We thank Referee 2 for their comments and help improving the manuscript. This document replies to those comments point by point, with R2's comments in green, our responses in black, additions to the manuscript in blue, and deletions in red.

Overall comments
This paper investigates the transport and evolution of chemical species through sea ice by comparing lab experiments with theoretical modelling work. The paper shows that simply assuming that PFAS evolve in the same way as salinity does not match observations. Three possible alternative models are tested, with two seeming plausible representations of this process. The rationales for some of these models could have been explained more and there are important presentational areas for improvement. But I think that the paper is a very good contribution, containing novel results of interest to the Cryosphere readership. Thus, it should be published subject to the minor corrections.

Minor (general) comments:
The presentation of results could be improved, both the in terms of the figures and the text. I make detailed line-by-line comments below, but I think the heart of the issue is that the figures may be in a sub-optimal order. The following is only a suggestion but may improve the readability of the paper.
Figure 3 is by far the easiest to understand, so I suggest it should go first, possibly with a second panel showing an example with a short chained PFAS as a contrast.
Then figure 1 could go second (or even by moved an appendix/supplement, which might allow for the model fitting to be explained in more detail). At present, it is not clear why the absolute value of b is plotted (or even the precise definition of b, which should be more clearly stated). Plotting b would make the sign of the bias apparent. The figure is far too small with very many lines (the choice of which is not precisely stated in the caption). One solution might be having a single panel (larger) in the main text explaining the calibration of a single example in greater detail, then move all the other calibrations to a supplement.
Then figure 2 (which was good) would move next, and finally figure 4. The latter could potentially be expanded to plot more of the correlations implicit in table 1. E.g., it could have a separate panel with N_c as the independent variable and $\alpha_{\{B,C,D\}}$ as dependent variables, normalized appropriately.
The reordering of the figures may also help the writing by making the ideas easier to visualize.
We thank the referee for these constructive comments.

On reflection, we have retained the original ordering of figures. Our reasoning is that:
Figure 1 shows tuning of the methods. It must therefore be placed before the other figures, which show data from tuned model results.
Figure 2 shows the scatter plots, which are described before the profile shapes are discussed in detail (for which we used Figure 3).
However, we have made adjustments to the figures and text to address the concerns of the Referee, and these have improved the readability of the manuscript. Figure 3 is referenced earlier, alongside Figure 2, so that readers are introduced to this easy to understand figure simultaneously with Figure 2. Figure 3 has also been placed in the Result section, rather than the Discussion. Also, in response to referee 1, the profiles in Figure 2 have been made more
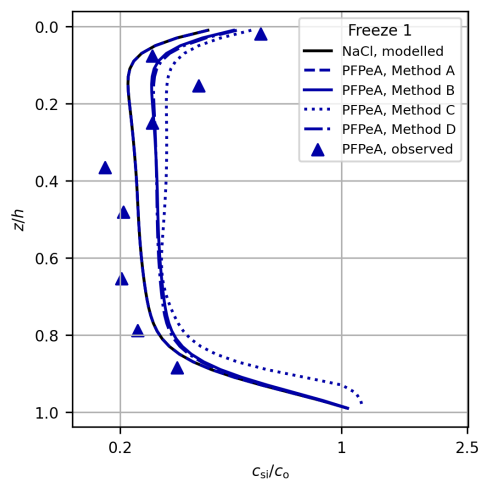
clear (pasted below).
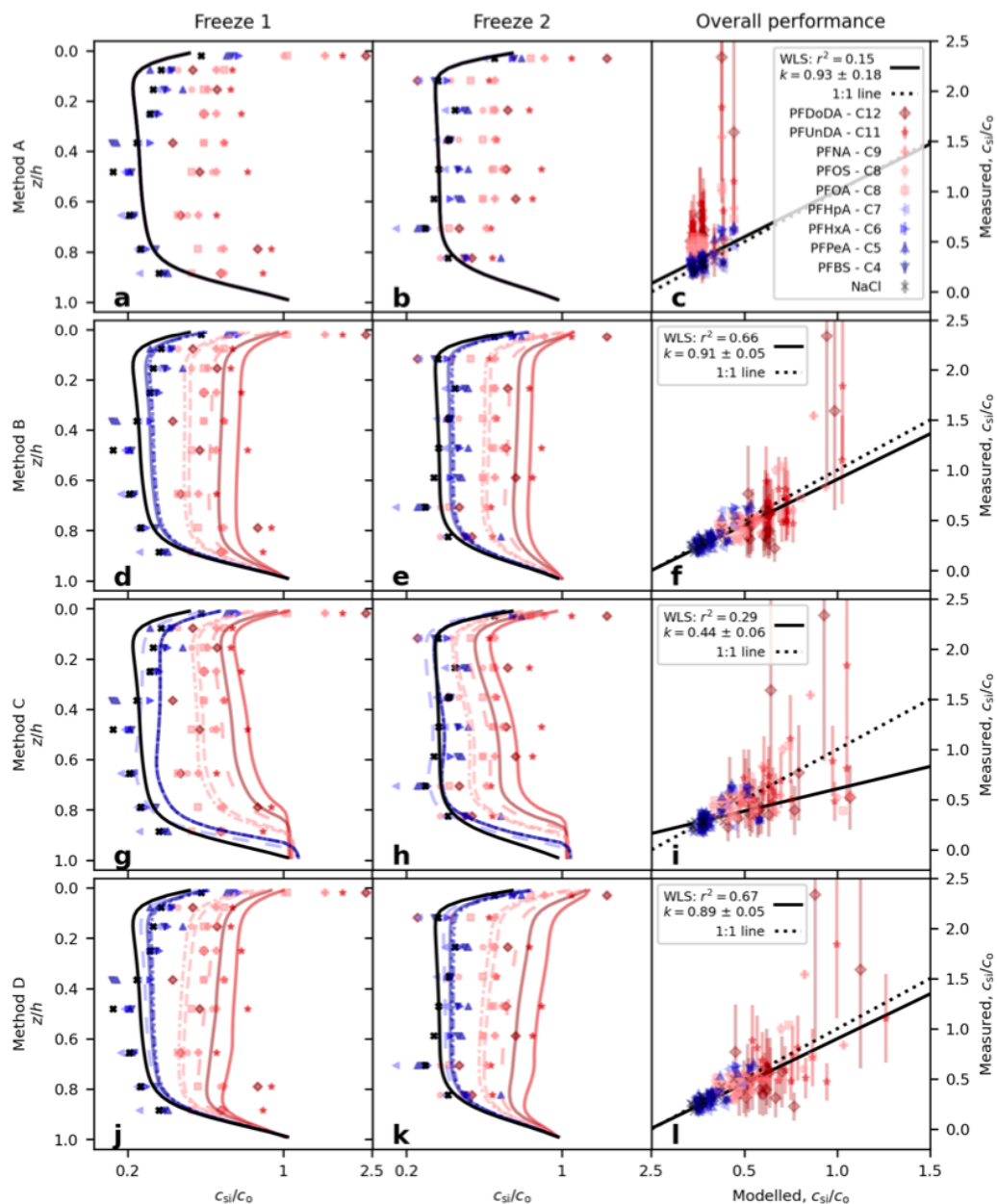
Figure 2. Comparison of modelled and measured concentration for: Method A, perfectly dissolved chemicals (a, b, c); Method B, simple ~~partitioning~~ decoupling (d, e, f); Method C, surface area adsorption (g, h, i); and Method D, ~~salting out~~ salinity mediated decoupling (j, k, l). Depth profiles are shown for freeze 1 (a, d, g, j) and freeze 2 (b, e, h, k). Modelled against measured concentrations are shown in panels c, f, i, and l, alongside the best fit weighted least squares (WLS) regression (black line, gradient k with one standard error and coefficient of determination r^2 shown in legend) and the theoretical 1 to 1 line for perfect model behavior (dotted black). Concentration for the profiles is given on a log scale to highlight separation between the profiles.

We thank the referee for highlighting the room for improvement in Figure 1. Figure 1 has been adjusted to 1) show bias, rather than absolute bias, as suggested; 2) have bigger panels; 3) have bolder lines; and 4) show the 0 bias line where we identify the best tuning parameters. The revised figure is pasted below. With these changes, the figure is more clear, and we retain all three panels in the main text rather than moving two to the supplement. Some of these changes were made in response to Referee 1, and more information about these changes are available in our response to Referee 1.
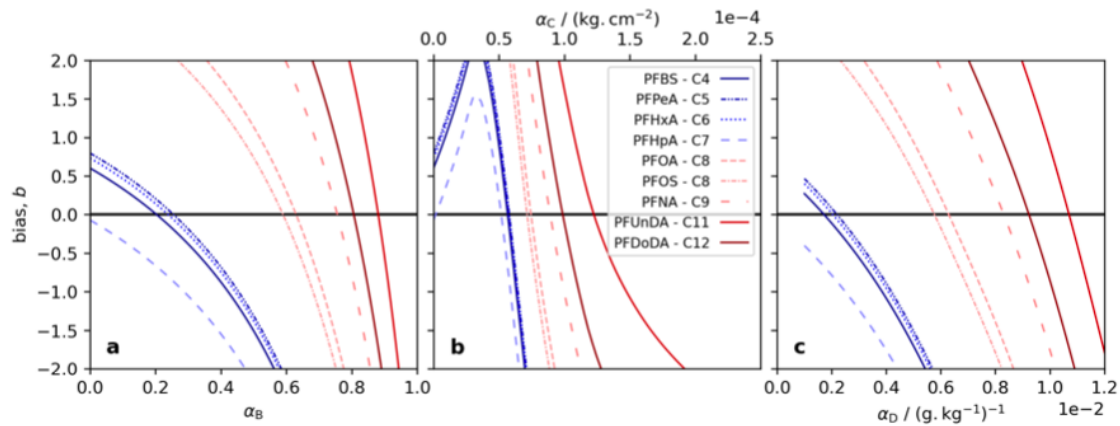
Figure 1. ~~Absolute~~ Model tuning for each PFAS. The bias, ~~|b|~~ b, was calculated as the sum of the difference between measurements and co-located model ~~with a range of~~ layers in three tuning ~~parameters, α, runs~~. Each panel shows results for ~~Methods B~~ a different decoupling method: a) simple decoupling, ~~C~~ tuned using α_B; b) surface area adsorption, tuned using α_C; and ~~D~~ c) salinity mediated decoupling, tuned using α_D. The best performing α give b closest to 0 (black line).

With Figure 3 we aim at communicating the differences between the methods. For short chained PFAS those differences are less clear (see figure pasted below), so we have not added a panel showing the short chain results. Several cosmetic changes to Figure 2 (in response to the other review) have made the profiles clearer.

We investigated different combinations of alpha_B and alpha_D regressed against K_OW and N_C (3 extra regressions, on top of alpha_B vs K_OW as shown in Figure 4). They are pasted below.
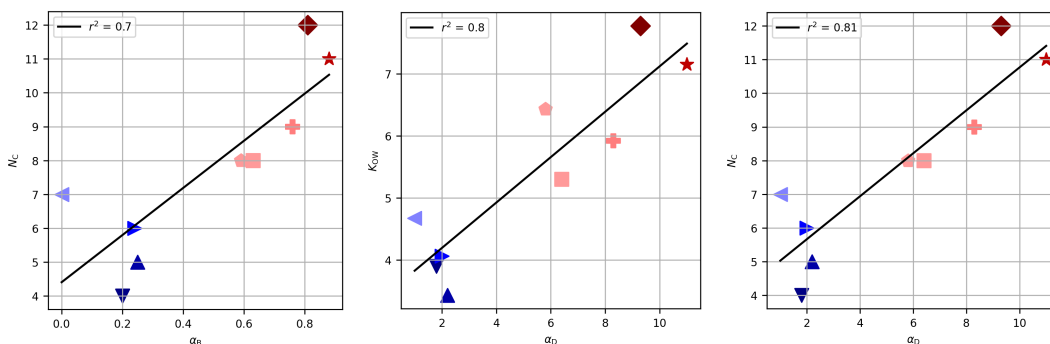
We did not investigate alpha_C as Method C did not perform satisfactorily. We state in the revised manuscript that:

indirect relevance to our experimental system). We also calculated this correlation for α_D (which also performed well) and for α_B and α_D against N_C (not shown). The correlations are all significant at 95 % confidence and range from $r^2 = 0.70$ to $r^2 = 0.81$. In our experimental system,

The other structural writing issue is the overlap between sections 3 and section 4. I think more of the detail should go into section 3 and then section 4 should be more summative (rather than restating the detail in section 3).

We have moved the start of section 4 to section 3, so that the comparison of the different methods is in the Results, and the Discussion starts with several questions. Some changes were also made in response to Referee 1, and these are detailed in our response to Referee 1.

Technical line-by-line comments:

- L5: could mention direction of bias (although this is clear later in the abstract)

  We have clarified that there is an underestimation (see also below).

- L6: "as a constant fraction" a bit vague (fraction of what, especially when it is being contrasted with "proportional to the brine salinity" which also has a constant proportionality coefficient)

  We have clarified:

  ice. With no decoupling the model performs poorly — failing to reproduce underestimating the measured concentrations of high chain-length PFAS. A decoupling scheme where PFAS are decoupled from salinity as a constant fraction of their brine concentration, and a scheme where decoupling is proportional to the brine salinity, give better performance and bring the model into reasonable agreement with observations. A scheme where the decoupling is proportional to the

- L7: consider putting the other poorly performing scheme earlier (before the good ones)

  We have retained the original order, which was based on scheme simplicity. While we agree that the story would be more linear if the schemes improved from A to D, we feel that Method B must come second as it is more simple than C and D.

- L34: this was interesting, it looked like none of the models would have been able to reproduce this?

  Actually the decoupling schemes can reproduce this effect. Method D, for example, gives PFUnDA concentrations of 1.25 near the surface. We mention this in the

manuscript already (including in the abstract and results), but have added a line to the conclusions regarding this point:

> A sea-ice brine convection model can reproduce observations of poly- and perfluoroalkylated substances (PFAS) freezing into sea ice, providing a term is introduced that partially de-couples PFAS from the moving brine. PFAS with longer car-
> 30 bon chains ~~behaved less conservatively~~ were enriched with respect to salinity~~and required larger decoupling parameters~~ ~~.~~, sometimes to levels above the underlying water, in observations. Larger decoupling parameters were required to simulate this behaviour. Decoupling methods with primary
> 35 dependence on the brine salinity performed well, and outperformed a decoupling method that depended on the internal sea-ice surface area. Our results demonstrate that brine convection models are powerful tools beyond pre-dicting sea-ice salinity. Complex, biogeochemical problems
> 40 can benefit from the accurate, physically-based advection of brine predicted by current parameterisations.

- L50: will the sampling bias be the same for salt vs PFAS? If the idea that some of the PFAS is stuck on the solid, then you might think there is a greater sampling bias for salt vs PFAS. Might be worth discussing

We have added a line to the discussion.

> 10 underestimate the sea-ice bulk salinity~~, causing the tuning~~. A brine loss driven sampling bias may impact PFAS differently to salt. If PFAS were preferentially retained, the observed enrichment in the sea ice would overestimate the in situ value. Also, tuning to samples affected by this bias would
> 15 cause the tuning parameters to give too much desalination (Thomas et al., 2021). Griewank and Notz (2015) present

- Table 1: could give a definition/formula for K_OW.

We have added the formula to the table and updated the notation to $\log_{10}K_{OW}$ throughout the manuscript.

- A more general point is that the terms 'decoupling,' 'partitioning,' 'fractionation' are being used for the same type of process. It may be worth spelling this out explicitly in the introduction.

We have searched for these terms in the manuscript and have revised their use to be consistent. 'Partitioning' now only refers to K_OW. 'Fraction(ation)' now refers either to a literal fraction, or to isotopic fractionation.

- Eq. (1): could explain that diffusion is assumed to be slow on these timescales

Rather than specifically mentioning diffusion we have cited Notz and Worster (2009) in two instances, stating that brine convection is the dominant redistributor of solutes in growing sea ice.

- L80: I would write out this formula. I would also make it more explicitly clear that c_br=c_si/phi (or similar expression using S for salinity).

We have added the formula (see also next comment).

> For salt or other fully-mobile tracers c_s ~~would be zero.~~ is zero, reducing to $c_{br} = \frac{c_{si}}{\phi}$ which is normally used
> 15 (e.g. Cox and Weeks, 1988).

- In equation (2), the meaning of this formula is not very clear. The usual lever rule for bulk concentration is c_si=c_br*phi+c_s*(1-phi), but this does not appear to be equivalent to equation (2).

We have written Equation two as follows :

Csi / φ = Cbr + Cs,

Which highlights that Cs and Cbr are to be read at the same level. We have also clarified that when C_s=0, this equation reverts back to the usual formulation and have cited Cox & Weeks (1988) as an example. We have also revised the text slightly to highlight that we made a choice to split the brine chemical pool.

$$\frac{c_{si}}{\phi} = c_{br} = \frac{c_{si}}{\phi} - c + c_s \qquad (2)$$

> where ϕ is the brine fraction. Equation 2 expresses ~~that the~~ the choice of splitting the dissolved-in-brine PFAS pool ~~is split~~ into a mobile (c_{br}) and a stationary phase (c_s).

- Relating to equation (3), subject to the comments about equation (2), it is possible to combine equations (2) and (3) to relate c_s to c_br, in which case the proportionality constant would be called a partition coefficient.

We agree that equations 2 and 3 could be combined. We have used the form we present in the manuscript because we need to introduce the stationary phase. If we combined equations 2 and 3 the stationary phase would disappear. While this would be a simpler formulation, the description in text becomes awkward.

- L104-105: could add a reference for this claim. Are there any independent experimental estimates of the strength of this effect?

While we are not aware of modelled or measured Setschenow (salting out) constants by sodium chloride for PFAS, the aqueous solubilities of these chemicals will be reduced in seawater/brine relative to pure water based on basic physico-chemical considerations. We have added a reference to Freire et al. (2005) who show this effect for Hexafluorobenzene.

- L122 add "(WLS)" as the acronym appears in figure legend but is otherwise undefined.

We have defined WLS in the figure legend and in the results text.

- L144: the differences are quite marginal

Please see new text discussing the similarities between Methods B and D (which was requested specifically by Referee 1).

brine salinity profile. Method D marginally fails one of our tests, where the (Figure 3). Despite this, Methods B and D are similar in terms of profile shape and quantitative performance. This similarity arises because Method B also includes a (implicit) dependence on S_{br}. The product $\gamma c_{si}/\phi$ (Equation 2) increases with S_{br} (for both methods) because ϕ linearly scales with S_{br}. After tuning, both methods simulate similar dynamics. The gradient of the modelled concentra-

- L165: needs a paragraph break (but also see general comments on rearranging sections 3 and 4).

We have added this paragraph break. We have moved some discussion text to the results.

- L207: presumably, how to handle the decoupling will vary between these distinct types of chemicals?

We agree, but think this paragraph is not the correct place to mention it. Here, we are discussing potential applications for brine convection modelling (and have avoided specific mention of decoupling). In the previous paragraph we discuss the generality of our results. We have made a change there, removing mention of a different gamma, and rather leave scope for more significant changes relative to the framework we propose.

be a useful approximation in natural seawater environments. For other chemicals, the different physico-chemical properties would necessitate re-tuning and possibly different functional forms for deriving γdecoupling, but the frameworks proposed for Method B (and possibly D) here would be a useful starting point.