

Answer to the Referee 3

General comments:

1. Much of the text is vague and unclear. Balance on details is lacking; in places there is an abundance of detail, but in others sufficient detail is lacking.

R: Based on your specific comments and on those given by Referee 1 and Referee 2, we tried to make our manuscript more consistent. Many details which were indeed missing have been added in answer to the three reviewers whose comments have been very helpful.

2. While the authors make a point of belonging to GAW, little or none of the data presented in the paper are available from the WMO world data center for GHGs (WD- CGG).

R: This is indeed true that only CO₂ and CH₄ measurements have been so far provided to WDCGG, as well as to GLOBALVIEW project, and not recently updated. It is not clear which strategy should be adopted when several instruments are overlapped and then replaced. There was recently discussions about the difficulty for users to decide which dataset using when several of them are provided (e.g. Schauinsland station in WDCGG). Ideally we should use such configuration to improve the estimation of uncertainties. Since the GC at Puy de Dôme will be stopped in 2015 the full time series will be provided to WDCGG within the next months.

3. Why "flux densities"; why not "fluxes"?

R: we replaced "flux densities" with "fluxes" in the whole manuscript

4. Typically, when a noun is used as an adjective to modify another noun, it does not take the plural. In the title, "greenhouse gas measurements" is correct. "greenhouse gases measurements", "flasks analysis", "fluxes uncertainties" (and many other examples) are not.

R: We corrected as much as possible all of the grammatical errors.

5. Use consistent units through out.

R: we converted the Imperial units in SI units when it was needed.

Specific comments:

p3123,12: gas measurements.

R: corrected

p3123,110: show that

R: the sentence has been deleted on the recommendation of referee 1.

p3123,111-12: what are the WMO recommendations? How could a reader outside the GAW community know what this means?

R: the sentence has been deleted on the recommendation of referee 1. The description of the WMO recommendations are given in the "Comparisons of different analyzers" part.

p3123,115: gas measurements

R: corrected

p3124,110: upper case "P" on Protocol

R: corrected

p3125,l24: If you are going to claim to be part of it, you should know the name: GAW = Global Atmosphere Watch

R: corrected

p3126,l16-17: SF₆ does not have the 4th largest RF among anthropogenically-emitted GHGs; it may not be 4th among Kyoto Protocol gases, with HFC-23 slightly ahead of it. A few ODSs (which are also GHGs) have greater RF.

R: we replaced the sentence: "... SF₆ mole fractions, which is ~~the fourth anthropogenic GHG in terms of radiative forcing~~—an extremely stable GHG having a global warming potential of (23,900 Forster et al., 2007)"

p3131,l11: ..in northern France...

R: corrected

p3131,l14: specify inside or outside diameter.

R: we specified outside diameter in the manuscript. We also added the diameter in SI unit.

p3131,l12: It is absurd to claim you "remove the remaining water vapor". What is the dew point of the air exiting the dryer?

R: We corrected the sentence. The dew point is approximately -50°C, depending on the room temperature as the temperature of the ethanol bath varies with it.

p3132,l11: Where are the sample loops? In the room? In the GC oven?

R: we specified that the sample loops are located in the room.

p3132,l10: What is 6.0 quality? It is jargon. Give the purity.

R: 6.0 quality correspond to a purity < 99,9999%. We removed the quality number and gave the purity instead. This has also been corrected in Table2.

p3132: This description of the chromatographic separation scheme combined with an incomplete schematic diagram is very difficult to comprehend. Show the different valve positions with solid and dashed lines, as done e.g., by Aoki (Tellus, 1992). Show example chromatograms. For an experimental paper in a journal largely dedicated to publication of experimental details, this section is very weak.

R: We have added the dashed lines on the valves 1, 2 and 4 indicating the "On" position. This has been specified in Table1.

We also have added a typical chromatogram from the FID and from the μ ECD with the following explanation:

"Figure 3 shows the typical chromatograms obtain by the two detectors used in the GC system. The top panel presents the FID's response in pA. The spike observed between the CH₄ peak and the CO₂ peak in the zoomed panel is caused by the switching of valve 4. The bottom panel of Fig. 3 presents the μ ECD's response in Hz . The first large peak observed at approximately 2.7 min is the O₂ peak which is following by the N₂O peak and finally by the SF₆ peak."

The "chromatogram figure" and the modified GC scheme have also been added at the end of this document.

p3132,l25: CO₂ molecules are not detected by the FID.

R: we corrected the concerned sentence:

“The CH₄ and CO₂ molecules are detected by an FID and a Ni-catalyst, which reduces CO₂ to CH₄.”

p3134,19: What does it mean that GC system performance is controlled with a graphical application? Do you mean performance is evaluated by looking at graphs of instrument parameters? Do you mean you control the instrument and set instrumental parameters from a graphical interface?

R: We clarified the sentence.

“Three to five times each week, a trained operator evaluates the performances of the GC through a dedicated graphical application enabling to draw graphics of the instrument parameters (see Sect. 3.2). Based on these graphics, flags are manually assigned to the data.”

p3136,127: What are these recommendations for?

R: The concerned sentence have been removed from the manuscript because it concerns data which have not been presented. Nevertheless, the recommendation goals are presented in the “Comparisons of different analyzers” part.

p3137,120: In places, English units of measure are used and in other places metric. It should be consistent through out and consistent with the journal’s policy. Give the alternate common unit in parentheses for clarity. Is the diameter o.d. or i.d.?

R: As suggested, we gave the meter units in parentheses when the Imperial units are used.
It is 35 mm outside diameter

p3138,18: What flows through the reference cell?

R: flows in the reference cell is 15ml min⁻¹. This was indicated p3138,111.

p3138,113-14: It does not seem sufficient to calibrate an NDIR twice per year (or even twice per day). Are the analyzer temperature, the flow rates, and the cell pressures controlled?

R: There was not enough full calibration. However there is also a regular calibration of the instrument with the REF measurements which enable to correct for short-term variation. The flow rates and the cell pressure were controlled. The analyzer temperature was not controlled and any variation of temperature is visible in the voltage baseline and is take into account for mole fraction calculation.

p3138,115: Is 3/8 in the length of the line?

R: we specified that it is the outside diameter and also gave the metric unit

p3138,117-18: Is gas from the reference cylinder also flowing through the reference cell of the NDIR?

R: The reference gas flows permanently through the reference cell. Every hour, the reference gas flows through the sample cell for 10 minutes. It has been clarified in the manuscript.

p3139,127: A filter will not prevent outliers, but it could be used to flag them and omit them from further analysis.

R: We changed the sentence:

“The 2-sigma filter has been used to flag the eventual outliers.”

p3141,121: How were the diurnal cycles computed?

R: We have first calculated the yearly trend from the flasks sampled at Puy de Dôme for the three year period considered in this manuscript. Then, we have subtracted the computed yearly trend to the ambient measurement from the January first, 2013. Finally, the diurnal cycles have been calculated by averaging every hour slot.

We changed added following sentence to the MS: “The GHG diurnal cycles were computed from the detrended hourly time series to the reference of January first, 2013. The mean yearly increase rate was subtracted from the time series, before computing the seasonal means diurnal cycles.”

p3142,l14: It is not this simple; sinks at the surface will affect CO₂ and result in lower values at the surface than above the BL.

R: we change the sentence as follow.

“The atmospheric mole fraction variabilities of trace gases are generally larger in the PBL because of the combination between its diurnal variability and the emissions from surface sources.”

p3143,l3-4: The amplitude of the diurnal cycle for N₂O in summer looks significant; why do you say it is nearly undetectable? It is difficult to explain.

R: we added the following sentence:

“except for the N₂O in summer which exhibits an amplitude of 0.25 nmol mol⁻¹”

p3144,l13: What is the value of lambda used?

R: as written in the manuscript, T_{1/2} for R_n is 3.8 days leading to a lambda of 2.1*10⁻⁶ s⁻¹. Nevertheless, we used the approximation given by Schmidt et al., 2003 leading to estimate the term in bracket (eq. 1) to 0.77.

p3147,l12: These look like absolute uncertainties; what makes them relative?

R: We corrected the mistake

p3149,l25-26: GCs run without interruption for years on end, and can often be quickly repaired without specialized technical skills when they fail. You mentioned that your picarro CRDS was sent back twice causing gaps, so how will these new technology analyzers result in fewer data gaps?

R: This is right and we adapted the sentence.

“Compare to the GC system presented in this study, the new analysis technique based on CRDS, FTIR or OA-ICOS achieve better precision and require less maintenance. In consequence, the use of these new technologies enables the development of dense measurement network, such as ICOS, which will further improve uncertainties in the flux estimates.”

p3157: How do "on" and "off" positions relate to the valve positions drawn in the schematic diagram?

R: as previously suggested, we added dashed lines in order to make the distinction between the two positions. We adapted the text in consequence.

p3160: State in table title that values are 1 sigma.

R: done

p3163: If the fluxes are based only night time measurements, the table description should state that. These estimated fluxes are difficult to compare without uncertainties on them.

R: We added the “night” mention and the respective uncertainties in the Table.

p3165: The schematic diagram seems incomplete. Some valves have nothing connected to some of their ports. The flow of gas through the catalyst, then to a tee, branching between an FID and a needle valve seems odd. Where is the supply of carrier gas to the ECD channel sample/inject valve?

R: We completed the diagram by adding the connection between the ECD carrier gas and the sample/inject valve. It is exact that we do not use all of the port on the valve #4.

We corrected the FID part.

p3168: What does "detrended based on 1 January 2013" mean?

R: Please, see explanation above (answer to p3141, l.21)

Additional figures:

