

Stable isotopes unveil ocean transport of legacy mercury into Arctic food webs

Corresponding Author: Dr Jens Søndergaard

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Review of NCOMMS-24-75705

"Stable isotopes unveil ocean transport of legacy mercury into Arctic food webs"

December 18, 2024

Summary: This study used mercury (Hg) stable isotopes to study numerous environmental processes in Arctic ecosystems. Using both new and historical samples and data from long-term monitoring by the AMAP program, the study generates several notable findings. First, the isotopic modeling suggests that ocean currents are the main transport pathway of legacy Hg to Greenland. This is in notable contrast to our traditional understanding of Arctic Hg cycling, which suggests the majority of Hg is deposited from the atmosphere (through AMDE events, which are discussed in the manuscript). Secondly, this study suggests that the dominant species deposited and accumulated is Hg(0), not Hg(II). Additionally, the authors support that photo-demethylation continues to be an important process impacting Arctic Hg cycling. Their work further supports growing evidence that, while Hg emissions are reducing, concentrations (and, according to their study, the sources of this Hg) are not responding similarly and are, in fact, increasing in some biota. Collectively, these findings have implications for long-term mercury trends and necessary monitoring, given that the lifetime of Hg in oceans is longer than atmospheric sources used in most temporal models to-date.

Overview: Overall, this is a strong paper that impacts our collective understanding of Hg cycling. The data are valuable and expansive, both in a temporal and geographic sense. Their methods are appropriately detailed, particularly around the various lab analyses. However, the manuscript is particularly dense, assessing processes from atmospheric to ecological, which has resulted in deficient details (e.g., statistical information) and discussion content in some places (e.g., photochemical impacts on bioaccumulation, see detailed comments below). In general, the results and discussion around abiotic cycling are much stronger and more complete than the discussions around trophic transfer and metabolism. This may be due, in part, to a firmer collective understanding of isotopic behavior during the former processes, but additional details are warranted in the latter, where possible. We additionally suggest the authors check their references to figure panels within the text, as we found several errors (noted below).

Specific points:

1. Line 55: Food webs cannot be slow-growing. We believe the authors may mean the biota within those food webs are slow growing, which we agree can increase mercury accumulation. We suggest some minor re-wording for clarification around this point.
2. Figure 1: What is the source of information for the ocean currents presented in part (a). Please include relevant citations.
3. Line 98: Should the reference to Figure 1C be 1D in this line?
4. Line 114: There is some confusing language in this line. You begin with "As for Peat" but discuss muscle results. You might consider rewording for clarity.
5. Jiskra et al. (2021) use their endpoints to study terrestrial environments. Can the authors comment on the relevance of these endpoints when translated to aquatic ecosystems? Are there any differences in fractionation that should be accounted for in the interpretation herein or would all the changes from atmospheric speciation be equally reflected in terrestrial and aqueous environments?
6. Lines 180-183: The results herein are important. Where can the data supporting this conclusion be seen? Figure 2, we assume. Please add a reference to relevant information to allow for independent reader interpretation.

7. Line 246: "Residual Hg from photochemical demethylation" would inherently not be methylmercury, which is commonly assumed to be the dominate form bioaccumulated in the biota and tissues reference herein. Thus, these estimates are surprising. The authors don't explore this or explain potential mechanisms. We suggest some discussion is added around these points within lines 242-250, or clarifications to your phrasing as we are mis-interpreting this sentence.
8. Lines 305-307: More support for this statement is needed. Were benthic organisms tested in this study to make this inference, or is this based on previous work? If the latter, citations are needed.
9. Lines 388-395: The direct link between water mass exchange, ecosystem size/location, and legacy Hg is unclear. Ocean spray and local atmospheric processes would surely impact coastal ecosystems, marine and freshwater alike. Could the authors elaborate on these points?
10. Line 521: Seabirds are used to make local inferences; how mobile is this gull species? Is there a confounding factor of movement in the interpretation of their Hg concentrations and isotopes that should be accounted for herein? The same could be asked of seals and polar bears.
11. Line 569: We would argue that blanks test for contamination, not accuracy of precision. This is a minor point, but important nonetheless. Otherwise, We appreciate the authors clear reporting of their QA/QC procedures and results.
12. Where possible, statistical details (e.g., p-values, and metrics of relationship strength) should be added to results. We find the method statement about p-values in line 585 to be insufficient. In particular, line statistics (including slopes, r^2 , and p-values) would be helpful in figures 4 and extended figure 2 and allow for more effective reader interpretations.
13. Line 578: The program used for statistical analyses and graphing should be noted. If R was used, any specific packages (and their versions) should also be noted, as this can affect the background settings and statistical inferences.
14. Line 332-334: Should the reference to Figure 4C be 4B? We suggest double-checking the figure references in this section.
15. Figure 4: Beyond diet, which only explains ~ 0.5 per mil of the differences in slopes, what other factors explains the difference in temporal response between seal and bears, two species which should be trophically linked. If this trend is otherwise geographically impacted, we would expect both species to show an impact.

Reviewer #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3

(Remarks to the Author)

The authors provide an in-depth and comprehensive assessment on spatio-temporal variation in mercury source and pathway dynamics in the Arctic using total Hg and Hg isotope measurements of peat and several key marine organisms. The authors attribute increases in Hg for some species to spatial differences in ocean current transport of legacy Hg up the food web. I found the methodological approach, experimental design and timeline investigated to be novel with results having high impact. I really enjoyed reading this well-written and well thought-out manuscript where the data and results support the main conclusions of the paper. This manuscript will definitely be of interest to readers of Nature Communications and the broader scientific community. I only have a few very minor comments.

- 1) In the Summary section, suggest modifying "internal metabolic fractionation" to "internal metabolism and isotopic fractionation."
- 2) Bottom of page 9 where the authors are inferring species trophic level from $\delta^{202}\text{Hg}$ values – I'm curious whether the authors investigated how well $\delta^{202}\text{Hg}$ correlates with $\delta^{15}\text{N}$ across species. $\delta^{15}\text{N}$ is an isotope that is very commonly used to estimate trophic level differences between species and this could be worthwhile to include in the supplementary materials section.
- 3) Top of page 10 when describing the foraging ecology of glaucous gulls – it's also worthwhile to state that they are also scavengers.
- 4) Top of page 10 when describing the foraging ecology of polar bears – it's also worthwhile to briefly state some of the other food items (i.e. bearded seals, narwhal, Atlantic walrus)

Reviewer #4

(Remarks to the Author)

The authors describe a study on total Hg concentrations and Hg isotopes in peat profiles and different key Arctic aquatic predators at different locations in Greenland. The major aims of this study are 1: to track Hg sources and pathways to Greenland 2: to show the development of total Hg and Hg isotopes in aquatic predator species in the past 40 years and how the development of the Hg content compares to that of anthropogenic atmospheric Hg emissions. 3: to show if there is internal metabolic fractionation of Hg isotopes in the different investigated aquatic species. 3:
The authors could show that there are distinct regional differences in total Hg and Hg isotopes ($\delta^{202}\text{Hg}$ signatures) between central western Greenland and northern-eastern Greenland which they suggest to be caused by different marine currents with

different dissolved Hg concentration rather than by different atmospheric sources. Moreover they observed that Hg concentrations in Arctic aquatic predatory species have increased in the past 40 years opposite to the decrease of atmospheric Hg emissions and atmospheric Hg concentrations. From this they concluded that rather delayed mobilised legacy Hg than direct atmospheric Hg deposition is the major pathway of Hg to arctic aquatic predators and Greenland soils/peat (e.g. through seaspray aerosols).

This study is innovative and of importance and shows important findings on the distribution of Hg contamination in the aquatic environment in Greenland. To my knowledge, this is the first and most comprehensive data-set of Hg concentrations and Hg isotopes from Greenland biota and from Arctic biota in general including monitoring data of aquatic species of the past 40 years, which I think is crucial to understand the biogeochemical Hg cycling in the arctic.

For me, the merit of this study lies in the large and unique data set of Hg concentrations and isotopes in Arctic biota, which gives new insights into the large variability in Hg enrichment and Hg isotope fractionation due to different food sources and through internal metabolic processes and indicates that processes and pathways of Hg and Hg isotope fractionation are less clear than previously assumed. The discussion here appears to be sound, although the authors clearly point out that more research is needed here.

However, even if I think that it is a plausible assumption, the conclusion that the major pathway of Hg to the marine and the terrestrial Greenland foodchains is Hg evasion from different ocean currents is not really supported by the data. The differences in dissolved Hg data which the authors have drawn from the literature may explain at least partly the differences observed in marine predatory species at different locations in Greenland, but cannot explain the differences in peat or landlocked Arctic char.

Frankly, I do not understand why the authors have included the Hg data from the peat/palsa profiles in this study. In the introduction the authors do not clearly explain why they believe that the approach of using Hg data of peat for Hg source tracing (marine versus atmosphere) is reasonable. First, the investigated peat deposits are quite different and as it seems, that they are not ombrothrophic (all elements atmospherically derived), which makes a solely marine Hg source appear doubtful. Moreover, instead of using Hg accumulation rates to estimate differences in Hg flux or Hg accumulation the authors base their interpretation only on peat Hg concentrations. Many studies have shown that Hg concentrations in peat alone cannot be used to derive differences in atmospheric fluxes as they are strongly influenced by peat decomposition, plant species or hydrology etc.. Similar, the interpretation of $\delta^{202}\text{Hg}$ in peat as a source proxy is doubtful as long as peat decomposition and related Hg isotope fraction has not been considered. The authors actually mention this limitations. Generally, $\delta^{202}\text{Hg}$ as a source proxy appears to be doubtful regarding that the authors show intense internal metabolic $\delta^{202}\text{Hg}$ fractionation in their biota.

I suggest that the authors focus on the Hg data in biota as this data set is unique and important e.g. the finding that Hg concentrations in hair and liver of polar bears show an increasing trend with time while Hg concentration in muscle decrease, is remarkable. I also believe that the authors should attenuate their conclusions regarding the role of different Hg currents as different Hg sources because the data supporting this conclusion is rather weak. Discussing the inverse trend of Hg accumulation in biota and the atmosphere is the most important finding here. In this context, I further propose to remove the peat data from the manuscript as it does not really support the discussion on Hg sources or transport pathways.

Please mention number of samples in figure captions, respectively.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Summary: We have re-reviewed NCOMMS-24-75705 "Stable isotopes unveil ocean transport of legacy mercury into Arctic food webs" including the response to reviewers file. Overall, we find the authors responses to be thorough and detailed, addressing most of our original concerns and corrected areas where we believed were lacking with sufficient details. The inclusion of more robust evidence and clarity on connections between the peat data and biological data is especially well written. We have one final point around the differences in temporal trends between polar bears and seals for consideration, detailed below.

Recommendation: Overall, we encourage publication of the manuscript.

Detailed points:

1. Our comments were addressed, largely through text modification and clarifications. This notably includes: (1) incorporating recently published literature (e.g., Huang et al. 2025); (2) a more detailed (i.e., well cited) paragraph around animal migration impacts near line 521; (3) more robust statistical details (p-value reporting and method information); (4) 2. For our (Review 1) point 15: In some ways, the authors' answer contradicts itself: differences in trends are due to changes in polar bear diets, from rings seals to hooded seals, but what are the diet differences in the seals themselves. We understand that you do not have data on that, but there are past studies done that could build a much more robust discussion around this point, in lieu of direct data. E.g., Schiøtt, S., Holland, D.M., Holland, D. et al. The diet and diving behaviour of the ringed seal (*Pusa hispida*) in Kangia (Ilulissat Icefjord), Greenland. *Polar Biol* 47, 1409–1422 (2024). <https://doi.org/10.1007/s00300-024-03310-4>.

a. More broadly, the authors also mention 3 sources of variability in their answer: sources, biogeochemistry, and diet. What about species-specific metabolism? That has been important in fish recently (as well as other groups like birds), with some taxonomic groups being identified as able to de-methylate MeHg or excrete high amounts of MeHg, relative to other species.

E.g., see the recent paper: Tepper, K., King, J., Manuneehi Cholan, P. et al. Methylmercury demethylation and volatilization by animals expressing microbial enzymes. *Nat Commun* 16, 1117 (2025). <https://doi.org/10.1038/s41467-025-56145-w>.

3. The authors responses to review 4's thorough comments are details and sufficient. While we do not disagree that the peat data made the original paper somewhat complex, we agree with the authors' holistic approach. Furthermore, we do not agree that the peat data is not inherently useful. While it may not serve as a direct measure of deposited Hg due to variations in decomposition rates and other factors, it remains a valuable data point when considered alongside biotic data. Given the distance between sites (1000 km shortest) there may be differences in biogeochemical properties in the area but that also occurring with similar changes in biota (internal metabolism) seem to be slim. Therefore, I believe the inferences drawn are reasonable and well-supported.

4. The added detail from Reviewer 4's comments seem to be redundant at times. More specifically, they added a lot of sentences that end with "...and that's why ocean currents seem to be the Hg source" The authors might consider compiling all of this evidence more concisely. However, we consider this a stylistic comment and defer to the editor on its applicability.

Reviewer #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3

(Remarks to the Author)

The authors have done a nice job addressing and incorporating reviewer comments. I recommend acceptance.

Reviewer #4

(Remarks to the Author)

The authors have generally improved the clarity of the manuscript and have addressed most of my concerns. However, they decided to keep the peat data in the manuscript although they now discuss the influence of peat decomposition etc. in more depth.

I still believe that the peat data is weak. The authors used only one record for each side of Greenland, respectively, and it remains doubtful that such a small data is representative. The variability of Hg uptake by peatland plants, and especially related Hg isotope fractionation, is not well understood and most existing data is from moss and not from vascular plants. Moreover, I don't think that the peat data (n=1 only) does add much valuable and robust information to the overall message of the study and I still think it should be removed. However, it seems that the information received from the peat data is not essential for the conclusion drawn why I think that it might be possible to publish the ms in its present form.

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Author's responses to reviewer comments (in green below)

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

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Author's response: We are grateful for the positive and thorough feedback. We find that the general comments have been elaborated in the specific comments below, so please find our responses here.

Specific points:

1. Line 55: Food webs cannot be slow-growing. We believe the authors may mean the biota within those food webs are slow growing, which We agree can increase mercury accumulation. We suggest some minor re-wording for clarification around this point.

#1: Author's response: We agree, and the sentence has now been changed to: *"Once Hg enters the Arctic, it biomagnifies in the long Arctic food web, creating high Hg loads in long-lived top predators such as toothed whales, polar bears (Ursus maritimus) and humans, thereby constituting a threat to biodiversity and health due to adverse neuroendocrine disruption."*

2. Figure 1: What is the source of information for the ocean currents presented in part (a). Please include relevant citations.

#2: Author's response: References are now inserted.²⁵⁻²⁷ (modified from ref. ²⁸)

3. Line 98: Should the reference to Figure 1C be 1D in this line?

#3: Author's response: We find that the sentence: *"The $\Delta^{199}\text{Hg}$ signatures are near-similar for Central West and Northeast Greenland in the litter and upper 5 cm active layer (medians, -0.17 and -0.15‰) (Fig 1b) and decrease with depth down to c. 50 cm (Supplementary Fig. 1c)"* refers correctly to Supplementary Fig. 1c.

4. Line 114: There is some confusing language in this line. You begin with "As for Peat" but discuss muscle results. You might consider rewording for clarity.

#4 Author's response: 'As for peat' was meant as 'Similar to peat'. The sentence has now been reworded for clarity:

"In the marine species, Hg stable isotopes in the muscle tissue of shorthorn sculpin and ringed seal reveal similar significant regional differences in $\delta^{202}\text{Hg}$ signatures between Central West Greenland, Northwest Greenland (Qaanaaq), and Central East Greenland (Ittoqqortoormiit) (Fig. 1c and d)."

5. Jiskra et al. (2021) use their endpoints to study terrestrial environments. Can the authors comment on the relevance of these endpoints when translated to aquatic ecosystems? Are there any differences in fractionation that should be accounted for in the interpretation herein or would all the changes from atmospheric speciation be equally reflected in terrestrial and aqueous environments?

#5: Author's response: Jiskra et al. (2021) used $\Delta^{200}\text{Hg}$ endpoints to study atmospheric mercury sources to the ocean (i.e. not terrestrial environments) (Jiskra, M. et al. Nature 597, 678-682 (2021)). Perhaps, it is a typing error, and the sentence should be reversed? However, regardless, $\Delta^{200}\text{Hg}$ is considered a conservative trace for atmospheric mercury deposition as $\text{Hg}(0)$ or $\text{Hg}(\text{II})$ to both marine and terrestrial environments. $\Delta^{200}\text{Hg}$ is thought to be produced by photochemical Hg redox reactions at or above the tropopause, and no relevant Hg transformations at the Earth's surface have thus far been shown to further result in changes in $\Delta^{200}\text{Hg}$. This is now elaborated in the introduction:

“Even-isotope MIF ($\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$) is considered linked to photo-oxidation of gaseous $\text{Hg}(0)$ high in the tropopause, and the $\Delta^{200}\text{Hg}$ signature is therefore commonly used as a conservative tracer of atmospheric Hg deposition pathways to both terrestrial surfaces and water bodies (i.e. deposition as $\text{Hg}(0)$ or $\text{Hg}(\text{II})$). ”

6. Lines 180-183: The results herein are important. Where can the data supporting this conclusion be seen? Figure 2, We assume. Please add a reference to relevant information to allow for independent reader interpretation.

#6: Author's response: Yes, correct, the conclusion points to the data shown in Fig. 2 and a reference to Fig. 2 has now been inserted:

“The $\Delta^{200}\text{Hg}$ signatures in the marine and freshwater species in this study indicate that between ~60 and ~87% of the Hg bioaccumulated in the species are atmospherically deposited as $\text{Hg}(0)$, while the remaining ~13 to ~40% are deposited via wet or dry $\text{Hg}(\text{II})$ deposition (Fig. 2).”

7. Line 246: “Residual Hg from photochemical demethylation” would inherently not be methylmercury, which is commonly assumed to be the dominate form bioaccumulated in the biota and tissues reference herein. Thus, these estimates are surprising. The authors

don't explore this or explain potential mechanisms. We suggest some discussion is added around these points within lines 242-250, or clarifications to your phrasing as we are misinterpreting this sentence.

#7: Author's response: We agree. The sentence should have been “Residual methyl-Hg from photochemical demethylation”. Now corrected.

8. Lines 305-307: More support for this statement is needed. Were benthic organisms tested in this study to make this inference, or is this based on previous work? If the latter, citations are needed.

#8: Author's response: We agree. Benthic organisms were not part of the study, but other studies have found that fish feeding on benthic organisms have lower $\Delta^{199}\text{Hg}$ than fish feeding on pelagic organisms. This is due to less light penetration and photochemical breakdown of methyl-Hg in the benthic environment. A reference to two of those studies (Perrot et al., 2012; Pinzone et al., 2021) is now provided in the text.

9. Lines 388-395: The direct link between water mass exchange, ecosystem size/location, and legacy Hg is unclear. Ocean spray and local atmospheric processes would surely impact coastal ecosystems, marine and freshwater alike. Could the authors elaborate on these points?

#9: Author's response: We agree. It is a good point to mention that ocean spray and local atmospheric processes such as oceanic evasion/redeposition will impact all coastal ecosystems, both marine and fresh water. Furthermore, since the manuscript was submitted, a study was published in Nature Communications showing that oceanic evasion and redeposition of gaseous elemental Hg is likely to drive transport to Arctic terrestrial ecosystems (Huang S. et al. Oceanic evasion fuels Arctic summertime rebound of atmospheric mercury and drives transport to Arctic terrestrial ecosystems, Nat. Commun. 16, 903 (2025)).

We have added the following sentence to the discussion: “However, sea spray and oceanic evasion/redeposition of Hg will impact all Arctic marine and coastal ecosystems and may also dominate summertime transport to Arctic terrestrial ecosystems⁴⁷.”

10. Line 521: Seabirds are used to make local inferences; how mobile is this gull species?

Is there a confounding factor of movement in the interpretation of their Hg concentrations and isotopes that should be accounted for herein? The same could be asked of seals and polar bears.

#10: Author's response: This is a good point, and we agree that the mobility of all species should be elaborated in the discussion. For the glaucous gulls, they come to East Greenland in spring and leave in late October and go to wintering areas in warmer latitudes. The wintering area for the East Greenland glaucous gull population is not known but is likely to include wintering areas in Iceland. This is now elaborated in the manuscript:

*“Sculpins are known as a sedentary benthic fish species with a generalist feeding strategy encompassing amphipods, polychaetes, decapods, molluscs, and other fish in their diet⁵⁰. Ringed seals in East and Northwest Greenland are known to feed mainly on polar cod (*Boreogadus saida*) and Arctic cod (*Arctogadus glacialis*), whereas ringed seals in Central West Greenland primarily prey on amphipods (*Parathemisto* spp.), capelin (*Mallotus villosus*), redfish (*Sebastes* sp.), and squid (*Gonatus* sp.)⁵¹. The ringed seal ecology is closely connected to the sea ice and most adult ringed seals are believed to be rather stationary, whereas juvenile ringed seals from different parts of the Arctic have been reported to travel over large distances (>1,000 km)⁵². Glaucous gulls are top predators and scavengers, and feed on a variety of prey items, including marine invertebrates, fish, bird eggs, and other birds⁵³. Glaucous gulls come to East Greenland in spring and leave in late October to winter at lower latitudes, possibly around Iceland⁵⁴. Polar bears are top predators and feed predominantly on ringed seals, hooded seals, and harp seals in East Greenland but occasionally also on other food items (e.g. bearded seals (*Erignathus barbatus*), narwhal (*Monodon monoceros*), and Atlantic walrus (*Odobenus rosmarus*))⁵⁵. The Central East Greenland polar bear subpopulation is one of 20 recognized subpopulations of polar bears in the world and the movement patterns cover >2,000 km of pack ice and fast ice along the East Greenland coast above 64°N⁵⁶.”*

11. Line 569: We would argue that blanks test for contamination, not accuracy of precision. This is a minor point, but important nonetheless. Otherwise, We appreciate the authors clear reporting of their QA/QC procedures and results.

#11: Author's response: We agree and have now adjusted the sentence: *“One blank sample and one NIST 8610 CRM were analyzed for at least every six samples to assess*

contamination/carry-over and accuracy/precision". We also updated the method section with a description of the methodology for the liver, hair, and kidney samples that were added to the manuscript late in the writing process along with QA/QC data for those.

12. Where possible, statistical details (e.g., p-values, and metrics of relationship strength) should be added to results. We find the method statement about p-values in line 585 to be insufficient. In particular, line statistics (including slopes, r², and p-values) would be helpful in figures 4 and extended figure 2 and allow for more effective reader interpretations.

#12: Author's response: *We agree and have now inserted trendline statistics in Fig. 4 and Fig. S-2 to allow for a more effective reader interpretation. To accommodate the line statistics in Fig. 4, we enlarged the figure to two column width. We chose to also display the individual points in the Fig. 4 (not only the medians) to visualize the variation in the data. Finally, since the original manuscript was written, we received 9 more specimens of ringed seal from Qeqertarsuaq collected in 2023 and 2024 and have updated the figures and text with data from those specimens (confirming the previous trends but now with 66 data points instead of 57, providing more solid conclusions).*

13. Line 578: The program used for statistical analyses and graphing should be noted. If R was used, any specific packages (and their versions) should also be noted, as this can affect the background settings and statistical inferences.

#13: Author's response: *Now inserted:*

"Statistical analyses were done using R version 4.2.3. One-way analysis of variance (ANOVA) was performed to determine differences in Hg stable isotope signatures between sites and species. Prior to the ANOVA, a Shapiro-Wilk test was conducted to test for normality of data, and a Brown-Forsythe test was undertaken to test for equal variances between groups. A Welch ANOVA test was applied due to unequal variances identified for some groups, and a Games-Howell post hoc test was used to assess individual group differences (using R-package 'rstatix 0.7.1'). Temporal trends in Hg stable isotope signatures were evaluated using the Mann-Kendall and Theil-Sen's slope analysis on subadult ringed seals (≤ 4 years) and polar bears (males ≤ 5 years; females ≤ 4 years) (using R-packages 'Kendall 2.2.1' and 'mblm 0.12.1')."

14. Line 332-334: Should the reference to Figure 4C be 4B? We suggest double-checking

the figure references in this section.

#14: Author's response: The section has now been checked and the references updated.

15. Figure 4: Beyond diet, which only explains ~ 0.5 per mil of the differences in slopes, what other factors explain the difference in temporal response between seal and bears, two species which should be trophically linked. If this trend is otherwise geographically impacted, we would expect both species to show an impact.

#15: Author's response: This is a good question. The $\delta^{202}\text{Hg}$ is affected by both source inputs to the ecosystem (i.e. oceanic, atmospheric and/or riverine) as well as biogeochemical transformation processes in the environment, and prey species/trophic levels. Consequently, the observed differences in $\delta^{202}\text{Hg}$ trends between ringed seals and polar bears could reflect different changes in diet across species. We can adjust for changes in diet in the polar bears as we have reported documented diet changes for the polar bears along with $\delta^{202}\text{Hg}$ data of the prey species. However, we do not have similar data available for the ringed seals. This is now mentioned:

"Part of the increase in $\delta^{202}\text{Hg}$ in the polar bears in Central East Greenland can be ascribed to a documented shift in diet during the period with consumption of significant less ringed seals with lower $\delta^{202}\text{Hg}$ (median, 0.28; $n = 82$) towards more hooded seals with higher $\delta^{202}\text{Hg}$ (median, 1.19; $n = 27$) (Supplementary Fig. 3)^{55,59}. When accounting for this change in diet composition, the increase in $\delta^{202}\text{Hg}$ is reduced from ~1.7‰ to ~1.2‰ (Fig. 4c). We do not have similar data for potential changes in diet for the ringed seals."

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3 (Remarks to the Author):

The authors provide an in-depth and comprehensive assessment on spatio-temporal variation in mercury source and pathway dynamics in the Arctic using total Hg and Hg

isotope measurements of peat and several key marine organisms. The authors attribute increases in Hg for some species to spatial differences in ocean current transport of legacy Hg up the food web. I found the methodological approach, experimental design and timeline investigated to be novel with results having high impact. I really enjoyed reading this well-written and well thought-out manuscript where the data and results support the main conclusions of the paper. This manuscript will definitely be of interest to readers of Nature Communications and the broader scientific community. I only have a few very minor comments.

1) In the Summary section, suggest modifying “internal metabolic fractionation” to “internal metabolism and isotopic fractionation.”

#16: Author’s response: We agree. Now changed.

2) Bottom of page 9 where the authors are inferring species trophic level from $\delta^{202}\text{Hg}$ values – I’m curious whether the authors investigated how well $\delta^{202}\text{Hg}$ correlates with $\delta^{15}\text{N}$ across species. $\delta^{15}\text{N}$ is an isotope that is very commonly used to estimate trophic level differences between species and this could be worthwhile to include in the supplementary materials section.

#17: Author’s response: This is a good suggestion, and we expect $\delta^{15}\text{N}$ to correlate well with $\delta^{202}\text{Hg}$ within sites as we see a very clear increase in $\delta^{202}\text{Hg}$ with trophic level in Ittoqqortoormiit, East Greenland (Fig. 3d). However, we do not have $\delta^{15}\text{N}$ data available for the sculpins and glaucous gulls from Ittoqqortoormiit measured for $\delta^{202}\text{Hg}$. This is certainly something we will consider for future work.

3) Top of page 10 when describing the foraging ecology of glaucous gulls – it’s also worthwhile to state that they are also scavengers.

#18: Author’s response: We agree and this is now added: “Glaucous gulls are top predators and scavengers, and feed on a variety of prey items, including marine invertebrates, fish, birds and bird eggs as well as e.g. polar bear left-over seal carcasses⁵³.”

4) Top of page 10 when describing the foraging ecology of polar bears – it’s also worthwhile to briefly state some of the other food items (i.e. bearded seals, narwhal, Atlantic walrus).

#19: Author's response: We agree and this is now added: *"Finally, polar bears are top predators and feed predominantly on ringed seals, hooded seals, and harp seals in East Greenland but occasionally also on other food items (e.g. bearded seals, narwhal, and Atlantic walrus)"*.

Reviewer #4 (Remarks to the Author):

The authors describe a study on total Hg concentrations and Hg isotopes in peat profiles and different key Arctic aquatic predators at different locations in Greenland. The major aims of this study are 1: to track Hg sources and pathways to Greenland 2: to show the development of total Hg and Hg isotopes in aquatic predator species in the past 40 years and how the development of the Hg content compares to that of anthropogenic atmospheric Hg emissions. 3: to show if there is internal metabolic fractionation of Hg isotopes in the different investigated aquatic species. 3:

The authors could show that there are distinct regional differences in total Hg and Hg isotopes ($\delta^{202}\text{Hg}$ signatures) between central western Greenland and northern-eastern Greenland which they suggest to be caused by different marine currents with different dissolved Hg concentration rather than by different atmospheric sources. Moreover they observed that Hg concentrations in Arctic aquatic predatory species have increased in the past 40 years opposite to the decrease of atmospheric Hg emissions and atmospheric Hg concentrations. From this they concluded that rather delayed mobilised legacy Hg than direct atmospheric Hg deposition is the major pathway of Hg to arctic aquatic predators and Greenland soils/peat (e.g. through seaspray aerosols).

This study is innovative and of importance and shows important findings on the distribution of Hg contamination in the aquatic environment in Greenland. To my knowledge, this is the first and most comprehensive data-set of Hg concentrations and Hg isotopes from Greenland biota and from Arctic biota in general including monitoring data of aquatic species of the past 40 years, which I think is crucial to understand the biogeochemical Hg cycling in the arctic.

For me, the merit of this study lies in the large and unique data set of Hg concentrations and isotopes in Arctic biota, which gives new insights into the large variability in Hg enrichment and Hg isotope fractionation due to different food sources and through internal metabolic processes and indicates that processes and pathways of Hg and Hg isotope fractionation are less clear than previously assumed. The discussion here appears to be sound, although the authors clearly point out that more research is needed here.

However, even if I think that it is a plausible assumption, the conclusion that the major pathway of Hg to the marine and the terrestrial Greenland foodchains is Hg evasion from

different ocean currents is not really supported by the data. The differences in dissolved Hg data which the authors have drawn from the literature may explain at least partly the differences observed in marine predatory species at different locations in Greenland, but cannot explain the differences in peat or landlocked Arctic char.

#20: Author's response:

We thank for the positive feedback and appreciate the comment. Based on the comment we find that we may not have explained properly how our data supports the conclusion that oceanic source Hg, either through direct uptake in the marine environment or through evasion/atmospheric re-deposition in the terrestrial coastal environment, is likely to be a major pathway to the marine and coastal/terrestrial food webs in Greenland (and why we consider the peat data important to include). We base this conclusion on the following:

- 1) We observe significant regional difference in Hg isotopes in Greenland with $\sim 0.5\text{‰}$ higher $\delta^{202}\text{Hg}$ in Central West Greenland compared to Northwest/Northeast/Central East Greenland (Fig. 1). This spatial trend contrasts with modelling data for atmospheric deposition showing roughly similar source contributions (i.e. $\delta^{202}\text{Hg}$) for these regions but is in line with the regions being dominated by different ocean currents (i.e. Central West Greenland influenced by Atlantic Ocean inflow via the Irminger Current, and the other sites dominated by Arctic Ocean water). Previously reported $\delta^{202}\text{Hg}$ values for the Atlantic and Arctic Ocean show higher $\delta^{202}\text{Hg}$ in the Atlantic Ocean in line with the higher $\delta^{202}\text{Hg}$ observed in Central West Greenland in this study influenced by Atlantic Ocean water. Importantly, we find the same regional $\sim 0.5\text{‰}$ difference in both the aquatic species (considered dominated by aquatic methyl-Hg uptake) and in peat (dominated by atmospheric Hg(0) uptake). This supports the conclusion that the difference in $\delta^{202}\text{Hg}$ is likely due to an overall source difference rather than regional differences in environmental transformation processes affecting the partitioning of Hg (although we cannot rule out the influence of that, which is now mentioned in the discussion). The spatial trend is consistent throughout the 1983-2024 period when ringed seals have been collected (Fig 4a) and on a long-term basis revealed by the peat data (Fig. 1b; Supplementary Fig 1b).
- 2) We find consistently lower total Hg in similar marine biota matrices in Central West Greenland influenced by Atlantic Ocean inflow compared to the other Greenland regions consistent with lower total Hg concentration reported for Atlantic Ocean water compared with Arctic Ocean water.

- 3) We observe increasing total Hg trends in most marine biota matrices in Greenland during the past 40 years in contrast to observed and modelled atmospheric deposition trends for the Arctic showing decreasing trends due to decreased global emissions. This discrepancy is an indication that ocean transported Hg may now be a dominant source. The average retention time of Hg in the ocean is much longer (>300 years) than in the atmosphere (approx. 0-5-1 year) and because of that, the marine species will show a much-delayed response to atmospheric variations. Due to the long retention time of Hg reported in the ocean, the ocean transported Hg will contain both recent and legacy Hg.

Consequently, we have updated and sharpened the discussion:

*“The consistent geographic difference in $\delta^{202}\text{Hg}$ signatures in peat profiles as well as in $\delta^{202}\text{Hg}$ and total Hg in shorthorn sculpin and ringed seal is surprising and considered to reflect long-term regional differences in Hg source inputs to Greenland. Overall, Central West Greenland displays ~ 0.5 to 0.6% higher $\delta^{202}\text{Hg}$ compared to similar matrices in Northwest, Northeast, and Central East Greenland, with consistent differences observed throughout the active layer in peat profiles (Fig. 1b) and through the last 40 years of sampling for ringed seal (Fig. 4a). Although the peat profiles are not dated, a similar peat profile (i.e. palsa consisting of alternating layers of segregated ice, peat, or mineral soil material) from Northeast Greenland is dated to $2,623 \pm 123$ BP at 50 cm depth²⁹, implying that the Hg in the upper c. 25 cm thick active layer of the peat should represent hundreds of years of deposition even though growth rates of palsas vary with time³⁰. However, using $\delta^{202}\text{Hg}$ as source tracers in peat profiles and biological tissue samples pose limitations. For peat, factors such as vegetation type and decomposition/leaching may influence $\delta^{202}\text{Hg}$ ³¹⁻³². Consequently, to minimize potential effects of vegetation type and decomposition on the $\delta^{202}\text{Hg}$ between sites, we chose two representative palsas, one from each region, both considered ombrotrophic and fully covered with a similar vegetation type dominated by sedges including *Carex* and *Eriophorum* species as well as a diverse moss flora. We observe the same spatial difference in $\delta^{202}\text{Hg}$ in the fresh litter layer and through the upper 25 cm of the active layer between the two regions (Fig. 1b; Supplementary Fig. 1b), indicating little/no effects from varying decomposition between the two sites. Thus, the consistently higher $\delta^{202}\text{Hg}$ signatures in Central West Greenland throughout the top 25 cm active layer compared to the Northeast Greenland profile is considered a result of persistent long-term regional differences in $\delta^{202}\text{Hg}$ inputs. For the biological tissue samples, $\delta^{202}\text{Hg}$ signatures are influenced not only by the Hg source input to the environment*

but also by all biogeochemical transformation processes (e.g. microbial methylation/demethylation)^{11,12} as well as prey species/trophic level³³⁻³⁴, and internal metabolism in the species^{12,16}. Here, we choose to compare $\delta^{202}\text{Hg}$ signatures in similar species (shorthorn sculpin and ringed seal) and tissue (muscle) across the three regions to minimize effects related to differences in prey species/trophic level and internal metabolism. The consistent ~0.5 to 0.6‰ higher $\delta^{202}\text{Hg}$ in Central West Greenland compared to the other Greenland regions for both peat (mainly influenced by atmospheric Hg(0) source uptake^{22,35}) and marine species (considered mainly influenced by marine methyl-Hg source uptake³⁶) suggests an overall difference in Hg source inputs between the regions. However, we cannot rule out that regional differences in environmental biochemical processes affecting the $\delta^{202}\text{Hg}$ can have additional minor effects.

The significant spatial difference in $\delta^{202}\text{Hg}$ for Greenland contrasts with recent model predictions for atmospheric Hg deposition in the Arctic showing regional differences in absolute deposition but relatively uniform source compositions, reflecting a relatively long lifetime of Hg in the atmosphere (~0.5-1 y)³⁷ and effective mixing of source inputs⁴. Atmospheric Hg transport to the Arctic follows a general pattern dominated by northern Eurasian sources in winter and mid-latitude Asian and North American sources in spring^{4,38}. The significantly higher $\delta^{202}\text{Hg}$ in all sample matrices at the Central West Greenland site, located only approximately 1,000 km from the Northwest Greenland site and both being exposed to the same overall atmospheric transport patterns, is therefore surprising. This suggests that direct atmospheric Hg deposition may not, as currently predicted⁵, be the dominant pathway for Hg pollution in marine and coastal areas in Greenland and perhaps also elsewhere in the Arctic. Inputs of Hg from rivers or glacial meltwater in Greenland are considered low³⁹⁻⁴¹ and therefore not a likely cause of the observed $\delta^{202}\text{Hg}$ variation. Instead, we propose that ocean currents may be the main Hg pathway explaining the significantly lower total Hg concentrations and higher $\delta^{202}\text{Hg}$ observed in marine and coastal areas in Central West Greenland than in Northwest, Northeast, and Central East Greenland. Central West Greenland is dominated by the relatively warm West Greenland Current, which is a mixed current comprising a branch of the Irminger Current from the North Atlantic Ocean and a branch of the East Greenland Current originating from the Arctic Ocean (Fig. 1a)²⁵. The other Greenland regions are all dominated by currents from the Arctic Ocean²⁶⁻²⁷. The total Hg concentrations measured in North Atlantic waters are lower than in Arctic waters^{42-43,54}, with North Atlantic-sourced water flowing into the Arctic having lower Hg (0.15 ± 0.07 ng/l) than Arctic Ocean surface waters (0.24 ± 0.12 ng/l)⁵.

Further, the few studies available on Hg stable isotopes in seawater report higher $\delta^{202}\text{Hg}$ in the North Atlantic waters (median, -0.45‰)¹⁹ than in Arctic Ocean waters (-2.85 to -1.10‰)⁴⁴. Consequently, we hypothesize that ocean currents are the main driver of Hg uptake in marine and coastal environments in Greenland, and other Arctic regions, either through direct uptake of Hg from ocean waters or through Hg evasion from the ocean surface followed by atmospheric redeposition and uptake, explaining the significantly lower total Hg in marine species and higher $\delta^{202}\text{Hg}$ in both marine species and peat profiles in Central West Greenland influenced by Atlantic Ocean water. It is interesting to note that the same regional difference in $\delta^{202}\text{Hg}$ as recorded for marine species occurs in peat profiles in Central West Greenland and Northeast Greenland, situated 800 m and 2 km inland, respectively. This may be due to a greater influence of atmospheric Hg deposition from sea spray and/or evasion/redeposition of Hg from regional ocean surface waters compared to long-distance atmospheric sources. The influence of sea spray on the vegetation in coastal zones is well known⁴⁵, and laboratory studies have shown that sea salt aerosols have a high capacity for uptake of reactive gaseous Hg⁴⁶. A recent study shows that oceanic evasion is likely the dominating source and pathway to atmospheric Hg(0) in the Arctic during summertime (~80%) driving the transport to Arctic terrestrial ecosystems⁴⁷. Likewise, evasion from the ocean surface waters is known to constitute a major part (~43%) of the global Hg flux budget⁸.”

Frankly, I do not understand why the authors have included the Hg data from the peat/palsa profiles in this study. In the introduction the authors do not clearly explain why they believe that the approach of using Hg data of peat for Hg source tracing (marine versus atmosphere) is reasonable. First, the investigated peat deposits are quite different and as it seems, that they are not ombrothrophic (all elements atmospherically derived), which makes a solely marine Hg source appear doubtful. Moreover, instead of using Hg accumulation rates to estimate differences in Hg flux or Hg accumulation the authors base their interpretation only on peat Hg concentrations. Many studies have shown that Hg concentrations in peat alone cannot be used to derive differences in atmospheric fluxes as they are strongly influenced by peat decomposition, plant species or hydrology etc.. Similar, the interpretation of $\delta^{202}\text{Hg}$ in peat as a source proxy is doubtful as long as peat decomposition and related Hg isotope fractionation has not been considered. The authors actually mention this limitations. Generally, $\delta^{202}\text{Hg}$ as a source proxy appears to be doubtful regarding that the authors show intense internal metabolic $\delta^{202}\text{Hg}$ fractionation in their biota.

I suggest that the authors focus on the Hg data in biota as this data set is unique and

important e.g. the finding that Hg concentrations in hair and liver of polar bears show an increasing trend with time while Hg concentration in muscle decrease, is remarkable. I also believe that the authors should attenuate their conclusions regarding the role of different Hg currents as different Hg sources because the data supporting this conclusion is rather weak. Discussing the inverse trend of Hg accumulation in biota and the atmosphere is the most important finding here. In this context, I further propose to remove the peat data from the manuscript as it does not really support the discussion on Hg sources or transport pathways.

#21: Author's response: We value the comment and agree that it is unusual to combine aquatic biota data and peat data in the same paper. We also agree that we need to better explain the reasoning behind this approach. However, after careful consideration, and based on the reasons elaborated below, we suggest keeping the most important peat data in the paper (i.e. the Hg isotope data from the litter/active layer representing the most recent deposition). The peat Hg isotope data ($\delta^{202}\text{Hg}$) in combination with the sculpin and ringed seals Hg isotope data show a similar spatial trend across different Hg pools (representing predominately Hg uptake atmospheric Hg (0) and aquatic methyl-Hg, respectively), providing evidence that the difference in $\delta^{202}\text{Hg}$ is likely to be due to overall differences in source input and most likely related to the ocean currents (see #20 pt. 1 above). Further, we use $\Delta^{200}\text{Hg}$ in the peat to describe long-term pathways of atmospheric Hg deposition as Hg(0) or Hg(II), which we consider a valuable addition to the paper. In contrast to $\delta^{202}\text{Hg}$, the $\Delta^{200}\text{Hg}$ signature in peat is not considered influenced by decomposition etc. and $\Delta^{200}\text{Hg}$ signature in peat is widely used as a conservative tracer for atmospheric deposition pathways (e.g. Enrico et al. Environ. Sci. Technol. 50, 2405–2412 (2016); Li, C. et al. Geochim. Cosmochim. Acta 341, 62-74 (2023)).

For the peat total Hg concentrations, we agree that since we do not have a dating of the individual peat layers, the total Hg cannot be used to conclude on absolute deposition rates and may therefore only be important as a reference in the Supplementary Information section. Due to these changes, Fig. 1, 2 and 3 and Supplementary Table 1 have been revised accordingly along with changes to the text throughout the manuscript (we refer to the track-changes version of the manuscript for these changes).

Further, we agree that indeed all the sample matrixes (in this case peat, sculpin muscle and ringed seal muscle) have limitations with regards to using $\delta^{202}\text{Hg}$ as a Hg source proxy, and we have now elaborated more on that in the manuscript. All biogeochemical

processes in the Hg cycle will result in changes in $\delta^{202}\text{Hg}$ (e.g. Blum, J.D. et al. *Annu. Rev. Earth Planet. Sci.* 42, 249-269 (2014)). Moreover, factors such as vegetation type and decomposition (for peat) and diet and internal metabolic fractionation (for biota) may affect $\delta^{202}\text{Hg}$. However, by comparing the same vegetation type across regions and using similar species and tissues, we can largely normalise for these influencing factors, although we cannot rule out the regional differences in environmental Hg transformation processes may also affect the $\delta^{202}\text{Hg}$. Both peat sites are considered ombrotrophic and no indication of decomposition effects on the $\delta^{202}\text{Hg}$ was observed as we see the same difference between regions throughout the upper profile (now added to the discussion). Consequently, we have elaborated on the limitations for using the $\delta^{202}\text{Hg}$ as a source tracer in the all sample matrices in the discussion and have attenuated the conclusions regarding the regional differences in $\delta^{202}\text{Hg}$ as a source tracer for the ocean currents, pointing out the regional differences in environmental Hg transformation processes may also have an influence (although several trends in the data suggest ocean currents as the driving source, see #20 pt. 1-3 above). Also, we mention that the increase in $\delta^{202}\text{Hg}$ during the last 40 years in ringed seals and polar bears may also be influenced by changes in environmental Hg transformation processes in addition to anthropogenic sources. We refer to the track-changes version of the manuscript for these changes.

Please mention number of samples in figure captions, respectively.

#22: Author's response: Good point – thanks. The numbers of samples are now inserted in figure captions or figure legends.

Author's responses to reviewer comments (in green below)

REVIEWER COMMENTS TO REVISION 1

Reviewer #1 (Remarks to the Author):

Summary: We have re-reviewed NCOMMS-24-75705 "Stable isotopes unveil ocean transport of legacy mercury into Arctic food webs" including the response to reviewers file. Overall, we find the authors responses to be thorough and detailed, addressing most of our original concerns and corrected areas where we believed were lacking with sufficient details. The inclusion of more robust evidence and clarity on connections between the peat data and biological data is especially well written. We have one final point around the differences in temporal trends between polar bears and seals for consideration, detailed below.

Recommendation: Overall, we encourage publication of the manuscript.

Detailed points:

1. Our comments were addressed, largely through text modification and clarifications. This notably includes: (1) incorporating recently published literature (e.g., Huang et al. 2025); (2) a more detailed (i.e., well cited) paragraph around animal migration impacts near line 521; (3) more robust statistical details (p-value reporting and method information); (4)

Author's response: We are very pleased to hear that.

2. For our (Review 1) point 15: In some ways, the authors' answer contradicts itself: differences in trends are due to changes in polar bear diets, from rings seals to hooded seals, but what are the diet differences in the seals themselves. We understand that you do not have data on that, but there are past studies done that could build a much more robust discussion around this point, in lieu of direct data. E.g., Schiøtt, S., Holland, D.M., Holland, D. et al. The diet and diving behaviour of the ringed seal (*Pusa hispida*) in Kangia (Ilulissat Icefjord), Greenland. *Polar Biol* 47, 1409–1422 (2024). <https://doi.org/10.1007/s00300-024-03310-4>.

Author's response: Diet differences between the seals species (as well as potential species-specific internal metabolism) are indeed reflected in the differences in Hg isotope signatures in the seal species tissues shown in Supplemental Fig. 3b and is the basis for diet adjustments of the polar bear Hg isotope time trends shown in Fig. 4c and d. We see that this was perhaps not adequately explained/elaborated in the previous version of the manuscript. Consequently, we have adjusted the paragraph

and added a new sentence and reference on documented stable isotope/diet differences between Greenland ringed seals, harp seals and hooded seals (ref. 60).

'Part of the increase in $\delta^{202}\text{Hg}$ in the polar bears in Central East Greenland can be ascribed to a documented shift in diet during the period with consumption of significant less ringed seals having lower $\delta^{202}\text{Hg}$ (median in muscle tissue, 0.28; $n = 82$) towards more hooded seals having higher $\delta^{202}\text{Hg}$ (median in muscle tissue, 1.19; $n = 27$) (Supplementary Fig. 3)^{55,59}. These differences in $\delta^{202}\text{Hg}$ between seal species are considered to mainly reflect differences in prey species⁶⁰.'

We also thank for the reference to the interesting paper from the Ilulissat Icefjord area. However, we are a bit reluctant to include the paper as reference in this study as the Ilulissat Icefjord represents an area with very special environmental characteristics, and with a unique and adapted ringed seal and harp seal population. Consequently, the conclusions drawn may not apply to other Greenland sites and we have therefore not included it. We hope the discussion on this topic is now sufficiently detailed, and we consider that a longer discussion on diet differences between seal species is beyond the scope of this study (which is also the reason why the Hg isotope data on the different seal species is placed in the Supplementary Information section). In our view, the main discussion should be focused on the changes in diet over time for the East Greenland polar bears (and ringed seals) and the differences in Hg isotopes between prey items to interpret/adjust the Hg isotope time-trend series for the influence of diet changes. We can do these diet adjustments for the East Greenland polar bears, but we do not currently have the data needed to do it for the ringed seals (as also discussed).

a. More broadly, the authors also mention 3 sources of variability in their answer: sources, biogeochemistry, and diet. What about species-specific metabolism? That has been important in fish recently (as well as other groups like birds), with some taxonomic groups being identified as able to de-methylate MeHg or excrete high amounts of MeHg, relative to other species. E.g., see the recent paper: Tepper, K., King, J., Manuneechi Cholan, P. et al. Methylmercury demethylation and volatilization by animals expressing microbial enzymes. Nat Commun 16, 1117 (2025). <https://doi.org/10.1038/s41467-025-56145-w>.

Author's response: Species-specific metabolism and internal Hg isotope fractionation is indeed an important aspect of the variability in Hg isotopes observed in the paper as presented and discussed in the section 'Environmental processes and trophic

transfer', especially for the mammals (ringed seals and polar bears). Species-specific metabolism leading the internal Hg isotope fractionation in tissues is shown in Fig. 3c and the resulting difference in Hg isotope signatures with increasing $\delta^{202}\text{Hg}$ shown in Fig 3d. However, species-specific metabolism is not considered a reason for the spatial and temporal trends observed in our study as we compare similar species and tissues to normalise for this effect. Finally, thanks for the interesting reference. However, for the scope of our study with focus on Hg isotopes, and considering that we already have 71 references, we consider the Li et al. (2022) review paper titled 'Internal dynamics and metabolism of mercury in biota: A review of insights from mercury stable isotopes' (ref. 16) more appropriate and sufficient to cover the topic.

3. The authors responses to review 4's thorough comments are details and sufficient. While we do not disagree that the peat data made the original paper somewhat complex, we agree with the authors' holistic approach. Furthermore, we do not agree that the peat data is not inherently useful. While it may not serve as a direct measure of deposited Hg due to variations in decomposition rates and other factors, it remains a valuable data point when considered alongside biotic data. Given the distance between sites (1000 km shortest) there may be differences in biogeochemical properties in the area but that also occurring with similar changes in biota (internal metabolism) seem to be slim. Therefore, I believe the inferences drawn are reasonable and well-supported.

Author's response: We are very pleased to hear that.

4. The added detail from Reviewer 4's comments seem to be redundant at times. More specifically, they added a lot of sentences that end with "...and that's why ocean currents seem to be the Hg source" The authors might consider compiling all of this evidence more concisely. However, we consider this a stylistic comment and defer to the editor on its applicability.

Author's response: We appreciate the comment and have read through the manuscript carefully again and made some minor changes. However, since this is considered a stylistic comment, we will leave it to the editor to decide if more adjustment is required.

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3 (Remarks to the Author):

The authors have done a nice job addressing and incorporating reviewer comments. I recommend acceptance.

Reviewer #4 (Remarks to the Author):

The authors have generally improved the clarity of the manuscript and have addressed most of my concerns. However, they decided to keep the peat data in the manuscript although they now discuss the influence of peat decomposition etc. in more depth.

I still believe that the peat data is weak. The authors used only one record for each side of Greenland, respectively, and it remains doubtful that such a small data is representative.

The variability of Hg uptake by peatland plants, and especially related Hg isotope fractionation, is not well understood and most existing data is from moss and not from vascular plants. Moreover, I don't think that the peat data (n=1 only) does add much valuable and robust information to the overall message of the study and I still think it should be removed. However, it seems that the information received from the peat data is not essential for the conclusion drawn why I think that it might be possible to publish the ms in its present form.

Author's response: We agree that the peat profiles only represent two sites, one from Central West Greenland and one from Northeast Greenland and therefore represent a limited spatial coverage. However, the sites were carefully chosen to represent comparable palsas fully covered with a similar vegetation type dominated by sedges including *Carex* and *Eriophorum* species as well as a diverse moss flora.

Consequently, we expect that any difference of Hg isotope fractionation related to vegetation type is minimal. We find it important here that we observe a similar difference in $\delta^{202}\text{Hg}$ between the two peat profiles ($\sim 0.5\text{--}0.6\text{‰}$) as in the aquatic species (muscle of sculpins and ringed seals) from the same regions (Central West Greenland vs. Northwest/Northeast/Central East Greenland, respectively). Hg uptake in vegetation is known to be dominated by $\text{Hg}(0)$, whereas Hg uptake in aquatic species is dominated by methyl-Hg. We find the peat data important to include in the paper since the data adds evidence and support overall differences in Hg sources between these regions in Greenland (i.e. and not just differences in biogeochemical processes affecting Hg isotope fractionation in methyl-Hg as also pointed out by

Reviewer #1). Furthermore, we observe the same consistent difference in $\delta^{202}\text{Hg}$ in the peat cores between the two regions throughout the upper 25 cm unfrozen active layer, from the litter layer to the permafrost table, indicating consistent long-term differences and making the conclusion more robust.