**Atmospheric
Chemistry
and Physics
Discussions**

Interactive
Comment

Interactive comment on "Analysis of $\triangle O_2/\triangle CO_2$ ratios for the pollution events observed at Hateruma Island, Japan" *by* C. Minejima et al.

Anonymous Referee #2

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The discussion paper by Mijima *et al.* ("Analysis of O2/CO2 ratios for the Pollution events observed at Hateruma Island, Japan") presents results from a study on O2/CO2 ratios of several synoptic events based on continuous O2 and CO2 measurement at Hateruma Island, Japan. O2 and CO2 concentrations are anti-correlated in the process of biomass burning, with different fuels resulting in different ratios between the two gas species. The O2/CO2 molar ratios for main fuel types, i.e. coal, gas and liquid fuels, are well-known which allows attributing differences in the ratios to the certain types of the fossil fuels burned. The authors present the continuous records of O2 and CO2 concentrations from Oct 2006 to Dec 2008 and calculate correlation plots for 67 pollution events. Using back trajectory analysis (METEX) they classify pollution areas into three main regions of origin: China, Japan/Korea, and other. The observed ratios are then compared to those obtained from the national fossil fuel inventories from the

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Carbon Dioxide Information Analysis Center (CDIAC). In addition, the authors use two different modelling approaches incorporating regional and global models (FLEXPART and the Coupled model) to compare the simulated ratios with the observational values. The results and discussion are structured and organised around the findings from the comparison results and discuss possible reasons for discrepancies.

From the technical perspective, the authors have a good continuous record for both O2 and CO2, with only a couple of gaps in the data collection over a two-year period. This is a good achievement by itself owing to the difficulties associated with O2 measurements. The reported O2 precision is typical for this measurement technique, whereas the CO2 precision could be potentially improved. However, the latter is not relevant to the present study. The authors report high flowrate at 9 L/min. I'd like to know whether they have any Tee-junctions in the system to reduce the flowrate prior to sending it to analysers, and if so, whether they observed or evaluated fractionation effects associated with such setups?

I have two main concerns about the study.

First is the method itself. Although, it is clearly a nice exercise to perform at a site where concurrent O2 and CO2 measurements are made, it can hardly be used on a larger scale, e.g. for proper verification of the countries' emissions. It is technically challenging to set up and maintain good quality O2 measurements, and there are only few of such sites/labs available worldwide. So it is unlikely that we will see a big increase in such sites in the near future. In addition, as the authors report themselves, the method is not entirely objective. For example, the beginning and the end of the pollution events were decided on as a result of visual inspection, which can vary when performed by different people/groups. Did you evaluate errors associated with this approach? The pollution events with r2 less than 0.8 were discarded and the rest were accepted. Why was this particular value chosen as such an important criterion?

40% of pollution events were discarded as they didn't meet the criterion above, and

67 events were analysed in total. This does look like a lot of manual work. Based on this I doubt that this approach will be widely used for the purpose proposed in the manuscript. As air masses tend to show disregard for the political and economical borders it would be difficult to separate the influence of fossil fuel sources in different regions/countries. This is in fact one of the finding of the study as the authors suggest that the simulated ratios for Japan/Korea tend to me underestimated (compared to those observed) because of the large influence of coal-dominated emissions from China. Thus it is practically impossible to use a single (or even several) station approach for such applications. If one looks at the model simulations (e.g., back trajectories) then there are lots of other uncertainties introduced into this analysis. However, this might be a more valid approach when applied more locally to minimise uncertainties associated with air mass transport.

The second issue with this manuscript is the presentation of the material, especially the Abstract and Introduction. The Abstract could be shortened and re-written in a way to highlight the main findings of the study and paying less attention to detail. At the moment those findings are buried somewhere in the results section. Although they are mentioned in the Abstract they do not attract attention as such. I think it is an important finding that the observed ratios are in such a good agreement with the ratios obtained from the fossil fuel inventories. The accuracy of such inventories will be questioned even more often in the future, and it is useful to have an independent tool to verify that. The other finding is a good agreement between the regional and global model simulations with respect to the O_2/CO_2 ratios, and particularly the fact that the simulated values for Japan/Korea were significantly underestimated compared to the observed ratios and those based on fossil fuel inventories. This brings us back to the problem that I've mentioned above about difficulties on implementing this method on a larger scale. As the authors point out the influence of the emissions from China most likely is the reason for the underestimated model results for the Japan/Korea region. It is also not possible to attribute those results to either Japan or Korea using the data only from this location.

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The Introduction is also not particularly exciting as it stands now. What are you planning to achieve with your study? As you mention other studies on the subject what would make yours different, and why do you think it will give us a fuller picture of the studied area? Do you think that the approach is a viable tool to verify the composition of countries' industrial emissions? If not, then is there room for improvement or practical solution for that?

I would certainly recommend the manuscript for the publication as long as the authors address at least some of the points above as this will make the manuscript to stand out on its own much more. The language is mostly good throughout the manuscript, although I would recommend making an effort to shorten and clarify some statements and sentences because some of them are too long and thus difficult to follow. This applies in particular to the modelling sections of the manuscript. There are also occasional grammatical mistakes which are easy to correct if ones re-read the manuscript carefully. The graphics are of good quality, with comprehensive captions.

Minor points:

Abstract:

- CDIAC, please write in full here as this is the first time you mention this abbreviation in the text.

- When talking about agreements between different ratios I would suggest giving the ratios here rather than just writing about them.

Introduction:

Page 15633, line 12: remove "changing";

Page 15633, line 20: remove "varying", change to "ratios";

Page 15634, line 8: Sentence starting with "In addition" is too long. I'd suggest breaking into a couple of separate sentences.

Page 15635, line 9: 4.8 per meg of O2. Remove "the mole fraction of" as you repeat yourself here.

Page 15635, line 22: Replace "relatively long-term" with seasonal.

You have a discrepancy when defining the length of your trajectories. Sometimes they are described as 5-day and sometimes as 8-day trajectories.

Results and Discussion:

Page 15639, line 5: for both regions, not events. Suggest saying "the ratios for China"instead of "the range for China" and "the ratios for Japan/Korea" instead of "the latter". You might consider re-writing the sentence as it is a bit awkward.

Page 15639, line 7: Therefore doesn't make sense here.

What are the other countries whose emission influences you detect at your station in the O2/CO2 ratios? How do you explain the fact that the average ratio for those countries derived from the inventories is higher than the observed and simulated ratios (although the uncertainties are close to overlapping).

Page 15640, line 11: the stochiometry of O2 and CO2 for specific processes is usually fixed in the models (in fact, r2 should probably be close to 1.0) so it would be expected to see a much higher correlation for simulated ratios compared to those based on observations.

Fig 6: mention the blue line in the caption.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 15631, 2011.