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Exploring Drivers of Historic Mercury Trends in Beluga Whales Using an Ecosystem Modeling Approach

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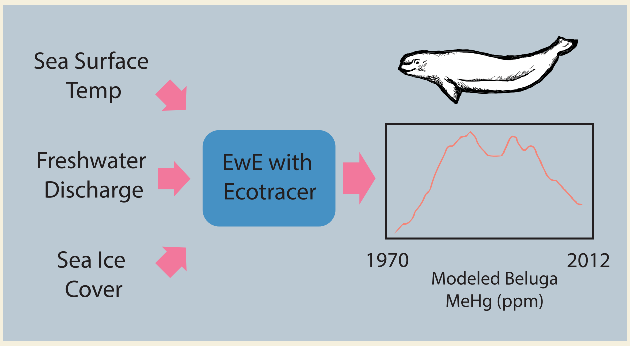

ABSTRACT: While mercury occurs naturally in the environment, human activity has significantly disturbed its biogeochemical cycle. Inorganic mercury entering aquatic systems can be transformed into methylmercury, a strong neurotoxicant that builds up in organisms and affects ecosystem and public health. In the Arctic, top predators such as beluga whales, an ecologically and culturally significant species for many Inuit communities, can contain high concentrations of methylmercury. Historical mercury concentrations in beluga in the western Canadian Arctic's Beaufort Sea cannot be explained by mercury emission trends alone; in addition, they could potentially be driven by climate change impacts, such as rising temperatures and sea ice melt. These changes can affect mercury bioaccumulation through different pathways, including ecological and mercury transport processes. In this study, we explore key drivers of mercury bioaccumulation in the Beaufort Sea beluga population using Ecopath with Ecosim, an ecosystem modeling approach, and scenarios of environmental change informed by Western Science and Inuvialuit Knowledge. Comparing the effect of historical sea ice cover, sea surface temperature, and freshwater discharge time series, modeling suggests that the timing of historical increases and decreases in beluga methylmercury concentrations can be better explained by the resulting changes to ecosystem productivity rather than by those to mercury inputs and that all three environmental drivers could partially explain the decrease in mercury concentrations in beluga after the mid-1990s. This work highlights the value of multiple knowledge systems and exploratory modeling methods in understanding environmental change and contaminant cycling. Future work building on this research could inform climate change adaptation efforts and inform management decisions in the region.

KEYWORDS: *mercury, bioaccumulation, Arctic, beluga, ecosystem modeling, climate change, Beaufort Sea*

1. INTRODUCTION

In the Arctic, methylmercury (MeHg) concentrations in recent decades have changed significantly in top predators, including beluga whales (*Delphinapterus leucas*).[1,2] These changes have implications for the ecosystem and Inuit communities who rely on such species for nutritional, cultural, and economic well-being.[3,4] In the Canadian Beaufort Sea, average mercury levels in older and larger whales of the Eastern Beaufort Sea (EBS) beluga population, tracked in muscle and liver tissues, significantly increased in the 1990s but declined in the 2000s.[5] While changes in mercury emissions could contribute to observed beluga MeHg concentrations, observed increases in beluga mercury concentrations during the 1970s and 1980s differ from most other long-term mercury trends in biota and mercury emission trends.[6] Further, Loseto et al.[5] examined mercury emissions to the atmosphere (as presented by Muntean et al.[7]) in relation to beluga mercury and found that they could not alone explain declining concentrations in the 2000s, when mercury emissions were increasing.[8] Mercury trends in beluga may, however, be driven by the impacts of climate change, including rising temperatures, higher freshwater flow from the Mackenzie River, increasing coastal erosion, accelerating sea ice melt and permafrost thaw, and increasing storms.[9−13] To add further complexity, the EBS beluga stock, numbering around 40,000 individuals, is migratory, spending May to late June in the Canadian Beaufort Sea and wintering in the Bering Sea.[14] As a result, climate

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impacts on the population are not limited to the Beaufort Sea, although that is the focus of this study.

Environmental change in the Beaufort Sea has been monitored by researchers since the latter half of the 20th century[15,16] and by Inuvialuit (Inuit of Canada's western Arctic) communities for millennia.[17] Rapid changes in the Arctic have been reported, including a shifting sea ice environment, increasing erosion and permafrost thaw, warming sea surface temperatures, and the impact of such changes on hunting and access to traditional foods.[18,19] Indeed, from 1990 to 2013, parts of the Beaufort Sea experienced an increase in mean surface air temperature by roughly 1 °C per decade.[20] From 1979 to 2012, the Beaufort Sea also experienced significant negative trends in sea ice concentration between June and October, and in 2012, it became ice free for the first time in the observational record.[21] In the Canadian Subarctic, changes have also been reported: For instance, from 1964 to 1984 in the Mackenzie Highway region, the southern limit of the discontinuous permafrost zone, where permafrost occurs over 50 to 90% of the land area, migrated northward by about 120 km, which coincided with a general warming trend.[22] Many of these same changes can have impacts on the biotic aspects of the ecosystem, such as microbial productivity, that can then affect MeHg bioaccumulation in diverging ways.[23] For instance, Stern et al.[24] detail that climate-driven changes such as enhanced nutrient inputs (from rivers, for example) and higher temperatures can promote microbial activity and thus increase methylation rates; however, other changes, such as decreasing sea ice cover, can increase demethylation rates via photodegradation.

While many effects of climate change on MeHg in the food web are still unclear and highly uncertain, ecosystem modeling can be used to explore hypotheses about how different environmental changes might impact bioaccumulation in the real world.[25−27] These studies have highlighted that environmental change-mediated shifts in mercury biogeochemical cycles and ecosystem structure can have significant impacts on biotic MeHg concentrations and that interpreting changes in biotic mercury therefore requires attention to both anthropogenic and environmental drivers.[11] An ecosystem model that can be used for this kind of investigation is Ecopath with Ecosim (EwE), which layers temporal dynamics (Ecosim) on top of a mass-balanced model (Ecopath).[28] Since Ecopath's original inception in the 1980s[29] and its subsequent development in the 1990s,[30,31] other modules have been added, such as Ecotracer, a contaminant module that allows the user to assess how contaminants like polychlorinated biphenyls and MeHg flow through the food web.[27,32−34] In the Canadian Beaufort Sea, EwE has been used to better understand ecosystem structure and function[35] and climate change effects on various species groups.[36] These past applications of EwE for the Beaufort Sea suggest that changes to the ecosystem were driven more by reductions in sea ice and increases in sea surface temperature (SST) compared to the harvest mortality impact[35] and that ecosystem stability (indicated by the trophic structure) has remained relatively stable over time.[36] Li et al.[34] developed an Ecotracer model to simulate MeHg bioaccumulation in the Canadian Beaufort Sea Shelf, which was able to capture real-world MeHg biomagnification patterns in the food web.

This study examines the effect of several historical environmental change scenarios on MeHg trends over time in the Beaufort Sea Shelf ecosystem, focusing on EBS beluga (hereafter referred to as beluga). It aims to advance understanding of how environmental change has affected mercury cycling in the food web through both ecological and mercury transport pathways using exploratory modeling with EwE. First, we review Western Science and Inuvialuit Knowledge literature to select three environmental drivers for further study (freshwater discharge, sea ice cover, and SST) and input these into EwE using time series data from 1970 to 2012 to force primary production and MeHg inputs into the system. We then use segmented regression to examine how modeled beluga MeHg trends compare to one another and to observed data.

2. METHODS

2.1. Study Area

The model area is the Canadian Beaufort Sea Shelf (BSS) (Figure 1). It is the largest North American shelf in the Arctic, though most of the BSS is shallower than 200 m.[37] The Mackenzie River, the longest river system in Canada with the second largest drainage basin of any river in North America (and the 13th largest in the world), flows into the BSS.[38] Importantly, the BSS is also within the homeland of the Inuvialuit and lies in an area of the Northwest Territories recognized today as the Inuvialuit Settlement Region (ISR), designated with the modern land claim, the Inuvialuit Final Agreement.[39] While the BSS model area is in the Northwest Territories, the ISR itself spans both the Northwest Territories and the North Slope of the Yukon Territory.[40] There are six Inuvialuit communities in the region: Aklavik, Inuvik, Tuktoyaktuk, Paulatuk, Sachs Harbour, and Ulukhaktok, which lie either directly on the BSS coastal area or harvest from the region.

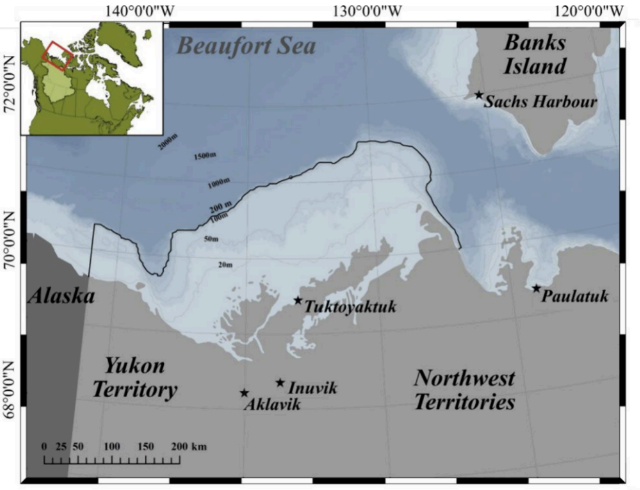

Figure 1. BSS model area, outlined in black. The stars indicate the location of the Inuvialuit communities in the region. Note that an additional community, Ulukhaktok, does not lie directly on the BSS but does harvest from the region. Reprinted or adapted with permission under a Creative Commons BY 4.0 from Hoover et al.[35] Copyright 2021 The Authors.

The marine ecosystem is home to several migratory mammals, including beluga, as well as various fish species, many of which are diadromous.[37] More detail on EBS beluga biology is included in SI Section 1. Sea ice plays an important role in the ecosystem, governing the inflow of river water into the BSS, phytoplankton blooms, and predation.[41] Like many other regions in the Arctic, the BSS has experienced a multitude of climate-related changes, including rising temperatures, decreasing sea ice extent, more erratic storms and winds, and increased coastal erosion and permafrost thaw.[42]

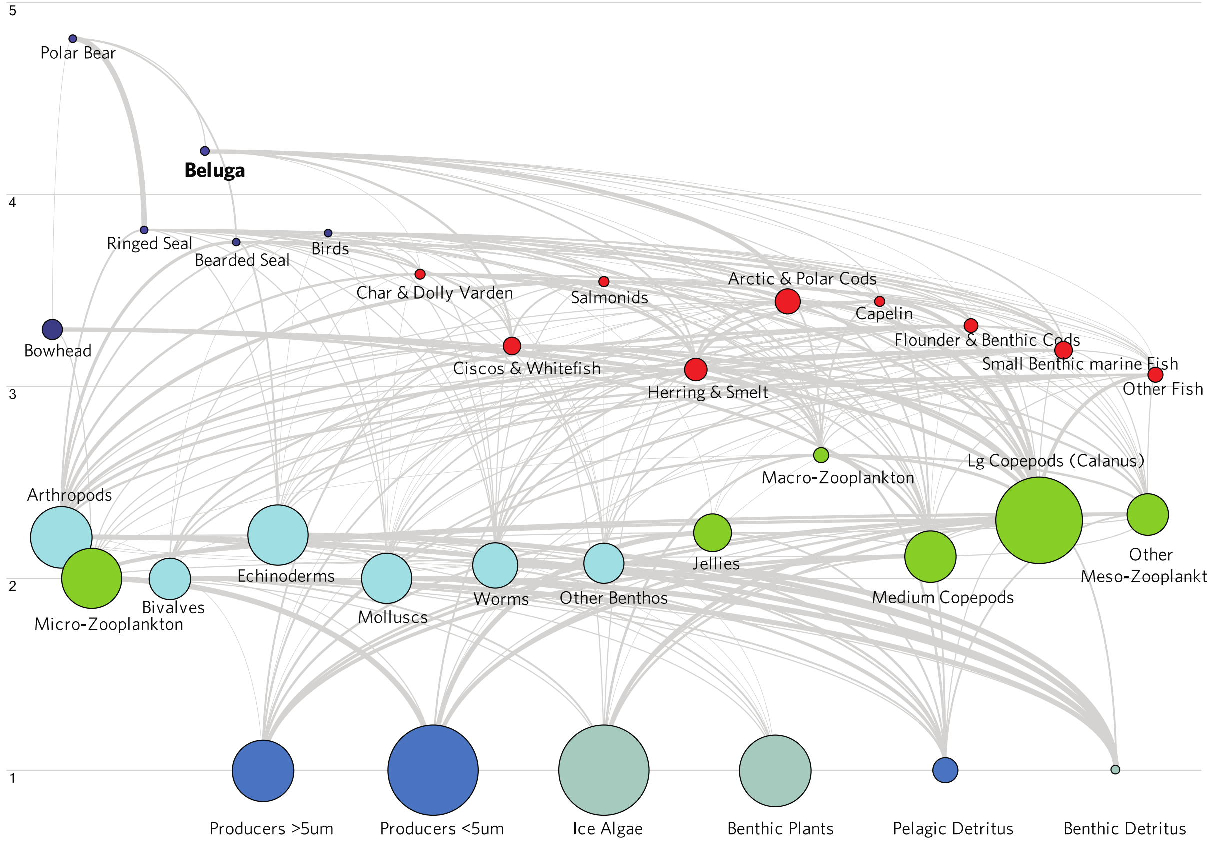

Figure 2. Graphical representation of the functional groups in the BSS EwE model. The size of circles represents biomass of each group, lines between circles represent flows of biomass, and the vertical axis represents the trophic level. The position on the horizontal axis is for visibility only.

Mercury data in the region are available for seawater,[43] producers,[44] zooplankton,[45] benthic species,[46] many fish species,[47] and ringed seals,[48] but by far, the most extensive data pertain to beluga.[5] Mercury levels have been monitored in beluga for roughly four decades via a collaborative community-based monitoring program led by the Fisheries Joint Management Committee (FJMC), Fisheries and Oceans Canada (DFO), and local Hunters and Trappers Committees. The FJMC is a regional comanagement body with members appointed by both the Inuvialuit Game Council (IGC), a body representing Inuvialuit interests regarding wildlife and wildlife habitat, and DFO, a department of the Canadian federal government.[39] We engaged all boards with this paper, described in Section 2.3.

2.2. EwE and Ecotracer Model Description

EwE is a widely used trophodynamic ecosystem model that can simulate mass and energy flows in the food web. The model uses information on biomass, production, consumption, and mortality rate of different species functional groups to represent a mass-balanced ecosystem structure, in which the biomass flows into a group via reproduction and immigration and flows out of a group via mortality (harvest and natural) and emigration.[28]

EwE has previously been used to represent the BSS ecosystem in the form of a model with a range of different functional groups in the ecosystem, including detritus, primary producers, zooplankton, fish, marine mammals, and birds,[37] with 33 functional groups in all. While some functional groups include multiple species, others comprise a single species. Figure 2 shows a graphical representation of groups in the BSS EwE model. Hoover et al.[37] provided more information on the types of organisms that comprise each functional group.

The Ecopath module within EwE represents a mass-balanced food web at a single point in time, whereas the Ecosim module adds time dynamics to the simulation.[28] Other modules can also be used on top of the base Ecopath food web representation to add specific layers of capabilities. Here, we use Ecotracer for modeling contaminant bioaccumulation, using the Ecotracer module for the BSS developed by Li et al.,[34] which itself built on the EwE model previously developed for the BSS.[37] In Ecotracer, the amount of MeHg in each functional group is based on predator−prey interactions, direct uptake of MeHg from seawater, internal metabolism, internal decay, and harvest (mortality). At a steady state, intake of MeHg (eq 1) must be equal to loss (eq 2).[49]

$$\text{intake } \alpha_i = \mu_i B_i C_o + \mathrm{AE}_i \sum_{j=\text{prey}} Q_{ji} C_j \tag{1}$$

where C_o is the seawater MeHg concentration (t/km^2); for group i, B_i is the biomass (t); μ_i is the direct absorption rate of MeHg from water (km^2/(t year)); AE_i is the assimilation efficiency (unitless). Q_{ji} is the consumption rate (t/year) of prey j by predator i, and C_j is the MeHg concentration in prey j (t of MeHg/t of biomass).

$$\text{loss } \beta_i = \left(\sum_{k=\text{predator}} \frac{Q_{ik}}{B_i} + H_i + \mathrm{MO}_i + E_i \right) B_i C_i \tag{2}$$

where Q_{ik} is the rate of consumption (t/year) of group i due to predation by k and $\frac{Q_{ik}}{B_i}$ is the fraction of group i consumed by predator k (unitless), H_i is the mortality rate due to harvests (/year), MO_i is the natural mortality rate (/year), and E_i is the elimination rate (/year).

In this study, we apply a version of the EwE model that was previously parametrized for the BSS.[37] To parametrize Ecotracer, we used a version of the EwE BSS model developed by Sora et al.,[36] representing the average food web structure and dynamics between 2008 and 2012. This version was created by using a balanced model for 1970 (presented by Hoover et al.[37]) and running it forward in time based on a fitted Ecosim model. The Ecotracer parametrization process was fully documented by Li et al.[34]

2.3. Development of Model Scenarios

Selection and development of historical environmental change scenarios were informed by a review of Western Science[1] (WS) and Inuvialuit Knowledge[2] (IK) literature. To confirm appropriate sources of previously documented IK for use in this research, we engaged with the IGC, FJMC, the Hunters and Trappers Committees (HTCs) of the ISR, and the Joint Secretariat Traditional and Local Knowledge staff. We focused on using previously documented IK sources to reduce research fatigue and engagement burdens, as knowledge holders in the North experience numerous and sometimes duplicative demands for their expertise.[52] In March 2022, this paper's lead author (E.J.G.) attended a meeting of the IGC, who recommended that the research team verify that the HTCs from each of the six communities in the ISR were comfortable with the sources being drawn upon. E.J.G. then contacted each HTC and received feedback and approval on selected sources from Aklavik HTC via Zoom meeting in June 2022, from Paulatuk HTC via phone call in July 2022, from Olokhaktomiut HTC via phone call in July 2022, from Sachs Harbour HTC via email in August 2022, and from Tuktoyaktuk HTC via Zoom meeting in August 2022. E.J.G. was not able to receive feedback from Inuvik HTC; thus, our review of sources from that community is limited. Please note that when we use the word "community" in this study, we are referring simply to community representatives with whom we engaged, rather than the entire community.

Key Inuvialuit Knowledge published sources included those from the communities of Aklavik, Inuvik, Paulatuk, Sachs Harbour, Tuktoyaktuk, and Ulukhaktok (e.g., refs 19 and 53–71). Meanwhile, key WS sources included those from various scientific disciplines, including marine ecology, mercury biogeochemistry, atmospheric sciences, and polar ecology (e.g., refs 9, 24, 27, 41, 72–83). A table summarizing the key environmental changes drawn from the literature review is presented in Table S1 in the SI.

The literature review highlighted sea ice cover, SST, and freshwater discharge as key historical changes with linkages to temporal mercury variability in the region. For instance, in the case of sea ice, modeling suggests that the ice volume could decline in the Arctic Ocean at a rate of 11% per decade over the period from 1979 to 2069.[85] Meanwhile, Inuvialuit communities have reported observations of later ice freeze-up and earlier breakup, thinner ice, and more open water in the winter.[19] Sea ice melt is a central element of Arctic ecosystems and is important for mercury cycling in several ways: For instance, it affects net deposition and evasion of mercury,[72,86] promotes primary productivity,[87] and drives photodemethylation.[88]

SST was selected as another driver of interest because it, along with air temperature, has been increasing over the past few decades in the region.[54] In the Beaufort Sea, warmer SSTs in the summer have led to delayed freeze-up in the fall, along with large surface air temperature anomalies.[82] Inuvialuit communities have linked observed changes in fish and primary producer populations to increases in water temperature.[89] In addition, higher temperatures stimulate microbial activity, thus increasing mercury methylation.[90]

Finally, freshwater discharge was selected as a driver given the important role of rivers as a source of mercury to the Arctic Ocean.[13,91] The Mackenzie River, Canada's longest and largest river system, empties into the Beaufort Sea.[38] The Mackenzie is also the largest source of sediment and fourth largest source of freshwater of any Arctic river to the Arctic Ocean,[92] and its flow has been variable over the past four decades due to climate variation.[93] River discharge from the Mackenzie River transports enormous amounts of carbon, mercury, nutrients, and heat to the BSS, affecting mercury concentrations, biota, and sea ice cover.[41] This has also been seen in other regions. For instance, nutrient inputs can increase phytoplankton productivity, in turn affecting mercury: A study comparing Atlantic Ocean and Mediterranean Sea marine sites found that lower primary producer biomass in the Mediterranean increased mercury biomagnification, while higher primary producer biomass in the Atlantic decreased mercury biomagnification, a phenomenon known as biodilution.[94] Similarly, a study on MeHg in black-legged kittawakes in Svalbard, Norway found that chlorophyll a (an indicator of primary productivity) was tightly related to MeHg.[95]

As sea ice cover, SST, and freshwater discharge were highlighted by the literature review and implementable as scenarios in EwE, they were selected as the drivers of interest for this study. We then designed simplified model scenarios to investigate the influence of the three selected environmental drivers of food web MeHg. The primary aim of these exploratory scenarios was to examine biotic MeHg trends in beluga, including the direction and timing of any changes, rather than the absolute magnitude of the MeHg concentrations themselves. Other groups were also examined to help understand the trends seen in beluga.

2.4. Implementing Scenarios in EwE

We implemented historical environmental change scenarios from 1970 to 2012, beginning with a version of the BSS EwE food web structure for the initial year (1970) and simulating changes forward. We use this time period to compare modeled MeHg concentrations to observed data presented by Loseto et al.,[5] in which beluga MeHg concentrations were measured until 2012. Ecotracer parameters for the BSS model (uptake rate, elimination rate, and assimilation efficiency) are described by Li et al.[34] We adjusted the initial MeHg concentration conditions in Ecotracer to reflect 1970 conditions, the starting point of the simulation. We set the initial concentrations for each group according to the concentration output from a parametrized model run at a steady state with the estimated 1970 environmental (i.e., water) concentration. The 1970 seawater concentration was estimated following a methodology used by Schartup et al.[26] and used historical MeHg concentrations in the subsurface Arctic Ocean.[96] Briefly, we applied the percent change in historical seawater MeHg concentration in 2010 compared to 1970[96] to back-calculate the 1970 seawater concentration based on the seawater MeHg level in 2010 from Li et al.[34] The base case Ecotracer parametrization and inputs are summarized in Tables S2 and S3 in the SI.

2.4.1. Scenarios. We implemented a range of scenarios representing individual environmental drivers and change pathways and combinations thereof. In this study, we focus on two pathways through which these environmental change drivers could influence beluga MeHg:[1] methylmercury inputs into the ecosystem and[2] primary production. Other potential pathways for change related to these drivers, such as dietary shifts,[97] are explored in Section 3.3. The scenarios are outlined below, and the assumptions and associated uncertainties are outlined in greater detail in Table S4 in the SI.

The three time series used as the basis for the environmental change scenarios are sea ice cover, freshwater discharge, and SST. All time series are on a monthly scale, from January 1970 to December 2012. Information on the time series data and their sources is provided in brief below, with more detail in SI Section 4.

Sea ice cover and SST were both extracted from the HadISST1 global data set, which combines in situ and satellite-derived observations.[98,99] Time series data for the 1° × 1° cells contained in the BSS model area were extracted and averaged across the model area.[37] Values for sea ice are presented as monthly average values of sea ice concentration (0–1) at a 1° latitude–longitude (1° × 1° cells) resolution, while values for SST are presented as monthly average values of SST (°C) at a 1° latitude–longitude (1° × 1° cells) resolution. We selected this data set because it provides sufficient spatial resolution to capture variations in both sea ice extent and SST trends between Arctic sub-basins (described by Steiner et al.[100]) and used a consistent interpolation methodology across the multidecadal study period. However, changes in data sources over this time period (e.g., incorporation of satellite data after 1982) and more limited data in polar regions, particularly presatellite, are noted limitations.[101] More information on how both data sets were created can be found in Rayner et al.'s work[98] and in SI Section 4. The two time series have been used in previous BSS EwE studies: by Suprenand et al.[104] to fit

Table 1. Summary of Environmental Change Scenarios

environmental change	EwE scenario type	implementation
increasing freshwater discharge	primary production	Pelagic producer biomass is linked to carbon and nutrient inputs, which are linked to freshwater discharge,[107] with upper and lower bounds.
	mercury inputs	Freshwater inputs are linked to increased MeHg inputs due to increased inorganic Hg and increased dissolved organic carbon, increasing methylation.[108] Terrestrial permafrost thaw could also contribute to increased mercury sources from freshwater inputs.[72]
increasing sea ice melt	primary production	Pelagic producer biomass is linked to light availability, which is in turn linked to ice cover,[109] with upper and lower bounds; ice algae are proportional to sea ice,[110] with an upper bound.
	mercury inputs	Sea ice cover decreases atmospheric deposition[96] but also limits evasion from seawater;[75] light availability, linked to ice cover, also affects inorganic mercury redox reactions at the air water interface.[84] Thus, decreases in ice cover are assumed to decrease mercury inputs.[84]
increasing sea surface temperature (SST)	primary production	The pelagic producer biomass production rate is linked to SST,[109] with a separate effect from sea ice melt, with upper and lower bounds.
	mercury inputs	SST increases the microbial activity and methylation rate of inorganic mercury to MeHg.[90]
multiple (e.g., SST and freshwater discharge)	primary production	Multiple environmental drivers force pelagic producer biomass (and sympagic producer biomass, if sea ice cover is included as a driver in the scenario).
multiple (e.g., SST and freshwater discharge)	mercury inputs	Multiple environmental drivers force mercury inputs.
one or multiple	multiple (e.g., primary production and mercury inputs)	One or multiple environmental drivers force both production and mercury inputs.

Table 2. Model Runs and Associated Scenario Names, Including Combinations of Different Environmental Driver Pathways

run	scenario name
base run	base run
MeHg inputs driven by freshwater discharge	freshwater-MeHg
pelagic production driven by freshwater discharge	freshwater-PP
pelagic production + MeHg inputs driven by freshwater discharge	freshwater-combined
pelagic and sympagic production driven by sea ice cover	sea ice-PP
MeHg inputs driven by sea ice cover	sea ice-MeHg
pelagic and sympagic production + MeHg inputs driven by sea ice cover	sea ice-combined
pelagic production driven by SST	SST-PP
MeHg inputs driven by SST	SST-MeHg
pelagic production + MeHg inputs driven by SST	SST-combined
production driven by sea ice cover + freshwater discharge + SST	PP-combined
MeHg inputs driven by sea ice cover + freshwater discharge + SST	MeHg-combined
production + MeHg inputs driven by sea ice cover + freshwater discharge + SST	all-combined

the BSS EwE model and by Sora et al.[36] to run historical climate change scenarios in the same model.

Freshwater discharge was extracted from Environment Canada's historical hydrometric data, sampled at several stations in the Mackenzie River near the Beaufort Sea.[105] The forcing function was created from data from the Mackenzie River (East Channel) at Inuvik. Values for freshwater discharge are presented as monthly mean values of freshwater discharge in m^3/s. We assumed that this time series was representative of all inflow into the Beaufort Sea, and this data input would be better constrained with additional station data. However, this time series, which showed an overall increasing trend, was comparable to broader freshwater discharge trends in the region. For instance, from 1984 to 2018, annual discharge rates from North American basins into the Arctic Ocean showed an overall increasing trend, though rates varied regionally.[106] In addition, Rood et al.[38] reported increasing annual discharge rates at several locations along the Mackenzie River between 1940 and 2014.

Overall, freshwater discharge (m^3/s) increased between 1970 to 2012 at a rate of 0.25 m^3/s per month ($p = 1.1 \times 10^{-4}$) (see Figure S1), assessed via the seasonal Mann−Kendall trend test and Theil−Sen slope,[102] as implemented by Hussain and Mahmud.[103] Meanwhile, annual sea ice cover (% of the model area) showed no statistically significant trend over the whole 1970 to 2012 period, though a significant decreasing trend (−0.18% per month, $p = 6.4 \times 10^{-4}$) was apparent after 2000, with sea ice cover reaching its lowest point (0%) in 2012 (see Figure S2). Finally, SST (°C) increased between 1970 and 2012 ($p = 2.6 \times 10^{-9}$, Figure S3); though with many data points at the freezing point of seawater, this significant trend yielded a Theil−Sen slope estimate of 0 °C/month.

In the baseline scenario, we include changes over time related to yearly subsistence harvesting (relevant for marine mammals like beluga) and biomass accumulation (relevant for bowhead whales, which encountered a massive population rebound after the 1970s),[37] but no other environmental forcings. The scenarios are summarized in Table 1. The model runs and their scenario names are outlined in Table 2.

2.4.1.1. Production Scenarios. The primary production scenarios were implemented using Ecosim environmental response functions. Briefly, an environmental response function relates environmental variables in the form of forcing functions to effects on biota. The user can define a relationship based on available data and assumptions so that each value of the forcing function has a defined effect on the relative foraging rate (for a consumer) or production rate (for a producer).

We used minimum and maximum thresholds of 0.5 and 1.5 times (respectively) the baseline level of producer biomass to make temporal changes in production more consistent with modeled estimates of biomass variability in the region and understandings of producer dynamics.[111] A completely linear response is often unrealistic as there can be threshold effects in primary producer biomass dynamics, where perturbations above and below an optimal zone have smaller effects.[112] We based the 0.5 and 1.5 thresholds on the simulated variability in phytoplankton biomass for pelagic producers and ice algae between 1979 and 2015 by Steiner et al.[111]

We used this logic to bound the production scenarios (i.e., to set the threshold values) for pelagic phytoplankton and ice algae so that the functional response varies by 50% from the baseline. When we assumed a simple linear relationship between each of the environmental drivers and production, the variability of biomass was much larger than this. Thus, in the absence of other data, we selected ±50% as realistic thresholds of relative primary production. Each of the production-forced scenarios, outlined below, are bounded by these thresholds.

1. Sea Ice Cover Forces Small and Large Pelagic Producer Biomass. Increases in pelagic producer biomass are linked to light availability, which is in turn associated with sea ice cover.[109] We assumed that as relative sea ice cover increases from 0.5 to 1.5, there is a corresponding proportional decrease in the environmental response of primary production; that is, if relative sea ice cover is 0.7 (a 30% reduction from its 1970 value), then the environmental response of relative primary production is 1.3 (a 30% increase relative to the base level of primary production). If sea ice cover was less than 0.5 (that is, the relative monthly sea ice cover has experienced over a 50% decline relative to 1970), we assumed that the environmental response of pelagic production does not rise above 1.5; similarly, if sea ice cover is more than 1.5 (that is, the relative monthly sea ice cover has experienced over a 50% increase relative to 1970), we assumed that the environmental response of pelagic production does not fall below 0.5 (Figure S4 in the SI).
2. Sea Ice Forces Ice Algae Biomass. Ice algae are producers found within the sea ice, and as sea ice is necessary for at least part of their life cycle,[110] sea ice cover can be correlated with ice algae abundance. Therefore, ice algae were assumed to have a linear relationship with sea ice, consistent with previous EwE modeling efforts in the BSS.[37] We assumed that as relative sea ice cover increases from 0 to 1.5, there is a corresponding proportional increase in the environmental response of primary production; unlike for pelagic producers, we assume that if sea ice cover falls below 0.5, the environmental response of ice algae production will continue to decline linearly because of their heavy reliance on sea ice (Figure S5 in the SI).
3. Freshwater Discharge Forces Small and Large Pelagic Producer Biomass. We assumed that more freshwater outflow from the Mackenzie River into the Beaufort Sea increases the production rate of small and large pelagic producers. Primary producer growth is controlled not only by light (which controls the timing of the blooms) but also by nutrient availability (which controls their abundance).[41] The Mackenzie River is an important source of heat,[113] inorganic sediment,[114] and dissolved organic carbon, nutrients, and mercury.[115] We assumed that as relative freshwater discharge increases from 0.5 to 1.5, there is a corresponding proportional increase in the environmental response of primary production, using the same rationale for the thresholds as previous primary production scenarios (Figure S6 in the SI).
4. SST Forces Small and Large Pelagic Producer Biomass. We assumed that higher SST increases the production rate of small and large pelagic producers. As outlined by Hoover et al.,[37] increases in SST have been associated with less sea ice and higher phytoplankton biomass in the open water season.[109] Declines in pelagic producer biomass have been linked to light availability, which have the same peak and decline patterns as SST.[109] We assumed that as relative SST increases from 0.5 to 1.5, there is a corresponding proportional increase in the environmental response of primary production (Figure S7 in the SI).

2.4.1.2. Mercury Scenarios. The mercury (MeHg-forced) scenarios were implemented using the environmental inflow forcing function in Ecotracer.

1. Sea Ice Forces MeHg Base Inflow. Higher sea ice cover decreases the amount of mercury deposited from the atmosphere but also increases the amount of mercury in the seawater due to decreased evasion to the atmosphere.[110] Sea ice cover, through its impacts on light availability, may also affect reduction and oxidation of inorganic mercury (with implications for evasion)[84] and methylation.[86]

   Following Schartup et al.,[84] we assumed that as relative sea ice cover increases, MeHg base inflow increases via the following relationship:

   $$1 + (\mathrm{SI} \times C_A) \tag{3}$$

   where SI is the percent change in sea ice cover and $C_A = 0.2$ is the contribution of atmospheric net deposition to the total flux of total mercury (THg) into the Arctic Ocean.[116] In the absence of other data, we assumed that MeHg fluxes are proportional to THg fluxes. This assumption is commonly made in bioaccumulation modeling studies, such as that of Schartup et al.[26] Because of the diverging effects on net deposition, there is high uncertainty in this scenario, not only in the form but also in the direction of the relationship. We address this uncertainty through an alternative functional form for eq 3, discussed as a sensitivity scenario.
2. Freshwater Discharge Forces MeHg Base Inflow. Higher freshwater discharge increases MeHg through two mechanisms: increased dissolved organic carbon inputs, thus increasing methylation,[79] and increased THg inputs, thus increasing the amount of THg available for methylation and the amount of MeHg entering the model area.[117] Permafrost thaw can also contribute to increased mercury sources from freshwater inputs and so is indirectly accounted for in this scenario.[72] We assumed that as relative freshwater discharge increases, MeHg base inflow increases via the following relationship:

   $$1 + (\mathrm{FW} \times C_{\mathrm{FW}}) \tag{4}$$

   where FW is the percent change in freshwater discharge and $C_{\mathrm{FW}} = 0.25$ is an estimate of the contribution of riverine sources to Arctic Ocean methylmercury.[116] Dastoor et al.[116] included glaciers and snowpack melt in this flux, which are not as influential for the Mackenzie River. However, we used this value as a means of also accounting for a potential pathway through increased carbon inputs and methylation.
3. SST Forces MeHg Base Inflow through Impacts on Methylation. We assumed that higher SST increases the net methylation rate of THg into MeHg primarily because it increases microbial activity.[90] While SST has other potential effects on MeHg concentrations, for instance, via its correlation with sea ice melt (discussed above) and mercury deposition, here, we focus just on the SST methylation pathway. Thus, we assumed that as relative SST increases, MeHg base inflow increases via the following relationship:

   $$1 + (\mathrm{SST} \times C_{\mathrm{ME}}) \tag{5}$$

   where SST is the percent change in SST and $C_{\mathrm{ME}} = 0.4$ is an estimate of the contribution of in situ methylation to the Arctic Ocean MeHg pool.[96] It should be noted that this mass balance study focused on the Arctic Ocean as a whole, rather than the BSS specifically; in reality, the multiplier likely depends on the context of a particular ecosystem.

2.5. Comparative Simulation Metrics

We ran segmented regressions on both observed beluga mercury time series and the modeled beluga MeHg time series to highlight the overall trends and breakpoints at which the slopes change. More detail on the regression analysis is provided in SI Section 5. We compared the modeled MeHg time series for beluga across scenarios and to observed mercury data. Beluga are culturally and ecologically important to Inuvialuit communities,[71] and there are extensive time

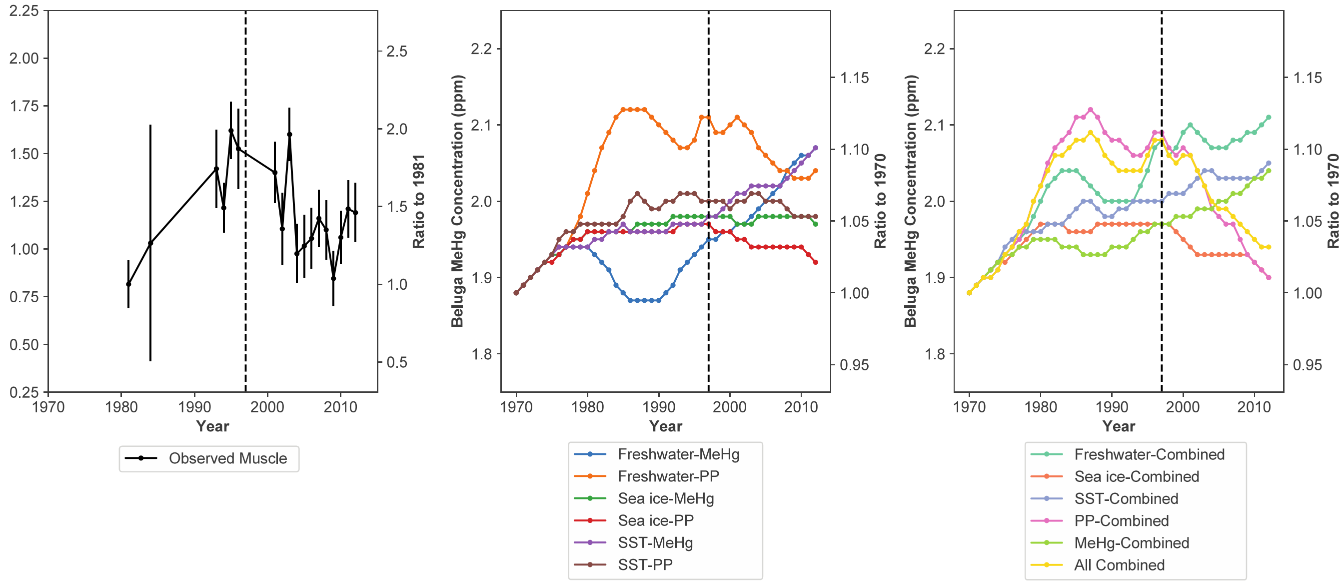

Figure 3. Observed and modeleld beluga MeHg. Left: Observed beluga MeHg (taken from muscle), using data from Loseto et al.[5] Bars represent the standard deviation. Center: Modeled beluga MeHg under each driver-pathway pairing, where drivers are freshwater discharge, sea ice cover, and SST, and pathways are MeHg inflow and primary production (PP). Right: Modeled beluga MeHg for combined scenarios. Combined scenarios for drivers (freshwater, sea ice, and SST) are the combination of MeHg and primary production pathways for that driver. Combined scenarios for pathways (MeHg and PP) are the combination of that pathway across a given environmental driver. The all-combined scenario represents the combination of all pathways and drivers. In all plots, the black dashed line represents the significant breakpoint year (1997) in the observed data at which point beluga MeHg concentrations began to decrease. The *Y*-axis scale differs between the left plot and center/right.

series for beluga mercury in the Beaufort Sea;[2,5] thus, beluga are the focus of the trend analysis, though other groups were also examined to help interpret the trends in beluga.

The observed MeHg time series data in beluga come from Loseto et al.,[5] in which the authors present temporal trends of THg in beluga liver and muscle from 1981 to 2012. The study analyzed 379 beluga (64 females and 315 males). As there were no significant differences in liver THg between male and female beluga, males and females were grouped together in the original study for the statistical analyses and are likewise here. While Loseto et al.[5] present two size groupings for their muscle THg trends, a medium size class (380 to 420 cm) and a large size class (above 420 cm), they also provide the mercury mean and standard deviation across all individuals for each year, which is what we based our segmented regression analysis on. Thus, our presentation of observed mercury trends is not divided into age nor size classes. This is consistent with how the beluga group is presented in the EwE model. In addition, although the two time series (in both the liver and muscle) show similar trends, we chose to focus the comparison between observational data and modeled concentrations on just the observed muscle trends, as most of the mercury in the liver is inorganic.[118] In contrast, the majority of mercury in the muscle is MeHg, making for a better comparison to the simulated concentrations. In addition, liver Hg has been shown to have a much longer half-life than muscle Hg and so does not accurately represent recent exposure.[118]

For comparing simulated time series against historical data, we focused on the following metrics: the direction of trends, assessed via the sign of the slope/linear regression coefficient; trend breakpoints, assessed via segmented regression; and, to a lesser extent, the magnitude of change, assessed via the magnitudes of regression coefficients. We also use these metrics and interannual variability (via calculating the coefficient of variation, or the variance between yearly averages) to compare the relative effect of modeled environmental change scenarios to each other.

3. RESULTS AND DISCUSSION

In this study, we use an EwE model of the BSS ecosystem to explore historical drivers of MeHg bioaccumulation in the Beaufort Sea food web, particularly in beluga. We compare the effect of SST, sea ice cover, and freshwater discharge on temporal trends of beluga MeHg via two pathways: ecological (in which the environmental driver forces primary production) and mercury transport (in which the environmental driver forces MeHg inputs to the system). We note that results from this study represent initial, exploratory estimates, which are primarily focused on examining the direction of change in MeHg rather than the magnitudes. As such, the functional form of the relationship between the environmental driver and MeHg concentrations is less emphasized than the direction of the correlation. Future work should explore this in more detail.

3.1. Comparing the Influence of Different Environmental Change Driver and Pathway Scenarios on Beluga MeHg

3.1.1. Comparing Primary Production and Mercury Transport Pathways. We find that the same environmental driver could yield diverging effects in simulated beluga MeHg concentrations, depending on whether it was forcing primary production or MeHg inputs. Figure 3 shows observed mercury concentrations in beluga (left) and modeled concentrations under each driver-pathway pairing (center) and combined driver-pathway pairings (right), where drivers are freshwater discharge, sea ice cover, and SST, and pathways are MeHg inputs (MeHg) and net primary production (NPP, hereafter expressed as simply PP, shorthand for primary production). When driven by the same environmental driver, production pathways and MeHg input pathways often led to diverging trends in beluga MeHg. For instance, Figure 3 illustrates that for freshwater discharge, the production pathway led to increasing beluga MeHg concentrations until 1985 before decreasing, while the MeHg input pathway led to the opposite pattern: decreasing beluga MeHg until 1988 followed by an increasing trend. Freshwater discharge decreased over the first part of the time series, which led to declines in primary

Table 3. Segmented Regression Results (Beluga MeHg against Time) of Selected Scenario Runs[a]

Scenario	Segment	Slope estimate (ppm/yr)	SE (ppm/yr)	CI (2.5%) (ppm/yr)	CI (97.5) (ppm/yr)	BP 1 (year)	BP 2 (year)
Base run	1	1.00×10^{-02}	8.98×10^{-04}	8.18×10^{-03}	1.18×10^{-02}	1975	1982
	2	4.16×10^{-03}	4.38×10^{-04}	3.27×10^{-03}	5.04×10^{-03}		
	3	1.39×10^{-03}	5.99×10^{-05}	1.27×10^{-03}	1.51×10^{-03}		
Freshwater-MeHg	1	7.81×10^{-03}	8.65×10^{-04}	6.06×10^{-03}	9.56×10^{-03}	1979	1988
	2	-1.03×10^{-02}	8.65×10^{-04}	-1.21×10^{-02}	-8.58×10^{-03}		
	3	8.54×10^{-03}	1.86×10^{-04}	8.18×10^{-03}	8.94×10^{-03}		
Freshwater-PP	1	1.67×10^{-02}	9.09×10^{-04}	1.49×10^{-02}	1.85×10^{-02}	1985	2002
	2	-1.10×10^{-03}	9.09×10^{-04}	-2.95×10^{-03}	7.41×10^{-04}		
	3	-6.64×10^{-03}	1.60×10^{-03}	-9.89×10^{-03}	-3.39×10^{-03}		
Freshwater-Combined	1	1.26×10^{-02}	1.08×10^{-03}	1.05×10^{-02}	1.47×10^{-02}	1984	1989
	2	-7.99×10^{-03}	4.96×10^{-03}	-1.80×10^{-02}	2.04×10^{-03}		
	3	4.45×10^{-03}	4.64×10^{-04}	3.53×10^{-03}	5.40×10^{-03}		
Sea Ice-PP	1	7.92×10^{-03}	4.45×10^{-04}	7.04×10^{-03}	8.83×10^{-03}	1979	1996
	2	5.73×10^{-04}	2.20×10^{-04}	1.27×10^{-04}	1.02×10^{-03}		
	3	-2.52×10^{-03}	2.01×10^{-04}	-2.93×10^{-03}	-2.12×10^{-03}		
Sea Ice-MeHg	1	8.50×10^{-03}	4.85×10^{-04}	7.52×10^{-03}	9.49×10^{-03}	1978	1994
	2	1.71×10^{-03}	2.25×10^{-04}	1.26×10^{-03}	2.17×10^{-03}		
	3	-1.06×10^{-04}	1.58×10^{-04}	-4.38×10^{-04}	2.14×10^{-04}		
Sea Ice-Combined	1	8.47×10^{-03}	6.46×10^{-04}	7.15×10^{-03}	9.78×10^{-03}	1980	1995
	2	2.35×10^{-04}	3.19×10^{-04}	-4.02×10^{-04}	8.83×10^{-04}		
	3	-3.65×10^{-03}	2.91×10^{-04}	-4.38×10^{-03}	-3.18×10^{-03}		
SST-PP	1	1.14×10^{-02}	1.14×10^{-03}	9.13×10^{-03}	1.37×10^{-02}	1977	1994
	2	3.06×10^{-03}	4.02×10^{-04}	2.24×10^{-03}	3.87×10^{-03}		
	3	-1.37×10^{-03}	3.10×10^{-04}	-2.00×10^{-03}	-7.41×10^{-04}		
SST-MeHg	1	1.00×10^{-02}	1.07×10^{-03}	7.88×10^{-03}	1.22×10^{-02}	1976	1996
	2	1.70×10^{-03}	2.18×10^{-04}	1.26×10^{-03}	2.14×10^{-03}		
	3	5.37×10^{-03}	3.06×10^{-04}	4.75×10^{-03}	5.99×10^{-03}		
SST-Combined	1	1.18×10^{-02}	1.40×10^{-03}	8.94×10^{-03}	1.46×10^{-02}	1977	NA
	2	2.42×10^{-03}	1.19×10^{-04}	2.18×10^{-03}	2.66×10^{-03}		
PP-Combined	1	1.65×10^{-02}	7.08×10^{-04}	1.50×10^{-02}	1.79×10^{-02}	1985	2000
	2	-3.00×10^{-03}	7.77×10^{-04}	-4.56×10^{-03}	-1.42×10^{-03}		
	3	-1.41×10^{-02}	1.09×10^{-03}	-1.62×10^{-02}	-1.18×10^{-02}		
MeHg-Combined	1	7.81×10^{-03}	5.37×10^{-04}	6.75×10^{-03}	8.91×10^{-03}	1979	1988
	2	-2.74×10^{-03}	4.56×10^{-04}	-3.65×10^{-03}	-1.79×10^{-03}		
	3	4.34×10^{-03}	1.23×10^{-04}	4.09×10^{-03}	4.60×10^{-03}		
All-Combined	1	1.51×10^{-02}	8.80×10^{-04}	1.34×10^{-02}	1.69×10^{-02}	1984	2000
	2	-1.17×10^{-03}	6.57×10^{-04}	-2.52×10^{-03}	1.57×10^{-04}		
	3	-1.06×10^{-02}	1.11×10^{-03}	-1.28×10^{-02}	-8.03×10^{-03}		
Observed Muscle	1	4.45×10^{-02}	1.19×10^{-02}	1.86×10^{-02}	7.08×10^{-02}	1997	2009
	2	-4.93×10^{-02}	2.19×10^{-02}	-9.71×10^{-02}	-1.61×10^{-03}		
	3	9.78×10^{-02}	1.20×10^{-01}	-1.64×10^{-01}	3.60×10^{-01}		

[a] Negative slopes are in bold. Scenarios that exhibited the same trend as the observed data—an increase in MeHg followed by a decrease—are highlighted in light blue, while scenarios that exhibited the same trend as the observed data and at the closest breakpoint to the observed data are highlighted in dark blue.

production (and subsequently more MeHg bioaccumulation) and declines in MeHg inputs. Meanwhile, over the second part of the time series, freshwater discharge increased, driving higher primary production (and subsequently MeHg biodilution) and higher MeHg inputs. Overall, changes in MeHg concentrations under the freshwater-MeHg scenario were positively correlated with the freshwater discharge time series, while changes in MeHg concentrations under the freshwater-PP scenario were negatively correlated with the freshwater discharge time series.

When comparing the combined effect of all three environmental drivers through either the production (PP-combined, see Table 2) or mercury transport pathway (MeHg-combined), simulations suggest that these environmental changes resulted in modest increases in beluga MeHg through the transport pathway during the majority of the study period, while the production pathway contributed more substantially to decreases in beluga MeHg. Figure 3 (right) shows modeled beluga concentrations for combined scenarios, for drivers (freshwater-combined, sea ice-combined, and SST-combined) and pathways (PP-combined and MeHg-combined). For the MeHg-combined scenario, beluga mercury concentrations were increasing for 35 of the 42 modeled years: 7.81×10^{-3} ppm/year from 1970 to 1979 and 4.34×10^{-3} ppm/year from 1988 to 2012 (see Table 3). In contrast, the PP-combined scenario estimated decreasing beluga mercury concentrations for 27 of 42 years: -3.00×10^{-3} ppm/year from 1985 to 2000 and -1.41×10^{-2} ppm/year from 2000 to 2012.

Overall, production and MeHg input pathway scenarios exhibited similar magnitudes of the coefficients estimated in

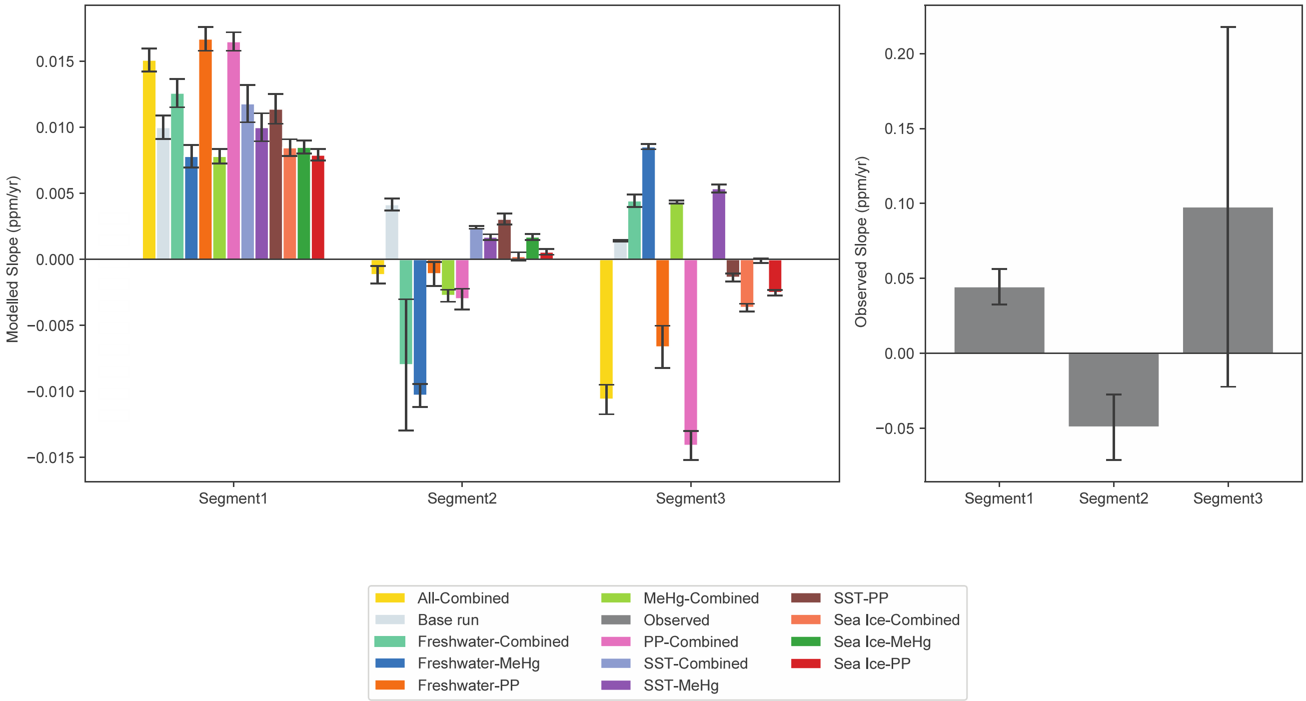

Figure 4. Comparison of regression coefficients from segmented regression (beluga MeHg against time) of selected scenario runs and observations. Two breakpoints were identified in segmented regression of observed beluga MeHg concentrations, yielding three segments on the right panel, gray bars). The regression coefficient (slope, ppm/yr) of each segment for observations and modeled scenarios is shown in bars, with standard error in the left panel.

the segmented regression analysis, emphasizing the importance of considering both changes to contaminant loading and ecosystem productivity when assessing the impacts of climate change on bioaccumulation. The biomass in BSS lower trophic levels also increased over time (from 1970 to 2010), largely due to increases in primary production.[36] While climate-induced impacts on mercury loading are critical to consider, these results suggest that additional attention to ecosystem productivity and methylation processes, both of which are poorly understood in the context of mercury,[11] is needed to understand historical and future climate impacts on biotic mercury concentrations.

3.1.2. Comparing Environmental Change Driver Scenarios. In the segmented regression analysis (Table 3), freshwater discharge consistently led to greater magnitudes of change in MeHg than other scenarios. The freshwater-PP scenario exhibited the greatest rate of change (slope = 1.67×10^{-2} ppm/year, $p = 3.57 \times 10^{-20}$) followed by the PP-combined scenario, from 1970 to 1985 (slope = 1.65×10^{-2} ppm/year, $p = 1.09 \times 10^{-23}$), and the all-combined scenario, from 1970 to 1984 (slope = 1.51×10^{-2} ppm/year, $p = 4.42 \times 10^{-18}$). Sea ice cover, on the other hand, consistently led to lesser magnitudes of change.

Overall, scenarios driven by freshwater discharge also showed higher interannual variability than other drivers. For instance, for individual MeHg scenarios, the coefficient of variation of the MeHg concentrations under the freshwater-MeHg scenario was 2.95, but it was 2.25 and 1.34 under the SST-MeHg and the sea ice-MeHg scenarios, respectively. The coefficient of variation under the individual production scenarios followed the same order (freshwater-PP = 3.56; SST-PP = 1.72; sea ice-PP = 1.12). Indeed, Arctic rivers have been highlighted as an important driver of mercury in Arctic marine ecosystems.[119] The Mackenzie River dominates the supply of inorganic sediment to the Beaufort Shelf, which has implications not only for productivity but for mercury inputs.[108,114] In other regions of the Arctic, flooding associated with hydroelectric development has been found to drive increases in biotic MeHg concentrations as a result of increasing stratification, likely an impact of increasing freshwater discharge and sea ice melt.[80] A study in Antarctica found that MeHg biomagnification increased at sites nearer to freshwater inputs.[120] Overall, freshwater discharge is important to consider moving forward, particularly as a driver of primary productivity.

3.2. Comparing Modeled Beluga MeHg Trends to Observed Data

For comparison between observed and modeled concentrations, we also performed segmented regression analysis on the observed data. Based on the segmented regression analysis (Table 3 and Figure 4), observed THg in the beluga muscle increased from 1981 to 1997 (slope = 4.45×10^{-2} ppm/year, $p = 2.85 \times 10^{-3}$); from 1997 to 2009, it decreased at a comparable slope (slope = -4.93×10^{-2} ppm/year, $p = 2.68 \times 10^{-3}$); finally, from 2009 to 2012, it increased again. However, this increase was not significant (slope = 9.78×10^{-2} ppm/year, $p = 0.25$). Thus, to compare the observed trends against the modeled ones, we chose to focus on the first breakpoint year (1997), rather than the second (2009).

Several scenarios showed a first breakpoint in the 1970s followed by a second in the 1980s or 1990s (and sometimes as late as 2001). Below, we highlight the scenarios that showed a breakpoint close in time to the observed breakpoint (1997) with the same change in direction as the observed 1997 breakpoint: an increase until the breakpoint followed by a decrease. Scenarios exhibiting this trend are highlighted in light blue in Table 3; those exhibiting the trend at a breakpoint closest to that of the observed data are highlighted in dark blue. The freshwater-PP, sea ice-PP, sea ice-MeHg, sea ice-combined, SST-PP, PP-combined, and all-combined scenarios exhibited this trend; these scenarios were mainly production-driven, although all sea ice scenarios were able to replicate the increasing and decreasing trend. When we examined the timing of changes in the modeled trends, all scenarios had second breakpoints between 1988 and 2001; in other words, all scenarios had a breakpoint within 9 years of 1997. While some MeHg-forced scenarios had breakpoints that were very close to 1997, only one of them showed a change in direction that was consistent with the observed data: the sea ice-MeHg scenario (BP = 1994). Two production-driven scenarios showed this trend, with breakpoints close to 1997: the sea ice-PP scenario (BP = 1996) and the SST-PP scenario (BP = 1994). The last scenario to exhibit this trend was when both production and MeHg inputs were driven by sea ice cover in the sea ice-combined scenario (BP = 1996). Out of the four scenarios that best matched the observed trend, three were driven by sea ice cover. Three other scenarios showed increasing to decreasing trends at earlier breakpoints and then a second breakpoint that led to a sharper decrease: the freshwater-PP scenario (BP 1 = 1985, BP 2 = 2002), the PP-combined scenario (BP 1 = 1985, BP 2 = 2000), and the all-combined scenario, in which both production and MeHg inputs were driven by all three environmental drivers (BP 1 = 1984, BP 2 = 2000).

Production pathway scenarios were more reflective of the timing of observed MeHg trends in beluga than MeHg input pathway scenarios, given that production pathway scenarios generally exhibited increases in MeHg followed by decreases, while MeHg input pathway scenarios typically exhibited statistically significant increasing trends only, as shown in Figure 3. The MeHg input pathway scenarios that reflected the observed beluga MeHg trends were driven by sea ice cover.

Though there are limited time series data for other high trophic species in the Beaufort Sea, some comparisons can be made between beluga MeHg trends and those of other predators. For instance, McKinney et al.[121] observed a decreasing trend in southern Beaufort Sea polar bear hair THg from 2004 to 2011, which overlaps with the period of decrease for beluga, attributed largely to changes in foraging ecology. These changes in foraging ecology may also be linked to some of the environmental drivers explored in this study, such as sea ice cover. Kannan et al.[122] also found a decreasing trend in liver mercury (Hg) in polar bears in Alaska from 1993 to 2002. Liver Hg in ringed seal in the Beaufort Sea exhibited no significant trend from 1973 to 2007,[42] though muscle Hg in Eastern Beaufort Sea seals has shown decreasing levels after 2009.[2]

In all, these results suggest that both production-driven effects and changes in mercury inputs related to changes in sea ice cover, SST, and freshwater discharge could contribute to observed increases in beluga MeHg up until year 1997 and decreases after 1997. In both cases, sea ice cover appears to be an important driver of the timing of observed trends, though freshwater discharge and SST resulted in greater magnitudes of change. These study results highlight the importance of food web effects but show that mercury inputs still play an important role. Indeed, a single driver is unlikely to be the sole cause of the changes in beluga MeHg. A 2002 workshop pointed to the Arctic Oscillation and potential atmospheric depletion events that might have led to increases in beluga mercury in the 1990s, but the workshop participants could not agree on a single driver that might be affecting beluga mercury.[123] Loseto et al.[5] contemplated the Pacific Decadal Oscillation as a possible driver, and other recent studies also highlight the importance of Pacific-based processes as important influences on EBS beluga.[34,124] Suprenand et al.[104] modeled the BSS ecosystem from 1970 to 2014 and highlighted a potential climatological tipping point in 1993 followed by a biological tipping point in 1998, in which biodiversity values preceding that year were significantly different to biodiversity values after. This coincides with our breakpoint year of 1997, when beluga mercury started to decrease (Figure 3).

None of the modeled scenarios capture the magnitude of change that was seen in the observed data. For instance, the modeled scenarios that were closest to capturing the observed magnitude of change were the freshwater-PP scenario and the combined-PP scenario, both of which exhibited a 12.8% increase in concentration, from 1.88 ppm in 1970 to a maximum of 2.12 ppm in 1985 and 1987, respectively. Meanwhile, the observed data exhibited a 98.8% increase in concentration, from 0.82 ppm in 1981 to 1.62 ppm in 1995. Some possible explanations are discussed below in the Limitations and Future Work section.

3.3. Limitations and Future Work

We simulated sensitivity scenarios to assess the potential impact of uncertainty about (1) the strength of the relationship between environmental change drivers (freshwater discharge, SST, and sea ice cover) and MeHg inputs and primary production (see SI, Figures S8 and S9) and (2) the direction of the relationship between sea ice cover and MeHg inputs, given the uncertainty associated with the relationship between sea ice cover and MeHg inputs[84,116] (see SI, Figures S10 and S11). More information on the sensitivity analyses is provided in SI Section 6.

Capturing the observed magnitudes of mercury concentrations was challenging largely because the EwE representation of the food web does not capture certain important dynamics, such as beluga migration into and out of the system, and because Ecotracer is unable to capture the complexities of the mercury cycle, such as methylation rates from inorganic mercury to MeHg. Parameter and structural uncertainty for this model was addressed by Li et al.,[34] who showed that MeHg concentrations in beluga were overestimated in the model, likely because the model does not account for beluga migration and feeding in different Arctic areas. The Bering Sea exhibits higher productivity than the Beaufort Sea,[5] which could lead to biomass dilution of MeHg in this beluga population's wintering habitat. Indeed, organisms like ringed seal, beluga, and polar bear in the Beaufort Sea have been found to have higher concentrations of mercury than organisms elsewhere in the Arctic,[1] and common prey items of beluga, such as Arctic cod and Pacific herring, are 2 to 2.7 times lower in the Bering Sea than in the Beaufort Sea.[34] Further refinements to this study's modeling efforts could lie in creating multiple spatially explicit models in Ecospace to account for the beluga stock's migration.

To assess parameter uncertainty in the base Ecopath model, Hoover et al.[35] compared modeled trophic levels with those derived from nitrogen stable isotope modeling and found that EwE performed well for most groups, though performance was worse for species groups representing broad taxonomic levels, groups with poorly documented diets, and anadromous fish. Meanwhile, Suprenand et al.[104] conducted sensitivity analysis of the Ecosim model via Monte Carlo analysis, examining how initial species group biomass estimates impacted the ecosystem's stability over time. They concluded that the model was robust for temporal analysis.[104] Scenario uncertainty could have originated from data limitations and assumptions. For instance, research has shown that the bioavailability of dissolved mercury is higher than that of particulate-bound mercury;[79] however, there were not enough observed data on this subject nor was there a specific mechanism in which to model this in Ecotracer. In addition, the environmental response functions were limited by our understanding and measurement of sympagic and pelagic primary production.[125,126]

While this study's selected drivers can indeed explain some aspects of the variability seen in beluga MeHg, other drivers not examined here could also contribute to observed changes. For instance, dietary shifts are important to consider for changing bioaccumulation trends, especially given that the Arctic is seeing climate-driven species invasions that often lengthen food chains and thus have the potential to increase bioaccumulation, particularly in apex predators.[97] Beluga in the Beaufort Sea primarily feed on Arctic cod, a species that is highly sensitive to climate change.[127] Prey item analysis in the region indicates that decreasing consumption of Arctic cod by beluga, and increasing consumption of another prey source, capelin, coincided with a decline in beluga condition.[127] While we did not examine dietary shifts and their effect on beluga mercury, we note that it should be prioritized for further study, especially given observed declines in the Bering Sea and possible impacts on beluga condition.[124] We also recommend that future studies prioritize erosion and permafrost thaw, which were raised as points of concern during our meetings with the IGC and HTCs in the ISR. Erosion has been increasing in the region[10,128] and has been shown to increase mercury inputs to aquatic systems.[129] Thawing permafrost, too, releases nutrients and organic carbon, which can stimulate methylmercury production, as well as primary production.[130]

We do not consider the impact of anthropogenic activity on MeHg inputs to the ecosystem, though human emissions also varied substantially during the modeled period. Estimates vary across studies; there is debate on global emission trends depending on the emission inventory.[6] For instance, a 1970–2008 trend analysis by Muntean et al.[7] estimated that global Hg emissions to the atmosphere increased steadily after 1970, rising to an estimated 1287 tons in 2008, 61% higher than in 1970. In an 1850–2010 decadal trend analysis, Streets et al.[131] estimated that global Hg emissions to the atmosphere increased from approximately 1.2 Gg/year in 1970 to 1.9 Gg/year in 2010, roughly 58% higher than in 1970. They estimated a slight decrease between 1990 and 2000 before emissions rose again.[131] Meanwhile, Streets et al.[132] estimated that global Hg releases to air, land, and water decreased slightly from 1970 to 2010.

Further, human emissions of mercury are estimated to be a key driver of Arctic Ocean mercury concentrations in the future, even when compared to the impact of climate-induced effects.[84] Thus, we recommend considering the combined impact of mercury emission scenarios and climate drivers in future studies.

4. CONCLUSIONS

Exploratory modeling of MeHg trends in beluga, as well as in other species in the Beaufort Sea, suggests that climate-induced impacts on primary productivity and prey rather than mercury inputs alone play a larger role in explaining historical MeHg trends. Production scenarios overall were able to better represent trends in beluga, particularly the declining trend in MeHg after 2000, and beluga MeHg was more sensitive to changes in freshwater discharge than any other environmental driver. That said, all three drivers could play a role in explaining beluga mercury over time, with sea ice cover driving trends closest to that of observed data.

We prioritized environmental drivers based on findings from both Western Science and Inuvialuit Knowledge. We found that the two knowledge systems supported one another, focusing on different yet complementary parts of the system. We hope that this study has provided space for Inuvialuit Knowledge in codesign and has shown the importance of considering IK more broadly when using ecosystem models. This research would have benefited from additional opportunities to cointerpret model outputs with Inuvialuit Knowledge holders; we recommend funding opportunities to support these activities for model-based investigations. For future work on environmental change and contaminant cycling in the Beaufort Sea, the FJMC recommended conducting similar engagement with Gwich'in harvesters in the region. Environmental changes in upstream territories (e.g., permafrost thaw) can also have important implications for the BSS, which further emphasizes the importance of wider engagement. Other opportunities for future work could lie in studying the drivers of the region's primary producers, which are important for explaining beluga MeHg trends but are also associated with uncertainty. Understanding climate-driven impacts through the beluga winter habitat in the Bering Sea and other Pacific influences are also critical next steps.

This exploratory modeling exercise demonstrated that Ecotracer is a useful tool for exploring the effects of environmental change on contaminant cycling and bioaccumulation. While not as complex as other contaminant models, its simplicity makes it well-suited to understanding the ecosystem processes that underlie bioaccumulation to begin with. Future development and work with Ecotracer could include building in methylation processes, multiple contaminant species, and environmental inflow forcing functions for not only the environmental compartment but for functional groups, too. EwE and Ecotracer could also be coupled to more quantitative models (e.g., using primary producer biomass time series from climate model outputs as inputs to EwE scenarios). Additional laboratory, mesocosm, and in situ field studies, as well as ecotoxicological tests applied to Arctic species, would support this model development. As shown here, modeling is a powerful tool, but it is all the more powerful when it informs and is informed by real-world monitoring and data collection.

■ ASSOCIATED CONTENT

Data Availability Statement

The data underlying this study are openly available in Zenodo at 10.5281/zenodo.7013095. The Ecopath with Ecosim modeling software suite is available at https://ecopath.org/downloads/.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsenvironau.3c00072.

> Material on Eastern Beaufort Sea beluga biology, key results from the Western Science and Inuvialuit Knowledge literature review, model parameters, scenario implementation, regression analyses, and sensitivity analysis (PDF)

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Notes

The authors declare no competing financial interest.

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ADDITIONAL NOTES

[1] Here, we use a definition of Western Science from the Intergovernmental Science-Policy Platform on Biodiversity and Ecosystem Services (IPBES): *"Western Science (also called modern science, Western scientific knowledge, or international science) is used...as a broad term to refer to knowledge typically generated in universities, research institutions, and private firms following paradigms and methods typically associated with the "scientific method" consolidated in Post-Renaissance Europe on the basis of wider and more ancient roots. It is typically transmitted through scientific journals and scholarly books. Some of its central tenets are observer independence, replicable findings, systematic scepticism, and transparent research methodologies with standard units and categories"*.[50]

[2] The Inuit Circumpolar Council defines Indigenous Knowledge as *"a systematic way of thinking applied to phenomena across biological, physical, cultural, and spiritual systems. It includes insights based on evidence acquired through direct and long-term experiences and extensive and multigenerational observations, lessons, and skills. It has developed over millennia and is still developing in a living process, including knowledge acquired today and in the future, and it is passed on from generation to generation"*.[51] Here, Inuvialuit Knowledge is used to refer specifically to knowledge from the Inuvialuit People.

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