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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "^{14}C observations of atmospheric CO_2 at Anmyeondo GAW station, Korea: Implications for fossil fuel CO_2 and emission ratios" *by* Haeyoung Lee et al.

Anonymous Referee #2

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This is a well-written paper with interesting data concerning atmospheric observations and validations of fossil CO2, and CO and SF6 emissions from Korea, and the Asian main land. Upon reading, I have made notes and comments that I present below. A more general remark (also given below) is that I invite/encourage the authors to more explicitly conclude what their observations show concerning the quality of the inventories, and if these inventories are thrusworthy or not. By drawing conclusions in that style, these data will be more accessible and valuable to policy makers, and might help to improve the inventories.

I recommend publication after the authors have dealt with my comments below.

Comments ACP-2020-122

page 5, lines 108-109. at what flow rate are the flasks filled, or rather: is the flask air composition an average over some period of time, or merely a point in time?

line 107 "Two pairs of flask-air samples (4 flasks total," In tabel S1 I see only one value per week. Is this an average? Flasks taken together for 14C mm?

line 129 "suggested" ??

line 245, 246 This largest positive Cbio is actually a single point. Which trajectory belongs to this point? The tic marks in the histogram of fig 2C do not correspond to those in the left part of fig 2C.

Lines 252-254. I doubt the explanation offered. Even though your sampling time is early afternoon, I expect still the average mixing height to be the most important player in the mixing ratios of Cff and Cbio, as it influences the flux-to-mixing ratios relation. There must be a seasonal effect in the mixing height no doubt. This must be taken into account in this dicussion. The fact that Cbio covaries with Cff also points to the importance of the (average) mixing height.

Line 270-271 I would say in general nobody expects the CO2 enhancements above background to be entirely due to Cff.

Line 280-282 "During the experimental period, the averages from Asian continent (sectors CE and CN) were higher than KL without the baseline level."

Without the baseline level?? What do you mean?

282 Does OB fit in this set? You call it ocean background, but at the same time you mention it crosses over Shanghai (213-214). In line 234-235 you indicate it again as being background, and then here (282) you take it along with the "real" continental trajectories. This needs to be clarified.

298-299 "we also see CN originated from northeast China and it was around

(10.6±6.9) μmol mol–1." I don't get the meaning or consequence of this sentence part.

Lines 300-302 Once more, I think average mixing height is the key player here. Are the weather patterns, and thus mixing heights different in the years 2009-2010 from 2014-2106? Did Turnbull et all also sample between 14 and 16 hours?

304 increase of 16.7% line306 "broadly consistent" I disagree for the China case, as you find way larger increases between 2010 and 2016. So you might hypothesize that your measurements indicate much higher increases in fossil fuel use? Of course what you state in 311-312 is very true...

Line 321 are these differences significant? I would say (KL,PL) > (CN,CE) > (CB,OB)

Line 331 To my opinion SF6 is not a good tracer/surrogate for fossil fuel CO2, as it is not produced in the same process. So SF6 actually traces specific industrial activities, and electricity use. Both are coupled to fossil fuel CO2, but not in a 1:1 (spatial,temporal) relation. CO, on the other hand is really co-produced with fossil fuel CO2 (and with biofuel CO2), albeit at a varying rate.

Line 332 "Even though" I don't see the contrast between the strong correlation and the differences

Caption figure S3: "From 2005,.. " -> "From 2005 onwards, .."

lines 347-349 Still, in spite of the still large uncertainty, I invite you to make a stronger statement here, namely that the SF6 inventory in EDGAR and in KNIR are too low given your measurements.

Line 351 also here, watch the significance. I would conclude from table 1 that CB=KL And indeed (see my point higher up), OB is mostly regional background air.

354 "CO...it is more closely related to fossil fuel CO2 emissions" yes, but also to bio-material combustion (compare the Cff to the CO excess)

357-358 I think you can safely erase the word "likely" here. 358 add "and the use of catalysers" ?

358-360 Indeed, biomaterial combustion must play a role, regarding the low Cff especially for CB.

366 Figure S2 -> Figure S1

369 Paragraph 3.4 I suppose you did a similar thing for the SF6 inventories. That means the either the start of this paragraph should be moved up into 3.3, or the SF6 inventory discussions should be taken form 3.3 and moved to this paragraph.

377 "The uncertainty of EDGAR4.3.2 emissions" -> "The uncertainty of EDGAR4.3.2 fossil fuel CO2 emissions"

397-399 if a difference is not significant, it is doubtful to discuss its possible causes.

403 "KNIR seems to have uncounted CO emissions," -> "KNIR suffers from a high number of missing CO emission sources," in other words: make this statement stronger, as the difference is huge: ≈2500 vs ≈700 Gg in 2012 ! And your data corroborate the Edgar emission ratios...

433-439 S. Korea: your RCO results are 1.2 times the Edgar results. That is hard to see in figure 4. Your value (from table 1) is 8±2, so a ±25% uncertainty, which makes this factor 1.2 not significant. The Chinese inventories, on the other hand, ARE significantly too low, even though the declining trend has been confirmed by atmospheric measurements. My guess would be that the lack of biofuels/biomaterial burning which is not present in the EDGAR CO inventory, explains the large difference in China, and is not so important in S. Korea.

441 (and also earlier and further) you express mean values ± standard deviations, whereas the way you write it suggests that this is the error in the mean value, which is in fact sqrt(#mm) lower. So in fact the mean value here is (-6.2±2.2) ‰ (I took N=70), with a spread of 19 ‰ In your case, most of the time the spread= the standard deviation

is the important feature, but if you compare in lines 446-447 to previous measurements at TAP it is important to know how many measurements those were, and thus what the mean and error in the mean are. Your statement: the average is twice as high strongly suggests that this difference is significant, but the reader can only judge that if you present the error in the mean in both cases. I advise to make this difference between standard deviation and error in the mean clear at the various points where it matters in the paper.

Lines 449-452 Yes, Cff really increased for the air masses from the Asian mainland. Do you conclude that this indicates stronger growth of fossil fuel use than the statistics say? If you think your data clearly point at that, mention that here.

lines 453-463 Based on your data I would (also) conclude the following: (1) 14C analysis is a reliable way of determining Cff in the mixing ratio of air masses (2) Then, the ratio of the emission of rare trace gases and Cff can be determined as well (3) As the inventories for various other trace gases/greenhouse gases are generally much less reliable than that of Cff, these inventories can be validated/verified using atmospheric measurements like ours. (4) I our case we conclude that the inventories for SF6 ... and for CO ...

In this way your results will probably be more valuable to policy makers.

I would also formulate (part of) this reasoning in the abstract.

Two more references suggested:

Page 2 I would suggest in addition the reference : van der Laan, S. et al. Observation-based estimates of fossil fuel-derived CO2 emissions in the Netherlands using Delta 14C, CO and 222Radon, Tellus B, 62(5, SI), 389–402, doi:10.1111/j.1600-0889.2010.00493.x, 2010.

page 3 line 64 "..correlate well..." I think the earliest 14C-based reference to this is Zondervan, A. and Meijer, H. A. J.: Isotopic characterisation of CO2 sources during

regional pollution events using isotopic and radiocarbon analysis, TELLUS SERIES B-CHEMICAL AND PHYSICAL METEOROLOGY, 48(4), 601–612, doi:10.1034/j.1600-0889.1996.00013.x, 1996.

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