

## ***Interactive comment on “In-situ observations of the isotopic composition of methane at the Cabauw tall tower site” by T. Röckmann et al.***

**Anonymous Referee #1**

Received and published: 9 March 2016

The authors present a comprehensive comparison and evaluation of two techniques for high-resolution, in situ measurements of atmospheric methane. Notably, the two techniques represent both traditional, mass-spectrometer-based measurements and newly-developed quantum-cascade laser spectroscopy measurements. The authors present a robust accounting of comparability of data measurements by each technique and the compilation of data produced by each instrument. Subsequently, the data measurements are compared at length to global and mesoscale models of methane emissions. Importantly, the authors present new techniques for and identify challenges to the data production and evaluation of the fine-scale time-series of methane isotope measurements.

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,

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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.

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.

The “moving” Keeling approach is quite clever, especially as applied source attribution for CH<sub>4</sub> from real-time data collections.

Specific comments

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

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)

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

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.

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.

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Line 222: capitalize “plus”

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

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

Line 237: “base” should be “basis”

Line 238: please define “target gas”

Line 245-257: would units of  $\mu\text{mol/mol}$  not be better considering the magnitude of the concentration, as well and bringing uniformity to the units reported for the laser reference gases?

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.

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.

Line 333: remove “see” in brackets.

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

Line 358: “Denmark, and Poland”

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

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

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-60, 2016.

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