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 #1

1) My only concern with the manuscript, at present, is the detrimental effect of the current length and breadth of the present manuscript on the overall readability and, possibly, even limiting further discussion of both (1) the data collection and (2) the data comparison with global and regional models. The manuscript would appear to be well-suited for division into two companion manuscripts, with one containing the description and comparison of the instrumentation and observational data, and a second manuscript containing the measurement-model discussion.

We agree that in principle the scope and findings of the manuscript could be separated into two companion publications, and we discussed this when starting on the manuscript. However, we concluded that the combination of the new high resolution $CH_4$ isotope data with the data interpretation using models in one manuscript is in fact one of the strengths of our work, and we therefore decided to keep these two aspects together in order to increase the impact of the work.

2) It appears as though the TREX-QCLAS collected only half as many measurements as the IRMS system (Fig. 2). Is there cause for concern regarding reliability, despite a moderate increase in throughput? I commend the authors for their discussion of the inter-calibration and comparison of the two measurement techniques, both with regards to isotopic and mole fraction measurements.

Both analytical systems were newly developed and for the first time applied in an extended field campaign, and there are periods without data, indeed more so for the QCLAS system than for the IRMS system, which is partly due to the fact that high quality measurements with the QCLAS system started later. The authors are convinced that there is no fundamental cause for concern regarding the TREX-QCLAS system as demonstrated by a significantly improved performance by the end of the campaign as mentioned in the method section on page 10 Line 283 – 289. Similar instrumentation developed by the authors has been applied for extended field campaigns (e.g. Wolf et al. Biogeosciences, 2015). The fact that the TREX-QCLAS system started later and that various kind of instrument malfunctions led to a number of interruptions for both analytical systems is mentioned in the result section of the manuscript on page 14 Line 417 – 422.

Specific comments:

3) Line 89: This is a poor topic sentence for this paragraph. This paragraph should be re-structured.

The respective sentence was moved to page 4 Line 97, after the wording: … and given in per mill (‰). $CH_4$ mole fractions …

4) Line 163 and elsewhere: Manufacturers need additional location information (e.g. line 163, Varian, Inc.) and sufficient information is missing for some manufacturers (e.g. VICI, manufacturer of Valco valves).

The following information was added:

Line 163: … by a Varian scroll pump (Agilent Technologies Inc., USA)

Line 178: … Thermo Delta Plus XP for carbon isotopes, both Thermo Fisher Scientific Inc., Germany)

Line 187: … using removable frits (CEF1F, Valco Instruments Company Inc., USA)

Line 189: … with Valco fittings (ECEF211.0F, Valco Instruments Company Inc., USA).

Line 202: … directed either Helium (He, BIP quality, Air Products and Chemicals Inc., USA) or …

Line 204: … by mass flow controllers (MFC, MKS Instruments Inc., USA).

Line 210: … by a mini pump (P200-GAS-12V, Xavitech AB, Sweden).

Line 241: A custom made LabVIEW software program (National Instruments Corp., USA) was …

5) Line 134: A summary of the contents of each section seems self-explanatory and does not add value to the manuscript.

The respective section was shortened to:

… and one QCLAS-instrument developed at Empa. The compatibility of both analytical techniques for $CH_4$ mole fractions, $\delta^{13}C$-$CH_4$ and $\delta D$-$CH_4$ is assessed and the obtained high-resolution isotope dataset is exploited using a novel moving Keeling plot method. Comparison of measurement results with calculations from two different models (TM5 and FLEXPART-COSMO) and two inventories (EDGAR, TNO-MACC) indicates the potential of this approach to better constrain on isotope source signatures and emissions in atmospheric models.

6) Lines 196-205: Are the valve abbreviations used for descriptive purposes or are they part numbers? If they are for descriptive purposes, it would be great if they also appeared in Fig. 1.

Figure 1 has been updated accordingly.

7) Line 191: the authors have described two traps, one for pre-concentration (“PreCon”) and a second for focusing (“Focus”). Please replace “focus units” with “trapping units” or something similar.

The respective sentence was changed to:

The PreCon and Focus trapping units were glued together with a PT-100 temperature sensor in heat-conducting two component epoxy on a brass standoff.

8) Line 222: capitalize “plus”

The actual IRMS product names from Thermo Fisher do not use capitals: Delta plus XL or Delta plus XP

9) Line 232: In Fig. 1, the temperature for the PoraPLOT Q is listed as 0 ◦C, not 5◦C. Please clarify.

The correct temperature is 5˚C, this was updated in figure 1.

10) Lines 227-228 and 234: repeatability estimate needs +/-; is the estimate reported as SD or SE?

The ± sign has been added and the estimate is reported as SD (also added).

11) Line 237: “base” should be “basis”

Done

12) Line 238: please define “target gas”

We have added the following sentence:

… was analyzed as a quality control tool in order to monitor the long term stability of the analytical technique.

13) Line 245-257: would units of umol/mol not be better considering the magnitude of the concentration, as well and bringing uniformity to the units reported for the laser reference gases?

The authors prefer to keep the units for $CH_4$ mole fractions in the reference air cylinder in nmol/mol, which is a common unit for close to ambient $CH_4$ mole fractions and also used in Figures 2 – 5 and Figure 9.

14) Line 253: I think something is confused here. I believe the authors intend to report no significant linearity, however, as worded (“no non-linearity”), the opposite is conveyed. If there was significant linearity, please report the change in isotopic value with concentration.

This confusion arose from the somewhat unconventional use of the term “non-linearity” in isotope research. The sentence has been clarified as follows

The linear response of the analytical system (independence of the δ value on the amount of $CH_4$ analyzed) was verified by injecting various volumes of reference air up to a volume equivalent to 2700 nmol/mol.

15) Line 276: A direct comparison of the throughput between systems would be helpful earlier in the manuscript. It is referenced later, but I was already wondering about the comparison at first presentation.

The authors agree and added the following sentence:

This translates into an analysis time of 54 minutes per sample of ambient or pressurized air.

16) Line 333: remove “see” in brackets.

Done

17) Lines 334-336: How were these source signatures “adjusted”? Please expand.

The sentence was misleading and has been modified as follows (see comment 10 from referee 2).

The emission inventory was built according to a double constraint: $1^{st}$, each source signature must be chosen within its own uncertainty interval, and $2^{nd}$, 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).

18) Line 358: “Denmark, and Poland”

Done.

19) Line 444: what might be instrumental causes of offsets between systems? Combustion/HTC versus spectroscopy? Further discussion here would be interesting.

For $\delta^{13}C-CH_4$ only a slight offset between TREX-QCLAS and IRMS results of 0.05 ± 0.03 ‰ was observed. TREX-QCLAS results for $\delta D-CH_4$ during the Cabauw campaign were on average -3.6 ± 0.4 ‰ lower than values obtained by IRMS and discrepancies were most pronounced for low $\delta D-CH_4$ values. Differences for $\delta D-CH_4$ between both techniques are irrespective of the fact, that recently a link was established between the $CH_4$ isotopic composition and the international reference materials VPDB and VSMOW, in the framework of the INGOS project (Sperlich et al., 2016) and both laboratories already performed inter-laboratory measurements in a previous campaign as published by Eyer et al. (2016). The enhanced discrepancies for low $\delta D-CH_4$ values, might be explained by a non-linear response of one of the applied techniques. To present the above argumentation the following wording was added to page 16 Line 463:

… as indicated by the red dashed lines (WMO, 2014). Individual measurement pairs, however, can show significantly larger deviations for aforementioned reasons. Differences for $\delta D-CH_4$ between both techniques are higher than expected as both laboratories refer their measurements to MPI-BGC, who recently established a link between the $CH_4$ isotopic composition and the international reference materials VPDB and VSMOW, in the framework of the INGOS project (Sperlich et al., 2016). Therefore, remaining differences can only be rationalized by uncertainties in propagating the scale or by instrumental issues. The enhanced discrepancies for low $\delta D-CH_4$ values might originate from a non-linear response of one of the applied analytical techniques. The mean offset …

20) Fig. 1. Labeling of valves and matching description in text would be helpful

Done. See also response on comment #6 of reviewer #1.