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

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Abstract. Global chemistry-climate models (CCMs) play an important role in assessing the climate and air pollution implications of aerosols and chemically reactive gases. Evaluating these models under past conditions and constraining
25 historical sources and sinks necessitates reliable records of atmospheric mixing ratios spanning preindustrial times. Such precious records were recently obtained for carbon monoxide (CO) documenting for the first time the evolution of this reactive compound over the industrial era. In this study, we compare the simulated atmospheric surface CO mixing ratios ([CO]) from two different sets of CCMs and emissions in the frame of CMIP5 and of CMIP6 (Coupled Model Intercomparison Project Phases 5 and 6) with recent bipolar ice archive reconstructions for the period spanning 1850 to present. We analyze how
30 historical (1850-2014) [CO] outputs from 16 (Atmospheric Chemistry and Climate Model Intercomparison Project) models and 6 AerChemMIP (Aerosol Chemistry Model Intercomparison Project) models over Greenland and Antarctica are able to capture both absolute values and trends recorded in multi-site ice archives. While most models underestimate [CO] at high northern latitudes, a reduction in this bias is observed from ACCMIP to AerChemMIP exercises. Over the 1980-2010 CE period, trends in ice archive and firn air observations and AerChemMIP outputs align remarkably well at high northern and
35 southern latitudes, indicating improved quantification of CO anthropogenic emissions and the main CO sink (OH oxidation) compared to ACCMIP. From 1850 to 1980 CE, AerChemMIP models and observations consistently show increasing [CO] in both the Northern Hemisphere (NH) and Southern Hemisphere (SH), suggesting a robust understanding of the CO budget evolution. However, a divergence in the [CO] growth rate emerges in NH between models and observations over the 1920-1975 CE period, attributed to uncertainties in CO emission factors (EF), particularly EF for RCO (Residential, Commercial

40 and Others) and transportation sectors, although we cannot totally rule out that the CO record based on Greenland ice archives may be biased high by CO chemical production processes occurring in the ice prior the measurements (i.e., in situ CO production). In the Southern Hemisphere, AerChemMIP models simulate an increase in atmospheric [CO] from 1850 to 1980 CE closely reproducing the observations (22±10 ppb and 13±7 ppb, respectively). Such agreement supports CMIP6 biomass burning CO emission inventories which do not reveal a peak in CO emissions in the late 19th century. Furthermore, both SH

45 models and observations reveal an accelerated growth rate in [CO] during 1945-1980 CE relative to 1980-1945 CE, likely linked to increased anthropogenic transportation emissions.

1 Introduction

Carbon monoxide (CO) is an air pollutant that affects the oxidizing capacity of the troposphere and contributes to ozone formation (Crutzen, 1973). Its primary loss is through reaction with OH with a small contribution from dry deposition, which

50 leads to an atmospheric lifetime of about 2 months (Khalil et al., 1999). Thus, CO is an excellent tracer of long-range transport and an integrator of atmospheric chemical processes. In the modern atmosphere, the main sources of CO are combustion (fossil fuel and biomass burning) and through atmospheric oxidation of methane (CH_4) and non-methane volatile organic compounds (VOCs). Methane oxidation currently amounts to 70-80 % of the atmospheric CO production (Duncan et al., 2007; Fortems-Cheiney et al., 2012). CO surface emissions include biomass burning (van der Werf et al., 2017), incomplete combustion of

55 anthropogenic fossil fuels and biofuels (Hoesly et al., 2018), and minor contributions from the ocean (Conte et al., 2019) and plant leaves (Bruhn et al., 2013). Incomplete fossil fuel combustion represents ~70% of CO primary emissions (Szopa et al., 2021, and references therein). A schematic overview of the modern global CO budget is given in Fig. 1.
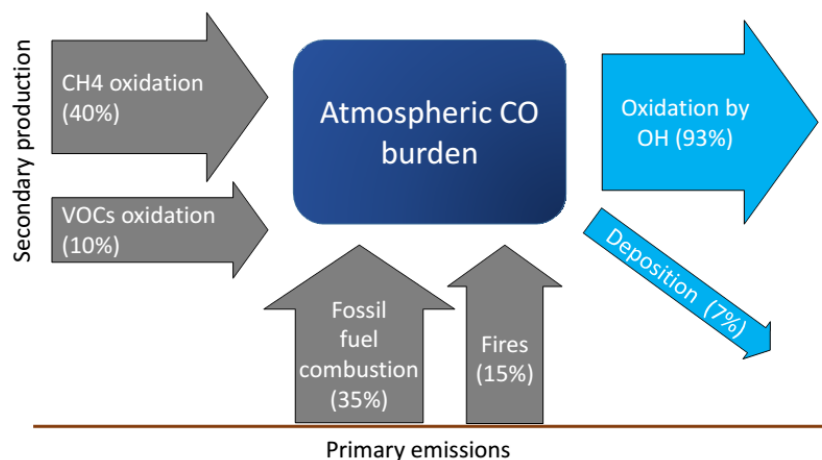
Figure 1: Modern budget of atmospheric CO. Primary emissions and secondary production each amount to ~50% of CO sources

60 **(Szopa et al., 2021). Primary emissions for plant leaves and oceans are small and not shown here, the proportion of emissions from fossil fuel combustion and fires is 35% and 15%, respectively (Szopa et al., 2021). Secondary CO production is dominated by CH_4 oxidation which amounts to ~35-40% of the global CO source, with VOCs oxidation accounting for ~10-15% (Duncan et al., 2007; Fortems-Cheiney et al., 2012). The main CO sink is through OH oxidation, which accounts for more than 90% of CO removal (Khalil et al., 1999; Stein et al., 2014).**

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Recent advances in satellite retrievals, ground based column observations, airborne platforms, surface measurement networks and assimilation products have improved the characterization of the present-day atmospheric CO distribution (Szopa et al., 2021, and reference therein). Typical annual mean surface CO concentrations range from ~120 ppb in the Northern Hemisphere (NH) to ~40 ppb in the Southern Hemisphere (SH) (Petron et al., 2023). A study using data assimilation techniques estimates

70   a global mean CO burden of 356 ± 27 Tg over the 2002-2013 period (Gaubert et al., 2017). There is high confidence that global CO burden has been declining since 2000 (Szopa et al., 2021, and references therein), although this global decreasing trend in CO has shown a slowdown over the last 5 years (Buchholz et al., 2021; Petron et al., 2023).

Recent analytical and modeling advances applied to ice archives (firn air and bubbles trapped in deep ice) from both Greenland (Petrenko et al., 2013; Faïn et al., 2022) and Antarctica (Faïn et al., 2023) have allowed the production of a new bipolar

75   reconstruction of atmospheric [CO] including preindustrial (PI) times and spanning 1700-1992 CE (resp. 1700-2016) in the SH (resp. in the NH). These reconstructions can be extended to present day using direct measurements of atmospheric [CO] at high latitudes, e.g. at Mawson or South Pole stations (Antarctica) and Cape Grim station (Australia) in the SH (Faïn et al., 2023; Loh et al., 2021).

Estimates of global anthropogenic emissions of both CO and its precursors have been revised recently for use in the Coupled-

80   Model Intercomparison Project 6 (CMIP6) (Collins et al., 2017). Hoesly et al. (2018) have extended these estimates of anthropogenic emissions back to 1750 and developed differentiation (source sector-wise) and gridding of emissions (Feng et al., 2020). Similarly, biomass burning CO emissions, which represent about 30% of present-day global CO emissions (i.e., about 15% of global CO sources, Fig. 1) have been revised in the framework of CMIP6 (van Marle et al., 2017). CMIP6 biomass burning CO emissions, which assume that fires scale up with population density, increase only slightly from 1750 to

85   2015 CE and peak during the 1990s after which they decrease gradually and consistently. However, fire emissions reaching higher levels in the PI compared to present day have been recently suggested (Hamilton et al., 2018; Liu et al., 2021; Rowlinson et al., 2020) as a result of a possible decline in burned areas with increasing population density due to land use changes (Andela et al., 2017; Knorr et al., 2014).

CMIP6 updated anthropogenic and biomass burning CO emissions inventories have been used by a set of state of the art global

90   chemistry-climate models (CCMs) to investigate the trends in global tropospheric hydroxyl radical and methane lifetime (Stevenson et al., 2020) and the evolution of the tropospheric ozone burden and budget terms (Griffiths et al., 2021) since 1850 CE. CCMs suggest that the global CO tropospheric burden has increased by ~120 Tg since 1850 (Fig. 1 in Griffiths et al., 2021), with: (i) a moderate increase rate from 1850 to 1950 CE, (ii) a fast increase rate from 1950 to 1990 CE, and (iii) a decrease trend from 1990 and 2015 which reproduces the observed decline in tropospheric [CO] (Szopa et al., 2021, and

95   reference therein).

In this paper we compare atmospheric surface [CO] simulated by two different sets of CCMs and emissions in the frame of CMIP5 (Lamarque et al., 2013) and of CMIP6 with recent reconstructions from polar ice archives for the period spanning 1850 to present. The aim of this work is to establish the periods of robust agreement between models and data, indicating a

robust knowledge of emissions and adequate fit-for-purpose of the models over such periods, and discuss the possible reasons

100   for disagreement over specific periods.

2 Bipolar [CO] reconstructions from polar ice archives

Ground based and satellite-derived CO data are only available for the last three decades. Ancient air preserved in glacial ice and firn is thus a unique archive for reconstructing the past atmospheric [CO] record prior to the 1990s. A large amount of air can be sampled from the interconnected open pores of the firn, which is the upper layer of an ice sheet where snow is slowly

105   transformed into ice. Mean ages of atmospheric gases increase with firn depth to about a century ago. Analysis of air trapped in bubbles in solid ice below the firn layer is required to extend reconstructions further back in time, and can now be conducted by Continuous Flow Analyses (CFA) which produce continuous, high resolution, datasets (Faïn et al., 2023, 2022, 2014).

By analyzing [CO] depth profiles collected from firn air at three different Greenland sites (NGRIP, Summit, and NEEM), Petrenko et al. (2013) obtained a reconstruction of atmospheric [CO] spanning 1950-2010 CE. Over the same period, stable

110   isotopes of CO (i.e., $\delta^{13}CO$ and $\delta C^{18}O$) were analyzed in firn air samples collected at the NEEM site to provide additional powerful clues for the interpretation of the changes in the CO budget (Wang et al., 2012). The reconstruction from Petrenko et al. (2013) has been recently extended back to 1700 CE with the CFA analysis of 4 different Greenland ice cores (Fig. 2, upper panel, NGRIP, PLACE, NEEM, and D4) (Faïn et al., 2022). Faïn et al. (2022) could not fully exclude the possibility that the Greenland ice archive CO reconstruction could be slightly positively biased by chemical processes (in situ production)

115   occurring within the Greenland ice. Therefore, the atmospheric [CO] history extracted from Greenland should be considered as an upper bound of the past [CO] in the Arctic (Faïn et al., 2022). The temporal changes depicted by this historical Greenland [CO] record probably have a larger spatial significance than the Arctic context alone, as suggested by the GAW (Global Atmospheric Watch) reactive gases measurement network (Petron et al., 2023; Schultz et al., 2015).

Ice archives from Antarctica were also investigated: CO was collected in firn air at seven sites (Lock In, DE08-2, DSSW19K,

120   DSSW20K, South Pole, ABN, and Berkner Island) to reconstruct [CO] in the Antarctic atmosphere from 1898 CE to present (Fig. 2, lower panel; Faïn et al., 2023). CFA analyses conducted on three ice cores (DC12, ABN, and TaldIce) extended this reconstruction back for three millenia starting 1897 CE (Faïn et al., 2023). Ice core and firn air measurements, as well as firn air and direct atmospheric measurements, show excellent agreements during intervals of overlap. These Antarctic CO records appear to be unaffected by in situ CO production or sampling artifacts. This new Antarctic CO reconstruction suggests that the

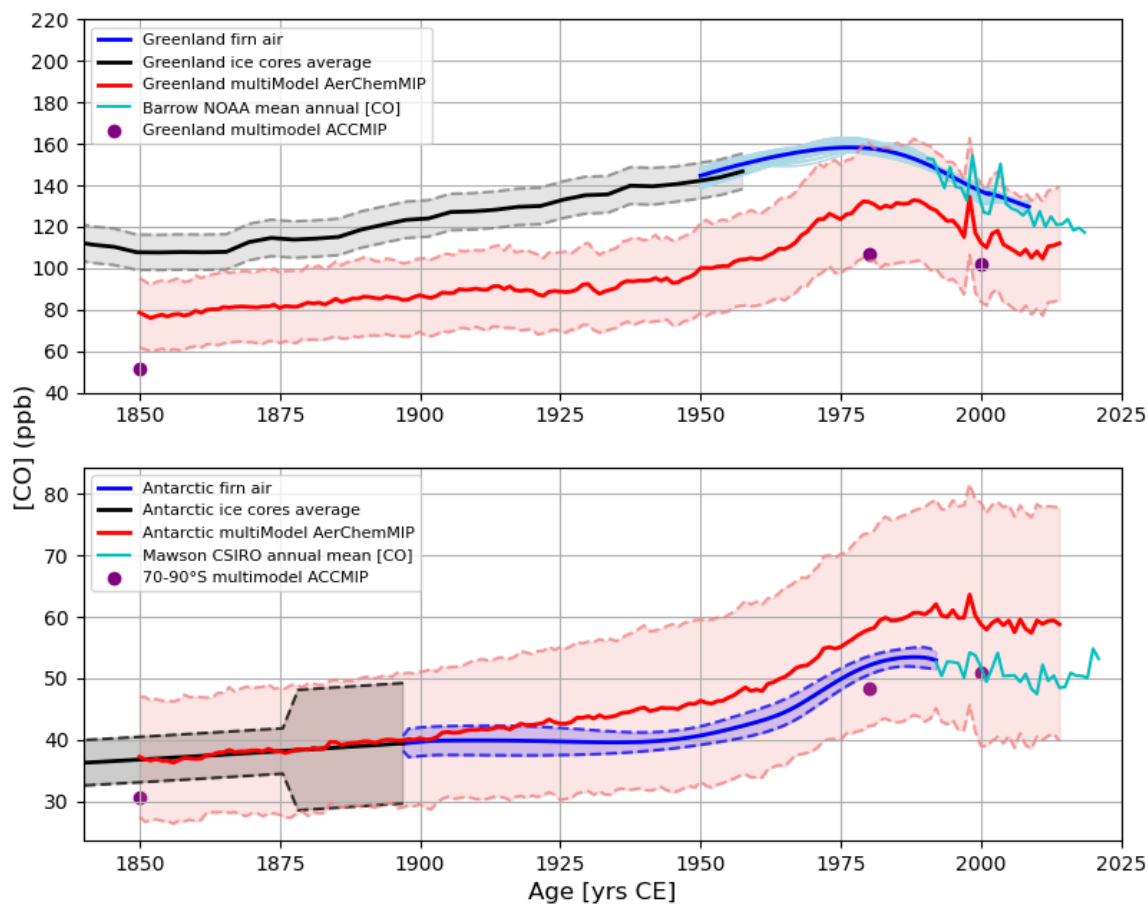
125   previously published Antarctic [CO] datasets (Haan et al., 1996; Haan and Raynaud, 1998; Wang et al., 2010) were likely biased high. The [CO] reconstruction based on Antarctic ice archives is representative of the 45-90°S atmosphere, with temporal changes having probably a larger spatial significance, including at least the 30-90°S latitudinal band (Faïn et al., 2023).

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Figure 2: Greenland (upper panel) and Antarctic (lower panel) atmospheric [CO] from ice cores and firn air reconstructions (Faïn et al., 2023, 2022; Petrenko et al., 2013), from NOAA and CSIRO atmospheric monitoring ; and from Multimodel AerChemMIP historical simulations (red line), with the red envelope showing a 1-sigma uncertainty. The multimodel ACCMIP [CO] is also reported for 1850 and 1900.

3 The ACCMIP exercice

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).

3.1 Emission inventories

140 **3.1.1 CO sources**

Anthropogenic and biomass burning CO emissions used in the frame of ACCMIP are taken from the RETRO and EDGAR-HYDE inventories (Lamarque et al., 2010). Natural emissions for CO were specified from different sources depending on models

3.1.2 CO sinks

145 Sinks for atmospheric CO include oxidation with OH radicals and CO uptake via oxidation by soil microbes. Reaction with OH radicals acts as the major CO sink while deposition in soils contributes about 10% to the global atmospheric CO losses (Duncan et al., 2007; Khalil et al., 1999; Stein et al., 2014). ACCMIP models exhibit a large diversity in the magnitude and sign of PI to present-day OH changes (ranging from a decrease of 12.7% to an increase of 14.6 %). Despite large regional changes, the multi-model global mean (mass-weighted) OH concentration changes little over the past 150 yrs (Naik et al.,
150 2013). For the 1980 to 2000 period, ACCMIP models find a slight increase in mean OH (3.5±2.2 %) (Naik et al., 2013).

3.2. Model outputs

In this study, we extract historical CO mixing ratio outputs from the 16 ACCMIP models (Naik et al., 2013) for three time slices (1850, 1980, and 2000 CE) over two areas (i) Greenland (60-20°W; 84-60°N), and (ii) Antarctica (90-70°S). For each time slice, a single mutlimodel mean [CO] is obtained by averaging CO outputs of the 16 models. Figure 2 (resp. Fig. 3) reports
155 the ACCMIP multimodel CO absolute levels (resp. the anomaly relative to the 2000 CE) for both Arctic and Antarctic atmosphere (in ppb).

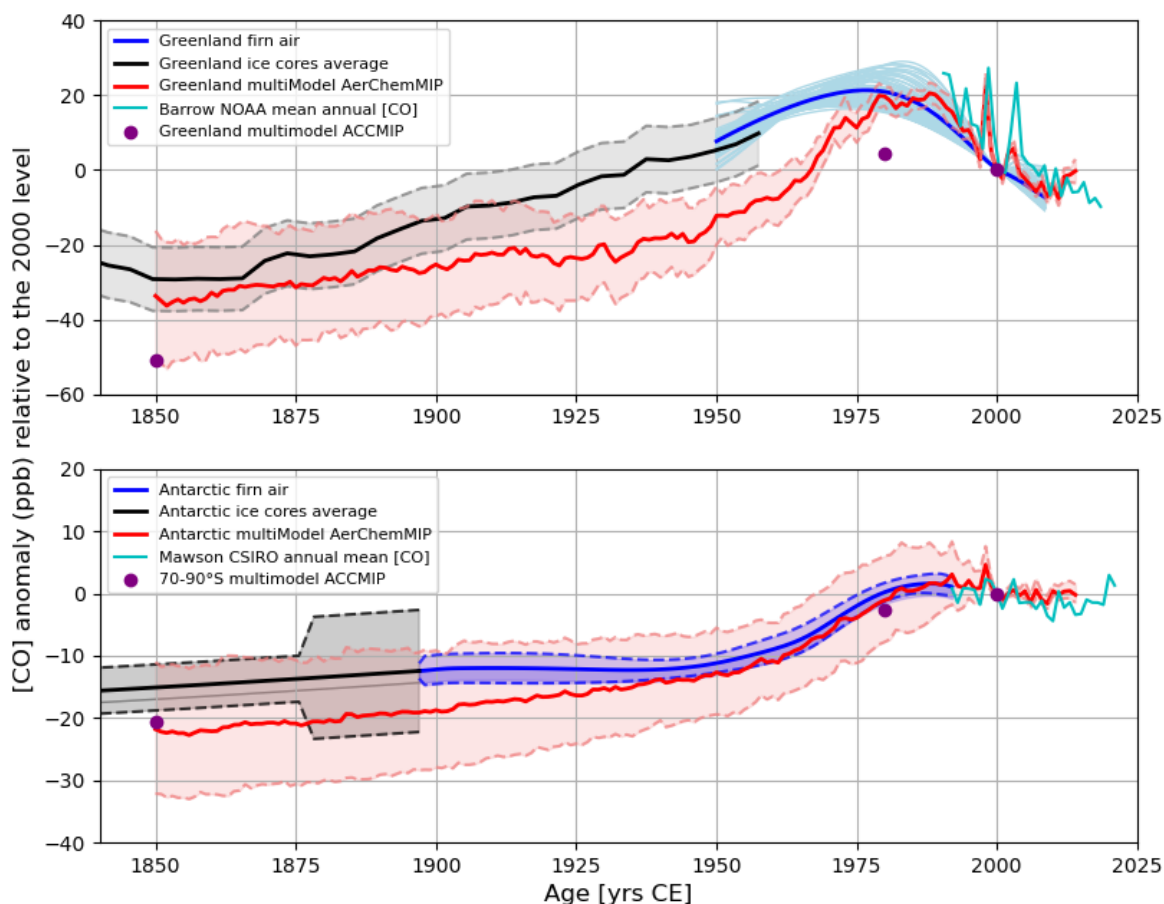



Figure 3. Anomaly of Greenland (upper panel) and Antarctic (lower panel) atmospheric [CO] from ice cores and firn air
160   **reconstructions (Faïn et al., 2023, 2022; Petrenko et al., 2013), from NOAA and CSIRO atmospheric monitoring ; and from**
Multimodel AerChemMIP historical simulations (red line), with the red envelope showing a 1-sigma uncertainty. The multimodel
ACCMIP [CO] anomaly is also reported for 1850 and 1900. All data are reported as anomalies relative to annually-averaged 2000
levels.

4. The AerChemMIP exercice

The Aerosol Chemistry Model Intercomparison Project (AerChemMIP) is part of CMIP6 and aims to quantify the climate and
air quality impacts of aerosols and chemically reactive gases, including CO (Collins et al., 2017). In this study, we use the
historical (1850-2014) CO mixing ratios outputs from six models involved in the AerChemMIP exercice: UKESM1-0-LL
(Archibald et al., 2020), GFDL-ESM4 (Dunne et al., 2020; Horowitz et al., 2020), CESM2-WACCM (Emmons et al., 2020),
170   BCC-ESM1 (Zhang et al., 2021), GISS-E2 (Bauer et al., 2020) and MRI-ESM2 (Yukimoto et al., 2019). No criteria were

applied to select specific models, and we included in this study all CO modeling results available on the ESGF datasets repository at the end of December 2021.

4.1. AerChemMIP CO budget

4.1.1. CO sources

175 Anthropogenic and biomass burning CO emissions used in the framework of AerChemMIP have recently been produced specifically for CMIP6 for the 1750-2014 CE period, and are common to all models. Anthropogenic CO emissions are reported by Hoesly et al. (2018) and include aircraft, energy production, industry, residential commercial and others, shipping, transportation on land, and waste (including waste burning). Open burning CO emissions are based on merging satellite observations with proxies and fire models (van Marle et al., 2017).

180 Biogenic sources are not constrained by the AerChemMIP protocol. Biogenic VOC (BVOC) emissions are interactive: CESM2 and GFDL models use the same emission model (MEGAN, Guenther et al., 2006), while UKESM1 applies a different model (iBVOC, Pacifico et al., 2011). The direct CO emission from biogenic sources (e.g., plant leaves) is low, but the emissions of biogenic VOCs (BVOCs), later oxidized into CO, is a large source of atmospheric CO (Fig. 1).

For all models, CH_4 mixing ratios are prescribed at ground levels using ice core reconstructions (Meinshausen et al., 2017).

185 CH_4-oxidation CO source is thus well constrained in terms of trend but can vary accordingly to absolute OH values differing from one model to another.

4.1.2. CO sinks

AerChemMIP models simulate stable global OH from 1850 to 1980, and a 10% increase in global OH since 1980, indicating that no changes in the major CO sink are modeled prior to 1980 (Stevenson et al., 2020). However, spatial distribution of the

190 OH radical, which differs among models (Stevenson et al., 2020), can have a large impact on the main CO sink (Strode et al., 2015).

Although much smaller, the CO sink driven by dry deposition into soils is included in the AerChemMIP models (e.g., Archibald et al., 2020; Emmons et al., 2020; Horowitz et al., 2020). However, this CO sink remains poorly constrained as little focus has been placed so far on the role of CO dry deposition in global modeling (Stein et al., 2014).

195 ### 4.2. Model outputs

In this study, we extract historical (1850-2014) CO mixing ratio outputs from the 6 AerChemMIP models mentioned earlier over the same two areas already defined for ACCMIP simulation (Sect 3.2): (i) Greenland (60-20°W; 84-60°N), and (ii) Antarctica (90-70°S). For each model, we first obtain a single timeseries by averaging all members available on the ESGF datasets repository at the end of December 2021. For each area (Greenland and Antarctica), model outputs are thus combined

200 to produce (i) a multimodel history of absolute [CO] (Fig. 2, red line), and (ii) a multimodel [CO] trend defined as [CO]

anomaly relative to the 2000 CE [CO] level (Fig. 3, red line). The envelopes reported on Fig. 2 and 3 (1-σ) represent the variability in simulated [CO] among the 6 models and can be interpreted as the uncertainty in how state-of-the-art CCMs can simulate historical [CO]. Individual model outputs are reported on Fig. S1 (Greenland) and Fig. S2 (Antarctica). Interestingly, while differences in transport dynamics and chemical schemes (e.g., Shindell et al., 2006) drive a wide range of CO burdens

205   among models, they do not drive a large diversity in modeled trends (Fig. S1 and S2). One of the 6 models, the BCC-ESM1 model, exhibits a larger CO burden, but excluding this "outlier" model has little effect on the multimodel mean results.

5. New insights into past CO budget

In this section, we compare observations (i.e., historical reconstructions retrieved from ice archives, Sect. 2) with ACCMIP and AerChemMIP multimodel outputs (Sect. 3 and 4), at both high northern and southern latitudes, to infer new constraints on

210   past CO sources and sinks.

5.1. Model bias of [CO] at high latitudes

We observe discrepancies in absolute [CO] values between model output and ice archive datasets (Fig. 1). AerChemMIP model mean is about 20% lower than ice archive datasets at high northern latitudes along the entire 1850-2014 period. At high southern latitudes, AerChemMIP model mean is in good agreement with ice archive dataset during the period spanning 1850-

215   1900, with a positive bias slowly increasing from 1900 to 1970, to reach modeled [CO] about 20% higher than ice core archives during the last three decades. Such discrepancies also exist for the recent two decades between surface ground observations (e.g., at Barrow AK and South Pole Antarctica, Fig. 1). However, AerChemMIP model mean agrees with the ice core record at Antarctica within its 1-σ variability envelope, and we thus focus here on the NH [CO] model bias.

Negative bias in the AerChemMIP model mean [CO] at northern mid to high latitudes (Fig. 1) is consistent with systematic

220   underestimation of observed [CO] at these latitudes, but the hypotheses to explain such bias vary between studies. Missing anthropogenic emissions from the inventories are commonly hypothesized to explain the negative CO bias (e.g., Emmons et al., 2020; Heimann et al., 2020; Miyazaki et al., 2020; Shindell et al., 2006; Stein et al., 2014). However, underestimation of the secondary CO production from chemical oxidation of methane or VOCs could also be involved (e.g., Archibald et al., 2020; Gaubert et al., 2017; Heimann et al., 2020), as well as CO losses via OH oxidation (Archibald et al., 2020; Strode et al.,

225   2015) or soil deposition (Stein et al., 2014). Recent studies have suggested mitigating the negative CO bias in the NH by assimilating multiple datasets of chemical observations (e.g., Gaubert et al., 2020; Miyazaki et al., 2020; Zheng et al., 2019). The causes of CO negative biases at northern mid to high latitudes in CCMs remain highly debated, and targeted sensitivity experiments are missing for bias attribution. However, how models represent absolute [CO] at northern latitudes has improved over the last decade, as shown by the reduction in negative bias in model mean [CO] between the ACCMIP and the

230   AreChemMIP exercises (Fig. 1). Furthermore, not all models exhibit a negative bias at high northern latitudes, for e.g. the

UKESM1 model accurately simulates the absolute [CO] observed at Barrow (AK) over the last 3 decades, or the BCC-ESM1 model shows a positive bias (Fig. S1).

5.2. Global decline in atmospheric [CO] since 1980 CE

Over the period spanning 1980-2010 CE, trends in observations and AerChemMIP multimodel outputs are in excellent

235   agreement at both northern and southern high latitudes (Fig. 3). Trends in atmospheric [CO] are defined here as variations in [CO] relative to the year 2000 CE. Trends are almost identical between observations (-0.85 ppb yr-1) and models (-0.80 ppb yr-1) in the NH during this period. Isotopic CO analysis conducted on Greenland firn air suggest that the decline in Arctic [CO] between the peak in the 1970s and 2000 is due almost entirely to reduced fossil fuel emissions, specifically reductions from road transportation (Wang et al., 2012). Over the recent decades, AerChemMIP models also successfully capture at

240   different latitudes the amplitude and phase of the [CO] seasonal cycle as well as the magnitude of interannual variability in atmospheric [CO] revealed by direct atmospheric monitoring (Archibald et al., 2020; Emmons et al., 2020; Horowitz et al., 2020). Figure 3 reveals how AerChemMIP models reproduce interannual variability in Arctic [CO], driven by biomass burning (e.g., during summer 1998 CE). Such variability is not captured by ice archives due to the temporal smoothing introduced by gas diffusion in polar firn.

245   Recent decades are when surface emissions of CO are the best constrained (Hoesly et al., 2018). NMVOCs emissions are also precisely quantified (Hoesly et al., 2018), and atmospheric [CH$_4$] is prescribed using ice core records (Meinshausen et al., 2017), thus CO secondary production is also well known. The good agreement in the trends between model outputs and observations suggest that the main CO sink (OH oxidation) is also accurately represented within models. AerChemMIP models report stable global levels of tropospheric OH from 1850 to 1980 CE, and a ~10% increase in OH from 1980 to 2014 CE (Fig.

250   2a from Stevenson et al., 2020). The good agreement in the trends between models and observations reported here lends credibility to the OH trend simulated by AerChemMIP models. It also suggests that the recent increase in anthropogenic NOx emission is well understood.

Figure 3 also includes a comparison between AerChemMip and ACCMIP modeling outputs, with ACCMIP temporal resolution being much more limited. The ACCMIP multi-model simulation of the Arctic [CO] is only 4 ppb higher in 2000

255   CE compared to 1980 CE (Naik et al., 2013). ACCMIP models were not able to reproduce the decline in Arctic [CO] reconstructed for the last decade. Petrenko et al. (2013) already reported a poor agreement between the trends from a firn air [CO] record and a CAM-Chem historical run, which was part of the ACCMIP exercise. One possible reason for such improvement in the framework of AerChemMip are the anthropogenic emissions, dominant over the last decades, which are better constrained now (Hoesly et al., 2018).

260   ## 5.3. Increase in Northern Hemisphere atmospheric [CO] from 1850 to 1980 CE

Along the time period spanning 1850-1980 CE, both AerChemMIP models and observations exhibit increasing [CO] (Fig. 2 and 3). At northern latitudes, the increase in [CO] simulated from 1850 to 1980 CE is ~55 ppb, in excellent agreement with

observations that reveal a ~50 ppb increase for this period. NH simulated and observed trends in [CO] agree within their 1σ
uncertainties prior to 1920 but diverge during the 1920-1975 CE period. While the ice core and firn based record exhibit an
265 increase in [CO] at a steady rate of ~0.5 ppb yr-1, the AerChemMIP multimodel average reveals slow growth from 1920 to
1945 at a rate of ~0.2 ppb yr-1, which quickens from 1945 to 1975 with a rate of ~1.1 ppb yr-1. At southern latitudes,
AerChemMIP models simulate a larger increase in atmospheric [CO] from 1850 to 1980 CE than observations (22±10 ppb
and 13±7 ppb, respectively), although model outputs and ice archive datasets always agree within their 1σ uncertainties. Both
models and observations also reveal an increase in [CO] growth rate in the SH, which is 2 times larger during 1945-1980 CE.
270 Finally, over the time period spanning 1850-1980 CE, the [CO] trends simulated by ACCMIP datasets are similar to the one
produced in the frame of AerChemMIP (Fig. 3). For this period, a key improvement of AerChemMIP compared to ACCMIP
is the annual resolution of the model runs.

The general agreement between simulated and observed [CO] trends, both in the NH and SH, for the period spanning 1850-
1980, indicate a good overall understanding of the evolution of the CO budget over this period. However, a mismatch in [CO]
275 trends remains between the AerChemMIP multimodel mean and the ice archive dataset in the NH for the period spanning 1920
to 1975 CE, with the multimodel mean showing lower trend before 1950, and higher trend after 1950 (Fig. 3). The CO record
based on Greenland ice archives should be considered as an upper bound of the past CO abundance, and we can not fully rule
out that in situ production artifact (Faïn et al., 2022) impacting ice and firn air samples explain such mismatch (Sect. 2).
However, the Greenland ice archives record is a combination of 5 ice core and 3 firn air records, which all support an increase
280 in [CO] at a steady state during the 1920-1975 CE period (Faïn et al., 2022). We thus investigate if uncertainties in descriptions
of CO sources and sinks used by AerChemMIP models can explain the differences in [CO] growth rate between models and
observations for the period spanning 1920-1975 CE.

5.3.1. OH-oxidation CO sink

A first hypothesis to explain the differences in trends between ice archive and model [CO] in the NH for the period spanning
285 1920-1975 could be an inaccurate estimation of the OH-oxidation CO sink by models. AerChemMIP simulations suggest that
global tropospheric hydroxyl radical changes little from 1850 to 1980 CE (Stevenson et al., 2020). Global OH reconstructions
are not easily comparable with Greenland [CO] reconstructions because OH is more abundant in the tropics whereas the
latitudinal band that is of most importance for polar CO is considerably narrower, likely ~30–90°, because of the relatively
short CO atmospheric lifetime. Ozone precursor (NOx, CO, and VOC) emissions have strongly influenced OH trends,
290 specifically the relative roles of changing emissions of CO and NOx have important competing consequences. Higher
concentrations of NOx increase OH through ozone photochemical production and the subsequent reaction with H_2O to produce
OH. On the other hand, higher concentrations of CO and VOCs (and CH_4) will reduce OH. Stevenson et al. (2020) report a
dominant role of NOx emission increases, whose impact overwhelms the impacts of increasing CO (up to the 1990s) and
VOCs emissions, which will have tended to reduce OH. Finally, the OH-oxidation CO sink modelled in the framework of
295 AerChemMIP is closely related to NOx emission inventories. To reconcile modeled and ice core-based NH CO trends, a

decrease in the OH-oxidation CO sink would be required during the period spanning 1870-1945 CE, which would mean lower NOx emissions. The CMIP6 NOx emissions remain low before 1945 CE (Fig. 1 in Stevenson et al., 2020). Considering the period spanning 1945-1980 CE, an increase in the OH-oxidation CO sink would be required to reconcile modeled and observed [CO] growth rates. Overall, if an inaccurate estimation of the OH-oxidation CO sink by models was the cause of the mismatch

300 between observed and modeled NH [CO] trend, it would imply centennial variation in global OH, with unlikely low OH levels in the early XXth century. A diversity of models is involved here (i.e. diversity of chemical schemes) and the drivers of variability in global OH can be very different between models (Wild et al., 2020). However, most CCMS models show buffered OH at global scale (Stevenson et al., 2020), and our comparison between modeled and observed CO record supports this conclusion.

305 ### 5.3.2. CO secondary production

If we hypothesize that the past evolution of OH levels is well understood for the period spanning 1920-1980 (Stevenson et al., 2020), CO sources should be investigated to explain the mismatch between modeled and observed CO over the time period 1920-1975 CE: secondary production and/or direct emissions may be involved. Secondary production represents in the modern atmosphere about 50% of the CO sources (Fig. 1, Szopa et al., 2021), and is largely driven by methane oxidation. CEDS

310 anthropogenic global methane emissions exhibit an 8-fold increase from 1850 to present (Stevenson et al., 2020), but for all AerChemMIP simulations atmospheric CH_4 levels are prescribed at the surface based on observations and ice core data (Meinshausen et al., 2017). Thus atmospheric CH_4 is well constrained, and the CH_4-oxidation CO source is accurately quantified by models. On the other hand, VOCs-oxidation represents only a small fraction of the modern CO sources (~10%, Fig. 1; Fortems-Cheiney et al., 2012). AerChemMIP models suggest no large changes in BVOCs emissions for the 1850-1980

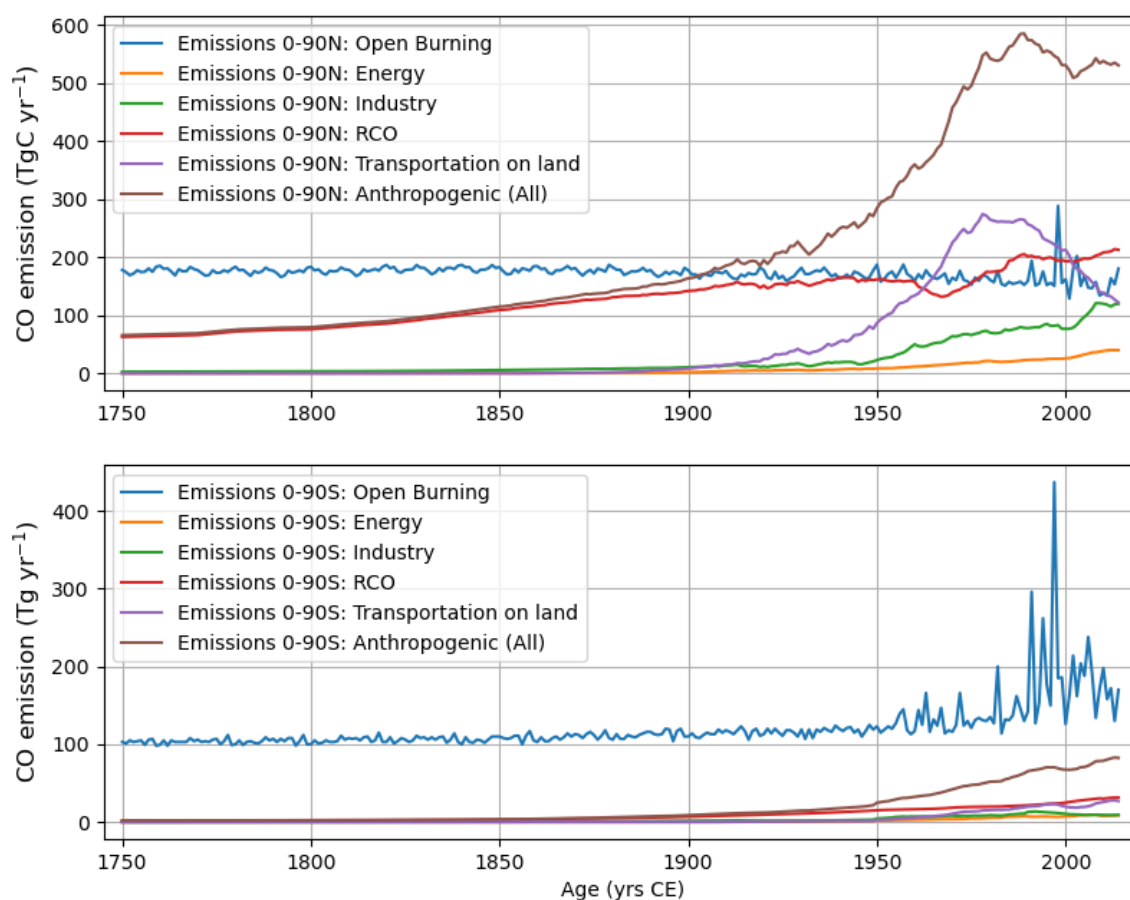
315 CE period (Griffiths et al., 2021). Using the dynamic global vegetation model LPJ-GUESS, Hantson et al. (2017) find that both isoprene and monoterpene, which represent the largest fraction of BVOCs, had higher emissions at the beginning of the 20th century than at present. AerChemMIP models suggest an increase in the growth rate of anthropogenic VOCs (AVOCs) emissions in the 1950s (Stevenson et al., 2020). However, AVOCs oxidation represents a minor source of CO and an underestimation of the AVOCs emission prior to 1950 CE is unlikely to explain the differences in modeled and ice archive

320 [CO] NH trends (Fig. 3).

5.3.3. CO Anthropogenic emissions

In the period spanning 1850-1975 CE, the multimodel mean of Arctic [CO] (Fig. 3) evolves with a similar rate as its direct emissions in the 30-90°N latitudinal band (Fig. 4). This direct emissions pattern is driven by change in fossil fuel emissions

325 (purple line on Fig. 4; Hoesly et al., 2018), as CMIP6 CO fire emissions exhibit little variation since PI times (brown line on Fig. 4; van Marle et al., 2017). Prior to 1945 CE, direct anthropogenic CO emissions are largely dominated by the RCO (residential, commercial, others) sector. In 1945 CE, CO emissions related to transportation start increasing, dominating other emissions sectors, including RCO (Fig. 4). Combustion of fossil fuels drives direct CO emissions, but also CO_2 emissions.

CO_2 atmospheric levels, evaluated by multiple ice core and firn records as well as direct atmospheric monitoring, reveals a
330   sharp increase in growth rates in ~1945 CE (Meinshausen et al., 2017; Rubino et al., 2019). Such a pattern demonstrates that
the volume of fossil fuel annually burnt abruptly increases after WWII. The atmospheric CO_2 record brings a strong constraint
on fossil fuel combustion inventories. On the other hand, Hoesley et al. (2018) report that anthropogenic CO EF prior to 1970
are not well constrained. Combustion EF for CO are held constant before 1900 at values drawn from a literature review
(Winijkul et al., 2016), and linearly interpolated between 1900 and 1970 by Hoesley et al. (2018). Consequently, we suggest
335   that the mismatch in [CO] trends observed between the AerChemMIP multimodel mean and the ice archive dataset in the NH
for the period spanning 1920 to 1980 CE (Fig. 3) may be related to uncertainties in CO emission factors (EF), specifically EF
for the RCO and transportation sectors which drive the direct CO emission pattern during the 1920-1980 CE period.

340   **Figure 4. CEDS anthropogenic emission estimates for the 30-90°N (upper panel) and 30-90°S (lower panel) latitudinal bands by
aggregate sectors (Hoesly et al., 2018): energy, Industry, RCO, and transportation. "RCO" stands for residential, commercial, and
other. "Fossil fuel combustion" combines all sectors listed before, and small contributions from the waste and shipping sectors (not
shown separately). Open burning emissions from the 30-90° latitudinal bands are from van Marle et al. (2017).**

5.4. Biomass burning emissions in the Southern Hemisphere since preindustrial times

Evolution of biomass burning emission inventories since PI times have been recently subject to debate. Biomass burning emission inventories, such as the CMIP6 inventories, commonly scale up fire emission with population (e.g., van Marle et al., 2017; van der Werf et al., 2013), suggesting lower biomass burning emission in PI compared to present day. On the other side, hypothesizing a decline in burned areas with increasing population density due to land use changes (Andela et al., 2017; Knorr et al., 2014) lead to biomass burning reaching higher levels in the PI compared to present day (e.g., Hamilton et al., 2018; Liu et al., 2021; Rowlinson et al., 2020). Notably, Liu et al. (2021) concluded that fire emissions remained relatively stable in the SH between 1750 and ~1920 CE followed by a 30% decrease until about 1990 CE. These authors support this conclusion by a comparison of simulated black carbon (BC) deposition fluxes with BC measurement conducted on an array of ice cores (14 Antarctica records, and one record from the Andes).

Our SH [CO] reconstruction from ice archives (Fig. 3b) do not show any decreasing trend in [CO] between 1850 and 1992 CE. Instead, atmospheric CO in the southern hemisphere exhibits an increase closely reproduced by AerChemMIP models. If the biomass burning inventories used in the frame of CMIP6 (Fig. 4b) were inexact, as suggested by Liu et al. (2021), this would mean that anthropogenic CO emissions (Fig. 4) would also be inexact (i.e., underestimated), compensating for an overestimation of CO fire emissions. This seems unlikely. Our data also disagree with the elevated [CO] levels reported previously by Wang et al. (2010) which concluded that biomass burning CO emissions peaked during the late 19th century at rates roughly 3 times modern levels (see Faïn et al., 2023, for a detailed comparison with the Wang et al. CO dataset). Finally, both models and observations reveal a [CO] growth rate in the SH a factor of 2 larger during the 1945-1980 CE time period. The modeled trend is mostly driven by an increase in anthropogenic transportation emissions (Fig. 4), and the Antarctica ice archives suggest that such CO anthropogenic emissions are well constrained.

6. Summary and conclusion

We have reported the first bipolar comparison of atmospheric CO mixing ratio extracted from Greenland and Antarctic ice archives with multimodel ACCMIP and AerChemMIP historical ensembles, for a period spanning 1850 to present days. Specifically, the ice archive [CO] records are composites based on multiple ice core and firn air records, respectively 8 and 10 datasets for Greenland and Antarctica (Faïn et al., 2023, 2022; Petrenko et al., 2013). We used historical CO mixing ratio outputs from the 16 ACCMIP models (Naik et al., 2013) for three time slices (1850, 1980, and 2000 CE), and from 6 models involved in the AerChemMIP exercise (Collins et al., 2017) for the historical period spanning 1850-2014 CE at annual resolution.

Both ACCMIP and AerChemMiP model ensembles present negative CO biases at high latitudes, particularly in the northern hemisphere, which are still poorly understood and could involve missing anthropogenic emissions, underestimation of the

375   secondary chemical production of CO, or excessive CO losses via OH oxidation. However, the model CO bias at high northern latitudes has decreased by ~50% between the ACCMIP and AerChemMiP exercises.

We used the atmospheric [CO] from Greenland and Antarctica ice archives as a benchmark for ACCMIP and AerChemMiP ensembles allowing to assess together the global chemistry-climate models and the emission datasets they used. Over the period 1980-2010 CE, CO trends from ice archives and AerChemMIP multimodel outputs are in excellent agreement at both

380   northern and southern high latitudes. Such agreement suggests that the trends in surface emissions of CO (including anthropogenic emissions), as well as in the main CO sink (OH oxidation), are now accurately represented within models.

Over the 1920 to 1975 CE, a mismatch in [CO] trends remains between the AerChemMIP multimodel mean and the ice archive dataset in the NH, with the multimodel mean showing lower trend before 1945, and higher trend after 1945. Although Greenland ice archive record should only be considered as an upper bound of the past CO abundance, as we can not fully rule

385   out in situ production artifacts (Faïn et al., 2022), we suggest that the mismatch between modeled and observed CO trends at high NH latitudes is related to uncertainties in anthropogenic CO emission factors, specifically EF for residential commercial and other, and transportation sectors. Our study indicates that a better evaluation of these EFs would be an area of improvement for modeling of the past NH [CO]. Finally, SH atmospheric [CO] exhibits an increase closely reproduced by AerChemMIP models, supporting the biomass burning inventories used in the frame of CMIP6 (van Marle et al., 2017). The hypothesis of

390   higher fire emissions at the onset of the XXth century (e.g., Liu et al., 2021) seems unlikely in the light of our results.

Although imperfect, state of the art global chemistry-climate models now exhibit good capabilities in representing trends in atmospheric [CO] at high latitudes. Further modeling sensitivity studies would provide a better understanding of the nature of the CO bias at high NH latitudes. Also, isotopic information can help to determine the various CO sources and their relative magnitudes (e.g., Röckmann et al., 2002). Including isotope data in modeling atmospheric chemistry schemes has been shown

395   to help constraining source strengths of CO (Park et al., 2015). Thus, more measurements of the stable isotopes of CO from firn and ice cores would help constrain relative CO source strengths in the past. This work offers a new constraint to better characterize emissions factors over the past century and thus could contribute to a revision of past tropospheric ozone or methane lifetime evolutions. On the other hand, new measurement-derived CO datasets from high-resolution glacial archives would enable to characterize decadal variations in atmospheric CO, and potentially contribute in constraining further CO

400   evolution of emissions and sinks during the industrial period. Improvement in the assessment of past OH trends (e.g., using innovative proxies such as ^{14}CO, Brenninkmeijer et al., 2022) would also provide powerful constraints on the past evolution of the main CO sink.

Competing interests

405   The contact author has declared that none of the authors has any competing interests.

Acknowledgements

The authors would like to thank all the drillers, field crews and scientists who participated in the collection, analysis and interpretation of ice core and firn air samples for CO, and who helped produce the CO dataset included in this study. We acknowledge the World Climate Research Programme, which, through its Working Group on Coupled Modelling, coordinated and promoted CMIP6, AerChemMIP, a CMIP6 endorsed MIP. We thank the climate modelling centers for producing and making available their model output, the Earth System Grid Federation (ESGF) for archiving the data and providing access, and the multiple funding agencies who supported CMIP6 and ESGF. We are also grateful to ACCMIP, an IGAC project under the International Geosphere-Biosphere Project (IGBP) and World Climate Research Program (WCRP), and the British Atmospheric Data Centre (BADC), which is part of the NERC National Centre for Atmospheric Science (NCAS), for collecting and archiving the ACCMIP data.

Financial support

This work has been supported by the French ANR projects RPD-COCLICO (grant no. 10-RPDOC-002-01; Xavier Faïn) and the French national program LEFE/INSU (project GreenCO, Xavier Faïn); by the EU FP7-IP (grant no. ENV-2010/265148; project Pegasos; Xavier Faïn) and FP7 ERC (grant no. 291062; project Ice and Lasers; Jérôme Chappellaz); Rachael H. Rhodes has received funding from the Isaac Newton Trust (grant no. LBZG/080).

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