https://egusphere.copernicus.org/preprints/egusphere-2024-745#RC1

This novel study employs ring wind tunnel experiments combined with stable water isotope analysis to investigate airborne snow particle metamorphism. A main finding is that vapour deposition drives snow particle growth and rounding supported by the observed isotopic fractionation and concurrent SSA decrease. It is inferred that particles and air inside the saltation layer are not in thermal equilibrium as is commonly assumed in blowing snow models. Any mechanical particle fragmentation or coalescence likely play a smaller role in the observed particle size changes as they would not induce isotope fractionation. In turn, the water stable isotopic fractionation induced by airborne snow metamorphism needs to be taken into account when extracting climate information from ice cores, especially at dry and windy locations.

We thank the reviewer for the detailed feedback and are delighted about the short, yet accurate and precise summary of our manuscript which agrees very well with our own perception of the main take-home messages of this study. We address the reviewer's comments individually and in detail below (answes in green). In addition to edits based on the reviewers' comments, we updated a few inconsistencies in the text and figures, such as the color code in Fig. 3 to be consistent throughout the manuscript. In summary, the major changes made are related to:
1) New Fig. 5 to describe the co-evolution of d18O and dD during and after snow introduction in more detail
2) the statistics of observed isotope changes in vapour and snow. We included a table (Table 2) to group the information and declutter the corresponding Sec 3.2
3) A short paragraph in the introduction to define temperature-gradient and isothermal snow metamorphism

General Comments

This is a carefully designed laboratory experiment, with sound methods and data analysis, and with some interesting conclusions, and should published after addressing minor comments listed below.

While this may be common knowledge a very brief description of isothermal versus temperature gradient snow metamorphism as relevant to this study is warranted in the introduction.
We followed the reviewer's recommendation and have included the following paragraph in the introduction: L.62: *"In this context, Walter et al., (2023) first introduced the term "airborne snow metamorphism" which summarises the multiple cycles of sublimation and vapour deposition on the suspended snow particle resulting in modifications of the snow particle size and shape during aeolian particle transport in analogy to metamorphism inside a stationary snowpack (Pinzer et al., 2012; Schleef et al., 2014b). The term snow metamorphism describes the recrystallization of snow grains*

in a snowpack that is driven by vapour pressure gradients (Colbeck, 1982). Snow metamorphism typically results in a decrease in SSA (growing of snow grains) and can be associated with density changes in a snowpack (Jafari et al., 2020; Kaempfer and Schneebeli, 2007). Based on the temperature regime of the snowpack a distinction can be made between isothermal and temperature-gradient metamorphism. The dominant physical processes creating the vapour pressure gradients are distinct: Under isothermal conditions, the microscale curvature effects (Kelvin equation) drive metamorphism (Colbeck, 1980), which are outweighed by macroscale temperature-gradient effects (Clausius-Clapeyron equation) in snowpacks with a temperature gradient (Marbouty, 1980). Temperature-gradient metamorphism typically results in higher recrystallization rates and thus faster SSA decay in snowpacks (Taillandier et al., 2007).

Thus, airborne snow metamorphism was proposed as a driving factor for PPP changes during aeolian transport of snow, yet the relative importance of the different processes involved and their combined effect on the snow microstructure is still unknown due to missing observations."

In particular to clarify the statement that a particle-air temperature gradient must exist to explain depositional particle growth and isotopic fractionation.
The alternative would be vapour fluxes (sublimation/deposition) across an individual particle but also between particles driven by the curvature (Kelvin) effect resulting in local water vapour pressure gradients and super(or sub)-saturation. These fluxes occur at thermal equilibrium and may also induce isotopic fractionation between solid and the remaining vapour phase. I may be convinced that the bulk isotopic composition of snow remains constant but some further discussion is warranted.

We fully agree with the reviewer that microscale curvature-driven pressure gradients exist which control local snow sublimation and vapor deposition that result in grain growth and grain rounding also under thermal equilibrium. In fact, we mention that analogy to our observations in L. 476: *"Notably, the evolution of sphere size distribution resembles the evolution observed during isothermal metamorphism (Legagneux and Domine, 2005; Flin et al., 2004)".* Yet, the time scales for such isothermal metamorphism are in the range of days while the observed airborne metamorphism happens within minutes to hours.
To clarify this important distinction, we have modified L. 559 which now reads: "*As discussed earlier, this condition of a particle-air temperature gradient that leads to supersaturation with respect to the particle surface is a requirement for the growth of particles through vapour deposition* ***on such short timescales*** *and the changes in* $\delta^{18}O$ *support this theory.*"
Regarding the necessity for a temperature-disequilibrium for isotopic fractionation, we also agree with the reviewer that isotopic fractionation, and thus potentially a noticeable change in snow isotopic composition can also happen under thermal equilibrium (driven by the Kelvin effect). The considerations to explain the changes

in snow $\Delta\delta$D (Fig. 6b) assume such equilibrium conditions. However, these considerations can not explain the observed $\Delta\delta^{18}$O. Thus, it was our intention to highlight in Discussion Section 4.2, that the specific change in bulk δ^{18}O that we observe in 4 experiments (negative $\Delta\delta^{18}$O) can ONLY be explained through kinetic fractionation under net supersaturation conditions (L. 558 and Fig. 6a). We then translate the necessary supersaturation value into a temperature difference between snow and air and obtain reasonable values that also agree with modeling estimates of Sigmund et al.. Based on these considerations we feel confident to postulate a snow-air temperature disequilibrium for larger snow particles. Smaller snow particles instead, may thermally equilibrate faster and then sublimate due to higher surface curvatures (convexities). Therefore, we argue that smaller particles disappear at the expense of the larger particles that grow resulting in the observed shift of the sphere size distribution shown in Fig. 2. The schematic in Fig. 7 is intended to summarize these processes. The corresponding text is: L. 491*:*
"The growing and rounding of the particles may thus be explained by airborne metamorphic growth with vapour being preferentially sublimated from convex sub-grain boundaries and entire small grains and preferentially deposited in concavities or on larger grains with lower curvature (Kelvin effect) in analogy to isothermal metamorphism in a snowpack (Colbeck, 1998, 2001; Wakai et al., 2005)."

To do this I'd suggest to better illustrate the temporal co-evolution of the stable H2O isotopes in both snow and also water vapour. E.g. add a similar figure as Fig.3 showing d18O, d2H and d-exx in the vapour phase. Some of the behaviour seen in experiment No.9 (Fig.4) is puzzling, e.g. O18 in vapour and snow shows correlation, whereas 2H shows anti-correlation (significant?). Was this behaviour observed also in other experiments and is this related to the mentioned non-equilibrium conditions?
We agree that the co-evolution of snow and vapour in each individual experiment is interesting but we had to synthesize the results to be able to draw general conclusions and to maintain a reasonable manuscript length. Figure 4 was intended to provide a visualization of the co-evolution to the reader as a trade-off. We decided to follow your suggestion (that was also mentioned by reviewer 2, comment 15) and we now incorporate a figure (Fig 5) to additionally visualise the co-evolution (net change) of the vapour isotopic composition vs. the change in snow isotopes. We color-coded the markers with the average experiment temperature to allow a good comparison to Fig 3.

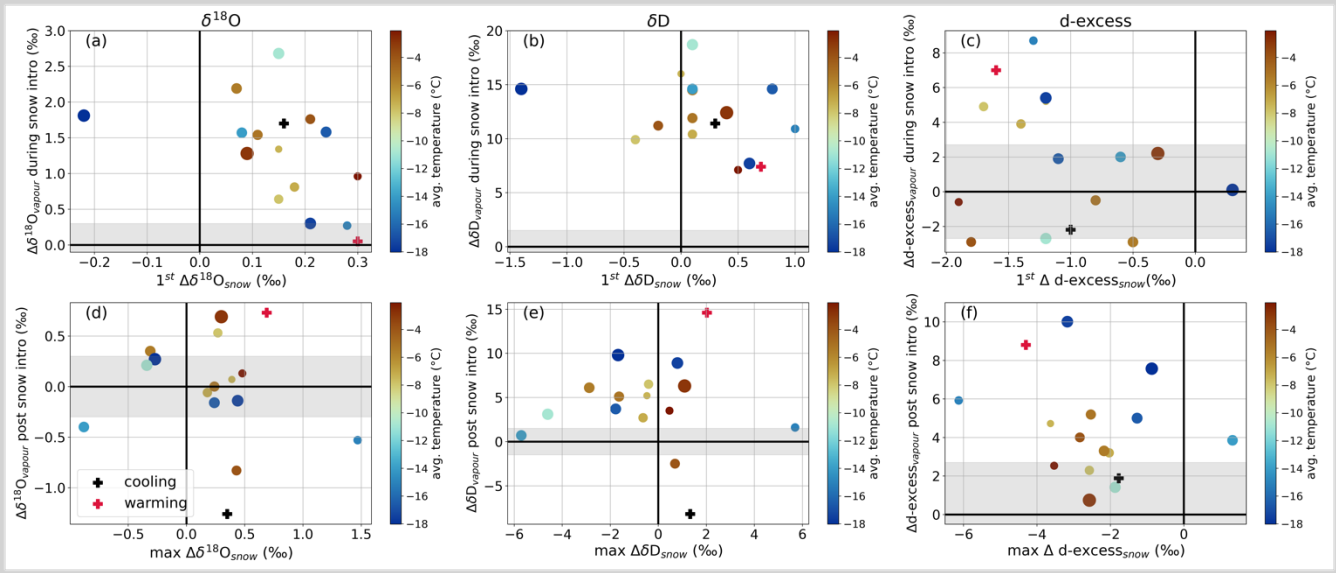

Figure 1 The co-evolution of changes in vapour and snow isotopes in all experiments with vapour isotope observations The change in the snow isotopes (x-axes) is plotted against the changes in the vapour isotopes (y-axes). The changes in vapour isotopes are calculated from the 3-min averaged data. The upper row (a, b, c) shows changes in vapour isotopes during the snow introduction plotted against the observed change in snow isotopes of the first airborne sample (see Fig. 3). The lower row (d, e, f) shows the subsequent change in vapour isotopes post snow introduction until the end of the experiment plotted against the maximum observed change in the snow isotopic composition. Shaded areas represent low variability in vapour isotopic composition (|Δδ18O| <0.3 ‰, |ΔδD| <1.5 ‰, (|Δd-excess| <2.7 ‰). Note that the colour code represents the average air temperature during the experiments and allows the comparison between upper and lower row and the results shown in Fig. 3. The cooling (black cross) and warming (red cross) experiments are exempt.

As the reviewer points out, the two isotope species ($\delta^{18}O$ and δD) do not necessarily show the same temporal evolution and trends. This is intriguing and explainable through the unique combination of vapour and snow isotopic composition and the resulting isotopic disequilibrium in our experiments. As we make snow from local tap water we create specific snow-vapor isotopic combinations that are not identical for the different experiments and not necessarily often observed in nature. However, the strong disequilibrium conditions allow us to draw conclusions, such as the existence of a snow-air temperature gradient, that would otherwise not be discernible. To better explain this to the reader we have modified L. 686ff which now reads: *"The experiments revealed that the isotopic signature of the wind-blown snow event in the wind tunnel was dependent on the combination of initial vapour and snow isotopic composition and the resulting disequilibrium. As we produced snow from tap water this disequilibrium was at times asymmetric between the two isotope species and more pronounced than generally assumed in nature. As the disequilibrium determines the isotopic evolution, it is not possible to unambiguously predict the expected changes in the snow isotope signal under wind influence without considering the water vapour isotope variability imposed by synoptic-scale atmospheric transport (Aemisegger et al., 2022; Bagheri Dastgerdi et al., 2021)."*

Detailed Comments

L145 - Mention here what temp was the wind tunnel set to?

Since the experiments were conducted at varying temperature regimes we decided to reference Table 1 at this point. The sentence now reads:

L158: "Care was taken to equilibrate the snow temperature to the target wind tunnel air temperature (Table 1) for 30–60 min before the start of the experiment."

Table 1: Clarify in the caption that DELTA T means change in mean wind tunnel T over the duration of the experiment
We changed that as recommended.

L182 - cm3
We assume the reviewer suggests adding information about the volume of the wind tunnel. We added it in L. 212: *"The wind tunnel has an air volume of ~ 0.5 m^3 and was sealed against snow loss with the help of insulation material."*

L337 - Be specific: significant enrichment by how many permil?
We added the statistics about the observed significant changes in a new table (Table 2.)

L395 - In order to illustrate the concurrent vapour isotopic composition change across all experiments I suggest a similar figure as Fig3. (see above)
We included a new figure and made that section more concise. Fig.5 shows the change in vapor isotopes during the snow introduction (upper row) and post snow introduction (lower row) plotted against the corresponding snow changes.

L431 - Shouldn't mechanic fragmentation lead to a SSA increase if it was the dominating process?
The reviewer raises a valid point. Depending on how much "new surface area" is created through the breaking of the crystal, the SSA value can increase if the volume stays the same. However, previous literature has claimed that mechanic fragmentation leads to lower SSA (Comola et al., 2017) which is conceptually wrong. We therefore included that statement but will adapt it to your suggestion now:
L.457: *"Note here, that simple mechanic fragmentation of snow particles alone does conceptually lead to an increase in sample SSA and can thus not explain the decrease in SSA."*

L441 - Please explain "higher SSA decay rates for isothermal snowpack metamorphism", how much higher? Higher than T-gradient metamorphism? reference?
We realise that this sentence was poorly phrased and thus wrongly understood. It was supposed to reference the well known temperature-SSA decay rate dependency in isothermal and temperature-gradient conditions. We did not observe such temperature dependency in our windtunnel experiments. The sentence is rewritten and references are added to L. 464: *"The driving processes for snow metamorphism are vapour pressure gradients, which are largely governed by the (absolute) temperature regime in a stationary snowpack (Kaempfer and Schneebeli, 2007; Taillandier et al.,*

2007). In airborne snow metamorphism however, other processes such as turbulent mixing of air and vapour, particle-air temperature gradients (see Sec. 4.2) and variability in saturation conditions might be dominating vapour pressure variability and therefore masking the simple absolute temperature dependency that is expected in stationary snowpack metamorphism."

L448 - Are particles in the saltation layer subject to a different metamorphism regime than those in the suspension layer? Please expand & add any relevant reference
The reviewer raises an interesting question. However, with the current experimental set-up and the available observations we don't think we can provide an adequate answer that is based on more than speculations. Since we sampled snow from the whole height of the air column our observations integrate the saltation and suspension layer. Because of the generally close-to-saturation levels and the transport of snow particles in general proximity to the outer wall or the floor, we concluded that our wind tunnel experiments are probably more likely to represent saltation layer regimes (see Section 4.3). We are not aware of other studies that have performed research on the concept of airborne snow metamorphism in suspension or saltation layer and can thus not provide relevant references.

L465 - What about the vapour flux between particles, i.e. sublimation of small snow particles, which may eventually disappear, followed by deposition to larger particles. See comment above.
We agree that we should mention this process specifically at this point. We discuss it already in L 717-718 and the process is also included in the schematic Fig 7. Following the reviewer's suggestion we added to L. 491: "The growing and rounding of the particles may thus be explained by airborne metamorphic growth with vapour being preferentially sublimated from convex sub-grain boundaries **and entire small grains** and preferentially deposited in concavities **or on larger grains with lower curvature (Kelvin effect)** in analogy to isothermal metamorphism in a snowpack (Colbeck, 1998, 2001; Wakai et al., 2005)."

L528 - Except that particle-to-particle vapour flux can occur also at T-gradient = 0 and RH_ice = 1 due to Kelvin effect (equivalent to isothermal metamorphism I think)
This is in relation to the reviewer's general comment which we have answered in detail above. We want to emphasize that the observed $\Delta\delta^{18}O$ changes can NOT be explained with isotopic equilibrium fractionation (i.e. RH=1).

Fig.6 - yes, this is related to the curvature (Kelvin) effect
We added an explanation of the Kelvin/curvature effect in the introduction and also in response to your comment above to L. 491, so it is easier now for the reader to understand Fig. 7 (previously Fig 6).

Conclusions - list here and possibly in the abstract the order of magnitude of the observed isotope fractionation attributed to airborne snow metamorphism in permil, a result relevant for the interpretation of field data.
Thank you for this valuable feedback. We added this sentence to the Conclusion L. 724: "*The change in the snow isotope signal that we attribute to airborne snow metamorphism was dependent on the vapour-snow disequilibrium and ranged from: -0.88 ‰ – +1.47 ‰ in δ18O, -5.7 ‰ – +5.7 ‰ in δD and -6.1 ‰ – +1.3 ‰ in d-excess.*"
And this sentence in the abstract: L. 24 "*Within transport times of 3 hours, we observed changes in the isotope signal of airborne snow of up to: +1.47 ‰ in δ18O, ± 5.7 ‰ in δD and -6.1 ‰ in d-excess.*"

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