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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The isotopic record of Northern Hemisphere atmospheric carbon monoxide since 1950, implications for the CO budget" *by* Z. Wang et al.

Anonymous Referee #2

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Overview This study presents measurements of [CO] and its stable isotope (d13C and d18O) from Greenland firn data. The authors reconstruct the northern high latitude records of [CO], d13C , and d18O and use an isotope mass balance model to infer changes in CO sources from 1950 to present. The authors find the most plausible explanation of their data is a reduction in CO emissions from fossil fuel burning due to the adaptation of catalytic converters starting in the mid-1970s which coincided with the reductions in leaded gasoline. The idea that CO emissions have declined due to catalytic converter use is not entirely new but this work provides important new data to reexamine this question. My comments mainly address the modeling aspect of the work.

Comments Authors reconstruct the fossil fuel and NMHC oxidation source history. Bottom-up inventories of both of these sources have large uncertainties so using a top-down constraint is needed. Inventories of CO emissions from biomass burning also have large uncertainties- thus there are three potential sources the authors could constrain. The limitations of the data mean only two can be solved for. Authors should motivate why they chose to solve for fossil fuels instead of biomass burning. Presumably this is because the authors believe the biomass burning is better known? If so, authors need to argue this.

How sensitive is the trend of derived fossil fuel [CO](Fig 7c) to trends in biomass burning emissions? Since our knowledge of biomass burning trends is poor is it possible that the [CO]ff trend could become statistically insignificant with a reasonable biomass burning trend? Sensitivity tests could be performed to test this.

Other authors have postulated that CO emissions from fossil fuel burning should have decreased when catalytic converters started to become popular but I see no mention of this literature in the paper. Authors should provide appropriate background and references.

In addition to providing the modeled [CO] partitioning for CO sources (Fig 7) it would be useful if the authors could provide CO fossil fuel emissions consistent with the [CO]ff record.

The authors' fossil fuel [CO] is about double that of [CO] derived from CO measurements and MOZART simulations (Fig 7c). This is a large difference. What is the authors' explanation as this lies outside the uncertainty envelope?

p30635, line 15 – Regarding the inversion, how do authors define "simplest solution"? For example, continuous first and second derivatives of the temporal trend?

p30636, line 18 – Presumably the reconstruction can capture seasonality but not seasonality with large amplitudes ?

p 30640, line 2- Explain why source signatures are different at different latitudes. Because of differences in d18O of O2? Or do these reflect the fractionation of CO after under going some oxidation by OH during transport?

p 30640, line 28 – Authors state that both source d18O signatures and [CO] were calculated with MOZART, but presumably d18O source signatures are determined from measured literature values.

p30645, line 18 – Authors remark that studies by Larmarque et al. 2010 and van Aardenne et al. 2001 indicate that CO emissions from fossil fuel have increased through 1990 but authors work suggests the opposite. Authors should attempt to provide explanation. For example do these studies use constant CO emissions factors for fossil fuel burning? If so, they are not taking into account the use of catalytic converters which is an important difference.

p30641, line 24 – Is this a gridded inventory? Specify spatial resolution. Have authors tested to see how sensitive their results are (i.e. [CO]ff trends to changes in the distribution of biomass burning emissions?

p30641, line 26 – Not clear how or why the modeled biomass burning at Iceland is scaled. Why not just use the biomass burning contribution to [CO] directly at the Greenland site? And why just for 1997-2004? Is that time of atmospheric measurements at Iceland? If so, how are measurements used to tease out the biomass burning contribution?

p30643, line 12 – Since the d18O data is used to determine the fossil fuel combustion, not vice versa, the causal relation in this sentence is inverted.

p30643, line 17 – Not too much needs to be said, but would be good to put this finding – no change in NMHC oxidation – into some context. Is it consistent with the literature?

p 30643, line 24-26 – To my eye from the graph (Fig 4) it appears that d18O is actually increasing (more enriched) from 1950 to the mid-1970s.

Editorial remarks Font size of all text in figures should be increased. p30629 line 2 – Replace “frame” with “framework”

p30633 line 16 – Remove “in”; line 21 – Remove “to”

p30634 line 1 – Start sentence with “The”

p30636 line 15 – Replace “until” with “up to”

p30646 line 22 – Remove “contributing to”

Fig2 caption – [CO] is plotted using green triangles not squares.

Fig5 caption – Grey lines are on left panels not right.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 30627, 2011.