Response to Referee #2

The authors appreciate the reviewer's constructive comments and detailed feedback for improving the manuscript. The point by point response is as follows:

For Reviewer's General comments:

In the paper, the authors mentioned several times about $\delta^{18}O$ but there is very little results not examples presented about isotope (in particular the record and associated trend of $\delta^{18}O$ is not shown, page 4012 they choose a dry period favorable for calibration of ^{18}O but nothing is shown!). This could be ca valuable addition to the paper as this isotope is even more sensitive than $\delta^{13}C$ to drift and to measure with a high precision.

As mentioned in the response to the Referee#1, although the traceability of $\delta^{18}O$ is the same as $\delta^{13}C$, originally the authors intended to focus on the trend of $\delta^{13}C$, which may be more relevant for anthropogenic sources. The authors agree with the referee that adding the $\delta^{18}O$ record in the paper will be appreciated by the community. In the revised paper, the $\delta^{18}O$ measurements at Alert will be included and the title will be modified to reflect the changes.

For Reviewer's specific comments:

In general, the paper would require and carful proof reading as there is a lot of typing, spelling, used abbreviations or misunderstanding errors and mistakes. In some cases these might be problematic for a good understanding of the paper (in particular for non specialists). On the contrary, it is sometime may be too detailed (acid digestion part for example).

The suggestions will be taken. Proof reading by native speakers of English will be conducted. The authors understand the importance of a clear and concise writing and will make a special effort to improve the manuscript as a whole.

I 'am not sure that Appendix at the end of the paper are fully useful (except may be appendix A). The second one is more a recipe and the last one can found elsewhere about error propagation method.

The suggestion will be taken.

The Appendix C will be removed and only the results will be used.

The Appendix B will be shortened to keep the only important part. The recipe is unique for obtaining the quality of the consistent data. For the reviewer's information, we were asked many times to provide the recipe.

P4005 L7-8: the significance of mbl is not straightforward! Please explicit marine boundary layer.

The suggestion will be taken.

Based on the information on the web (http://www.esrl.noaa.gov/gmd/ccgg/about/global_means.html)

"MBL" sites are from a subset of NOAA network sites where samples are predominantly representative of a large volume of the atmosphere. These sites are typically at remote marine sea level locations with prevailing onshore winds. The use of MBL data results in

a low-noise representation of the global trend and allows us to make the estimate directly from the data without the need for an atmospheric transport model.

P4005 L22: The's why, please reword properly for example " this is the reason why".
The suggestion will be taken.

P4006 L12: The time span … is derived, please reword that sentence, this is not clear!
The sentence will be reworded to be clearer. The authors tried to say that to derive a reliable atmospheric trend for a certain period of time, it is required to present a stability of standard used during the same period of time.

P4006 L18-19: Style a bit too telegraphic! Please reword in a more "written style"
The content will be reworded.

P4006 L25: miss one opening parenthesis before "directly'.
The missed parenthesis will be added.

P4007 L11: Add "is defined as THE "property …"
The "THE' will be added.

P4007 18-19: please add a reference for the NBS19/VOD value cited here
The suggestions will be taken and the following references will be added in the revised version.

Friedman, I., O'Neil, J., Cebula, G. (1982): Two New Carbonate Stable Isotope Standards. Geostandards Newsletter, 6 (1), 11-12.
Hut, G. (1987): Consultants' group meeting on stable isotope reference samples for geochemical and hydrological investigations, Report to the Director General, International Atomic Energy Agency, Vienna, April 1987.
Coplen, T. B., Brand, W. A., Gehre, M., Gröning, M., Meijer, H. A. J., Toman, B., Verkouteren, R. M. (2006): New Guidelines for ^{13}C Measurements. Analytical Chemistry 78 (7), 2439-2441.

P4007L22: what is an "instrument liberality"? Not clear what the authors are meaning here.
This was a typo. This should be "instrument linearity".

P4008 L2-3: "the other ARE secondary" there are three standards mentioned just after.
The mistake will be corrected.

P4008L7: NBS instead of NBA!
The typo will be corrected.

P4008L13: "… discussed later. As shown on the …"
The suggestion will be taken. The text will be modified as "… (which will be discussed later). As shown on …".

P4009 L5: delete the “closing parenthesis”.
This was a typo. The closing parenthesis will be moved just after the “ ‰”.

P4009L15: “ to decreases or increases of OF the magnitude …”
The typo will be corrected.

P4009 L16: The author claim about “ideal clean conditions” for IRMS but without precision. What are those ideal clean conditions? How to evaluate them? When and how do you say it is clean? This is an important point for reader that would like to apply and evaluate this method elsewhere.
The clean condition is a relative status for each individual IRMS instrument. Theoretically, when the high vacuum reading is at the lowest, the mass readings at background scanning (e.g., 18, 28, 30, 32, 40, 44) are the lowest, the cleanest condition for an IRMS should be reached and the largest Big Delta value should occur. An ideal clean condition (i.e., the cleanest status) may never be reached. For MAT252, one criterion is the background count of mass 44. The electronic offset for mass 44 is ~ 100mV (90-100mV), which is about 200 counts after the signals-pulse conversion (Merritt and Hayes, 1994). This indicates that the electronic zero in background count is 200. The “Background Count” is listed in Table 4 to show the relationship with the Big Delta values. As shown on the Table, the relatively clean conditions (~ 200) were reached and maintained during the periods of 2007-2011.

Reference:
Merritt and Hayes (1994) Factors controlling precision and accuracy in isotope-ratio-monitoring mass spectrometry. Anal Chem. 1994 Jul 15; 66(14):2336-47.

P4010L2-4: There are inconsistencies between the “consistent values” given in the text and in the table A1 page 4033 (example: Cal1-Cal2 about -46 permil given in the text and about -45 permil What is the exact value used at the end?). Same for Cal2.

These were overlooks. The averaging result over 10 year (2011- 2011) shown on Table 2 and 3 should be used for the exact values in $\delta^{13}C_{VPDB\text{-}CO2}$, i.e. -45.81‰ and – 2.60‰ for Cal1 and Cla2, respectively.

P4010 L8: QA/QC
The suggestion will be taken.

P4010L10: add references for the NBS18 literature value!
The suggestions will be taken.
Friedman, I., O'Neil, J., Cebula, G. (1982): Two New Carbonate Stable Isotope Standards. Geostandards Newsletter, 6 (1), 11-12.
Stichler, W. (1995): Interlaboratory Comparison of new materials for carbon and oxygen ratio measurements. Proceedings of a consultants meeting held in Vienna, 1 - 3. Dec. 1993, IAEA-TECDOC-825, 67-74.

Verkouteren, R. Michael, Donna B. Klinedinst (2004) Value assignment and uncertainty estimation of selected light stable isotope reference materials: RMs 8543-8545, RMs 8562-8564, and RM 8566, In NIST Special Publication 260-149, 2004 Edtion.

P4012 L14: “between February AND April”
The “to” will be replaced by “and”.

P4012 L21: delete “that” which is double.
The “that” will be deleted.

P4012 L25: delete of before greater
The suggestion will be taken.

P4013 L15: delete “are” at the end of the line!
The “ are ” will be deleted.

P4013 L23: I am not sure I understand well. For me there is only one term on the left side of Eqn. 3.
Sorry, this was a mistake. The left side should be “the right side”.

P4013 L25: Finnigan
The misspelling will be corrected.

P4014 L2-5: consider rewording, the sentence is quite a bit difficult to follow. Is there a physical explanation for this effect in the machine?
The suggestion will be taken.
Due to a certain degree of cross contamination between the sample and the reference, the measured isotopic values (sample or reference) are always the results of mixing. As a Big Delta is derived from the measured values, thus, the uncertainty (or the variation) of the Big Delta depends on the uncertainties of the measured values. It is known that the cross contamination shows a relatively large effect on the measured sample and reference with a large difference in isotopic ratio, in comparison with a relatively small effect on those with a small difference in isotopic ratio. In order to address the cross contamination issue fundamentally, only using a WRG (working reference gas) with a δ value close to the sample’s value is not enough, the IRMS should be kept as clean as possible all the time. To do so, a large Big Delta should be used as a criterion for the ideal clean conditions.

P4014 10-14: sentence stating with “as mentioned … is not fully readable, there is a t alone in the middle of the sentence … not self-understanding.
Sorry, this is a typo. The “t” should be replace by “it”.

P4014 L15-17: Couldn’t this effect be linked with the fact the Cal2 is the most used standard and then the one where the air is the most circulated then potentially minimize fractionation of missing effect that might arise in the manometer, or inlet lines?
It is not sure if the question is well understood.

For the referee's information, the same number of ampoules for Cal1 and Cal2 were produced by acid digestion reactions (always as a pair), even though the Cal1 was not measured as often as Cal2 by IRMS.

P4015 L19: delete "and" after time.
The suggestion will be taken.

P4016 L1: here the IRMS uncertainty is given as 0.01 whereas it was shown and written to be 0.02 in the previous sections of the paper. This is not coherent.
The 0.02 ‰ mentioned in the previous sections are not the analytical uncertainties of the IRMS (MAT252) but the overall uncertainties which associate with the whole procedures, including acid digestion, CO_2 extraction and IRMS analysis. For one dual-inlet run (8 blocks of sample and reference), the analytical uncertainty of the MAT252 is usually at the level of 0.01 ‰ (always < 0.02‰).

P4016 L7: Yr!
The "yr" has been used consistently in the whole manuscript at the suggestion of publication Production Team from Copernicus Publications.

P4016 L10: Could you please add a reference as "it is known"
The suggestion will be taken.

P4016 L13-15: consider re-wording this sentence which is difficult to follow.
Thanks for pointing this out! The author intended to say that the absolute value of "R" (i.e. $^{13}C/^{12}C$) can not be precisely measured because it easily varies depending on measurement conditions and instruments. Only the ratio of R (i.e., R_A/R_B, where A and B are two different materials) can be precisely measured.
As suggested, the sentence will be rewording.

P4016 L27: Yr!
The "yr" has been used consistently in the whole manuscript. The reason has been mentioned above.

P4017L20: yr
The "yr" has been used consistently in the whole manuscript. The reason has been mentioned above.

P4018L3 and 25: QA/QC
The suggestions will be taken.

P4020 L9: Passed
The mistake will be corrected.

P4020 L16: delete will and correct associated.
The corrections will be done.

P4021 L4: QAQC
The suggestion will be taken.

P4021 L5-8: sentence not understandable
The content will be reworded to improve the readability.

P4025: L 14, 22: yr
The “yr” has been used consistently in the whole manuscript. The reason has been mentioned above.

P4029: Table 1
Could it be possible to add and compare some of the reported literature values for NBS18. This could give more strength to the table and section.
The suggestion will be taken.

P4031: Table 3
Columns are not well aligned in the ave. for IRMS
Thanks for your careful reading! It was likely that this was due to the format conversion since the original table was correct. This correction will be done.

P4032 Table 4.
It seems to me that there is a little drift of the big deltas toward larger values over time and for both instruments. This seems correlated with cleanliness of the MAT252. What about the IsoPrime? Is there any other explanation for this? Is surprising that in the meantime there is no drift when Cal1 and Cal2 are calibrated against NBS19 or NBS18?

Yes, as shown on Table 4, the little drifts of the Big Deltas (Cal2 vs. Cal1) over time were partially due to the cleanliness of the MAT252. As shown on Table 2, there is a slight drift of the Big Deltas (Cal1 vs. NBS19) over time for both instruments too. The drifts in two sets of Big Deltas suggest that Cal1 might be also changing over time, while the cleanliness of the MAT252 was changing. Fortunately, it is also implied that the change of Cal2 has been small over time.

P4033: Table A1: inconsistencies between the “constant values” given in the text and in the table. Measurement order 5 please delete the second “and” on the purpose text. In the figure caption, “ the batch of APB2”, APB2 is not defined and is this information useful that way.
Table A2: Again there are different and incoherent values for Cal1 (-4 !!! instead of -45!) and Cal2 (-2 instead of -2.6 initially in the text)
The inconsistencies of the “constant values” have been addressed and the mean values over the 10 yr for both Cal1 and Cal2 are used in Table A1 and A2.

In measurement order 4 from Table A2, what happens to the samples QC if the Big Delta values are not within the 2 standard deviation range? Are they invalidated?
Yes, the Cal2 is invalid and a new Cal2 or a new pair of Cal1 & Cal2 will be measured until the Cal2 is valid.

There are inconsistencies in the notations WRG or WR! Are those gases the same (I suppose they are all WRG)?
Yes, the inconsistencies will be addressed. All “WR” will be changed into “WRG”.

P4036: Figure 2 and caption: the WR is not defined in the legend and is referred too as WRG in the text page 4008, L19 (working reference gas). Why not adding the relative position of $\delta^{18}O_{VPDB\text{-}CO2}$?
The inconsistency of using “WR” and WRG” will be addressed throughout the whole manuscript. The relative position of $\delta^{18}O_{VPDB\text{-}CO2}$ will be added.

The figure on the left panel is not easy to follow and the caption doesn’t help too much (too more details and not enough description and explanation of figure itself).
The caption will be shortened to improve the conciseness of the content. The Figure 2 will be modified to improve the explanation of the figure itself.

P4038: Figure 3:
Why not adding big delta 46? Replace C by c in coefficient.
The big delta 46 will be added. Originally, the authors’ intention was focusing on $\delta^{13}C$. The typo of the “C” will be corrected.