Reply to Interactive comment on “Six years of high-precision quasi-continuous atmospheric greenhouse gas measurements at Trainou Tower (Orléans Forest, France)” by M. Schmidt et al.

Reply to Referee #1,

We would like to thank to the two referees for their very helpful comments and suggestions which will improve our manuscript. We provide here explicit responses to all reviewer comments. In the following, the referee comments are in black color, followed by our replies in *blue color*.

Anonymous Referee #1
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Reliable monitoring of greenhouse gas concentrations is one of the most important goals of atmospheric chemistry and all the technical solutions and inventions should be published as soon as possible. From the other hand, a field evaluation of the analytical instruments requires a lot of time and usually applicability of the technique is proofed after few years. In this light I find this paper very much expected especially in scope of AMT. I hope that the further remarks will not be treated as an overall criticism of the paper but rather critical comments to which referee is obligated.

General remarks: 1) It is not easy to understand the aim of the article. - The paper is rather balanced to present technical aspects of instruments measuring trace gases at different level of the tower. For this purpose one would expect definitely more detailed description of the instruments and validation of the sampling techniques itself (side instruments, valves, cause of their malfunctions and repair routines, validated opinions about used gc, generators, gases, deep statistical analyses commonly used in technical sciences etc...) rather than simple descriptions of records. By this I don’t mean that the record itself is not valuable, in opposite, its detailed discussion would be very interesting but not in AMT. I would suggest to remove the part of the article describing trend analysis but remain the record for overview of the instrument work. Much more interesting are results of target gases and working standard measurements presented in paper. Authors may refine it with more statistical analysis and a bit deeper discussions (e.g. noise analysis or proper detailed nonlinearity).

Referee #1 proposed to delete the paragraph with the trend analysis whereas referee # 2 recommended expanding it. Hence, we decided to keep the discussion as it is, but we clarified that we simply provide an update on the record presented and discussed in parts earlier. This is because we wanted to show the complete picture of available in-situ data for Trainou tower and its variability for different sampling heights.

- CARIBOU is a kind of modifications of commercial instrument, in this case comparison of original and modified instrument would be reliable for evaluating of proposed solutions. Actually the most common technique for high accuracy CO2 measurement is CRDS, authors should

compare both instruments aiming at possible applicability of presented technique, also comparison of Loflo would increase value of the paper. Any
new instrument shell be validated in some way. If the CARIBOU instrument is not recommend for this measurement in future please indicate possible field of application.

At the time the the CARIBOU was developed and installed no commercial analyser provided a sufficient precision. In the manuscript we present the technique used at Trainou tower during the last 8 years and which is still running. However, the CARIBOU will be replaced by a CRDS in the near future and a comparison through parallel measurements has been started, but is not yet completed.

2) Some serious statistical mistakes have to be corrected, what is especially important in case of technical articles. It regards both: notation and calculation. Authors of technical papers dealing with metrology should apply the commonly accepted way of uncertainty expression: e.g. JCGM (former GUM : fttp://www.iso.org/sites/JCGM/GUMintroduction. htm) to obtain the consistency of applied methodology.

We have modified our manuscript according to that guideline.

Detailed comments: -Do the authors posses the footprint calculation or any other model validation of the impact of the surrounding (or regional) area on the results obtained at different levels of the tower, can it be included? Without that the site description is not validated.

No, we do not have a model with sufficient resolution to compute the footprints for different levels at the moment. Such tool would be very helpful for the future and we are working on this issue.

- From the abstract, but later at the instrument description there are constantly mistakes in rounding of the value and standard deviation also notation may be a bit misleading as it is not expanded uncertainty. Notation 1080±0.03 is incorrect.

We carefully verified and corrected the numbers and rounding in the manuscript.

-Pump is not a process variable itself (however pressure, voltage and angular momentum might be).

We replaced the “process variable” on page 578 line 11 with “hardware and parameter settings”.

-Page 8 line 11: add concentration or mixing ratio (if applicable) after CO2 in parenthesis.
done

-Standard gases are very short description - please explain what cylinders (volume, alloy, producer) and pressure regulators etc. are applied.

We added the following two sentences:
“These standard gases are produced by Deuste Steininger (Mühlhausen, Germany) in a gas matrix of so called natural air (mixture of Nitrogen, Oxygen and Argon) and filled in 10L

aluminum cylinders (Luxfer). At Trainou station we use two stage nickel plated brass pressure regulators (Model 14, Scott Specialty Gases, Breda, The Netherlands).

-Chapter 3.2.2: application of averaging values of reference gases measured 5 years from each other seems to be unjustified especially if the changes are small and might be due to the drift of the real air composition inside a cylinder.

We revisit the cylinders again and the difference is for 5 cylinders smaller than 0.01 ppm and for one 0.03 ppm. Regarding our calibration accuracy of 0.02 ppm we cannot apply a drift correction. The calibration cylinders thus show a clear stable behavior.

-Chapter 3.2.2: What is an order of the drift correction, can it be presented on the graph?

From the Licor data sheet, we know that the original instrument has a drift of 1 ppm/24 hours and an accuracy of 1 ppm (datasheet). This is not sufficient for our precision goals and modifications are needed. We modified the sentence P575 Line 20 to " The thermal and pressure control of the instrument minimize the drift of NDIR analyzer of 1 ppm per day specified by the constructor (Li-6252 Instruction manual) to approximately 0.07 ppm per day. In addition we analyse the reference gas during 10 min every hour, and use this results to correct for the instrument drift. After drift correction we obtain a residual drift in the order of 0.001 ppm per day allowing calibrations to occur once per week."

- Why ambient air is not measured at the level 5m by the CARIBOU, what are the purposes to include only one 100m analyses in each pyramid while 50 and 180 are measured twice.

The CARIBOU was designed to measure up to 3 sampling levels. The decision to include the 5m sampling level came when the CARIBOU was already running several years. In the setup with 3 levels we wanted to measure at least the lowest 50m and the highest 180m twice in 1 hour.

-Chapter 3.3: What method was used for test of catalyst efficiency, how variable is it? Why temperature of catalyst was not higher (Agilent gc can control temperature zone up to 400)?

We compared the ratio of the FID signal (CO_2/CH_4 when measuring a calibration cylinder) with the ratio of the mole fraction of this cylinder (CO_2/CH_4). The efficiency was very constant over time during the 6 years of measurement. Agilent recommend 375°C and we increased this to 390°C keeping a security limit of 400°C.

- Indicate the letter Q of Haysep columns (or make the reference to the table) in ECD branch. It is also important to put the supplier as there are few different companies producing this columns (unequal grade).

done

-Page 10 line 16,28 - not analysed but detected (analysis require reference, integration etc.)

done

-Page 10 line 27- Backflush system are commonly used for purpose presented by authors but in this special case both columns are of the same kind. Only one aim of backflush application in this case is to shorten the analyse time.

We rephrase the sentence to “This avoids that compounds with longer retention time elute with the next injection and shorten the analysis time.”

-Linearity of ECD: When authors apply the linear correction function also intercept is important what is its value, what are uncertainties of both parameters.

The slope of the non-linearity is only given for other applicants to see the importance of this issue. In our case we use the two-point calibration to correct for the non-linearity.

-Chapter 3.4. Why meteorological sensors are placed only at 2 heights while authors are suggesting importance of temperature gradient influence on trace gas measurements.

This was more a logistical/budget decision, but it is a helpful suggestion to revisit the possibility to install temperature sensors at all sampling levels.

- There are no 14C results presented as well no ABL heights obtained from Lidar.

We want to show in this paper all measurement and sampling performed at Trainou station, even if these data will be made available to the scientific community by other groups or to a later date.

-Wrong rounding of flask-gc differences.

done

-Chapter 4. There are no uncertainties of: gradients, diurnal variability and trends.

We added two sentences describing the way how we compute the seasonal cycle and the trend and added the uncertainties for the trend.

-can authors comment if the CARIBOU was exchanged in 2013, what are the plans for CARIBOU?

We modified the sentence P589 Line 4-6 to “In June 2013 the station was upgraded with a CRDS analyser to measure CO2, CH4 and CO, in 2015 a second CRDS analyser for N2O will be added. The CARIBOU will be continued in order to have more than one year of overlap between the different analysers. “

-Table 1. What is the value of drift before correction.

From the Licor data sheet, we know that the original instrument has a drift of 1 ppm/24 hours and an accuracy of 1 ppm (datasheet). This is not sufficient for our precision goals and modifications are needed. We modified the sentence P575 Line 20 to “ The thermal and pressure control of the instrument minimize the drift of NDIR analyzer of 1 ppm per day specified by the constructor (Li-6252 Instruction manual) to approximately 0.07 ppm per day. In addition we analyse the reference gas during 10 min every hour in analysis cell, and use this

results to correct for the instrument drift. After drift correction we obtain a residual drift in the order of 0.001 ppm per day allowing calibrations to occur once per week."

-Table 1. Sentence "The reproducibility of the standard gas is 0.02 ppb." is perhaps in wrong place. Which cylinder, why this information is placed here. values of parameter (not only standard deviation) are interesting.

We deleted the sentence "The reproducibility of the standard gas is 0.02 ppb" and add the value of the parameters in table 1.

-Table 2. What does "regulation" refers to?
We deleted "regulation" in the capture of table 2.

-Table 2. Molecular Sieve 5A instead of Å or indicate the problem to the editor?
Done

-Table 2. What "30" means?

We corrected table 2.

-Table 3. Hydrogen most actual scale is MPI 2009.

Modified.

-Table 4. Maybe it is coincidence but for 100m level overall average 2007-2012 is equal to average of yearly mean values - please check the calculation as for other levels results are different due to the different representation of each mean.

We verified it and it is coincidence.

-Table 4. Is overall stdev (2007-2012) really so low while for 2007 it was 1.75, only 2011 is lower than 1.00 but not substantially.

We verified the values and the standard deviations are a bit smaller for the whole period as we applied a two sigma filter.

-Fig.1 Why model is used while wind rose and distribution might be obtained from direct measurements for the years when measurements were performed.

As we have only one wind sensor at the tower, this measurement is influenced by the wind shadow of the tower, and therefore not representative. We add following sentence to paragraph 2: " As we have only one wind sensor at the tower the measured wind direction and velocity is biased by the wind shadow of the tower.

-Fig.1 Is any important information coming from the wind speed cumulative distribution while the distribution is presented?

No, we remove part b of this figure.

-Fig 2. Ethanol instead of "ethalon"

Done

-Fig 3. A lot of text with no apparent reference in the text of the article. complexity of the figure is OK but this require more detailed description inside the paper.

We described figure 3 in a more concise way

-Fig 4. Two valves marked #4
-Fig 4. It would be good to mark the separately heated zones.

We modified Figure 4 to frame the inlet part and the heated zones with different color.

-Fig 5. Scale for CO2 is to big

We kept the same scaling for the GC and the CARIBOU, in order to show the better CO2 performance of the CARIBOU compared to the GC.

-Fig 5. Target value of H2 is constantly drifting (aprox 15ppb/year)- there's no comment to this in the paper.

We add following sentence in paragraph 3.3.2: The Hydrogen mole fraction of the target cylinder shows clearly a drift-like behavior with decreasing values. As explained in paragraph 3.3 the non-linearity of the RGD-system changes for H_2 mole fraction above 800 ppb. The target cylinder has a H_2 concentration of 841 ppb which is much above typical high ambient air values of 650 ppb at Trainou. With our non-linearity test we were not able to correct such high values. Outside of the calibrated range has this target cylinder only a limited suitability for hydrogen and should be interpreted with care.

-Fig 6. "(Electron capture detector)." not necessary.

deleted

-Fig 6. Why the fig5 and fig6 have the different x-scale labelling?

We harmonized the x-scaling labeling of both figures.

-Fig 7. The diurnal cycles from neighbouring months should not be connected with lines (no meaning).

We modified this figure.