

Anonymous Referee #1

General comments:

Overall, this is a well written paper describing and interpreting carefully made measurements. The scope of the paper is somewhat narrow, but it is still very much worthy of publication in ACP because it is essentially demonstrating a new source of data and presenting some very nice interpretation. I think some of the interpretation, especially that of the CO:CO₂ ratios and the $\delta^{13}\text{C}$ signatures can be improved by taking account of more known processes that affect them. I have specific suggestions to that effect below. On the other hand, the interpretation of the d18O signals is excellent, and I have just one suggestion to make another estimate of the d18O of soil water from observations.

Specific comments:

24564, 14: As admitted later in the paper, it's a bit dangerous to refer to JFJ as 'free troposphere', because while this is true most of the time, it sometimes may in fact be in the PBL as shown by high pollution signals. I suppose it is an open question as to whether or not JFJ is actually in the PBL or just receiving venting from the PBL. Nonetheless, the data presented here puts at least some doubt in the statement that JFJ is 'free troposphere'.

Since this study focuses on the winter period we think it is well justified to refer to the observations as being representative for the free troposphere. During winter, air pollutant concentrations at JFJ do not exhibit a measurable diurnal cycle, which is different from the summer period when the PBL often rises above the altitude of JFJ in the afternoon. We have added a reference on Zellweger et al. (2000) and briefly discuss in Section 2.1 the frequency of occurrence of different types of air masses in different seasons.

“Given its high elevation and year-round accessibility, the station is an excellent place for long-term monitoring. Observations at JFJ are mostly representative of the free troposphere, particularly during autumn/winter when about 70% of the air masses can be characterized as undisturbed free tropospheric, 20% as free tropospheric but disturbed by synoptic uplift of polluted air, and only about 10% by more local uplift and convection (Zellweger et al., 2000).”

24564, 121: The current fossil fuel emissions are above 8 GtC/yr, not 6. See http://cdiac.ornl.gov/trends/emis/tre_glob.html (this has a doi and is referenceable.)

Value updated and Reference added as suggested.

24565, 15: The value of ~19 per mil refers only to C3 plants and doesn't account for the ~20% of plants that are C4, and have a fractionation of about 4 per mil. Overall, the global average fractionation for terrestrial plants should be more like 15 or 16 per mil. (see e.g. Scholze et al, 2003, GRL; Suits et al, 2005, GBC).

During the writing we were focused on Europe and tacitly considered the C3/C4 plant distribution for the foot-print area seen from Jungfraujoch only. At this scale, the contribution of C4 plants is < 2% (e.g. R. P. Collins and M. B. Jones, *Vegetatio*, 64, 121-129, 1985) and as such negligible. However, in the context of the introductory part a global value is more adequate. Therefore, we changed the value as proposed.

24567 14: Insert 'a' in front of 'mid-infrared'.

Done.

24567 1 28: What is NABEL? Explain or provide a reference.

The abbreviation is defined with its first appearance (24566 L18).

24568 11: The units of m³/hr are quite odd. More normal is L (or dm³) per minute as is later used. Additionally, it should be stated explicitly that this is an STP volume by saying so parenthetically or by using ‘standard liters per minute’ (or SLM).

Changed as suggested.

24568 13: ‘through a 15m stainless steel tubing’ should be changed to: ‘through 15m of stainless steel tubing’ or ‘through a 15m length of stainless steel tubing’.

Done.

24568 17: When discussing the dilution effect of water, it would be helpful to read what the requirement of drying needed to be to avoid a bias of xx ppm. E.g., in order to keep CO₂ dilution biases below 0.1 ppm, the dewpoint of the airstream needs to be kept below about -35°C. Was the water vapor measured after drying? And if so, what was the typical dewpoint of the air? I wouldn’t expect a water vapor dilution effect for the isotope ratios, but what is the sensitivity to broadening?

The dilution effect is given by: $[\text{CO}_2]_{\text{dry}} = [\text{CO}_2]_{\text{wet}} / (1 - [\text{H}_2\text{O}])$, i.e. 1% change in the water mixing ratio will induce 1% change in the CO₂ mixing ratio. Thus, for a bias < 0.1 ppm in CO₂ the water mixing ratio must be < 0.00025 %v (or 250 ppm), which corresponds to a dew point temperature of -37°C. The nafion drier (PD-100T-48) in our sampling setup is specified to achieve a dew point of -45°C at a gas flow of 0.5 slpm. In another study (Tuzson et al., 2010), we did laboratory tests to investigate the dilution effect (including drying efficiency at different flow rates and water content) with a QCL based spectrometer, which measures the water mixing ratio. We found a good agreement with the specifications and, thus, expect that a dew point of < -35°C is obtained under the sampling conditions at the JFJ.

Regarding pressure broadening due to the presence of water vapour, the referee is rightly assuming that little or no effect is expected for the isotope ratios. This is due to the fact that the pressure broadening coefficients for the main and minor CO₂ isotopic species at 4.3 μm are very similar (see e.g. Sung et al., *Can. J. Phys.*, 87, 469–484, 2009). Note that this is a special case and should not be taken as a general rule.

Text modified:

“... to eliminate volumetric effects on mixing ratio determinations. Potentially, water vapour could also have an impact on the isotope ratio through collisional broadening (Tuzson et al., 2010). However, in this very specific case, the effect is likely to be small because the pressure broadening coefficients for the main and minor isotopic species at 4.3 μm are very similar (Sung et al., 2009).”

24568 129: Remove ‘exact’ here. Isotopic ratios (as stated later in the sentence) are always relative to a standard reference material, whose isotopic composition is not known to absolute accuracy.

Modified as suggested.

24569 18: The ‘CO₂ converter’ is more commonly referred to, in my experience, as a ‘methanizer’.

Done.

24569 125: The compromise between the actual altitude and the model surface height seems reasonable, but it would be worth it to re-run the trajectories at the actual elevation of the measurements to see if there is any difference in footprint region (these simulations are very fast in my experience). The relief around JFJ is very steep so that using the actual height also seems reasonable – in other words, treat JFJ as an extremely tall tower above Switzerland.

Even if the slopes are steep, the topography around Jungfrauoch has a strong influence on the air flow which is very different from the situation of a tall tower. The compromise is based on extended tests with different release altitudes which indicated that neither the true station altitude nor the model topography, but rather an intermediate level, provides best results (in terms of agreement between simulated and measured carbon monoxide concentrations, where simulated CO was obtained by combining the FLEXPART footprints with an emission inventory). The chosen altitude is also

based on comparisons of specific humidity values measured at JFJ with corresponding observations over flat terrain from the nearby radiosonde station Payerne.

We changed the sentence to:

“Particles were released at 3000 m a.s.l. which is a compromise between the model surface altitude (2170 m asl) and the real station height (3580 m asl). This choice is based on sensitivity tests with different release altitudes and on a comparison of specific humidity measurements at Jungfraujoch with vertical profiles acquired from routinely launched soundings at the nearby station Payerne (46.8°N, 6.9°E) over flat terrain. The best agreement with the sondes was found for an altitude of about 3000 m a.s.l. indicating that air reaching Jungfraujoch is experiencing significant uplift due to topographic effects which can not be resolved by the model.”

24570 122: The statement that 10 minute averaging can smooth out random measurement error is odd considering the 50s minimum in the Allan variance plots mentioned earlier. It would seem that 1 minute averaging is all that is needed to smooth out measurement error. Smoothing out environmental variability, on the other hand, could benefit from longer averaging times. The actual environmental variability and instrument variability (if I am reading correctly) should not be conflated.. We agree and have removed the sentence to avoid this ambiguity. The choice of 10 minute averages is to smooth out short-term variability, because we are mainly interested in the pollution events, which have much longer time scales. The text was modified to include this.

“High resolution time series of CO₂ mixing ratio and the corresponding $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values are shown in Fig. 3. Since we are mainly interested in the pollution events rather than the short-term environmental fluctuations, the data points were averaged for 10 min, which is consistent with the sampling/calibration routine.”

24570 126: For figure 2, again, why is 2 minutes chosen for averaging, when the Allan variance suggests 1 minute can be used with equal precision. Also, Fig. 2 would benefit from ^{13}C being added. I (and possibly other readers) would be interested to see these data.

Two minutes were chosen because this is consistent with the calibration routine for tank A. We consider anything between about 50 s and 150 s to be a good integration time with respect to the instrumental precision. One should note that the results from an Allan variance plot have a relatively large uncertainty. This is illustrated in Tuzson et al., (2010), where the range and average of 32 Allan plots (30 min each) are shown in the same graph. Two minute averages is close to the minimum in the Allan variance plot and was chosen here to account for the two minutes calibration interval.

Regarding the $\delta^{13}\text{C}$ plot, it was skipped from the paper because of space and placement symmetry considerations and our expectation that most readers will be interested in the more “difficult” to measure $\delta^{18}\text{O}$ data. Of course, we appreciate the interest in the ^{13}C data and have added it to Fig. 2.

24571 11: ‘precision’ should be used here instead of ‘accuracy’, because the 0.08 ppm (e.g.) comes from the width of the distribution, not the deviation of the median from the assigned value of Tank B. But, in fact, it would be very nice to know what the accuracy or bias is here, by comparing the field determinations of Tank B to its calibrated or assigned value. Can this be provided?

The instrumental precision for CO₂ mixing ratio is about 0.03 ppm, defined by the Allan variance minimum. We argue that the 0.08 ppm mentioned in the text for CO₂ corresponds to the repeatability of the measurements including the uncertainty of the calibration procedure. As such, the dispersion of the values reflects the measurement uncertainty (expressing the closeness of agreement between a measurement result and the value of the measurand) and provides information about both precision and trueness (bias). The histogram is centred over the true value and only for the delta values can a minor bias be observed (e.g. the $\delta^{13}\text{C}$ determined by IRMS was $-11.05(2) \text{‰}$, while the histogram of the QCLAS data was centred over $-10.99(1) \text{‰}$.)

24571 19: The work of Turnbull et al (2010) which is cited here clearly demonstrates that one cannot assume zero biological contribution to CO₂ signals, even in winter. Furthermore many other studies (e.g. Monson et al, 2006, Nature), show that respiration can be significant even in very cold environments. However, I believe it would still be reasonable to say that biological activity during this period is likely to be smaller than in other months, minimizing but not eliminating its effects on the interpretation of pollution signals.

We understand the concern of the reviewer. However, there is a significant difference between what is considered as biological CO₂ in our paper and the above mentioned work of Turnbull et al (2010). They use the following expression: $CO_{2bio} = CO_{2obs} - CO_{2bg} - CO_{2ff}$, where CO_{2ff} refers to fossil fuel combustion only, excluding biomass burning. They found up to 8 ppm ± 2 ppm contribution from above and below ground respiration, biofuel use and wood combustion for home heating. In our case, biofuel use and wood combustion (likely to be dominant) are included in the anthropogenic term.

Moreover, the climate in Sacramento, California is rather Mediterranean with no snow cover and seldom freezing events. The ambient temperature during the flights was above 5°C (up to 16°C) with significant rainfall between the sampling episodes (see <http://www.wrh.noaa.gov/climate>). In fact, one might expect significant ecosystem respiration in this case. In contrast, the northern European countries had temperatures below 5°C (mean February temperature around -2 °C). Only Italy (events I and II) had rather mild weather conditions (between 5 and 15°C, average 7°C) comparable with Sacramento. However, these events actually show steeper CO:CO₂ slopes than those with footprints of northern Europe.

We have clarified the text accordingly.

“(1) the biological activity (photosynthesis/respiration) is presumably the smallest for this period of the year, thus, minimizing but not eliminating its effects on the interpretation of pollution signals.”

24571 112: Define or reference ‘Foehn events’.

Changed sentence to “pollution events associated with a variety of pronounced meteorological conditions including frontal lifting and south Foehn events typical for the situation at Jungfraujoch (Zellweger et al. 2003).”

24571 114-16: Change from ‘. . . allowed to . . . and determining. . . ’ to ‘allowed us to . . . and determine. . . ’

Done.

24571 1 29: change ‘with partly’ to ‘partly with’

Done.

24572 19: Using data from Spivakovsky, JGR, 2000 and the JPL rate constant data indicate that at 44 N in Jan in the lower half of the atmosphere, the lifetime of CO with respect to OH averages more 12 months. This doesn’t significantly change your point, but I would change ‘several months’ to ‘about a year’

We agree and modified the text as suggested.

24572 19: While CO can be used as a tracer for fossil fuel, it is only quantitative if the flux-weighted average of all the emission ratios are known and there is no biological contribution. And, as several of the studies you cite show, (especially those using ¹⁴C), the emission ratio can not simply be assumed from inventory data. I think ‘quantitative’ should be stricken; it’s enough to say that it is a tracer.

Agreed and modified the text as suggested.

24572 113-14: Change: ‘ratios are indicative for’ to ‘ratios indicate’

Done.

24572 118: Were the regressions calculated using a ‘model-II’ regression (see Pataki, 2003 GBC and Miller, 2003, *Tellus*) which is necessary to avoid biases in these kinds of plots?

Yes. We used the orthogonal distance regression method to account for the errors in both CO and CO₂ data. Furthermore, these plots were also checked for autocorrelation.

The text was adapted accordingly. “Here, the orthogonal distance regression method was applied using the total CO mixing ratio and the CO₂ above tropospheric background (ΔCO_2), the latter being calculated as the difference between observed and background concentration level and referred to as fossil-derived CO₂.”

24572 123-24: While it might appear that one could count on the background taking out the influence of the biosphere, I think this is unlikely. The enhanced pollution events seen at JFJ are coming from the surface, and this is exactly the location where biospheric respiration contributions are coming from as well. So, anytime pollution enhancements are seen, these are likely to be accompanied by respiration enhancements as well. One nice way to look at this would be using ¹⁴C. (n.b. While Ingeborg Levin’s group collects samples ¹⁴C at JFJ, these are unfortunately two-week integrated samples from which the background and the pollution events cannot be disentangled.)

We agree that there may be some bias in the anthropogenic CO₂ due to biospheric respiration and this contribution cannot be ascertained from the data. The tight correlation between CO and CO₂ strongly limits the amount of such enhancements. The ¹⁴C method is definitely the best way to determine CO₂ originated from fossil fuel combustion processes. However, the uncertainty of the $\delta^{14}\text{C}$ determination (5 -10 ‰) involves an uncertainty in the CO₂ff of about 2-4 ppm (see e.g. Levin, I. and Karstens, U., *Tellus*, 59B, 245–250, 2007). Furthermore, it excludes the contribution from biomass burning. See also discussion at 24571 19.

We have slightly adapted the text.

“In this approach, the contribution of respired CO₂ from the biosphere is **partly** accounted for in the background air as a seasonal effect **and** the y-intercept given by the regression in Fig. 4 should represent the CO mixing ratios of continental background air for the specific season.”

24573 13: Unfortunately, high correlations are not necessarily an indication that CO:CO₂ emission ratios are well constrained. As shown in Turnbull et al, 2010, (and as explained in the comment directly above), biological CO₂ can be co-transported with fossil CO₂ and fossil CO preserving tight correlations, but giving biased apparent emission ratios. While the ratios for event II III and IV may correspond well to the Swiss tunnel study cited, an examination of the footprints in Fig. 7 shows that only for event IV, is the footprint more Swiss than of any other nation. Events II and III show a much stronger influence from Italy and France, respectively, than Switzerland. This is significant because other studies give higher auto emission ratios: e.g. close to 0.020 ppm/ppm for Germany (as cited in Vogel et al, 2010, *Tellus*). I do not know if Switzerland stands out as having exceptionally clean burning cars within Europe, but given that JFJ is receiving air from all over Europe, the observed CO:CO₂ slopes of ~0.010 ppm/ppm could also be indicative of higher fossil fuel emission ratios then diluted by a biological contribution. Similarly, the hypothesis of wood burning in event I is highly speculative, without knowing the emission ratios for cars and other industrial sources in central Italy. I think the conclusions presented here are premature without considering emission ratios throughout the regions of influence: not just Switzerland, and not just for cars. In terms of this paper, I do not insist on such an analysis (though it would be interesting to see), but rather that the authors not ascribe the apparent emission ratios (i.e. slopes) to Swiss auto emissions and acknowledge these other factors.

Finally, the analysis of ¹⁸O later in the paper is consistent with the presence of a significant amount of biological CO₂ reaching JFJ. Briefly, in order to explain the source signatures for ¹⁸O, the authors conclude that air must have had a significant residence time near the surface for invasion to have occurred. While invasion is an abiotic process, it requires liquid, not frozen, water to occur – the same conditions that are required for soil respiration.

We agree with the reviewer that much more work and longer time series are needed to improve the data analysis. The assumption of biogenic respiration as a contributing term in the observed CO₂ in-

crease opens interesting possibilities to better understand the observed values for the CO:CO₂ slopes as well the $\delta^{13}\text{C}$ Keeling-plot intercepts. This is, however, beyond the scope of this study, and a follow up paper is planned to deal with this and other topics not covered by the present manuscript. A data set covering more than two years of continuous measurement should shed light and possibly quantify such terms.

With respect to the paper of Vogel et al. (2010), the cited 0.020 ppm/ppm was only observed in summer, and as Vogel et al. admit, it is a challenge even to speculate on the cause of such surprisingly elevated ratio. During winter time a mean CO/CO₂ ratio of 0.0129 ppm/ppm was observed.

We have, nevertheless, modified the text to underline the preliminary nature of these interpretations. “The correlation found for all selected time periods is significant, with r^2 as high as 0.985. However, these tight correlations are not necessarily a clear indication for specific CO:CO₂ emission ratios. Since biogenic CO₂, originating mainly from above and underground respiration, can be co-transported with anthropogenic CO₂ preserving the tight correlations between fossil CO₂ and CO, a certain bias in the emission ratio cannot be excluded. However, the slope values of the regressions compare well with the results of tunnel measurements representing local traffic and transport on the Swiss highways, which report a vehicle weighted mean CO:CO₂ [ppm/ppm] ratio of 0.008 ± 0.003 linked to diesel and gasoline vehicles (Vollmer et al 2007). It should also be mentioned that Vogel et al. (2010) report for Heidelberg, located in the industrialized upper Rhine valley in southwestern Germany, a winter time CO:CO₂ emission ratio of 0.0135 ± 0.0025 . This higher value could be indicative that the pollution events investigated in this study may contain significant (up to 20 – 40%) contribution from biogenic sources.”

24573 18: What is the parenthetical (3) after 0.008? If an uncertainty, please write as 0.008 ± 0.003 . Modified as suggested.

24573 124-25. The >75 ppm range necessary for good statistics on Keeling plots is actually a function of the analytical precision. This range doesn't apply to the very nice precision of the QCL system in question here and this repeated statement from Pataki et al is a bit misleading and unnecessary.

Agreed and changed text.

24574 5-17: The interpretation here is a bit simplistic. The range of isotopic signatures across events could represent geographic variability in fuel mixtures, likely a variable amount of natural gas combustion (which is isotopically depleted relative to coal and oil). The number given by Andres et al (2000), is, I believe (I don't have access to the reference) a global number and is not specific to W. Europe, where natural gas usage is higher than the global average (see fuel type consumption in the cdiac reference given above). Along these lines, just because European emissions have not changed much, it does not mean the mix has been stable. In fact, looking at Switzerland (<http://cdiac.ornl.gov/trends/emis/swi.html>), we see a stable total masking a big rise in natural gas usage over the past 20 years. Furthermore, for the same reasons that the CO:CO₂ ratios probably reflect a combination of fossil and biological sources, so to do these Keeling plot intercepts. These complexities should be acknowledged in the discussion – not merely as complications, but as opportunities to leverage the footprint information to gain insight into potential regional differences in combustion processes.

We agree and have adopted the text to include the suggested complexity.

“Furthermore, it is tempting to say that the range of isotopic signatures across events could represent geographic variability in fuel mixtures, likely a variable amount of natural gas combustion. However, a clear distinction between various types of combustion processes is hindered by the same complexity as mentioned in the case of CO and CO₂ correlation. Similarly, the Keeling plot intercepts probably reflect a combination of fossil and biological sources, thus giving biased (by up to 2 %) apparent source signatures.”

24575 127: While the regression approach seems reasonable, I would also check the IAEA isotope hydrology data to see if it agrees with the current approach. Overall, I think the 18O analysis is very good.

We considered the Global Network of Isotopes in Precipitation (GNIP) database and surveyed the oxygen isotopes data in precipitation using the Water Isotope System for Data Analysis, Visualization, and Electronic Retrieval (WISER) application. However, the latest data we were able to find was December 2008 from Switzerland and December 2007 for other countries. This motivated the use of the regression approach which should closely reflect the real situation, but within a given uncertainty.

24577 112: Capitalize Foehn here.

Done.

24577 120: As mentioned earlier about the regional $\delta^{13}\text{C}$ of fuels, the Netherlands and Belgium use about 30-40% natural gas (compared to Switzerland about 16%), which could help explain the -30 per mil signature from this event.

We agree that a detailed analysis of the emission inventories of the countries covered by the footprint area and the inclusion of biogenic respiration may improve our data interpretation and give more insight for the CO:CO₂ emission ratios as well as for the $\delta^{13}\text{C}$ signatures. However, this is beyond the scope of this manuscript and we would prefer to apply such investigation on a more extended time series.

24580 11: 'illustrates' to 'illustrate'

Done.

Fig2. Change Date (tt.mm.yyy) to Date (dd.mm.yyyy)

Done.

Fig 4. Explain more clearly (if this is the case) that the number in parentheses refers to the uncertainty in the last digit.

Text changed as suggested.

Fig5 and 6. Here the uncertainties are presented differently. I suggest choosing a uniform presentation.

Text modified to reflect this case.

Anonymous Referee #2

The use of a QLAS system for quasi-continuous CO₂ in air stable isotope measurements is demonstrated to work at a precision level of ± 0.05 per mill level ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$). The Jungfraujoch station has been selected to test the system in a "free troposphere" environment over 2 years. Data are presented for a limited time window of 30 days in early 2009. During this time pollution events from local sources occurred, which altered the CO₂ concentration and isotopic signature. The major advantage of the presented system is the ability to analyze stable isotope data continuously (at a rate of about 70 data points per hour). This allows to infer sources of air masses to be traced down to their presumed origin using a Keeling plot end-member technique. However, this can work only for two clearly distinct components (like a general C₃-derived source of CO₂ mixing into background air), but it will not allow to constrain this a lot further when several sources are involved or the sources have similar isotopic compositions. For 18O the Keeling plot technique does not provide conclusive results, owing to the decoupling between source concentration and isotopic signature. The paper is well written and the information given is clear and concise. Publication is recommended after addressing a number of specific questions and comments:

24564, L21: The 6 GtC/yr needs updating (e.g. LeQuere et al, Nature Geoscience 2 (2009) 831; Friedlingstein et al, Nature Geoscience 3 (2010) 811)

Done.

24565, L5: the discrimination of $\delta^{19}\text{O}$ for C3-photosynthesis only

See our answer to Referee #1

24565, L18: except for sea-water, which is largely uniform. However, in contrast to terrestrial exchange equilibration is slow (not catalyzed by enzymes). The resulting CO_2 $\delta^{18}\text{O}$ is also a function of water temperature.

This topic is discussed in details later in the manuscript (see Eq.2).

24565, L25: Please provide scale for the -17 ‰ presumably the VPDB-CO₂ scale.

Across the manuscript the delta values are given on VPDB-CO₂ scale (see 24569 L5)

24567, L7: While “simultaneous” gas calibration can also be done in series (provided the time intervals do not become excessively long), the simultaneous recording of the isotopologue absorptions is more important for stable isotope ratio precision.

Actually we are doing both, simultaneous sample/gas measurement and isotopologue absorption recording. This approach is described in details in the given references (see 24566 L25).

24567, L24: The 50s integration time presumably applies to a complete cycle including the measurement of all 3 isotopologues. Are these measured in sequence? If so, could this be improved to a simultaneous analysis within identical time windows? How much time is spent on each absorption feature?

The absorption features of all three CO₂ isotopologues are recorded within one spectral scan. The laser is scanned at 3.7 kHz repetition rate through the absorption lines and the acquired spectra are averaged in the DAQ for one second. In other words, the one second spectrum is already the result of averaging 3700 individual spectra. The 50s integration time indicates that actually the data can still be further averaged for this amount of time to smooth out random noise components. Detailed information is given in the references; however, we have also slightly modified the text to make this aspect clearer.

“Briefly, a pulsed quantum cascade laser (QCL, Alpes Lasers, Switzerland) emitting at 2310cm^{-1} is used as mid-infrared light source and scanned at 3.7 kHz repetition rate over the absorption lines of the three CO₂ isotopic species.”

24568, L1: Replace ‘50 m³ h⁻¹) with ‘833 L min⁻¹’ (use consistent units throughout the paper)

Done.

24568, L19ff: The exact way of reporting data is not clear. I assume that the short-term data use Tank A and the reference cell. Is Tank A then used as the longer term calibration anchor point when it is used for flushing the sample cell? What are the differences for the Tank A ‘sample’ against the reference cell Tank A?

Modified text to avoid ambiguity:

“The instrument’s sampling system periodically switched between air sample and calibration gases. The latter were measured as follows: (1) standard Tank A (every 15 min); (2) standard Tank B (every two hours); (3) standard Tank C (every 12 h).”

24569, L4: The correct reference would be ‘P Ghosh et al, Calcite-CO₂ Mixed into CO₂-free Air: A New CO₂ in-Air Stable Isotope Reference Material for the VPDB Scale, Rapid Comm. Mass Spectrom., (2005) 19: 1097-1119’

Done.

24569, L11: Please provide isotope scales information
[See reply on 24565, L25](#)

24569, L14: . . . values are better. . .
[Done.](#)

24571, L19: I do not understand this statement. Why is scatter “much accentuated at the permil scale”? Compared to what? Is there another scale available for stable isotope ratios? Or do you compare to something else (mixing ratios?). Then you would compare apples with pears.
[This paragraph seems to be misleading; we have modified the text.](#)

24574, L24: I do not see this as clear cut as stated. Why should the -27.4 ‰ intercept in figure 5 a be more specific for coal or biomass combustion, but not oil? It could be anything but natural gas, provided we can exclude a more complex mixture.
[We agree that this is not clear cut but merely “consistent” with the CO:CO₂ ratio and the foot-print calculations, as stated in the text.](#)

24576, L8: Here, the scale is misleading. +46.6 is the fractionation effect for any CO₂ equilibrated with water at 0 °C.
[The absolute \$\delta^{13}\text{C}\$ and \$\delta^{18}\text{O}\$ value of the J-RAS is closely linked to the V-PDB scale and is used to serve as a primary scale anchor for CO₂-in-air measurements. Therefore, all data given in this text is given in the V-PDB scale if not stated otherwise.](#)

24576, L23: scale missing
[See above.](#)

24589ff, Fig 4 to 6: Please provide an identification with the events from Fig. 3
[Done.](#)

24587, Fig 2: The $\delta^{18}\text{O}$ stability data still exhibit a clear sinusoidal signature, which is not seen in the CO₂ mixing ratio. Where does this originate from? How can this be improved? Are these data used for correcting the $\delta^{18}\text{O}$ record?
[The seemingly sinusoidal signature is probably without any physical meaning since its amplitude is within the reported uncertainty of the measurements.](#)