Impact of stratospheric atomic chlorine on the modelling of total methane and its isotopic ratios at global scale - Supplementary Information

Joël Thanwerdas[1,*], Marielle Saunois[1], Antoine Berchet[1], Isabelle Pison[1], Didier Hauglustaine[1], Michel Ramonet[1], Cyril Crevoisier[2], Bianca Baier[3, 4], Colm Sweeney[4], and Philippe Bousquet[1]

[1]Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, IPSL, Gif-sur-Yvette, France.
[2]Laboratoire de Météorologie Dynamique, École Polytechnique, IPSL, Palaiseau, France.
[3]Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado-Boulder, Boulder, CO 80305
[4]NOAA Earth System Research Laboratory Global Monitoring Division, Boulder, CO, USA 80305

Correspondence: J. Thanwerdas (joel.thanwerdas@lsce.ipsl.fr)

The supplementary information contains details on the litterature used to assess the source isotopic signatures associated to the different sectors, on the sector source intensities used in the model, on the AirCore profiles and regions used to infer regional δ^{13}C isotopic signature values for wetlands, gas and biomass burning. The methane global emissions time-series for the 2000-2018 period are plotted in Figure S2. The individual AirCore vertical profiles plots with the associated simulated profiles are shown in Figure S3. Regional isotopic signature values for three sectors are given in Table S5. Regions are plotted in Figure S4.

1 Text S1

Data from litterature have been compiled to assess a global isotopic signature for the multiple sectors (regional for three of them) used in the paper. These references are given here.

1.1 Wetlands

After reviewing literature on isotopic methane signatures from sources in North America (Alstad and Whiticar, 2011; Burke et al., 1988; Chasar et al., 2000; Happell et al., 1995, 1994; Hornibrook et al., 1997, 2000; Martens et al., 1992; Popp et al., 1999; Santoni et al., 2012), South America (Chanton et al., 1989; Devol et al., 1996; Holmes et al., 2015; Smith et al., 2000; Tyler et al., 1987), Africa (Tyler et al., 1988), Europe (Bowes and Hornibrook, 2006; Galand et al., 2010; Sriskantharajah et al., 2012; Woltemate et al., 1984) and Asia (Itoh et al., 2008; Nakagawa et al., 2002; Sugimoto and Fujita, 2006), the following regional values were decided: a global base δ^{13}C value of -60 ‰ , -65 ‰ in boreal regions, -60 ‰ in temperate regions, and -55 ‰ in tropical regions. In Africa, -50 ‰ was used, following (Tyler et al., 1988).

1.2 Termites

A wide range of δ^{13}C signatures have been reported in the literature, from -93.8 ‰ to -54 ‰ (Bréas et al., 2001). For these simulations, δ^{13}C was taken to be -63 ‰

1.3 Oceanic fluxes

Surface δ^{13}C values from various oceans in the world were found in a small range: roughly -44 ‰ to -40 ‰ (Brunskill et al., 2011; Holmes et al., 2000; Sansone et al., 2001). A value of -42 ‰ was used.

1.4 Geological fluxes

No litterature have been found. A value of -50 ‰ was used (personal communication with Philippe Bousquet). Geological sources contribute only a small fraction of the total methane sources so uncertainties on this value do not largely affect final results.

1.5 Livestock

Livestock is a large sector and includes two different source categories : enteric fermentation ($\sim$ 90 % of the livestock sector total methane sources) and manure management ($\sim$ 10 %). Values of -62 ‰ for entering fermentation and -52 ‰ for manure management were taken (Levin et al., 1993; Klevenhusen et al., 2010; Bilek et al., 2001), resulting in a flux- and area-weighted value of -61 ‰

1.6 Rice cultivation

Bréas et al. (2001) cited literature δ^{13}C values from rice paddies, ranging from -68 ‰ to -48 ‰ . For this work, -63 ‰ was used (Bousquet et al., 2006).

1.7 Oil, Gas, Industry (OGI)

OGI encompasses five very different source categories : gas ($\sim$ 68 %), oil ($\sim$ 28 %), industrial processes ($\sim$ 1 %), road transportation ($\sim$ 2 %) and non-road transportation ($\sim$ 1 %). Regional values for gas from -50 ‰ to -36.8 ‰ (see Table S5) (Levin et al., 1999; Lowry et al., 2001; Zazzeri et al., 2015; Phillips et al., 2013; Aali et al., 2006), -40 ‰ for oil (Monteil et al., 2011), -52.6 ‰ for industrial processes (overall signature), -20 ‰ for road transportation and also for non-road transportation (Levin et al., 1999; Nakagawa et al., 2005; Chanton et al., 2000) were taken, resulting in a flux- and area-weighted value of -39.7 ‰

1.8 Biofuels and Biomass

Biofuels and Biomass sector encompasses four very different source categories : biomass burning ($\sim$ 35 %), biofuel burning ($\sim$ 40 %), energy use ($\sim$ 20 %) and fossil fuel fires ($\sim$ 5 %). Regional values for biomass burning from -26.5 ‰ to -15.46 ‰ (see Table S5), -20 ‰ for biofuel burning (Chanton et al., 2000), -30 ‰ for energy use and -30 ‰ for fossil fuel fires were taken, resulting in a flux- and area-weighted value of -25.8 ‰

1.9 Waste

Waste sector encompasses three different source categories : waste water ($\sim$ 49 %), solid waste ($\sim$ 49 %), agriculture waste burning and ($\sim$ 2 %). Values of -48 ‰ for waste water, -52 ‰ for solid waste and -23.6 ‰ for agriculture waste burning (Levin et al., 1993; Yamada et al., 2006; Townsend-Small et al., 2012; Bergamaschi et al., 1998; Chanton et al., 1999) were taken, resulting in a flux- and area-weighted value of -49.7 ‰

1.10 Coal

For coal emissions in mines, a δ^{13}C value of -35 ‰ was used, as in Monteil et al. (2011).

2 Figure S2

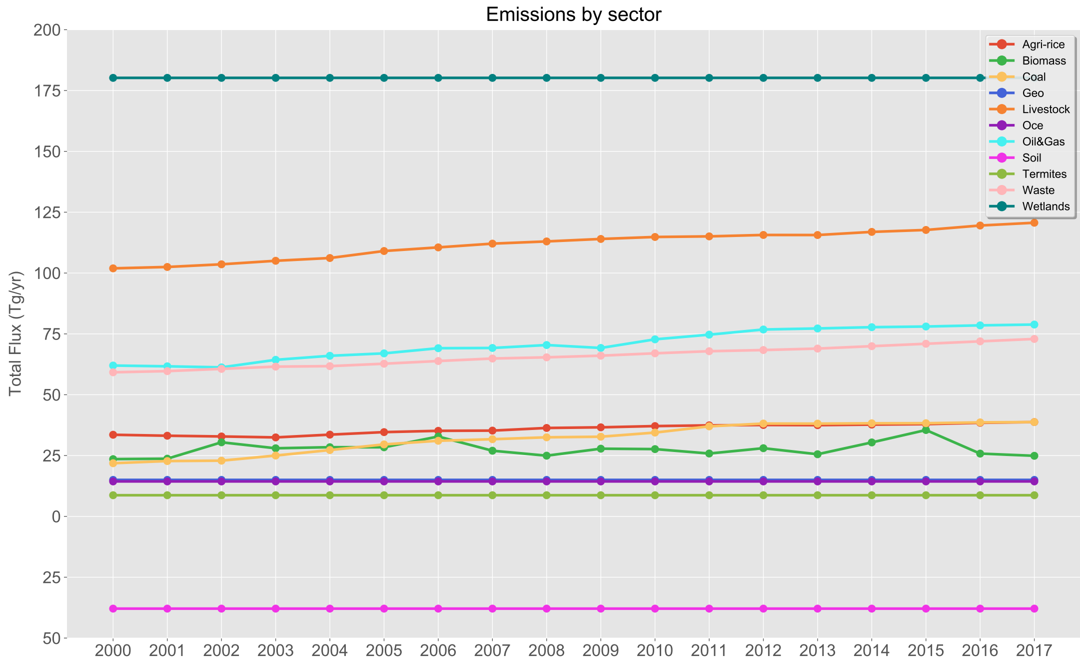

Figure S2. Sector emissions for the 2000-2017 period in Tg/yr. The 2018 emissions are set equal to 2017.

3 Figure S3

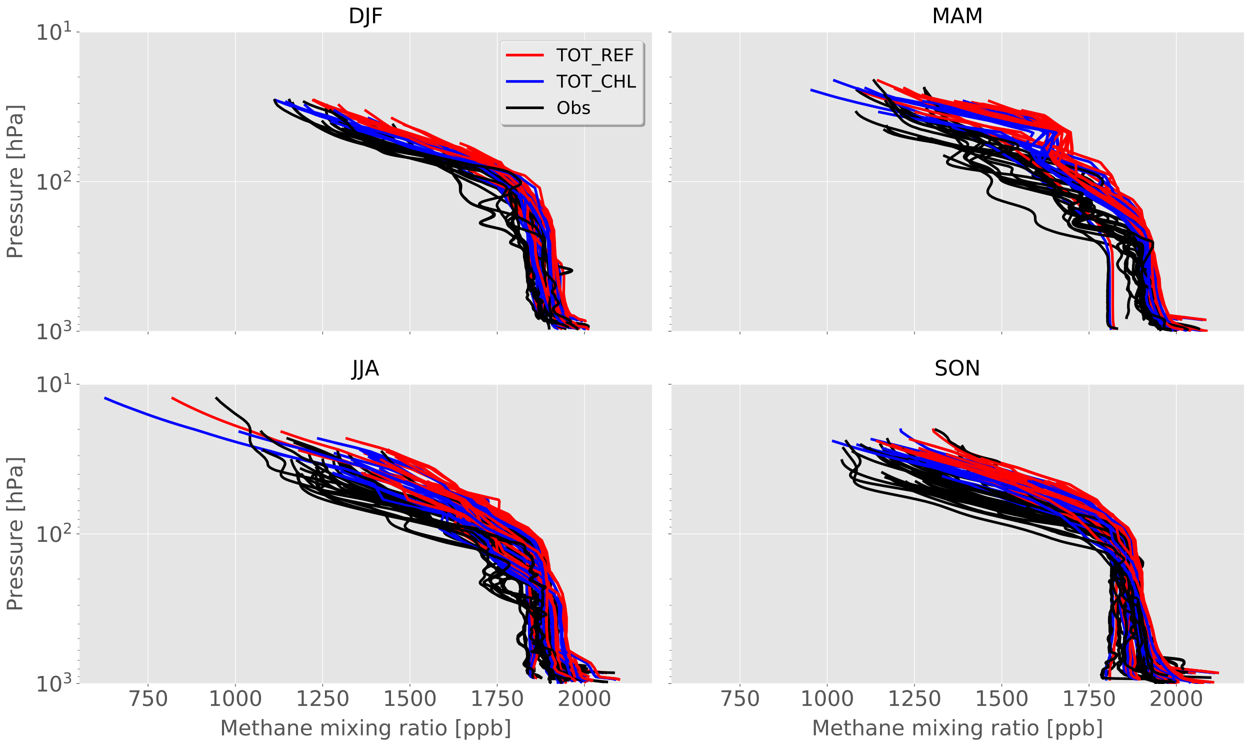

Figure S3. Methane vertical profiles for each season. Simulated profiles from TOT_REF and TOT_CHL are indicated by red and blue lines, respectively. The dark lines are the raw data retrieved using AirCore technique.

4 Figure S4

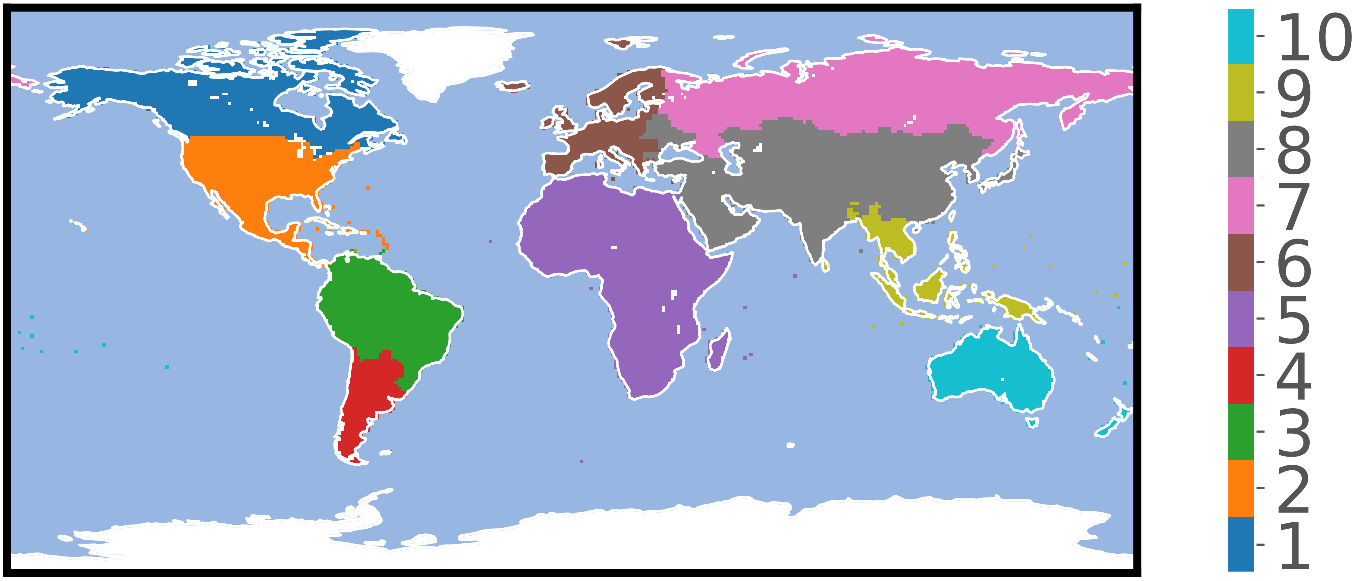

Figure S4. Regions used to infer regional δ^{13}C isotopic signature values for wetlands, gas and biomass burning.

5 Table S5

Region	Wetlands	Gas	Biomass burning
Global default	-60.0 ‰	-40.0 ‰	-26.5 ‰
Boreal North America	-65.0 ‰	-36.8 ‰	-26.5 ‰
Temperate North America	-60.0 ‰	-36.8 ‰	-26.5 ‰
Tropical South America	-55.0 ‰	-40.0 ‰	-18.15 ‰
Temperate South America	-60.0 ‰	-40.0 ‰	-19.53 ‰
Africa	-50.0 ‰	-40.0 ‰	-15.46 ‰
Europe	-60.0 ‰	-44.0 ‰	-26.5 ‰
Boreal Asia	-65.0 ‰	-50.0 ‰	-26.82 ‰
Temperate Asia	-60.0 ‰	-40.0 ‰	-19.16 ‰
Southeast Asia	-55.0 ‰	-40.0 ‰	-20.65 ‰
Australia	-60.0 ‰	-40.0 ‰	-26.5 ‰

Table S5. Regional δ^{13}C isotopic signature values for wetlands, gas and biomass burning.

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