General comments:

The manuscript of Pieter P. Tans et al. with the title “Abundances of isotopologues and calibration of CO_2 greenhouse gas measurements” is reporting the development of a method to calculate amount of substance fractions for individual CO_2 isotopologues, based on X_{CO2}, $\delta^{13}C$ and $\delta^{18}O$ values. The new method is applied in combination with a new CO_2 calibration system, consisting of three laser spectrometer with different technology and sensitivity for isotopologues: CRDS ($^{16}O^{12}C^{16}O$), OA-ICOS and QC-TILDAS ($^{16}O^{12}C^{16}O$, $^{16}O^{13}C^{16}O$, $^{18}O^{12}C^{16}O$, $^{17}O^{12}C^{16}O$), to account for isotopic differences among standards.

The topic is very timely and of very high interest for a large number of readers of Atmospheric Measurement Techniques involved in atmospheric monitoring of greenhouse gases (especially CO_2) and their isotopic composition. The manuscript is a fundamental conceptual and technical description on the WMO CO_2 calibration scale and therefore a basic document to define the state of the art. As mentioned by the authors, the developed technique can be applied to other molecules, where isotopologues-specific values are desired or isotopic differences affect analytical techniques. Furthermore, the technique to calculate fractional distribution of isotopologues of CO_2 (and other target substances, e.g. CH_4, N_2O) will be of great benefit for users of optical isotope laser spectroscopy (e.g. Edgar Flores et al., Analytical Chemistry (2017), DOI: 10.1021/acs.analchem.6b05063).

The manuscript is very carefully written, concise and clear, and it can be published with very minor revisions. The authors might consider a small number of suggestions to further increase the readability / impact of their work.

Specific comments:

Page 12 Line 5: Eq. 21 + 22: The two formulas for P(826) and P(726) are incorrect as the second denominator is supposed to be squared.

Although the mathematical equations are clearly presented, it would be very helpful for the reader to have a sample calculation, for example as a supplementary file, on how to calculate mole fractions of isotopologues (X(626) etc.) from $\delta^{13}C$, $\delta^{18}O$ and X_{CO2}. Implementing the equations of the presented manuscript (Eq. 12 – 15 or 19 – 22) on an example from Flores et al. (Analytical Chemistry (2017), DOI: 10.1021/acs.analchem.6b05063, e.g. Table 1 mixture 1) results in different mole fractions of isotopologues than given by Flores – please state on the differences in the calculations, and cite the work of Flores at al. in your manuscript.

Minor revisions/suggestions:

Page 2 Line 17 – 20: The role of the world calibration centres (WCCs) to independently verify the implementation of the calibration scales at laboratories or monitoring stations could be mentioned here or elsewhere in the text.

Page 3 Line 11: Results of most recent key comparisons (CCQM) with national metrological institutes (NMIs) could be included here.

Page 4 Line 20 (Eq. 1): Multiplication with “ x 1000” is frequently used, but should be avoided according to Tyler Coplen, RCM (2011), DOI: 10.1002/rcm.5129.

Page 5 Line 32: Scott Marrin Inc. offers “ultrapure air” and “cryogenic ultrapure” air but not “ultra high purity air” please specify accordingly.

Page 6 Line 11: The sub-sentence “… to properly address isotopic issues when …” is colloquial and might be rephrased.

Page 9 Line 24: Is error the correct wording in this context, or should it be bias? Please check.

Page 11 Line 6: Replace 0.194 with 0.195.

Page 14 Line 1-2: Please specify the wavelength region used for the analysis of CO_2 isotopologues as different possibilities exist and this might vary from instrument to instrument. Was there any additional temperature stabilization implemented for the optics / electronics of the QC-TILDAS or is the laboratory air-conditioned?

Page 14 Line 11: What is the reason that $\delta^{17}O$-CO_2 cannot be calibrated independently, no IRMS measurements?

Page 14 Line 35 – Page 15 Line 1: The sentence “The solenoid valve fails to the idle gas …” is unclear and might be rephrased.

Page 16 Line 13-20: The memory effect might cancel out, but it will add to the uncertainty of the isotope analyzers. Has this been quantified? Are improvements possible, such as longer flushing times, or optimization of the flow scheme?

Page 18 Line 24: The sentence “Isotopic standards should be calibrated by IRMS measurements” could be valid for the given example but is not a correct general statement, please specify the sentence.