Reply to reviews for "*Preindustrial to present-day changes in atmospheric carbon monoxide: agreements and gaps between ice archives and global model reconstructions*"

We are grateful to both reviewers for their time and consideration of our manuscript.
Here we reproduce the reviewers' comments in blue with our responses given in black. Line numbers, sections and figure numbering included in our response refers to the revised manuscript and SI. Where figures are explicit to this document they are references by letters (e.g., Fig. A).

1. Preamble

Two changes have been made to our article, in addition to those needed to address the reviews.

1. During the review process, we were contacted by Twan van Noije, who asked us to include the EC-Earth3 model outputs in our study. The EC-Earth3 model outputs were indeed made available later than our upload from the ESGF dataset repository.
   The [CO] model output from the EC-Earth3 model, and the description of this model have been added to our manuscript and the upload date of the CO modeling results on the ESGF data repository has been updated (Sect. 4). The inclusion of the EC-Earth3 model does not affect our conclusion since there is no significant change in the multimodel CO trend as illustrated in Fig. A, which compares atmospheric [CO] from AerChemMIP multimodel historical simulations with and without the EC-Earth3 model.

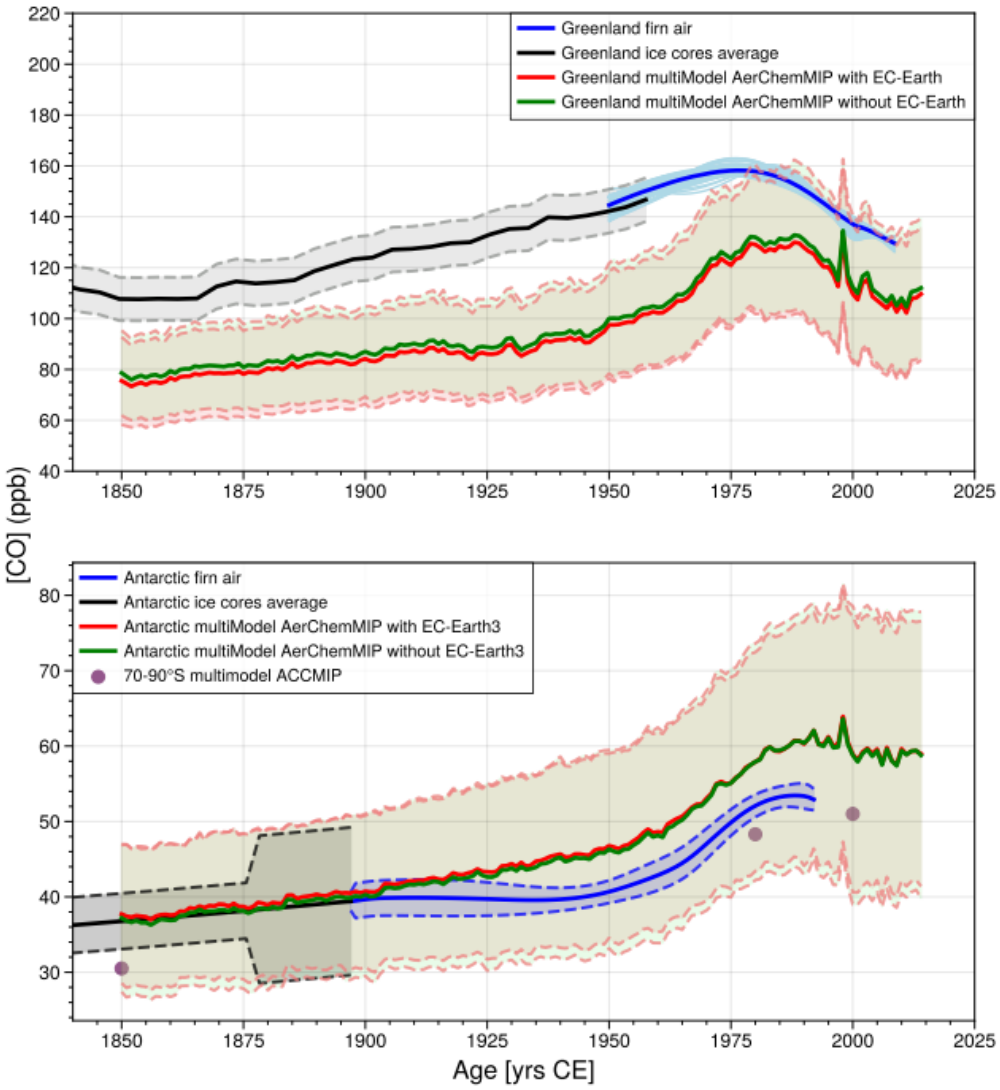

Figure A. Greenland (upper panel) and Antarctic (lower panel) atmospheric [CO] from Multimodel AerChemMIP historical [CO] simulations not including the EC-Earth3 model (green line, data reported during the first submission of the article) and including the EC-Earth3 model (red line, data included in the revised manuscript). The green and red envelopes on the AerChemMIP simulations show 1σ uncertainties. Ice cores and firn air reconstructions (Faïn et al., 2023, 2022; Petrenko et al., 2013) are also shown (see the revised manuscript for further description on ice core and firn air data uncertainties).

2. We have added to the discussion a recent paper published by Strawson et al. (2024), which reports a new multi-ice core CO record extracted from high accumulation Antarctic Peninsula sites, spanning 1821-1995 CE. The new record from Strawson et al. supports the previously published record from Faïn et al. (2023) discussed in this study and has been included in Fig. 2.

2. Reply to review #1

Main comments:

By comparison with the recorded CO concentrations ([CO]) in multi-site ice archives, this study analyzes the model performance of 16 ACCMIP models and 6 AerChemMIP models on simulating historical (1850-2014) [CO] over Greenland and Antarctica, focusing on both absolute values and trends of [CO], and points out that the model is biased in simulating high-latitude CO concentrations. The results are meaningful for improving the long-term simulations of [CO] in a GCM model. However, the analysis of the specific causes of these biases may be insufficient. More quantitative and in-depth analysis is needed to identify sources of the bias.

The aim of this article is to draw the modeling community's attention to areas where further analysis is needed to improve long-term simulations of [CO] in GCMs. To this end, we have identified gaps between existing information from updated models and polar archives. The main comments of reviewer 1 suggest that we have achieved this objective. However, this article is not a modeling article per se, and it does not provide in-depth quantitative analyses of the causes of [CO] bias in GCMs: this would require specific protocols to perform additional simulations, which is outside the scope of this study.

Specific comments:

Section 3.2, for the ACCMIP models, why were only 3 time slices selected in this study?

As stated in the first paragraph of the section 3 (line 151), ACCMIP consisted of a series of few time slice experiments: "*The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), which was part of CMIP5, consisted of a series of time slice experiments to investigate long-term changes in atmospheric composition between 1850 and 2100 CE, and how composition changes impact radiative forcing (Lamarque et al., 2013).*"

Time slices investigated in the framework of ACCMIP, for the historical period, include: 1850, 1930, 1980, and 2000 CE. In our study, we chose to include only time slices for which all the participating models were available (i.e., 1850, 1980 and 2000 CE) to avoid skewing the multimodel mean (see Table 2 in Lamarque et al., 2013).

Overall, we use in this paper the most recent modeling results made available by the MIP exercises, each time, and we are therefore entirely dependent on the choices made by others and the availability of model outputs.

Sections 3 and 4, It is suggested that in the text or in an annex, a table be added to give the anthropogenic and biogenic emission inventories and OH radicals used in each model.

About anthropogenic and open burning CO emissions:

As stated in Sect. 4.1.1, anthropogenic and open burning CO emissions used in the framework of AerChemMIP have been produced specifically for CMIP6 for the 1750-2014 CE period, and are common to all models (line 203). Anthropogenic CO emissions are reported by Hoesly et al. (2018); open burning CO emissions are based on merging satellite observations with proxies and fire models (van Marle et al., 2017).

About BVOC sources and emission inventories:

Biogenic VOC sources are not constrained by the AerChemMIP protocol. Section 4.1.1 provides model-specific approaches involved in interactive emissions of BVOC with more details in the revised manuscript (lines 207-213). Furthermore, Griffith et al. (2021, Fig. 1) reports BVOC emission for the period spanning 1850-2014 for 5 of the 7 models involved in our study (all models but EC-Earth3 and BCC-ESM1): this is now stated in the manuscript.

About OH radicals:

Tropospheric OH burden and vertical distributions vary between models, and would be extremely difficult to describe with a table. The manuscript includes all the citations needed for the reader to learn more about OH fields simulated by the 7 AerChemMIP models included in our study (Sect. 4.1.2).

In summary, including here a table is not relevant since these quantities are dynamically computed by each model but Sect 4.1 has been extended to better describe how is handled by the various AerChemMIP models and to refer to Fig. 1 in Griffith et al. (2021) to see the BVOC emissions.

Lines 230-231, It is recommended that the differences between the models are analysed in more detail to see why the UKESM1 model can accurately simulate the [CO], the BCC-ESM1 model overestimates [CO] and the other models underestimate it.

Anthropogenic and open burning emissions, which represent 50% of the CO sources (Fig. 1) are identical for all AerchemMIP models (Sect. 4.1.). The inter-variability in [CO] between models is thus related to the OH sink, and/or the secondary CO sources.

Inter-model variability in the CO OH-sink can be observed in the dataset reported by Griffith et al. (2021). Fig. 1 of Griffith's study compares the evolution for the period spanning 1850 CE to present of both CO emissions and CO burden, for 5 of the 7 models included in this study. Interestingly, models with larger CO emissions can exhibit lower CO burden (e.g., CESM2), suggesting a more efficient CO OH-sink for those models. Indeed, models vary in their simulation of the distribution and trends of OH radicals over the historical period (Naïk et al., 2013; Zhao et al., 2019; Stevenson et al., 2020).

Factors controlling inter-model OH discrepancies can be complex as differences in model emissions, chemistry, physics, and dynamics can together impact [OH]. In the frame of ACCMIP, several reasons have been ascribed to the variability in OH across models which then have implications for simulated CO trends and variability. Specifically, Naïk et al. (2013) find that the preindustrial to present-day OH trend simulated by a model depends linearly on the ratio of the change in global mean tropospheric CO and NOx burdens (ΔCO/ΔNOx – approximately represents changes in OH sinks versus changes in OH sources). Zhao et al. (2019) reported that the inter-model differences in tropospheric OH burden and vertical distributions are mainly determined by the differences in the nitrogen oxide (NO) distributions, while the spatial discrepancies between OH fields are mostly due to differences in natural emissions and volatile organic compound (VOC) chemistry.

Furthermore, Griffiths et al. (2021) also demonstrate that secondary CO sources and OH sink are strongly related. Although all CO emission datasets reported by Griffiths et al. (Fig. 1) show the same trends, they show different absolute levels (e.g., CO emissions are 33% lower for GISS compared to GFDL). This inter-model variability shows that differences in transport dynamics and chemical schemes can largely impact CO secondary emissions, and ultimately the atmospheric CO burden. As an example, the CH_4-oxidation CO source is well constrained in terms of trend but this source can vary according to absolute OH values differing from one model to another. It can also vary according to different yields of CO from CH_4 oxidation (e.g., removal of CH_2O before formation of CO).

The multiple and complex inter-relations between factors controlling inter-model OH discrepancies would require a thorough analysis which should be performed with coordinated process-oriented evaluation of models. Such evaluation is out of the scope of this study, as clarified in Sect. 5.1 and 5.2 (line ~300) of the manuscript but the need of such study and the possibility to use CO trends deduced from ice core analysis if better highlighted in the perspectives of the paper (Sect. 6, line 460).

In addition, there are significant differences in [CO] between AerChemMIP and ACCMIP, what's the reason for these differences. Overall, the authors provide a lot of analysis of deviations from modelled and observed trends, but there is a lack of explanation of the reasons for the deviations between modelled and observed absolute concentrations.

As discussed previously, modeled trends in atmospheric CO concentrations are determined by the trends in its sources and sinks in the models. Fig. B reports the total anthropogenic emissions prescribed in ACCMIP (CMIP5) and AerChemMIP (CMIP6): trends in anthropogenic emissions are similar for ACCMIP and AerCheMMP models. However, the trends in secondary CO sources (oxidation of VOCs) in the models are difficult to compare across the different generations of models because of the lack of required output. Since models vary in their representation of the chemical mechanisms (and therefore VOC chemistry), we expect this to be a driving factor in the diversity of CO trends similar to that for O_3 (Young et al., 2013, Griffith et al., 2020). Additionally, trends in OH (see Sect. 3.1.2 and 4.1.2 of the manuscript), which is the primary sink for CO, also vary across the models (Naik et al. 2013, Stevenson et al., 2020).

Overall, the focus of the paper is not to analyze the inter-exercises (ACCMIP vs AerChemMIP) and inter-model's differences in atmospheric [CO] levels because it would require more outputs from the models that have not been archived at the time of the intermodal exercise (and would sometimes require extra simulations) but showing both AerChemMIP and ACCMIP allows to point progress in the modeling ability to reproduce CO trends.

Sect. 5.2. has been modified to discuss the differences between ACCMIP and AerChemMIP in more detail, and also to highlight the limitations of comparing ACCMIP and AerChemMIP outputs (lines 295-302).

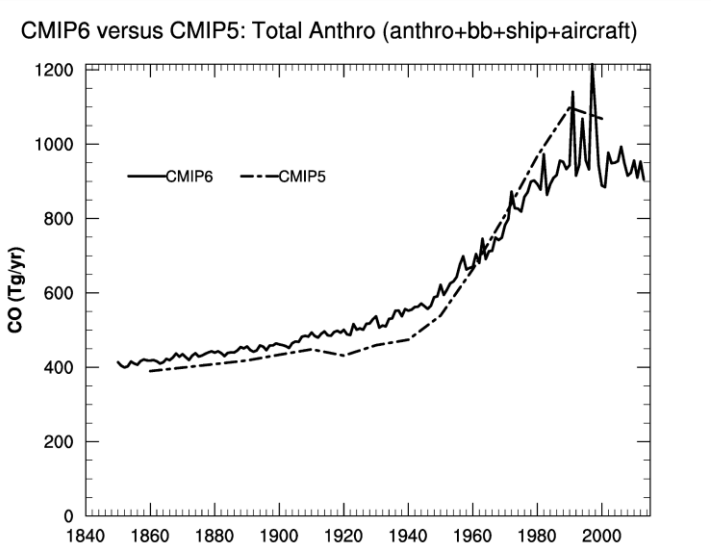

Figure B. Total CO anthropogenic emissions prescribed in the frame of CMIP5-ACCMIP (Lamaraque et al., 2010) and CMIP6-AerChemMIP (Hoesly et al., 2018). The strong variability in CMIP6 comes from biomass burning emissions over the satellite era.

Lines 335-337, "the mismatch in [CO] trends … may be related to uncertainties in CO emission factors (EF)", This conclusion is too arbitrary. Although the author describes that the [CO_2] has a sharp increase in growth rates in ~1945 CE, as shown in Fig 3a, for [CO], there is also a sharp increase in ~1945 CE. It is recommended that the authors make some quantitative comparisons rather than just qualitative descriptions.

CEDS anthropogenic emissions are calculated as the product of activity data and emission factors (Hoesly et al., 2018). Anthropogenic emissions of both CO_2 and CO largely originate from fossil fuel combustion, involving similar activity dataset. In Sect. 5.3.3, we introduce CO_2 in the discussion as a proxy and constraint on fossil fuel combustion activity dataset. Indeed, uncertainty on emission inventories is among the lowest for CO2 (Hoesly et al., 2018). If we hypothesize that fossil fuel combustion activity dataset are constrained by CO_2, then "the mismatch in [CO] trends … may be related to uncertainties in CO emission factors (EF)". This conclusion is supported by Hoesly et al. (2018) who states that combustion EF for CO are uncertain before 1970 CE. A quantitative comparison of CO and CO_2 trends and emissions would not support further our reasoning here. We clarified this point in Sect. 5.3.3 which has been largely modified.

Section 5.3.3, Many “top-down” inversions (e.g., Müller, and Stavrakou, 2005, doi:10.5194/acp-5-1157-2005; Feng et al., 2020, doi: 10.1029/2019JD031808.) have shown that the present bottom-up inventories underestimate the anthropogenic CO emissions in the Northern Hemisphere. Therefore, I suggest that the author can cite some inversion studies to support that the anthropogenic CO emissions have been underestimated.

To our understanding Feng et al. (2020) published a description of the methods used for generating gridded datasets produced for use by the modeling community, particularly for CMIP6. They did not produce a different global emission inventory for the historical period.

We agree with the reviewer that many “top-down” inversions suggests that bottom-up inventories underestimate CO emissions in the Northern Hemisphere: this point is discussed in section 5.1 (line 259), citing some more recent related papers by Gaubert et al. (2020), Miyazaki et al., (2020) or Zheng et al. (2019).

Line 30, “outputs from 16 (Atmospheric” => “outputs from 16 ACCMIP (Atmospheric”

This typo was corrected.

Line 75, “[CO]”, full name is needed here.

The manuscript was corrected accordingly.

Line 143, Missing period.

This typo was corrected.

Line 253 and 258, ‘AerChemMip’=>‘AerChemMIP’

This typo was corrected.

Line 269, 2 times larger than what?

The sentence was rephrased as follow: “Both models and observations also reveal an increase in [CO] growth rate in the SH, which is twice as large between 1945 and 1980 CE as it was between 1850 and 1945 CE”

Line 301, “XXth”

This typo was corrected.

Line 325, in Figure 4, fossil fuel emissions are shown as brown lines instead of purple lines, and fire emissions are shown as blue lines instead of brown lines.

The manuscript (Sect. 5) has been corrected to refer to the appropriate colors when discussing Fig. 4. Also, the legends of the Fig.4 subplots have been corrected to better match with the figure caption: “Anthropogenic (all)” has been replaced by “Fossil fuel combustion”.

Figure 4, Labels of a and b should be added to the top left corner of the two subfigures. In addition, the text of 0-90N and 0-90S in the figures is inconsistent with the description in the figure title of 30-90N and 30-90S. Meanwhile, it is recommended that the timeframe of this graph be consistent with that in Figures 2 and 3.

Figure 4 has been improved as suggested by the reviewer : its time frame is now 1850-2014 CE; a and b labels have been added. The figure captions have been corrected to mention 30-90 instead of 0-90.

Line 331, “WWII”, full name is needed here.

Full name has been added.

Line 332, “EF”, full name is needed here.

Full name has been added.

3. Reply to review #2

The study presents a comparison of modeled atmospheric carbon monoxide (CO) against reconstructions of CO from ice cores and firn air. The authors focus on comparisons for high latitudes in the Northern Hemisphere (NH) and Southern Hemisphere (SH), investigating the multi-model mean for models used in ACCMIP and AerChemMIP. They find improvement of CO representation from ACCMIP to AerChemMIP. CO trends are generally well reproduced by the multimodel mean, with potential reasons for discrepancies discussed.

Overall the manuscript is well written and the analysis rigorous. The evaluation of model results against measurement is important, and timely as a benchmark for future modeling studies. I have several comments to be addressed below.

Main Comments:

Fig. 2 and Fig. 3 – the different lines in the firn air record for the NH need defining. Originally, I thought the spread was a measure of uncertainty shading, but it looks like multiple lines are plotted from the NH firn data. Please clarify. Also, describe other uncertainty bounds in the caption.

The NH firn air record is reported directly from Petrenko et al. (2013). The different lines in the NH [CO] firn air record represent 61 scenarios that can successfully fit with the multisite firn air data, through inverse modeling of gas transport in the firn (Witrant et al., 2012). This approach is described in detail by Petrenko et al. (2013) who used the spread of these 61 scenarios as a metric of uncertainty for the mean [CO] scenario. Captions of both Fig. 2 and 3 were modified to clarify this point.

Introduction: the discussion of methane oxidation was a little confusing around line 53 onwards. Consider clarifying by first mentioning secondary CO production accounts for about 50% of CO globally, and then state methane oxidation is the dominant secondary source.

The manuscript was modified accordingly.

Section 3.2 and Section 4.2 – Please reference if there is a public data repository where you obtained the ACCMIP and AerChemMIP data, or explain how you obtained the data model output.

The following information on data repository have been added to the manuscript:

ACCMIP: https://catalogue.ceda.ac.uk/

AerChemMIP: (https://aims2.llnl.gov/search)

Section 5.2: There are several additions/clarifications needed for the trend analysis.

- Please clarify whether trends were defined 1980 to 2020 or from 2000?
- What method was used to determine trends? Please add uncertainties to your trend estimates.
- What is being used to define “excellent” agreement?
- Please also state the SH trends in addition to the NH.

In Sect. 5.2, we define [CO] trends as follows: ”*Trends in atmospheric [CO] are defined here as variations in [CO] relative to the year 2000 CE*” (line 269).

The trends provided in the draft manuscript in Sect. 5.2 were only qualitative. We have carefully recalculated trends for NH multimodel, firn air and atmospheric monitoring [CO], for the period 1990-2010 CE:

- Trend in multi-modeled NH [CO]: -01.22 ±0.16 ppb yr^{-1}, (1σ), p<0.01
- Trend in Greenland firn air NH [CO]:-1.14±0.01 ppb yr^{-1} (1σ), p<0.01
- Trend in Barrow [CO] atmospheric monitoring: -1.27±0.26 ppb yr^{-1} (1σ), p<0.01

[CO] trends for the SH has also been calculated:

- Trend in multi-model SH [CO] :-0.15±0.04 ppb yr^{-1} (1σ), p<0.01
- Trend in Antarctica firn air NH [CO] : not calculated, time series end up in 1992.
- Trend in Mawson [CO] atmospheric monitoring : -0.20±0.07 ppb yr^{-1} (1σ), p<0.01

The manuscript has been updated with these trends.

We use the term 'excellent' because all the trends for the period spanning 1990-2010 CE computed from the model outputs agree are in the uncertainty ranges of the reconstructed ones, for both NH and SH.

L254-256: The statement "… 4 ppb higher in 2000 CE compared to 1980 CE. ACCMIP models were not able to reproduce decline in Arctic [CO]" seems inconsistent with Fig.2 where the 2000 value seems lower than the 1980 value in the NH.

The manuscript was corrected as follows : "… 4 ppb *lower* in 2000 CE compared to 1980 CE. ACCMIP models were not able to *fully* reproduce decline in Arctic [CO]" (line 290).

Section 5.3: The time bound is 1945 on line 266, but 1950 on line 276. I suggest to chose one time bound for ease of understanding.

The appropriate time bound here is 1945, and the manuscript was corrected accordingly.

Fig. 4: I suggest the time range begin at 1850 to be consistent with other figures and the main text arguments.

Fig. 4 has been modified accordingly.

Section 5.4: Consider whether there is also some potential overestimation in emission sources for the SH between 1900 to 1950 – the model trend seems higher than the measurements during this time period in Figs. 2 and 3.

Over the period 1900 to 1950 CE, the AerChemMIP model mean and the Antarctic ice core record agree within their uncertainty envelopes. Consequently, we have chosen not to discuss further the differences between modeled and observed SH trends for this period.

Technical Corrections:

Fig 2. and Fig 3. caption: 1850 and 1900 should be 1850, 1980 and 2000.

This typo was corrected.

3.1 Heading – consider changing to “ACCMIP CO budget” to be consistent with section 4.1.

The manuscript was modified as suggested.

Line 302: Check CCMS is defined

CCMs is now defined in the introduction.

Fig. 4: I suggest to rename “Open Burning” in the legend to “Biomass Burning” to be consistent with the main text.

As suggested, we have renamed “Open Burning” to “Biomass Burning” in the entire manuscript.

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