

I feel obligated to ask the authors to redo the same revision again, as the revisions they make often miss the point and are incomplete. I was, and still am, quite enthusiastic about this paper and trust its results. But perhaps the authors took the positive reviews from both referees as license to perform very minimal edits to the manuscript (the editor had asked for a major revision).

Evaluating this revision has been made more challenging by the fact that the line numbers listed in the author response are incorrect (as in, the line numbers provided for the corrections do not actually match those in the text, neither the regular text nor the version with tracked changes). In other cases (such as their response to my point 3b) the authors claim to have made changes without actually doing so – the proposed “revised” text is already in the original.

All of this does not inspire confidence in the reviewer that their comments were considered carefully. The authors will need to submit another revision of this manuscript in which they engage more deeply with the actual substance of the original review comments. Below I add clarification in response to the author’s comments.

The following revisions have been made to the paper:

- 1) Only model results using model configurations that accurately capture neon enrichment are presented.
- 2) Tunu is excluded from the revised manuscript because there is no neon data from that site.
- 3) Instead of describing “alternative models” we instead use “model configurations” with different tuned pore close-off fractionation parameterizations.
- 4) We avoid physical justification for the pore close-off fractionation tunings (see below).
- 5) The model was updated and re-tuned to include backflow. The text and figures have been updated accordingly.

The final results (Figure 9) are essentially unchanged, but the revised methods are clearer, and the scientific reasoning is strengthened.

Regarding my original point (1)

The manuscript still feels schizophrenic. The authors agree that the “altered” scenarios are more realistic and that Figure 9 represents the best estimate of the atmospheric history of H<sub>2</sub>, yet most of the manuscript still discusses the regular UCI\_2 simulation as the main result and model solution. For example, the abstract (lines 22-26), and sections 4-6 still discuss the UCI\_2 scenario as if it were the main result of the paper.

As an additional note, it is confusing that the name of the firm model (UCI\_2) is identical to that of the baseline scenario. I would recommend renaming the scenario. The altered scenarios are also run on the UCI\_2 model – it is not the model that is at fault, but rather the calibration of the model.

I further do not agree that the altered scenarios are ad hoc. To me, in order to perform atmospheric reconstruction of H<sub>2</sub> one needs to calibrate the firm air transport model to correctly simulate the

expulsion of fugitive gases during close-off – very much in the same way that we calibrate the diffusivity of regular gases in the diffusive zone to simulate their movement.

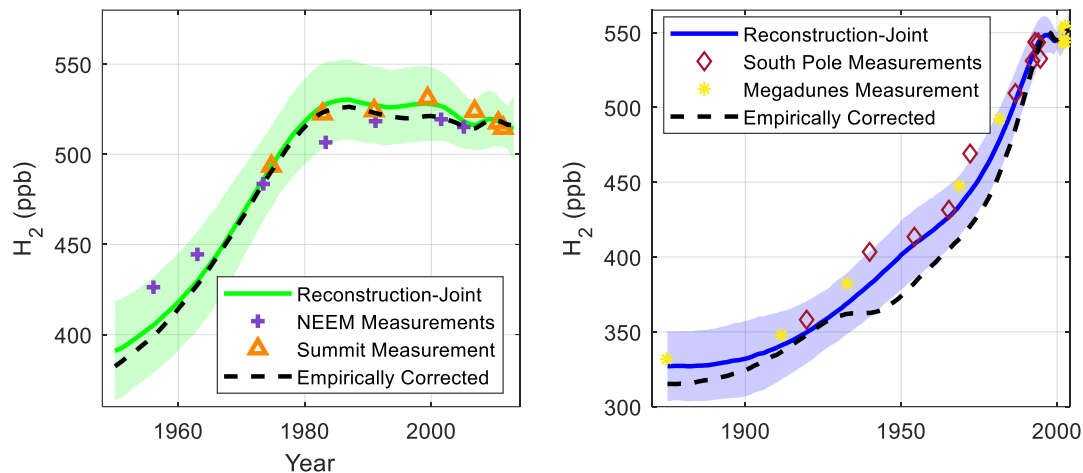
Only results for H<sub>2</sub> from model configurations that accurately capture neon enrichment are included in the revised manuscript. We have clarified our treatment of pore close-off fractionation in Section 3.2, and instead of referring to “alternative models,” we describe different model configurations for porosity partitioning and pore close-off fractionation. The 3 model configurations discussed in Section 3.2 are:

- 1) Goujon et al. (2003)- this is presented as a starting point to demonstrate that “typical” partitioning between closed and open porosity will not accurately model the neon enrichment at NEEM and Summit.  $s_{co}$  used for the Goujon configuration is given in Table 3
- 2) *Mitchell\_optimized*- an empirically tuned porosity partitioning that uses the Mitchell et al. (2015) formulation. This configuration shows good agreement between modeled and measured neon at NEEM and Summit. For the remainder of the paper, we consider this the “default” configuration. But, the optimized closed porosity profile is qualitatively different from previously observed and modeled closed porosity profiles, and it is important to demonstrate that our results are not entirely dependent on a strange porosity profile.
- 3) *Compression*- an empirically tuned 2-stage porosity partitioning that is forced to be more “realistic” than the “*Mitchell\_optimized*” configuration. In order to accurately model the neon measurements with a “realistic” closed porosity profile, we must also invoke a reduction in bubble compression (discussed further below). This configuration shows good agreement between modeled and measured neon at NEEM and Summit. We include this configuration to demonstrate that our results are insensitive to how we model pore close-off fractionation as long as it is modeled accurately.

Regarding my original point (2)

The authors did not perform the test I suggested. They show two age distributions in Fig. S7, which is unrelated to the suggestion I made. I suggested that they apply an empirical correction for close-off fractionation based on dNe/N<sub>2</sub> and then treat the gas as a normal non-fugitive gas. This should be very straightforward. I am curious whether it agrees with the Figure 9 reconstruction.

Pore close-off fractionation affects the age of the calculated age distribution, not just the integrated area. To properly conduct an inversion on empirically corrected H<sub>2</sub> measurements, one would need to use a normalized version of the age distribution plotted in blue in Figure S3 (i.e. one that integrates to 1 instead of 1.06). Turning off pore close-off fractionation yields the distribution plotted in pink. That is, the age distributions used to invert the empirically corrected measurements would be too young. The effect on age is somewhat more dramatic at the Antarctic sites than the Greenland sites, and we have chosen to plot Megadunes age distributions in Figure S3 to better illustrate the effect. In our earlier work on Antarctic firn air, we pursued empirically correcting the measurements before realizing the effects pore close-off fractionation has on the age. We prefer not to suggest to readers that empirical correction of H<sub>2</sub> measurements is valid. We have plotted the results of such an inversion below, together with the joint reconstructions. The Greenland results are fairly similar, but the Antarctic results are quite different.



Regarding my original point (3)

The updated Figure 8 (thanks for that!) sheds a lot of light on what is happening in the model. The reason these three scenarios give the same  $d\text{Ne}/\text{N}_2$  solution is that they have the same bubble pressure profile  $R$ . I fear that this is very much obscured by the text which seeks to discuss the processes. In reality, the authors just tune the closed porosity until the  $d\text{Ne}/\text{N}_2$  data fit. In each scenario, the authors essentially increase the closed porosity at shallower depths, which increases the bubble pressure  $R$ . If I may be blunt, the discussion of physical processes is just window dressing for a fitting exercise. I am totally fine with brute force calibration/tuning, if it fits the data. Given the uncertainties in porosity this seems justified to me. If anything, the scenarios demonstrate that as long as one gets  $R$  right, one fits  $d\text{Ne}/\text{N}_2$ . The details of how this is done seem to be secondary.

Note: the relevant figure is now Figure 2. We have clarified the treatment in section 3.2. We have largely jettisoned the discussion of physical processes in favor of tuning the porosity partitioning. In the *Compression* model configuration, we still optimize the rate of bubble pressurization, but that is just a convenient way to model the neon data. The revised manuscript describes the rate of bubble pressurization as a tuning lever. We direct the reviewer towards the following passages:

L276-277: Here, we use the rate of bubble pressurization as an additional tuning lever to examine the sensitivity of the model results to the specific physical parameterizations in the model.

L295-296: The alternative model configurations are considered empirical tuning methods to fit the Ne data and should not be used to draw conclusions about the underlying firn physics.

L662-663: The *Mitchell\_optimized* and *Compression* model configurations are empirical tuning methods and do not address uncertainty around the physics underlying the observed pore close-off induced enrichment in Greenland firn air

In my comment 3b I was really suggesting that the "reduced compression" scenario is basically mislabeled, as the real reason the fit is improved is the closed porosity parameterization (the reduced compression actually makes things worse). By changing two parameters at once, the test is hard to interpret. In their response they claim to clarify the text, but I see no changes to section 7.2. Such textual clarifications (which were not made, as far as I can tell) also do not fix the bigger issue of mislabeling and/or unclear experimental design (maybe I should have been more explicit about this). The reader will come away from this section thinking that reduced compression somehow improves the simulation of fugitive gases, but in fact the opposite is true. Fig. 8 shows that to compensate for the reduced compression, the authors have to increase the shallow trapping even more such that the R is correct. There MAY be reduced compression of bubbles, but the tests performed cannot say anything about this as they compensate with changes to the porosity profile.

The revised manuscript clarifies this point. The reduced bubble compression (now the *Compression* configuration) is simply a way to model the data using a fairly realistic closed porosity profile. The problem, broadly described, is that with a realistic closed porosity profile, R increases too rapidly with depth to capture the neon data. That is why we cannot use only adjust  $s_{co}$  for the Goujon parameterization or  $\rho_{co}$  for the Mitchell parameterization. To match the neon data, either bubble close-off must take place over a broader depth range (as in the *Mitchell\_optimized* configuration), or we must invoke some other process. Here, we have chosen the rate of bubble compression as a convenient lever (as mentioned above). We first describe our attempt to model neon with a realistic porosity partitioning. Only when that fails do we introduce reduced bubble compression:

L265-278: "We also examined one additional model configuration, referred to as the "Compression" configuration, in which we force the closed porosity profile to have a more typical complete close-off depth. In this configuration, the closed porosity above some optimized critical depth ( $z_{crit}$ ) is parameterized as in Goujon et al. (2003; their equation 9) with an optimized  $s_{co}$  parameter. Below the critical depth, closed porosity increases linearly with depth, reaching complete close-off at the same depth as in the Goujon et al. (2003) base-case (79.2 m at NEEM and 81.5 m at Summit). A similar two-stage closed porosity parameterization was used in Severinghaus & Battle (2006). The resulting closed porosity profiles are more similar to previously published profiles and probably more realistic (Figure 2). However, even with an optimized  $z_{crit}$  and  $s_{co}$ , we found that R increases too rapidly with depth to capture the  $\delta^{22}\text{Ne}/\text{N}_2$  at both the top and the bottom of the lock-in zone. Therefore, to generate the necessary R profile while maintaining a realistic closed porosity profile, some other physical process must be modified in the model in addition to the closed porosity profile. Possible candidates include the rate of pressurization of bubbles, the rate of densification, and the rate of bubble close-off. A detailed observation-based investigation of these processes at these sites would require field data that do not currently exist. Here, we use the rate of bubble pressurization as an additional tuning lever to examine the sensitivity of the model results to the specific physical parameterizations in the model."

It is important to include this configuration to demonstrate our results are not dependent on the strange *Mitchell\_optimized* closed porosity profile. We agree that the tuned model configurations cannot say anything about the underlying firm physics and direct the reviewer to the following passages:

L295-296: The alternative model configurations are considered empirical tuning methods to fit the Ne data and should not be used to draw conclusions about the underlying firn physics.

L662-663: The *Mitchell\_optimized* and *Compression* model configurations are empirical tuning methods and do not address uncertainty around the physics underlying the observed pore close-off induced enrichment in Greenland firn air

In comment 3c I was suggesting that the NEEM closed porosity parameterization they start out with (the UCI\_2 run) is not reliable. They respond by adding a comment that the Mitchell parameterization didn't improve the fit - not exactly a full response. What does "not optimal" mean here? Surely it performed better than the UCI\_2, and perhaps by tuning the 3 parameters in this parameterization one can also get a good fit to the dNe/N2 data?

We are now using an optimized Mitchell parameterization as the default model configuration. The optimized parameters are very different from what Mitchell recommends, and, as shown in Figures 2a and 2c, the optimized closed porosity profile is qualitatively different from "typical" closed porosity profiles with bubble close-off taking place over a much broader depth range and a deeper complete close-off depth.

L259-264: The *Mitchell\_optimized* parameters are significantly different from the recommendations of Mitchell et al. (2015). Furthermore, the resulting closed porosity profiles are qualitatively different from other measured and modeled closed porosity profiles and are probably not physically realistic (Figure 2). Bubble close-off takes place over a much broader depth range in the *Mitchell\_optimized* configuration, and complete close-off (i.e.  $s_c > 0.999 s_{total}$ ) does not occur until depths of 111.0 m (NEEM) and 112.5 m (Summit). Firn air could only be sampled to a depth of 75.9 m (NEEM) and 80.1 m (Summit), suggesting that complete close-off is actually significantly shallower than in the *Mitchell\_optimized* configuration.

In my comment 3d, please add some text to the discussion stating that as long as one gets R right, one gets the dNe/N2 right.

L245-252: At equilibrium, modeled enrichment of Ne and H<sub>2</sub> is controlled primarily by the ratio of the volume-weighted average pressure in the open and closed pores to the ambient pressure, adjusted for mixing. We define a new parameter (*R*) to describe this ratio:

$$R = (P_{bubble} s_c / s_{total} + P_{ambient} s_o / s_{total}) / P_{ambient} \quad (8)$$

When previously published parameterizations of partitioning between open and closed pores are implemented in the model, *R* begins to increase too deep in the firn to capture the shallower  $\delta^{22}\text{Ne}/\text{N}_2$  measurements (Figure 1 and Figure 2).

In response to my comment 3e, could you please just extend the plot further down? The alternative scenarios provide the best atmospheric reconstruction (by the author's admission), so it seems that we

should have the details to evaluate them. None of them go all the way to a closed pore fraction of 1, which is what prompted my curiosity

For the Goujon et al. (2003) base-case and the *Compression* configuration, Figure 2 now shows a closed porosity ratio of 1. The *Mitchell\_optimized* configuration reaches 1 much deeper, and we would prefer not to plot it here as it is probably not realistic. In the text we have listed the complete close-off depth for all three configurations.

L260-264: Furthermore, the resulting closed porosity profiles are qualitatively different from other measured and modeled closed porosity profiles and are probably not physically realistic (Figure 2). Bubble close-off takes place over a much broader depth range in the *Mitchell\_optimized* configuration, and complete close-off (i.e.  $s_c > 0.999 s_{total}$ ) does not occur until depths of 111.0 m (NEEM) and 112.5 m (Summit).

L268-269: Below the critical depth, closed porosity increases linearly with depth, reaching complete close-off at the same depth as in the Goujon et al. (2003) base-case (79.2 m at NEEM and 81.5 m at Summit)

The response to my Eq. 4-6 comment is insufficient. The way the equations are written they cannot be solved sequentially. So this needs to be clarified.

We have added additional subscripts to equations 5 and 6 and clarified the text.

(L211-219):

$$x_{n(bubble)\_eq} = P_n / P_{bubble} \quad (5)$$

$$x_{n(firn)\_eq} = P_n / P_{ambient} \quad (6)$$

Where  $P_n$  (Pa) is partial pressure of gas  $n$  ( $H_2$  or Ne; See section 5),  $x_n$  is mole fraction of gas  $n$ ,  $P_{bubble}$  (Pa) is the total bubble pressure,  $P_{ambient}$  (Pa) is the ambient pressure in the open pores, and  $s_c$  is closed porosity. The subscripts *bubble* and *firn* distinguish between the closed and open porosity. Equation 4 is executed at the end of each time step in each grid cell in the model. Then the mole fraction of gas  $n$  in the bubbles and firn air is updated to its equilibrium value using equations 5 and 6 before proceeding to the next timestep ( $x_{n(bubble)\_eq}, x_{n(firn)\_eq}$ ).

In their response to L201-206, they neglect a critical process (backflow) because it improves the fit to two tracers - however, the correct approach here would be to include the physical process and calibrate the diffusivity profile to fit the tracers. They claim the model tracks air content, but this cannot be correct if you neglect the backflow.

We updated the model using equation A27 in Severinghaus & Battle (2006) and re-tuned. The text and figures have been updated accordingly.

In their response on Line 267. How do they pick the beta parameter? My main point is that instead of an arbitrary smoothness parameter, they impose a autocorrelation parameter – can you explain why this is less arbitrary? Isn't the beta parameter effectively a smoothness parameter also?

Beta is not explicitly prescribed. The source of concern may have been this sentence: “ $\beta$  is a positive scalar which may be specified or varied as a free parameter.” In the results presented here, Beta is only varied as a free parameter. Therefore, we have deleted the sentence. Additionally, we have changed  $\beta$  to  $\alpha$  so that our nomenclature directly aligns with Aydin et al., (2020).

The beta parameter quantifies how much atmospheric  $H_2$  can change from one year to the next and can take on a range of values that allow an ensemble of different trajectories for  $H_2$  with different variances in time. Larger values of beta result in higher the year-to-year change in  $H_2$  in each atmospheric trajectory, with no direct relevance to the smoothness of the posterior distributions, i.e. the presented firn inversion results. The smoothness directly emerges out of the structure in the measurements and the magnitude of the associated error bars, and for multi-site inversions, also the agreement between the data from different sites. The MCMC optimization algorithm selects the most likely Beta from a uniform prior given the firn air data. In this way, Beta depends on the firn air data instead of an arbitrary user-selection.