

Anonymous Referee #1

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The authors have presented an evaluation of the CAMS prediction system, focusing on CO and CO<sub>2</sub>, during the KORUS-AQ campaign. They evaluated three different CO and CO<sub>2</sub> forecast and analysis products: 16-km CO and CO<sub>2</sub> forecasts, 9-km CO and CO<sub>2</sub> forecasts, and analyses of CO and CO<sub>2</sub> at 80 km and 40 km, respectively. The CAMS products were compared to the KORUS-AQ aircraft data as well as to ground-based and satellite measurements of CO and CO<sub>2</sub>. They found that CAM overestimated CO<sub>2</sub>, suggesting a positive bias in background CO<sub>2</sub>, whereas it underestimated CO, with the underestimate confined mainly to the lower troposphere. The authors also found that CAMS underestimates the outflow of pollution from China, possibly due to an underestimate of Chinese emissions. The study is a nice evaluation of CAMS CO and CO<sub>2</sub> under unique conditions. I have no major concerns about the analysis.

Response: Thank you!

My main concern is about the appropriateness of the manuscript for ACP. As a model evaluation study, I think it is better suited for GMD than ACP. My comments below are relatively minor, but must be address before the manuscript can be accepted for publication, if the Editor decides it is suitable for ACP.

Response: Thank you. We understand the reviewer's concern. But we think that our manuscript is still within the scope of ACP for the following reasons. We double-checked the main subject areas of ACP, which includes atmospheric modelling, field measurements, remote sensing, and laboratory studies of gases, aerosols, clouds and precipitation, radiation, and so on. In this study, we assess the performance of the Copernicus Atmosphere Monitoring Service (CAMS) global prediction system using field measurements from aircraft, ground sites, and ships, and remote sensing data during the KORUS-AQ field campaign. In addition to model evaluation, this study also addresses a few scientific topics on atmospheric chemistry and physics, including (1) anthropogenic combustion characteristics in Korea and China (as well as how well CAMS captures it), (2) impacts of different model configurations and environmental conditions on CO simulations, and (3) implications for CO emissions in CAMS. Thus, this manuscript is in line with the research focuses of ACP. Furthermore, we believe that the findings of this manuscript will be of interest not only to CAMS developers and users, but also to the general atmospheric chemistry community. Therefore, ACP is a perfect platform for us to share these results with the community. We sincerely hope that the editor will consider publishing this manuscript in ACP.

#### Comments

1. There is no mention of the **CAMS OH field**, which is critical for the simulation of CO. What is the global mean OH from the analyses and forecasts? On page 5, lines 28, it is mentioned that the 16-km CO forecasts use a linear chemistry scheme. A brief description of the scheme, either in the manuscript or in the supplement, would be helpful.

Response: The two high-resolution forecast products (FC16s and FC9s) employ a linear chemistry scheme, without the direct use of model OH.

The OH fields are only used in the CAMS ANs for CO which has full chemistry. In the ANs, the global and Northern Hemisphere mean are  $0.98 \times 10^{-6}$  molecules/cm<sup>3</sup> and  $1.20 \times 10^{-6}$  molecules/cm<sup>3</sup> during May 2016, respectively. We have extended description of the linear chemistry scheme at the end of Section 2.1. Specifically, we extended

*“A linear chemistry scheme is used in FC16s for CO (C-IFS-LINCO) for computationally expediency (Claeyman et al., 2010; Flemming et al., 2012; Massart et al. 2015; Eskes et al., 2017). Key aspects of the three CAMS configurations evaluated in this study are listed in Table 1.”*

to:

*“ANs for CO use the on-line implemented chemical mechanism (C-IFS-CB05, Flemming et al., 2015) that is an extended version of the Carbon Bond mechanism 5 (CB05, Yarwood et al., 2005). Because hydroxyl radical (OH) is an important sink for CO, modeled OH is critical for the simulation of CO (Gaubert et al., 2016, 2017). In the ANs for CO, the global and NH mean of air mass-weighted OH are  $0.98 \times 10^{-6}$  molecules/cm<sup>3</sup> and  $1.20 \times 10^{-6}$  molecules/cm<sup>3</sup> during May 2016, respectively (calculated following recommendations from Lawrence et al. (2001)). The mean OH from the ANs for CO is consistent with previous studies (e.g., Lawrence et al., 2001; Lelieveld et al., 2016; Gaubert et al., 2016, 2017). A linear chemistry scheme is (C-IFS-LINCO) used in FC16s and FC9s for CO for computationally expediency (Claeyman et al., 2010; Flemming et al., 2012; Massart et al. 2015; Eskes et al., 2017). C-IFS-LINCO computes CO sources and sinks using the approach developed by Cariolle and Déqué (1986) and updated by Cariolle and Teyssède (2007), without direct use of modeled OH. C-IFS-LINCO is less computationally demanding than the full chemistry, permitting simulations at higher resolutions (Massart et al. 2015). Key aspects of the three CAMS configurations evaluated in this study are listed in Table 1.”*

2. It is stated that the overestimate in CO<sub>2</sub> is associated with the bias correction in the biogenic source of CO<sub>2</sub>, but there is no discussion of this “bias correction”. Furthermore, in Agusti-Panareda et al. (2016) CAMS CO<sub>2</sub> was underestimating CO<sub>2</sub> observations from the surface in situ network and from TCCON, which the “bias correction” (the biospheric flux adjustment) reduced. Why is CAMS overestimating CO<sub>2</sub> here? A discussion is needed about the **treatment of the biospheric fluxes in CAMS and its possible impact on the modeled CO<sub>2</sub> over Korea**.

Response: First of all, we appreciate the reviewer for noticing this. According to Agusti-Panareda et al (2016), in the Northern Hemisphere there is a growing overestimation of the atmospheric CO<sub>2</sub> at the end of winter and throughout spring (from March to May); while at the end of the growing season in both the Northern Hemisphere and the Southern Hemisphere (August and March, respectively) there is a growing negative bias, i.e. an overestimation of the sink based on observations from NOAA/ESRL and TCCON (Section 5.1 of Agusti-Panareda et al (2016)). This is consistent with our finding. Agusti-Panareda et al (2016) also implies that the CO<sub>2</sub> overestimation by CAMS is enhanced in the BFAS simulation (Section 5.1 of Agusti-Panareda et al (2016)).

However, we note that the statement “As found by Agusti-Panareda et al (2016), the overall overestimation of CO<sub>2</sub> is associated with the biogenic bias correction” is inappropriate. We have changed it to “Agusti-Panareda et al. [2016] also suggests CO<sub>2</sub> is overestimated by CAMS in the Northern Hemisphere at the end of winter and throughout spring”.

We have also included more discussions on the bias correction section 2.1 (where we introduce biogenic flux adjustment scheme (BFAS)), including treatment of the biospheric fluxes in CAMS, and its possible impact on the modeled CO<sub>2</sub> over Korea.

3. On page 11 it was shown that the model produced steeper vertical gradients in CO and CO<sub>2</sub> than observed over Seoul, which the authors suggested may be due to weak boundary layer mixing. Since CAMS seems to perform better over Taehwa, it would be interesting to compare the vertical

gradients over Seoul and Taehwa in CAMS and in the observations to see if the issue is mainly an inability of CAMS to capture the PBL heights over the Seoul urban environment.

Response: We compared the observed vertical gradient of CO<sub>2</sub> over Seoul and Taehwa. We also analyzed the PBL height derived from observations and CAMS.

We have added the following analysis in the revised manuscript (Section 3.3.1):

*“We further find that compared with the Seoul metropolitan, the observed vertical gradient of CO<sub>2</sub> over Taehwa (~0.03 ppmv/hPa) below 925 hPa is smaller, which is relatively better captured by CAMS (0.02–0.12 ppmv/hPa). This again implies the possible inefficient boundary layer mixing in CAMS over the Seoul urban environment.”*

*“CO over Taehwa is more likely to be due to regional transport, as Taehwa is not a strong CO source region. Thus, the vertical gradient of CO over Taehwa does not necessarily reflect the impact of boundary layer mixing over Taehwa.”*

4. I am surprised that the analyses are not much better than the forecasts. Indeed, it seems as though the 9-km forecast is better than the analyses in some cases. I think it would be helpful for the reader if the authors expanded the description of the analyses to give the reader **more information about the configure and quality of the analyses**. Figure S1 and the brief text on page 5 are not enough.

Response: Analyses of both CO and CO<sub>2</sub> (ANs) do show improvement from the free running simulation (i.e., FC16s). However, the new 9-km forecast product (i.e., FC9s) is expected to have a better performance of FC9s because:

- (1) FC9s is initialized with the analysis product every 24 hours (i.e., it incorporates information of analyses every 24 hours);
- (2) FC9s has a much higher horizontal resolution (9 km) than the analyses (80 km for CO and 40 km for CO<sub>2</sub>).

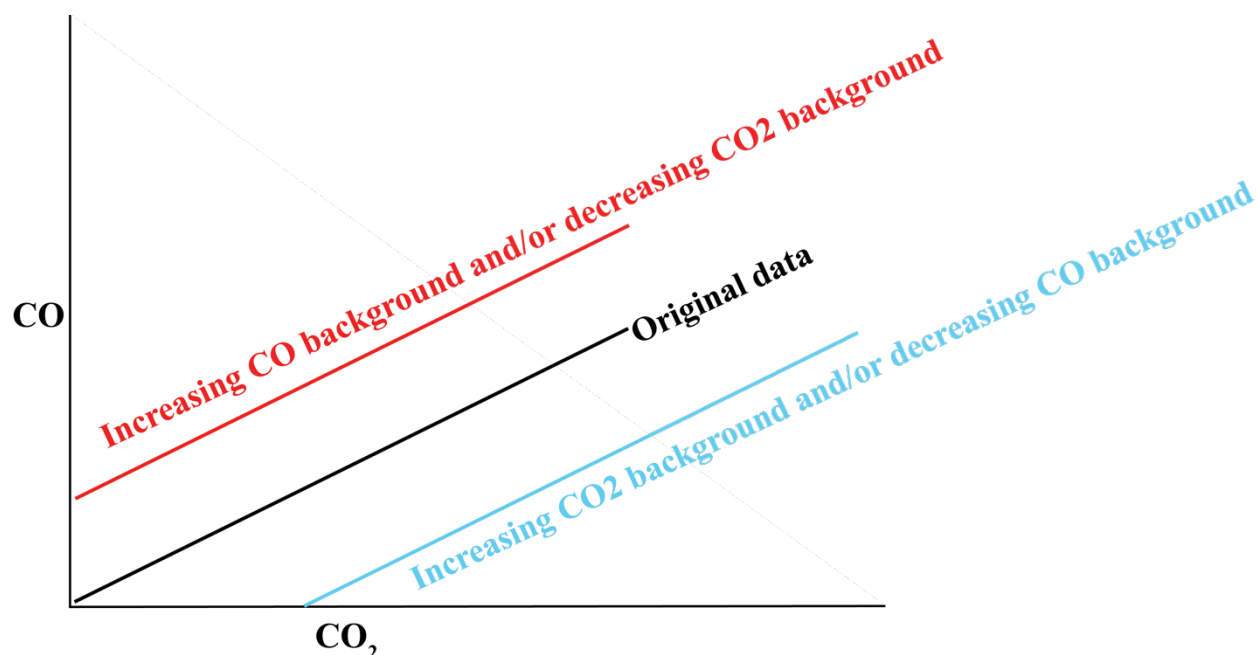
In addition to Figure S1 (i.e., the new Figure S2), Table 1 also summarized configurations of CAMS global atmospheric composition products including the analysis product of CO (AN\_CHEM) and CO<sub>2</sub> (AN\_GHG). We have also added more information about the configure and quality of the analyses:

*“Observations of both CO and CO<sub>2</sub> are assimilated in 12-hour assimilation windows. Inness et al. (2015) found that CO total column field, vertical distribution, and concentrations in the lower troposphere are improved by assimilating the CO total column from MOPITT. Assimilation of the GOSAT XCO<sub>2</sub> lead to improvements in mean absolute error and bias variability in XCO<sub>2</sub> fields during the year 2013 (Massart et al., 2016).”*

5. The discussion of **enhancement ratios** is confusing. It is unclear if the authors are using the slope of the CO/CO<sub>2</sub> relationship or the slope of delta CO/delta CO<sub>2</sub> relationship. The two approaches are different. The description in the text suggests that they are using the RMA regression of CO/CO<sub>2</sub> to assess the combustion sources, but throughout the text there is use of the delta CO/delta CO<sub>2</sub> notation. If they are indeed calculating an enhancement ratio (delta CO/delta CO<sub>2</sub>) above the background, how is the background being calculated? How sensitive is the analysis to the definition of the background?

Response: Thank you for pointing this out. We used slopes of the CO to CO<sub>2</sub> regressions as our enhancement ratios.

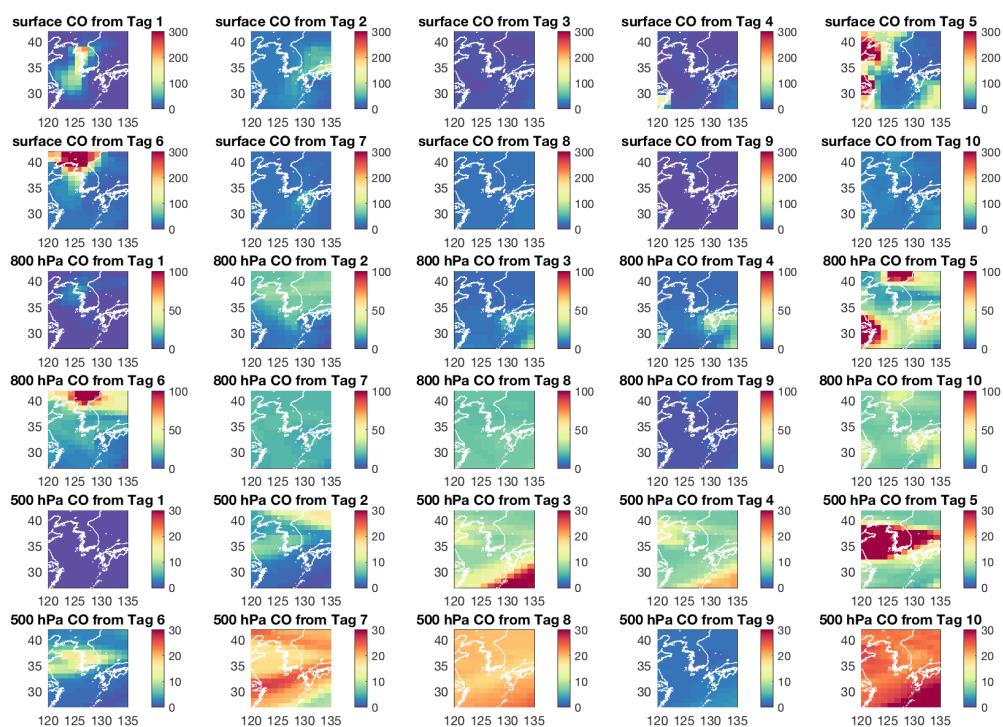
The estimated regression slope in the RMA corresponds to enhancement ratio of CO and CO<sub>2</sub> (dCO/dCO<sub>2</sub>). No background values are used here. In fact, the definition of the background does not change the regression slope. Please see the following figure for a demonstration.



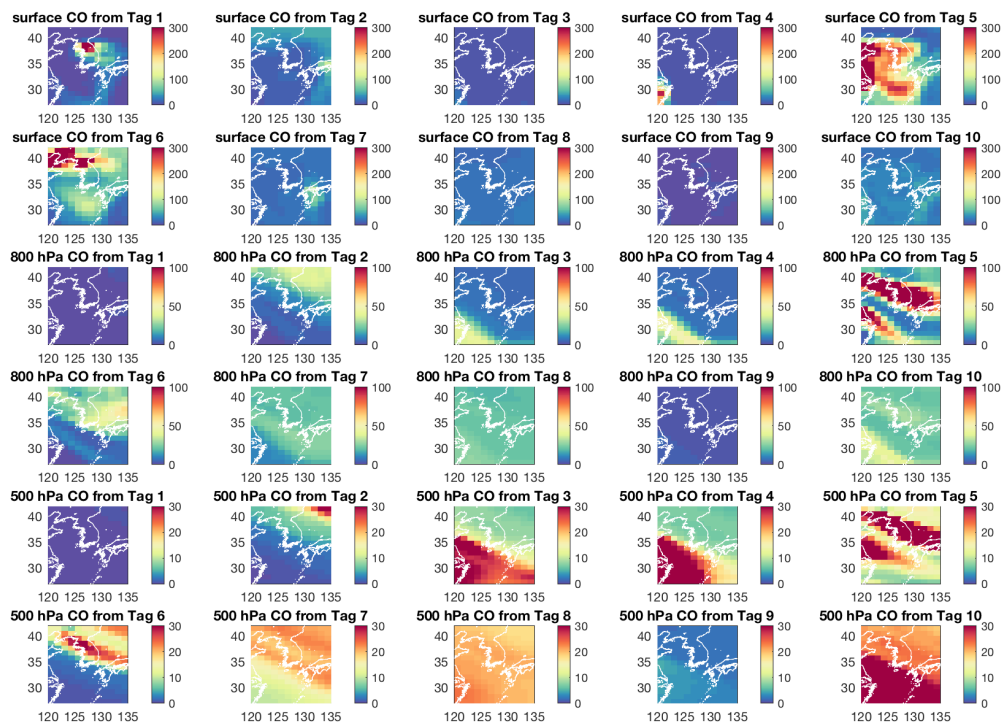
Nevertheless, we agree with the reviewer that the usage of  $\Delta\text{CO}/\Delta\text{CO}_2$  in this manuscript could be misleading, and have changed  $\Delta\text{CO}/\Delta\text{CO}_2$  to  $d\text{CO}/d\text{CO}_2$ .

6. The authors found that CAMS underestimated CO during China outflow events, but overestimated it under normal conditions. What are the **different source regions** for air reach the West Sea during “outflow” and “normal” conditions? To what degree is the model bias due to CAMS not capturing this difference in transport as compared to it not have the correct balance of emissions in China?

Our recent work with Community Atmosphere Model with chemistry (CAM-chem) tagged CO tracers studied the different source regions. Taking condition on June 5<sup>th</sup> (corresponding to the June 4<sup>th</sup> flight) as an example of normal conditions, both China and Korea contribute to the CO over the West Sea at surface. At 800 hPa, Japan, Russia, China, the rest of the world, and ocean all contribute to CO over the West Sea. However, CO concentrations over the West Sea is relatively low at these conditions. The following figure (Fig. S6 of Tang et al., 2018) shows spatial distributions of the tagged CO (ppbv) on June 5<sup>th</sup>, 2016 at model surface, 800 hPa, and 500 hPa (Tag 1: Korea; Tag 2: Japan+Russia; Tag 3: Indonesia+India; Tag 4: EA-S; Tag 5: EA-M; Tag 6: EA-N; Tag 7: the rest of the world+ocean; Tag 8: CH<sub>4</sub> oxidation; Tag 9: biogenic; Tag 10: chemical production besides CH<sub>4</sub>):



During outflow events (e.g., conditions during the May 30<sup>th</sup> flight), contribution from China are largely enhanced and becomes dominant at surface, 800 hPa, and 500 hPa. CO concentrations over the West Sea is relatively high during China outflow events. The following figure (Fig. S7 of Tang et al., 2018) shows spatial distributions of the tagged CO (ppbv) on May 31<sup>st</sup>, 2016 (corresponding to the May 30<sup>th</sup> flight) at model surface, 800 hPa, and 500 hPa (the Tags are the same as in the Fig. S6).



In Tang et al. (2018), we found that estimates of CO emissions in China rather than transport is potentially the main source of model bias over the West Sea. We have included the discussions and reference in the revised manuscript:

*“More elaborate analysis of source contributions during KORUS-AQ is beyond the scope of this study and can be found in Tang et al. (2018), which suggested that during China outflow events, the contribution from Chinese direct emissions to CO over the West Sea is largely enhanced and dominant.”*

#### Technical Comments

1. Page 4, line 26: add a comma between “forecast” and “CO2”.

Response: We have added the comma.

2. Page 4, line 28: add “the” between “on” and “free-running”.

Response: Thank you. We have edited accordingly.

3. Page 5, line 1: Is Figure 1 really necessary? I don’t think it adds much to the manuscript. Since there are already 11 figures, I would suggest removing Figure 1.

Response: We have moved Figure 1 to supplement (the New Figure S2).

4. Page 6, lines 1 and 2; add “the” before “South Korean peninsula”.

Response: Correction made.



5. Page 6, line 7: remove “including” before the list of the three questions.

Response: Thank you. We have deleted the “including”.

6. Page 6, line 8: The wording for question (3) needs improving. The English is not quite correct.

Response: Thank you for pointing this out. We have changed the sentence to “(3) *how well do models perform and what improvements are needed to better represent atmospheric composition over Korea and its connection to the larger global atmosphere (Kim and Park, 2014, KORUS-AQ White Paper).*”

7. Page 8, lines 22 and 23: The revisit and overpass times seem to be used interchangeably here. The revisit time of OCO-2, for example, is 16 days since it is in the A-Train orbit. However, the local overpass time is around 1:30 pm. For GOSAT the revisit time is 3 days.

Response: Thank you. We have changed “revisit” to “overpass”.

8. Page 8, line 26: Change 0.09e18 from e-notation to standard SI notation.

Response: We have changed the notation accordingly.

9. Page 9, line 5: Please add “is” between “CO<sub>2</sub>” and “associated”.

Response: We have deleted this sentence.

10. Page 10, line 1: The variance in CO in the May 3rd data does not seem larger than average to me. In fact, it seems to be smaller than average.

Response: We have changed the sentences “*For example, the flights in May 3rd, May 17th, May 24th, May 29th, and May 30th were specifically designed to capture Chinese pollution outflow. In these days, the variances in CAMS biases for CO (but not CO<sub>2</sub>) are larger than the average*” to

“*For example, parts of flight tracks on May 3rd, May 17th, May 24th, May 29th, and May 30th were specifically designed to capture Chinese pollution outflow. In these days, the variances in CAMS biases for CO (but not CO<sub>2</sub>) are generally larger than the average except for the flight tracks on May 3rd when Chinese influences were expected to be weak*”

11. Page 10, lines 12 and 14: Please change “tale” to “tail”.

Response: Thank you. We have changed it.

12. Page 12, line 4: It is unclear what is meant by the statement that “the wind speeds dominate the transport flux variations in CO<sub>2</sub>.” It the argument here that the meteorological uncertainty is the dominant contribution to the uncertainty in the forecast and analysis fields? If so, how does one come to that conclusion from Figure 7?

Response: Because the variations in CO<sub>2</sub> density are very low relative to CO<sub>2</sub> background, the pattern of the CO<sub>2</sub> fluxes in the Figure (CO<sub>2</sub> fluxes = wind speed × CO<sub>2</sub> density) mostly display pattern of wind speed instead of pattern of CO<sub>2</sub> density. However, we find the implication of “*the wind speeds dominate the transport flux variations in CO<sub>2</sub>*” to be redundant with previous sentences, and this sentence itself is confusing. We have deleted it.

13. Page 15, line 23: Section 4.2 only discusses the comparison to ship data of CO, not CO and CO<sub>2</sub>.

Response: Thank you. We have deleted “*and CO<sub>2</sub>*”.

14. Page 15, line 23: change “shop tracks” to “ship tracks”.

Response: Correction made.

15. Page 17, line 7: Change “size of CO data” to “amount of CO data”.

Thank you. We have changed “*size*” to “*amount*”.

16. Page 40, Table 3: The title for the table is wrong. This is the same title as for Table 4.

Response: Thank you for pointing this out. We have changed the title to “**Table 3.** *Enhancement ratios of CO to CO<sub>2</sub> (ppbv/ppmv), CO and CO<sub>2</sub> correlations, and bias of CO to bias of CO<sub>2</sub> correlations from airborne measurements, CAMS FC16s, ANs, and FC9s.*”

17. Figure S1: Should the labels “FX9s” and “FX16s” in the figure be “FC9s” and “FC16s”?

Response: Thank you for noticing this. The labels should be “*FC9s*” and “*FC16s*”, and we have corrected them.