

We would like to thank the referees for reviewing our manuscript and for their helpful comments and suggestions. Our point-by-point responses are given below. Page and line numbers given refer to the reviewed version of the manuscript. The original reviewer comments are typeset in **gray**, the replies in **black**, while the proposed changes in the text are in **blue**.

Anonymous Referee #2

1) Bag samples from the Royal Holloway University of London laboratory are presented in Figure 2, but not referenced anywhere that I can find in the main text. I think these measurements should either be discussed in the body of the manuscript, or removed from Figure 2.

The IRMS data from RHUL were removed from Figure 2 in order to not further increase the complexity and length of the manuscript.

2) I don't know if this is a convention in the literature, but my understanding of the nomenclature used in this paper is that "isotopic signature" is used to describe the isotope ratio of the CH₄ emitted from a particular source, whereas "isotope source signature" or just "source signature" is used to describe the combined isotopic signature of the sources that might have contributed to a measured air mass. I found this a very confusing (e.g. see L378 - 380), and perhaps not the most descriptive set of terms. Perhaps I am confused, but I wonder whether the authors could come up with a naming system that describes these terms more clearly. Mainly, when I read the term "source signature" I think that it must refer to the delta value that is the property of a particular source. This does not seem to be its use in this paper.

The terms "isotopic signature" and "isotopic source signature" were used to describe both the isotopic composition of a "single process" CH₄ source and a "combined or mixed source", which may indeed lead to confusion. To resolve this, signatures of combined or mixed sources are now referred to as "mean isotopic signature" or "isotopic composition". To clarify the wording the following section on page 13 Line 383 – 391 was rephrased:

The isotopic composition of CH₄ emissions were estimated using the Keeling plot technique (Keeling, 1961; Pataki et al., 2003). This method allows determining the isotopic signature of a single source process or the mean isotopic signature of combined source processes that mix into a background reservoir from the observed ambient isotopic composition and mole fraction. An implicit assumption of the Keeling plot approach is that the isotopic composition and mole fraction of the background reservoir and the isotopic composition of the source or the combined source stay constant over the time range of the analysis. This may not always apply as the relative contribution of individual CH₄ sources or their isotopic signature may change over time.

In addition, the terms "isotopic source signature" and "isotopic signature" were replaced on

- page 13 Line 379: "isotopic source signatures" replaced by "mean $\delta^{13}\text{C}$ and δD isotopic signatures"
- page 18 Line 526: "source signatures" replaced by "mean δD signature"
- page 18 Line 549: "isotopic signature" replaced by "mean isotopic signatures"
- page 18 Line 552: "isotopic signatures" replaced by "mean isotopic signatures"
- page 18 Line 554: "isotopic signature" replaced by "mean isotopic signature"
- page 19 Line 556: "cumulative source signature" replaced by "mean isotopic signature"
- page 19 Line 560: "source signature" replaced by "mean isotopic signature"
- page 19 Line 571: " $\delta^{13}\text{C}$ source signatures" replaced by "mean $\delta^{13}\text{C}$ signatures"
- page 19 Line 572: " δD source signatures" replaced by "mean $\delta^{13}\text{D}$ signatures"
- page 19 Line 575: " $\delta^{13}\text{C}$ and δD source signatures" replaced by "mean $\delta^{13}\text{C}$ and δD signatures"
- page 19 Line 577: "source signatures" replaced by "mean isotopic signature"
- page 19 Line 582: "isotopic source signatures" replaced by "mean isotopic signature"
- page 20 Line 589: "isotope source signature" replaced by "mean isotopic signature"
- page 20 Line 595: "source signature" replaced by "signature"
- page 20 Line 595-596: "source signature" replaced by "signature"

page 20 Line 601: "source signatures" replaced by "mean isotopic signatures"
page 21 Line 621: "isotopic source signatures" replaced by "isotopic composition"
page 21 Line 624: "isotopic source signatures" replaced by "mean isotopic signatures"
page 21 Line 629: "isotopic source signature time series" replaced by "time series for the mean isotopic signatures"
page 21 Line 631: "source signatures" replaced by "mean isotopic signatures"
page 21 Line 633: "isotope source signatures" replaced by "mean isotopic signatures"
page 21 Line 635: "source signatures" replaced by "mean isotopic signatures"
page 21 Line 635-636: "source signatures" replaced by "signatures"
page 21 Line 637: "source signatures" replaced by "mean isotopic signatures"
page 21 Line 639: "CH₄ isotope source signature distributions" replaced by "mean CH₄ isotopic signatures"
page 21 Line 646: "source signatures" replaced by "mean isotopic signature"
page 22 Line 651: "isotope source signatures" replaced by "mean isotopic signatures"
page 22 Line 654: "source signatures" replaced by "mean isotopic signatures"
page 22 Line 665: "isotopic source signatures" replaced by "mean isotopic signatures"
page 22 Line 670: "source signatures" replaced by "mean isotopic signatures"
page 22 Line 677: "source signatures" replaced by "mean isotopic signatures"
page 22 Line 678: "source signatures" replaced by "mean isotopic signatures"
page 35 Line 798: "isotopic source signatures" replaced by "mean isotopic signatures"
Page 36 Line 809: "source signatures" replaced by "mean isotopic signatures"

Furthermore the title of section 4.2 was changed to:

[Isotope identification of the mean CH₄ source](#)

Specific comments:

3) L51: "representative of"

The wording "...at multiple sites that are representative for the entire European domain." seems to be appropriate.

4) L94: Shouldn't this be "Pee Dee Belemnite"?

Done

5) L159: What does "limited air conditioning mean"?

Wording replaced by:

[Since this room is not commonly used as laboratory, it has an air-conditioning with limited cooling capacity and ...](#)

6) L162: Define "o.d" in this section.

Done

7) L271: "a lower offset in d13C of 1.58permil". I think this statement would benefit from spelling out a little more clearly. What was the offset before, and how has it changed?

The reasoning for the lower offset was already given in the text a few lines before. To clarify the issue and to add details on the offset during previous measurements the text on page 9 Line 268 ff was changed too:

Due to the lower heating power, the duration of the desorption step had to be extended, which led to an improved separation from residual bulk gases (e.g. N₂ and O₂). Lowering the O₂ enhancement in the gas matrix is also the main reason for a lower offset in $\delta^{13}\text{C}$ of 1.58 ‰, with respect to the MPI-scale, as compared to 2.3 ‰ in previously published results (Eyer et al., 2015).

8) L286 - 288: What adjustments were made? Can you provide general details?

The following information has been added on page 10 Line 286:

Additional adjustments in the preconcentration procedure and in the analytical routine for isotope analysis improved the repeatability to 0.18 ‰ and 0.85 ‰ for $\delta^{13}\text{C}$ and δD in the last month of the campaign. One example is the improved temperature control of the trap during adsorption, which in turn stabilized the O₂ content in the measuring gas and thereby reduced variations in $\delta^{13}\text{C}\text{-CH}_4$.

9) L318: ERA interim

Done

10) L334: I presume this is following some spin up period? Are the delta-values at ~ steady state before the model run?

We did a long (20 years) spinup run for the year 2005. The atmospheric CH₄ growth rate had been stable for a few years then and thus it can be assumed that the atmosphere was relatively close to equilibrium with the emissions. We used this spinup as a starting point for our simulations, which cover the period 2005-2015. This way the imbalance of the atmospheric CH₄ and $\delta^{13}\text{C}\text{-CH}_4$ in 2014 should be reasonably accounted for. The paragraph was modified accordingly as follows.

... source signatures (see Table 1). The emission inventory was built according to a double constraint: 1st, each source signature must be chosen within its own uncertainty interval, and 2nd, the resulting global average source signature must be compatible with the global source signature that is inferred from the observations (and that is known with a much better precision than the individual source signatures). In a second set of simulations, anthropogenic emissions in a regional domain centered on Cabauw were replaced by emissions from the European TNO-MACC_2 inventory, which was used as the standard inventory in the FLEXPART-COSMO simulations (see below). Outside the regional domain covered by TNO-MACC_2, the EDGAR emissions were used.

Atmospheric removal of CH₄ was modeled as described in Monteil et al. (2013), using kinetic fractionation factors $\alpha = k(^{12}\text{C})/k(^{13}\text{C})$ of $\alpha_{\text{OH}} = 1.0055$, $\alpha_{\text{Cl}} = 1.0066$ and $\alpha_{\text{O}(^1\text{D})} = 1.013$ for the reactions between CH₄ and OH (Sander et al., 2006), Cl (Saueressig et al., 1995) and O(¹D) (Saueressig et al., 2000), respectively. The simulations were initialized at steady state (obtained via a spin-up run) in 2005, and simulations of the period 2005-2015 were used to calculate a realistic state of the atmosphere at the start of the measurement campaigns, including the imbalance between emissions and atmospheric CH₄ mixing ratio/isotopic composition in 2014. Time series were extracted from model-simulated mole fraction fields after interpolation to the horizontal coordinate and height of the Cabauw tower air inlet.

11) L369: This sentence is a little confusing and doesn't quite follow from the previous one. What was "done separately" for the SNAP categories?

To clarify this, the sentence on page 13 Line 369 was replaced by:

Methane mole fractions were computed separately for a number of SNAP (Standardized Nomenclature for Air Pollutants) source categories with specific isotopic signatures as summarized in Table 2.

12) L385: "This method allows ... to be determined"

Done

13) L429: I don't think "Inter-calibration" is the correct term here. I think you mean "Inter-comparison"? This section doesn't describe a calibration exercise.

Done

14) L466 and Figure 3: It looks to me like there is some non-linearity in the comparison between the two instruments for delta-D? The values at the lower end of the scale appear to be significantly lower for the TREX-QCLAS. Is this true? E.g. if a 2nd order polynomial were fit, it looks to me like it would come out with some curvature. I think this deserves a comment.

The response to this comment has been combined with the response to comment #19 of reviewer #1.

15) L498 - 499: TM5 does indeed seem to perform very well. However, I think this line is somewhat subjective (e.g. use of the term "remarkable") and should be removed.

Done

16) L508: I'm a little unclear what this line means. Do you mean simply that this offset is seen globally?

Yes, at least it is present in the whole Northern Hemisphere.

... offset by about 1 ‰, but this offset is also present at clean background sites in the Northern hemisphere.

17) L520: "showed, that" ... I don't think the comma is needed.

Done

18) L569 and Figure 7: I'm a little concerned about the points in Figure 7. My understanding of the MKP is that it is an hourly "running" 12-hour Keeling plot. Each point in Figure 7 is then the 6-hourly average of the "12-hour running intercepts". Therefore, it seems to me that what we're seeing at each point is a doubly smoothed estimate of the source signature during some ~ 18-hour period. Wouldn't this analysis be more transparent if a "normal" Keeling plot were calculated during a block 6 or 12-hour period? Otherwise it becomes more difficult to understand what these points really show and what time period they correspond to.

The reason for the additional 6-hour averaging is that in some cases the time periods of the MKP for $\delta^{13}\text{C}$ and δD are not identical, e.g. because of instrument failure or because of the data filtering procedures. To increase the number of pairs of MKP intercepts the additional 6-hour averaging was added.

19) L644 - 645: delete "even"

Done

20) L654: This is a good example of where the term "source signatures" is confusing. I'm not sure whether you're referring to the particular isotopic ratio of the methane emitted from a source, or of something about the bulk source mix that you've inferred from the air mass.

This has been updated accordingly, see response to comment #2.

21) Figure 7: At the point where this Figure appears and is referenced, it's not clear what the "a" and "b" refer to in the event labels. I think the Figure caption should be expanded to explain when these events were.

The color coding, numbering and characters (a, b) of event labels of Figure 7 refer to Figure 9 as mentioned in the legend. In Figure 9 more details on the events are given. Nonetheless the additional information has also been added to Figure 7:

... Large colored symbols indicate data from the three events (event 1: 10th – 12th March, event 2: 16th – 18th March, event 3: 22nd to 24th March) that are highlighted in detail in Fig. 9. The labels a and b refer to day 1 and day 2 of the two-day events, respectively.

22) Figure 7: I think these points should have error bars.

Error bars have been added.

23) Figure 7: The reference to Rigby et al. (2012). The original “mean” values are from Snover et al (2000), which I think should be cited. The ranges are those assumed (somewhat arbitrarily) for the inversion in Rigby et al (2012).

The reference of Snover et al. (2000) was added to the legend of Figure 7:

... from recent literature (Snover et al., 2000; Rigby et al., 2012)