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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The CO₂ release and Oxygen uptake from Fossil Fuel Emission Estimate (COFFEE) dataset: effects from varying oxidative ratios" *by* J. Steinbach et al.

Anonymous Referee #2

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In this paper, the authors broadly address the question "What is the impact of the (changing) mix of combusted fuels on the amount of oxygen in the atmosphere?"

They begin by creating the spatially and temporally resolved "COFFEE" dataset. This is done by combining information on usage of various fuels by type with the oxidative ratios for each of these fuel types. With the dataset in hand, they use two atmospheric transport models (TM3 and REMO) to compare their results at specific locations with the widely used assumption of a temporally and spatially uniform oxidative ratio. They conclude by using the TM3 model to conduct an inversion study using both real and fictitious datasets to determine whether inferred ocean fluxes would be significantly

more accurate if the COFFEE values for oxidative ratios were used.

Overall, I find the paper a well written, clearly organized, and thorough investigation of a set of questions that are definitely worthy of attention. The authors have created a useful dataset, made it publicly available, and taken valuable first steps in using it to answer scientifically interesting questions. The paper is appropriate in scope and content for ACP and I believe it should be published after some relatively minor revisions. I would like to see my concerns below addressed.

Substantive questions/comments:

P6190 L24 and later: The term "categories" is general enough that when it reappears later, it becomes confusing. Please adopt a more descriptive term such as "usage types".

P6191 L7-16: As written, I found this paragraph confusing. Please spell out more clearly exactly how the UN data connect to the EDGAR data. If my understanding is correct, it should read more like "While the EDGAR data break down fossil fuel consumption by usage type, the oxidative ratios we seek are determined by fuel type. Therefore, we use the energy statistics compiled by the United Nations Statistics Division (UN, 2009) to connect fuel type with usage type. This dataset, currently available..." Note my replacement of "category" with "usage type" (see comment above).

P6192 L18-19 Two questions here: First, how were "unrealistically large variations" identified? Did you have any quantitative criteria? Secondly, you say that you are using only the data from 1996 onward. As I understand it, this is exclusively data from the extension period. Are these data as rigorously vetted as the original EDGAR dataset? They appear to be based on a personal communication from 2004. Is there no better or more current citation?

P6193 L1 Were seasonal cycles *completely* suppressed? For which usage types? Please be more specific. What evidence do you have to support your choices (i.e.

what constitutes "reasonable")?

Section 3.2: As far as I can tell from this description, the variation of OR for the entire planet on timescales of a year or less is given by the variation in the Netherlands (with a phase difference of π in the Southern Hemisphere). Furthermore, the seasonal usage patterns had been reported as square waves and these were smoothed out with a mathematically (rather than mechanistically) based algorithm.

These are all reasonable assumptions, and given the limitations of what is available, I cannot necessarily come up with a better alternative. Nonetheless, they are fairly substantial assumptions and I would like to see an argument justifying the extensive discussion of the sub-annual variability of OR that is present in this paper, in light of this (at least apparently) tenuous foundation.

P6196 L6-24 My feeling is that these two paragraphs are overemphasized. I suppose it is good to check that the BP and CDIAC datasets are consistent with EDGAR. However, as you yourselves point out, these datasets are hardly independent. Given the weakness of this comparison, I'd suggest simply a line or two saying something like "As a consistency check, we have confirmed that the oxidative ratios that result from applying our method to the British Petroleum (British Petroleum, 2009) and CDIAC (Boden et al., 2009) datasets are not significantly different from those based on EDGAR." Anything more is excessive.

P6198 L17 Please be a bit more specific about how the 5-day timescale was chosen, and how sensitive the results are to this choice.

P6198 L19-20 When I look at the plots, it actually does look to me like the black trace is consistently a little higher than the blue. Please address this.

P6199 L11 Strictly speaking, you don't know that OR_{ffp} is changing at Hateruma; you only know that OR_p is changing, and you use other lines of evidence (such as back-trajectory analysis) to attribute this to fossil fuels.

P6199 L23ff The authors make a fairly compelling case that for somewhere like Hateruma, a high-resolution regional model such as REMO is more appropriate. However, REMO's domain didn't include Hateruma. Therefore, it would have been more informative and convincing if we could see results for another site with strong local influences that is within the REMO domain (perhaps Ochsenkopf or Jungfraujoch) or maybe an adjustment of the REMO domain to include Hateruma (although this might be prohibitively difficult).

P6200 L1-12: I would like to see a more focused discussion of the months of November and December. This is the time with well constrained observations and (in early November), a noticeable disagreement between the model and data. What do the authors think is going on here? It would also be good if the authors could spell out how much better things would be at Hateruma if they were to use a constant, but locally correct value for OR_{ff} (instead of the globally appropriate 1.4).

P6201 L15: What about Kumukahi? This shows variations in OR comparable to ALT and BRW but is not discussed at all.

P6204 L6-8 I am probably simply failing to understand something here, but I am not entirely convinced by figure 7b and 7c. The authors use Figure 7b to claim that there is an increase in the interannual variability of APO fluxes when a variable OR_{ff} is used. What puzzles me is that the black and red curves in Figure 7c show very similar amounts of variability (at least to my eye). If 7b really does show the variability in APO fluxes that result from introducing variable OR_{ff}, and values around Hateruma are positive, shouldn't Figure 7c reflect this increased variability?

P6207 L23: As discussed earlier in my review, because there really isn't much independence of the BP, CDIAC and EDGAR datasets, I'm not sure the agreement of these results is worth highlighting in the conclusions.

Suggested text edits:

P6188 L20-21 should read "Photosythesis **in** the terrestrial..."

P6188 L22 should read "Only **ocean-related** changes..."

P6188 L23 should read "allowing **use of** measurements..."

P6189 L2 and throughout the paper: The authors move freely back and forth between OR and α. I know that both are used in the literature, but within a single paper, there should be a single choice. Please pick one and stick with it.

P6189 L11 "...is used:" By whom? Presumably this is the one you will use, so say instead "...therefore we adopt the following simplified formula:"

P6189 L22 should read "have already been observed. In the Netherlands..."

P6189 L27 should read "Japan $(OR_{ff} \sim 1.37)$, with.."

P6190 L1 Who has calculated the OR_{ff} values for China, Korea and Japan? As stated, it's not at all clear whether these values are calculated by Tohjima Yamagishi, the authors of this paper, or Boden *et al.*

P6190 L25 I'm pretty sure this should read "until the year **1995**, with **an** extension **for** 1996-2001 (calculated..."

P6191 L23 should read "is assumed. Therefore, the resulting..."

P6192 L6 should read "and gas the **aformentioned oxidative** ratios..."

P6192 L7-8 should read "..was taken. This value was determined by applying Eq. (1) to each of the biofuels present in the dataset and weighting their contributions by amount used.

P6192 L14 should read "ratios were interpolated from..."

P6192 L23 The Brenkert citation is not in the bibliography.

P6193 L8 The acronym "COFFEE" is a charming one, but it's actually a little bit decep-

tive, since the dataset includes biomass burning (not a *fossil* fuel). While I don't expect you to abandon your COFFEE acronym, you should at least point out this bit of artistic license.

P6193 L21 should read "representative **of** the fuel..."

P6194 L13 should read "over three latitudinal bands: Extratropical Northern Hemisphere (NH $+20^\circ$ to $+90^\circ$), the Tropics (TR, -20° to $+20^\circ$) and the Extratropical Southern Hemisphere (SH -90° to -20°). The upper row..."

P6194 L22 should read "this effect is more pronounced in the **high latitudes** than in the tropics (18%), **as expected**. Seasonal..."

P6195 L9 should read "at night **should** not be..."

P6196 L2-5 should read "in the year 2005. This increase is caused by strong increases in emissions from countries which rely primarily on coal such as China."

P6196 L9 Spell out "BP" both in the text, and in the citation, since it appears as "British Petroleum" in the bibliography.

P6197 L5 should read "$PgCyr^{-1}$ This is consistent with Manning and Keeling's (2006) statement that..."

P6198 L22 should read "the difference **between** the fossil..."

P6198 L23 should read "from COFFEE **and the O2 signal calculated using** a constant..."

P6201 L12 should read "average are **generally** larger for. . ."

P6201 L27 should read "is used, the fossil-fuel-related atmospheric signals. . ."

P6202 L17: Please give a citation as an example where you say "usually accounted for"

P6203 L5 should read "1992), Princeton University (Bender. . ."

P6204 L7 should read “in the Hateruma region **(7b). In addition**, most of the other. . .”

P6204 L11 should read “To assess the significance of these effects, we. . .”

P6204 L16 should read “ “standard run” **to** an ensemble. . .”

P6204 L19 italicize “a priori”

P6204 L25 should read “area are **comparable to** the uncertainty. . .”

P6205 L7 should read “will probably include data from. . .”

P6206 L25 The 2nd clause here (reading “as the results for all monitoring. . .”) is confusing. I *think* it should read “as the results from the “all monitoring stations” run do not show any significant differences.” However, this leads to the question: no significant differences from what? From the “inversion stations” run? Or from the constant OR_{ff} run? My guess is the former, but the prose should be more explicit.

P6207 L20: In what sense is this “average”? Is it a spatial average, a temporal one, or both? Please be explicit.

P6208 L14 should read “emission sources depends on the individual. . .”

P6208 L15 should read “Winter), as the size of the. . .”

P6208 L23 should read “An exception is the station. . .”

P6208 L27 should read “might also play a significant role at other European. . .”

P6209 L8-9 should read “the possibility **that** variations in OR_{ff} **could** be mistaken. . .”

P6209 L 21 should read “inversion. Thus, we recommend taking into account. . .”

P6209 L23 should read “variable oxidative ratios even more important.”

Figure 3b: Somewhere (either in the legend or the figure caption), there should be an explanation of the “BP” and “CDIAC” curves.

Figure 6: It may be clearer if you were to make the map Fig. 6a and then move the other two to the right side of the panel and relabel as 6b and 6c. Also, line 7 of the caption should read “REMO is used when available. . .” Finally, why are the whiskers in 6a solid lines, while those in 6b are dashed?

Figure 7: Line 6 of the caption should read “using the station set from. . .”. Also, the axis labels in 7b and 7e should have Δ instead of d.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 6187, 2011.