Response to anonymous referee #1 on "Evaluation of four years continuous d13C(CO2) data using a running Keeling approach"

We want to thank this anonymous reviewer for very helpful suggestions and comments, which have helped to improve the manuscript. We have revised the manuscript and outline the changes in the following.

General:
The manuscript deals with a four year combined record of $\delta_{13}C(CO_2)$ and CO_2 from Heidelberg in Germany. An of running Keeling plot approach has been applied in order to estimate the source signatures from the data.
The approach including their set criteria were tested using a STILT model dataset representing the Heidelberg conditions as good as possible. The agreement between the known source signature in these modelled dataset and those retrieved from it using their running Keeling approach is surprisingly well. The application of their approach to the four years observed dataset yields a clear seasonality of the retrieved source signature between quite well defined limits using a 100 hours smoothing filter. Then they discussed the shortcomings of the method to disentangle the different unknowns, namely the fossil fuel share and its isotope composition as well as the isotope signature of biosphere source. They conclude that it is only possible to retrieve robust results under quite strict conditions, i.e. (i) a monotonous CO_2 increase of at least five ppm over a five hours interval and (ii) an uncertainty of below two permil for the source signature. This restricts their derived source signature dataset by 85%, which is very substantial, which is somewhat a disadvantage.
Furthermore, they nicely document that the biosphere source signal can only reliably be estimated during summer. The fossil fuel source signature is in contrast only reliable during winter, when only $\delta_{13}C(CO_2)$ and CO_2measurements are available. I really enjoyed reading this manuscript and I suggest accepting it with only minor revisions.

Detailed comments:
Abstract: L 4: ...opening the door to the quantification of CO_2 shares...or opening the door to quantify CO_2 shares...

We have added "the", such that it reads: "…opening the door to the quantification of…".

L8: Disentangling this seasonal source signature into shares of source components is, however, ...

We have changed this in the revised manuscript.

L13: ..., such as D14C(CO2) or oxygen/carbon dioxide concentration ratios.

As oxygen/carbon dioxide concentration ratios have not been used quantitatively to distinguish between fossil fuel and biospheric CO_2, we have decided to not include this sentence here in the abstract. For consistency, we also remove $^{14}C(CO_2)$ in the abstract. However, we mention both tracer methods in the conclusion of the revised manuscript instead.

Main text
P2, L6-7: style, two times insight into ....reformulate one

We have changed the second one to "This may be used to study biospheric responses…".

P2, L32-33: eq. 2 and 3 are equivalent, therefore the about equal has to be changed to an equal sign in eq. 3.

We use an equal sign in the revised manuscript.

P3, L10ff and L23ff is referring to the same topic, namely what kind of regression analyses should be used. These two parts should be combined. I personally would move the second part up.

L10ff refers to the difference between Keeling plot approach and Miller-Tans plot approach and l. 23ff refers to the fitting algorithm. We agree that these two topics should be discussed together and move the second part up, as suggested by the reviewer.

P3, L 20f: This statement is two strict and has not been mentioned like this by Miller and Tans (2003). Otherwise, the comparison between regression filtered and STILT filtered source estimates would not be as good since most of the time simultaneous occurring sinks and sources are present.

Miller and Tans (2003) state that "counter-intuitive results can occur any time fluxes of opposing sign are combined and then sampled in the atmosphere" and that "the precision […] depends on choosing our measurement environment to match closely the assumptions in our models". We agree that the statement "the determination of source mix is not per se possible" is too strict and instead write that biases may be introduced when fluxes of opposing sign occur simultaneously.

P3, L25: What is WTLS? Is it the same as geometric mean regression (GMR) as discussed in Zobitz?

WTLS (weighted total least square fit) is similar to a ODR (orthogonal distance regression) used by Zobitz et al. (2006). It has been developed by Krystek and Anton (2007) as stable algorithm for line fitting and takes into account the uncertainty in x and y direction. We give the citation and add a comment in the revised manuscript.

P4, L1: occurring

We have changed this in the revised manuscript.

P4, L4: this approach leads to a strong auto-correlation of the source signature values.

We have changed this in the revised manuscript to “this approach leads to a strong auto-correlation of neighboring source signature values.”

P4, L5: maybe reformulate to something like: We choose five hours as a compromise between maximal number of data points and source mix constancy.

We have reformulated this sentence to “We choose five hours as a compromise between number of data points and thus, of robust regression, and source mix constancy.”

P4, L21: ...as a decrease would be due ...(delete of)

We have deleted “of”.

P4, L19ff: Why do you not apply a simple r2 criteria? Your criteria yield a significant reduction of data and corresponds to r2 larger than 0.9.
What is the benefit of using your criteria of source signal uncertainty?
R2 would also be independent on the regression method applied, the retrieved slopes and intercepts not. Maybe the errors are again independent, I have not checked it.

As the reviewer points out, R^2 is the same for different regression models. The reason is that R^2 is independent of the uncertainty of $1/CO_2$ and $\delta^{13}C$. However, the standard deviation of the offset (and of the slope) in the WTLS-fit takes into account the errors in x and y (see Eqs. 24 c,d in Krystek and Anton, 2007). We therefore prefer using the standard deviation of the offset instead of R^2.

In preliminary work, which we do not show in the manuscript, we have also tried using a $R^2>0.9$ criteria to filter the data and have found no significant differences to using the standard deviation of the offset. Both criteria seem to be similarly valid in the studied catchment area.

Section 2.3:
(structure) For the reader it would better to improve the visibility of the actual criteria in use: maybe with (i) ...(ii)

We have adapted this numbering at the end of section 2.3 in the revised manuscript to summarize the filter criteria.

P5, L11: ...are 0 - 2‰ more enriched than the “filtered” source signatures (blue) as expected from our criteria.

We feel that the insertion “as expected from our criteria” is not necessary here, as this is explained in the following sentence.

P5, L16: ...Keeling method and the used filter criteria on the model...

We have changed this in the revised manuscript.

P5, L26: about instead of ca.?

We now use “about” in the revised manuscript.

P6, L12f: this finding is in excellent agreement with a previous source seasonality estimate by Sturm et al, which should be mentioned:
Sturm, P., M. Leuenberger, F.L. Valentino, B. Lehmann, and B. Ihly, Measurements of CO2, its stable isotopes, O2/N2, and Rn-222 at Bern, Switzerland, Atmospheric Chemistry and Physics, 6, 1991-2004, 2006.

We have added this reference by Sturm et al., 2006 in the revised manuscript together with Schmidt, 1999.

P6, L27 delete sub-title 4.2.1

We have deleted the subtitle 4.2.1. Note that upon suggestion of reviewer #2, we have applied a new and more classical structure to the manuscript.

P7, L17: maybe it is better to use whether instead of if

We use “whether” in the revised manuscript.

P7, L24: ...the mean measured isotope signature.

Ok.

P7, L25: delete significantly

We have deleted significantly in the revised manuscript.

P8, L29 and 33: Why are the values different (1.5 ‰ and 1‰)?

They should actually be the same and be 1.0 ‰. We have changed this in the revised manuscript.

P9, L2: Assuming constant isotopic end members over the course of one year, we would be able....

We have made this stylistic change in the revised manuscript.

P9, L6: ....to the change in the fraction of respiration...

We have added “the” in this sentence.

P9, L6: is it correct to say that in principle photosynthesis would also lead to an isotopic change but since you are analysing only positive CO2 gradients, i.e. CO2 release, you restricted it to respiration only. You might state this explicitly.

Yes, this is correct. We make a respective comment in the revised manuscript (Sect. 2.3).

P9, L14: ...into the fuel CO2 share.

We added “share” at the end of this sentence.

P9, L17: ..there is a need of either ...at the sources.

We have deleted this subsubsection on request of reviewer #2, and have embedded it into the conclusions. There we now use “there is a need of” instead of “must be”, as recommended.

P9, L15ff (4.2.5): Nothing is said about the possibility to use oxygen measurement. A clear distinction between biospheric and fossil fuel sources can be calculated based on the different oxidation ratios for these two sources. Furthermore, calibrated CO/CO2 measurements are helpful as well as already documented in various studies.

In this study, we want to focus on using $\delta^{13}C$ and CO_2 only as tracer. However, it is true that O_2/N_2 measurements provide an additional promising tracer to separate between fossil fuel, biogenic and oceanic sources (e.g. Keeling, 1988; Bender et al., 2005; Steinbach et al., 2011). To our knowledge, using O_2/N_2 as tracer for fossil fuels on a regional scale has not been comprehensively studied so far. On the other hand, studies using CO/CO_2 at regional scale are various (e.g. Levin and Karstens, 2007; Vogel et al., 2010; Vardag et al., 2015b). We have added a short comment in the conclusion, but we do not ponder upon the different tracers, as this is not the scope of the manuscript.

P9, L27: ..aiming at an improved quantitative ...

We have made this stylistic change in the revised manuscript.

P9, L29: ...and CO2 records for a potential partitioning of source contributions.

We have changed this in the revised manuscript.

P9, L29f: this last sentence is not clear, please reformulate or delete it.

We have deleted this sentence in the revised manuscript.

P10, L26: ditto as P8, L29 and L33.

We change these numbers that both read 1.0 ‰ in the revised manuscript.

Appendix A: P11, L3: ...air parcel originated from.

We have added “from” in the revised manuscript.

P11, eq. A1: still not clear to me why one has to use absolute concentration values. It leads to different delta values.

Indeed using the absolute values leads to different numbers. However, as Miller and Tans (2003) point out and as we elaborate in this manuscript (see Fig. 1c), as soon as negative fluxes occur, the resulting source signature does not lie within the range of the source signature end members anymore. Respective results are, thus, not interpretable as gross-flux weighted mean source signature anymore. As we are interested in determining the gross-flux weighted mean source signature, we take absolute values for the calculation of the reference mean source signature. In this way, we can check if the computed source signature equals the gross-flux weighted mean of all sources; with that is an interpretable and intuitive measure.

P18, Fig1c: The lengths of the red and green arrows is not the same since one has to balance CO_2 and not $1/CO_2$. However, it might be not visible

Upon impulse of this reviewer, we have checked and can confirm that the difference is not visible for typical CO_2 (and $1/CO_2$) ranges.

P18, Caption: ...or wind direction change (transportation)

We have changed this in the revised manuscript.

P18; Caption, line 3: what do you mean with correct isotope signature, it is still a mixture and it has not been split up yet.

Here, we mean the flux-weighted mean isotopic source signature (following eq. A1 in the manuscript). We have added a comment in the figure caption of the revised manuscript.

P20, It would be worthwhile to have the CO2 changes along with these graphs (at least for b and c).

We add the CO_2 changes to Fig. 3 in the revised manuscript.

P21: high values in 2011? Correct or artefact due to calibration issues?

Even though 2011 was the very first year of our measurements, we do not have any hints (e.g. target gas measurements or other), which would explain any artefact due to calibration issues. Therefore, we suggest that this is a real effect.

P21: It would be nice to add the modelled curve for the year 2012.

We have now added the modelled curve for the year 2012 in Figure 4.

P22: Why don't you use the radiocarbon that you have available and base your fossil fuel on inventory estimates?

Good point: We have actually considered including this record initially. However, we have decided to leave $^{14}CO_2$ out because of three main reasons:
1) We felt that the main statement of the manuscript, which concerns the usefulness and pitfalls when evaluating and interpreting continuous $\delta^{13}C$-CO_2 measurements, would be weakened by including an additional tracer.
2) Many monitoring stations do not have ^{14}C measurements available. Therefore, our study is more representative if not including the ^{14}C-CO_2 measurements, but using (generally available) emission inventory data instead.
3) We only have integrated samples of $^{14}C(CO_2)$ available, which cannot be compared to continuous $\delta^{13}C$-CO_2 measurements directly due to the integration effect (described in Vardag et al., 2015b).

P23: are the lower and upper 5% important? Have you used this filtering?

They are not used in the manuscript. We have removed these in the revised manuscript.

References used in this reply:

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