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Comment on acp-2021-736

Anonymous Referee #2

Referee comment on "Towards reconstructing the Arctic atmospheric methane history over the 20th century: measurement and modelling results for the North Greenland Ice Core Project firn" by Taku Umezawa et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-736-RC2, 2021

Umezawa et al. used a suite of gas measurements from NGRIP firn air (CO2, CH4, SF6, CH3CCl3, CFC-11, CFC-113, and CFC12) in combination with a firn model to reconstruct the atmospheric history of CH4 in the northern hemisphere (NH). Although the firn air samples were collected close to 20 years ago (in 2001), a great care has been taken to use state-of-the-art (or close to state-of-the-art) measurement techniques to achieve analytical precisions that are comparable or better than present-day modern atmospheric measurements. This is not a trivial merit and I think the authors should be commended. Following precedents set by previous studies of firn air (e.g., Rommelaere et al., 1997; Trudinger et al., 2002; Witrant et al., 2012; Buizert et al., 2012), Umezawa et al. used a forward gas transport firn model that takes in a "known" atmospheric history of a certain gas as an input and produce the expected mole fraction of that gas vs. depth profile in the open porosity of the firn. The difference between the expected mole fraction depth profile vs. measurements is then used to tune the "effective diffusivity" for this particular firn air sampling borehole (which is the Japanese firn sampling borehole at NGRIP).

A previous study by Buizert et al. (2012) set a precedent by including CH4 as part of the suite of gases used to tune the effective diffusivity at the NEEM ice core site. Buizert et al. (2012) achieved this by first making an educated guess about the "known" atmospheric history of CH4 in the NH. However, in this study Umezawa et al. challenge this assumption, treat the NH atmospheric history of CH4 as an unknown, and only used the other six gas measurements (CO2, SF6, CH3CCl3, CFC-11, CFC-113, and CFC12) to tune the effective diffusivity profile for NGRIP. As a result, the atmospheric CH4 history reconstructed by Umezawa et al. has larger uncertainties; from this, Umezawa et al. argue that we cannot take the NH CH4 history for granted as a known variable to tune effective diffusivity profile for ice cores collected in the northern hemisphere and to certain extent, we also do not know the true atmospheric history of NH CH4 before the 1970s.

The main conclusion from of Umezawa et al. study (to which precision do we know the NH

atmospheric history of CH4) is potentially an important one, so I would recommmend the manuscript for publication if the following comments are sufficiently addressed.

Major comments:

1. As reviewer #1 pointed out, it is not immediately clear whether the atmospheric histories for the other six gases outside of CH4 (CO2, SF6, CH3CCl3, CFC-11, CFC-113, and CFC12) used to tune the effective diffusivity profile are sufficiently known as well. Why focus on CH4 and not say, the uncertainty on NH CO2 history? I think a discussion or even a specific section addressing this question is warranted. Fortunately, given the current state-of-science knowledge, I think Umezawa et al. should be able justify their assumption in using CO2, SF6, CH3CCl3, and CFCs to tune effective diffusivity profile. Meinshausen et al. (2017) took a great care in synthesizing all available data from historical atmospheric measurements, firn and ice cores from several sites to best reconstruct the GHGs (including CO2, CH4, SF6, CH3CCl3 and the CFCs measured by Umezawa et al.) mole fraction, interhemispheric gradient, and seasonal variabilities for the purpose of CMIP6 model runs. This would be a great starting point. The justification for treating the NH histories of CO2, SF6, and the CFCs as "known" parameters, or at least better known parameters than NH CH4 history in my opinion should revolve around a discussion about the interpolar gradients of these gases (which are relatively small owing to their long atmospheric lifetimes), but I will leave the exact formulation of this argument to Umezawa et al.

I think a sensitivity analysis comparing what mole fraction should we expect in the open porosity of NGRIP firn if we put in NH vs. SH history from Meinshausen et al. (2017) for CO2, SF6, and the halocarbons is warranted to further drive the point home. I might be wrong, but I would expect the mole fraction vs. depth profiles for these suite of gases in the firn open porosity would not be as sensitive to NH vs. SH difference, at least relative to their respective measurement precisions compared to CH4 given their long atmospheric lifetimes and relatively low interhermispheric gradient. Given Umezawa et al. already had their forward firn model setup, hopefuly this does not require a lot of additional work. Furthermore, as a more general comment, I would also recommend Umezawa et al. to use gas histories from Meinshausen et al. (2017) for their overall firn gas transport and effective diffusivity tuning because the GHGs histories proposed by Meinshausen et al. (2017) represent more updated, better-educated "guesses" than the gas histories previously used by Buizert et al. (2012).

2. The suite of CFCs measurements (CFC-11, CFC-113, and CFC12), CH3CCl3 and SF6 do not provide good constraints for reconstructing effective diffusivity for the deep firn just because the concentration of these gases are all very low and close to zero. Usually, the gases that are most useful to reconstruct the effective diffusivity in this firn region are CH4, CO2, and 14CO2 due to their respective unique atmospheric histories. 14CO2 is especially useful as its atmospheric history can be validated from tree rings and historical atmospheric measurements. Furthermore, 14CO2 has a unique profile from the "bomb pulse" in the 1950s that provides a strong and unique constraint on the effective diffusivity. Unfortunately, 14CO2 measurements for NGRIP are not available. Because the CH4 history in this study is treated as unknown, the effective diffusivity in the lower part of the NGRIP Japanese borehole presented by Umezawa et al. is almost solely constrained

by CO2 data. This made me question whether the conclusion obtained by Umezawa et al. regarding how we cannot accurately reconstruct NH CH4 history from firn air samples is a unique problem pertaining to NGRIP (and its suite of gas measurements) or is it more general problem to other Greenland ice core sites as well. I don't think the current version of the manuscript sufficiently answer this question and additional work might be warranted to justify the conclusion put forward by Umezawa et al.

In particular, I think it would be especially useful to revisit the NEEM data from Buizert et al. (2012) with the same firn model and iterative dating algorithm presented in this study, but also excluding CH4 as part of the suite of gases to tune the effective diffusivity of the NEEM site. This would provide a more fair comparison rather than putting in the atmospheric history reconstruction from likely underconstrained NGRIP site into NEEM with a forward firn model. It would be interesting to see whether additional constraints from 14CO2 data at NEEM will allow for reconstruction of NH CH4 history with a better uncertainty and to what extent the uncertainty is better. For this experiment, I would recommend using the updated "known" 14CO2 history from Graven et al. (2017). Given Umezawa et al. already had their firn model tuned for the NEEM EU borehole as part of their model validation, I don't think this extra calculation would require significant amount of additional work.

3. I think the uncertainty analysis/discussion regarding the conclusion is a bit lacking. It is not immediately clear to me whether conclusion reached by Umezawa et al., that NH CH4 history in general should be considered preliminary and should not be used to tune effective diffusivity is sufficiently justified. From the study, it is clear that reconstructing NH CH4 history from NGRIP firn air samples, when CH4 is excluded from the suite of gases used to tune the effective diffusivity result in large uncertainties. But I think we know the NH CH4 history slightly better than just the reconstructed history from NGRIP firn air presented in this study.

Meinshausen et al. (2017) decided against providing uncertainties to the reconstruction of GHG histories that they did, arguing that the CMIP6 models would not have the computational resources to run multiple scenarios and sensitivity analysis from multiple GHG histories on top of the envisaged SSPs. I think an assessment about the uncertainty of historical CH4 reconstruction is very valuable and Umezawa et al. is in a unique position to take a first attempt at this. How about reconstructing NH CH4 history from NEEM (with its additional 14CO2 constraint) like discussed above, how about combining NGRIP, NEEM history inversion results to make a best-estimate of NH CH4 history and its uncertainties, and how about including CH4 in the suite of gases used for effective diffusivity tuning, but through iterative method starting first with larger uncertainty for the RMSD calculation to account for uncertainty in the CH4 history? There are still many avenues to explore beyond the reconstructed NH CH4 history from NGRIP firn samples before one can conclusively claim that we don't know the NH CH4 history to such a degree that it should not be included in the suite of gases used to tune effective diffusivity in firn profiles. I don't demand Umezawa et al. to do all of the above, as it might constitute a whole different study entirely, but a preliminary exploration on this and an honest assessment about how well can we reconstruct the NH CH4 history would significantly strenghten the manuscript and provide very valuable insights to the community.

Minor comments:

I find that in general, the description about the firn gas transport models and the iterative method is very brief and might be bit hard to follow. The brevity is fine for the main manuscript, but the authors might want to consider a supplementary material where they will have more room to describe the gas transport model, iterative methods, and especially additional data treatments. For example

Line 212 "Effective age at each sampling depth was calculated..." Several steps are clearly skipped here. It is not immediately clear to me, from the description of the model and equations above how one can determine the effective age at each sampling depth, as all the description before this line only pertains to the forward firn model. Did Umezawa et al. calculated a depth-age transfer function similar to Rommelaere et al. (1997) or through other means? Either way this needs to be elaborated.

Fig.3. From the text it says "Figure 3 presents the initial simulations …" Does this mean this is the initial effective diffusivity profile? It might also be beneficial to have the other effective diffusivity profiles like Fig.5 shown in Fig. 3.

There are several data treatment steps that is missing/the authors did not explain in sufficient details, or if the authors didn't do it, it is not well justified why they choose not to. For example, in their supplementary material Buizert et al. (2012) discussed how they added additional uncertainties for CO2 to account for possible in situ production and bubble close-off fractionation. In Buizert et al. (2012), uncertainty in atmospheric histories is accounted during the tuning of effective diffusivity by running the uncertainties through the forward model when the tuning of effective diffusivity is near complete to transfer the uncertainties from time domain to depth domain. I might miss it somewhere, but I think it is not immediately clear to me how the uncertainties of "known" atmospheric gases used to tune the effective diffusivity is treated in this study.

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