

**Authors' responses to reviewer's comments follow. A copy of the reviewer comment is given (with comment 'number') followed by a response (blue font).**

## **Response to referee 1**

### 1. General comments

The manuscript discusses radiocarbon estimated fossil fuel CO<sub>2</sub> emissions from local South Korean sources as well as from the Asian continent based on samples taken at the GAW station Anmyeondo in Korea. Additionally, they calculated the emission ratios of CO/CO<sub>2</sub> and SF<sub>6</sub>/CO<sub>2</sub> and draw conclusions about improved oxidation efficiency in both the Asian continent as well as Korea. They also state based on a comparison between top-down and bottom-up (inventory) methods that there is a mismatch of estimated emissions to the point that inventory-based methods lead to up to 1.8 times lower emissions. The paper is well written, easy to follow and well-illustrated with graphs. I suggest publications of this manuscript after minor revision:

We thank you for your comments on the paper's value. We also appreciate your helpful comments to improve our manuscript. According to your specific comments, we revised our manuscript.

2. L:434 In South Korea and China, atmosphere-based RCO values are 1.2 times and (1.8±0.2) times greater than in the inventory, respectively. Please add also an uncertainty for the Korean value.

Thank you for the comment. We calculated each uncertainty for each sector. And we revised the sentence below.

**Line 480: In South Korea and China, atmosphere-based R<sub>CO</sub> values calculated by this study are (1.2±0.3) times (with KL), (1.6±0.4), (1.7±0.4), (2±0.1) and (1.7±0.2) times greater (with CB, CN, CE and OB) than in the inventory, respectively (Figure 4).**

Also in the abstracts

**Line 33: ...originating in China showed (1.6±0.4) to (2±0.1) times greater R<sub>CO</sub> than...**

In summary as well,

**Line 516: For CO, our values are (1.2±0.3) times and (1.6±0.4) to (2±0.1) times greater ...**

3. L: 38 the CO<sub>2</sub> increase rate seems very high to me with a large uncertainty, 2.4±0.5 ppm.

Thank you for the comment. Recently atmospheric CO<sub>2</sub> growth rate increased faster than the early measurement period, 1960s (0.8±0.3 ppm/year). ±0.5 is not uncertainty, rather the standard deviation of the annual increases. The value of S.D was a typo and should be ±0.4.

**Line 41: atmosphere at (2.4±0.4) μmol mol<sup>-1</sup> a<sup>-1</sup> in a recent decade globally (where 0.4 is the standard deviation of annual growth rates; [www.esrl.noaa.gov/gmd/ccgg/trends/](http://www.esrl.noaa.gov/gmd/ccgg/trends/), last access: 6 December 2019).**

From 2010 to 2019, the CO<sub>2</sub> global annual increase

Year	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	Mean (±S.D.)
ppm/year	2.4	1.7	2.4	2.4	2.0	3.0	2.9	2.1	2.4	2.6	2.4±0.4

These values are from [www.esrl.noaa.gov/gmd/ccgg/trends/](http://www.esrl.noaa.gov/gmd/ccgg/trends/), as we cited in the manuscript.

4. L: 56 . . . , since those (not clear what you mean here, I guess CO<sub>2</sub>)

Corrected.

**Line 60: We revised it from ‘those emissions’ to ‘fossil fuel CO<sub>2</sub> emissions’**

5. L:82-83 Why was the station location changed between the previous and the present study?

The TAP station does not belong to the KMA/NIMS and this paper focuses only on data from AMY. Therefore we think our data can give the information of this region with recent data since AMY is close to TAP (28 km away from AMY). We did not add that specific information in the manuscript.

6. L:126-127: what about permeation problems associated with glass flasks? To which pressure are the flasks filled? Under which conditions are the flask stored until measurement take place? How long does it take to be analysed?

The flasks have undergone extensive laboratory testing to ensure they maintain sample integrity for storage times up to one year. Comparison of flask-air samples with in situ measurements at South Pole have revealed storage offsets of up to 0.2 ppm after a year, but storage times at AMY are much less, and the difference in pressure between the flask and outside air (the main driver of preferential diffusion through the Teflon o-rings) is also less.

Flask-air sampling steps are as follows;

Using a semi-automated sampler, flasks are flushed at 5-6 L/min for 10 min then pressurized to 5 – 6 psig. After the pump turns off, falling pressure indicates a leak at the connectors. In that case, flasks are reconnected and the sample collected again. To prevent a slow leak through the pump, we close the stopcocks from the pump and to the exhaust first ([esrl.noaa.gov/gmd/ccgg/psu/manuals/psu\\_manual\\_1.6.pdf](https://esrl.noaa.gov/gmd/ccgg/psu/manuals/psu_manual_1.6.pdf)). We store the collected samples in the laboratory and send them to NOAA about every two months.

Another reason we are sure there is no permeation problem is that we compare the flask-air CO<sub>2</sub> data to KMA continuous measurements. We confirmed the differences are small close to GAW's compatibility goal ( $\pm 0.1$  ppm; Lee et al., 2019).

We added the sentence in the manuscript

**Line 114:** Two pairs of flask-air samples (4 flasks total, 2 L, borosilicate glass with Teflon O-ring sealed stopcocks) were collected about weekly from a 40 m tall tower at AMY, regardless of wind direction and speed from May 2014 to August 2016, generally between 1400 to 1600 local time (Table S1) using a semi-automated portable sampler. A pair of flasks was flushed for 10 min at 5-6 L min<sup>-1</sup> then pressurized to 5.5 psig in less than 1 min. A second pair is collected shortly after the first (within 20 min). The portable sampler was checked for leaks after pressurizing by observing the pressure gauge before closing the stopcocks. Batches of sampled flasks were shipped to Boulder, CO, USA every two months.

**Line 136:** When we compare NOAA's CO<sub>2</sub> measurements from flask-air with quasi-continuous measurements by KMA at AMY, the difference was -0.11±2.32 μmol mol<sup>-1</sup> (mean±1 σ), close to GAW's compatibility goal for CO<sub>2</sub> (±0.1 ppm for Northern Hemisphere measurements, Lee et al., 2019).

Reference: [www.esrl.noaa.gov/gmd/ccgg/flask.html](http://www.esrl.noaa.gov/gmd/ccgg/flask.html)

7. L:164-166: It might be worthwhile to give a upper limit estimate for this influence. Maybe, also CO<sub>2</sub> flux values for the Yellow and Japanese Sea would be helpful for the reader to underpin your conclusion.

We agree with the reviewer It would be great to test whether the samples were affected by ocean fluxes, but this is well-beyond this study. So we added more references. There is a reference value of a flux that estimate for the East China Sea of -4.2 mmol/m<sup>2</sup>/day (Song et al., 2018). This value is very negligible. Turnbull et al. (2009) reported no significant bias from oceans in the Northern Hemisphere, even at coastal sites, while this bias is very important in the Southern Hemisphere. Also Turnbull et al. (2011a) mentioned that ocean exchange was negligible at TAP. Therefore we just added this reference in the manuscript and explained the bias from the ocean can be negligible.

**Line 184:** It was also demonstrated there is no significant bias from the oceans including

**East China Sea (Song et al., 2018), even at coastal sites in Northern Hemisphere (Turnbull et al., 2009).**

8. Eq. 4-6 I guess these equations are well-known and not necessary to be shown again. I would skip it and only reference on a paper describing this or to the software tool that you have used to calculate the regressions.

We deleted and added the reference. On the other hand, to make readers understand easily, we described the equations in the supplementary materials.

**Line 201: we use reduced major axis (RMA) regression analysis (Sokal and Rohlf, 1981)**

**Line 205: The relevant equations are presented from Equ. S1 to Equ. S3.**

**Delete the equations from Line 207 to 217**

9. L:309-310 This is an important issue to be discussed in more detail, since this relevant with the conclusions drawn from the data about Asian emissions.

It is very clear that even the  $C_{ff}$  from CB sector in this study increased compared to TAP far-field samples from 2004/2010. CB sector is the cleanest sector in this study with high wind speed (median value is 5 m/s and maximum of 10.2 m/s) and high PBL (median value is 600 m and maximized up to 1700 m). Therefore we assumed that any contamination could not affect the samples due to synoptic condition. For other sector which are originated from China, not only this study but also other studies showed the increased values compared to the Turnbull et al.(2011a).

But we did not totally ignore the possibility. As reviewer mentioned, it would be great to mention about those factors in more detail and to consider for further study.

**Line 347: On the other hand, those values from this study showed large variability with small sample numbers due to different sampling strategy, environment, and synoptic**

conditions such as boundary layer height at the sampling time from reference studies. Further study will be necessary to understand those increased values.

10. L:314 what about correlation between SF<sub>6</sub> and CO?

When we implement RMA analysis for CO/SF<sub>6</sub>, the correlation is very weak (R=0.18). And to consider only the outflow of Asian continent, R was 0.24. Only CE and OB whose CO and SF<sub>6</sub> had a good correlation with C<sub>ff</sub> showed good correlation (R>0.6).

11. L: 337 what about a contamination from the local SF<sub>6</sub> emissions on the ratio assigned to the Asian continent? Could you get an handle on it from SF<sub>6</sub>/CO ratios?

We considered this idea, when analyzing the R<sub>gas</sub> values. But it was not possible due to several reasons. 1) as we explained, basically the correlation between SF<sub>6</sub> and CO was weak. 2) To select the data with SF<sub>6</sub>/CO ratio, the ratios of sample-by-sample should be constant (or Gaussian). However the data characteristics did not show that.

Therefore to reduce local SF<sub>6</sub> effects, after cluster analysis we select the data again for wind speed > 3 m/s, as described in section 3.2. As seen in Table 1, the mean value and standard deviation of SF<sub>6</sub> in outflow from the Asia continent is smaller than for South Korea. This also means that SF<sub>6</sub> values was not be affected by local effects as shown by relatively constant values. We have high confidence that ratios from the Asian continent are less affected by local pollution.

12. Fig. 2 How sensitive are the results on the selection of the background values? To use NWR as background sounds rather strange as the two stations are very far apart and the authors mention explicitly Chinese station as well. Alternatives would be a Japanese location? or a European station. Or even lower bound values of the AMY station based on Hysplit selection.

When we selected the baseline station, there were only a few possible stations where <sup>14</sup>C in CO<sub>2</sub> data were available. Asian stations would be a good option for this study but there is no available <sup>14</sup>C data. And even if a data set existed, when the sampling/analysis methods are different, the

data uncertainty can be increased. Therefore we used NWR data, which are located at similar latitude to AMY with the same sampling/analysis method used as at AMY. And the analysis for  $^{14}\text{C}$  was conducted by the same institute, INSTAAR, thus decreasing uncertainty that might occur if measurements from different laboratories were used (Miller et al., 2013)

According to Turnbull et al. (2011a), choice of background values did not significantly influence derived enhancements due to the large regional and local signal at TAP, 28 km from AMY. It was also described on Line 160. We hope the reviewer can understand that  $^{14}\text{C}$  data are limited, and this is the one of reasons which makes this paper valuable.

Reference: Miller et al., (2013), Initial results of an intercomparison of AMS-Based atmospheric  $^{14}\text{CO}_2$  measurements, Radiocarbon, Vol.55, Nr 2-3, 2013, 1475-1483.

13. Table 1 strange that  $r$  is low for PL trajectories. Has it to do with only a few values, since there is a much larger addition of fossil fuel  $\text{CO}_2$  present.

When sampling well-mixed air masses, we can clearly see correlations among the gases. Under stagnant conditions, due to micro scale meteorology, the gases showed correlated weakly.