603–27630, 2010

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Bousquet, P., Peylin, P., Ciais, P., Le Quere, C., Friedlingstein, P., and Tans, P. P.: Regional changes in carbon dioxide fluxes of land and oceans since 1980, *Science*, 290, 1342–1346, 2000.

Bousquet, P., Hauglustaine, D. A., Peylin, P., Carouge, C., and Ciais, P.: Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform, *Atmos. Chem. Phys.*, 5, 2635–2656, 2005, <http://www.atmos-chem-phys.net/5/2635/2005/>.

Bousquet, P., Ciais, P., Miller, J. B., Dlugokencky, E. J., Hauglustaine, D. A., Prigent, C., Van der Werf, G. R., Peylin, P., Brunke, E. G., Carouge, C., Langenfelds, R. L., Lathiere, J., Papa, F., Ramonet, M., Schmidt, M., Steele, L. P., Tyler, S. C., and White, J.: Contribution of anthropogenic and natural sources to atmospheric methane variability, *Nature*, 443, 439–443, 2006.

Bousquet, P., Yver, C., Pison, I., Li, Y. S., Fortems, A., Hauglustaine, D., Szopa, S., Rayner, P., Peylin, P., Novelli, P. C., Langenfelds, R., Steele, L. P., Ramonet, M., Schmidt, M., Foster, P., Morfopoulos, C., and Ciais, P.: A 3D synthesis inversion of the molecular hydrogen cycle: sources and sinks budget and implications for the soil uptake., accepted, *J. Geophys. Res.*, 2010.

Chen, Y. H. and Prinn, R. G.: Estimation of atmospheric methane emissions between 1996 and 2001 using a three-dimensional global chemical transport model, *J. Geophys. Res.-atmos.*, 111, D10307, doi:10.1029/2005JD006058, 2006.

Chevallier, F., Fisher, M., Peylin, P., Serrar, S., Bousquet, P., Bréon, F.-M., Chédin, A., and Ciais, P.: Inferring CO₂ sources and sinks from satellite observations: Method and application to TOVS data, *J. Geophys. Res.*, 110, D24309, doi:10.21029/22005JD006390, 2005.

Chevallier, F., Bréon, F.-M., and Rayner, P. J.: Contribution of the Orbiting Carbon Observatory to the estimation of CO₂ sources and sinks: Theoretical study in a variational data assimilation framework, *J. Geophys. Res.*, 112, D09307, doi:10.1029/2006jd007375, 2007.

Dentener, F., Peters, W., Krol, M., van Weele, M., Bergamaschi, P., and Lelieveld, J.: Interannual variability and trend of CH₄ lifetime as a measure for OH changes in the 1979–1993 time period, *J. Geophys. Res.-Atmos.*, 108, 4442, doi:10.1029/2002JD002916, 2003.

Dlugokencky, E. J., Bruhwiler, L., White, J. W. C., Emmons, L. K., Novelli, P. C., Montzka, S. A., Masarie, K. A., Lang, P. M., Crotwell, A. M., Miller, J. B., and Gatti, L. V.: Observational constraints on recent increases in the atmospheric CH burden, *J. Geophys. Res. Lett.*, 36, L18803, doi:10.1029/2009GL039780, 2009.

Emission Database for Global Atmospheric Research (EDGAR), release version 4.0. European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), <http://edgar.jrc.ec.europa.eu>, 2009.

5 Frankenberg, C., Bergamaschi, P., Butz, A., Houweling, S., Meirink, J. F., Notholt, J., Petersen, A. K., Schrijver, H., Warneke, T., and Aben, I.: Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT, *Geophys. Res. Lett.*, 35, L15811, doi:10.1029/2008GL034300, 2008.

Gilbert, J.-C., and Lemaréchal, C.: Some numerical experiments with variable-storage quasi-Newton algorithms, *Math. Programm.*, 45, 407–435, 1989.

10 Hein, R., Crutzen, P. J., and Heimann, M.: An inverse modeling approach to investigate the global atmospheric methane cycle, *Global Biogeochem. Cy.*, 11, 43–76, 1997.

Hoerling, M. and Kumar, A.: The perfect ocean for drought, *Science*, 299, 691–694, 2003.

Hourdin, F. and Talagrand, O.: Eulerian backtracking of atmospheric tracers. I: Adjoint derivation and parametrization of subgrid-scale transport, *Q. J. Roy. Meteorol. Soc.*, 132, 567–583, 2006.

15 Hourdin, F. D., Couvreux, F., and Menut, L.: Parameterization of the dry convective boundary layer based on a mass flux representation of thermals, *J. Atmos. Sci.*, 59, 1105–1123, 2002.

Houweling, S., Kaminski, T., Dentener, F., Lelieveld, J., and Heimann, M.: Inverse modeling of methane sources and sinks using the adjoint of a global transport model, *J. Geophys. Res.-Atmos.*, 104, 26137–26160, 1999.

20 IPCC: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment : Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, UK and New York, NY, USA, 996 pp., 2007.

25 Kaminski, T., Rayner, P. J., Heimann, M., and Enting, I. G.: On aggregation errors in atmospheric transport inversions, *J. Geophys. Res.-Atmos.*, 106, 4703–4715, 2001.

Kaplan, J. O.: Wetlands at the Last Glacial Maximum: Distribution and methane emissions, *Geophys. Res. Lett.*, 29, 1079, doi:10.1029/2001gl013366, 2002.

30 Krinner, G., Viovy, N., de Noblet-Ducoudre, N., Ogee, J., Polcher, J., Friedlingstein, P., Ciais, P., Sitch, S., and Prentice, I. C.: A dynamic global vegetation model for studies of the coupled atmosphere-biosphere system, *Global. Biogeochem. Cycles*, 19, GB1015, doi:10.1029/2003GB002199, 2005.

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Krol, M. and Lelieveld, J.: Can the variability in tropospheric OH be deduced from measurements of 1,1,1-trichloroethane (methyl chloroform)?, *J. Geophys. Res.-atmos.*, 108, 4125, doi:4110.1029/2002JD002423, 2003.

Langenfelds, R. L., Francey, R. J., Pak, B. C., Steele, L. P., Lloyd, J., Trudinger, C. M., and Allison, C. E.: Interannual growth rate variations of atmospheric CO₂ and its delta C-13, H-2, CH₄, and CO between 1992 and 1999 linked to biomass burning, *Global Biogeochem. Cy.*, 16, 1048, doi:10.1029/2001GB001466, 2002.

Matthews, E. and Fung, I.: Methane emissions from natural wetlands, global distribution, area and environmental characteristics of sources, *Global Biogeochem. Cy.*, 1, 61–86, 1987.

National Climatic Data Center databasis, <http://www.ncdc.noaa.gov/climate-monitoring/>, 2008.

Olivier, J. G. J. and Berdowski, J. J. M.: Global emissions sources and sinks, in: *The Climate System*, edited by: Berdowski, J., Guichert, R., and Heij, B., 33–37, 2001.

Peylin, P., Baker, D., Sarmiento, J., Ciais, P., and Bousquet, P.: Influence of transport uncertainty on annual mean and seasonal inversions of atmospheric CO₂ data, *J. Geophys. Res.-Atmos.*, 107, 4385, doi:4310.1029/2001JD000857, 2002.

Peylin, P. and Bousquet, P.: Multiple constraints on regional CO₂ flux variations over land and oceans, *Global. Biogeochem. Cycles*, 19, GB1011, doi:10.1029/2003GB002214, 2005.

Pison, I., Bousquet, P., Chevallier, F., Szopa, S., and Hauglustaine, D.: Multi-species inversion of CH₄, CO and H₂ emissions from surface measurements, *Atmos. Chem. Phys.*, 9, 5281–5297, doi:10.5194/acp-9-5281-2009, 2009.

Prigent, C., Matthews, E., Aires, F., and Rossow, W. B.: Remote sensing of global wetland dynamics with multiple satellite data sets, *Geophys. Res. Lett.*, 28, 4631–4634, doi:4610.1029/2001GL013263, 2001.

Prigent, C., Papa, F., Aires, F., Rossow, W. B., and Matthews, E.: Global inundation dynamics inferred from multiple satellite observations, 1993–2000, *J. Geophys. Res.-Atmos.*, 112, D12107, doi:10.1029/2006JD007847, 2007.

Prinn, R. G., Huang, J., Weiss, R. F., Cunnold, D. M., Fraser, P. J., Simmonds, P. G., McCulloch, A., Harth, C., Salameh, P., O'Doherty, S., Wang, R. H. J., Porter, L., and Miller, B. R.: Evidence for substantial variations of atmospheric hydroxyl radicals in the past two decades, *Science*, 293, 1048–1048, 2001.

Prinn, R. G., Huang, J., Weiss, R. F., Cunnold, D. M., Fraser, P. J., Simmonds, P. G., McCulloch, A., Harth, C., Reimann, S., Salameh, P., O'Doherty, S., Wang, R. H. J., Porter, L. W., Miller, B. R., and Krummel, P. B.: Evidence for variability of atmospheric hydroxyl radicals over the

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past quarter century, *Geophys. Res. Lett.*, 32, L07809, doi:07810.01029/02004GL022228, 2005.

Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J., Cunnold, D. M., Steele, L. P., Krummel, P. B., Weiss, R. F., O'Doherty, S., Salameh, P. K., Wang, H. J., Harth, C. M., Muhle, J., and Porter, L. W.: Renewed growth of atmospheric methane, *Geophys. Res. Lett.*, 35, L22805, doi:10.1029/2008gl036037, 2008.

Ringeval, B., de Noblet-Ducoudre, N., Ciais, P., Bousquet, P., Prigent, C., Papa, F., and Rossow, W. B.: An attempt to quantify the impact of changes in wetland extention methane emissions at the seasonal and interannual time scales, *Global Biogeochem. Cy.*, 24, GB2003, doi:10.1029/2008GB003354, 2010a.

Ringeval, B., Friedlingstein, P., Koven, C., Ciais, P., de Noblet, N., Decharme, B., and Cadule, P.: Climate-CH₄ feedback from wetlands and ist interaction with the climate-CO₂ feedback, *Tellus*, in review, 2010b.

Ringeval, B., Decharme, B., Piao, S. L., Ciais, P., Papa, F., De-Noblet Ducoudré, N., Prigent, C., Friedlingstein, P., and Gouttevin, I.: Modelling sub-grid soil moisture saturation in the ORCHIDEE global land surface model: evaluation against river discharges and remotely sensed data, in review, *J. Geophys. Res.-Biogeochem.*, 2010c.

Global precipitation analysis products of the GPCC, Global Precipitation Climatology Centre (GPCC), DWD, Internet Publikation, 1–12, 2008.

Saulnier, G.-M. and Datin, R.: Analytical solution to a bias in the TOPMODEL framework balance *Hydrol. Process.*, 18, 1195-218, 2004.

Schneider, U., Fuchs, T., Meyer-Christoffer, A., and Rudolf, B.: Global precipitation analysis products of the GPCC, Global Precipitation Climatology Centre (GPCC), DWD, Internet Publikation, 1–12, 2008.

Simmonds, P. G., Manning, A. J., Derwent, R. G., Ciais, P., Ramonet, M., Kazan, V., and Ryall, D.: A burning question. Can recent growth rate anomalies in the greenhouse gases be attributed to large-scale biomass burning events?, *Atmos. Environ.*, 39, 2513–2517, 2005.

Uppala, S. M., Koallberg, P. W., Simmons, A. J., U. Andrae, V. da Costa Bechtold, M. Fiorino, J. K. Gibson, J. Haseler, A. Hernandez, G. Kelly, X. Li, K. Onogi, S. Saarinen, N, S., R. P. Allan, E. Andersson, K. Arpe, M. A. Balmaseda, A. C. M. Beljaars, L. van de Berg, J. Bidlot, N. Bormann, S. Caires, F. Chevallier, A, D., M. Dragosavac, M. Fisher, M. Fuentes, S. Hagemann, H, E., 'olm, B. J. Hoskins, L. Isaksen, P. A. E. M. Janssen, R. Jenne, A. P. A. McNally, J.-F. Mahfouf, J.-J. Morcrette, N. A. Rayner, R. W. Saunders, P. Simon, A. Sterl, K.

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Van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano, A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos. Chem. Phys.*, 6, 3423–3441, doi:10.5194/acp-6-3423-2006, 2006.

Van Weele, M., Neef, L., and van Velthoven, P.: Observation-based constraints on the present-day atmospheric methane budget, *Geochimica Et Cosmochimica Acta*, Switzerland, 21 June, 73, A1374-A1374, 2009.

Viovy, N. and Ciais, P.: A combined dataset for ecosystem modelling, <http://dods.extra.cea.fr/data/p529viov/cruncep/readme.htm>, 2009.

Walter, B. P., Heimann, M., and Matthews, E.: Modeling modern methane emissions from natural wetlands 2. Interannual variations 1982–1993, *J. Geophys. Res.-atmos.*, 106, 34207–34219, 2001.

Wania, R., Ross, I., and Prentice, I. C.: Implementation and evaluation of a new methane model within a dynamic global vegetation model: LPJ-WHyMe v1.3.1, *Geosci. Model Dev.*, 3, 565–584, doi:10.5194/gmd-3-565-2010, 2010.

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Table 1. Methane surface fluxes averaged for the 1999–2006 period, in $\text{TgCH}_4\cdot\text{yr}^{-1}$. From left to right, CH_4 fluxes are given: for natural wetlands from ORCHIDEE model, for four categories of emissions inferred by INV1 (wetlands, other natural, biomass burning, total), and for the total emissions inferred by INV2. From top to bottom: global, $<30^\circ\text{N}$, $30\text{--}50^\circ\text{N}$, $>50^\circ\text{N}$. Total fluxes are the sum of surface emissions and soil uptake. The uncertainty assigned to INV1 is the residual uncertainty returned by the inversion procedure (1-sigma). For INV1, regional anomalies are not shown for sources other than wetlands as generally very small. *: ORCHIDEE global emissions for the 1999–2006 period ($205\text{ TgCH}_4\cdot\text{yr}^{-1}$) were scaled to the wetland emissions estimate from scenario S_0 of INV1 for the same period ($165\text{ TgCH}_4\cdot\text{yr}^{-1}$). **: INV2 global total flux for the 1999–2006 period ($529\text{ TgCH}_4\cdot\text{yr}^{-1}$) was scaled to the global total flux from the scenario S_0 of INV1 for the same period ($515\text{ TgCH}_4\cdot\text{yr}^{-1}$).

$\text{TgCH}_4\cdot\text{yr}^{-1}$		1999–2006 period					
REGION	ORCHIDEE	INV1 NATURAL WETLANDS	INV1 OTHER NATURAL	INV1 BIOMASS BURNING	INV1 ANTHROPIC	INV1 TOTAL	INV2 TOTAL
GLOBAL	165*	165±9	17±9	36±5	297±15	515±3	515**
<30° N	110	115±9	–	–	–	304±6	290
30–50° N	25	19±4	–	–	–	136±6	143
>50° N	29	31±5	–	–	–	76±4	86

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Table 2. Methane surface flux anomalies for 2007 **(a)** and for 2008 **(b)**, relative to the 1999–2006 period (see Table 1), in $\text{TgCH}_4 \text{ yr}^{-1}$. From left to right, CH_4 fluxes are given: for natural wetlands from ORCHIDEE model, for four categories of emissions inferred by INV1 (wetlands, other natural, biomass burning, total), and for the total emissions inferred by INV2. From top to bottom: global, $<30^\circ \text{ N}$, $30\text{--}50^\circ \text{ N}$, $>50^\circ \text{ N}$. Total fluxes are the sum of surface emissions and soil uptake. The first uncertainty assigned to INV1 is the residual uncertainty returned by the inversion procedure (1-sigma). The second uncertainty is the spread of the 11 inversions of INV1 (1-sigma). For INV1, regional anomalies are not shown for sources other than wetlands as generally very small.

$\text{TgCH}_4 \cdot \text{yr}^{-1}$		a: 2007 anomaly					
REGION	ORCHIDEE	INV1 NATURAL WETLANDS	INV1 OTHER NATURAL	INV1 BIOMASS BURNING	INV1 ANTHROPIC	INV1 TOTAL	INV2 TOTAL
GLOBAL	26	$12 \pm 9 \pm 3$	$0 \pm 1 \pm 1$	$2 \pm 5 \pm 1$	$2 \pm 6 \pm 2$	$16 \pm 4 \pm 4$	21
$<30^\circ \text{ N}$	22	$8 \pm 9 \pm 3$	–	–	–	$12 \pm 6 \pm 4$	12
$30\text{--}50^\circ \text{ N}$	–4	$-1 \pm 4 \pm 1$	–	–	–	$1 \pm 6 \pm 2$	3
$>50^\circ \text{ N}$	8	$5 \pm 5 \pm 1$	–	–	–	$3 \pm 4 \pm 4$	5
$\text{TgCH}_4 \cdot \text{yr}^{-1}$		b: 2008 anomaly					
REGION	ORCHIDEE	INV1 NATURAL WETLANDS	INV1 OTHER NATURAL	INV1 BIOMASS BURNING	INV1 ANTHROPIC	INV1 TOTAL	INV2 TOTAL
GLOBAL	38	$-3 \pm 6 \pm 5$	$0 \pm 1 \pm 1$	$1 \pm 5 \pm 2$	$8 \pm 6 \pm 4$	$6 \pm 4 \pm 4$	20
$<30^\circ \text{ N}$	30	$0 \pm 9 \pm 4$	–	–	–	$5 \pm 7 \pm 4$	3
$30\text{--}50^\circ \text{ N}$	3	$-2 \pm 3 \pm 1$	–	–	–	$1 \pm 7 \pm 2$	20
$>50^\circ \text{ N}$	5	$0 \pm 5 \pm 1$	–	–	–	$-1 \pm 5 \pm 1$	–3

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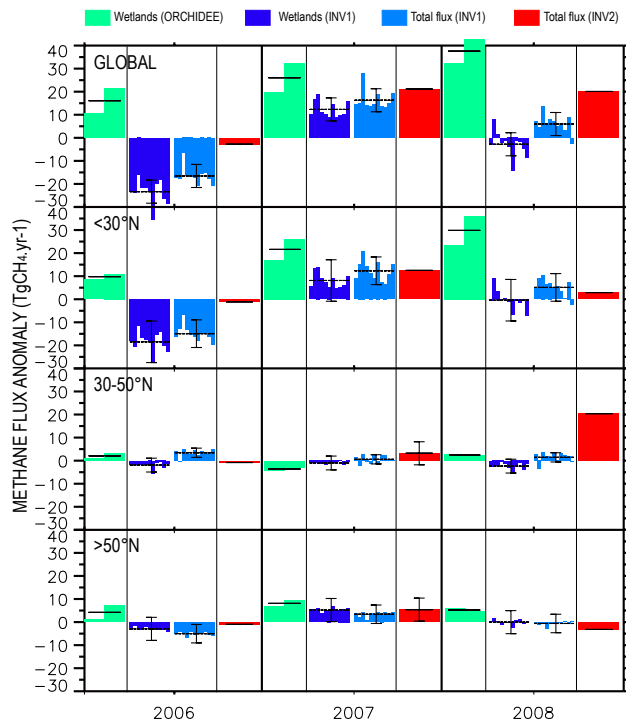


Fig. 1. Anomalies of annual methane emissions in $\text{TgCH}_4 \text{ yr}^{-1}$ for 2006 to 2008. The reference period for the anomaly calculation is 1999–2006. From left to right, each year: wetland anomalies from ORCHIDEE model (green), wetland anomalies from INV1 (11 inversions, dark blue), total anomalies from INV2 (11 inversions, light blue), and total anomalies from INV2 (red). ORCHIDEE is plotted in green with two simulations: CRU/NCEP+ECMWF precipitations (left green bar), and CRU+NCEP (right green bar). The dark line for each model represents the mean of the performed simulations. From top to bottom: global, $<30^\circ \text{N}$, $30\text{--}50^\circ \text{N}$, $>50^\circ \text{N}$. Error bars represent the residual uncertainty returned by the inversion (1-sigma).

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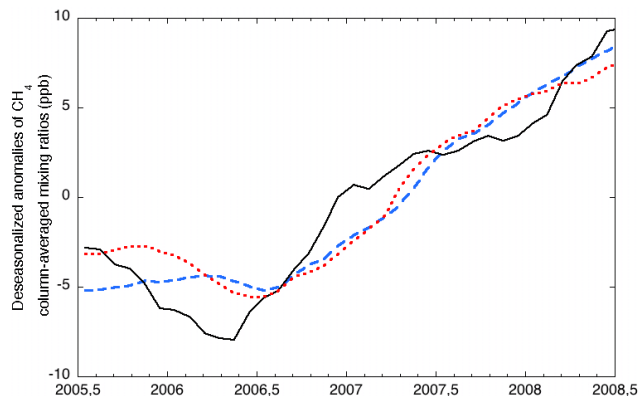


Fig. 2. Mean CH_4 column-averaged mixing ratio anomalies for South America for the 2005–2008 period (in ppb), as retrieved by SCIAMACHY on ENVISAT (solid black), by INV1 (dashed blue) and by INV2 (dotted red). A 12-month running mean was applied to the monthly column-averaged mixing ratios, and their mean over 2006–2008 was subtracted to calculate the anomaly.

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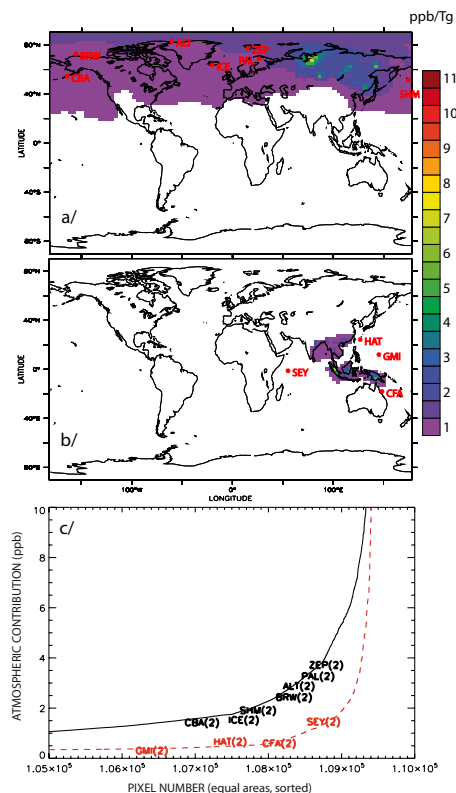


Fig. 3. Impact at the surface of a 1TgCH₄ pulse emitted from two regions (see text): boreal Asia and tropical Asia. **(a)** Mean cumulative plume after one year (1 month of emissions + 11 months of transport in the atmosphere) in ppb/TgCH₄ for boreal Asia. **(b)** Same as (a) for tropical Asia. **(c)** Maximum increase in mixing ratio detected for all transport model pixels, at the surface, for emissions in boreal Asia (black line) or in tropical Asia (red line). Model pixels have the same surface and are sorted by increasing values for each emitting region. Only pixels with the largest mixing ratios are kept for the plot. Maximum values at model pixels containing atmospheric measurement stations used in the inversion are plotted above the lines, as the name of the stations. The month when the maximum is reached is plotted as a number in parentheses. Locations of the stations are shown in (a) and (b).