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## ***Interactive comment on “Temporal changes in the emissions of CH<sub>4</sub> and CO from China estimated from CH<sub>4</sub> / CO<sub>2</sub> and CO / CO<sub>2</sub> correlations observed at Hateruma Island” by Y. Tohjima et al.***

### **Anonymous Referee #2**

Received and published: 29 October 2013

This paper analyse high-frequency CO<sub>2</sub>, CH<sub>4</sub> and CO measurements from the Hateruma site, located on the south-eastern side of mainland China and the two Ko-reas. They have applied reduced major axis regression analysis on the observed and simulated synoptic scale variations. Proven measurements and modelling tool have been used throughout the manuscript. The article is generally well written. However, I have reservations on some of their interpretations as listed below. The manuscript can be accepted for publication after a major revision.

Specific comments: \_\_\_\_\_

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Abstract. Please reorganise a bit. One suggestion is to spell out the use of FLEXPART or other modelling tools in one sentence before the sentence starting with "Although the..." in line#6. Also is this sentence alright? in the first part you say 'gradual decrease' but later identify two 'no trend' periods. May be using shorter sentences would improve clarity.

p.22894, l.25 : third most, after H2O and CO2

p.22895, l. 4 : "Recent" systematic ...

p.22895, l.10 : What about other losses, such as, soil absorption, Cl and O1D in the stratosphere?

p.22895, l.16 : just 'species' or 'gases' instead of 'greenhouse gases' to make it more general

p.22895, l.18 : does CO always acts as a precursor of tropospheric ozone production? then '..., CO could ...' may be changed to '..., CO acts ...'

p.22895, l.22 : ... and 'air' pollutants...

p.22895, l.25 : may be cite REAS (Ohara et al., 2013?) / EDGAR4.2 / CDAIC here

p.22896, l. 2 : I thought the CH4 emission increase is from coal mining too. Could you be more specific (also commented again later)

p.22896, l. 5 : any reference to support this 'consideration'?

p.22896, l.10-13 : join these two sentences, e.g., "In the downwind from source regions, synoptic-scale variation (SSV) in ..."

p.22896, l.15 : delete 'known'

p.22897, l. 8 : any specific reason for using hourly data here or daily data in the previous work?

p.22897, l.19 : suggest to modify as "only a brief description"

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p.22900, l. 22-24 : I tend to believe the CO/CH<sub>4</sub> seasonality comes from changes in winds between the winter (north-westerly, isentropic) and summer (south-easterly, convective), and may also contributed to some extent by the significantly faster chemistry of CO in the summer. (I find later you have addressed this in p.22903, thus this statement needs some modification).

The difference in the phase and amplitudes of CH<sub>4</sub>/CO<sub>2</sub>, CO/CO<sub>2</sub> and CO/CH<sub>4</sub> may also indicative of differences in spatial patterns of CO<sub>2</sub>, CH<sub>4</sub> and CO emissions. Could you make any comment on the spatial pattern aspects, which is not addressed amply.

p.22901, l. 8 : Please clarify whether these five months are same as the period used for Tohjima et al. (2010) analysis or NOT

p.22903, l. 8 : I think, the degree of similarity of the emissions is not very clear. Yes, they are roughly similar. It would have been great to have some pattern analysis here, although this is not an essential demand.

p.22904, l.19-20 : I am not sure. The narrowing of the histogram can also happen due to the more colocation of emissions. Because more and more population is becoming urban, particularly in the past decade, in China. Could you confirm this by analysing the emissions maps used during the corresponding years for panels a,c,e and panels b,d,f, respectively.

p.22906, l. 2 : Why the CO/CH<sub>4</sub> emission ratios are constant with time? Although CH<sub>4</sub> emissions are relatively constant in 2000s, I thought the EDGAR CO emissions have increased significantly in the same period.

p.22906, l.13 : I had an impression that FLEXPART is run using multi-year winds. Please clarify why you say "no IAV in the meteorological fields"

p.22907, l. 7 : I could not follow this argument very well. Do you mean that this method can robustly estimate "Emissions from Chinese part of the EFA". To support the existing sentence, please show the emissions for the region "EFA outside China".

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p.22907, l.16 : Do you mean "black open squares". If correct, change Fig. 7 to '7c' for better clarity.

p.22908, l.15 : This conclusion also came out of the TransCom-CH4 experiment (ACP, 2011). Please check.

p.22908, l.16 : Please do a sectoral emission analysis to conclude this or to be specific which sectors are contributing to this increase, e.g., coal burning, coal mining, etc.

p.22909, l.17 : I do not know the validity of this conclusions. The difference between EDGAR and all other emission estimated could arise from content of the values themselves. I think, EDGAR is reporting only the primary CO emissions, and what yours (and most likely others) estimated is the net CO release from primary emissions as well as oxidation products (from CH4, VOCs etc.).

If you agree, some part of the discussions in this sub-section needs major revision, for the comparisons of CO emission strengths and interannual variations.

Figure 1.: Either add USA to all panels and change the legend accordingly. I recommend remove USA line from Fig. 1a as your point is well stated in the introduction already.

Figure 4c: Y-axis units : ppb/ppb?

Figure 5. caption, last line : footprint (one word)

Figure 5. caption, first line : slop'e's

Figure 7c: What are these open squares? Not defined here, and also I could not find explanations in the main text.

Why not use the same colour for 'Obs.' in all the panels?

Figure 9.: REAS - 'S' not 'D'