We would like to thank all reviewers for their helpful advice and edits. We have tried to answer and fulfill their suggestions to the best of our abilities. Overall, we have made additional phrasing and typographical edits to improve readability and clarity. We have also switched phrasing of δD to δ^2H and use in figures of d-excess to *dxs* for consistency. Other changes are detailed below.

Improved Wind Data

During the revision process, we found and included an improved wind data source. Both the initial and improved wind datasets come from the same observation station (THU airport). Our initial data set gave the wind data (speed and direction) as hourly data, and we thought that this was the direct data from the observation site. However, we later discovered that the airport observations are not necessarily hourly, and additional sub-hourly observations are often included (possibly when flights are arriving or departing). The initial data we used in the original manuscript appears to have aggregated all these values into hourly observations to produce a consistent hourly database; however, the aggregation method used seems to have simply averaged all observations in each hourly block. For wind directions, this causes problems due to the degree nature of observations (e.g., simply averaging a 350° north wind and 10° north wind will result in a 180° SOUTH wind observation). Our improved wind data source includes all the original observations from the airport, and we could then aggregate the data to hourly values correctly (through conversion to u and v components).

The resulting improved dataset does not greatly differ from our initial set and no major changes are made to our paper's conclusions. The greatest change is that, previously, the correlations between katabatic deviation (i.e., wind direction) and isotopic variables dropped to near zero in July, and we struggled to explain this phenomenon. This now appears to have been an artifact of the poor initial wind database, as the correlations calculated in the improved data set show a smooth pattern throughout summer as expected.

RC1 Response

The paper introduces a new valuable data set of water vapor isotopic observations in the coastal high Arctic Greenland. Such a data set in a region where long term monitoring is still relatively scarce would be very useful to the community in particular to the understanding of the processes influencing water isotopic composition. The overall article is well written and clearly presented. The results are presented in detail, together with a precise analysis of the local meteorology and climate. The article presents a clear statistical analysis of the influences of multiple parameters and their interactions on the water vapor isotopic composition at multiple timescales and is therefore potentially beneficial to the fields of research of paleoclimates using water isotopic proxies or of present day atmospheric moisture cycle in the Arctic region. However, some very important aspects of the calibration procedure of the water vapor isotopic observations on which all the analyses rely are not totally clear and need to be clarified in order to validate this data set. The calibration method description is sometimes a little enigmatic and some aspects should really be clarified as several of the recommendations for long term measurements of vapor isotopes do no seem to be totally respected (Bailey et al. 2015). Technical issues did not allow applying a normal calibration procedure and therefore choices have been made not to apply all the usually recommended corrections. These choices seem to have been made in favor of a larger temporal data coverage, but the data quality is probably affected. The deviations which could be introduced

by a low quality calibration are not necessarily sufficient to invalidate the data set analyses, as the ambient air variability is very strong. But at the end, a clear overall estimate of the precision and accuracy of the data set is needed to justify subsequent analyses. If the precision is too low, some of the analyzed variations might not be significant. I believe that these questions must be answered before publishing these data set and analysis.

L.29: "and past reductions should be similarly preserved in local glacial ice": this is based on the hypothesis that the local precipitation and vapor isotopic composition are similarly affected by the local sea ice cover and that the isotopic signal of precipitation is preserved in ice cores. This hypothesis is better detailed in the "implications" section, which is sufficient. I believe this affirmation is rather strong for an abstract, as this has not been proven, and should be removed here or expression with more caution.
L29: Rephrased to be more clear that this is only a proposed concept by this paper. This concept has been independently supported by other regional ice core research such as Osterberg 2015, and we have not fully eliminated this from the abstract as we believe it highlights an important connection between our research and the broader paleo community.

L.38: Casado et al. 2018 is about the Antarctic environment, not the Arctic and should therefore be referred differently.
L 38: Removed Casado citation.

L. 140-142: What is the elevation of the inlet compared to the ground level? How long is the inlet tubing? Is the inlet tubing heated and if yes, at which temperature? This last information is important as there might be condensation occurring in a non-heated tube, in particular in very cold environments. Does the setup include any protection at the inlet to prevent snow flakes or rain from entering the system?
L 140-142: Clarified to answer questions and add additional information.

L. 144 to 148: Part of this paragraph could be moved to the previous section describing the local climate.
L 144-148: Paragraph reworked as requested

L. 155 to 157: The two liquid water isotopic standard have isotopic values of -2.24 and -29.80 ‰in _18O, whereas your ambient air measurements have values ranging from around -20 to -50 ‰. If the calibration scale does not encompass the values which are measured, how can you be sure of the validity of this calibrationÂˇa?
L 155-157: This is indeed a known issue. When the observation system was originally set up, the plan was to observe values only in summer. However, the extension of the system's observations through winter months does require standards with lower values for a properly robust calibration. Such standards are planned to be added at the next visit to the station. As for the potential impact on our results, it is true that the absolute values of observations below our standard water range risk an accuracy offset or bias that is not detected in our current calibrations. A paragraph has now been added to acknowledge this. However, we believe that the overall impact of a possible low-value bias on our study is fairly small because our conclusions and analysis largely focus on relative value changes and not formulas/equations that require absolute accuracy. Thus, even if the lowest observations are skewed in value somewhat too high or too low, this bias would very likely be graduated in degree with increasingly lower isotopic values.

Thus, a single observation's relative position (higher or lower) to other observed values (what we base our analyses on) would be preserved even if a bias exists.

L. 157 to 162: To justify the standard injection duration, which is lower than in many other studies, the stability of the isotopic values over the averaging period of the injection should be verified. A statistical analysis of the stability of _18O and _D values or a figure showing their evolution over the 10 minutes should be added to the supplementary materials at least.
L 157-162: A table is now added to the supplement (Table S3) that provides the slopes of isotopic values vs. time for each of the calibration runs, and summarized in the main text. This shows that the isotopic change after 10 minutes is acceptably flat. While the mean trends are slightly positive for three of the calibration isotopic values (USGS 45 $\delta^{18}O$ and USGS 46 $\delta^{18}O$ and δD), the mean value is within one standard deviation of zero. We judged that any possible improvements to calibration accuracy from extending the calibration time would be very small and not worth the loss of dry air from the extended run durations.

L. 168 to 169: If I understand yell, you finally estimated the stability of the system over two months and did not apply any correction over the two years. However, you can hardly justify that there is no drift of the instrument sensitivity at time scales longer than a few months with these observations. The instrument sensitivity can also be different each time it is restarted or the monitoring program is launched and a new calibration scale should be calculated. Has the instrument never been restarted over two years, or can you justify its stability? I believe this is a very important point regarding the quality and accuracy of your dataset.
L 168-169: Some clarifying lines are added to the discussion here. Comparison of isotope-isotope and isotope-climate relationships across the period of record fail to show any clear evidence of long term drift or changes in sensitivity. The machine has not been moved or shut down for extended times, with the only restarts occurring after brief power outages. It is still possible that some degree of sensor drift has occurred over our period of record, but we do not see evidence of anything large enough to alter the study's conclusions that are largely based on isotopic changes of >5-20‰.

L.177 to 186: If the humidity sensitivity has been estimated based on a experiment performed in July 2019, the dry air system had not yet been installed. If the humidity response function has been estimated based on measurements performed with the DRIERITE as a dry air source, it is highly possible that the remaining moisture in the dry air source is strongly influencing the measurements at low humidity. This can lead to strong deviations at humidity values below a few thousand ppms and could explain a very large part of the deviations depicted on Figure S1. It would be helpful to have an idea of the background humidity levels when injecting dry air produced via the DRIERITE without any liquid standard injection, in order to evaluate the validity of the humidity sensitivity corrections. If you apply a correction of the humidity sensitivity based on a experiment biased by a low quality air source, you will introduce this bias into your calibrated data set. Furthermore, more details should be given on the procedure to apply the humidity response correction has been applied.
L 177-186: This comment is drawn from a misunderstanding of the humidity response calibration. The reviewer believes it to predate the installation of the dry air system based on our previously confusing wording in the manuscript. This wording has been altered to make it clear that the humidity response calibration was performed with the

dry air system. A line was added to make the application of the correction more explicitly clear.

L. 196 to 197: How did you add the CRDS mixing ratio values to the values recorded by the weather station? Did you use these measurements to fill gaps in the weather station records, or did you use any kind of average between the two sensors? Did you apply any calibration of the mixing ratio measured by the CRDS analyzer, or did you compare it with measurements performed by the weather station?
L 196-197: Line added to clarify. Mixing ratios from the Picarro were added as an independent weather variable, but mixing ratios were also calculated from the SMT weather station data for comparison. The two mixing ratio datasets have a Spearman correlation of +0.99.

L. 580 – 604: The unique diel cycle observed in March in water isotopes is very similar to the observations of Bonne et al. 2020 in Siberia, which is cited in the previous section. The environment is a little different as there can not be any influence of katabatic winds in the Siberian sector, but this spring diel cycle is also attributed to the sublimation of the snow deposited earlier in winter, which has an isotopic composition different than the spring water vapour isotopic composition. Similarly, they don't see any significant diel cycle in other seasons. I think this would be worth including a short comment relating both situations.
L 580-604: Commenting lines added, thanks for the suggestion.

Technical comments: L. 122: Change "kmh□1" into "km h□1".
L. 139: "The L2130-i + SDM uses cavity ring-down spectroscopy". It does not seem necessary to add "+SDM", as the SDM is the calibration samples injection module, not the spectrometer itself.
L.140: Wouldn't "suited" be more suited than "amenable"?
L. 252: "over 30‰(-49.5‰to -17.5‰) 230‰(-377‰to -142‰), and 55‰(-7.6‰to 47.5‰)": correct positions of the commas "over 30‰(-49.5‰to -17.5‰), 230‰(-377‰to -142‰) and 55‰(-7.6‰to 47.5‰)"
L. 271: "the effects of the these factors"
L. 307: "predominate" > "predominant"
L. 318: our analysis reveals that a large amount
L. 342: "data's" > "data's"
L. 360-361: winds source have isotopically light vapor / winds source isotopically heavy vapor
L. 367: "on the L2130-i" is not necessary
L. 385: "composition, (Casado et al., 2018; Kopec et al., 2019)" : comma is not needed
L. 469: very warm observations at Thule come during this period
L. 590: "snow grains that have equilibrated"
L. 692: "adding additional": formulation could be improved
Corrected as suggested

RC2 Response
The article presents a 2-year record of water vapor isotopic measurements at a coastal station in Northwestern Greenland. This is part of a larger effort to build a network of water isotopic observations at different Arctic sites. The continuous record allows to analyze the isotopic variability at the diel, synoptic and seasonal time scales. Isotopic variations are interpreted in

terms of meteorological factors. Implications for paleoclimate studies are discussed. The paper reads very well. The rationales are sound. I only have minor comments.

section 4 “general overview of isotopic results”: I found it difficult to read because it’s only text and there is no reference to any figure or table. I suggest to add references to figure 2 and/or table 1 and to relate the text to them. Also, this section is very short and does not add much to the paper. Maybe it could be moved into an introductory sub-section of the next section?
Section 4: This section is now largely integrated into the next section, with one part moved to new Supplement Section 3. Table 2 now has the correlations in one place.

• l 255-265: this paragraph does not add much to the paper. This information is reused in the next sections. This paragraph could be removed and the information could be added when needed in the next sections. The correlation numbers could also be gathered in a table, which would make them easier to read and to refer to in the text.
L 255-265: Paragraph removed and necessary information integrated into following section and table 2, as suggested.

l 365: “personal field observations on multiple occasions”: can we see them in Figure 2? It would be more convincing to directly show it in the Figure (or in a zoom of this figure) than to refer to personal observations.
L 365: This sentence was deleted in edits, as the citing of anecdotal evidence was not needed as the previously stated statistics provide the necessary evidence. Visual supporting evidence is not easily clear at the temporal resolution of Figure 2, but is seen and discussed in more detail in the diel section and Figure 8.

l 391: Is this effect supported by observations? Or is it too small to be seen? If so, clarify that this effect is expected to be weak?
L 391: We are not sure which effect this comment is referring to. We assume it references the effect of plant transpiration, and we mentioned that we expect it to be very weak because plant cover is so sparse at Thule. We have rephrased the sentence to be more explicitly clear that this is an assumption of ours, but it is based on the cited work that quantified how little plant productivity exists in this landscape.

• section 6 “Annual isotopic cycle”; the title does not represent the content of this section. This section looks at correlations within each month, so it is rather synoptic variability. This section could be renamed “Synoptic-scale isotopic variability”.
Section 6 (now 5): We have changed the title and some organization of this section to highlight its focus on seasonal changes in isotopic relationships and variability. However, we don’t agree that the overall section is best described as focusing on synoptic patterns. Within this section, we discuss why the annual cycle observed in the isotopes emerges as a result of changes in the environment in different seasons. Some of these environmental differences include changes in the general synoptic patterns and thus we examine the isotopes and their relationships at monthly scales. Still, the overall focus of the section is not analyzing synoptic changes independently of seasonal identity (such as examining how particular synoptic atmospheric patterns consistently alter isotopic values whether in winter or summer, as we highlight in the following section), but rather specifically in the context of the annual isotopic cycle.

l 429: can we see it in a Figure? If so, refer to it.

L 429: Reference to figure 2 added

• Same l 436.
L 436: Reference to figure 2 added

• l 442: “isotopic variables”: only dxs, isn’t?
L 442: The referenced figures show that the correlations are very strong for both $\delta^{18}O$ and dxs, though one is strongly negative and the other strongly positive.

l 444-449: is this paragraph about diel variability? If so, maybe this can be moved to a later section on diel variability?
L 444-449: While this does mention changes occurring on a diel cycle, the focus here is that the sea ice breakup in spring directly leads to the sea breeze beginning which produces the strong katabatic deviation correlations of spring. The actual mechanics of the diel sea breeze and closer examination of the diel cycle in isotopes related to it are covered in the later diel section.

• l 469: “com”?
L 469: Edited

section 7: this section is actually a deeper analysis of the previous paragraph. Therefore, I suggest to move this section into a last sub-section of section 6. It could be renamed ”6.5. Moisture pulse events”. The content can be merged with the last paragraph of section 6.4 to avoid redundancy
Section 7 (now 5.6): With the reframing of section 6 (now 5), this moisture pulse section was added as a final subheading of that section.

l 611: Why can’t it just be the effect of boundary layer mixing? At midday, boundary layer mixing is more intense, bringing more depleted and high-dxs water vapor from the free troposphere down to the surface, e.g. the mechanism that you describe in lines 490-493.
L 611: Line added to include this possibility. However, local evaporation is still a likely contributor as well, so both options are presented here.

l 661: Start a new paragraph?
L 661: Paragraph structure altered

Figure 3: what are the white and magenta lines? Explain in caption
Figure 3: Clarification added to caption (the lines show the 95% and 5% sea ice concentration)

Figure 6: what is the time scale for the correlations? 10 minute, 1 hour or daily? Explain in caption.
Figure 6: Resolutions used in calculations clarified in caption.

RC3 Response
This paper presents a two-year time series of high time resolution water vapour isotope measurements from Thule in the northern Baffin Bay with the aim to investigate the synoptic drivers of the isotope variability measured in the region of the Greenland high Arctic. Five interacting factors are presented, that are thought to determine the isotope signals’ variability at the daily to annual timescales. These factors include mainly local environmental conditions

(temperature, marine moisture availability, surface winds, NAO and the contribution of land evaporative sources). The relative contribution of the different factors is thought to change with the seasonal cycle and in the coming years with the interannual variability in the extent of the sea ice. Overall, I found this well-written paper inspiring to read, it presents good quality measurement data, shows carefully compiled figures and several interesting analyses. I found the discussion related to the role of sea ice particularly interesting. I have three major comment on the science as well as a few minor comments listed below.

The analysis on the five factors determining the stable water vapour isotope variability measured at Thule at different timescales is very interesting. However, I had difficulties to evaluate the independence of these five factors and also found them to be chosen in a subjective way. The authors give no motivational framework of the basic physical mechanisms that would justify choosing these 5 factors as basic variables that determine isotopic variations. Could the authors provide a more thorough introduction into why they think these five factors are the relevant ones to be studied? Others would be just as relevant such as e.g. the relative humidity with respect to sea surface temperature, cloud condensation temperature or sea surface temperature, which are the traditional variables that are studied as environmental controls of stable water isotope variability. For these traditional variables, physical frameworks exist that explain why they are relevant: e.g. the formation of clouds during moist adiabatic ascent of air parcels (Rayleigh distillation framework, Dansgaard 1964) for cloud condensation temperatures and the Craig and Gordon 1965 ocean evaporation model for SST and the relative humidity with respect to SST.
These five factors were the variables that emerged as primary influences on and/or strongly related with the water vapor isotopic variability in our analyses. Our phrasing is altered slightly throughout the manuscript to be more clear of this fact. As our study was based on the comparative analysis of high frequency field observations (isotopes, weather), our results will of course highlight factors drawn from those observations. We do not exclude the influence of additional factors that we did not personally have observations for (e.g., cloud condensation temp, moisture source conditions), and these factors are mentioned in our discussion of the five factors. For example, we discuss Rayleigh distillation in the temperature section and the local marine moisture and synoptic flow/NAO sections both focus heavily on how different environmental conditions at the moisture source produce changes in the water vapor isotopes.

To develop a database of SST and RH at the moisture source and cloud condensation dynamics at a high temporal resolution would require an extensive amount of modeling and computational effort. We do not dispute that such an analysis would be very intriguing and informative, but it falls outside the realm of our study and its focus on observational data. We have highlighted this concept in the implications and conclusions section as valuable potential follow up research. We have also added more context for the moisture sourcing discussion on SST and RH in section 4.2.

To me highlighting the importance of the atmospheric circulation and at the same time underlining the relevance of local environmental conditions is somewhat contradictory. Many previous studies have used trajectory analysis to show the relevance of environmental conditions at the moisture source for the variability of stable water isotope measurements in water vapour (e.g. Pfahl and Wernli 2008; Aemisegger et al. 2014, Aemisegger 2018; Thurnherr et al. 2020). Here the authors say the circulation and the local conditions are key. I would find it useful, if there was a comment on this apparent contradiction in the paper. Or if

it is not a contradiction, then to resolve the misunderstanding and explain why the results of this paper are in agreement with these previous studies.
We do not see a contradiction in our argument that both the synoptic circulation and local conditions play a role in determining the final isotopic signature observed at Thule. Moisture source conditions are indeed very important to determining the vapor isotopes at a location, particularly when determining the mean values over longer periods of weeks or seasons. We fully acknowledge this in our discussion and particularly in sections 4.2, 4.3, 5.1, and 5.6.

However, on shorter scales it is certainly well-documented that local variables, such as sea breezes (Kopec 2014, Bréant 2019), dew condensation (Bastrikov 2014), and vapor exchange (Casado 2016) play a minor to major role in further modifying the isotopic composition of a moisture parcel upon arrival, particularly for $\delta^{18}O$ and $\delta^{2}H$ even if *dxs* better preserves the original moisture source signal. Additionally, our data highlight that many of these local variables are important only for parts of the year (e.g., sea breezes in summer) and thus may not be well-captured as an isotopic control in studies that focus mainly on factors that consistently can explain isotopic variability throughout the year or at coarser time resolutions. While atmospheric circulation and initial moisture source conditions may be able to explain a significant portion of longer-term isotopic variability, we believe it is a worthwhile endeavor to consider whether local environmental variability can explain the 'noise' left unexplained by only considering atmospheric circulation as a control. At their core, most of these local factors are, in fact, moisture source and routing effects, but examined at a more localized scale than traditional moisture source back trajectory-based studies. For example, the sea breeze-katabatic isotopic dichotomy is based on two different moisture sources, but back trajectory analysis may fail to capture this and other important mesoscale climatology influences due to coarse global circulation models and reanalysis data.

Additionally, we feel that highlighting the influence of local factors helps illustrate how the isotopic signature from a moisture source/specific circulation gets expressed/enhanced/preserved at the local scale. As we display in Figure 5, the local factors are tightly integrated with atmospheric circulation, and while one could simplify the system by subsuming all the local factors into the initiating atmospheric circulation variable, we feel this does a disservice to understanding the inner mechanisms of a complex system.

My third major comment is a more technical one: the presentation of the calibration and postprocessing framework of the exceptionally long and very valuable Arctic water vapor isotope time series lacks some details in particular on the total uncertainty of the measurements (see also my minor comments 5-11, below).
We have attempted to address issues with the calibration and improve the transparency of our uncertainties and unknowns throughout. See specific comments below and in other reviewers for details.

Minor Comments
P. 1, L. 1: The sea ice extent seems to come out as the most important factor controlling if moisture is mainly sourced from the local environment or if it is transported from further away. This could be mentioned more clearly in the abstract before the five controlling factors. In my opinion it comes a bit late in the current version.
Abstract reworded to emphasize sea ice role earlier.

P. 2, L. 50: There were many studies investigating the quality of laser spectrometric measurements in the early 2010s, add "e.g." and maybe Sturm and Knohl 2012 and Aemisegger et al. 2012 could be cited as well, since the latter study particularly focused on the capability of laser systems to resolve the synoptic timescale variability of water vapour isotopes.
References added.

P. 2, L.50: The Yale database could be cited here, since it groups most of the already published water vapour isotope data: Wei et al. 2019.
Reference added.

P. 3, L. 75 "critically giving a second set of observations to derive annual patterns and anomalies" not sure if I understand this correctly. What do the authors mean here?
Changed to: "permitting comparative multi-year analysis of isotopic patterns and anomalies". We are highlighting that we can better determine what is typical and atypical isotopic variability since we have more than just a single year's observations.

P. 5, L. 140: Did the authors test the response times of their system using "Tygon tubing". Several early studies (e.g. Sturm and Knohl 2010; Tremoy et al. 2011; Aemisegger et al. 2012) showed that certain tubing materials induce very large residence times and unwanted strong interactions between the tubing wall and the sample gas.
We observed no clear evidence of tubing effects on our observed data. This is now clarified in the methods.

6) P. 5, L. 140: What was the residence time of the sampled air in the tubing, how long was the tubing, was it heated, was the inlet shielded? These are all very important points for performing high quality stable water vapour isotope measurements especially in extreme environments such as in northern Greenland.
Information added to methods. Also, more detailed information on the tubing set up was found and corrected in the manuscript.

7) P. 5, L. 151: Introduce the delta notation and the normalisation to the international VSMOW-VSLAP scale.
Included.

8) P.6, L. 155: The standards' isotopic composition does not bracket the measured isotope signals. The authors should explicitly mention this and comment on the expected impact of this extrapolation on the total uncertainty of their measurements.
This is now explicitly mentioned and commented on.

9) P. 6, L. 175: Even though the drift of the Picarro laser spectrometers is limited regular calibrations should be carried out to 1) survey the good functioning of the system and 2) to provide a long term assessment of the total uncertainty of the measurements (see, Aemisegger et al. 2012; Thurnherr et al. 2020). In particular, Thurnherr et al. 2020 shows that different post processing procedures lead to substantial changes in the isotope data, in particular, with respect to the treatment of the water vapour mixing ratio dependent isotope bias correction.

We acknowledge the clear benefits that regular calibrations bring and did not intend to come across as dismissing their importance. The initial operational plan did have twice daily calibrations, but unfortunately the Drierite system failed to adequately work and assembling the funding and logistics to install a dry air system at Thule took a long time. We have rephrased the section to hopefully be more clear that this is a source of unquantified possible uncertainty.

10) P. 6, L. 181: The precision (Allan variance, or standard deviation of a constant water vapour isotope signal) strongly depends on the water vapour mixing ratio (see Aemisegger et al. 2012 and Sodemann et al. 2017). Please indicate the total uncertainty of the measurements as a function of water vapour mixing ratio. This is very important, given the very low levels of humidity observed at Thule in winter.
Estimates of precision from humidity response are now given in the manuscript and with more detail in S2 and Table S5.

11) P. 7, L. 195: Was the water vapour mixing ratio of the L2130 calibrated using a dew point generator or another humidity sensor installed in parallel? Without calibration the reading of the laser spectrometric volume mixing ratio may be biased.
The mixing ratio of the L2130 compares very well with mixing ratio calculated from our weather station data. We have added additional information to show this and clarified the consistent factor difference between the L2130 mixing ratio and the weather station ratio (x1.23).

12) P. 8, L. 225: Please indicated the horizontal and vertical grid resolution of the MERRA-2 reanalysis data. Note that HYSPLIT is not a model but a post-processing tool, thus it cannot be "forced". I would suggest to write: "… with air parcel backtrajectories calculated based on three-dimensional MERRA-2 wind fields…". I am not convinced that choosing only 10 days per months produces a robust two-year climatology. But given the limited use that is made of the trajectory climatology in this paper, the approach is ok.
Phrasing suggestions and grid resolution added. "Robust representation" changed to "acceptable representation" to signify that we were not attempting to perform an intense analysis with the trajectory results, but rather add exploratory context.

13) P. 9, L. 254: "The magnitude of irregular hourly to weekly variations" do you mean synoptic timescale variations?
This section has been changed somewhat, but this particular sentence now makes it clear that we attribute these irregular hourly-weekly variations to synoptic events. "The water vapor isotopes also have substantial sub-seasonal variability, and the magnitude of irregular hourly to weekly variations caused by synoptic scale weather events can approach 30–50% of the entire annual isotopic ranges."

14) P. 10, L. 291: What is meant by "temperature-driven equilibrium fractionation"? I think the cited literature is a bit misleading. Dütsch et al. 2017 shows that the condensation temperature indeed has a certain impact on the deuterium excess. Pfahl and Sodemann et al. 2014 discuss the effect of the SST.
Edited to have cited literature and statements more clear and in agreement.

15) P. 10, L. 293: Interesting that a negative correlation between the deuterium excess and the temperature is found! Did the authors also look at the correlation with

nearby SSTs? In climate reconstructions based on ice cores a positive relation between the deuterium excess and moisture source SST is assumed (e.g. Johnsen et al. 1989; Vimeux et al. 1999; Stenni et al. 2001). However, in a detailed analysis of the correlation behaviour between the deuterium excess and SST a recent study (Aemisegger and Sjolte 2018) found different regions (in particular at high latitudes) that are expected to exhibit a negative correlation based on the Craig Gordon model and the closure assumption. It is thought that this negative correlation arises from a positive feedback mechanism between the SST and the relative humidity with respect to SST. Such a negative correlation regime is expected to be dominant particularly in regions where the variability in air-sea interactions is mainly driven by variability in atmospheric circulation and not primarily by variations in ocean circulation. If the deuterium excess also shows a negative correlation to the nearby SST at Thule this would be evidence for such a behaviour. Of course, the time series at Thule is too short to look at this in detail. But still, I find the negative correlation between air temperature and deuterium excess that is found here very interesting and since it is of opposite sign with respect to the traditional interpretation of the deuterium excess in ice core studies, I would find it worthwhile to shortly discuss this.

This is an interesting and intriguing angle of study, but we did not compare our dxs with SST, because determining the exact moisture source and extracting a SST from that moisture source in a consistent, high temporal resolution manner across our entire record was beyond the bounds and abilities of our study. However, we would highlight for you our finding from Figure 6a, where the relationship between dxs and air temperature is opposite between summer and non-summer months. In summer, we do observe a strong positive correlation between air temperature and dxs while outside of summer, it has a strong negative correlation.

The interpretation of dxs in ice cores you mentioned largely pertains to reconstructing past SSTs, and not reconstructing past air temperature. At Thule, air temperature is likely a poor proxy for moisture source SST because the strong seasonal insolation variations and local topographic influences such as downsloping and katabatic winds. As a result, it is difficult to robustly link dxs-air temp correlations and patterns to dxs-SST interpretations with our data. We have included a reference to your proposal in our conclusion as a direction of possible future research.

16) P. 11, L. 320: The relative humidity is very low above forming ice? Can the authors show some evidence or cite some literature?

This comes from the cited Kurita 2011, but the sentence has been rephrased to be more clear about the origin of the dry air in these regions.

17) P. 11, L. 323: Phase changes in the NAO might be confusing in the context of isotopes and water phase changes. Is there another way to formulate the change in the NAO sign?

Phase is the most common and recognized term for this, and we struggled to come up with an alternative that was as clear. However, we acknowledge the potential for confusion with water phases, and some of the phrasing is altered to make it clear that it is the NAO phase being discussed.

18) P. 12, L341-L356: Very interesting discussion on the role of the NAO and the sea ice extent!

19) P. 14, L. 394: I think the relations to the "traditional driving variables" SST and relative humidity with respect to the SST should at least be shortly mentioned.
This is now mentioned earlier in the discussion.

20) P. 14, L. 396 and 399: "southerly" flow instead of "southern" flow.
Changed throughout manuscript.

21) P. 15, Section 6.3 How large is the influence of more distant land sources of e.g. northern Canada? From Fig. 3 one might think that they may play a significant role.
The moisture uptake from the land regions is generally light compared to that of open ocean and Baffin Bay. Additionally, moisture arriving at Thule from these land areas would have crossed substantial open water and potentially isotopically exchanged to a high degree. A focused follow up research project could attempt to key in on specific times where the dominant moisture transport to Thule was from Canadian land and look for anomalous isotopic responses (and in fact, some work related to this is close to submission by one of the co-authors). However, since our quasi-climatology was simply constructed to give broad context on moisture sourcing, we do not have such day-specific analytical data in our database.

22) P. 18, Section 7: Additionally, I suspect that Arctic anticyclones would play a key role in the synoptic timescale variability at Thule (in Greenland blocking situations). A case study of an Arctic Blocking event (over northern Russia) is shown in Schneider et al. 2019, which highlights the very large horizontal gradients resulting from the subsidence induced drying at the core of the anticyclone and the progressive atmospheric moisture uptake along the anticyclone edges.
Anticyclones now more prominently mentioned in synoptic circulation discussion.

23) P. 18, L. 542: I doubt that in the case of sea ice much water vapour at Thule originates from the deep tropics or subtropics. The analysis shown in Fig. 3 does not support such a statement. Here I think one can safely write "advection from the midlatitudes".
Edited.

24) P. 21, L. 609: I don't understand why an input of low $d_{18}O$/high dxs from evaposublimation results in a $d_{18}O$ maximum and dxs minimum at midday.
The latter max and min were erroneously switched in the text. It is in fact a $\delta^{18}O$ min and dxs max that occurs at midday, and this has been corrected in the text.

25) P. 21, L. 627: It would be great to clearly mention that evaporation of meltwater and sublimation of snow might not carry the same isotopic composition (see e.g. Christner et al. 2017).
This is now mentioned in the initial paragraph discussing evaposublimation.

26) P. 22, L. 664: Maybe not single extreme events but years with high frequency of occurrence of warm advection events, e.g. due to a northward shift of the storm track?
Edited to include this possibility.

27) P. 23: In the conclusion it would be very nice to mention the great use of this dataset

for the validation of high-resolution isotope-enabled simulations in the Arctic to study the importance of air-ice and air-sea interaction processes in more detail. Here the great value of the data is their long temporal coverage, thanks to which modelbased sensitivity experiments could be performed to test the importance of different driving factors (similar to the climatological sensitivity study over Europe by Christner et al. 2018).

Mentioned in the conclusion now.

Baffin Bay sea ice extent and synoptic moisture transport drive water vapor isotope ($\delta^{18}O$, $\delta^{2}H$, deuterium excess) variability in coastal northwest Greenland

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Abstract

At Thule Air Base on the coast of Baffin Bay (76.51° N 68.74° W), we continuously measured water vapor isotopes ($\delta^{18}O$, $\delta^{2}H$) at high frequency (1 s^{-1}) from August 2017 through August 2019. Our resulting record, including derived deuterium excess (*dxs*) values, allows analysis of isotopic–meteorological relationships at an unprecedented level of detail and duration for High Arctic Greenland. We examine isotopic variability across multiple temporal scales from daily to interannual, revealing that isotopic values at Thule are predominantly controlled by sea ice extent in northern Baffin Bay and synoptic flow pattern. This relationship can be identified through its expression in five interacting factors: a) local air temperature, b) local marine moisture availability, c) the North Atlantic Oscillation (NAO), d) surface wind regime, and e) land-based evaporation/sublimation. Each factor's relative importance changes based on temporal scale and in response to seasonal shifts in Thule's environment. Winter sea ice coverage forces distant sourcing of vapor that is isotopically light from fractionation during transport while preventing isotopic exchange with local waters. Sea ice breakup in late spring triggers a rapid isotopic change at Thule as the newly open ocean supplies warmth and moisture that has ~10‰ and ~70‰ higher $\delta^{18}O$ and $\delta^{2}H$ values, respectively, and ~10‰ lower *dxs* values. Sea ice retreat also leads to other environmental changes, such as sea breeze development, that radically alter the nature of relationships between isotopes and many meteorological variables in summer. On synoptic time scales, enhanced southerly flow promoted by negative NAO conditions produce higher $\delta^{18}O$ and $\delta^{2}H$ values and lower *dxs* values. Diel isotopic cycles are generally very small as a result of a moderated coastal climate and counteracting isotopic effects of the sea breeze, local evaporation, and convection. Future losses in Baffin Bay sea ice extent will likely shift mean annual isotopic compositions toward more summer-like values, and local glacial ice could potentially preserve isotopic

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evidence of past reductions. These findings highlight the influence that the local environment can have on isotope dynamics and the need for dedicated, multi-season monitoring to fully understand the controls on water vapor isotope variability.

1. Introduction

The Arctic environment is rapidly entering a new state dominated by warmer air temperatures in all seasons, accompanied by dramatic sea ice loss, ecological changes, and ice sheet mass loss. Improving our knowledge of how the Arctic water cycle responds to regional warming and develops feedbacks within the changing climate is key to planning for a more resilient Arctic environment and economy (Meier et al., 2006; Vihma et al., 2016). While many modeling and satellite-based studies are examining this issue (NOAA, 2020), ground-based observations are still critical to understanding the present-day hydrological cycle and tracking ongoing changes in local environments (e.g., Steen-Larsen et al., 2014; Bonne et al., 2015; Klein et al., 2015). Despite this need, the Arctic is data-sparse in terms of spatial coverage and temporal span of quality surface observations. More environmental monitoring efforts are needed across the region and particularly at the highest latitudes to better capture the hydroclimate processes and moisture transport across the Arctic domain.

The stable isotopes of water are well-established environmental tracers of the water cycle, reflecting both local weather conditions as well as moisture history and synoptic patterns (Craig, 1961; Dansgaard, 1964; Rozanski et al., 1993; Gat, 1996). Although less commonly studied than lower latitude regions, analyses across the Arctic have also shown that local observations of water isotopes can reveal important connections to wider atmospheric parameters such as teleconnections and storm patterns (e.g., Moorman et al., 1996; Welker et al., 2005; Theakstone, 2011; Bailey et al., 2015b; Puntsag et al., 2016; Putman et al., 2017). While our knowledge of isotope dynamics in the hydrosphere is largely based on studies of precipitation and surface waters (e.g., Dansgaard, 1964; Rozanski et al., 1993; Welker, 2000; Gurney and Lawrence, 2004), the development of field-deployable infrared laser spectrometers has fostered a number of recent studies focused on water vapor. With their ability to analyze water isotopes with high frequency (>1 min^{-1}) in a continuous vapor flow, these spectrometers are well-suited to long-term monitoring studies (e.g., Sturm and Knohl, 2010; Aemisegger et al., 2012; Bailey et al., 2015a; Wei et al., 2019). Several studies at land-based sites in the high latitudes have reported continuous water vapor isotopic observations for periods ranging from a single season to multiple years (Table S1), and their data sets are proving highly useful in understanding polar hydroclimate dynamics.

These observational studies are critical to tracking and understanding the ongoing climate changes in high latitude regions, especially as the impacts of amplified polar warming on global weather patterns are hotly debated (Francis and Vavrus, 2012; Francis et al., 2018; Pithan et al., 2018; Nusbaumer et al., 2019; Cohen et al., 2020). Already, the published results from these polar water vapor isotope sites have highlighted the potential in tracking shifting moisture sources during extreme weather events (Bonne et al., 2015; Klein et al., 2015) as well as the importance of local geography in short-term isotopic variability (e.g., Kopec et al., 2014; Bréant et al., 2019). However, these sites reflect only a small portion of the vast and diverse polar

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environment, and fewer than half report data covering multiple years. More spatial coverage and longer periods of record are needed to fully harness water vapor isotope monitoring to resolve unanswered questions on polar hydroclimate processes and to accurately predict and detect future changes.

Greenland is particularly important to our efforts to better understand past, current, and future climate, as its immense ice sheet has archived millennia of past climate changes (e.g., Steffensen et al., 2008) and environmental feedbacks from the increasing Greenland Ice Sheet loss reverberate globally (e.g., Box et al., 2012; Nghiem et al., 2012; Castro de la Guardia et al., 2015). The declining extent and duration of sea ice in its surrounding oceans are altering atmospheric moisture fluxes and transport across the Arctic (Gimeno et al., 2019; Nusbaumer et al., 2019), restructuring marine and terrestrial ecology (Bhatt et al., 2017; Laidre et al., 2020), and harming the health and welfare of native communities (Meier et al., 2006). Northwest Greenland in particular is one of the fastest warming regions on Earth with massive ice loss observed from glacial retreat and surface ablation (van As, 2011; Carr et al., 2013; Noël et al., 2019). Despite Greenland's importance, water vapor isotope monitoring has only been reported from four locations on the island (Steen-Larsen et al., 2013; Bonne et al., 2014; Kopec et al., 2014; Bailey et al., 2015a), and one record (Summit) has not had its data published.

We present here a new multi-year dataset from Thule Air Base in northwest Greenland that has recorded stable oxygen and hydrogen ratios ($\delta^{18}O$ and $\delta^{2}H$, respectively) of ambient water vapor nearly continuously from 04 August 2017 through 31 August 2019. As a result, this record is the first reported for Greenland that continuously spans over two years, permitting comparative multi-year analysis of isotopic patterns and anomalies. Thule Air Base has been a focus of High Arctic research for many years (e.g., Schytt, 1955; Mastenbrook, 1968; Sullivan et al., 2008; Rogers et al., 2011; Leffler and Welker, 2013; Schaeffer et al., 2013), and our observing station on northern Baffin Bay allows a focus on how changing seasonal and interannual sea ice coverage affect the local climate and water vapor isotopes.

Our research addresses how interactions between the hydroclimate, cryosphere, and ocean are manifested in water vapor isotopes, and we focus here on identifying how these broad interactions are expressed and detected in local weather observations. The long period of record and very high frequency of isotopic (one observation per second, aggregated to 10 minutes) and meteorological observations (one observation per 10 minutes) enable us to study these water vapor isotope dynamics across varying temporal scales from daily to interannual. This Thule dataset and our continuing observations are part of the MOSAiC (Multidisciplinary drifting Observatory for the Study of Arctic Climate) project's Arctic Water Isotope Network (Welker et al., 2019; MOSAiC, 2020), a series of nine pan-Arctic sites simultaneously observing water vapor isotopes. To that end, our research at Thule provides a focused examination of the many local and regional environmental controls on water vapor isotopes at a coastal High Arctic site in northwest Greenland.

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2. Field description

2.1 Local landscape

Our water vapor isotope observation station is located at Thule Air Base on the Pituffik Peninsula in far northwest Greenland (Fig. 1). Landmarks are referred to in this paper by their locally-known English names with Greenlandic names also listed where known. The main base and airfield, established in 1951 by the United States and Denmark, occupy a low-lying (< 100 m a.s.l.) region along the North River (Pitugfiup Kûgssua) facing North Star Bay and Bylot Sound, small arms of Baffin Bay that are semi-protected by Saunders and Wolstenholme Islands (Appat and Qeqertarsuaq). The main base is bracketed to the south and north by two broad ridges (summits: 300 and 240 m a.s.l., respectively) locally called South Mountain (Akínarssuaq) and North Mountain. The land-terminating margin of the Greenland Ice Sheet is approximately 14 km southeast of the main base and extends as low as 300 m a.s.l. This local section of the ice sheet, known as the Tuto Ice Dome, has a summit greater than 800 m a.s.l. and a mass balance that is semi-independent from the main Greenland Ice Sheet (Schytt, 1955; Hooke, 1970; Reeh et al., 1990).

Other military installations, both abandoned and presently occupied, are scattered throughout the wider Thule Defense Zone that covers the northern half of the Pituffik Peninsula. Near the southern edge of the Thule Defense Zone lies higher terrain including P-Mountain (Pingorssuit), the highest point of the peninsula at 815 m. In addition, rugged terrain and mountains over 600 m a.s.l. run along the southern coastline of the peninsula toward Cape York. Wolstenholme Fjord (Uummannap Kangerlua) forms the northern coast of the Pituffik Peninsula, separating it from Steensby Land and the North Ice Cap (summit: >1200 m). The three marine terminating glaciers that feed into the fjord have severely retreated and thinned in the past century, including a retreat of over 8 km for the largest glacier (Harald Moltke) since the 1940s (Mock, 1966, Hill et al., 2018).

2.2 Local climate

Thule has a polar desert/semi-desert climate (Gold and Bliss, 1995; Sullivan et al., 2008), with a mean annual temperature of -10.0°C and mean monthly temperatures ranging from -23.7°C in February to +6.5°C in July (2000–2018 observations, USAF, 2019). The high latitude (76° N) produces long periods of polar night (November–February) and midnight sun (May–August), and daylength changes by 15–30 minutes each day during transitional months. Extended periods of extremely cold temperatures below -25°C are frequent in winter, and frosts and snowfall are possible in all months. Mean monthly temperatures are above freezing for only the three summer months, but summer can be surprisingly mild under continuous insolation with inland temperatures sometimes rising above 10°C. Coastal sea ice and the seasonal snow pack develop by October and last through May to early June (Barber et al., 2001; Fetterer et al., 2017; Stroeve and Meier, 2018; USAF, 2019). At early spring maximum, sea ice covers Baffin Bay to an extent over 1000 km south of Thule, except for the biologically important North Water Polynya located to the northwest (Barber et al., 2001; Tang et al., 2004; Heide-Jørgensen et al., 2016).

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Annual precipitation is 130 mm water equivalent with half of this precipitation falling mostly as rain in June–August (USAF, 2019). Moisture in western Greenland primarily is sourced from the North Atlantic, but local sources of moisture, such as Baffin Bay, increase in importance during sea ice retreat in summer and early fall (Sodemann et al., 2008; Gimeno et al., 2019; Nusbaumer et al., 2019). Although the wind at Thule makes accurate snow measurements difficult and prone to overestimation (Chen et al., 1997), existing records report an average annual snowfall of 900 mm and October–December as the snowiest months (USAF, 2019). Synoptic storm systems dominate short-term weather variability with the prevailing storm track consisting of extratropical cyclones that form to the south in Labrador Bay before tracking north and strengthening over Baffin Bay (Chen et al., 1997). These cyclones can be very intense, including a 333 km h^{-1} observation at Thule in 1972 that is among the highest winds ever recorded on Earth (Stansfield, 1972; Moore, 2016). Atmospheric rivers (narrow corridors of strong horizontal moisture advection) have outsized effects on polar weather (Woods et al., 2013; Liu and Barnes, 2015; Wille et al., 2019), and periods of intense ice sheet melt and mass loss in Greenland, such as July 2012, often coincide with atmospheric rivers impacting the ice sheet (Neff et al., 2014; Bonne et al., 2015; Mattingly et al., 2018; Ballinger et al., 2019; Oltmanns et al., 2019).

Differential radiative heat loss between the ice sheet and coastal region drives katabatic winds (van As et al., 2014) from the east and southeast that dominate the wind regime in Thule outside of the summer months. From April through September, a west to northwest sea breeze develops along the coast due to differential warming between the ocean and the local area of snow-free tundra (Atkinson, 1981), and it brings cooler marine surface air, often associated with fog, several kilometers inland. This sea breeze generally strengthens in the afternoon and weakens at night, although continuous summer insolation often prevents the full shift to nightly katabatic flow that has been observed at other coastal polar sites (e.g., Kopec et al., 2014). These two wind patterns at Thule produce an overall bimodal wind distribution with azimuth peaks at 100° (katabatic) and 270° (sea breeze).

3. Methods

3.1 Equipment setting

A Picarro L2130-i with Standards Delivery Module (SDM) setup was installed in a temperature-controlled building on the crest of South Mountain (76.514° N 68.744° W, 229 m a.s.l.) in October 2016, with quality data observations beginning in August 2017 after a system reset. Local air flow on top of South Mountain is unimpeded by topographic barriers from any direction within 20 km. Potential anthropogenic impacts to observations at the monitoring site are limited as the main base lies 2 km to the north and 200 m lower in elevation. Typically, less than 20 vehicles a day travel the access road that is 200 m from the building housing the L2130-i, and visitors are uncommon.

The L2130-i uses cavity ring-down spectroscopy to measure $\delta^{18}O$ and $\delta^{2}H$ in ambient water vapor and is particularly suited to long-term monitoring with limited human interaction. The vapor collection point is five meters above the ground surface on

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the building's roof-edge, away from parking zones and building exhaust, and outfitted with a plastic cap to prevent precipitation entering the inlet. Five meters of Bev-A-Line IV EVA tubing (3/4 in diameter) connects the inlet to the L2130-i with an air residence time of less than three minutes. The tubing is not independently heated, but it is routed immediately into the heated building interior which is always warmer than ambient outside air, even during extreme warm events in summer. This limits potential condensation within the tubing, and data quality control included a cull on observations with abnormal isotopic values that suggested possible condensation or precipitation contamination.

Certain tubing types, such as Synflex tubing, are known to adversely affect water vapor isotopic values by a slower vapor response time, increased fractionation of δ^2H, and general smoothing of δ^2H values at timescales less than 30 minutes (Sturm and Knohl 2010; Tremoy et al., 2011). We see no evidence of substantial tubing effects on our data, as the δ^2H has the same response time and degree of variability as $\delta^{18}O$ in our 10 minute resolution data, and the strongest $\delta^{18}O$–δ^2H correlations are with zero time lag. Additionally, we do not observe any significant differences in the $\delta^{18}O$–δ^2H relationship between 10 minute, hourly, and daily aggregations which supports that both isotopic values are varying at a similar short-term frequency.

3.2 Isotopic observations and calibrations

Our L2130-i at Thule took an observation of the oxygen and hydrogen isotopic ratios in ambient water vapor approximately once per second, and these ratios are expressed in δ-notation relative to the Vienna Standard Mean Ocean Water (VSMOW) standard as:

$$\delta = \frac{R}{R_{VSMOW}} - 1 \quad (1)$$

where R is the measured ratio of rare to abundant isotopologue (^{18}O / ^{16}O or ^2H / ^1H) and R_{VSMOW} is the matching isotopic ratio of the reference standard water VSMOW. Deuterium excess values (*dxs*) were calculated from each isotope observation by $dxs = \delta^2H - 8 * \delta^{18}O$ (Dansgaard, 1964). Observations were continuous from August 2017 through August 2019 except for one period when the analyzer suffered power failure (05–13 Sep 2018). Some additional gaps in the data were introduced as a result of quality checking (S1) and standard calibrations, but these gaps were typically less than 24 hours in duration. We acknowledge that technical difficulties and some initial system design choices have prevented our data from being calibrated to the maximum quality level recommended by peer publications, and we attempt to be fully forthright with these issues here.

Every 25 hours, two water standards, USGS 45 ($\delta^{18}O$ = -2.24‰, δ^2H = -10.3‰) and USGS 46 ($\delta^{18}O$ = -29.80‰, δ^2H = -235.8‰), were injected sequentially by the SDM into a stream of air supplied by a dry air canister in order to normalize to the VSMOW/SLAP (Standard Light Antarctic Precipitation) scale. Each standard was injected for ten minutes, and the mean of the last 200 seconds of each standard injection was kept as the final isotopic value for each standard calibration. Preliminary tests at Thule suggested that acceptably stable isotopic value readings were reached in less than ten minutes, and in 51 calibrations that were run in August and September 2019 (Table S2), the final 200 observations (i.e., the data used to calculate

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calibration values) have flat trends suggesting acceptable isotopic stability (Table S3; mean slope ± 1 standard deviation (σ) for USGS 45: $\delta^{18}O$ = -0.0003 ± 0.0016‰ s^{-1}, $\delta^{2}H$ = 0.0026 ± 0.0062‰ s^{-1}; USGS 46: $\delta^{18}O$ = 0.0014 ± 0.0016‰ s^{-1}, $\delta^{2}H$ = 0.0026 ± 0.0064‰ s^{-1}). Our calibration interval and duration is longer and shorter, respectively, than in other similar studies (e.g., Bastrikov et al., 2014; Bailey et al., 2015a; Bréant et al., 2019), but deemed a necessary compromise to extend the life of the dry air canister and standard waters at the remote location while still allowing an estimate of the machine's accuracy drift.

The water standards used in calibration were originally chosen to cover the summer water vapor isotopic values at Thule before a project extension, but unfortunately most non-summer isotopic values are below the standard waters' value coverage. As a result, our reported values for non-summer observations may suffer from undetermined accuracy biases relative to the VSMOW/SLAP scale, particularly at the lowest isotopic values. The potential impact on our analyses is likely limited as long as any bias is internally consistent and/or small in magnitude, and comparison of our reported isotopic values to other polar sites did not reveal any clear sign of major inaccuracies. However, anyone performing a future comparison between our Thule data and the data from another site should certainly take into account the potential for unquantified accuracy bias below the isotopic values of USGS 46. A wider range of standard waters is planned for continued future operation of the Thule observation station.

At lower water vapor mixing ratios (<1500 ppmv), the L2130-i loses accuracy and precision, and a humidity response curve must be developed in order to correct for any analytical bias in isotopic values (Steen-Larsen et al., 2013; Bastrikov et al., 2014; Bailey et al., 2015a). In July 2019, the two standard waters were each injected for 10 minutes into dry air at ten different flow rates to produce a sequence of standard observations between 500 and 7000 ppmv. Humidity response curves were created using nonlinear regression to relate isotopic value offsets due to sensor biases at low humidity with water vapor mixing ratios, and ambient isotopic data were then corrected for sensor bias with these curves based on the ambient mixing ratio at the time of observation (S2, Fig. S1, Table S4). Ambient observations with a mixing ratio less than 500 ppmv (fewer than 500 individual 1 s^{-1} observations) fell outside our humidity response observations and were thus excluded from further analysis.

The analyzer has a consistent accuracy offset of +1.9‰ and +1.1‰ in all $\delta^{18}O$ and $\delta^{2}H$ observations, respectively, at mixing ratios higher than 1500 ppmv. The accuracy bias changes below 1500 ppmv, and the offset for $\delta^{18}O$ and $\delta^{2}H$ is -4.5‰ and -20‰, respectively, at 500 ppmv. Ambient data were corrected for these offsets, and 95% confidence intervals for humidity response corrections for $\delta^{18}O$ ranged from ±0.24‰ (500 ppmv) to ±0.09‰ (all observations >1500 ppmv) and for $\delta^{2}H$ ranged from ±1.6‰ (500 ppmv) to ±0.6‰ (all observations >1500 ppmv). Precision linearly decreases with the reciprocal of the water vapor mixing ratio (S2, Fig. S1, Table S5). Standard errors of the mean $\delta^{18}O$ values for each flow rate in the humidity response curve ranged from 0.57‰ (500 ppmv) to <0.05‰ (>5000 ppmv) while the standard errors for $\delta^{2}H$ ranged from 3.7‰ (500 ppmv) to <0.4‰ (>5000 ppmv). In comparison, each ambient water vapor data point from the highest resolution used in our analysis is mean value of 600 observations (i.e., 10 minutes) compared to the 200 observations in the humidity response curve,

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and thus the precision of our ambient data should as precise or greater than observed in the humidity response curves. After the humidity response corrections, data were quality checked again and offending data (e.g., extreme outliers, residual calibration vapor impacts, extreme moisture spikes unrelated to weather) removed. The final quality checked isotope data were aggregated into 10 minute, hourly, and daily means.

The calibration system described as above was installed in August 2019. Before this installation, standards were injected into an air stream dried by a Drierite column rather than supplied by a dry air canister. However, the Drierite column did not fully remove all ambient moisture and increasingly lost effectiveness over time at Thule. The residual ambient moisture had a significant effect on observed isotopic values during calibrations, and very few of the calibration runs prior to the dry air canister installation accurately recorded the standard values needed to correct for machine analytical drift. As a result, we do not have accurate daily calibrations for most of our observation period and cannot apply a correction to account for possible analytical drift.

We believe that the database can still provide robust analytical results despite the lack of drift correction. Although we cannot be certain that an estimate of recent stability is representative of the stability over the entire observation record at Thule, the calibrations completed after the dry air installation show consistently high precision and limited day to day sensor drift (Table S2). The standard deviation of the 51 calibration means for $\delta^{18}O$ and $\delta^{2}H$ were 0.2‰ and 0.7‰, respectively, with deviations the same for both standard waters, and the maximum difference between any two calibration run means were 1.2‰ and 3‰ for $\delta^{18}O$ and $\delta^{2}H$, respectively. This magnitude of recent drift at Thule and the magnitude reported at other similar stations (e.g., Steen-Larsen et al., 2013; Bréant et al., 2019) is very small relative to the natural short-term isotopic variability and large seasonal patterns that we observe in the ambient water vapor. As our study focuses on these large isotopic changes, the risk of substantial impact on our analyses and conclusions from undetected short-term sensor drift is minor. Comparison of isotope-climate relationships across the period of record and before/after unexpected shutdowns show no major changes that suggest long-term drift or post-restart shifts in instrument sensitivity that would negatively affect our analyses.

3.3 Meteorological observations

An automated weather station (SMT, for South Mountain), located on the roof of the building that houses the L2130-i, took a reading of air temperature, relative humidity (with respect to the saturation vapor pressure over ice), and station barometric pressure every 10 minutes throughout the duration of isotopic observations (Muscari, 2018). For analysis at different temporal resolutions, these data were also aggregated into hourly and daily means. The L2130-i takes a reading of the water vapor mixing ratio with every isotopic observation, and observations of this variable were added to the SMT datasets as an independent meteorological observation. The mixing ratios recorded by the L2130-i have a very strong linear relationship (r^2 = 0.99) with mixing ratios calculated from the SMT observations, but the L2130-i mixing ratios are greater by a consistent factor of 1.23. For analytical simplicity, all mixing ratio values used in analyses and reported here are the unaltered observations from the L2130-i.

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Hourly mean near-surface wind speeds and azimuths (USAF, 2019) were taken at the Thule Airport (THU), aggregated to daily values, and joined to the hourly and daily SMT databases. Wind azimuths were converted into u and v components for accurate aggregation and then reconverted back into azimuths. While the THU recording station is 170 m lower than SMT, the two stations are less than 1 km apart and their meteorological data agree well ($\rho_{temperature} = 0.98$, $\rho_{dew.point} = 0.98$, $\rho_{station.pressure} = 0.96$). Daily climate teleconnection indices for the North Atlantic Oscillation (NAO) and Arctic Oscillation (AO) were downloaded from the US National Weather Service Climate Prediction Center (NOAA, 2019) and joined with the daily SMT database. Because the AO did not produce significant correlations or results in our analyses, its discussion is limited here and largely subsumed under discussion of the NAO. Daily sea ice extent for Baffin Bay was obtained through the National Snow and Ice Data Center (NSIDC) Multisensor Analyzed Sea Ice Extent (MASIE) product (Fetterer et al., 2010).

To reduce issues with the radial nature of azimuth data during correlation analysis, wind azimuths were converted to 'katabatic deviations' where an azimuth of 100° (i.e., the mean katabatic wind azimuth) was defined as zero, and other observations were redefined as their minimum absolute degree distance from 100°. The katabatic deviations therefore range in value between 0° and 180°. While this does not fully solve the radial issue (e.g., a katabatic deviation of 80° can represent both a northly 20° wind or a southerly 180° wind), it fares well in distinguishing between the bimodal "easterly katabatic" and "westerly sea breeze" regimes.

3.4 Statistical analyses

Spearman correlations between isotopic and meteorological variables were calculated across the full datasets at all three temporal resolutions. Strong correlations that arise between variables in this analysis, though, may be spurious due to common responses to seasonal change. Likewise, variables that do not have a seasonal cycle, such as relative humidity, may have stronger correlations once the interference from seasonality is removed. To examine this, we removed the strong seasonal cycles of the isotopic values, air temperature, and mixing ratio by sinusoidal curves fitted to the annual cycle of each full data time series. With these seasonally-adjusted (SA) data, we calculated a second set of correlations.

Data at each temporal resolution were also binned by month, and correlations then calculated for each month to examine how isotope-climate relationships change over the course of a year. To assess temporal covariance and possible lead/lag relationships between teleconnection indices (NAO, AO) and isotopic variables, we performed cross-correlation analysis (Addinsoft, 2020) with a first differencing methodology (Peterson et al., 1998) to remove autocorrelations and avoid cross-correlation bias (Olden and Neff, 2001; Runge et al., 2014).

3.5 Back trajectory analysis

We compiled a Thule-specific moisture sourcing and transportation "quasi-climatology" with air parcel back trajectory analysis using the HYSPLIT tool and based on three-dimensional MERRA-2 wind field data that has a horizontal grid

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resolution of 0.5° latitude / 0.625° longitude and 72 hybrid-eta vertical levels from the surface to 0.01 hPa (Stein et al., 2015; Gelaro et al., 2017). To build this "quasi-climatology", we randomly selected 10 days from each month during 1980–2018 as we judged 10 days to provide an acceptable representation of the true climatology while remaining within reasonable computation and data storage limits. We then initiated 10 day back trajectories for each randomly selected day from the MERRA-2 grid point nearest Thule at the four synoptic hours (0000, 0600, 1200, and 1800 UTC) on 6 vertical levels (10, 100, 200, 500, 1000, and 1500 m above ground level), resulting in 2880 total trajectories. Following previous studies (e.g., Sodemann et al., 2008; Molina and Allen, 2019), the moisture uptake algorithm implemented in the PySPLIT python library (Warner, 2018) infers moisture uptake into the parcel when specific humidity change (Δq) is greater than +0.2 g kg^{-1} and infers precipitation when Δq is less than -0.2 g kg^{-1}. Later moisture uptakes are weighted more heavily than uptakes occurring prior to precipitation (Sodemann et al., 2008). A domain covering Baffin Bay from the Davis Strait to Nares Strait was defined, and the percentage of moisture uptake occurring within and outside this domain was calculated with raw values at each grid cell multiplied by the cosine of latitude to compensate for smaller grid cell area with northward extent.

4. Environmental controls on water vapor isotopes at Thule

The mean annual water vapor $\delta^{18}O$, $\delta^{2}H$, and *dxs* values at Thule are -33.2 ± 6.1‰, -249 ± 43‰, and +16.2 ± 8.1‰, respectively (10 min data, mean ± 1 σ). The isotopic records reveal strong annual cycles, with the highest values for $\delta^{18}O$ and $\delta^{2}H$ in summer and lowest values in winter and early spring, while *dxs* values show the opposite pattern with minimum values in summer and maximum values in winter (Fig. 2, Table 1, S3). The isotopic values also have very wide overall ranges, with $\delta^{18}O$, $\delta^{2}H$, and *dxs* spanning over 30‰ (-49.5‰ to -17.5‰), 230‰ (-377‰ to -142‰), and 55‰ (-7.6‰ to 47.5‰) respectively, in our 10 minute database. Temporal aggregation reduces the overall range and variability of isotopic data by reducing extreme values, but this does not greatly affect the distribution or means. Conclusions drawn from one level of aggregation are consistent across all levels of aggregation.

The water vapor isotopes also have substantial sub-seasonal variability, and the magnitude of irregular hourly to weekly variations caused by synoptic weather events can approach 30–50% of the entire annual isotopic ranges. These large isotopic changes are typically tied to the different sourcing and transport of warm and cold sector moisture during cyclone passage, as well as changes in air temperature, surface winds and cloud cover (Gedzelman and Lawrence, 1990; Coplen et al., 2008; Dütsch et al., 2016). Similarly, strong anticyclones and blocking events can also dominate local weather and alter atmospheric flow throughout the polar region (Davini et al., 2012; McLeod and Mote, 2016; Wernli and Papritz, 2018). Diel cycles in isotopes are very small (generally <1‰ for $\delta^{18}O$ and *dxs*), but they help reveal which environmental factors are driving isotopic changes at the local scale.

Based on our statistical analyses (Table 2) and field observations, the variability of water vapor isotope ratios at Thule can be explained largely by five interacting factors: a) local air temperature, b) local marine moisture availability, c) the NAO, d)

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4. General overview of isotopic results¶
The mean annual $\delta^{18}O$ and δD values at Thule are -33.0 ± 5.9‰ and -248 ± 41‰, respectively (10 min data, mean ± 1 σ). Over the full dataset, $\delta^{18}O$ and δD have a strong linear relationship with low parameter standard error: δD = 6.959 ± 0.003 * $\delta^{18}O$ - 18.07 ± 0.09‰ (r^2 = 0.98, n=111138, 10 min data). Overall, this value is comparable to other slopes observed at other high latitude sites, such as 6.8 at Ivittuut, Greenland, (Bonne et al., 2014), 6.5 at NEEM, Greenland, (Steen-Larsen et al., 2013), 6.0–6.5 at Dome C, Antarctica (Casado et al., 2016), and 6.95 from the vapor mixing line at Kangerlussuaq, Greenland (Kopec et al., 2014). Changes in $\delta^{18}O$ are thus closely mirrored in δD, and most differences are only detectable on very short timescales (i.e., less than hourly) when some minor lead-lag between relative maxima and minima may occur. ¶
The *dxs* at Thule is negatively correlated with both $\delta^{18}O$ and δD (ρ_{10min} = -0.76 and -0.67, respectively). Temporal aggregation reduces the overall range and variability of isotopic data by reducing extreme values, but this does not greatly affect the distribution or means. Generally, conclusions drawn from one level of aggregation are consistent across all levels of aggregation, and no exceptional cases were identified in this study.¶
The isotopic records show a strong annual cycle, with the highest values for $\delta^{18}O$ and δD in summer and lowest values in winter and early spring, while *dxs* values have the opposite pattern with minimum values in summer and maximum values in winter (Fig. 2, Table 1). The isotopic values also have very wide overall ranges, with $\delta^{18}O$, δD, and *dxs* spanning over 30‰ (-49.5‰ to -17.5‰) 230‰ (-377‰ to -142‰), and 55‰ (-7.6‰ to 47.5‰) respectively[4]
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surface wind regimes, and e) the evaporation/sublimation of local surface waters and snow. These factors interact and compete with each other, with the dominance of particular factors varying over the course of a year with the changing seasons. Together, these factors can be considered as specific expressions of the broad environmental responses to changes in Baffin Bay sea ice extent and polar synoptic flow. To clarify this complex situation, this discussion will first detail the effects of these factors specifically in the Thule environment. The impact of these factors on isotopic systematics will then be examined across seasonal, synoptic, and daily scales to reveal how observed isotopic values are produced.

4.1 Local air temperature

Our back trajectory analysis shows that most water vapor is supplied to Thule from evaporation in more southerly locations and then advected north along the western coast, with a substantial fraction of this water vapor having previously passed over the southern Greenland Ice Sheet (Fig. 3a). As the air mass cools in this northerly transport, water vapor will condense into clouds and precipitation that selectively removes isotopically heavier water molecules (Dansgaard, 1964; Rozanski et al., 1993). This Rayleigh fractionation results in an air mass with water vapor that is increasingly isotopically lighter (i.e., depleted in ^{18}O and ^{2}H) as it travels to Thule, and colder conditions at Thule create a steeper temperature gradient versus the more southerly moisture source that enhances the degree of fractionation (e.g., Rozanski et al., 1993; Bonne et al., 2014; Kopec et al., 2019). This connection is supported by the strong correlations of $\delta^{18}O$ and $\delta^{2}H$ with air temperature ($\rho_{\delta 18O.10min} = +0.76$, $\rho_{\delta 2H.10min} = +0.73$), which are surpassed in strength only by correlations with the closely linked water vapor mixing ratio (Table 2).

However, strong annual correlations between water isotopes and temperature often result from common responses between air temperature and other variables that also follow seasonal patterns (e.g., Akers et al., 2017). This also appears true for Thule, as the correlations are much weaker when seasonal cycles are removed ($\rho_{SA\text{-}\delta 18O.10min} = +0.24$, $\rho_{SA\text{-}\delta 2H.10min} = +0.20$). Most likely, much of the strength of the non-seasonally-adjusted correlations is due to the common response with broader environmental changes linked to seasonally-covarying sea ice extent. Despite this, the weaker seasonally-adjusted correlations show that a temperature effect is still expressed to a weaker degree in shorter-term, intra-seasonal isotopic variations (Table 2).

Local air temperature is not typically seen as a primary driver of *dxs* variability; rather, variables related to environmental conditions at the initial moisture source, such as sea surface temperature and relative humidity, are the principal controls (e.g., Merlivat and Jouzel, 1979; Vimeux et al., 1999; Pfahl and Sodemann, 2014). However, *dxs* values at Thule have a strong correlation with air temperature ($\rho_{dxs.10min} = -0.74$), and the correlation using seasonally-adjusted data is actually stronger ($\rho_{SA\text{-}dxs.10min} = -0.31$) than the correlations observed in $\delta^{18}O$ and $\delta^{2}H$. Temperature-driven changes on *dxs* have been reported (Jouzel and Merlivat, 1984; Dütsch et al., 2017; Kopec et al., 2019), particularly for very cold conditions, and it is possible that this is contributing to the short-term correlation between *dxs* and air temperature as well as the strong negative correlations between

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dxs and both $\delta^{18}O$ and $\delta^{2}H$. However, it is also possible that the correlation of *dxs* with temperature is, like with $\delta^{18}O$ and $\delta^{2}H$, actually responding to another environmental factor that covaries with temperature.

4.2 Local marine moisture availability

The isotopic composition of water vapor in an air mass is initially determined by the isotopic composition of the source water and by the environmental conditions during vapor uptake. At synoptic scales, the relative humidity near the sea surface strongly determines the *dxs* values for the resulting moisture parcel, with higher humidity producing lower *dxs* values and vice versa (Craig and Gordon, 1965; Pfahl and Sodemann, 2014) while, at longer timescales, sea surface temperature has traditionally been cited as a primary control on *dxs* values (e.g., Merlivat and Jouzel, 1979; Vimeux et al., 1999). If these isotopic signatures are well-preserved through later moisture transport, isotopic analysis of water vapor or precipitation can be used to remotely infer climate changes at the moisture source and/or shifts to different moisture sources (e.g., Merlivat and Jouzel, 1979; Feng et al., 2009; Bonne et al., 2015; Dütsch et al., 2017; Bonne et al., 2019). This has been applied most notably to ice cores in order to reconstruct moisture source changes extending back deep through time (e.g., Vimeux et al., 1999; Steffensen et al., 2008; Steen-Larsen et al., 2013; Osterberg et al., 2015).

Local polar waters have high surface relative humidity when open in summer (Fig. 3c) and supply Thule with water vapor that has high $\delta^{18}O$ and $\delta^{2}H$ values and low *dxs* values. At Thule, changes in the seasonal sea ice extent permit or restrict the delivery of local moisture from the nearby ocean, and this availability is a primary control on mean isotopic values. Our back trajectory analysis shows that the regions of predominant water vapor uptake for air masses arriving at Thule vary substantially over the year (Fig. 3b), supporting the role of sea ice in determining moisture sourcing. Baffin Bay is the dominant evaporative moisture source from the late spring through early winter, contributing ~50% of water vapor transported to Thule (Table S6). Substantial contributions from the Labrador Sea, Denmark Strait, Hudson Bay, and Canadian Arctic Archipelago regions are also observed during this time.

From January until May, however, local seas are extensively ice covered and the majority of water vapor present at Thule originates from more distant sources in the Labrador Sea and North Atlantic (Fig. 3b). Large evaporation events in the North Atlantic tend to occur in the cold sector of extratropical cycles (Aemisegger, 2018; Aemisegger and Papritz, 2018) where the dry air (Fig. 3c) produces water vapor with high *dxs* values. As this moisture travels north to Thule, fractionation from rain out during transport results in isotopically light water vapor, and sea ice coverage in Baffin Bay and other local Thule waters prevent isotopic exchange that would mitigate some of this fractionation. Interestingly, our analysis reveals that a large amount (>30%) of Thule's winter water vapor is supplied by sections of southern Baffin Bay and the Labrador Sea with 5–95% climatological mean sea ice cover. Moisture originating from such an environment often has very high *dxs* values from the frequent invasions of very dry Arctic air originating over expanses of continuous sea ice (Kurita, 2011) and/or vapor sourcing from snow accumulated on top of the sea ice (Bonne et al., 2019).

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4.3 The North Atlantic Oscillation

Phase shifts in the NAO can serve to enhance or limit moisture transport from the south to Thule (Fig. 3a) as atmospheric mass is redistributed between the Arctic and North Atlantic, and changes in water isotopes resulting from these atmospheric shifts have been detected in Arctic snow and ice (e.g., Vinther et al., 2003; Vinther et al., 2010; Zheng et al., 2018) and plants (Welker et al., 2005). In western Greenland, the negative phase of the NAO (NAO-) is associated with enhanced southerly flow that brings warmer temperatures and greater regional snow, while the positive phase (NAO+) has stronger westerlies that limit the northward penetration of southerly air masses (Sodemann et al., 2008; Bjørk et al., 2018). These effects are manifested in our observations where more negative NAO indices are correlated with a stronger Greenland anticyclone (i.e., higher Thule station pressure, $\rho_{NAO.pres}$ = -0.60).

When southerly advection along the western edge of the anticyclone is promoted during NAO- phases, Thule tends to be warmer ($\rho_{NAO.SA\text{-}temp}$ = -0.31) and have a higher water vapor mixing ratio ($\rho_{NAO.SA\text{-}MR}$ = -0.27). Cross-correlation analysis (Fig. 4, Fig. S2) reveals that shifts to more negative NAO indices produce isotopically heavier water vapor observations with lower *dxs* values two to three days later at Thule, presumably as air advected north over Baffin Bay picks up local moisture and/or the warmer conditions reduce the degree of fractionation during transport. We did not find noteworthy isotopic correlations with the AO at Thule, likely because changes in AO phase relate to pan-Arctic conditions while the NAO changes are more focused on Greenland and the North Atlantic.

The relationship between water vapor isotopes and NAO phase across interannual timeframes at Thule is uncertain from our data's limited period of record, but climatology and ice core studies have argued for clear impacts on Greenland's precipitation, glacial mass balance, and sea ice at this scale (Stern and Heide-Jørgensen, 2003; Vinther et al., 2003; Sodemann et al., 2008; Bjørk et al., 2018). NAO indices in summer 2018 were almost continuously positive, coinciding with relatively cool conditions at Thule and below average melt of the northwestern Greenland Ice Sheet. In contrast, summer 2019 was dominated by NAO- conditions and was nearly 4°C warmer than summer 2018 at Thule (USAF, 2019). Greenland as a whole recorded near-record ice sheet melt over the 2019 melt season, linked in part to extensive warm and sunny conditions over the ice sheet (Maslanik and Stroeve, 1999). Despite these markedly different conditions, mean $\delta^{18}O$ and $\Delta^{2}H$ values are practically identical between the two summers (<0.01‰ and <1‰ different, respectively) and mean *dxs* values differ only by 1‰. This suggests that changes in synoptic flow patterns have little impact when local seas are free of sea ice, as isotopic exchange with local marine moisture can buffer any potential isotopic changes from longer-distance synoptic transport shifts.

However, spring 2018 was also characterized by NAO+ indices and colder conditions while spring 2019 had lower NAO indices and warmer weather. In contrast to the summer observations, $\delta^{18}O$ and $\Delta^{2}H$ values are 4.4‰ and 34‰ higher, respectively, in spring 2019 than spring 2018, and *dxs* values are 1.7‰ lower. As sea ice is still largely intact through spring near Thule, the limited potential for local water input and exchange at this time appears to allow an NAO signal to be expressed

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in the mean isotopic values. Additional seasons and years of observation with different NAO conditions will help clarify the magnitude and seasonal extent of interannual effects from the NAO on Thule water vapor isotopes.

4.4 Surface wind regimes

Between April and September, the surface winds at Thule alternate between east-southeasterly katabatic winds off the nearby ice sheet and a west-northwesterly sea breeze from Bylot Sound, although not necessarily on a regular daily cycle. Such wind shifts at other coastal sites are isotopically identifiable because katabatic winds supply isotopically light vapor from ice sheets while sea breezes supply isotopically heavy vapor from the nearby ocean (e.g., Kopec et al., 2014; Bréant et al., 2019). After adjusting for the seasonal biases at Thule where katabatic winds dominate in colder months and the sea breeze dominates in warmer months, a weak correlation between katabatic deviation (i.e., sea breeze occurrence) and the water vapor isotopes ($\rho_{SA.\delta18O.hr}$ = +0.19, $\rho_{SA.dxs.hr}$ = -0.18) indicates that on a given day the sea breeze will bring isotopically heavier water vapor with lower *dxs* values than katabatic flow.

Although the difference in mean isotopic values are statistically significant between the two wind regimes for all isotopic species ($p < 0.001$, Welch's t-test), there is substantial overlap in the overall range (May–September data, mean ± 1 σ: $\delta^{18}O_{sea.breeze}$ = -26.8 ± 3.5‰ vs. $\delta^{18}O_{katabatic}$ = -28.5 ± 3.1‰; $\delta^{2}H_{sea.breeze}$ = -207 ± 25‰ vs. $\delta^{2}H_{katabatic}$ = -219 ± 22‰; $dxs_{sea.breeze}$ = 7.6 ± 4.8‰ vs. $dxs_{katabatic}$ = 9.3 ± 4.8‰; Fig. S3). The isotopic difference between the two wind regimes may be smaller at Thule than other studied sites because of Thule's location on the west-pointing Pituffik Peninsula. Much of the southeasterly katabatic flow here is not sourcing air directly off the main Greenland Ice Sheet, but rather air that is traveling over Baffin Bay along the western coast and potentially over nearby De Dødes Fjord (Fig. 1). Thus, both the katabatic winds and the sea breeze are bringing water vapor from locations where it has been able to isotopically exchange with local ocean waters.

The slightly lower water vapor isotope values during katabatic flow are then likely due to the vapor's brief passage over topographic highs of Cape York and the Tuto Ice Dome while the sea breeze carries moisture onshore unimpeded from Bylot Sound. Additionally, we note that as our wind data comes from a lower elevation site than our isotopic sampling, it is possible that particularly shallow sea breezes could produce wind observations identified as 'sea breeze' while the air at the isotopic sampling site on the South Mountain ridgetop is still katabatic sourced. However, no consistently clear signs of different air masses affecting the two stations in summer were found when examining periods with anomalous observations (e.g., sea breeze azimuth with high air temperature).

4.5 Evaposublimation of local snow and surface waters

Evaporation and sublimation of local surface waters and snowpack (referred here as "evaposublimation") can also alter local water vapor isotopic values by supplying vapor that is isotopically lighter with higher *dxs* values relative to the source snow/ice/water's isotopic composition (Casado et al., 2018; Kopec et al., 2019). Most of the snow at Thule falls in autumn and early winter (USAF, 2019) and surface waters in Thule generally match the isotopic composition of the winter snowpack

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(Csank et al., 2019). Thus, evaposublimation during warmer months will generally serve to lower $\delta^{18}O$ and $\delta^{2}H$ values and raise *dxs* values of water vapor in the local environment, although these two processes may fractionate the water isotopes different, and the exact isotopic composition of supplied vapor will depend on the relative balance of evaporation to sublimation (Christner et al., 2017). Additionally, very warm events in summer induce surface melt and sublimation across the Tuto Ice Dome and Greenland Ice Sheet (Box and Steffen, 2001; Nghiem et al., 2012; van As et al., 2012; Neff et al., 2014), and any of this moisture reaching Thule would be depleted in ^{18}O and ^{2}H and have a higher *dxs* relative to typical summer ocean-sourced moisture as it originates in a higher and colder location on the ice sheet. Isotopic effects from plant transpiration (Gat and Matsui, 1991; Farquhar et al., 2007; Aemisegger et al., 2014) are assumed to be very weak and inconsequential due to the sparse plant cover at Thule (Gold and Bliss, 1995).

4.6 Summary of environmental drivers of water vapor isotopes at Thule

To summarize, observations at Thule when the water vapor $\delta^{18}O$ and $\delta^{2}H$ values are higher and *dxs* values are lower are linked to: a) warmer local air temperature, b) more local marine moisture source, c) synoptic pattern that favors more southerly flow (i.e., NAO-), d) sea breeze surface winds, and/or e) lower evaposublimation. Likewise, lower $\delta^{18}O$ and $\delta^{2}H$ values and higher *dxs* values are linked to: a) colder local air temperature, b) more distant marine moisture source, c) synoptic pattern that restricts southerly flow (i.e., NAO+), d) katabatic surface winds, and/or e) higher evaposublimation. These different factors operate on different temporal scales (Fig. 5a). For example, shifts in local versus distant moisture sourcing affect isotopic composition at all timescales, but evaposublimation is mostly important only on shorter timescales like diel cycles.

These five factors should not be seen as independent drivers of isotopic change. Rather, these factors are detectable facets expressed in our observational data of the diverse environmental changes controlled by variations in Baffin Bay sea ice extent and synoptic moisture transport to the Arctic. As such, these factors greatly interact with each other, and isotopic variability cannot and should not be reduced down to a single predominant driver. However, these factors largely interact constructively to enhance relationships between environmental conditions and water vapor isotopic values (Fig. 5b, c). For example, the NAO- enhances southerly advection to Thule, which results in warmer local air temperature and potentially reduced sea ice extent that exposes local waters. Similarly, the sea breeze is enhanced during warmer periods of the year when sea ice extent is low, and it directly supplies local moisture to Thule from Bylot Sound. All these factors support isotopically heavier water vapor isotopes with lower *dxs* values when sea ice extent is low and synoptic conditions favor southerly flow.

The main exception to these constructive interactions is evaposublimation, which supplies isotopically light vapor with high *dxs* during warmer periods which would otherwise favor higher $\delta^{18}O$ and $\delta^{2}H$ values and lower *dxs* values as previously described. Due to the ample supply of marine moisture in the coastal Thule setting, evaposublimation is not expected to be a predominant moisture source or primary driver of isotopic variability. However, periods where water vapor isotopes are lighter

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and *dxs* values higher than expected for a given temperature may be a good identifier of an evaposublimation effect, and such analysis could be used to better quantify ice sheet vapor flux during large surface melt events.

5. Seasonal changes in the drivers of isotopic variability

The strong annual cycle in water vapor isotopes at Thule (Table 1) is directly tied to seasonal changes in weather and the regional environment driven by the presence or absence of local sea ice. While the annual sea ice breakup directly influences vapor isotopic composition by allowing local marine moisture supply, it also produces a cascade of other environmental changes that isotopically alter water vapor at Thule (Fig. 5). Beyond simply affecting the mean values of water vapor isotopes, the dramatic environmental transformations that occur at Thule after sea ice breakup also modify correlative relationships between isotopic and meteorological variables (Fig. 6). As a result of all these factors, the seasonal growth and breakup of the Baffin Bay sea ice is readily identifiable in our water vapor isotope record, particularly in the *dxs* data (Fig. 2).

5.1 Spring and sea ice breakup

In both spring seasons covered by our record, the isotopic and meteorological variables had a very abrupt shift from winter-typical values to summer-typical values (Fig. 2). During these shifts that began on 14 May 2018 and 29 Apr 2019, temperatures rose more than 15°C in a few hours, followed by a ~10‰ increase in $\delta^{18}O$ and ~10–15‰ drop in *dxs* over the next few days. Sea ice concentrations and satellite imagery from NSIDC (Fetterer et al., 2017) and MODIS (Hall and Riggs, 2015) show that these abrupt spring shifts were associated with the breakup of sea ice near and to the northwest of Thule as well as a general reduction in sea ice concentration throughout Baffin Bay (Fig. S4). Similar isotopic responses to sea ice breakup have been previously reported in the Arctic Ocean as well (Klein and Welker, 2016). Before the breakups in 2018 and 2019, the NAO index dropped 2 and 4 points, respectively, and the resulting extreme isotopic shifts create very strong correlations in May between isotopic variables and both Baffin Bay sea ice extent (Fig. 6e) and the NAO index (Fig. 6f).

Sea ice breakup and environmental warming is followed in short order by the first sustained sea breeze developments of the year that also aid the delivery of newly available local vapor to Thule. At this time in late spring, radiative heat loss at night drives a semi-consistent diel wind cycle between afternoon sea breezes and nightly katabatic flow, and synoptic storm systems may bring multiple days when winds predominantly come from either the ocean or over the ice sheet, depending the relative position of the storm system to Thule. The environmental contrast between the relatively warm open ocean and still frigid Greenland Ice Sheet and tundra maximize isotopic differences in the two wind regimes, and, as a result, the strongest correlations between isotopic variables and katabatic deviation are observed in April through June (Fig. 6d).

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During winter polar night when Baffin Bay is largely frozen over, the water vapor isotopic system at Thule is relatively simple. Mean values for winter $\delta^{18}O$ and $\delta^{2}H$ are lower and *dxs* values higher in the absence of a local marine moisture supply (Fig. 3b), and isotopic variability largely reflects extratropical cyclone impacts and broader synoptic flow. Strong winter correlations between isotopic species and both air temperature and mixing ratio (Fig. 6) reflect this as warmer and moister air transported to Thule produces higher water vapor $\delta^{18}O$ and δD values and lower *dxs* values. ¶
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5.2 Summer

The height of summer brings striking changes in isotope-climate relationships. Most notably, the $\delta^{18}O$ and $\delta^{2}H$ correlations with air temperature switch from positive to negative in summer months (mean $\rho_{non\text{-}summer}$ = +0.38 vs. mean ρ_{summer} = -0.32 for $\delta^{18}O$) with *dxs* showing a similar, though opposite, pattern (mean $\rho_{non\text{-}summer}$ = -0.53 vs. mean ρ_{summer} = +0.40) (Fig. 6a). As a result, the highest $\delta^{18}O$ and $\delta^{2}H$ values and lowest *dxs* values are not observed when air temperatures are greatest, but rather near 0°C (Fig. S3a, S3d) During the same summer period, isotopic correlations with mixing ratio greatly weaken (Fig. 6b) while correlations with relative humidity strengthen (Fig. 6c). The relationship between air temperature and mixing ratio, strongly correlated with a positive slope through most of the year, decouples in summer, and temperatures warmer than 5°C largely result in no higher water vapor content (Fig. S5)

Summer in Thule is typically cool and humid under shallow marine air from local open seas, helped onto land by the sea breeze. As the local ocean never warms much above freezing, most of the warmest periods at Thule (i.e., >5–8°C) require a high pressure ridge over Greenland, often associated with southerly air flow up along the western Greenland coast and aided by NAO- conditions. In many cases, the position of this high favors large-scale downsloping and subsidence off the northwestern ice sheet (Fig. 7). This pattern was exceptionally persistent in summer 2019 (Tedesco and Fettweis, 2019), and this period supplied most of the warmest observations in our database. Moisture arriving at Thule through southerly advection and/or downsloping in these warm events is isotopically lighter and less humid than local marine air as it is more distantly sourced and also must cross topographic highs. When contrasted against cool local marine air that has isotopically heavier vapor with low *dxs* values, this produces the observed flip in summer correlations between isotopic species and temperature as well as the strengthened correlations with relative humidity.

Correlations between isotopes and water vapor mixing ratio greatly weaken in summer because mixing ratios do not clearly differentiate between local marine and southerly advected air. Although the shallow marine air has a higher relative humidity, it is also colder than the southerly advected air and thus has a lower maximum mixing ratio. In contrast, the relative humidity of southerly advected air is lower, but its warmer temperature means that the actual water content of the air can be very similar to that of local marine air. As a result, the relationship between temperature and mixing ratio decouples (Fig. S5), and isotopic correlations with mixing ratio approach zero (Fig. 6b).

Two additional processes that emerge in summer may also lead to these relationship changes. First, the very warm conditions that occur during strong ridging over Greenland and enhanced southerly advection to Thule are also associated with intense surface melt and sublimation on the Tuto Ice Dome and greater Greenland Ice Sheet (Nghiem et al., 2012; van As et al., 2012; Neff et al., 2014; McLeod and Mote, 2016; Ballinger et al., 2019). As the ice sheet surfaces are higher in elevation and colder than Thule, water vapor coming from their snow and glacial ice would be particularly isotopically light with high *dxs* values (Steen-Larsen et al., 2013; Kopec et al., 2014; Bréant et al., 2019), and this would enhance the isotopic signature of southerly advected air as it passes over the ice sheet on its way to Thule.

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Second, the high pressure of these warm events often brings sunny weather to Thule that promotes intense heating of the local tundra and exposed rocky surfaces. Outside of polar night, water vapor mixing ratio typically follows the daily temperature cycle and peaks in the early afternoon. However, on days when the mean temperature is greater than 10°C, the mixing ratio drops as temperature rises and hits its minimum daily value around local noon. This loss of water vapor suggests that daytime heating on the warmest days induces convection (Duynkerke and van den Broeke, 1994) that vertically mixes drier air aloft down to the shallow marine surface layer. This results in a net upward water vapor transport (Sherwood et al., 2010; Kiemle et al., 2013; Homeyer et al., 2014), and the drier air that mixes downward from the free troposphere brings vapor with lower $\delta^{18}O$ and $\delta^{2}H$ and higher *dxs* values (Bailey et al., 2013). The surface heating and drying during midday could also increase local evaporation from the land and surface waters that, while not enough to overcome the net upward vapor transport from convection, would be an additional source of isotopically light water vapor to near surface moisture.

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5.4 Autumn and sea ice growth

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The transition to winter-type isotopic and meteorological conditions is much more gradual than the spring transition at sea ice breakup (Fig. 2), in accordance with the comparatively steady growth of sea ice in autumn. The $\delta^{18}O$ and $\delta^{2}H$ values slowly decrease while *dxs* values slowly increase from October through November, and values through winter do not generally overlap summer values. While isotopic correlations with sea ice extent are near zero throughout summer (likely because most changes to sea ice extent at this time are occurring too far north or west to directly impact Thule), the autumnal growth of sea ice results in moderately strong correlations in October (Fig. 6e) as the region of sea ice formation again affects Thule. Although small in magnitude, correlations with the NAO index also strengthen in autumn (Fig. 6f), and this is likely due to the close relationship between NAO phase, sea ice extent, and local marine moisture availability. The return of true day–night cycles in September creates a surface wind regime similar to that of spring, and correlations between isotopes and katabatic deviation strengthen in the autumn months until the sea breeze ceases due to polar night in November (Fig. 6d).

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5.5 Winter

During the polar night of winter, the water vapor isotopic system at Thule is relatively simple because northern Baffin Bay and all surface waters are frozen over. Mean values for $\delta^{18}O$ and $\delta^{2}H$ in winter are lower and *dxs* values higher than other seasons in the absence of a local marine moisture supply (Fig. 3b), and isotopic variability largely reflects extratropical cyclone impacts and broad synoptic flow changes. Strong winter correlations between isotopic species and both air temperature and mixing ratio (Fig. 6) reflect this as warmer and moister air transported to Thule produces higher water vapor $\delta^{18}O$ and $\delta^{2}H$ values and lower *dxs* values.

The *dxs* values in winter are also notably more variable than during summer (Fig. 2). Since nearly all moisture arriving to Thule in summer has to pass over the open water of Baffin Bay, vapor exchange and uptake probably helps stabilize the summer *dxs* variability. In contrast, when Baffin Bay is frozen, the *dxs* values from different moisture sources may be preserved

better and contribute to the higher winter variability in *dxs* values. More focused back trajectory analysis on winter *dxs* variability in the future may help quantify the variability and effects of different moisture sources.

Interestingly, $\delta^{18}O$ and $\delta^{2}H$ have very strong positive correlations with the late winter sea ice extent in February and March, while *dxs* has very strong negative correlations with sea ice extent in February (Fig. 6e). These relationships are opposite to those observed in spring and autumn, when the presence or absence of sea ice plays a clear isotopic role through local water availability. It is not entirely clear what is driving the strong correlations in late winter, as sea ice during these months is near its maximum extent and largely complete in coverage near Thule. These unusual correlations may be due to coincidental extreme events. Shortly after the maximal extent of sea ice was reached in 2018, a period of enhanced southerly moisture advection supplied abnormally heavy water vapor isotopes with very low *dxs* values. In 2019, the isotopes also become unusually heavy with low *dxs* for two weeks after the peak in sea ice extent, but no associated increase in southerly moisture was identified and the root cause is unclear. With only two years of record, it is difficult to conclude whether the observed correlative strengths in late winter accurately represent a true change in isotopic character after a certain sea ice threshold is exceeded, or if it is simply due to coincidental occurrence with two unusual late winter weather events. Additional years of observation may help clarify this uncertainty.

5.6 Cold season moisture pulse events

During winter months, rapid shifts to extremely low NAO indices often coincide with intense poleward transport of southerly heat and moisture to Thule that lasts 1–5 days. We refer to these distinct episodes as "moisture pulse events" as they appear very clearly in the Thule mixing ratio time series (Fig. 2). During these events, air temperature rises 10–15°C and peaks near or above freezing while water vapor concentrations can reach four times greater than mean winter levels. These pulses appear to notably reduce or slow the growth of sea ice across Baffin Bay (Fig. 2j), although the sea ice stays largely intact in the northern reaches near Thule. Although not all these events meet the defined criteria of an atmospheric river (Mattingly et al., 2018), all have similar impacts to an atmospheric river event due to their anomalously high moisture and heat transport.

Unsurprisingly, such extreme weather changes are reflected by impressive water vapor isotopic responses. During these moisture pulse events, the $\delta^{18}O$ and $\delta^{2}H$ rise 6–10‰ and 50–100‰, respectively, to reach values more typical of late spring and early summer. Concurrently, the *dxs* drops 15–25‰, and the minimum *dxs* values observed in these moisture pulse events match or exceed the minimum values observed at the height of summer. As sea ice coverage prevents the uptake of isotopically heavy moisture with low *dxs* values from local waters during these moisture pulse events, the anomalous isotopic values must signify the presence of deep southerly moisture transport (Bonne et al., 2015). A focused analysis of these events is currently underway.

The rapid isotopic and environmental shifts coinciding with sea ice break up in 2018 and 2019 also fit the general pattern of a moisture pulse event, except the isotopic and meteorological variables do not revert back to their preceding values after 1–5

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These unusual correlations may then be due to coincidental extreme events. Shortly after the maximal extent of sea ice was reached in 2018, a major and extended moisture pulse event occurred which gave abnormally heavy water vapor isotopes with very low *dxs* values. While 2019 did not have an identified moisture pulse at maximal sea ice extent, the isotopes become unusually heavy with low *dxs* for two weeks (with an unclear cause) shortly after the sea ice peak. Because both periods in our record with the highest sea ice extent coincide with these events which isotopically resemble warmer conditions, it is difficult to conclude whether the observed correlative strengths in late winter are due simply to these two events or accurately represent a true change in isotopic character after a certain sea ice threshold is exceeded. Additional years of observation may help clarify this uncertainty.¶

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days. Indeed, the shift in 2019 is associated with an atmospheric river event impacting western Greenland and an extreme drop in the NAO index from +2 to -2 while the 2018 shift coincided with an NAO index drop from +2.63 (the highest observed in our record) to +0.40 and an intense pulse of southerly moisture advection. In typical cold season moisture pulses, sea ice remains intact around Thule and the isotopic and meteorological effects from the southerly moisture advection are short lived as the moisture pulse air mass moves on or mixes out. In these two spring events, however, local sea ice coverage is broken, and the new ample supply of local water vapor allows isotopic and meteorological values to remain elevated.

6. Diel cycles

6.1 Diel cycles overview

Most polar sites report diel cycling of water vapor isotopes in summer (Steen-Larsen et al., 2013; Bastrikov et al., 2014; Bonne et al., 2014; Kopec et al., 2014; Casado et al., 2016; Ritter et al., 2016; Bréant et al., 2019), but this cycling is attributed to different causes including katabatic wind cycling (Kopec et al., 2014; Bréant et al., 2019), vapor exchange between snow and air (Steen-Larsen et al., 2013; Casado et al., 2016), and dew formation (Bastrikov et al., 2014). In contrast to these sites, Thule exhibits relatively limited isotopic diel cycling in all months except March, with total cycle magnitudes less than 1‰ for both $\delta^{18}O$ and *dxs*, even when observations with possible cyclonic system impacts (i.e., lower than average station pressure) are excluded. Additionally, the existence of these small diel cycles is only clearly evident when the larger synoptic variability is removed by averaging multiple days together. This subdued diel cycling is probably due to Thule's moderated coastal climate (Bréant et al., 2019; Bonne et al., 2020) while lengthy periods of midnight sun/polar night also reduce the day-night contrasts that power diel cycles at lower latitude sites.

Looking broadly at the entire Thule isotopic record, the magnitude of isotopic diel cycles is not a significant driver of isotopic variability. However, we believe that analyzing these cycles gives better insight into the broader environmental controls on water vapor isotopes, including those at synoptic and annual scales. Based on observed isotopic and meteorological patterns, we divide diel cycles at Thule into four regimes: polar night, March, summer, and transition (Fig. 8). These regimes are superimposed on the broader seasonal changes in isotopic variability and control previously discussed. Out of the major drivers of isotopic change at Thule, air temperature, surface wind regime, and evaposublimation vary enough on hourly timescales to contribute to diel isotopic cycles, while the NAO and local marine moisture availability are expressed at longer timeframes and/or not on a consistent daily cycle. We note that the magnitude of daily change in the isotopic variables is typically within the confidence intervals of the mean with the exception of the March regime. We believe the daily patterns of variable change are still informative, but discussion of these trends should be viewed as more speculative than other conclusions made in this study.

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6.2 Polar night regime

The polar night regime includes the months from October through February, when daylight is very short (October, February) or nonexistent (November–January). With the lack of a strong insolation cycle, meteorological variables change little over a day, and winds are strong and consistently katabatic. Without any diel changes in the factors that drive isotopic variability (i.e., air temperature, surface wind regime, evaposublimation), the water vapor isotopes show no diel cycling in the polar night regime (Fig. 8). A very slight hint of a rise or fall in values can be seen around local noon, but this is primarily attributed to data from the end of February that resemble the upcoming March regime.

6.3 March regime

The observed isotopic cycles in March (maximum diel range: $\delta^{18}O$: 2.4‰, $\delta^{2}H$: 16‰, *dxs*: 5.1‰) are by far the largest magnitude of any month of the year at Thule (Fig. 8), although similar cycles with smaller amplitudes are also observed at the end of February. This heightened isotopic response appears due to several coinciding factors: a maximized daily insolation cycle near the equinox, an extensive snowpack to supply water vapor, and the enhanced impact of snow-supplied vapor in the very cold and dry environment (aided by the maximal seasonal extent of sea ice that limits external moisture input). This environment approximates the summer ice sheet setting of two other water vapor isotope observation sites at NEEM, Greenland (Steen-Larsen et al., 2013), and Dome C, Antarctica (Casado et al., 2016), and Thule's March isotopic diel regime appears similar to the cycles reported from those sites.

Moisture at Thule in March predominantly arrives from distant transport through katabatic flow with resulting low $\delta^{18}O$ and $\delta^{2}H$ values and high *dxs* values. However, daytime heating of the snow surface across the landscape promotes release of water vapor held between snow grains that have equilibrated with the local snow isotopes (Steen-Larsen et al., 2013; Casado et al., 2016). Because most snow in Thule falls in late autumn/early winter and is sourced from Baffin Bay, the equilibrated vapor is isotopically heavier and has a lower *dxs* than the katabatic-supplied moisture, producing the observed $\delta^{18}O$ and $\delta^{2}H$ peak and *dxs* minimum around local noon. As insolation and air temperature drop in the evening, this vapor release ceases and potentially reverses, allowing the vapor isotopic composition to revert back to one of katabatic origin with lower $\delta^{18}O$ and $\delta^{2}H$ and higher *dxs* values. The lack of a large drop in relative humidity coinciding with the midday temperature rise suggests that additional vapor is being supplied, and the local snowpack is the only likely source with all local waters frozen. A large isotopic diel cycle predominating in spring has also been reported in the Lena River delta and similarly attributed to the release of water vapor from the preceding winter snowpack (Bonne et al., 2020).

A parallel isotopic regime does not reappear around the autumnal equinox at Thule or at the Lena Delta site (Bonne et al., 2020). This is likely due to little to no extensive snowpack and a much higher water vapor content from warmer temperatures and open nearby seas that buffer any minor potential input from a snowpack vapor exchange. One exception to this lack of autumnal diel cycles, unique in the Thule record, occurred from 20–25 September 2017 when an isotopic diel cycle very

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similar to the March regime appeared (Fig. S6). This cycling occurred shortly after a snowfall and coincided with a diel surface wind cycle where the katabatic winds calmed each afternoon but did not switch to a sea breeze. This appears to have allowed vapor released from the recent snow to raise isotopic values in the afternoon before the returning katabatic winds mixed the snow-derived moisture out and dropped isotopic values in the evening. In other September periods with a similar wind regime but without a recent snow cover, no isotopic cycling was recorded.

6.4 Summer regime

In the summer months (June–August), the midnight sun reduces the diel insolation cycle. Combined with the moderating influence of fully ice free local seas, most meteorological variables are more stationary over the course of a day. With this reduced environmental variability, diel cycles in isotopes are subdued. The subtle daily patterns in $\delta^{18}O$ and *dxs* that do exist appear largely attributable to evaposublimation cycles: daily warming supplies low $\delta^{18}O$/high *dxs* water vapor through increased evaposublimation, with the resulting $\delta^{18}O$ minimum and *dxs* maximum around midday (Fig. 8). Increased boundary layer mixing at midday may also supply moisture from the free troposphere with low $\delta^{18}O$ and high *dxs* values to produce a similar isotopic effect to evaposublimation.

At Kangerlussuaq (Kopec et al., 2014) and Dumont d'Urville (Bréant et al., 2019), the local surface winds have a clear diel cycle between daily sea breezes and nightly katabatic winds in summer. At Thule, the sea breeze has a diel cycle where it strengthens in the afternoon and weakens at night (Fig. 8g), but there is not a full switch back to katabatic flow at night in summer. This dominance of the sea breeze at Thule is likely due to 24 hour summer insolation that can fuel a sea breeze even at "night", while Kangerlussuaq and Dumont d'Urville still have true night at their lower latitudes. As a result, sea breeze-supplied local water is likely present throughout a typical day and night at the height of summer in Thule, and the nightly weakening of the sea breeze has limited isotopic effect without a full shift to katabatic flow.

6.5 Transition regime

The seasonal transition months of April, May, and September have a true day–night cycle that gives meteorological variables a greater diel amplitude than the summer or polar night regimes (Fig. 8). Yet unlike March, these transitional months are much warmer and more humid with moisture supplied from an open or opening Baffin Bay. Winds exhibit a strong diel cycle as surface heating leads to afternoon sea breeze development (Fig. 8g), and radiative heat loss over the ice sheet at night strengthens katabatic flow. We might expect a clear diel response in water vapor isotopes due to these heightened diel meteorological cycles, but the isotopes have little to no daily cycle (Fig. 8a–b).

This limited isotopic cycling is likely due to competing effects of diel surface wind regime and evaposublimation cycles. While the sea breeze-sourced vapor coming directly off Bylot Sound in afternoon is isotopically heavy, midday heating also supplies isotopically light moisture from evaposublimation and boundary level mixing. Cooling at night brings a wind shift to

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isotopically lighter katabatic flow but also suppresses evaposublimation and boundary level mixing while promoting dew condensation. As a result, no clear diel isotopic cycle emerges, although the subtle late evening peak in $\delta^{18}O$ may arise because evaposublimation rates are reduced while the sea breeze is still ongoing.

7. Implications

The improved understanding of water vapor isotopic variability granted by our Thule observations can aid the interpretation of regional ice cores. The primary drivers of isotopic changes in water vapor are likely to also affect local precipitation, although there are many additional processes involved in the transfer of an isotopic signature from vapor to precipitation and eventually to ice core that must be considered (Steen-Larsen et al., 2014; Casado et al., 2018; Madsen et al., 2019). Changes in $\delta^{18}O$ and $\delta^{2}H$ have long been used to reconstruct climate change from deep ice cores, typically interpreted as local temperature variability in Greenland (e.g., Dansgaard et al., 1969; Grootes and Stuiver, 1997; Johnsen et al., 2001). While air temperature is strongly correlated with isotopic change at Thule, much of this strength appears to arise as a common response to seasonal change between air temperature and sea ice extent. Air temperature might still be robustly reconstructed from isotopic archives based on our observed strong correlations, but the lack of a causative relationship risks misinterpretation if applied back far through time when the local environment may have significantly changed. Of particular warning is our observation that the basic relationship between air temperature and water vapor isotopes inverts in summer, leading to very different isotopic interpretations depending on season. While this may be a local and/or coastal effect that is not expressed on ice sheets (Ballinger et al., 2019), we still advise caution.

More recently, additional consideration has been given in ice core analysis to secondary isotopic variables like *dxs* and other environmental drivers such as moisture source, sea ice extent, and atmospheric circulation (e.g., Grumet et al., 2001; Vinther et al., 2003; Steffensen et al., 2008; Landais et al., 2018; Kopec et al., 2019). While our two-year Thule record is too short to statistically determine the strongest drivers of interannual isotopic variability, changes in the duration of sea ice coverage and mean NAO phase appear most likely to control year-to-year differences in mean isotopic composition. In line with recent interpretations in regional ice cores (Osterberg et al., 2015), our results suggest that past periods with decreased sea ice extent in Baffin Bay will have increased local marine moisture sourcing that produces higher $\delta^{18}O$ and $\delta^{2}H$ and lower *dxs* values.

In recent years, Baffin Bay has had later sea ice freezes, earlier breakups, and a decrease in overall sea ice extent that accounts for 22% of all recently observed loss in March Arctic sea ice (Onarheim et al., 2018). We expect that the resulting changes in the regional hydroclimate will be isotopically preserved in future glacial ice in Greenland. However, predicting the isotopic responses to these changes is challenging. The future of the Nares ice bridge between Greenland and Ellesmere Island is one potential complication: should it consistently fail to form in a warmer world, the North Water Polynya may see increased sea ice cover that slows the overall sea ice loss impact on Thule (Barber et al., 2001; Puntsag et al., 2016; Vincent, 2019). Reductions in Arctic sea ice coverage and duration may also shift precipitation seasonality (Kopec et al., 2016) and atmospheric

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While both the sea breeze and katabatic flow are likely sourcing similar southern moisture originating or passing over Baffin Bay, the sea breeze-sourced vapor coming directly off Bylot Sound in afternoon is isotopically heavier than the nighttime vapor that must travel over the Tuto Ice Dome and coastal mountains through katabatic flow. However, midday heating also increases the evaposublimation of surface waters and residual snow, and this vapor is isotopically lighter with high *dxs* values. The competing isotopic effects of the wind regime shifts and the evaposublimation cycle largely suppress any isotopic diel cycle from emerging, although the subtle late evening peak in $\delta^{18}O$ may arise because evaposublimation rates are reduced while the sea breeze is still ongoing.

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circulation (McLeod and Mote, 2016; Ballinger et al., 2018; Francis et al., 2018), changing the annual isotopic balance of precipitation. More open winter waters in Baffin Bay could decrease annual $\delta^{18}O$ and $\delta^{2}H$ values as the relative fraction of isotopically lighter winter precipitation events increases, but this may be counter-balanced in part by the greater sourcing from isotopically heavy local waters.

Enhanced southerly moisture advection aided by NAO- conditions would also favor higher $\delta^{18}O$ and $\delta^{2}H$ and lower *dxs* values in northwestern Greenland, though some caution might be warranted for isotopic complications from greater ice sheet surface melt and vertical atmospheric mixing similar to those we have observed during summer NAO- phases. Additionally, the positive relationship between NAO phase and Baffin Bay sea ice extent (Mysak et al., 1996; Grumet et al., 2001; Stern and Heide-Jørgensen, 2003) makes it difficult to quantifiably split their influences on regional water isotopes. The extreme isotopic values we observe during moisture pulse events suggest that any changes in their frequency and magnitude (and more generally of related atmospheric river events) will have an outsized effect due to the sheer volume of moisture and precipitation they can bring to Greenland. For highly-resolved ice cores, it may be possible to identify particularly strong moisture pulse events or years with high frequency of these events as extreme minima in *dxs* values. Taken as a whole, it is clear that in the absence of additional clarifying evidence from other ice core proxies or nearby records, isotopic interpretations should be cautious in assigning cause solely to one individual environmental factor without further analysis to tease the many highly-integrated potential factors apart.

8. Conclusions

Our two years of water vapor isotope monitoring in northwest Greenland have produced a record of unprecedented extent and resolution for High Arctic Greenland and one of the longest records of any polar site reported to date. The variability in water vapor isotopes at Thule is explained by five interrelated environmental mechanisms: local air temperature, local marine moisture availability, the NAO, surface wind regime, and evaposublimation. The relative importance of each mechanism changes over the course of the year and overall produces a clear annual isotopic cycle that is closely linked to sea ice extent in Baffin Bay and synoptic polar moisture transport. On top of these seasonal trends, local environmental drivers such as sea breeze development, vapor supply from surface water and snow, and convection can subtly modify near surface water vapor isotopes through diel cycles.

This analysis highlights the importance of local geography and climate in isotope systematics. Compared to other high latitude water vapor isotope studies, Thule is more substantially impacted by sea ice fluctuations, both seasonally and interannually. This results in a clearer identification of the isotopic effects of sea ice extent which is particularly valuable for the interpretation of deep ice cores. Additionally, some aspects of isotopic variability, such as the changes in correlative relationship strength and direction during spring and summer, are so far uniquely reported for Thule, and it is unclear at this time if these seasonal

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relationship patterns are present at other polar sites. As a result, conclusions based on data at one high latitude site should not be applied indiscriminately to other sites without extensive validation that such a comparison is warranted.

Caution should also be made when making strong conclusions about isotopic controls based on observations of short duration (i.e., a single year or less). If data is only taken during a single season, any observed relationships may be specific only to that season and should not be assumed to be applicable to the entire year. For variables with relatively long-term variability, such as teleconnection indices, data from a single year is likely not long enough to reveal fully robust correlations. In our Thule data, we were fortunate that the synoptic patterns were different enough between 2018 and 2019 to allow useful analytical comparisons of the NAO at monthly and seasonal resolutions. However, even two years of data are not enough to fully clarify the relationships between teleconnections and water vapor isotopes for all months, and additional years of isotopic data from continued observation are needed for an improved understanding.

Moving forward, this isotope and meteorological database offers many opportunities for more in-depth and focused analyses of specific weather events and atmospheric patterns. As previously stated, our identified moisture pulse events are a focus of additional research, and we encourage the use of our data in other focused case studies. An expansion of our "quasi-climatology" approach could be used to model moisture source environments for direct comparative analysis with the water vapor isotope data, which can help resolve existing questions relating to the nature of the *dxs*–sea surface temperature relationship in polar regions (Vimeux et al., 1999; Aemisegger and Sjolte, 2018).

Water vapor isotope observations at Thule continue, and additional months and years of data will help refine and verify our conclusions made here. The long temporal coverage of our database makes it an excellent option for validating high resolution isotope-enabled simulations of air-ice and air-sea interactions in the Arctic. These results from Thule will help greatly in interpreting isotopic variability at other sites in the Arctic Water Isotope Network (Welker et al., 2019), and future collaborative analysis across this network will allow tracking the effects of synoptic weather patterns on water vapor isotopes in real time across the circumpolar region.

Data availability

Isotopic data is available at https://doi.org/10.18739/A21J9779S. Meteorological data for SMT is available at https://doi.org/10.1594/PANGAEA.895059 and in associated followup datasets by G. Muscari also on PANGAEA. Meteorological data for THU is available upon request of the 821st Weather Squadron of the US Air Force.

Author contribution

JMW and ESK installed the L-2130i; PDA and BGK installed the dry air calibration system and performed maintenance on the L-2130i; PDA wrote the code in R for organizing and calibrating data and for statistical analyses; KSM performed back

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trajectory and water vapor transport analyses; DC performed cross-correlation analyses; PDA prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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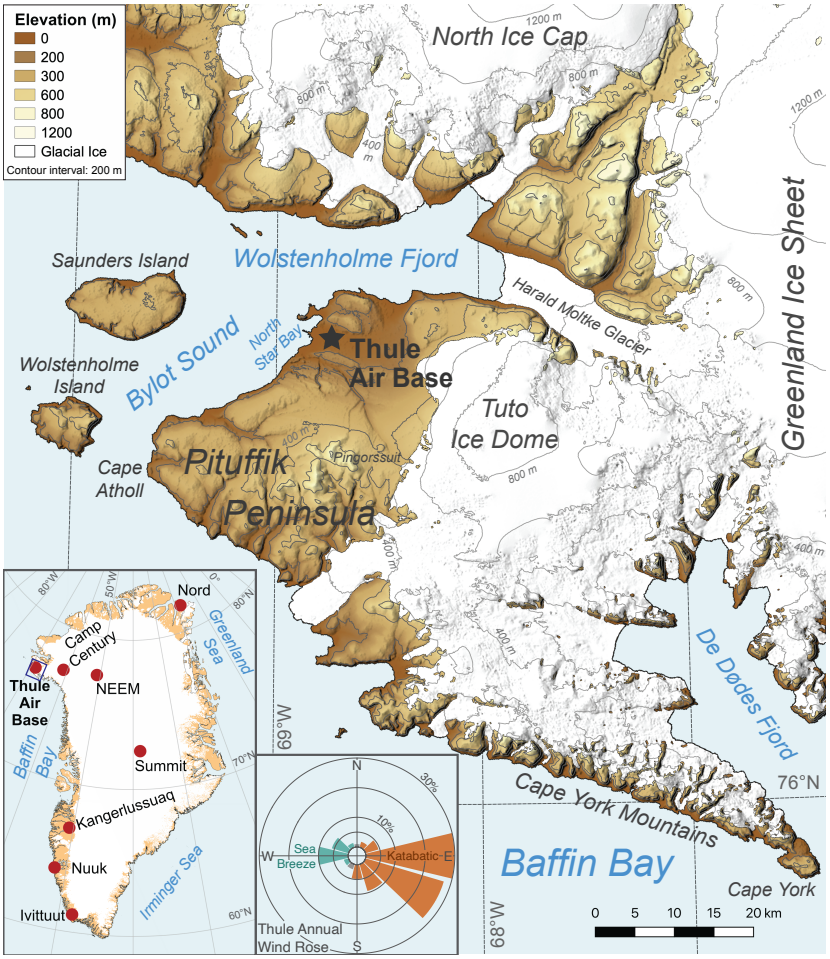

Figure 1. Map of the local geography around Thule Air Base, the site of water vapor and meteorological measurements for this study. The inset map at lower left shows the location of Thule Air Base in Greenland and other important Greenland sites. Elevation data was taken from ArcticDEM, Polar Geospatial Center (Porter et al., 2018), and ice sheet, land, and ocean extent data were taken from Greenland Ice Sheet Mapping Project, NSIDC (Howat et al., 2014; Howat, 2017). Wind rose created from Thule airport data covering Aug 2017-Aug 2019 (USAF, 2019).

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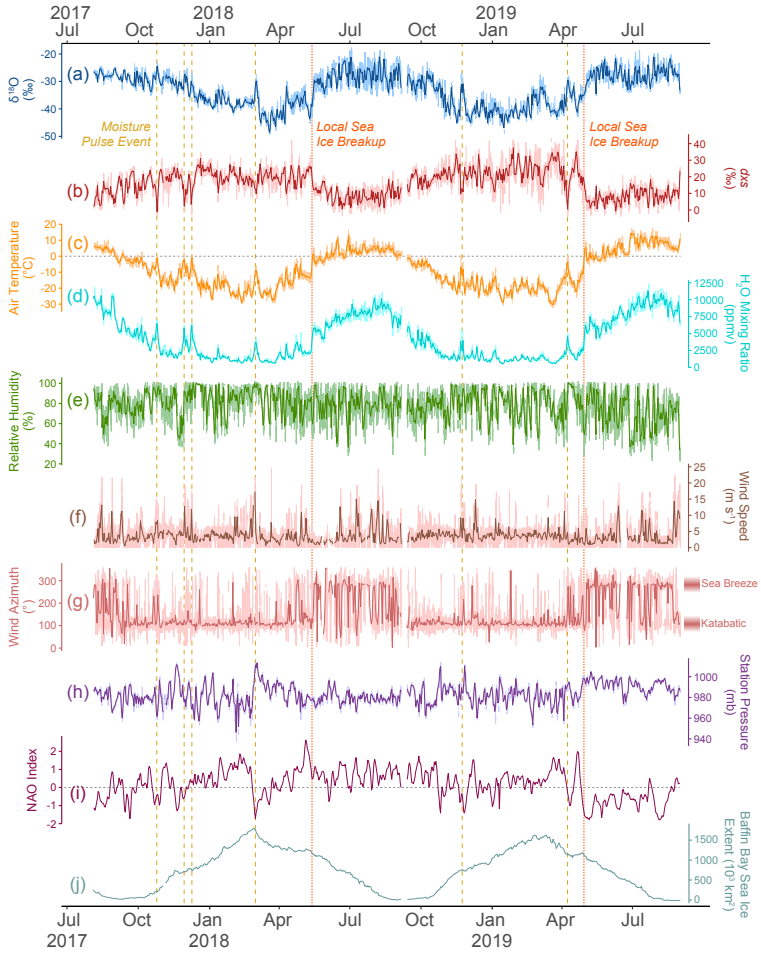

Figure 2. Observed data from Thule, Greenland, of (a) water vapor $\delta^{18}O$, (b) water vapor *dxs*, (c) mean air temperature, (d) water vapor mixing ratio, (e) relative humidity, (f) wind speed, (g) wind azimuth, (h) station pressure, (i) NAO index (NOAA, 2019), and (j) Baffin Bay sea ice extent (Fetterer et al., 2010). The time series of $\delta^{2}H$ is very similar to $\delta^{18}O$ and not shown. Data shown are daily mean values with hourly values as lighter backdrop line for higher resolution variables (a–h). All observations were taken at the

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SMT site except for wind speed and azimuth which were observed at the THU airport. Yellow dashed vertical lines indicate moisture pulse events and orange dotted vertical lines indicate the timing of sea ice breakup near Thule.

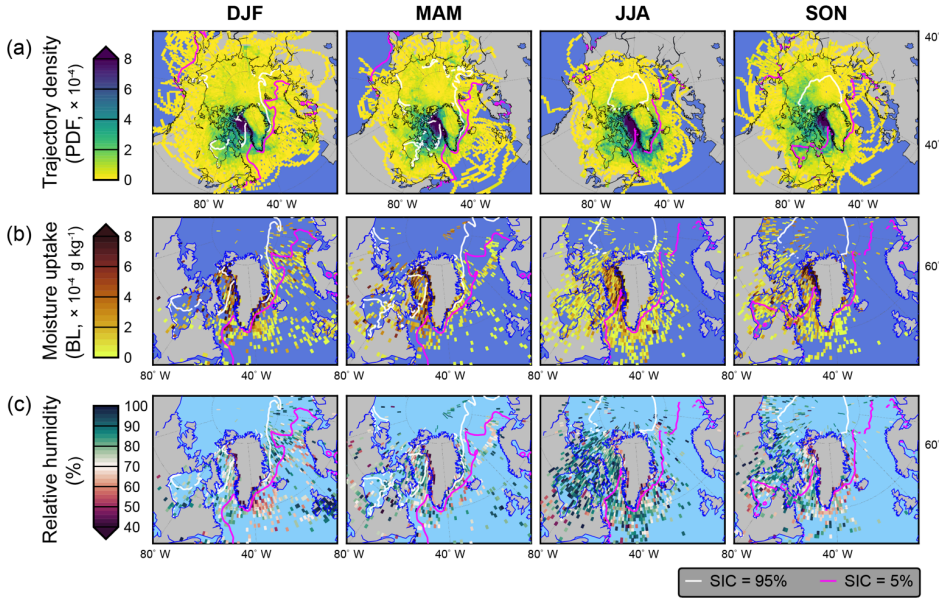

Figure 3. Back trajectory analysis results for air parcels arriving at Thule for each meteorological season, including (a) trajectory density, (b) regions of water vapor uptake from the boundary layer (BL), and (c) relative humidity for these regions of vapor uptake. Analysis based on a "quasi-climatology" that sampled 10 random days from each month during 1980–2018. Trajectory points are binned onto a 1° latitude/longitude grid as (a) probability density functions or (b, c) mean values within each grid cell. White and magenta lines show the extent of 95% and 5% sea ice concentration (SIC), respectively, for the given season.

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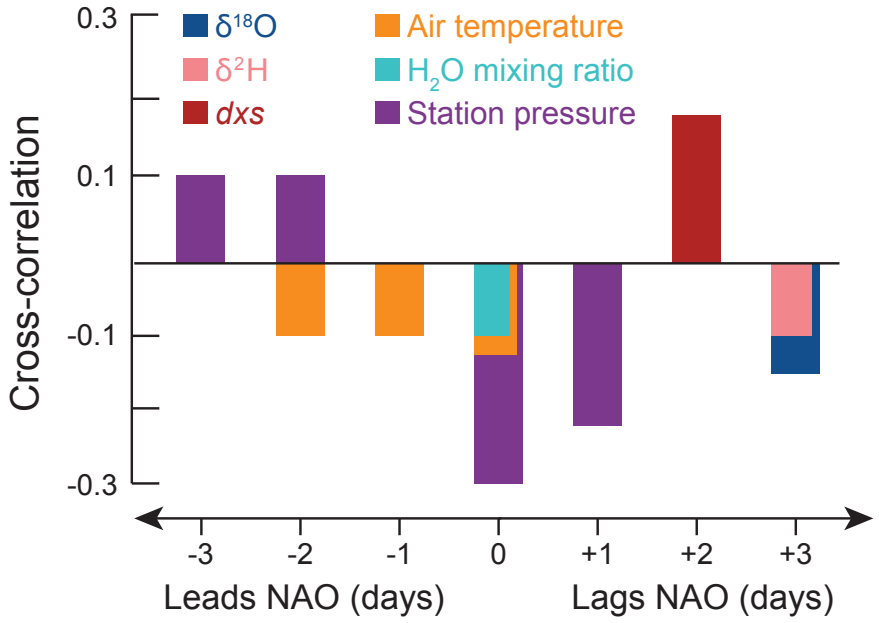

Figure 4. Cross-correlation analysis results between NAO index and selected isotopic and meteorological variables. Results shown are all statistically significant at $p < 0.05$ ($n = 737$), and non-significant results are not illustrated.

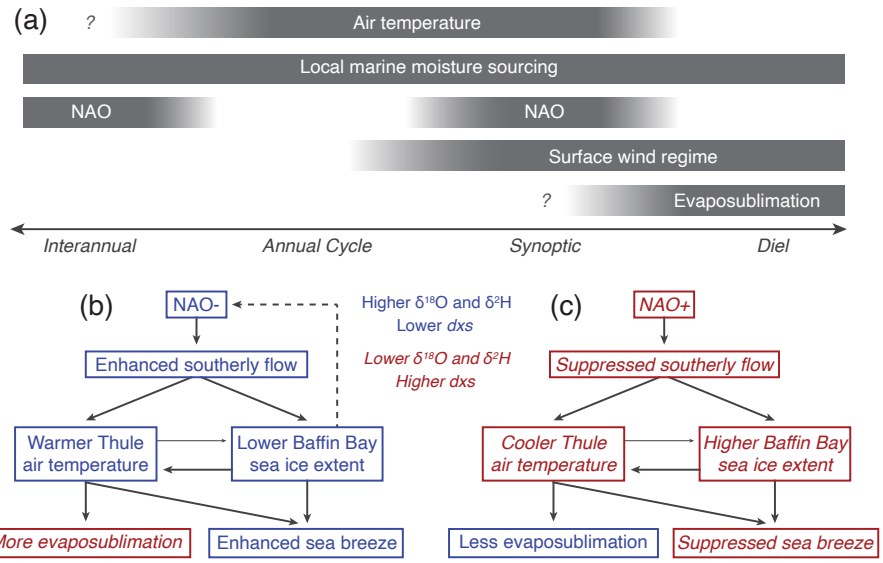

Figure 5. Summary of effects of environmental factors affecting water vapor isotope composition at Thule. (a) Illustration of temporal scales when each factor is most impactful. Question marks indicate periods of uncertain degree of impact. (b) Interactions between environmental factors during NAO- conditions, with factors that produce higher $\delta^{18}O$ and $\delta^{2}H$, values and lower *d*xs values in blue, and factors that produce lower $\delta^{18}O$ and $\delta^{2}H$, values and higher *d*xs values in italicized red. The dashed line refers to the possibility that low sea ice extent reinforces the NAO- phase (e.g., Petrie et al., 2015). (c) Same as (b), but for NAO+ conditions.

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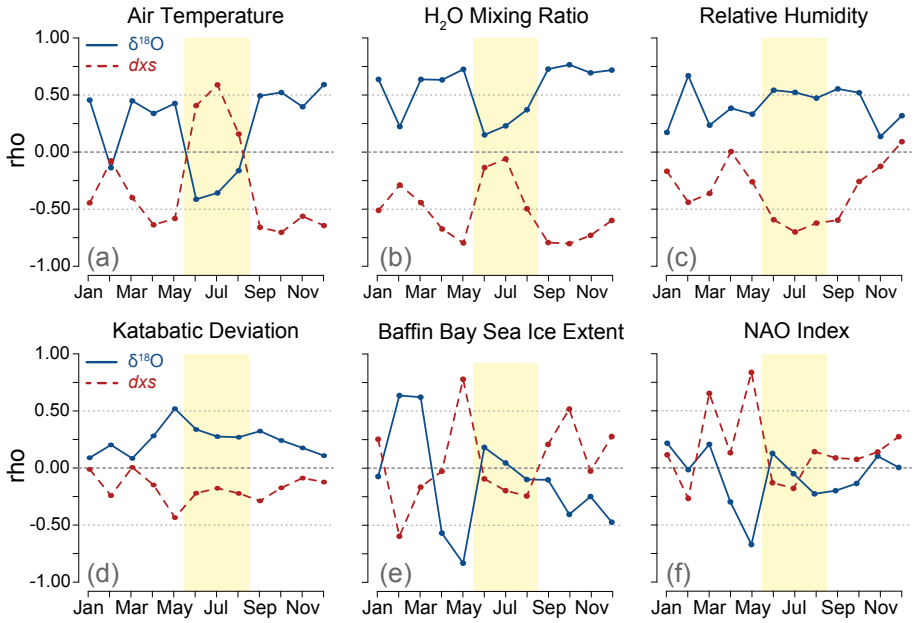

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Figure 6. Spearman correlations of meteorological variables (a-f) with $\delta^{18}O$ (blue, solid) and *dxs* (red, dashed) with data binned by month. The yellow box highlights the summer season (JJA) and horizontal dotted lines show rho values of 0.00 and ±0.50 for reference. Correlation patterns for $\delta^{2}H$ (not shown) are very similar to those of $\delta^{18}O$. Correlation values were calculated using the highest available data resolution: 10 minute for (a–c), hourly for (d), and daily for (e–f).

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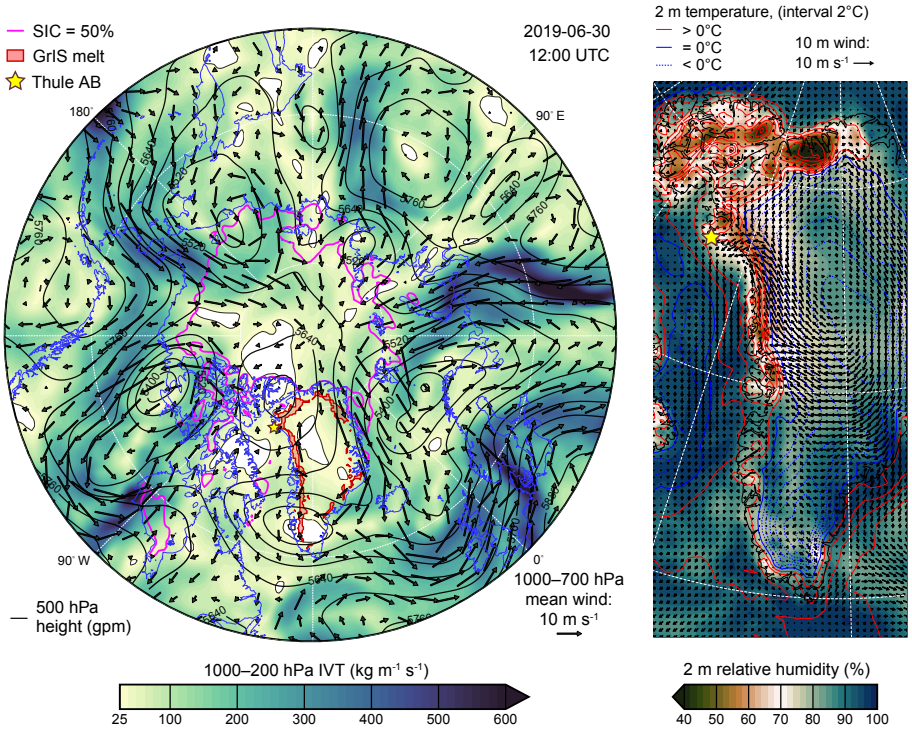

Figure 7. Synoptic pan-Arctic (left) and regional Greenland-focused (right) atmospheric conditions during an anomalously warm period on 30 June 2019, when a ridge over northwest Greenland directed downsloping air toward the observing site at Thule Air Base. Synoptic map shows integrated water vapor transport (IVT), 1000–700 hPa mean wind (arrows), and 500 hPa height (gray isolines), as well as the 50% sea ice concentration (SIC) boundary (pink line) and areas of Greenland Ice Sheet (GrIS) surface melt (red shaded zones). Areas in white have less than 25 kg m^{-1} s^{-1} IVT. Regional map on right shows 2-meter relative humidity (primary shading) and temperature (red and blue isolines) along with 10-meter wind (arrows). All data are from MERRA-2 (Stein et al., 2015; Gelaro et al., 2017) except for ice sheet surface melt, which is from the NSIDC MEaSUREs Greenland Surface Melt daily data set derived from passive microwave satellite observations (Mote, 2014).

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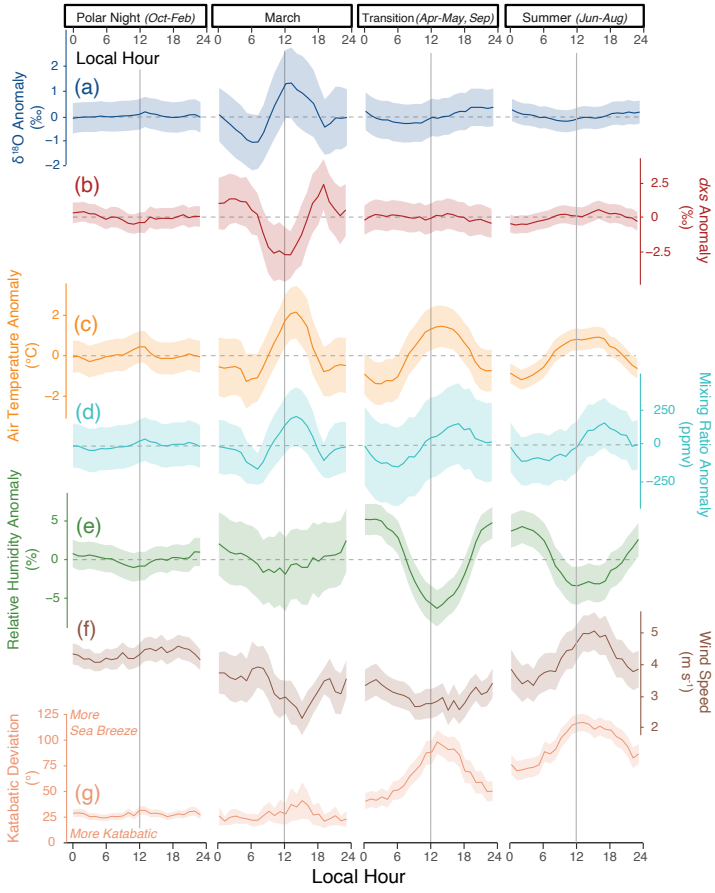

Figure 8. Diel patterns in isotopic and meteorological variables in Thule, Greenland, showing the mean hourly value (solid thick line) and 95% confidence intervals of mean estimates (colored shading) for each diel regime. Diel patterns for δ^2H are very similar to $\delta^{18}O$ (a) and not shown. Variables other than wind speed and katabatic deviation are expressed as deviations from the mean value in each regime, with zero deviation indicated as a dashed horizontal line. Note that because the winds in Thule are binary (katabatic or sea breeze), the diel means of katabatic deviations should be viewed as a probability of being katabatic or sea breeze at a given time and not that the winds are smoothly transitioning from east to west or vice versa. Additionally, because katabatic deviations increase in value both directions away from 100° (e.g., winds from 80° and 120° both have a katabatic deviation of +20°), the natural variance of purely katabatic winds produces an average value of ~25° rather than 0°.

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Table 1. Annual and seasonal mean ± 1 standard deviation values of selected water vapor isotopic and meteorological variables for Thule, Greenland, over the analytical period of this study. Values calculated using 10 minute resolution data.

	$\delta^{18}O$ (‰)	$\delta^{2}H$ (‰)	*dxs* (‰)	Air temperature (°C)	H_2O mixing ratio (ppmv)	Relative humidity (%)
Annual	-33.2 ± 6.1	-249 ± 43	+16.2 ± 8.1	-8.1 ± 11.1	4180 ± 3000	78 ± 16
Spring (MAM)	-35.9 ± 5.8	-270 ± 40	+17.7 ± 8.5	-12.1 ± 9.2	2850 ± 1940	79 ± 16
Summer (JJA)	-27.6 ± 3.5	-212 ± 24	+8.6 ± 4.9	+5.4 ± 4.1	8310 ± 1290	73 ± 18
Autumn (SON)	-31.3 ± 4.4	-233 ± 31	+17.7 ± 6.0	-8.5 ± 6.4	3600 ± 1740	78 ± 13
Winter (DJF)	-38.5 ± 3.6	-287 ± 26	+21.6 ± 6.4	-18.9 ± 5.2	1550 ± 900	83 ± 16

Table 2. Spearman correlation values between isotopic and meteorological variables at Thule. Two sets of correlations are reported here: analyses from the original data and analyses from the seasonally-adjusted data where the seasonal cycle was removed for isotopic, air temperature, mixing ratio, and sea ice data. Seasonally-adjusting the data weakens nearly all correlations by removing the common response to seasonal change, but strengthens isotopic correlations with relative humidity. Correlations are given for all available levels of temporal aggregation, but some variables do not have data at finer resolutions. Temporal aggregation slightly strengthens correlations, but does not generally change the order of variables when ranking by correlation strength. Due to the large sample sizes, all correlations are significant at $p < 0.05$ except for a few with very weak correlations (*italicized*).

	ρ (10 min)			*ρ (hourly)*			*ρ (daily)*		
Original	$\delta^{18}O$	$\delta^{2}H$	*dxs*	$\delta^{18}O$	$\delta^{2}H$	*dxs*	$\delta^{18}O$	$\delta^{2}H$	*dxs*
Air temperature	+0.76	+0.73	-0.74	+0.76	+0.73	-0.75	+0.81	+0.77	-0.79
H_2O mixing ratio	+0.83	+0.80	-0.79	+0.83	+0.80	-0.79	+0.86	+0.83	-0.82
Relative humidity	+0.06	+0.06	-0.07	+0.06	+0.06	-0.07	*0.00*	*-0.01*	*-0.01*
Station pressure	+0.09	+0.08	-0.12	+0.09	+0.08	-0.12	+0.10	+0.09	-0.13
Wind speed	-	-	-	-0.08	-0.07	+0.11	-0.14	-0.13	+0.16
Katabatic deviation	-	-	-	+0.50	+0.48	-0.50	+0.48	+0.47	-0.41
NAO	-	-	-	-	-	-	-0.25	-0.23	+0.29
AO	-	-	-	-	-	-	*-0.06*	*-0.04*	+0.17
Baffin Bay ice extent	-	-	-	-	-	-	-0.69	-0.71	+0.47
Seasonally-adjusted	$\delta^{18}O$	$\delta^{2}H$	*dxs*	$\delta^{18}O$	$\delta^{2}H$	*dxs*	$\delta^{18}O$	$\delta^{2}H$	*dxs*
Air temperature	+0.24	+0.20	-0.31	+0.25	+0.20	-0.32	+0.32	+0.27	-0.38
H_2O mixing ratio	+0.55	+0.49	-0.58	+0.55	+0.49	-0.58	+0.57	+0.51	-0.60

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Relative humidity	+0.46	+0.43	-0.44	+0.38	+0.36	-0.33	+0.32	+0.30	-0.29
Station pressure	+0.04	+0.03	-0.07	+0.04	+0.03	-0.07	*+0.03*	*+0.02*	-0.07
Wind speed	-	-	-	*-0.03*	*-0.02*	+0.06	*-0.03*	*-0.03*	*+0.06*
Katabatic deviation	-	-	-	+0.19	+0.17	-0.18	+0.10	+0.09	*-0.03*
NAO	-	-	-	-	-	-	-0.14	-0.12	+0.20
AO	-	-	-	-	-	-	*-0.04*	*-0.02*	+0.17
Baffin Bay ice extent	-	-	-	-	-	-	-0.02	-0.03	+0.05

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S1 Quality checking and instrument stability issues

The raw data from the L2130-i was passed through a series of quality checks prior to humidity response correction to remove readings that were well outside typical values observed at Thule. Most of these erroneous readings appear due to liquid water contamination of the intake tubing, likely from precipitation during intense cyclones. These quality checks identified and removed data where:

- L2130-i diagnostic data (e.g., chamber temperature, pressure, status, etc.) were unstable or out of typical ranges
- Water vapor mixing ratio >15000 ppmv
- $\delta^{18}O$ values >15‰ AND *dxs* values <20‰
- Standard deviation for five minute aggregates of $\delta^{18}O$ >8‰

An additional quality check was performed after humidity response calibration. Visual inspection was used to identify clearly abnormal isotopic or mixing ratio values (e.g., very large and/or abrupt changes not supported by meteorological data), and these observations were removed.

The machine was initially used at a tundra-based field site in Thule for a summer project in 2015, and it was installed its present location in October 2016. However, there were issues with cavity pressure stability and irregular isotopic readings which culminated in a full systems crash in May 2017. The system was restored on 04 Aug 2017 with stable cavity pressure that has continued through present. Data from before the system restoration has poor correlation in water vapor mixing ratio between the L2130-i and the SMT weather station. Winter isotopic values and mixing ratios are also much higher in the pre-restoration data than the next two winters despite generally similar winter weather. Out of caution, we have restricted our analyses and discussion to only post-restoration data.

S2 Humidity response calibrations

To correct for $\delta^{18}O$ and $\delta^{2}H$ accuracy and precision bias at low water vapor mixing ratios, we injected standard waters for ten minutes at ten different flow rates. The last 200 observations of each injection were saved, and a nonlinear regression was performed on the δX vs. mixing ratio relationship, where δX is either $\delta^{18}O$ or $\delta^{2}H$, to determine accuracy corrections. The nonlinear regression was of the form:

$$\delta X_{correction} = a + \frac{b}{q} \quad (S1)$$

where $\delta X_{correction}$ is the difference between the observed isotopic value and the actual standard isotopic value, q is the water vapor mixing ratio, and a and b are constants. Calculated values for regression parameters are given in Table S4. Confidence

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intervals for predicted humidity response corrections were estimated using the predictNLS function from the *propagate* package in R.

Changes in analytical precision at low water vapor mixing ratios were calculated with a nonlinear regression of the form:

$$\delta X_{precision} = a + \frac{b}{q} \quad \text{(S2)}$$

where $\delta X_{precision}$ is the standard error of the mean isotopic value for a given flow rate, q is the water vapor mixing ratio, and a and b are constants. Calculated values for regression parameters are given in Table S5.

S3 Isotope–isotope relationships

Over the full dataset, $\delta^{18}O$ and $\delta^{2}H$ have a strong linear relationship with low parameter standard error: $\delta^{2}H = 6.959 \pm 0.003 * \delta^{18}O - 18.07 \pm 0.09‰$ ($r^2 = 0.98$, n=111138, 10 min data). Overall, this value is comparable to other slopes observed at other high latitude sites, such as 6.8 at Ivittuut, Greenland, (Bonne et al., 2014), 6.5 at NEEM, Greenland, (Steen-Larsen et al., 2013), 6.0–6.5 at Dome C, Antarctica (Casado et al., 2016), and 6.95 from the vapor mixing line at Kangerlussuaq, Greenland (Kopec et al., 2014). Changes in $\delta^{18}O$ are thus closely mirrored in $\delta^{2}H$, and most differences are only detectable on very short timescales (i.e., less than hourly) when some minor lead-lag between relative maxima and minima may occur. The *dxs* at Thule is negatively correlated with both $\delta^{18}O$ and $\delta^{2}H$ (ρ_{10min} = -0.78 and -0.70, respectively).

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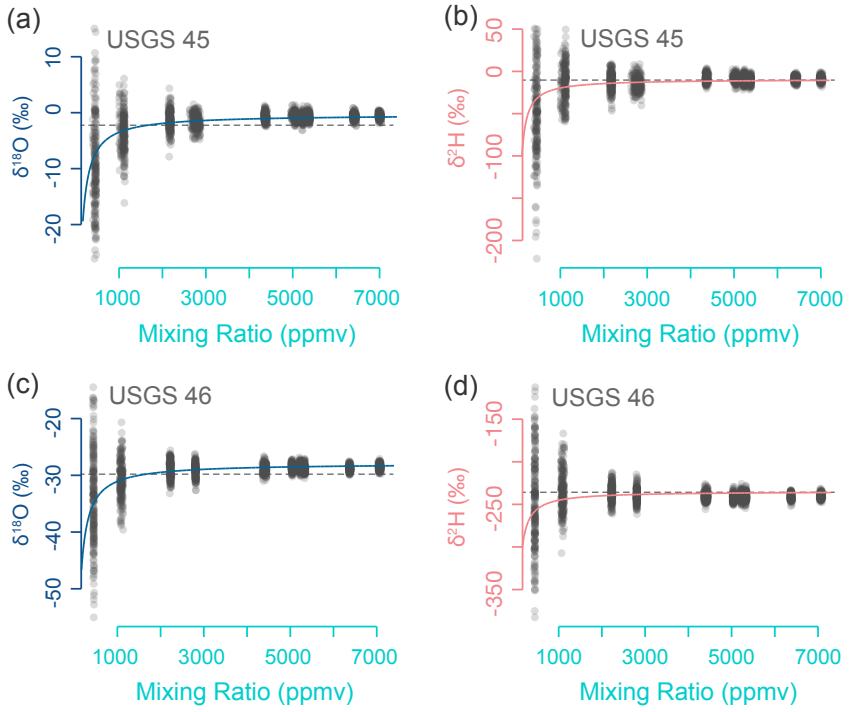

Figure S1. Results of the humidity response calibrations for $\delta^{18}O$ (a, c) and $\delta^{2}H$ (b, d) for two standard waters (USGS 45: top row and USGS 46: bottom row). Points show individual 1 s^{-1} observations, while solid lines show the nonlinear regression of the data (blue: $\delta^{18}O$, pink: $\delta^{2}H$). Dashed horizontal lines show the actual isotopic value of the standard waters.

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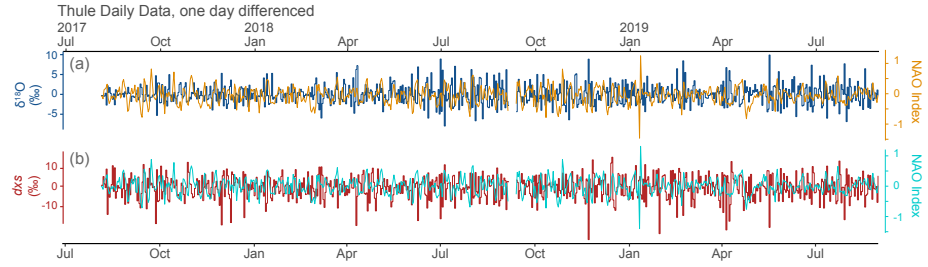

Fig S2: Comparison between time series of (a) $\delta^{18}O$ and (b) *dxs* with NAO index, after all data have been differenced by one day.

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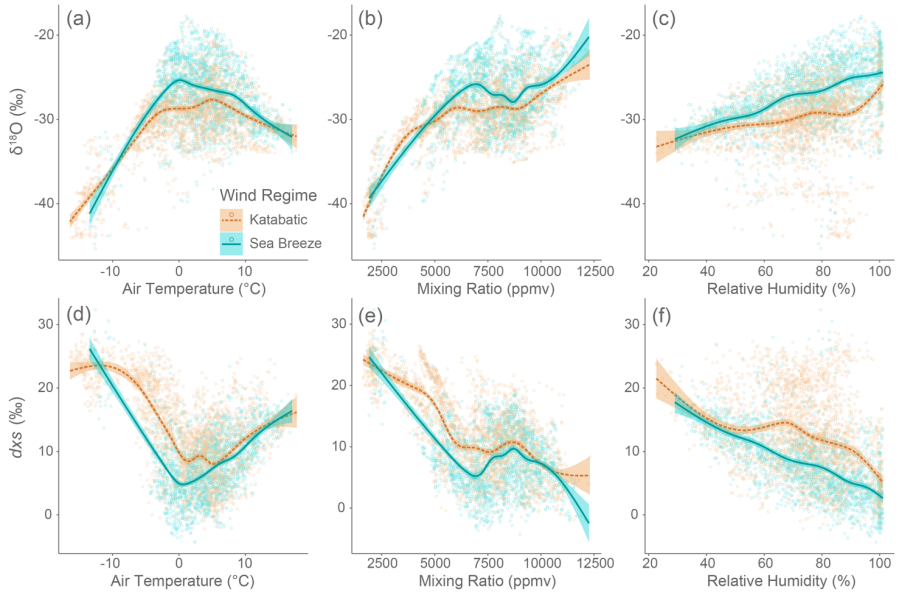

Figure S3. Water vapor $\delta^{18}O$ (top row; a–c) and *dxs* (bottom row; d–f) compared to meteorological variables for the period April through September. Data are split by whether the wind regime was katabatic (azimuth > 40° & < 180°) or sea breeze (azimuth > 240° & < 360°). A generalized additive smoothing model with 95% confidence intervals is overlaid to show trends of the mean values for each wind regime.

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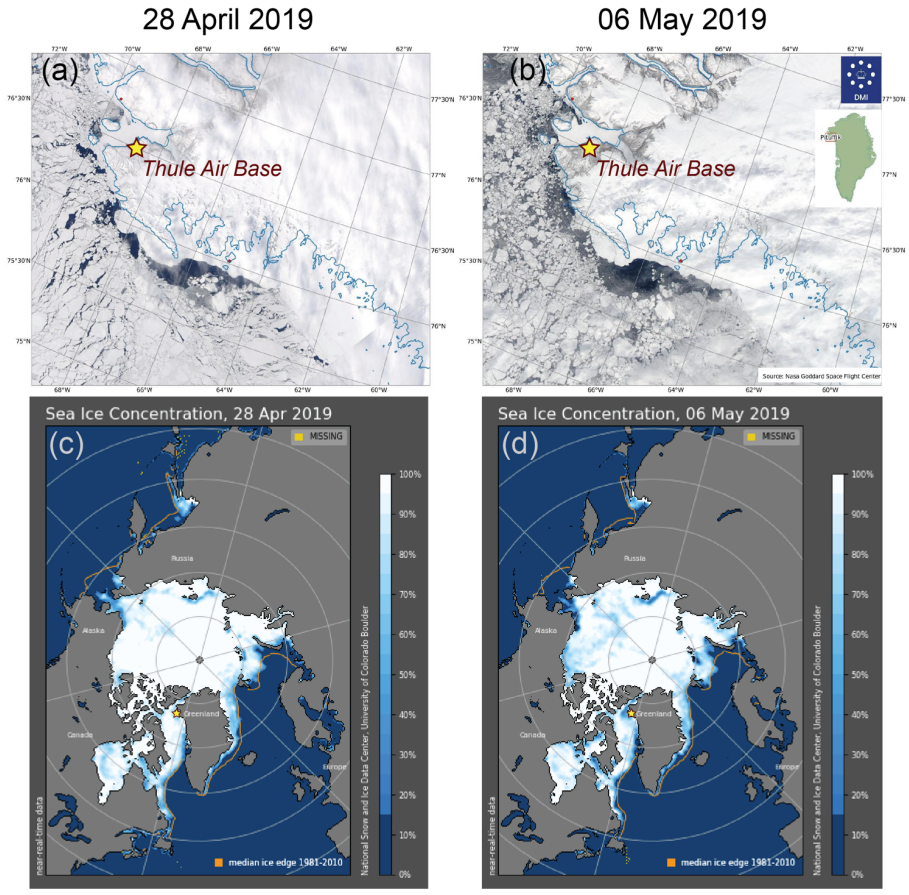

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Figure S4: Satellite imagery (top row; a, b) and sea ice concentration (bottom row; c, d) illustrating sea ice conditions before (left column; a, c) and after (right column; b, d) the late spring shift in isotopic and meteorological values at Thule in 2019. The images illustrate the opening of local oceans and snowpack melt as a result of local sea ice breakup. Satellite imagery provided by MODIS (Hall and Riggs, 2015) and sea ice concentration by the NSIDC (Fetterer et al., 2017).

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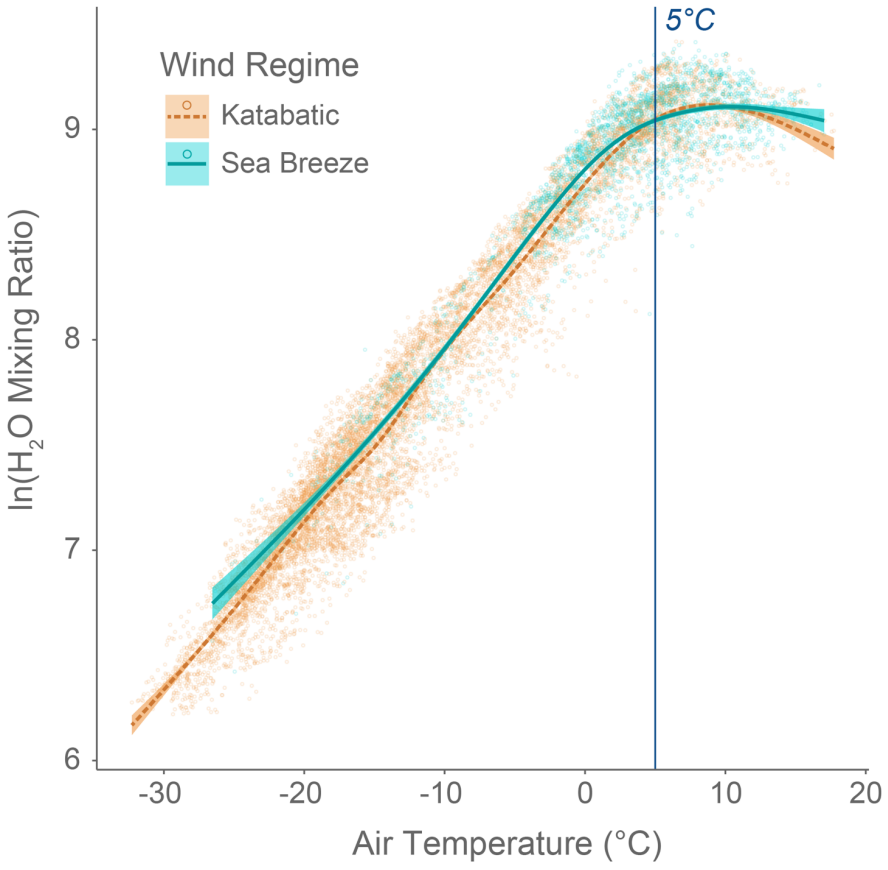

Figure S5. Relationship between air temperature and water vapor mixing ratio (here, log-transformed), illustrating the decoupling that occurs above 5°C (vertical line). Data are split by whether the wind regime was katabatic (azimuth > 40° & < 180°) or sea breeze (azimuth > 240° & < 360°). A generalized additive smoothing model with 95% confidence intervals is overlaid to show trends of the mean values for each wind regime. The relationship decoupling exists in the data from both wind regimes.

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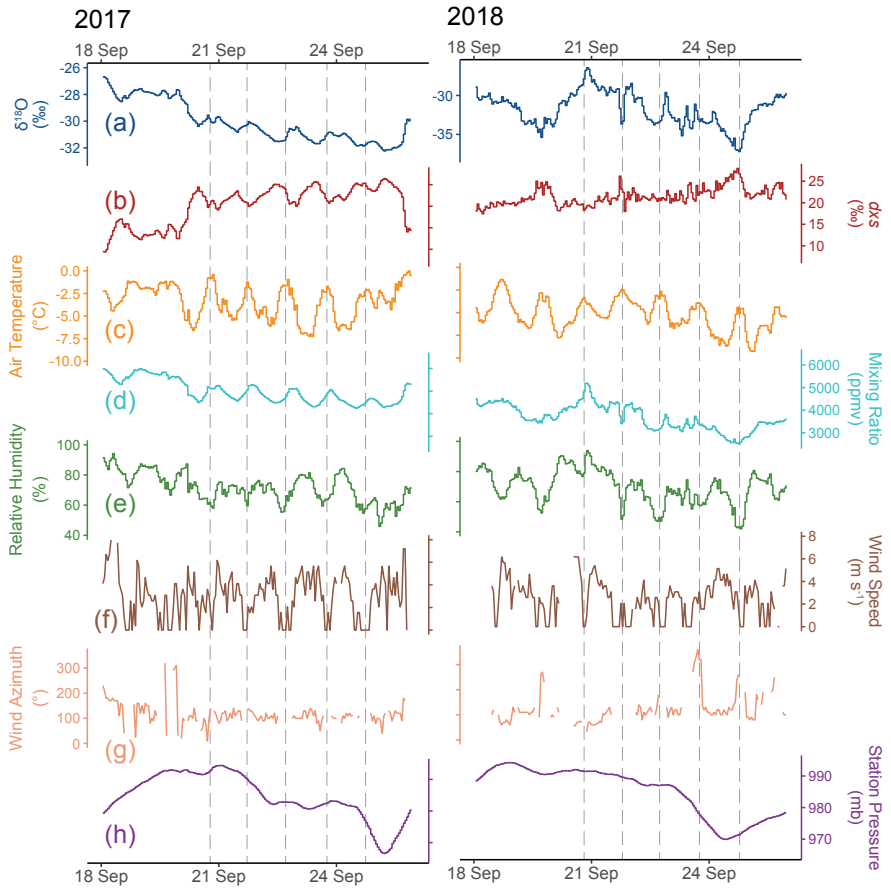

Figure S6. Comparison of isotopic and meteorological data during the period 18–26 Sep for 2017 (left) and 2018 (right). A diel cycle is observed in air temperature, relative humidity, and wind speed in both years, but in the isotopes and mixing ratio only for the year 2017. Y-axes are scaled the same for both years with the exception of $\delta^{18}O$. Dashed gray vertical lines indicate the time of daily thermal max from 20–25 Sep. The 2018 example, when no little to no isotopic cycling is observed, is representative of nearly all other autumn periods at Thule when diel cycles in temperature and relative humidity can be clearly observed in the general time series data.

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Table S1. High latitude stationary sites with continuous observations of water vapor isotopes longer than two weeks reported in scientific literature.

Site	Coordinates	Elevation	Setting	Period of record	Citation
Thule, NW Greenland	76.51°N, 68.74°W	229 m a.s.l.	Coastal	08/2017 to 08/2019	This study
Ivittuut, S Greenland	61.21°N, 48.17°W	30 m a.s.l.	Coastal	09/2011 to 05/2013	(Bonne et al., 2014)
Kangerlussuaq, W Greenland	67.02°N, 50.69°W	49 m a.s.l	Coastal	07/2011 to 08/2011	(Kopec et al., 2014)
NEEM, NW Greenland	77.45°N, 51.05°W	2484 m a.s.l.	Ice sheet	05/2010 to 07/2010; 07/2011 to 08/2011; 05/2012 to 08/2012	(Steen-Larsen et al., 2013; Steen-Larsen et al., 2014)
Summit, Greenland	72.58°N, 38.46°W	3216 m a.s.l.	Ice sheet	Summer 2011 to summer 2014	(Bailey et al., 2015)
Selvogsviti, Iceland	63.83°N, 21.47°W	0 m a.s.l.	Coastal	11/2011 to 04/2013	(Steen-Larsen et al., 2015)
Kourovka, W Siberia	57.04°N, 59.55°E	300 m a.s.l.	Inland	09/2012 to 08/2013	(Bastrikov et al., 2014)
Samoylov Island, E Siberia	72.37°N, 126.48°E	0 m a.s.l.	Coastal	07/2015 to 06/2017	(Bonne et al., 2020)
Toolik Lake, N Alaska	68.63°N, 149.60°W	760 m a.s.l.	Inland	05/2013 to 08/2013	(Klein et al., 2015)
Dumont d'Urville, E Antarctica	66.65°S, 140.00°E	10 m a.s.l.	Coastal	12/2016 to 02/2017	(Bréant et al., 2019)
Syowa, E Antarctica	69.00°S, 39.58°E	0 m a.s.l.	Coastal	12/2013 to 02/2014; 12/2014 to 02/2015	(Kurita et al., 2016)
Kohnen, E Antarctica	75.00°S, 0.07°E	2892 m a.s.l	Ice sheet	12/2013 to 01/2014	(Ritter et al., 2016)
Dome C, E Antarctica	75.10°S, 123.39°E	3233 m a.s.l	Ice sheet	12/2014 to 01/2015	(Casado et al., 2016)

Table S2. Standard waters calibration results after dry air system installation, performed roughly every 25 hours. Mean values and standard deviations of the last 200 observations for each calibration are given here. Some days do not have data due to failed calibration from a clogged injection needle or stuck injection piston. Some of the calibrations included here extend beyond the limit of ambient data discussed in the study, but station operation has continued without interruption.

	USGS 45							USGS 46					
	Mixing ratio (ppmv)		$\delta^{18}O$ (‰)		$\delta^{2}H$ (‰)			Mixing ratio (ppmv)		$\delta^{18}O$ (‰)		$\delta^{2}H$ (‰)	
Date	Mean	SD	Mean	SD	Mean	SD		Mean	SD	Mean	SD	Mean	SD
2019-08-01	5367	20	-0.8	0.6	-11	4		5325	14	-28.8	0.6	-241	4
2019-08-02	5378	16	-0.9	0.6	-9	4		5345	10	-28.9	0.6	-239	4
2019-08-04	5412	19	-0.7	0.7	-10	4		5396	14	-28.7	0.6	-240	4
2019-08-05	5421	18	-0.8	0.6	-10	4		5406	14	-28.8	0.6	-241	4
2019-08-06	5450	15	-0.7	0.6	-10	4		5446	17	-28.7	0.6	-240	4
2019-08-07	5459	15	-0.6	0.6	-10	4		5430	18	-28.6	0.6	-240	4
2019-08-09	5480	16	-0.7	0.6	-10	4		5431	17	-28.8	0.6	-240	4
2019-08-10	5438	12	-0.9	0.6	-10	4		5448	19	-28.7	0.6	-239	4
2019-08-11	5467	15	-0.8	0.6	-10	4		5476	59	-28.5	0.6	-239	4
2019-08-12	5392	57	-0.8	0.7	-10	4		5531	91	-28.5	0.7	-240	4

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2019-08-14	5520	20	-0.7	0.6	-9	4		5524	18	-28.6	0.7	-239
2019-08-15	5523	13	-0.8	0.6	-10	4		5528	21	-28.8	0.6	-240
2019-08-16	5574	19	-0.6	0.6	-10	4		5568	20	-28.5	0.6	-240
2019-08-17	5599	31	-0.6	0.6	-9	4		5535	38	-28.4	0.6	-239
2019-08-18	5492	38	-0.5	0.7	-9	4		5545	24	-28.8	0.7	-240
2019-08-19	5478	61	-0.9	0.6	-10	4		5573	20	-28.6	0.6	-239
2019-08-20	5610	18	-0.5	0.6	-9	4		5544	79	-28.8	0.6	-240
2019-08-22	5488	43	-1.1	0.6	-11	4		5564	15	-28.6	0.6	-240
2019-08-23	5605	13	-0.6	0.6	-9	4		5622	18	-28.6	0.6	-240
2019-08-24	5566	17	-0.9	0.6	-10	4		5591	17	-28.6	0.6	-240
2019-08-25	5642	9	-0.5	0.5	-10	4		5602	38	-28.3	0.7	-239
2019-08-26	5519	32	-1.0	0.6	-11	4		5641	19	-28.6	0.6	-240
2019-08-27	5674	18	-0.5	0.6	-9	4		5620	24	-28.6	0.6	-240
2019-08-28	5616	14	-0.5	0.6	-10	4		5602	64	-28.8	0.6	-241
2019-08-29	5642	13	-0.5	0.6	-9	4		5449	104	-29.1	0.7	-241
2019-08-31	5632	11	0.0	0.7	-9	4		5655	18	-28.7	0.7	-241
2019-09-01	5719	14	-0.6	0.7	-10	4		5668	17	-28.8	0.6	-240
2019-09-02	5727	29	-0.7	0.6	-10	4		5739	15	-28.6	0.6	-240
2019-09-03	5628	25	-0.5	0.7	-9	4		5671	20	-28.5	0.6	-241
2019-09-05	5687	27	-0.7	0.6	-10	4		5744	18	-28.5	0.6	-240
2019-09-06	5787	17	-0.5	0.6	-10	4		5734	21	-28.6	0.6	-241
2019-09-08	5769	15	-0.2	0.7	-9	4		5798	21	-28.6	0.6	-240
2019-09-09	5739	23	-0.3	0.7	-9	4		5762	20	-28.4	0.6	-241
2019-09-10	5729	13	-0.5	0.6	-9	4		5714	43	-28.6	0.6	-240
2019-09-11	5810	9	-0.5	0.6	-9	4		5749	19	-28.5	0.6	-240
2019-09-12	5737	20	-0.4	0.7	-9	4		5787	57	-28.4	0.6	-240
2019-09-13	5793	29	-0.9	0.6	-10	4		5788	28	-28.1	0.6	-239
2019-09-14	5871	17	-0.1	0.7	-8	4		5837	27	-28.4	0.6	-240
2019-09-16	5689	12	-0.6	0.6	-10	4		5852	24	-28.5	0.6	-240
2019-09-17	5723	37	-0.8	0.7	-10	4		5817	21	-28.3	0.7	-239
2019-09-18	5855	20	-0.3	0.6	-9	3		5901	35	-28.4	0.6	-240
2019-09-19	5864	11	-0.5	0.6	-9	4		5811	30	-28.7	0.7	-241
2019-09-20	5948	10	-0.5	0.6	-10	4		5966	14	-28.7	0.6	-241
2019-09-22	5942	12	-0.5	0.6	-9	4		5938	11	-28.6	0.6	-241
2019-09-24	5999	35	-0.4	0.6	-9	4		6009	17	-28.5	0.6	-240

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2019-09-25	5933	7	-0.4	0.6	-9	4		5929	15	-28.6	0.6	-240
2019-09-26	5887	15	-0.6	0.7	-9	4		5967	18	-28.4	0.6	-239
2019-09-27	5792	17	-0.9	0.6	-9	4		5855	21	-29.2	0.7	-242
2019-09-28	5887	13	-0.4	0.6	-9	4		5880	13	-28.5	0.6	-240
2019-09-29	5832	21	-0.3	0.5	-10	4		5866	16	-28.7	0.6	-241
2019-09-30	5815	32	0.1	0.6	-8	4		5837	19	-28.5	0.6	-241

Table S3. Isotopic value trends for standard water calibrations performed after dry air system installation (Table S3). Trends are calculated as the slope of the last 200 observations taken during a calibration versus time, with one observation per second. The mean and standard deviation of all calibration runs is given at the bottom of the table.

	USGS 45		USGS 46	
Date	$\delta^{18}O$ (‰ s^{-1})	$\delta^{2}H$ (‰ s^{-1})	$\delta^{18}O$ (‰ s^{-1})	$\delta^{2}H$ (‰ s^{-1})
2019-08-01	0.0000	0.0047	0.0006	-0.0043
2019-08-02	0.0004	0.0098	0.0001	0.0056
2019-08-04	0.0019	0.0036	0.0001	-0.0064
2019-08-05	0.0021	0.0008	0.0021	0.0037
2019-08-06	0.0015	-0.0063	0.0011	0.0089
2019-08-07	0.0008	-0.0014	0.0009	0.0004
2019-08-09	0.0014	0.0089	-0.0006	-0.0015
2019-08-10	0.0014	0.0016	0.0001	0.0018
2019-08-11	-0.0002	0.0033	0.0004	0.0061
2019-08-12	-0.0032	0.0059	-0.0003	-0.0030
2019-08-14	0.0013	0.0055	-0.0001	-0.0027
2019-08-15	-0.0001	-0.0106	0.0030	0.0096
2019-08-16	0.0006	0.0052	0.0050	0.0064
2019-08-17	0.0007	0.0038	0.0015	-0.0016
2019-08-18	0.0028	0.0040	0.0039	0.0110
2019-08-19	0.0018	0.0122	0.0012	0.0085
2019-08-20	0.0001	0.0112	0.0008	0.0045
2019-08-22	0.0011	0.0115	0.0019	-0.0067
2019-08-23	-0.0014	0.0010	0.0014	0.0042
2019-08-24	-0.0005	0.0079	0.0002	-0.0020
2019-08-25	-0.0002	0.0122	0.0049	0.0058

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2019-08-26	-0.0023	0.0001	-0.0002	0.0041
2019-08-27	0.0015	0.0033	0.0010	0.0060
2019-08-28	-0.0009	0.0078	-0.0023	-0.0082
2019-08-29	-0.0003	0.0017	0.0009	-0.0018
2019-08-31	-0.0026	-0.0083	0.0035	0.0009
2019-09-01	-0.0031	0.0004	0.0017	0.0037
2019-09-02	0.0009	0.0113	0.0022	-0.0043
2019-09-03	0.0011	-0.0011	0.0006	0.0021
2019-09-05	-0.0011	0.0021	0.0026	0.0085
2019-09-06	-0.0002	-0.0034	0.0019	0.0014
2019-09-08	-0.0014	0.0030	0.0025	-0.0038
2019-09-09	-0.0014	-0.0119	0.0018	0.0101
2019-09-10	0.0002	-0.0063	0.0018	-0.0027
2019-09-11	-0.0017	-0.0009	0.0034	0.0017
2019-09-12	-0.0013	0.0051	0.0005	0.0064
2019-09-13	-0.0021	-0.0136	0.0022	0.0015
2019-09-14	-0.0018	0.0093	0.0017	0.0103
2019-09-16	-0.0013	-0.0036	0.0012	0.0079
2019-09-17	-0.0036	0.0061	0.0046	0.0107
2019-09-18	-0.0014	0.0007	0.0017	0.0033
2019-09-19	-0.0018	-0.0035	0.0066	0.0243
2019-09-20	0.0037	0.0074	-0.0007	-0.0059
2019-09-22	-0.0025	-0.0006	-0.0004	-0.0009
2019-09-24	0.0000	0.0086	0.0015	0.0040
2019-09-25	0.0012	0.0037	0.0013	0.0059
2019-09-26	-0.0023	-0.0024	0.0030	0.0114
2019-09-27	0.0013	0.0107	-0.0032	-0.0111
2019-09-28	0.0000	0.0014	0.0016	0.0032
2019-09-29	-0.0016	0.0032	0.0018	-0.0081
2019-09-30	-0.0017	0.0058	0.0004	0.0048
Mean	-0.0003	0.0026	0.0014	0.0026

Standard deviation	0.0016	0.0062	0.0018	0.0064

Table S4. Parameters for nonlinear regression of the humidity response accuracy corrections (Equation S1).

Isotopic species	Parameter	Estimate	Standard error	t value	Pr(>|t|)
$\delta^{18}O$	*a*	1.92	0.061	31.6	<0.001
	b	-3190	80	-42.1	<0.001
$\delta^{2}H$	*a*	1.13	0.41	2.8	0.005
	b	-10500	510	-21.0	<0.001

Table S5. Parameters for nonlinear regression of the humidity response precision (Equation S2).

Isotopic species	Parameter	Estimate	Standard error	t value	Pr(>|t|)
$\delta^{18}O$	*a*	-0.003	0.002	-1.4	0.177
	b	260	3	102.2	<0.001
$\delta^{2}H$	*a*	-0.019	0.016	-1.2	0.238
	b	1690	20	85.7	<0.001

Table S6. Percentage of water vapor uptake attributed to Baffin Bay and the Labrador Sea for moisture arriving at Thule, based on back-trajectory analysis and split by meteorological season.

Domain	DJF	MAM	JJA	SON
Entire Baffin Bay (Davis to Nares)	35.6	44.4	51.2	47.2
Labrador Sea	13.7	16.3	12.2	11.7
Entire Baffin Bay + Labrador Sea	49.3	60.7	63.4	58.9

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