Review on Thanwerdas et al., acp-2019-925

General comments

The paper examines effect of chemical reactions with Cl on modelling atmospheric methane and is ^{13}C:^{12}C isotopic ratios. The global chemistry transport model LMDz is used to model the atmospheric concentrations, and observations from AirCore, balloon and surface network is used as evaluation. They examined the effects on different layers of atmosphere (troposphere and stratosphere), and on long-term trends and seasonal differences.

Such study is highly valuable for modelling community, who are trying to understand the cause of atmospheric methane increase. Accounting for correct atmospheric chemistry is extremely important in e.g. atmospheric inversion systems, where surface fluxes are constrained based on modelled concentrations and observations. If the chemistry is not accounted properly, then it influences the flux estimates directly.

Their finding is only from one model and a single Cl reaction fields, but still this study gives ideas on the importance of including Cl reactions in modelling atmospheric methane and its ^{13}C:^{12}C isotopic ratio, which can be a good guidance to other modellers.

Specific comments

Section 2.3: First two paragraphs can be in Introduction, as this does not describe “method”.

Equation 5 and L15-17: Could you explain this equation further? Simply substituting numbers does not seem to provide $\delta^{13}C_{eff}$ to be – 65.9 ‰. (1 – 47.2)/1.02 – 1= – 46.3, and following equations presented in Snover and Quay (2000), $\delta^{13}C_{soil} = \delta^{13}C_{atm} + (1/KIE_{soil} -1)*1000$, it will lead to $\delta^{13}C_{soil}$= – 66.8 ‰ (assuming $\delta^{13}C_{atm}$= – 47.2 ‰ and KIE_{soil}=1.02).

Section 3.1, 3.2: Did you find any effect on seasonal cycle by including Cl? The reaction with Cl possibly induces stronger seasonal cycle. It would be valuable to give discussion on this.

P15 L16-20: The sentences are not clear. You explain reduction ratio in range (%) and in absolute value (ppb), and different seasons for different layers of atmosphere. This might be more readable if consistent metric is used.

P15 L27: What do you mean by “small variations”? Variations as std, or differences from the observations?

P15 L28-29: “Those may be induced by small scale filaments of methane coming from polar regions, that cannot be adequately reproduced by our low-resolution model and seasonal averaging.” This can be checked with meteorological data.

P19 L15: “anthropogenic sources” may be not appropriate here, as agriculture, waste-related and biofuels sources are biogenic. It would be better to explain as e.g. simply “fossil fuel”.

P20 L15-16: The impact is actually on the forward run, which then influences the inversion results on flux estimates. In addition, the value “0.27 ‰” is dependent on model and its setups, but the sentence is phrased as if it it a general statement. Please consider rephrasing.

Figure 8: The observed seasonal cycle amplitude presented in the figure is much smaller than those modelled, but “std” of the observations show clear seasonal cycle. Did you use different averaging for model and observed values? In addition, it might be better to present figure with two panels, one on deseasonalised trends and another for monthly values.

Technical comments

Use same notations for “minus signs” (indicating negative values) and decimal points (there are ‘.’ and ‘,’) following publication guidelines.

Table 2: Could you add min. max. of the isotopic signatures for sources that varies between regions?

Figure 4 caption:
– What is (a) in “Dry-air column average mole fraction of methane **(a)** and differences”?
– I assume a) and b) are for the total column. Please specify.
– Add “from” in a) before the name of the simulation, i.e. “average mole fraction of methane **from** TOT_CHL”

P13 L16 “over those high regions” → “over those high latitudes”

P13 L29: The value r can be negative. So it would be better to phrase “The smaller the value of r,” → “The closer the value of r is to zero”, or change the Equation (6), such that it assesses the absolute differences.

P15 L15-16: Please remove the sentence “The displayed value in each frame is the mean value of these differences over the considered region, namely troposphere or stratosphere.”, or change to definition of how you defined troposphere and stratosphere for the analysis.

P15 L20: “each region of the atmosphere” → “each layer of atmosphere”

Figure 5: “line” → “row”

P16 L3: (Locatelli et al., 2015) → Locatelli et al. (2015)

P18 L10 – P19 L2: This explanation is better fit to the method section.

P20 L8: 5%/decade → 5% per decade

P20 L11: d13_CHL and the scenario S10 → d13_CHL, and the scenario S10 (i.e. add “,”)

P20 L15: “if $\delta^{13}C\text{-}CH_4$ constraint are to be used for sources characterization in long-period inversion runs” → “if $\delta^{13}C\text{-}CH_4$ observations are used as constraint for sources characterization in long-period inversion runs”

P21 L5: “10 forward simulations (including 6 sensitivity test runs) have been assessing the impact of”
→ “10 forward simulations (including 6 sensitivity tests) have been carried out to assess the impact of”

P21 L17-L28: Please move this to earlier section, and only include some summary on this in the Conclusion.