Spatiotemporal patterns of the fossil-fuel CO_2 signal in central Europe: Results from a high-resolution atmospheric transport model

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We would like to thank the reviewer for the efforts to review this manuscript and the thoughtful comments and remarks. All of the referee's comments have been carefully examined and addressed in the revised manuscript.

Referee #2

General comments*:*

The authors firstly presented the spatiotemporal patterns of the atmospheric fossil fuel CO2 concentration over the central and southern Europe, and then conducted a sensitivity study on the impact of fossil fuel emission on the atmospheric CO2concentration varying with emission reduction and regions.
Overall, this study is interesting, fits the scope of ACP, and shows enough findings to be published.

> **Authors:** Many thanks for the positive remarks.

However, it is quite confusing to me sometimes that what the authors referring is fossil fuel emissions or fossil fuel CO2 signals (concentration). One is a flux based concept, another is mixing ratio. The authors need to make it clear.

> **Authors:** The reviewer is absolutely correct in that we clearly need to distinguish between the two. We went through the text again and ensured that this is taken care of. Specifically, we added "signal" or "concentration" whenever we refer to the atmospheric concentration and "emission" whenever we refer to the fluxes.

The authors conducted a reasonable amount of the statistics to support their findings, i.e., Table 1. However, based on their results, they used quite a few statements like "good agreement", "agree well", etc. These kinds of statements have no reference in

the entire manuscript: no inter-comparison with other studies, and/or no significant test, which makes these statements are too vague and scientifically meaningless.

Authors: We agree with the reviewer that care needs to be taken when using such qualitative statements. But we feel that we had (mostly) adhered to common scientific standards, e.g., those established by IPCC, by using "good/well" when the probability exceeds 66% (likely in the parlance of IPCC) and very good/very well if it exceeds 90% (very likely for IPCC). We have checked the paper again and made sure that we use such statement only when they are fully justified.

It will be very helpful if the authors list more information in the figure caption. For example, it's not clear to me that which figure is based on the all time series, which based on the daytime afternoon. The bottom line is that readers can understand the figures without checking the text.

Authors: We added the time period to the caption of almost all figures. We extended the caption also with regard to other elements, such as the layer considered in the plots (also responding to a comment by the first reviewer).

Overall, the authors did model evaluation (comparison to observations) based on the daytime values but did the annual mean based on the all time series. The models are struggled with simulating the nighttime CO2 in general. The all time series will very likely bias the entire results of annual mean. There are quite a lot details needed to be fixed and listed below as the specific comments. I would recommend the authors to work on the consistency of the statement and correct those comments before they resubmit the manuscript.

Authors: The reviewer is correct that current generation atmospheric models tend to deviate more strongly from observations at night compared to the day, largely owing to the difficulties of correctly modeling the night time shoaling of the atmospheric boundary layer. This is why we were evaluating the model simulated CO_2 against daytime observations only in the paper. But while we feel that this does not lead to any inconsistencies with our showing later results reflecting a true daily average, we nevertheless decided to make all evaluations and results consistent, i.e., being based on true daily averages. A key reason for this choice is also that the difference is actually not particularly large.

We thus delete the sentence (line 236/37)

"In order to minimize the impact of local influences, we use the average CO2

concentrations between 12:00 and 18:00 local time, i.e., the time of day of maximum vertical mixing."

and add "daily averaged" in front of "measurements" to read
"by comparing them to daily averaged measurements" (line 231)

Furthermore, we changed all values in Table 1. No change is needed for Figure 3, as this was already based on a daily average.

Specific comments:

1. *Line 22, what do you mean the last sentence of the abstract?*

Authors: It means that changes in the standard deviation of atmospheric CO2 might be used as a method to detect changes in fossil fuel emission. We re-read the abstract and found it to be clear. Thus, no changes were made to the text.

2. Line 50, Newmant et al., (2016, ACP) also studied the fossil fuel section based on the observation approach.

Authors: Thanks for pointing this out**.** This reference was added to the text

3. Line 63, it can't be true that the model dynamic is fully resolved. The subgrid processes should be parameterized.

Authors: This was indeed not carefully worded. Of course, no model can fully resolve all scales of motion. In response, we change the sentence to "A key advantage of this set of approaches is that the spatiotemporal dynamics is fully resolved to the limit provided by the resolution of the transport model."

4. Line 87-93, the authors is explaining the possibility to detect the fossil fuel signal, which I was assuming that it referred CO2 concentration. However, the two approaches – bottom up and top-down – are used to quantify CO2 fluxes in general. This confusion appeared a lot throughout the entire paper and needs to be fixed.

Authors: Here, in fact, we are referring to methods that either detect the signal (concentration) or the fluxes. In response, we changed the sentence to
"In fact, several studies already explored the possibilities to detect the fossil fuel signal and the emissions driving them."

5. Line 132, in the model setup, the flux components are called surface flux and, can be

considered as surface boundary condition. The prescribed global model output providing the advection on the boundaries of model domain is lateral boundary conditions. The statement regarding these drivers is not clear to me.

Authors: Yes, the description of how we treated the surface and lateral boundary conditions was not entirely clear (see also main comment by the 1st reviewer). In response we have added quite some text to this effect. In addition, we added "surface" to the specific line mentioned by this reviewer to read:

" we employ a regional high-resolution atmospheric transport model for the European domain and prescribe lateral and surface boundary conditions for the various components that constitute atmospheric CO_2".

6. Line 170, the authors used time function to scale the monthly emission in time. I recommend the authors to make a figure that shows the time series of the scaled emission components and the total emission on top of Figure 2a. I also would like to see how the authors reconcile the discontinuity between weeks and between months. (plot the month and weeks?)

Authors: It seems as if we had confused the reviewer regarding our approach. Our starting point are the annual totals (line 171). These annual totals were then scaled with the product of three time functions, i.e., f(t) = f_hour(t) * f_week(t) * f_season(t), where f_hour is the diurnal time function, f_week is the weekly one, and f_season the seasonal one. As these are time-continuous functions, there is no discontinuity between weeks and months. In order to make our approach clearer, we added this equation to the methods. Namely, the text now reads:

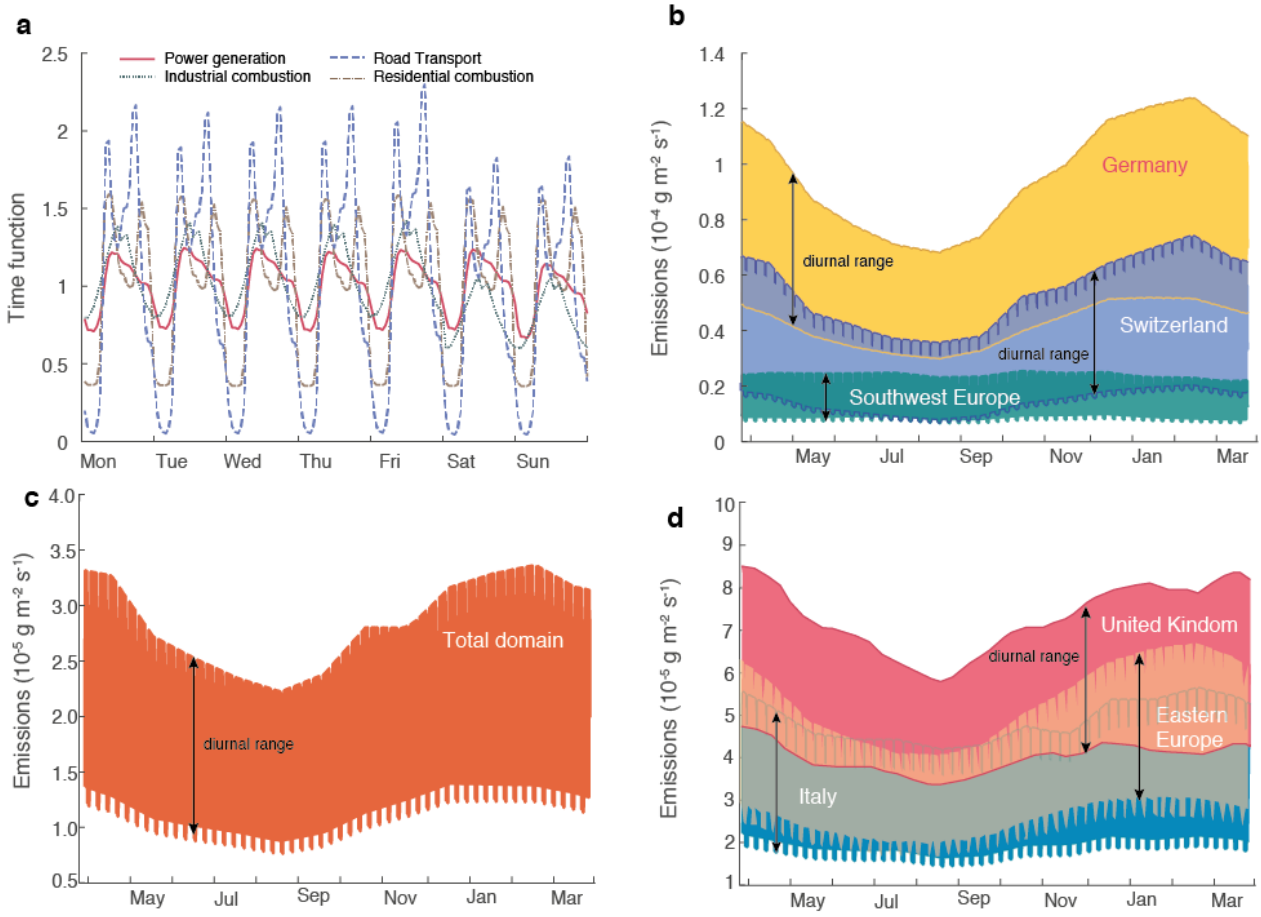
"The scaled emission flux (E(t)), was then given by

$E(t) = E_{ann} * f_{hour}(t) * f_{week}(t) * f_{season}(t)$,

where t is the time, E_{ann} is the annual integrated fossil fuel emission, $f_{hour}(t)$ is the diurnal time function, $f_{week}(t)$ is the weekly one, and $f_{season}(t)$ is the seasonal one."

We are also not entirely sure about how to respond to the request by the reviewer to add the totals, since this is what is already shown in Figure 2b, albeit for a few countries only. What we propose to do is to add two plots showing the average emission density for the domain, as well as that for a few

more countries (see below). Since we computed the impact of the sectorial emissions with time-constant emissions, it makes no sense to show these scaled emissions.



Proposed modified Figure 2: We have added the total emissions for the domain (panel c), and for the three additional countries/regions (panel d).

7. Line 207, a table to describe all of the fossil fuel tracers will be very helpful for readers to follow.

Authors: We propose to add the following table to the text.

Name	Character	Time function
CO2_C_tot	*Tracer of total emissions*	constant
CO2_C_he	*Tracer of emissions from heating*	constant
CO2_C_in	*Tracer of emissions from industry*	constant

CO2_C_pp	*Tracer of emissions from power plants*	constant
CO2_C_ro	*Tracer of emissions from road transport*	constant
CO2_C_re	*Tracer of emissions from other sources*	constant
CO2_p_tot	*Tracer from total emissions*	Time varying
CO2_p_CH	*Tracer of emissions from Switzerland*	Time varying
CO2_p_GE	*Tracer of emissions from Germany*	Time varying
CO2_p_FR	*Tracer of emissions from France*	Time varying
CO2_p_IT	*Tracer of emissions from Italy*	Time varying
CO2_p_AU	*Tracer of emissions from Austria*	Time varying
CO2_p_NL	*Tracer of emissions from the Netherlands and Belgium*	Time varying
CO2_p_UK	*Tracer of emissions from the United Kingdom*	Time varying
CO2_p_SW	*Tracer of emissions from southwestern countries (Spain and Portugal)*	Time varying
CO2_p_EA	*Tracer of emissions from eastern European countries*	Time varying
CO2_p_other	*Tracer of emissions from other regions (e.g. maritime emissions by shipping)*	

8. Line 207 and 226 contradict each other. Please double check how many fossil fuel tracers used, and correct it if it's not.

Authors: 17 is the correct number. We thus changed "15" into "17" in Line 226

9. Line 225, see item 5 above.

Authors: we changed “the lateral and boundary conditions” to “the lateral and surface boundary conditions”.

10. Line 235, does R>0.7 mean significant correlation?

Authors: In these comparisons, any correlation with an R>0.7 is highly significant. We adjusted the text by adding this information.

11. Line 248, “underestimate” or overestimate? All of the bias values in Table 1 are positive but the one at Mace Head.

Authors: Thanks for spotting this mistake. It should be "*overestimate*". But please note that since we changed the basis of our evaluation, all numbers in Table 1 have changed.

12. Line 248 – 259, I can’t find those value in the Table. It’s confusing.

Authors: We changed the sign in table 1.

13. Line 261, in Table 1 the STD for the model are ~4 ppm and ~9ppm at Mace Head and Hungary, respectively. Please correct them and carefully check the values are stated in the manuscript.

Authors: They are correct, because we are describing the model results in Line 261. But please note that since we changed the basis of our evaluation, all numbers in Table 1 have changed.

5. Line 300, see item 10.

Authors: Compared to biases from other models, e.g., those from Bozhinova et. al., (2014), these comparisons are good to excellent. We thus consider our statement as warranted.

15. Line 314, what is the criteria for “the good to excellent evaluation”? Could the authors quantify them?

Authors: (see also item 14) Of course, such statements are always relative to an expectation formed by how well previous modeling studies were able to fit the observations. Using the results of Bozhinova et. al., (2014), as a benchmark, our

results are indeed very encouraging, leading us to conclude that our model is doing a good to excellent job. Nevertheless, in response to this comment, we deleted "good to excellent"

16. Line 317 – 319, "It is particularly ….". " The presence of an overall …" These two sentence contradict each other.

Authors: We deleted the sentence "It is particularly encouraging to note the good agreement not only for the fossil fuel CO_2 component, but also for total atmospheric CO_2. "

17. Line 320, the statistics were made sometimes based on the daytime values sometimes based on the all time series. Overall, the authors did model evaluation (comparison to observations) based on the daytime values but did the annual mean based on the all time series. The models are struggled with simulating the nighttime CO2 in general. The all time series will very likely bias the entire results of annual mean.

Authors: We refer back to our response above. We do not share this reviewer's concern about his/her perceived inconsistency between the evaluation and the results section. But we agree that it is somewhat awkward to use two different averaging periods. We thus now show true daily means for both evaluation and results.

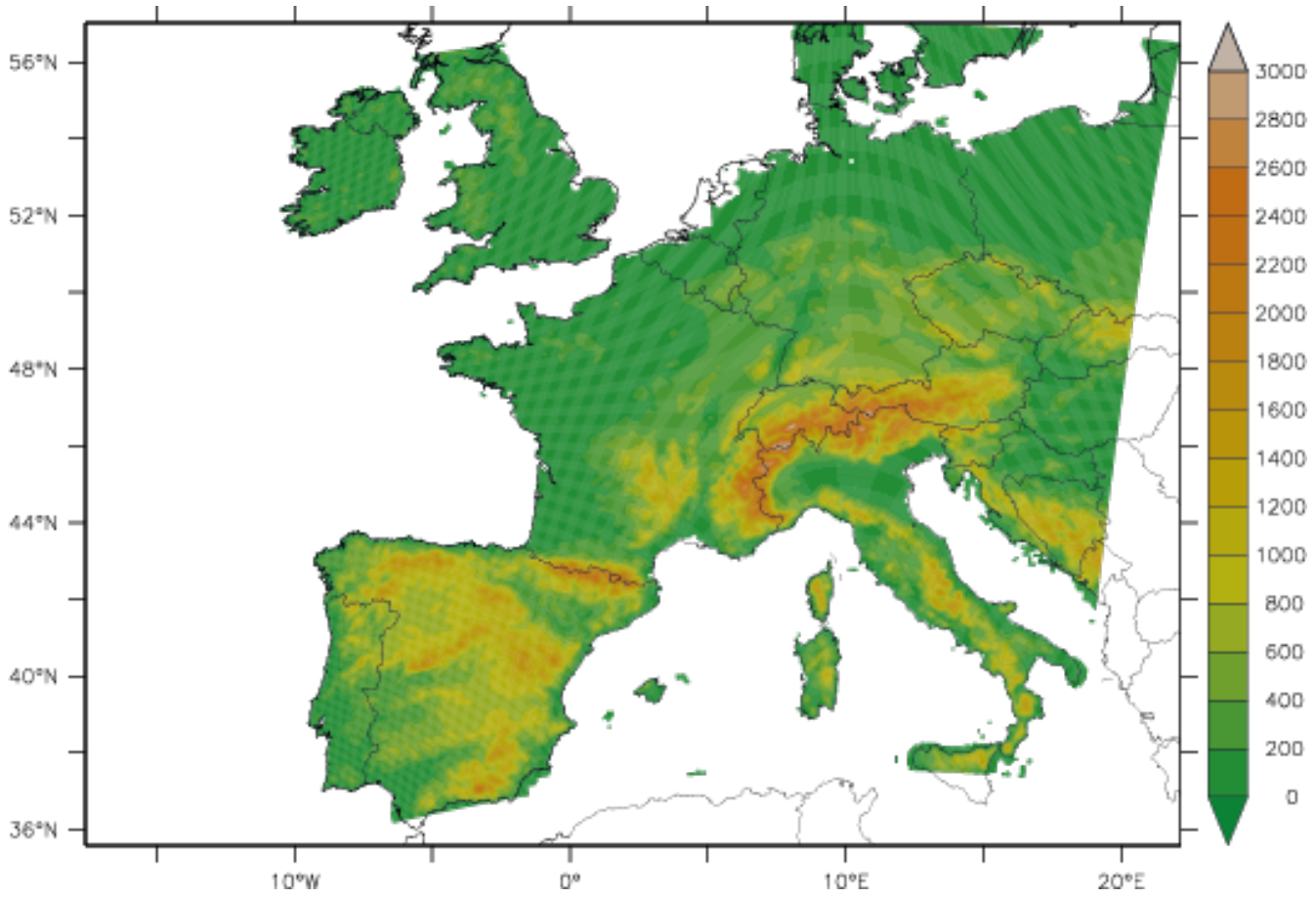
In response, we added a sentence to this effect to the results section. It reads: "We computed this mean using data from all times of the day in order to fully reflect the annual mean."

18. Line 333, more explanation needed on "suggesting a somewhat limited efficiency of atmospheric transport and mixing to disperse the signal laterally". More explanation needed.

Authors: What was meant was that the emissions tend to get trapped near their sources, i.e., that the transport is not very effective in mixing these signals aloft and in lateral directions. In order to reflect this, we reformulated the sentence to "...suggesting a somewhat limited effectiveness of atmospheric transport and mixing to disperse the signal aloft and in lateral directions".

19. Line 334, a map of terrain for the region of interest will be very helpful.

Authors: We will add the following topographic map of the model to the supplementary section of the paper.



20. Line 360, "while the biospheric signal is stronger". Biospheric signal can be positive and negative, which will cause completely opposite effect on the total CO2. Please clarify it.

Authors: We reformulated these sentences to read "At the same time, the biospheric signal changes sign in the south and becomes positive. This compensates for the smaller fossil fuel signal there and results in a relatively uniform spatial pattern of atmospheric CO_2 across Europe". We hope this clarifies this.

21. Line 369, the column averaged values are smaller than the surface ones, mainly because the signals are at surface and they are diluted in the column as averaged out. The statement is incorrect.

Authors: The reviewer is correct that the dilution of the surface signals with the air aloft leads to the lower concentration. Our statement does not contradict this at all. It rather emphasizes why the concentrations aloft are smaller. One element that we had not mentioned before is the role of the lateral boundary

conditions, whose role is more important aloft than at the surface. We thus revised our statement accordingly. It now reads:

"An additional reason is a much stronger influence of the lateral boundary conditions, which result in a dilution of the fossil fuel components."

22. Line 400, what does lateral gradient mean specifically here?

Authors: It refers to the spatial gradient in horizontal direction. We believe that the use of the expression "lateral" is accurate and we thus decided to keep it.

23. Line 419-428, Figure 8 and the relevant statement doesn't make sense to me. I don't understand that why the contribution of each component (b-d) would be larger than the total (a).

Authors: This - at first surprising - effect can arise as a result of potentially compensating effects between the different components. This happens when the different components are anti-correlated. To clarify this, we added a short statement to the text. It reads:

"To determine the contribution, we then computed the fractional variance of each component relative to the total variance. Since the different temporal components can compensate for each other, the sum of the fractional variance can actually exceed unity."

24. Line 487-555, the authors made misleading statement regarding the relationship between meteorological matric (such as PBL, etc.) and fossil fuel emission. Fossil fuel emission is strongly correlated to the anthropogenic activities. The increase and decrease of the fossil fuel emission is not affected by transport and/or mixing; the atmospheric co2 concentration is. At the beginning, I thought it ("emission") is a typo, which is an easy fix. However, I realized that this relationship is not clear at all to the authors when I saw Figure 9b.

Authors: It appears as if we confused the reviewer. We are, of course, not implying a causation between atmospheric transport and emissions. Rather we are implying that their correlation causes a net signal in atmospheric CO_2 in the presence of a net zero flux. This phenomenon, coined rectification effect by Denning et al., (1995) has been thoroughly discussed in the literature. Most of the literature was concerned about the terrestrial rectification, arising on diurnal and seasonal timescales from the correlation between atmospheric

(vertical) mixing and the net exchange fluxes of the terrestrial biosphere, but more recently, this term has also been used for fossil fuels (e.g. Zhang et al., 2016). We re-read what we have written, and could not identify where we went wrong. Thus, no changes were made to the text.

25. Line 670, the authors selected three location to do further analysis for Figure 15. Why these three? Please explain.

Authors: The exact locations of these three locations were chosen somewhat arbitrarily, but were identified on the basis of us seeking examples of the three different cases of interest. (i) The changes in the average concentration are larger than those of the standard deviation, (ii) the changes in the average are of similar magnitude as those of the standard deviation, and (iii) the changes in the average are smaller. We clarified this in the text.

26. Line 700, yes, the authors were being optimistic as they admitted. All of the analyses were based on the model results. The cloud contamination was considered in the discussion. However, the more important errors are very likely caused by model transport errors and error estimate of biogenic flux. It will be great if the author can expend the discussion on the impact of those error on their findings.

Authors: We absolutely agree. In response we added the following two sentences to the text "We assumed here also "perfect transport", i.e., no errors in how the emission reductions manifest themselves in a change in the concentration field. In fact, errors in this transport are, perhaps, next to the lack of observations that largest impediment to detect changes in fossil fuel emissions."

27. Line 724, where does "110%" come from? Is it a new? The authors shouldn't include any new results in the conclusions.

Authors: Thanks for pointing this out. While this result is actually not new, we nevertheless deleted it.

28. Figure 1, please put the labels on the color bars.

Authors: *added as requested.*

29. Figure 2, I would like to see the time series of the scaled emission components and the total emission. (plot total scaling factors)

Authors: See our reply to point 6 above.

30. Figure 4, 1) enlarge the font size. 2) what year is it? 3) night time model results included? If so, 10 m above ground is too late. Models are struggled at nighttime, especially for such low level. 4) Keep the color bars in the scale and same range for a and c, b and d.

1) enlarge the font size.
Authors: changed
2) what year is it?
Authors: The whole paper is based on simulations from one year, as stated in the method part, i.e., from end of March 2008 till March 2009
3) night time model results included?
Authors: Yes.
4) Keep the color bars in the scale and same range for a and c, b and d.
Authors: We use the same scale for a and c, b and d, and we keep the same range for all the 4 figures.

31. Figure 5, the figure labels (a, b, c, d) are missing.

Authors: Added to the figure.

32. Figure 6, use the same color scale as Figure 4 does.

Authors: As Figure 6 shows a different quantity than Figure 4 (column averaged XCO_2 instead of the surface mixing ratio), and a quantity whose spatial gradients are much smaller, we decided to the keep range. This advantage clearly outweighs the benefit being able to directly compare Figures 4 and 6.

33. Figure 8 doesn't make sense to me as I pointed out before. The contribution of each component can't larger than the total.

Authors: Please see our explanation above.

34. Figure 9a, which minus which? Figure 9b doesn't make sense to me. The fossil fuel emission is related to anthropogenic activities; it won't be affected by transport and/or mixing. If it is concentration instead of emission. The correlation of the concentration and PBL height should be negative. It's confusing.

Authors: Please see our explanations above. We are indeed concerned here about the temporal correlation between fossil emissions and vertical mixing/transport. But please recall that a correlation does not imply a causation. We clarified in the caption the sign of the signal, i.e., that the plot is showing the difference between the time varying and the time constant emission case, i.e., "(time varying minus time constant)". Figure 9b is correlation between the emission and PBL height. This figure just helps us to explain the map in Figure 9 a.

35. Figure 11, what countries are they exactly included in SW?

Authors: Portugal and Spain. We clarified this in the caption.

36. Table 1, it's a confusing statement that "the 3 hourly means between 12 – 18 PM"

Authors: It is indeed confusing. We thus changed "the 3 hourly means between 12 – 18 PM" changed into "the mean between 12 and 18 PM based on the 3 hourly output of the model."

37. Figure 13, enlarge the font size. Does it in percentage? Can the authors overlay the wind field on top?

Authors: We changed the font size. Yes, the numbers refer to percentage, as we had stated in the caption "relative contribution". As the figures contain already too much information, we decided against the overlaying of the wind field.

38. Figure 14, it's not clear to me to look at the standard deviation changes between two cases.

Authors: The idea here is analyze whether changes in the variability of atmospheric CO_2 can be used to detect changes in fossil fuel emissions in addition to changes in the mean concentration. However, Figure 14 suggests that this method might be not so promising when using the column averaged CO_2.

39. Figure 15, I can only find three green dots in Figure 14b, but Figure 15b shows four lines. Please make correction accordingly.

Authors: Thanks for spotting this. The two dots (corresponding to blue and cyan line) were overlaying each other due to their short distance. We changed this in the figure by using opacity.

References:

D. Bozhinova, M. K. Van Der Molen, I. R. van der Velde, M. C. Krol, S. van der Laan, H. A. J. Meijer, and W. Peters. Simulating the integrated summertime Δ14CO2 signature from anthro- pogenic emissions over Western Europe. Atmos. Chem. Phys., 14(14):7273–7290, 2014

Houweling, S., Aben, I., Breon, F.-M., Chevallier, F., Deutscher, N., Engelen, R., Gerbig, C., Griffith, D., Hungershoefer, K., Macatangay, R., Marshall, J., Notholt, J., Peters, W., and Serrar, S.: The importance of transport model uncertainties for the estimation of CO2 sources and sinks using satellite measurements, Atmos. Chem. Phys., 10, 9981-9992, doi:10.5194/acp-10-9981-2010, 2010.

F. R. Vogel, B. Thiruchittampalam, J. Theloke, R. Kretschmer, C. Gerbig, S. Hammer, and I. Levin. Can we evaluate a fine-grained emission model using high-resolution atmospheric transport modelling and regional fossil fuel CO2 observations? Tellus B, 65:1–16, 2013. doi: 10.3402/tellusb.v65i0.18681.