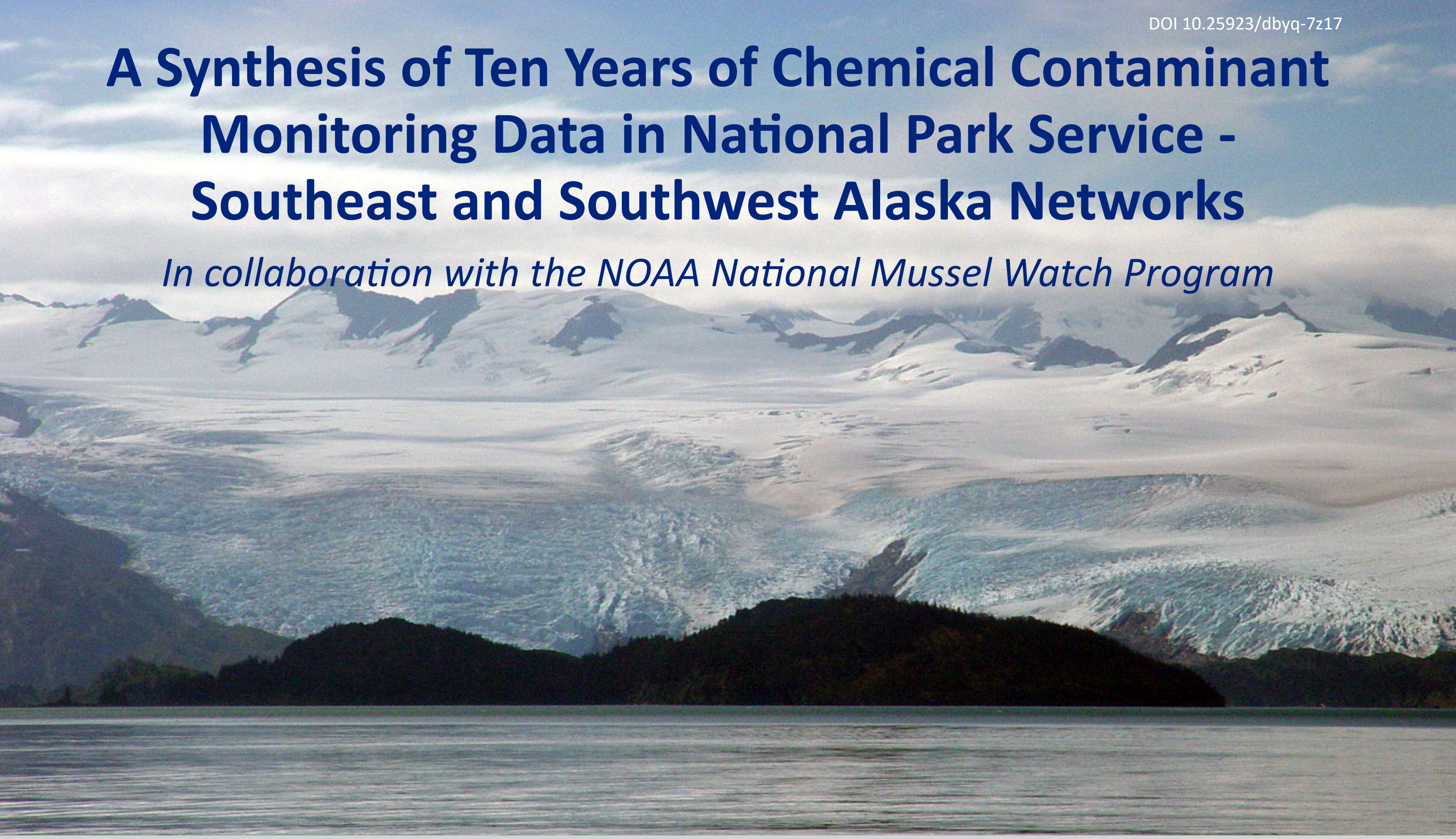
DOI 10.25923/dbyq-7z17

A Synthesis of Ten Years of Chemical Contaminant Monitoring Data in National Park Service - Southeast and Southwest Alaska Networks

In collaboration with the NOAA National Mussel Watch Program



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July 2020

NOAA TECHNICAL MEMORANDUM NOS NCCOS 277

NOAA NCCOS Monitoring and Assessment Branch

Front Page Images (Clockwise): Alaska. Credit: NOAA; Blue mussels from Glacier Bay, AK. Credit: AK NPS; Seward, AK. Credit: NOAA; Blue mussels from Nanwalek, AK. Credit: NOAA

Back Page Image: Alaska. Credit: NOAA

Acknowledgements

This project was supported by the National Park Service (NPS). We would like to acknowledge the NPS' Southeast and Southwest Alaska Networks and their partners for conducting the initial fieldwork and data collection. This report could not have been completed without the cooperation, time and effort contributed by many, whose collective input has resulted in a document far superior to that which we envisioned. We would also like to thank all of the reviewers and collaborators for their invaluable assistance.

Felipe Arzayus	National Oceanic & Atmospheric Administration
A.K. Leight	National Oceanic & Atmospheric Administration
Kevin McMahon	National Oceanic & Atmospheric Administration
Ed Wirth	National Oceanic & Atmospheric Administration

The Southwest Alaska Networks' research described in this publication was supported by the Exxon Valdez Oil Spill Trustee Council. However, the findings and conclusions presented by the authors do not reflect the views or position of the Trustee Council.

Suggested Citation

Rider, M., Apeti, D.A., Jacob, A., Kimbrough, K., Davenport, E., Bower, M., Coletti, H. and Esler, D., 2020. A Synthesis of Ten Years of Chemical Contaminants Monitoring in National Park Service - Southeast and Southwest Alaska Networks. A collaboration with the NOAA National Mussel Watch Program. NOAA Technical Memorandum NOS NCCOS 277. Silver Spring, MD. 110 pp. DOI 10.25923/dbyq-7z17

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NOAA Technical Memorandum NOS NCCOS XXX

United States Department of Commerce	National Oceanic and Atmospheric Administration	National Ocean Service
Wilbur L. Ross, Jr. Secretary	Neil Jacobs Administrator, Acting	Nicole LeBoeuf Assistant Administrator, Acting

Contents

Abbreviations

DDT	Dichlorodiphenyltrichloroethane
EPWS	Eastern Prince William Sound
g	gram
GLBA	Glacier Bay National Park and Preserve
GWA	Gulf Watch Alaska
HCH	Hexachlorocyclohexane
KATM	Katmai National Park and Preserve
KBAY	Kachemak Bay
KEFJ	Kenai Fjords National Park
KLGO	Klondike Gold Rush National Historical Park
MDL	method detection limit
µg	microgram
MWP	Mussel Watch Program
NS&T	National Status & Trends
ng	nanogram
NOAA	National Oceanic and Atmospheric Administration
NPS	National Park Service
NPWS	Northern Prince William Sound
PAH	Polycyclic aromatic hydrocarbon
PBDE	Polybrominated diphenyl ether
PCB	Polychlorinated biphenyl
ppm	parts per million
SE	standard error
SEAN	Southeast Alaska Network
SITK	Sitka National Historical Park
SWAN	Southwest Alaska Network
US	United States
WPWS	Western Prince William Sound

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Executive Summary

This report summarizes the contaminant results from marine sampling conducted between 2007 and 2018 by the National Park Service (NPS Southeast Alaska Network (SEAN and Southwest Alaska Net-work (SWAN) and their partners in association with the NOAA National Mussel Watch Program (MWP). As part of their monitoring efforts to determine the status and trends of chemical contaminants in near-shore and coastal areas within their park boundaries, the SEAN and SWAN have partnered with the MWP to measure a comprehensive suite of contaminants in bay mussels (*Mytilus trossulus*) at targeted sites across the networks. Sampling also occurred outside of parks through external partnerships with the Gulf Watch Alaska (GWA) marine monitoring program and the Municipality of Skagway to provide a regional perspective on contaminant levels. The goal was to create a robust baseline of contaminant concentrations from this region and to compare these concentrations to national MWP data. This report represents a rich source of information to help support the parks' responses during unforeseen catastrophic marine pollution events and guide future sampling site choices and frequency.

For this report, 42 sites and 120 samples located across southern Alaska were combined to characterize the extent of contamination and bring both temporal and spatial variations into context. Mussel samples were analyzed for trace metals (Arsenic, Cadmium, Copper, Lead, Mercury and Nickel), total Butyltins, total Chlordanes, total Chlorobenzenes, total DDTs, total Dieldrins, total Endosulfans, total HCHs, Mirex, Chlorpyrifos, total PAHs, total PBDEs, total PBBs and total PCBs. Contaminant concentrations from this study were summarized and compared to the long-term NOAA National Status and Trend (NS&T) moni-toring data for each group of contaminants.

With the exception of PAHs and trace metals, which were detected at 100% of the sites, all of the other contaminants were detected at varying frequencies. PBBs, Mirex and Endosulfans were not detected in any of the samples and Chlorpyrifos was only detected in five samples across four sites. Chlordanes were present at 79% of the sites while Butyltins were only detected at 20% of the sites. Overall, the majority of the concentrations can be considered to be at background levels when compared to the long-term NOAA National Status and Trends (NS&T) monitoring data for blue mussels nationwide. The relatively high concentrations of cadmium, copper, and nickel in comparison to the NS&T national groups could be a combination of natural inputs and anthropogenic sources. The natural exposure and weathering of rocks in southern Alaska can contribute to elevated background concentrations of these metals. Sample concentrations, compositions and/or trends for Total DDT, Total Dieldrins and Total HCHs suggest that these contaminants are no longer bioaccumulating at detectable levels. Total Butyltin concentrations were low compared to the NS&T national concentrations, but the presence of tributyltin (TBT) in recent years at Sitka Visitor's Center (SITK) and Skagway Harbor (SKWY) indicates that fresh sources of Butyltin are still entering these environments, probably through vessel traffic at these sites. The PAH profiles and higher concentrations at SITK, SKWY and Nahku Bay East Side (NBES) suggest that these sites are receiving anthropogenic sources of PAH contamination.

The results included in this report help to provide a greater understanding of general background contamination in NPS SWAN and SEAN parks, as well as other monitoring sites, including range, trends and variability. Future monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale through periodic sampling as well as focusing on areas of interest that could shed further insight on range and variation (see supplemental material).

Seward, AK. Credit: NOAA

Introduction

Nanwalek, AK. Credit: NOAA

Introduction

This report summarizes contaminant results from marine samples collected by the National Park Service (NPS) and its partners between 2007 and 2018 in association with the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Program (MWP). The Southeast Alaska Network (SEAN) and Southwest Alaska Network (SWAN) are two of 32 National Park Service (NPS) Inventory and Monitoring networks nationally. The SEAN is comprised of four coastal park units: Glacier Bay National Park and Preserve (GLBA), Klondike Gold Rush National Historical Park (KLGO), Sitka National Historical Park (SITK) and coastal portions of Wrangell-St Elias National Park and Preserve (WRST). Coastal portions of WRST were recently added to the SEAN monitoring program in 2019, but given a lack of marine samples from these areas, the scope of this report will be limited to the three original SEAN units (GLBA, SITK, KLGO). The SWAN is comprised of five park units: Alagnak Wild River, Aniakchak National Monument and Preserve, Katmai National Park and Preserve (KATM), Kenai Fjords National Park (KEFJ), and Lake Clark National Park and Preserve (Figure 1).

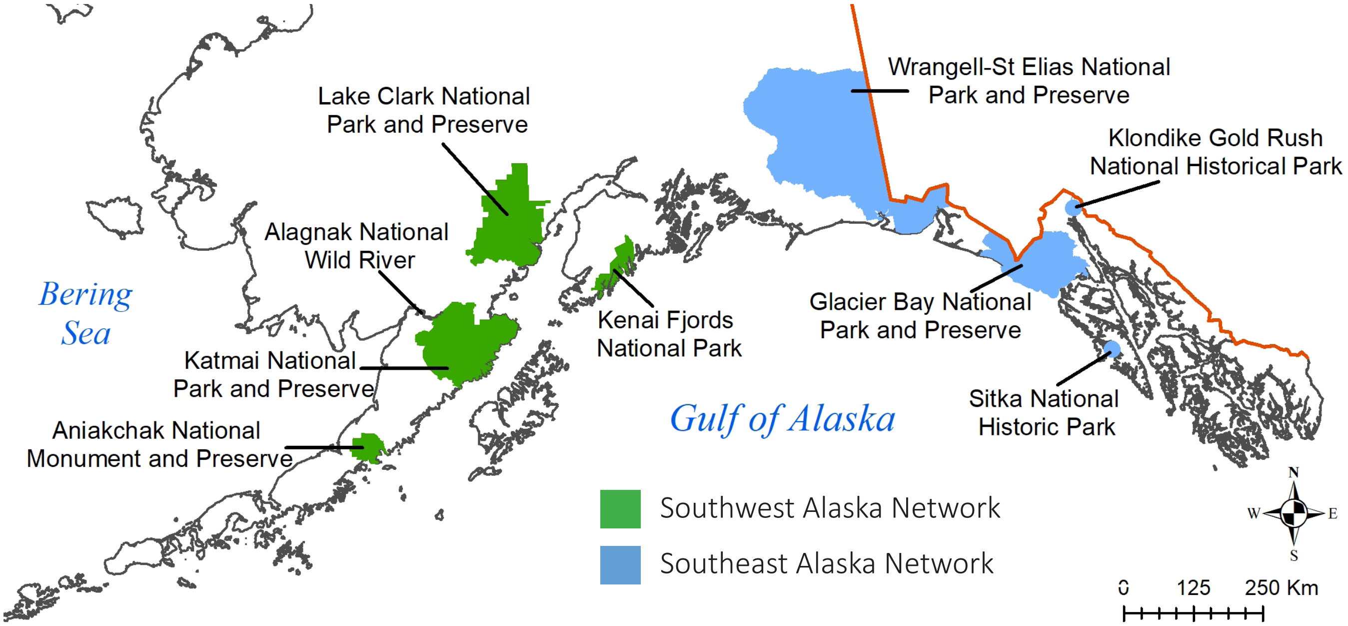

Figure 1. Map of southern Alaska showing the five park units in Southwest Alaska Network (SWAN) and four park units in Southeast Alaska Network (SEAN). Wrangell-St. Elias National Park and Preserve is supported by both the Central Alaska Network (inland areas) and SEAN (coastal areas).

The marine coastlines in these parks are susceptible to chronic pollution as they receive cumulative contamination from diverse diffuse sources. They are also susceptible to catastrophic releases, as in the case of the Exxon Valdez oil spill in 1989. With no known industrial point sources of pollution in the vicinities of these national parks, current sources of coastal contamination may include wastewater discharge, marine activities associated with commercial and recreational fishing, commercial shipping, and marine current transports, and the release of legacy contaminants from melting glaciers. Historically, seafood canning operations and the mining and export of coal and minerals have generated shoreline and watershed contaminant inputs in the region. Lately, concern regarding pollution originating from local sources associated with marine transportation and tourism have increased. Additionally, natural sources of pollution, particularly metals, may be associated with river runoff. Sources can also include long-range atmospheric transport that results in widespread global distribution by means of the "grasshopper" effect, where cycles of evaporation and distillation carry pollutants from low to high latitudes (Wania and Mackay, 1996).

INTRODUCTION

Contaminants, particularly those that are lipophilic, can biomagnify in the coastal food chain, such as shellfish and fish, with increasing concentrations in predatory wildlife and humans. Excessive levels of contaminants in the coastal areas, whether of natural or anthropogenic origin, can pose ecological and human-health risks. The presence of contaminants in coastal ecosystems can lead to loss of biodiversity through degraded habitats, biomagnification of contaminants in the coastal ecosystem, and human consumption of contaminated fish and wildlife. Thus, characterizing coastal pollution is often viewed as an important goal of coastal resource management. This is particularly important in Alaska where subsistence food contamination is a health concern, especially in rural areas where large amounts of these foods are consumed as a primary source of protein (Wolfe, 1996). Additionally, polluted marine shorelines could have immediate relevance to park management since catastrophic events and chronic exposure may be caused, in part, from permitted visitor activities such as cruise ship excursions.

In an effort to construct a broad-based understanding of the status and trends of park resources as a basis for making decisions and working with other agencies and the public for the long-term protection of park ecosystems, the NPS has developed a long-term ecological monitoring program consisting of 32 eco-regional networks of parks including the SEAN and SWAN in Alaska. This program encompasses a broad spectrum of research and monitoring studies to evaluate ecosystem health in the parks. The SEAN and SWAN both implement long-term monitoring of natural resources in their respective coastal park units in Alaska. In both monitoring networks, the marine contaminants found in nearshore mussels play an important role as “vital signs” of park marine health (Bennett et al., 2006; Moynahan et al., 2008). As part of their monitoring efforts, to determine the status and trends of chemical contaminants in nearshore and coastal areas within their park boundaries, the SEAN and SWAN have partnered with the NOAA na-

Mussels from Glacier Bay, AK. Credit: AK NPS tional MWP to measure a comprehensive suite of contaminants in bay mussels (*Mytilus trossulus*) at targeted sites across the networks. Sampling also occurred outside of parks through external partnerships with the Gulf Watch Alaska (GWA) marine monitoring program and the Municipality of Skagway. GWA is supported in part by the Exxon Valdez Oil Spill (EVOS) Trustee Council, a council created to oversee the restoration of injured ecosystems and resources from the EVOS. GWA sampling included Prince William Sound and Kachemak Bay sites. The partnership with the Municipality of Skagway supported sampling at sites in proximity to communities including Homer, Port Graham, Nanwalek, Seward and Skagway. This allowed for a regional perspective on contamination levels across a large portion of the Gulf of Alaska.

The NOAA national MWP has monitored over 300 chemical contaminants in bivalve tissue at over 300 sites since 1986 and maintains the longest running contaminants sampling program in the US (Kimbrough et al., 2008). The MWP design is based on the periodic collection and analysis of bivalves (oysters and mussels) and sediment from a network of monitoring sites located throughout the nation’s coastal zones including Alaska. The MWP provides unique long-term data that is vital to evaluating the health of the nation’s coastal waters regional and nationally. The partnership with the MWP was envisioned during the early phases of both SEAN and SWAN’s monitoring program development as an efficient means of marine contaminants monitoring in southern Alaska (Bennett et al., 2006; Moynahan et al., 2008). This approach was further recommended as a simple means to contextualize results from southern Alaska following a broad scale inventory of mussel contaminant loads conducted in 2007 (Tallmon, 2012). Since 2007, at least 125 mussel and sediment samples have been collected from shorelines within or adjacent to national park units in southern Alaska, but the laboratory results had not been summarized in a consistent manner or examined for potential trends. There was a strong need to consolidate existing data in one document and create a template for analyzing future results. The goal was to create a robust baseline of contaminant concentrations and compare them to known human and ecological health thresholds. Such a baseline report creates a rich information source available for park responses to catastrophic marine pollution events and guides future sampling site choices and frequency.

Methods

Seward, AK. Credit: NOAA

STUDY AREA DESCRIPTION

Southeast Alaska Network (SEAN)

The three original SEAN units (GLBA, SITK, KLGO) encompass nearly 3.3 million acres and represent the full range of landscapes within Southeast Alaska (Figure 2). Maritime influence and steep topography create abundant precipitation in this region. Weather patterns are dominated by a strong Aleutian Low in the northern Gulf of Alaska in the fall, winter, and spring, and by weak high pressure systems in the summer. Low elevations are dominated by temperate rainforest while higher elevations in GLBA and KLGO have significant and rapidly changing glacial coverage. Some coastal and high-elevation locations in GLBA and SITK exceed 330 cm (130 in) of rainfall annually (Shulski and Wendler, 2007), while KLGO is influenced by a rain shadow effect and receives as little as 76 cm (30 in) each year. These atmospheric patterns affect the transport of contaminants to terrestrial, freshwater, and marine systems.

Marine waters cover over 2,000 km^2 within GLBA boundaries. The park's approximately 1,900 km of coastline is the longest in NPS jurisdiction. Although coastal-estuarine portions of KLGO and SITK are very small in comparison to GLBA, both parks are directly influenced by marine activity (Moynahan et al., 2008). All three parks are exposed to vessel traffic ranging in size from small private boats to large cruise ships; thus, there remains a persistent potential for pollution events such as oil spills. In GLBA, more than 95% of visitors enter the park via cruise ship during the peak tourism period of June through August (Mölders and Gende, 2015). In addition to the potential for oil spills, cruise ships emit gas and particulates that may deposit contaminants within park boundaries. With the adoption of scrubbers, which remove sulfur dioxide (SO_2) from exhaust gases, there is the greater potential for discharge of wash water that can be high in PAHs.

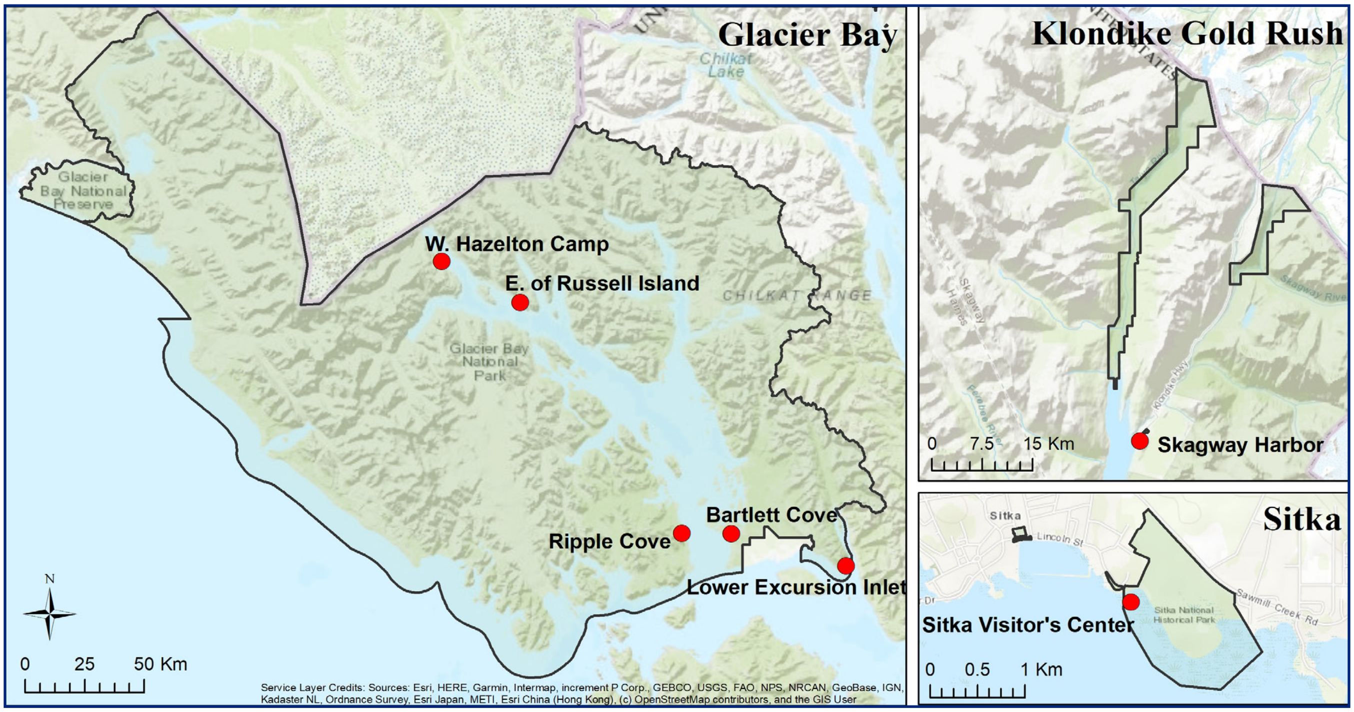

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Figure 2. Map of SEAN's six long-term marine contaminants monitoring stations in Glacier Bay National Park and Preserve, Klondike Gold Rush National Historical Park, Sitka National Historical Park, and one site in Skagway Harbor (sampled in partnership with the Municipality of Skagway). Red dots represent bay mussel tissue analysis collection sites.

Southwest Alaska Network (SWAN)

SWAN is comprised of five parks and encompasses approximately 9.4 million acres. Parks in SWAN that are included in contaminant monitoring are Katmai National Park and Preserve (KATM) and Kenai Fjords National Park (KEFJ) (Figure 3). Mussel samples were collected in conjunction with ongoing nearshore marine monitoring as part of Gulf Watch Alaska (https://gulfwatchalaska.org/), which is designed to monitor many aspects of marine ecosystems in the northern Gulf of Alaska region affected by the 1989 Exxon Valdez oil spill (Coletti et al., 2016). A valuable attribute of SWAN participation in Gulf Watch Alaska is that the program generates nearshore monitoring data from outside of the SWAN parks, allowing broader contrasts and regional perspectives on changes that are observed within parks. This rationale is also relevant for mussel contaminant sampling as Gulf Watch Alaska contaminant monitoring has occurred within the two parks mentioned above as well as Kachemak Bay (KBAY) and western, northern and eastern Prince William Sound (WPWS, NPWS and EPWS respectively).

The biogeography and climate of SWAN and Gulf Watch Alaska sites are similar to those described above for SEAN sampling sites. SWAN/Gulf Watch Alaska sites vary in their degree of human activity, with Kachemak Bay having considerable shoreline development and higher vessel traffic than any of the other sites. Prince William Sound was the site of the 1989 Exxon Valdez oil spill and WPWS beaches

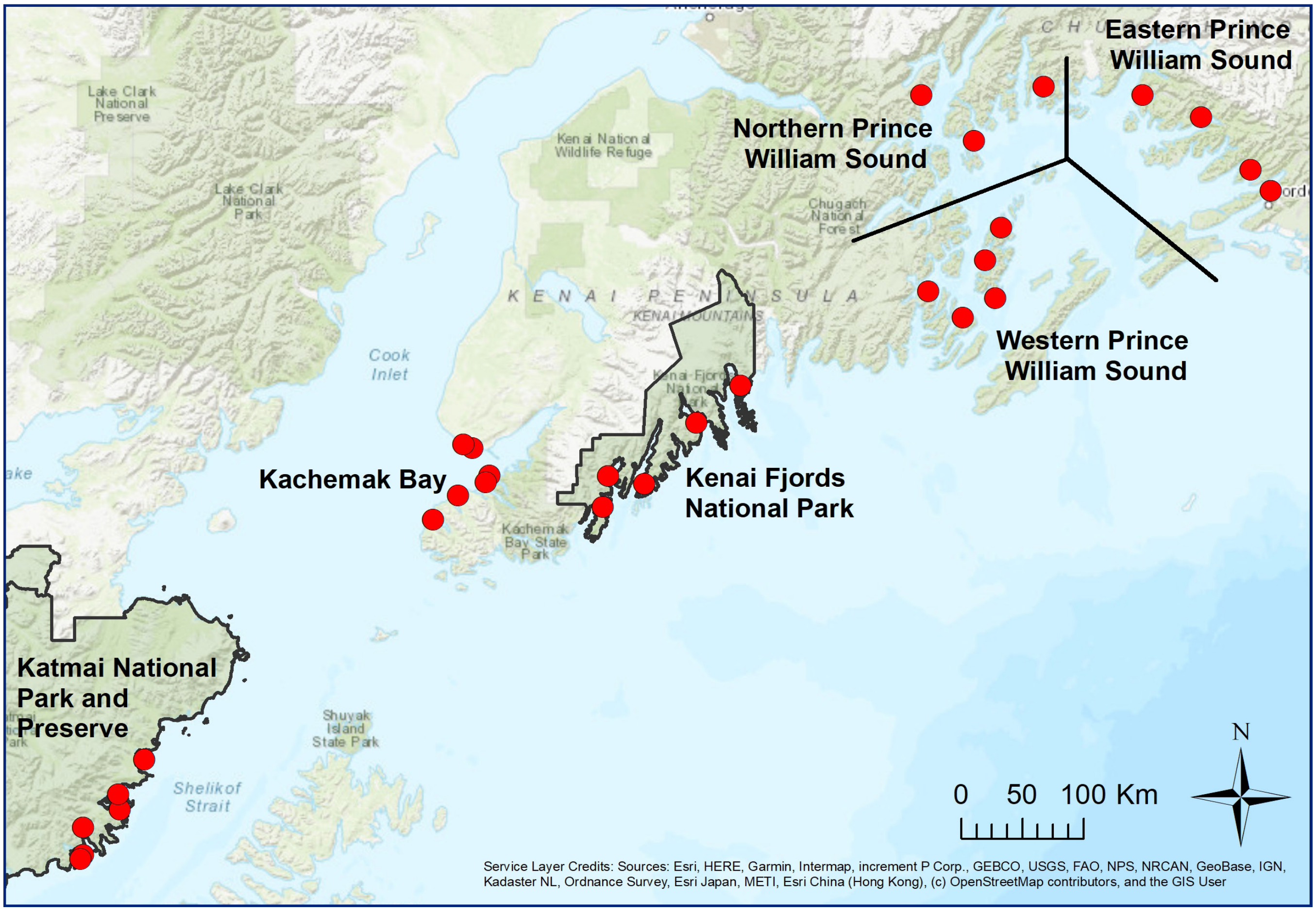

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Figure 3. Map of SWAN long-term marine contaminants monitoring stations in Katmai National Park and Preserve and Kenai Fjords National Park, and sites in Prince William Sound and Kachemak Bay sampled in partnership with Gulf Watch Alaska. Red dots represent bay mussel tissue analysis collection sites.

received a large proportion of the spilled oil (Galt et al., 1991). Some oil was also stranded on beaches outside of Prince William Sound, including KEFJ and KATM (Wolfe et al., 1994). Mussels have been assessed for PAH concentrations since the spill and some investigators indicate that effects of the spill abated quickly. However, oil remained in intertidal sediments for decades (Li and Boufadel, 2010; Irvine et al., 2014), as did exposure in higher trophic levels (Esler et al., 2016).

SAMPLING DESIGN

Sample design and collection was conducted following standard protocols utilized by the national MWP (Apeti et al., 2012). Each site sample consists of a composite of 30-60 mussels collected from different stations around the site. The MWP field activities are designed in a manner so as not to have any significant impact on the environment.

In 2007, the SEAN conducted an extensive baseline inventory of contaminants in mussel tissues at 53 sites in and adjacent to parklands (48 in GLBA, two in KLGO, and three in SITK) (Tallmon, 2011). These sites represented a range of relatively pristine to heavily human-modified sites. Since that initial 2007 survey, a subset of six sites has been monitored every two years (five in GLBA, one in SITK) (Figure 2, Figure 4). Four of the five long-term sites in GLBA follow the cruise ship path from lower to upper bay, while one site is near a fish processing plant in Excursion Inlet that may potentially contribute pollution to intertidal areas within park boundaries. SITK encompasses a small marine shoreline relative to the other parks; therefore, one monitoring site is sufficient for characterizing marine contaminants within the park. The 2007-2011 results from this sampling effort were summarized and published in Tallmon, 2012. One additional site (Skagway Harbor) was sampled in 2017 in partnership with the Municipality of Skagway, though it is not part of the SEAN's long-term monitoring design. While coastal portions of WRST were recently added to SEAN in 2019, no sample collection has yet been completed there. SEAN is currently planning an inventory of contaminants in bay mussels in this area to establish baseline conditions and will be selecting a number of sites to be incorporated into their long-term marine contaminants monitoring plans.

Nearshore sampling sites for SWAN and Gulf Watch Alaska (Figure 3, Figure 4) were selected using a randomized process within blocks that allows extrapolation at the block level (Bennett et al., 2006), with the exception of KBAY, where the sites were selected based on high diversity. Some blocks are sampled annually (KEFJ, KATM, KBAY, and WPWS) and each of the sites within these blocks had mussel samples collected during 2018 (Table 1). Other blocks are monitored intermittently (EPWS, NPWS) and were not sampled during 2018, but some sites were sampled in 2007 or 2012-2013 (Table 1).

The MWP began routinely monitoring in Southern Alaska at the conception of the program in 1986. It sampled two sites annually until 1993 and then added three more site in 1995, which it sampled biennially until 2007 at which point funding restrictions limited the ability to maintain the exisiting national sampling design. As a result, the MWP has developed a rotating regional design that leverages partnerships with local agencies and stakeholders such as SEAN and SWAN. During the 2017 SEAN sampling, the historic MWP site Nahku Bay East Side was sampled due to its proximity to KLGO. Additionally, two historic MWP sites located in Cook Inlet were sampled in 2018 in collaboration with the SWAN/GWA monitoring program. In this report, the MWP data from southern Alaska is used to bring a historic perspective to the SEAN and SWAN data, and provide further data to contextualize the NPS results.

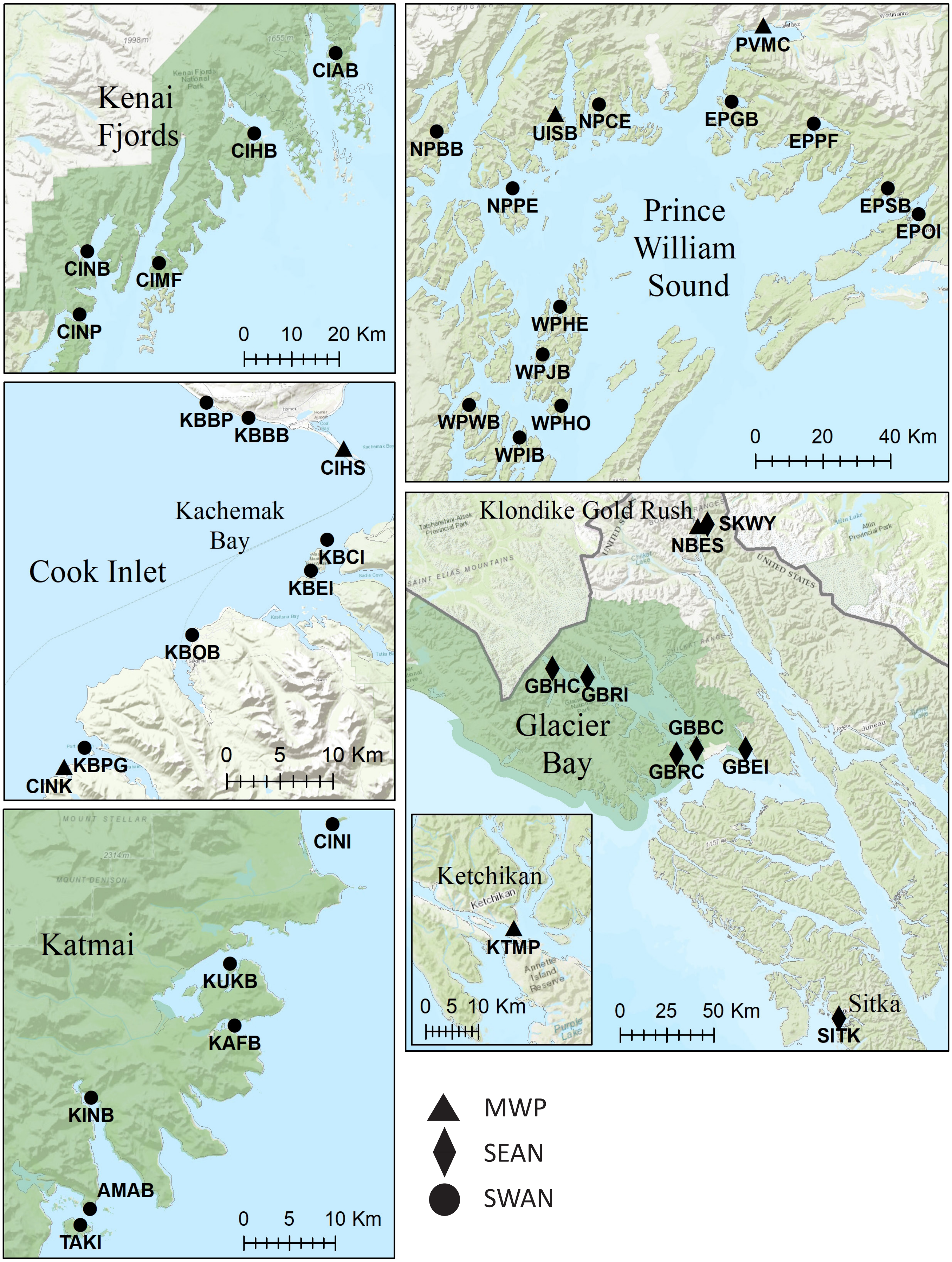

Figure 4. Site maps showing the combined site locations for Mussel Watch Program (MWP), Southwest Alaska Network (SWAN) Southeast Alaska Network (SEAN) and their partners.

Table 1. Description of 42 Mussel Watch Program (MWP), Southeast Alaska Network (SEAN) and Southwest Alaska Network (SWAN) and partner sites and the sampling years included in this report. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Map Code	Site Name	Program	Latitude	Longitude	1986	1987	1988	1990	1991	1992	1993	1995	1997	1999	2001	2003	2005	2007	2009	2012	2013	2015	2017	2018
CIHS	CI Homer Spit	MWP	59.61450	-151.44417								●	●	●	●	●	●	●						●
CINK	CI Nanwalek	MWP	59.35800	-151.93000														●						●
KTMP	Ketchikan Mountain Point	MWP	55.29383	-131.54800								●	●	●	●	●	●	●	●					
NBES	Nahku Bay East Side	MWP	59.45333	-135.33650								●	●	●	●	●	●	●	●				●	
PVMC	Port Valdez Mineral Creek Flats	MWP	61.13283	-146.46100	●	●	●	●	●	●	●	●	●	●	●	●	●	●						
UISB	Unakwit Inlet Siwash Bay	MWP	60.96083	-147.64600	●	●	●	●	●	●	●	●	●	●	●	●	●	●						
GBBC	GB Bartlett Cove	SEAN	58.44984	-135.89995																	●	●	●	
GBEI	GB Lower Excursion Inlet	SEAN	58.38483	-135.46693																	●	●	●	
GBHC	GB W. Hazelton Camp	SEAN	58.99271	-136.99306																	●	●	●	
GBRC	GB Ripple Cove	SEAN	58.45086	-136.08684																	●	●	●	
GBRI	GB E. of Russell Island	SEAN	58.91107	-136.69620																	●	●	●	
SITK	Sitka Visitor's Center	SEAN	57.04780	-135.32060																	●	●	●	
SKWY	Skagway Harbor	SEAN	59.44890	-135.32188																			●	
AMAB	KA Amalik Bay	SWAN	58.07922	-154.46600														●						●
CIAB	KF Aailik Bay	SWAN	59.87664	-149.63289														●						●
CIHB	KF Harris Bay	SWAN	59.73766	-149.95837														●						●
CIMF	KF McCarty Fjord	SWAN	59.50853	-150.34178														●						●
CINB	KF Nuka Bay	SWAN	59.53723	-150.60713														●						●
CINI	KA Ninagiak Island	SWAN	58.45150	-154.01430														●						
CINP	KF Nuka Passage	SWAN	59.42071	-150.64696														●						●
EPGB	EPWS Galena Bay	SWAN	60.94349	-146.67668																●				
EPOI	EPWS Observation Island	SWAN	60.59523	-145.73479																●				
EPPF	EPWS Port Fildalgo	SWAN	60.86214	-146.24434																●				
EPSB	EPWS Simpson Bay	SWAN	60.67212	-145.88406																●				
KAFB	KA Kaflia Bay	SWAN	58.25694	-154.19769														●						●
KBBB	KB Bishop's Beach	SWAN	59.64278	-151.60178																				●
KBBF	KB Bluff Point	SWAN	59.65670	-151.67117																				●
KBCI	KB Cohen Island	SWAN	59.53913	-151.47695																●				●
KBEI	KB Elephant Island	SWAN	59.51396	-151.50588																				●
KBOB	KB Outside Beach	SWAN	59.46423	-151.70850																●				●
KBPG	KB Port Graham	SWAN	59.37333	-151.89417																				●
KINB	KA Kinak Bay	SWAN	58.18656	-154.46575														●						●
KUKB	KA Kukak Bay	SWAN	58.31658	-154.20658														●						●
NPBB	NPWS Bettles Bay	SWAN	60.94331	-148.30196																	●			
NPCE	NPWS Cedar Bay	SWAN	60.97401	-147.40210																	●			
NPPE	NPWS Perry Island	SWAN	60.77780	-147.91574																	●			
TAKI	KA Takli Island	SWAN	58.06350	-154.48406														●						●
WPHE	WPWS Herring Bay	SWAN	60.45989	-147.71745																●				●
WPHO	WPWS Hogan Bay	SWAN	60.20197	-147.75984																●				●
WPIB	WPWS Iktua Bay	SWAN	60.13004	-147.99828																●				●
WPJB	WPWS Johnson Bay	SWAN	60.33995	-147.83482																●				●
WPWB	WPWS Whale Bay	SWAN	60.22661	-148.25105																●				●

METHODS

CHEMICAL ANALYSIS

Table 2. Distribution of contaminant groups analyzed in each year of data collection.

Years	Butyltins	Trace metals	PAHs	PBB/PBDEs	Other organic compounds
2005 & earlier	●	●	●		●
2007		●	●	● (only MWP sites)	●
2009	●	●	●		●
2012	●	●	●	●	●
2013	●	●	●		●
2015	●	●	●		●
2017	●	●	●	●	●
2018	●	●	●		●

The national MWP standard analytical protocols are applied for the SEAN and SWAN monitoring efforts. The list of chemical contaminants routinely monitored in mussel tissue samples is presented in Table 3. Chemical analyses in mussel tissue followed procedures routinely used in the NOAA NS&T Program (Kimbrough et al., 2007; Kimbrough and Lauenstein, 2007). The organic contaminants (OC) analyzed in this is study includes three Butyltins, seven Chlordanes, five Chlorobenzenes, six Dichlorodiphenyltrichloroethane (DDTs) related compounds, three Dieldrin related compounds, three Endosulfans, four Hexachlorocyclohexanes (HCHs), Mirex, Chlorpyrifos, 16 Polycyclic aromatic hydrocarbons (PAHs), 51 Polybrominated diphenyl ethers (PBDEs), 19 Polybrominated biphenyls (PBBs) and 18 Polychlorinated biphenyls (PCBs). Additionally a total of 6 major and trace metals were measured: Arsenic (As), Cadmium (Cd), Copper (Cu), Lead (Pb), Mercury (Hg) and Nickel (Ni). All metal analyses represent the "total" metals. Because of difference in programmatic funding and goals, not all of the contaminant groups were analyzed each sampling year (Table 2). An overview of each class of the chemical contaminants is provided in each section below including environmental fate and transport, and environmental health effects. Laboratory results were subjected to regular NS&T performance-based quality assessment and quality control procedures for data accuracy and precision. Data is presented in grams of contaminant per gram of dry tissue. The data in this report will be archived and publicly available at NOAA's National Centers for Environmental Information (NCEI) (https://www.ncei.noaa.gov/).

NPS scientists collecting blue mussels at the GBHC site in Glacier Bay. Credit: AK NPS

Table 3. List of chemical contaminants measured in Alaska and used to calculate total contaminant group concentrations.

Contaminant Group	Contaminants
Butyltins	Monobutyltin, Dibutyltin, Tributyltin
Chlordanes	Heptachlor, Heptachlor-Epoxide, Oxychlordane, Alpha-Chlordane, Gamma-Chlordane, Trans-Nonachlor, Cis-Nonachlor
Chlorobenzenes	1,2,3,4-Tetrachlorobenzene, 1,2,4,5-Tetrachlorobenzene, Hexachlorobenzene, Pentachloroanisole, Pentachlorobenzene
Dichlorodiphenyltrichloroethane (DDTs)	2,4'-DDD, 4,4'-DDD, 2,4'-DDE, 4,4'-DDE, 2,4'-DDT, 4,4'-DDT, DDMU
Dieldrins	Aldrin, Dieldrin, Endrin
Endolsulfans	Endosulfan I, Endosulfan II, Endosulfan Sulfate
Hexachlorocyclohexanes (HCHs)	Alpha-HCH, Beta-HCH, Delta-HCH, Gamma-HCH
Other	Mirex, Chlorpyrifos
Polycyclic aromatic hydrocarbons (PAHs)	Acenaphthene, Acenaphthylene, Anthracene, Benz(a)anthracene, Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(e)pyrene, Benzo(ghi)perylene, Benzo(kj)fluoranthene, Chrysene/Triphenylene, Dibenzo(ah)anthracene, Fluoranthene, Fluorene, Indeno(123-cd)pyrene, Phenanthrene, Pyrene
Polybrominated diphenyl ethers (PBDEs)	PBDE-1, PBDE-2, PBDE-3, PBDE-10, PBDE-7, PBDE-11, PBDE-8, PBDE-12, PBDE-13, PBDE-15, PBDE-32, PBDE-30, PBDE-17, PBDE-25, PBDE-33, PBDE-28, PBDE-35, PBDE-37, PBDE-75, PBDE-49_71, PBDE-47, PBDE-66, PBDE-77, PBDE-100, PBDE-119, PBDE-99, PBDE-116, PBDE-118, PBDE-126, PBDE-85, PBDE-155, PBDE-154, PBDE-153, PBDE-138, PBDE-166, PBDE-183, PBDE-181, PBDE-190, PBDE-202, PBDE-201, PBDE-204, PBDE-197, PBDE-198_199_203_200, PBDE-196, PBDE-205, PBDE-194, PBDE-195, PBDE-208, PBDE-207, PBDE-206, PBDE-209
Polybrominated biphenyls (PBBs)	PBB-1, PBB-2, PBB-3, PBB-4, PBB-10, PBB-7, PBB-9, PBB-15, PBB-30, PBB-18, PBB-26, PBB-31, PBB-53, PBB-52, PBB-49, PBB-103, PBB-80, PBB-77, PBB-155
Polychlorinated biphenyls (PCBs)	PCB8/5, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101/90, PCB105, PCB118, PCB128, PCB138/160, PCB153/132/168, PCB170/190, PCB180, PCB187, PCB195/208, PCB206, PCB209
Trace elements (TEs)	Arsenic (As), Cadmium (Cd), Copper (Cu), Lead (Pb), Mercury (Hg), Nickel (Ni)

DATA ANALYSIS

Data management and analysis were conducted using a combination of R version 3.4.4 (R Core Team, 2013), Microsoft Excel (2016), JMP® version 12 (SAS Institute Inc., Cary, NC, 1989-2019) and ArcGIS (ESRI, 2011). Concentrations were blank corrected by subtracting the method blank from the sample concentrations to correct for signal originating from the reagent or solvents. Concentration values for individual contaminants that were below the method detection limit (MDL) were qualified as undetected and were assigned a value of zero. Organic compounds were grouped by classes of contaminants (Table 3) and the "totals" of each group were derived as the arithmetic sum of all the individual compounds or congeners within a group. In order to visualize the data, if a site was sampled multiple times over the years, a site mean was calculated for each site. Although some of the individual contaminants and years are discussed in the results section because of temporal trends, concentration value deemed as an outlier, or toxicological importance, all of the figures and clusters are based on the mean values for each site. Individual year concentrations for each site can be found in the appendices. The site means were clustered using the Ward Hierarchical Cluster Analysis. This analysis clusters contaminant concentrations into significantly different groups such that values contained within a group are more like each other than any other value of a different group. The categories derived from the clusters were not representative measurements that have exceeded any regulatory thresholds; rather, they denoted concentrations that were significantly higher than the preceding category. Clusters were limited to three groups for easier visualizations unless one of the three clusters contained an individual site with the highest concentration, in which case it was labeled as an outlier and four clusters were used.

Contaminant concentrations in mussel samples from this study were compared to the national long-term NOAA National Status and Trend (NS&T) monitoring data for each group of contaminants. The NS&T data used for comparison comprises blue mussel tissue sample concentrations collected by the national MWP since the initiation of the program in 1986. For certain groups of contaminants, to maximize the number of analytes included in each class, earlier years with fewer analytes were removed from analysis. A maximum of three significantly different data clusters (low, medium and high) were calculated using the "mclust" package in R which uses model-based clustering which reduces the influence of outliers. Trends were analyzed using Spearman Rank Correlations on sites with more than seven data points (KTMP, NBES, PVMC, UISB, CIHS). Significance was determined as p-value <0.05 and rho >0.7. Concentrations in parts per million (ppm) wet weight were calculated using "percent dry" for samples collected after 2005 for comparison to the United States Food and Drug Administration's (US FDA) and United States Environmental Protection Agency's (US EPA) safety guidelines (US FDA, 2011). Many of these safety levels represent "the point at or above which the agency will take legal action to remove products from the market" (US FDA, 2011).

PBBs, Endosulfans, and Mirex were not detected in any of the samples for which they were tested and Chlorypyrifos was only detected five times, primarily in older MWP samples. For these reasons, these chemical groups were not attributed their own section, however are inlcuded in the appendix.

Results

Bear Cove, AK. Credit: NOAA

Arsenic (As)

CHEMICAL DESCRIPTION

Arsenic is a toxic metal that occurs naturally in the Earth's crust. Arsenic is found in the environment at high levels as a result of natural sources and industrial production. Products that contain arsenic include preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. In the recent past, as much as 90% of arsenic in industrial production was used for wood preservation (ATSDR, 2007a). Atmospheric sources of arsenic include smelting, fossil fuel combustion, power generation, and pesticide application.

Arsenic is toxic at high concentrations to fish, birds and plants. In animals and humans prolonged chronic exposure is linked to cancer (Goyer, 1986). Inorganic arsenic, the most toxic form, represents approximately 10% of total arsenic. Less harmful organic forms, such as arsenobetaine, predominate in seafood (Edmonds and Francesconi, 1977, 1988, 1993; Phillips, 1990; US FDA, 1993a). The MWP measures total arsenic, including both the inorganic and the organic forms. Safety guidance levels for arsenic in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

Centuries of human activities have changed the natural biogeochemical cycle of arsenic resulting in contamination of land, water, and air. Movement of arsenic to coastal and estuarine water occurs primarily from river runoff and atmospheric deposition. The major source responsible for ap-parent elevated levels of arsenic in the nation is natural crustal rock. This is important because it affects concentrations on the regional level. Concentrations of arsenic exceeding the current EPA drinking water standard (10 parts per billion) have been documented in Interior Alaska, Seward and Kenai peninsulas, Mat-Su Valley, and Anchorage (Athey et al., 2018).

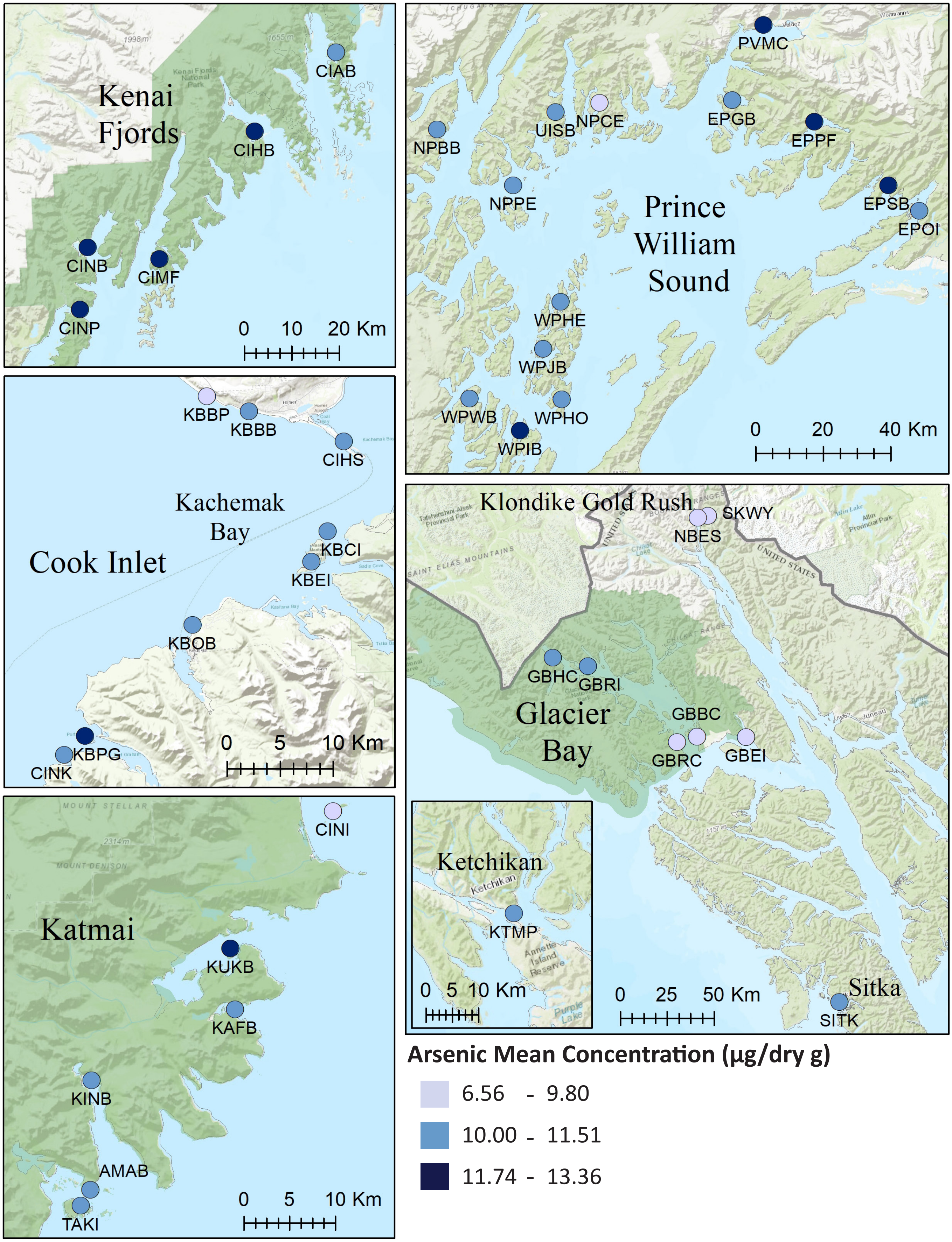

Figure 5. Mean concentration of Arsenic in mussel tissue at each site.

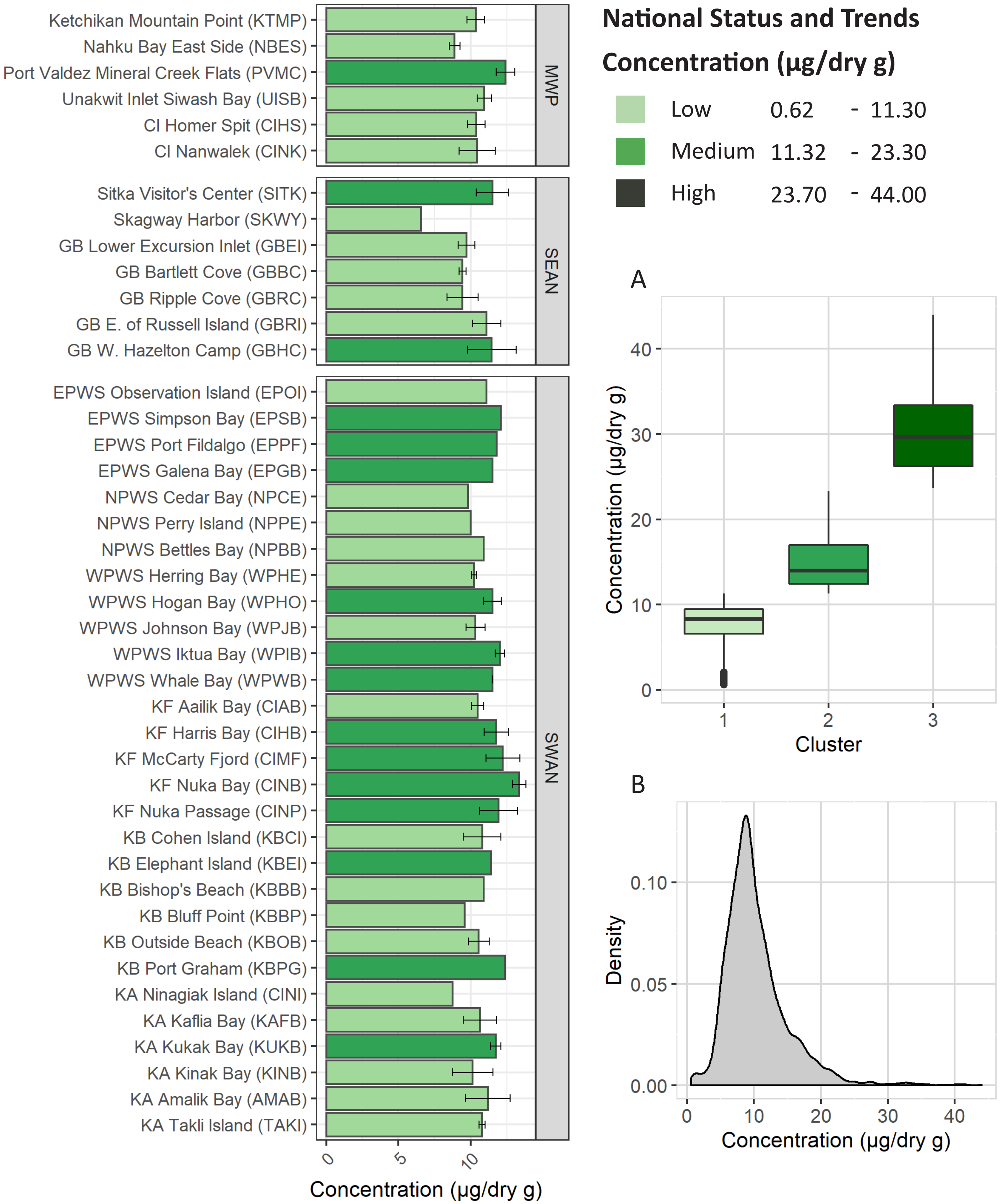

Figure 6. Bar plot showing the mean concentrations of Arsenic in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Arsenic

- Concentration range: 6.56 - 17.30 µg/dry g
- Highest mean concentration: 13.36 µg/dry g (Kenai Fjords Nuka Bay - CINB)

The highest individual year concentration was at Port Valdez Mineral Creek Flats (PVMC) in 2003 (17.3 µg/dry g) followed by PVMC in 2007 (16.7 µg/dry g) (Appendix 1). The only site trend for arsenic was at Nahku Bay East Side (NBES) which had a decreasing trendline over time (rho = -0.85, p = 0.006). However, all of the concentrations detected at NBES were within the NS&T low group (Figure 6) and the concentration range was only 2.69 µg/dry g indicating a very small decrease over time. The limited range of the arsenic concentrations and the low site based variability (Figure 5, Figure 6) suggest no particular point source of contamination but rather a background level of contamination due to sources such as volcanic sources and natural crustal rock erosion.

Cadmium (Cd)

CHEMICAL DESCRIPTION

Cadmium occurs naturally in the Earth's crust as complex oxides and sulfides in ores but is not regarded as an essential element for human life. Environmental contamination of cadmium in coastal and estuarine environments can be linked to both natural and non-point anthropogenic sources (Roesijadi, 1984). Natural sources can be linked to river runoff from cadmium rich soils, leaching from bedrock, and upwelling from marine sediment deposits (Sokolova et al., 2005). Industrial sources and uses include zinc, lead and copper production, electroplating and galvanizing, smelting, mining, fossil fuel burning, waste slag, and sewage sludge (ATSDR, 1999a; US FDA 1993b). In addition to abundant industrial applications, other products that contain cadmium include batteries, color pigment, plastics, and phosphate fertilizers. As a result of fossil fuel burning, erosion, and biological activities, cadmium becomes airborne and is transported by atmospheric processes. Land-based runoff and ocean upwelling are the main conveyors of cadmium into coastal environments.

Respiration and food represent the two major exposure pathways for humans to cadmium. Exposure to high levels occurs primarily as a result of occupational exposure. Cadmium is toxic to fish, especially salmonoid species and juveniles, and chronic exposure can result in reductions in growth. Safety guidance levels for cadmium in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

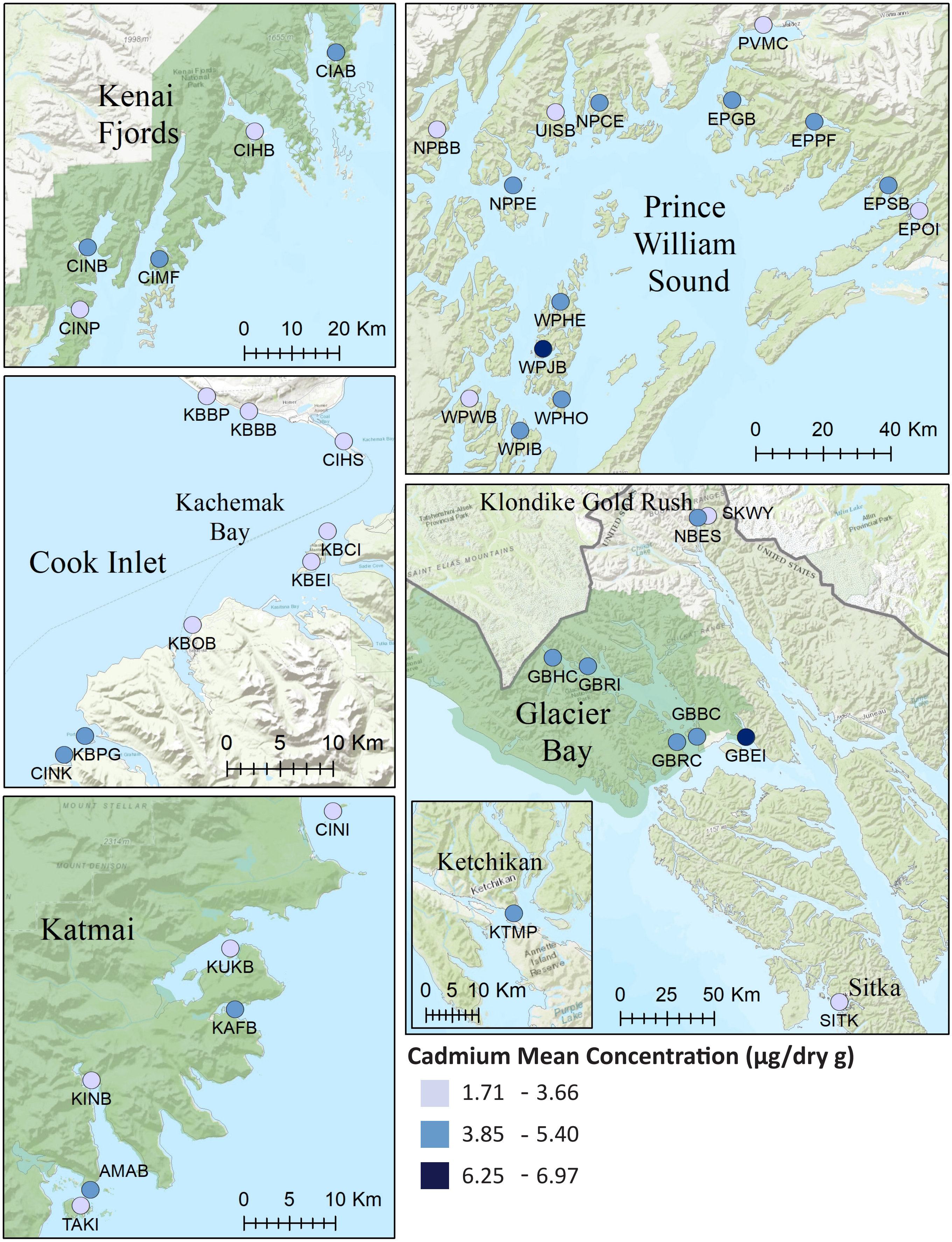

Figure 7. Mean concentration of Cadmium in mussel tissue at each site.

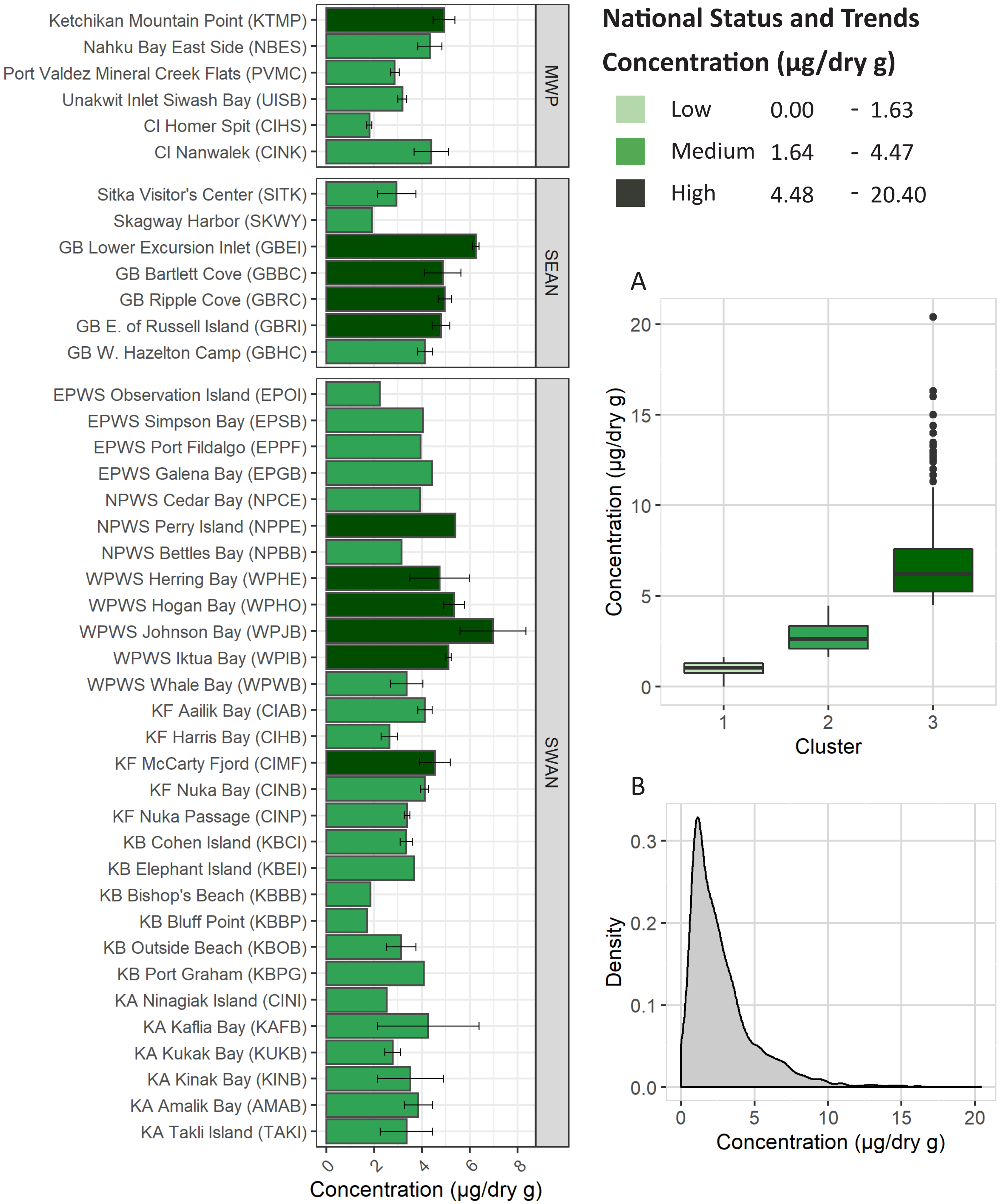

Figure 8. Bar plot showing the mean concentrations of Cadmium in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Cadmium

- Concentration range: 1.37 - 8.34 µg/dry g
- Highest mean concentration: 6.97 µg/dry g (Western Prince William Sound Johnson Bay - WPJB)

WPJB had both the highest individiual year concentration (8.34 µg/dry g, 2012) and the highest mean site concentration (6.97 µg/dry g) (Appendix 1). All of the mean site concentrations fell within the NS&T medium and high groups (Figure 8) indicating a regionwide persistent souce of cadmium. There were no site trends for cadmium. The presence of cadmium at every site suggests that natural sources are driving concentrations with some more localized influences potentially affecting a small number of sites, such as WPJB, thereby creating the minimal observed site and regional variability (Figure 7, Figure 8).

NPS scientists collecting blue mussels at the GBRI site in Glacier Bay. Credit: AK NPS

Copper (Cu)

CHEMICAL DESCRIPTION

Copper is a naturally occurring element that is ubiquitous in the environment. Trace amounts of copper are an essential nutrient for plants and animals. Anthropogenic sources include: mining, manufacturing, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and vehicle brake pads. The US EPA phase-out of chromated copper arsenate (CCA) wood preservatives and the 1980s restrictions on tributyltin marine antifouling paint has stimulated a transition to copper-based wood preservatives and marine antifouling paint.

Copper can be toxic to aquatic organisms; juvenile fishes and invertebrates are much more sensitive to copper than adults. Although copper is not highly toxic to humans, chronic effects of copper occur as a result of prolonged exposure to large doses and can cause damage to the digestive tract and eye irritation (ATSDR, 2004). There is no recommended US FDA safety level for copper in fish and fish products. The most common form of copper in water is Cu (II) which is mostly found bound to organic matter. Transport of copper to coastal and estuarine water occurs as a result of runoff and river transport. Atmospheric transport (Denier van der Gon et al., 2007) and deposition of particulate copper into surface waters may also be a significant source of copper to coastal waters.

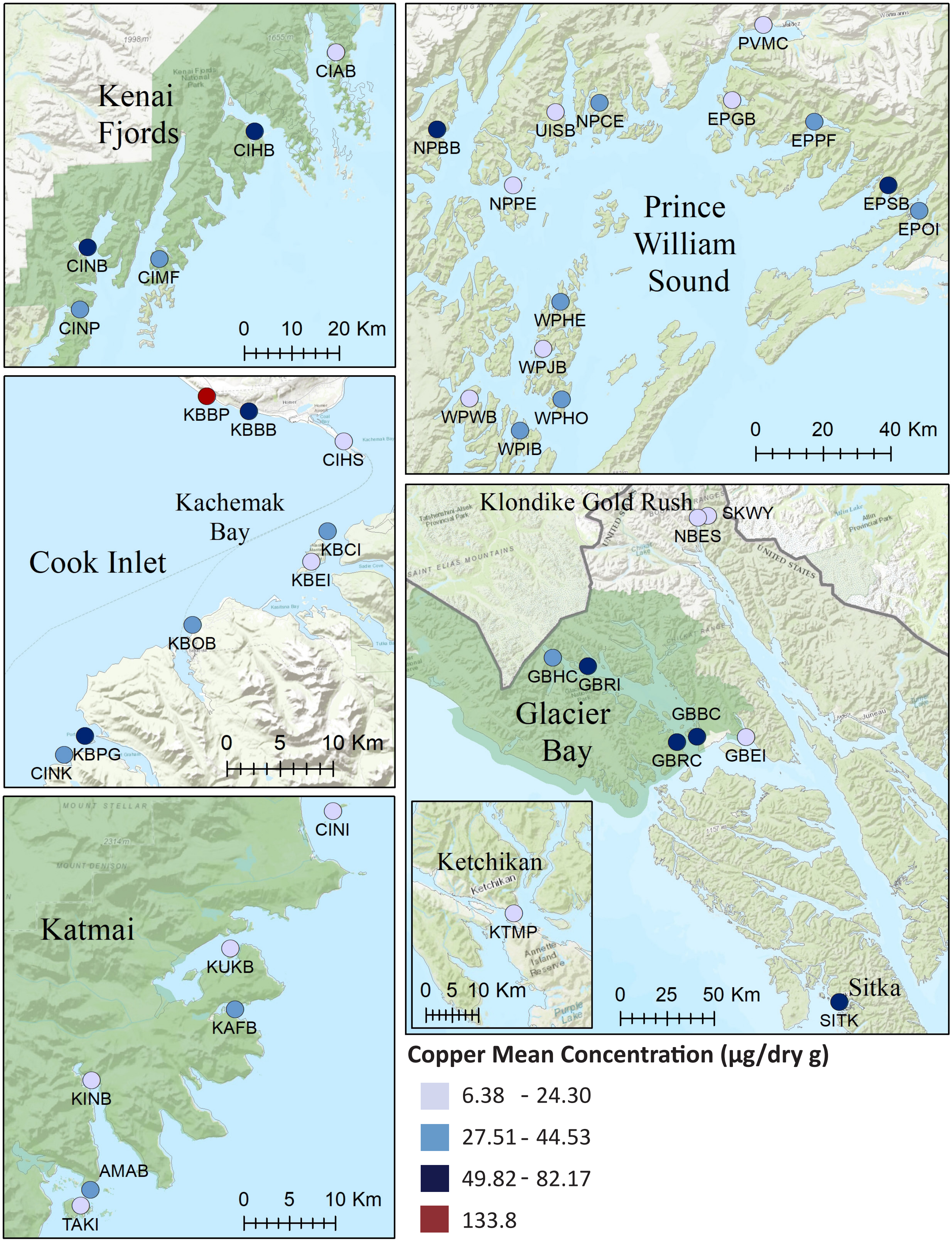

Figure 9. Mean concentration of Copper in mussel tissue at each site.

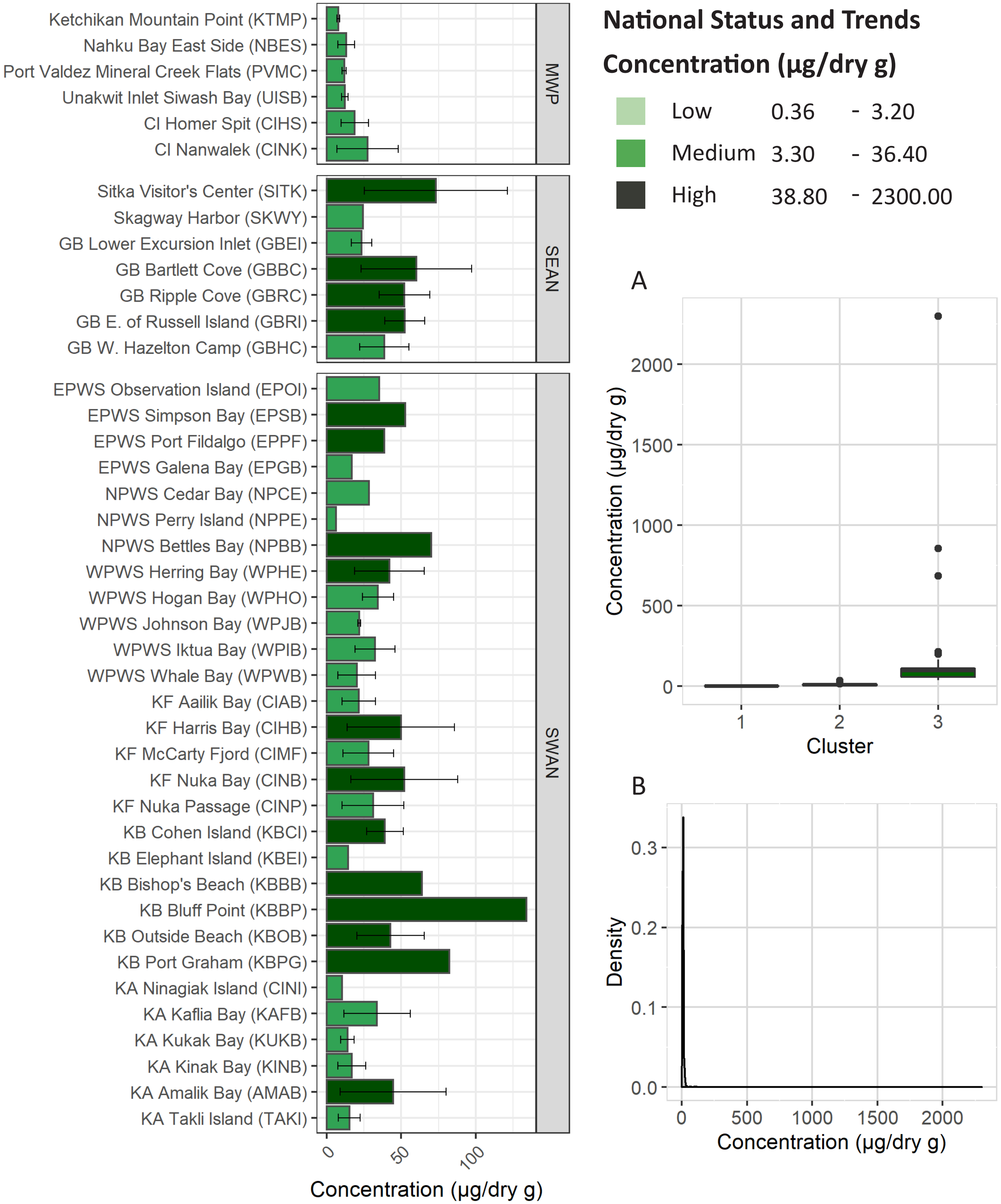

Figure 10. Bar plot showing the mean concentrations of Copper in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Copper

- Concentration range: 5.41 - 167.00 µg/dry g
- Highest mean concentration: 133.80 µg/dry g (Kachemak Bay Bluff Point - KBBP)

Copper concentrations showed greater variability within and between sites than arsenic or cadmium. However, like cadmium, all of the site means fell within the medium and high NS&T clusters (Figure 10). There were no site based trends in copper over time. KBBP was an outlier for the highest mean site concentration of copper (133.80 µg/dry g) within the Alaska data but Sitka Visitor's Center (SITK) had the highest individual year concentration at 167.00 µg/dry g (2013) followed by Glacier Bay Bartlett Cove (GBBC) at 134.00 µg/dry g (2017) (Appendix 1). There are no US FDA guideline for copper in shellfish. Although there is not enough data to calculate a trend, many of the sites that were sampled in 2007 or 2012 had increases in their copper concentrations when sampled more recently in 2018. Since copper continues to be used in both marine and terrestrial industries, it would be prudent to continue to monitor these concentrations over time.

Lead (Pb)

CHEMICAL DESCRIPTION

Lead is a ubiquitous metal that occurs naturally in the Earth's crust. Loadings of lead into coastal waters are primarily linked with wastewater discharge, river runoff, atmospheric deposition and natural weathering of rock. Lead can be found in air, soil and surface water (ATSDR, 2007b). Environmental levels of lead increased worldwide over the past century because of leaded gasoline use (ATSDR, 2007b). Significant reductions in source and load resulted from the regulation of lead in gasoline and lead based paints. High levels found in the environment are usually linked to anthropogenic activities such as manufacturing processes, paint and pigment, solder, ammunition, plumbing, incineration and fossil fuel burning. In the communications industry, lead is still used extensively as protective sheathing for underground and underwater cables, including transoceanic cable systems (USGS, 2008).

Lead has no biological use and is toxic to many organisms, including humans. Exposure of fish to elevated concentrations of lead results in neurological deformities and black fins in fish (Mance, 1987). Lead primarily affects the nervous system, which results in decreased performance and inhibits typical mental developmental in humans. Exposure to lead may also cause brain and kidney damage and cancer (IARC, 2006). Safety guidance levels for lead in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

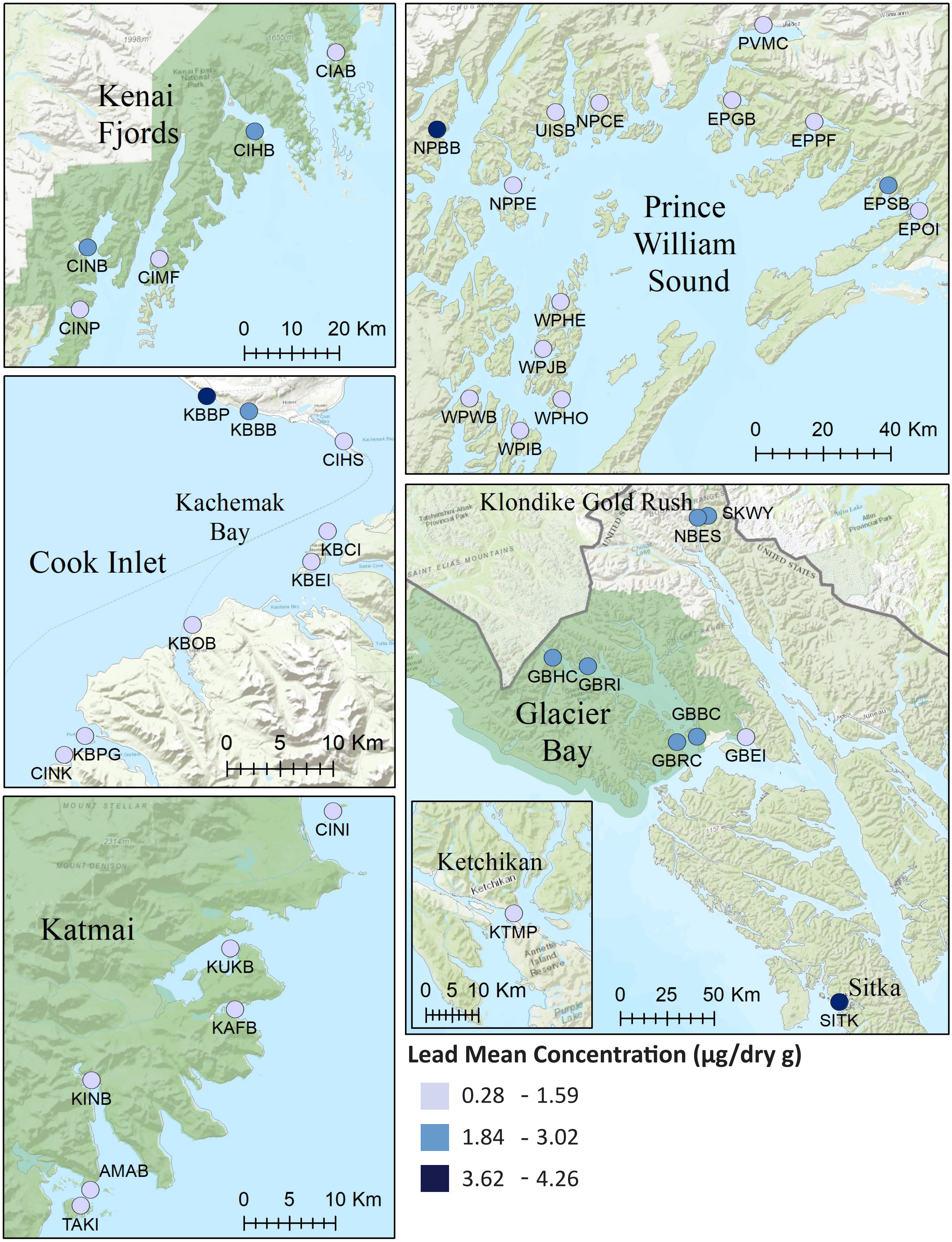

Figure 11. Mean concentration of Lead in mussel tissue at each site.

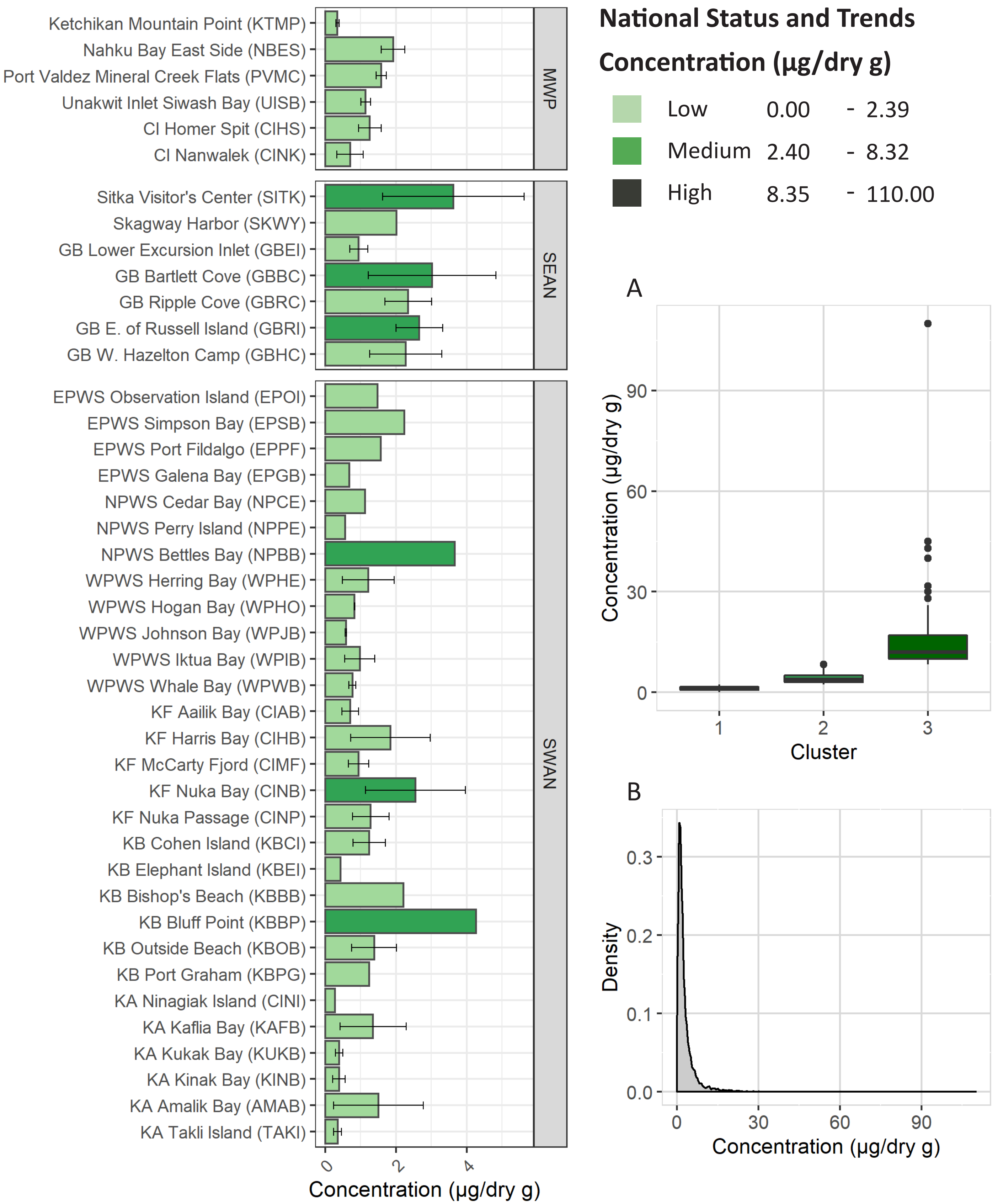

Figure 12. Bar plot showing the mean concentrations of Lead in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Lead

- Concentration range: 0.214 - 7.62 µg/dry g
- Highest mean concentration: 4.26 µg/dry g (Kachemak Bay Bluff Point - KBBP)

Similar to copper concentrations, lead concentrations showed more site based variability over time than arsenic or cadmium concentrations (Figure 12). Sitka Visitor's Center (SITK), which had the highest individual year concentrations for copper, also had the highest individual year concentration of lead (7.62 µg/dry g, 2013) and KBBP, which had the highest mean site concentration of copper, also had the highest mean concentration of lead (4.26 µg/dry g) (Appendix 1, Figure 11). Furthermore, GBBC had the second highest individual year concentration of lead (6.61 µg/dry g) (Appendix 1), as it did for copper. There were no trends in lead concentrations over time. However, like with copper concentrations, lead concentrations were higher in 2018 for a majority of those sites that were also sampled in 2007 or 2012, emphasizing the need for continued monitoring of lead concentrations.

Bald eagle in Homer Spit, AK. Credit: NOAA

Mercury (Hg)

CHEMICAL DESCRIPTION

Mercury is a highly toxic, non-essential trace metal that occurs naturally. Elevated levels occur as a result of human activity (ATSDR, 1999b). In the US, coal fired-electric turbines, municipal and medical waste incinerators, mining, landfills and sewage sludge are the primary emitters of mercury into the air. Mercury is a human neurotoxin that also affects the kidneys and developing fetuses. The most common human exposure route for mercury is the consumption of contaminated food. The US FDA has not established a safety level for mercury but has set a safety level of 1.0 ppm wet weight for methyl mercury, the form most likely to impact animal and humans (US FDA, 2011). The Mussel Watch Program measures total mercury, of which methyl mercury is only one component. Children, pregnant women or women likely to become pregnant are advised to avoid consumption of swordfish, shark, king mackerel and tilefish and should limit consumption to fish and shellfish recommended by US FDA and US EPA.

In the environment, mercury may change forms between elemental, inorganic, and organic. Natural sinks, such as sediment and soil, represent the largest source of mercury to the environment. Estimates suggest that wet and dry deposition accounts for 50-90% of the mercury load to many estuaries, making atmospheric transport a significant source of mercury worldwide (NADP, 2020).

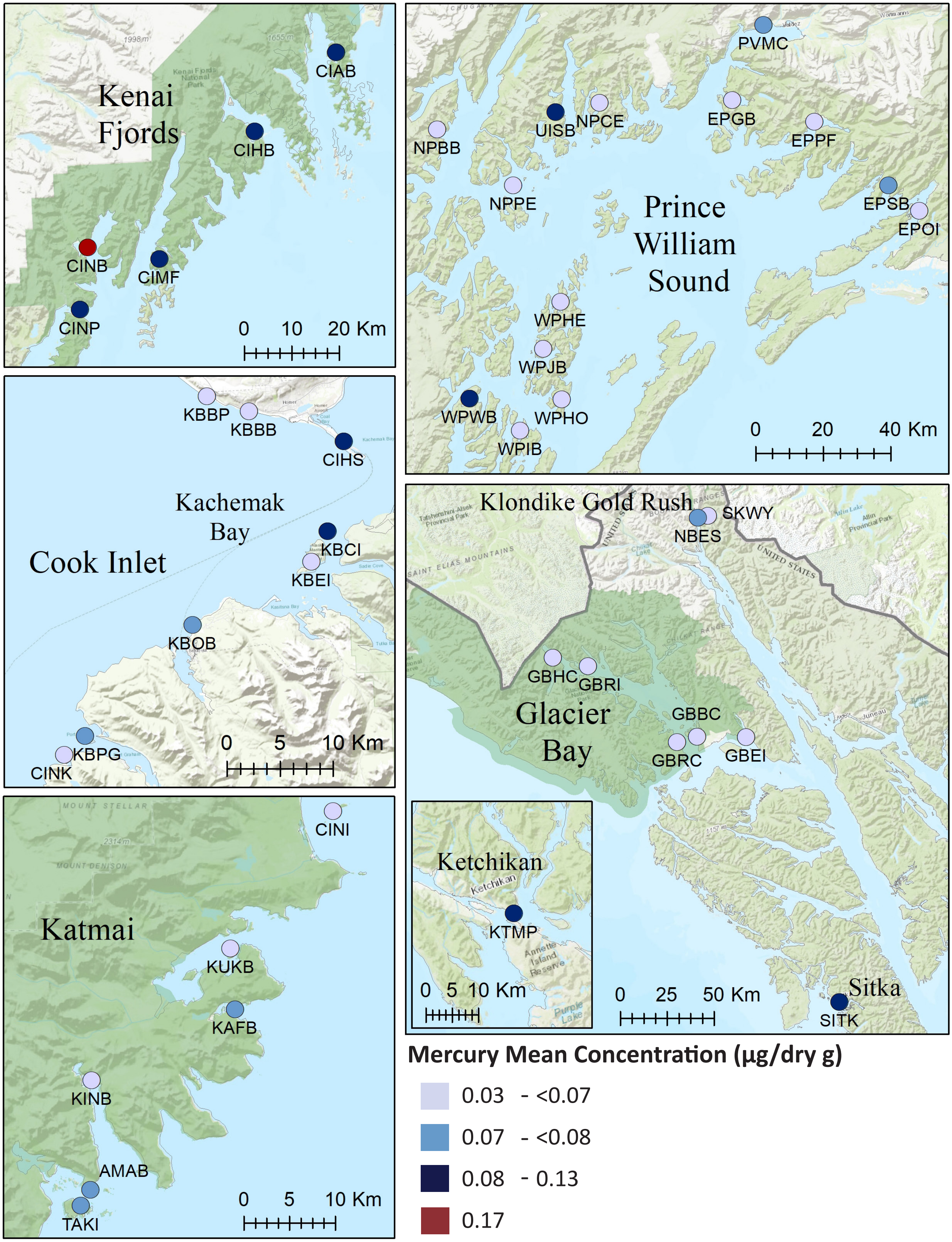

Figure 13. Mean concentration of Mercury in mussel tissue at each site.

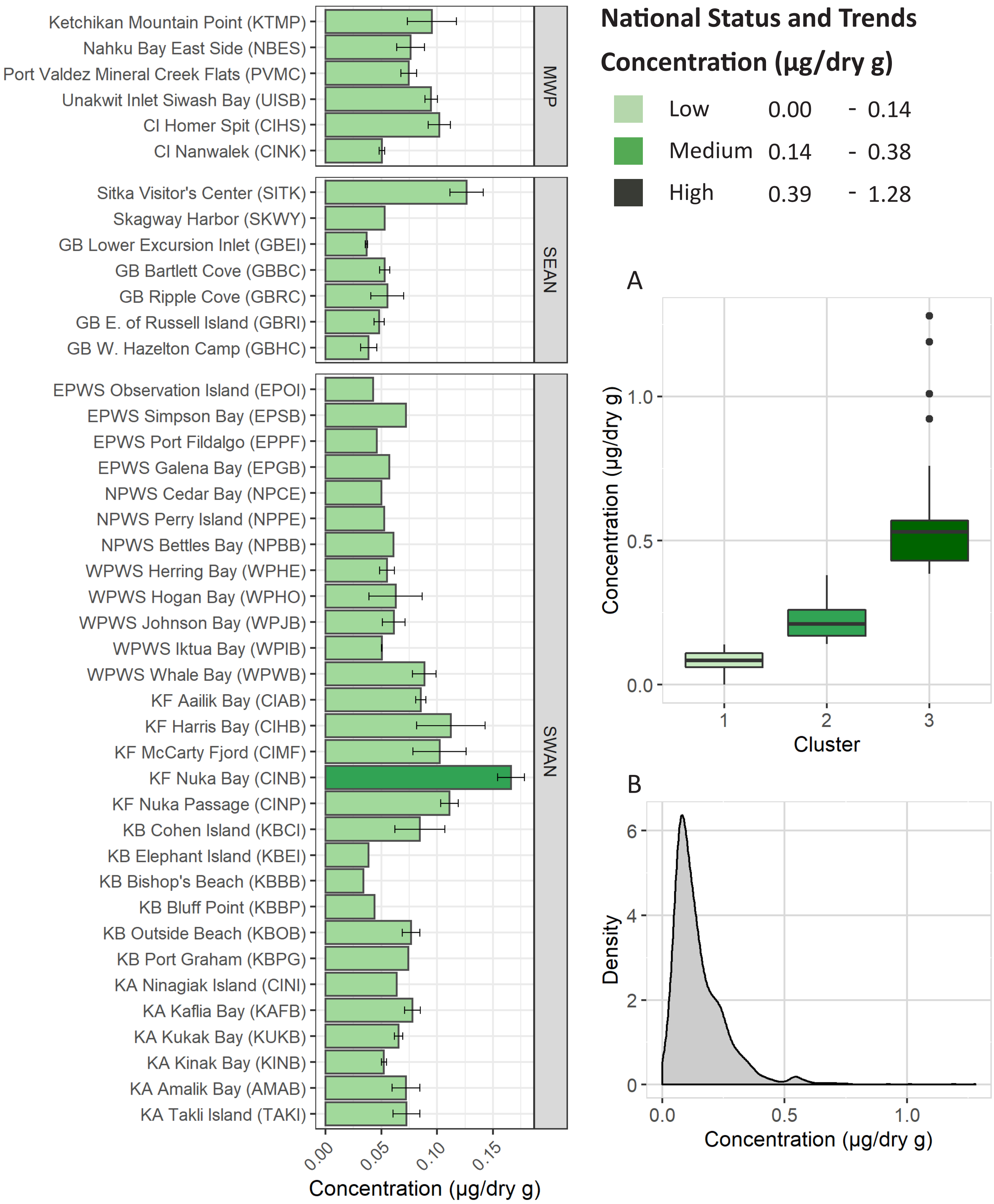

Figure 14. Bar plot showing the mean concentrations of Mercury in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Mercury

- Concentration range: 0.00 - 0.22 µg/dry g
- Highest mean concentration: 0.17 µg/dry g (Kenai Fjords Nuka Bay - CINB)

The range in mercury concentrations was very small (0.22 µg/dry g) indicating low spatial variability and the individual site concentrations of mercury showed less temporal variablility then copper and lead (Figure 14). There were no significant site trends for mercury. Ketchikan Mountain Point (KTMP) had the highest individual year mercury concentration (0.22 µg/dry g, 1999) followed by CINB (0.18 µg/dry g, 2007) (Appendix 1). CINB also had the highest mean site concentration (0.17 µg/dry g) (Figure 13). With the exception of CINB, all of the mean site concentrations of mercury were within the NS&T low cluster. Nuka Bay has a history of gold mining, which could help explain the higher concentrations of mercury found there since it is a substance traditionally used in that industry. Even though these results represent total mercury, of which methyl mercury is only one component, the highest concentration in wet weight was 0.03 ppm which is below the US FDA guidelines for methyl mercury of 1.0 ppm wet weight.

Nickel (Ni)

CHEMICAL DESCRIPTION

Nickel is a naturally occurring, biologically essential trace element that is widely distributed in the environment. It exists in alloy form in combination with other metals and as a soluble element. Nickel is found in stainless steel, nickel-cadmium batteries, pigments, computers, wire, coinage and is used for electroplating (ATSDR, 2005a). Nickel derived from weathering rocks and soil is transported to streams and rivers by runoff. It accumulates in sediment and becomes inert when it is incorporated into minerals. River and stream input of nickel are the largest sources for oceans and coastal waters. Atmospheric sources are usually not significant, except in the Great Lakes where the atmospheric input of nickel accounts for 60-80% of the total anthropogenic input to Lake Superior, and 20-70% of total inputs to Lakes Erie and Ontario (Nriagu et al., 1995).

Food is the major source of human exposure to nickel (ATSDR, 2005a). Exposure to large doses of nickel can cause serious health effects, such as bronchitis, while long-term exposure can result in cancer. There is no evidence that nickel biomagnifies in the food chain (McGeer et al., 2003; Suedel et al.,1994). Safety guidance levels for nickel in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

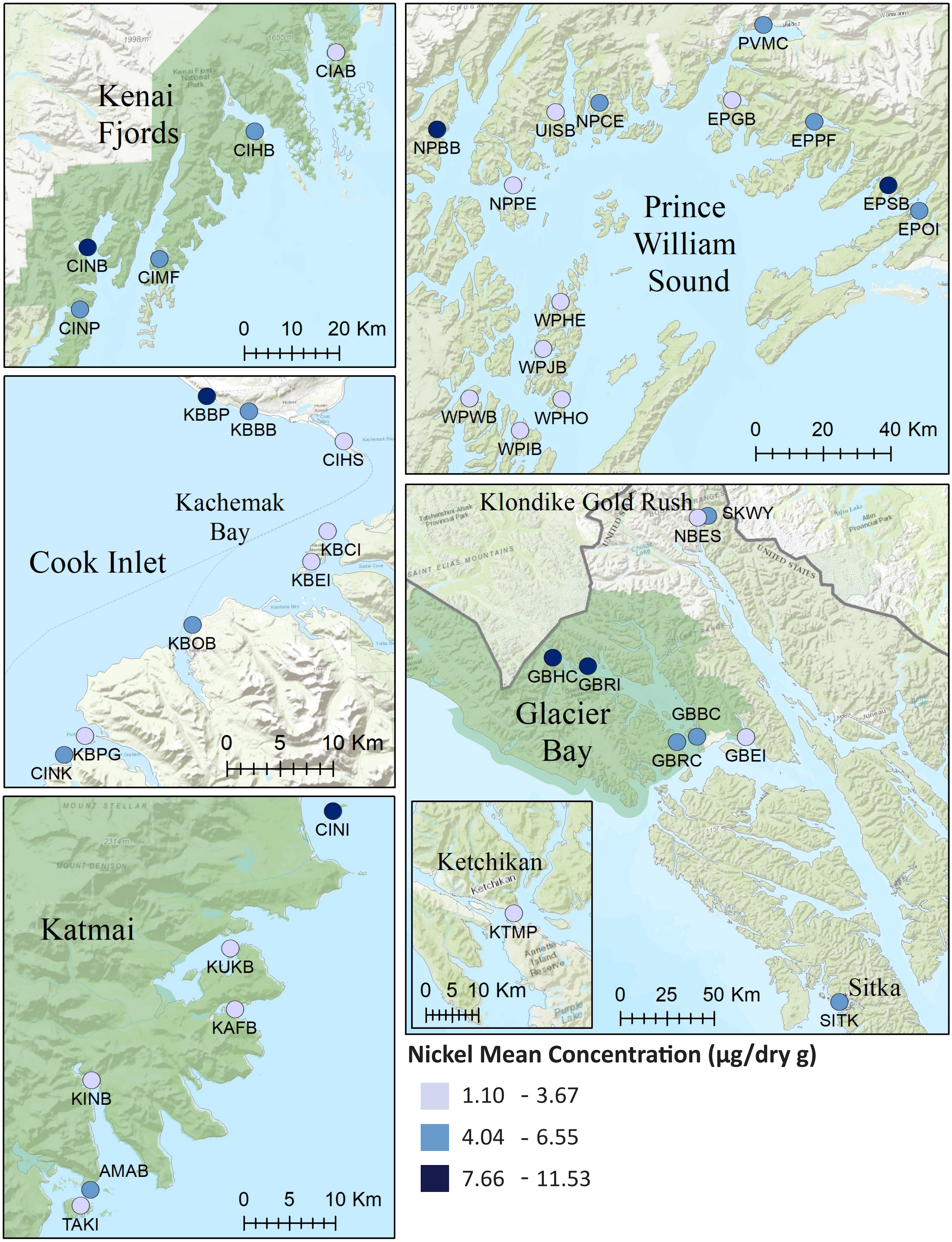

Figure 15. Mean concentration of Nickel in mussel tissue at each site.

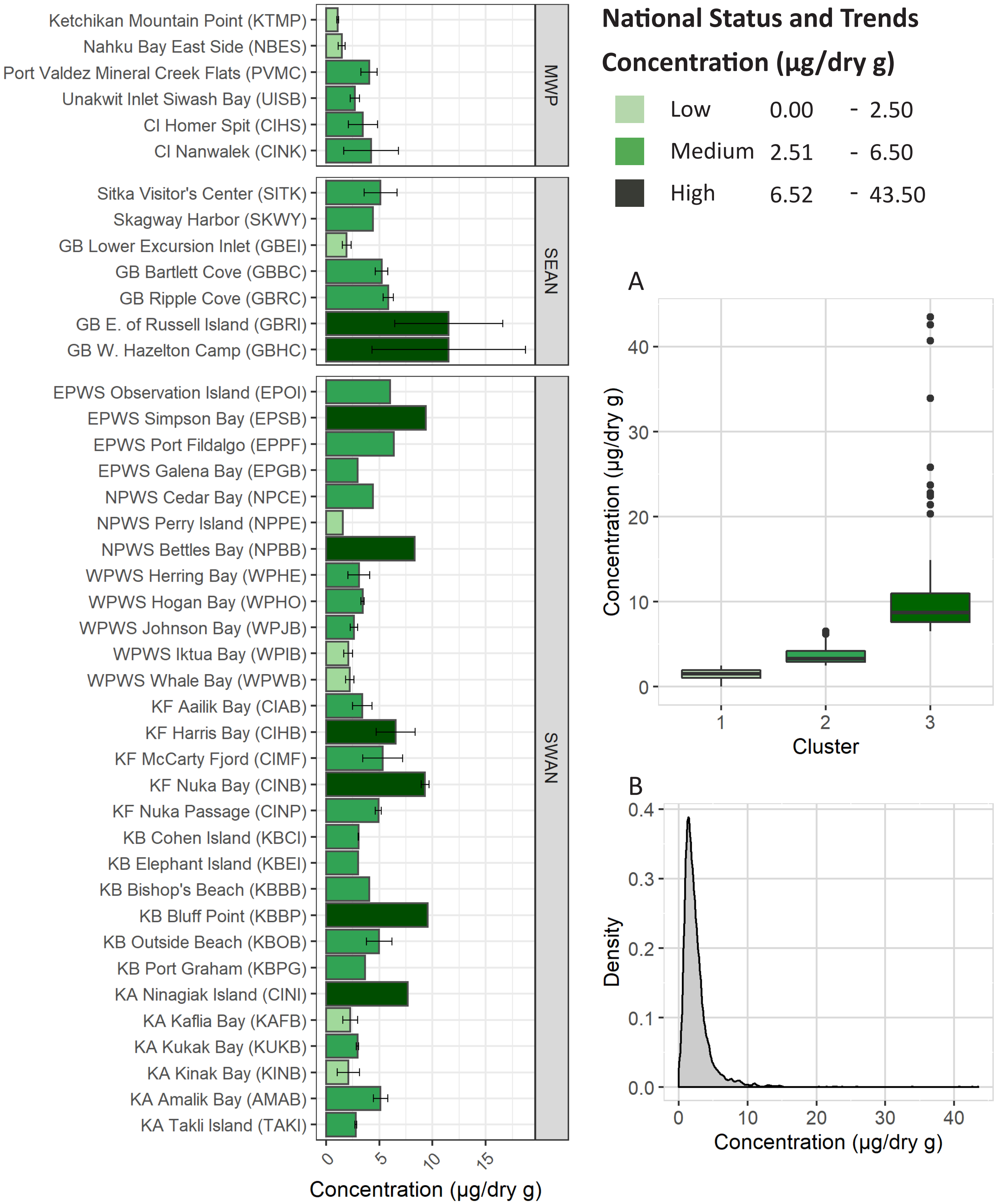

Figure 16. Bar plot showing the mean concentrations of Nickel in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Nickel

- Concentration range: 0.00 - 25.8 µg/dry g
- Highest mean concentration: 11.53 µg/dry g (Glacier Bay E. of Russell Island - GBRI)

Nickel concentrations vary both temporally and spatially with a concentration range of 25.8 µg/dry g (Figure 15, Figure 16). There were no site based temporal trends in nickel concentrations. Mean concentrations of nickel per site were mostly within the medium range of NS&T concentrations. The highest individual year concentration was 25.8 µg/dry g at Glacier Bay W. Hazelton Camp (GBHC) (2015), closely followed by 21.4 µg/dry g at GBRI (2013) (Appendix 1). GBRI had the highest mean site concentration (11.53 µg/dry g). Nickel, like cadmium and copper, appears to be elevated compared to the national NS&T concentrations.

Seward, AK. Credit: NOAA

Total Butyltins

CHEMICAL DESCRIPTION

For this document, Total Butyltins is the sum of three organometallic compounds: tributyltin (TBT), the parent compound, and two of its less toxic transformation products (dibutyltin and monobutyltin). TBT has had a variety of uses ranging from biocides in antifouling paints to catalysts and glass coatings (Bennett, 1996; Birchenough et al., 2002). These chemicals can be highly toxic in multiple forms as they naturally degrade in the environment. TBT was first shown to have biocidal properties in the 1950's (Bennett, 1996; Evans, 1970). In the late 1960s, TBT was incorporated into an antifoulant polymer paint system, quickly becoming one of the most effective paints ever used on boat hulls (Birchenough et al., 2002). In the aquatic environment, TBT is degraded by microorganisms and sunlight (Bennett, 1996). Experiments have shown that the half-life of TBT is on the order of days and degradation to monobutyltin (MBT) takes approximately a month. However, in deeper anoxic sediments, the half-life of TBT appears to be on the order of 2-4 years or longer (Batley, 1996).

The presence of TBT in the environment has been linked to endocrine disruption. In the mid-1970s, the use of TBT was linked to abnormal shell development and poor weight gain in oysters, and more recently to an imposex (females developing male characteristics) condition in marine gastropod mollusks (Batley, 1996; Strand et al., 2009). Beginning in 1989, the use of TBT as an antifouling agent was banned in the US on non-aluminum vessels smaller than 25 meters in length (Gibbs & Bryan, 1996). TBT and its metabolites continue to be detected in many components of the environment.

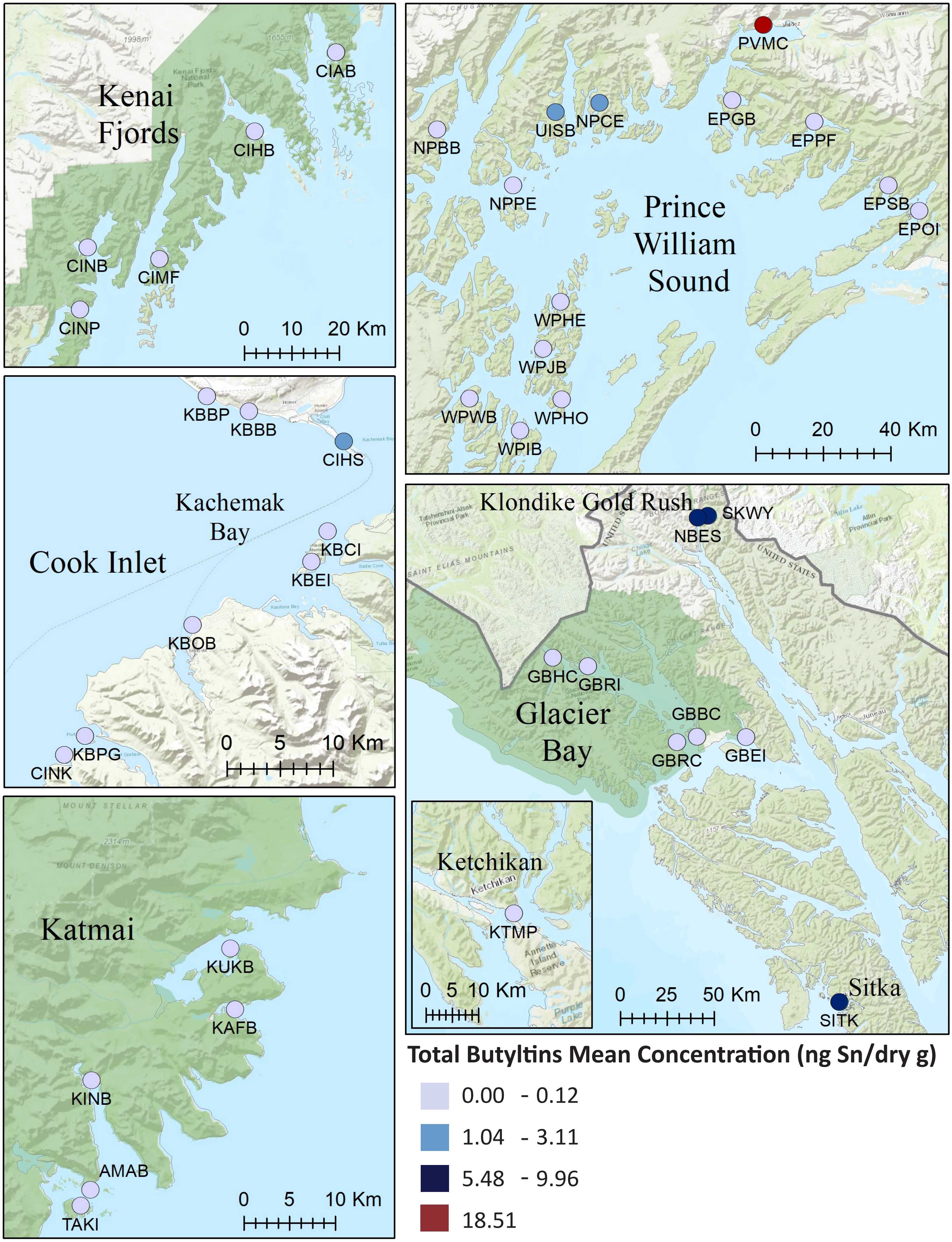

Figure 17. Mean concentration of Total Butyltins in mussel tissue at each site.

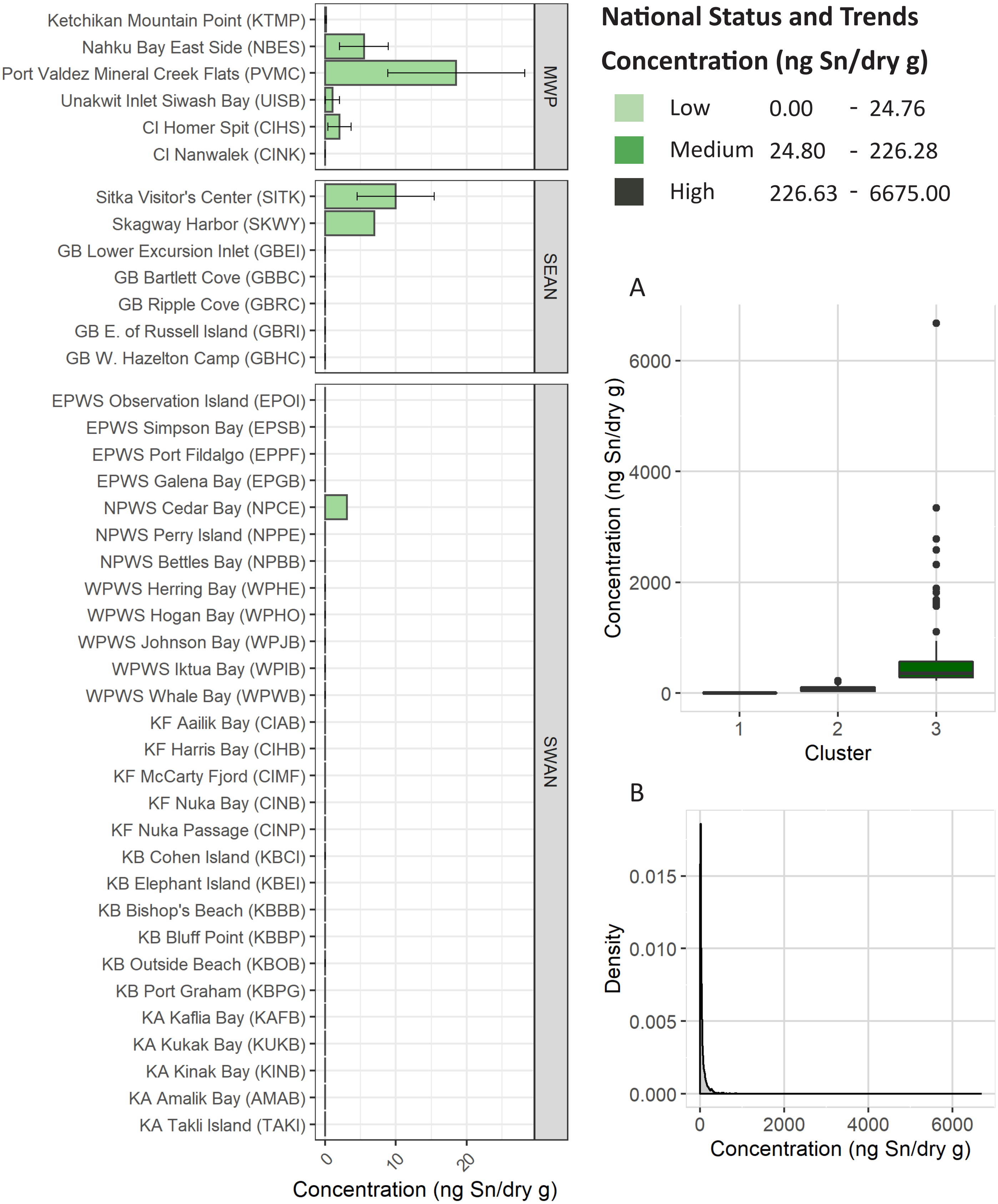

Figure 18. Bar plot showing the mean concentrations of Total Butyltins in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total Butyltins

- Concentration range: 0.00 - 107.00 ng Sn/dry g
- Highest mean concentration: 18.51 ng Sn/dry g (Port Valdez Mineral Creek Flats - PVMC)

Total Butyltins were detected at eight of the 41 sites at which they were tested and concentrations varied over both space and time (Figure 17, Figure 18). Both the highest individual year concentration (107.00 ng Sn/dry g, 1991) (Appendix 1) and highest mean site concentration (18.51 ng Sn/dry g) occurred at PVMC. Although the site mean concentration for PVMC stood out as an outlier in the Alaska data, the most recent samples from 2005 and 2007 at this site did not contain any detectable Total Butyltins, and the mean value is reflecting higher concentrations detected in the 1990's. There were no site trends for Total Butyltins, however, all of the detects at the five MWP sites analyzed for trends occured in 2003 or earlier. This data represents Total Butyltins samples starting in 1990, a year after the ban of TBT on vessels smaller than 25 meters in length. Therefore, this observation in the Total Butyltins data is probably reflecting a decreasing trend in environmental butyltins as a result of regulations. Furthermore, study-wide, only four of the 19 individual Total Butyltins detections occurred after 2003 and they were at SITK (2013, 2017), SKWY (2017), and NPCE (2013). Both SITK and SKWY are subjest to large vessel traffic that could serve as an ongoing fresh source of butyltins, as evidenced by the predominance of the parent TBT compound in these samples. All of the site mean concentrations in southern Alaska were in the NS&T low group.

Total Chlordanes

CHEMICAL DESCRIPTION

Chlordane belongs to a group of organic pesticides called cyclodienes. It is a technical mixture whose principle components are alpha-chlordane, gamma-chlordane, heptachlor, and nonachlor. Chlordane as reported here is the sum of seven prominent compounds, including: heptachlor, heptachlor-epoxide, oxychlordane, alpha-chlordane, gamma-chlordane, trans-nonachlor, and cis-nonachlor.

Technical chlordane, an insecticide, was used in the US from 1948-1983 for agricultural and urban settings to control insect pests. It was also the predominant insecticide for the control of subterranean termites. Agricultural uses were banned in 1983 and all uses were banned by 1988. These compounds are some of the most ubiquitous contaminants measured by the Mussel Watch Program. The US FDA has established a safety level of 0.3 ppm wet weight for both chlordane and heptachlor/heptachlor epoxide in all fish (US FDA, 2011).

Exposure to chlordane can occur through eating crops from contaminated soil, fish, and shellfish from contaminated waters or breathing contaminated air. Chlordane can enter the body by being absorbed through the skin, inhalation, and ingestion. At high levels, chlordane can affect the nervous system, digestive system, brain, and liver and is also carcinogenic. Chlordane is highly toxic to invertebrates and fish.

Removal from both soil and water sources is primarily by volatilization and particle-bound runoff. In air, chlordane degrades as a result of photolysis and oxidation. Chlordane exists in the atmosphere primarily in the vapor-phase, but the particle-bound fraction is important for long range transport. Chlordane is prevalent in the Arctic food web (Hargrave et al., 1992). Chlordane binds to dissolved organic matter further facilitating its transport in natural waters.

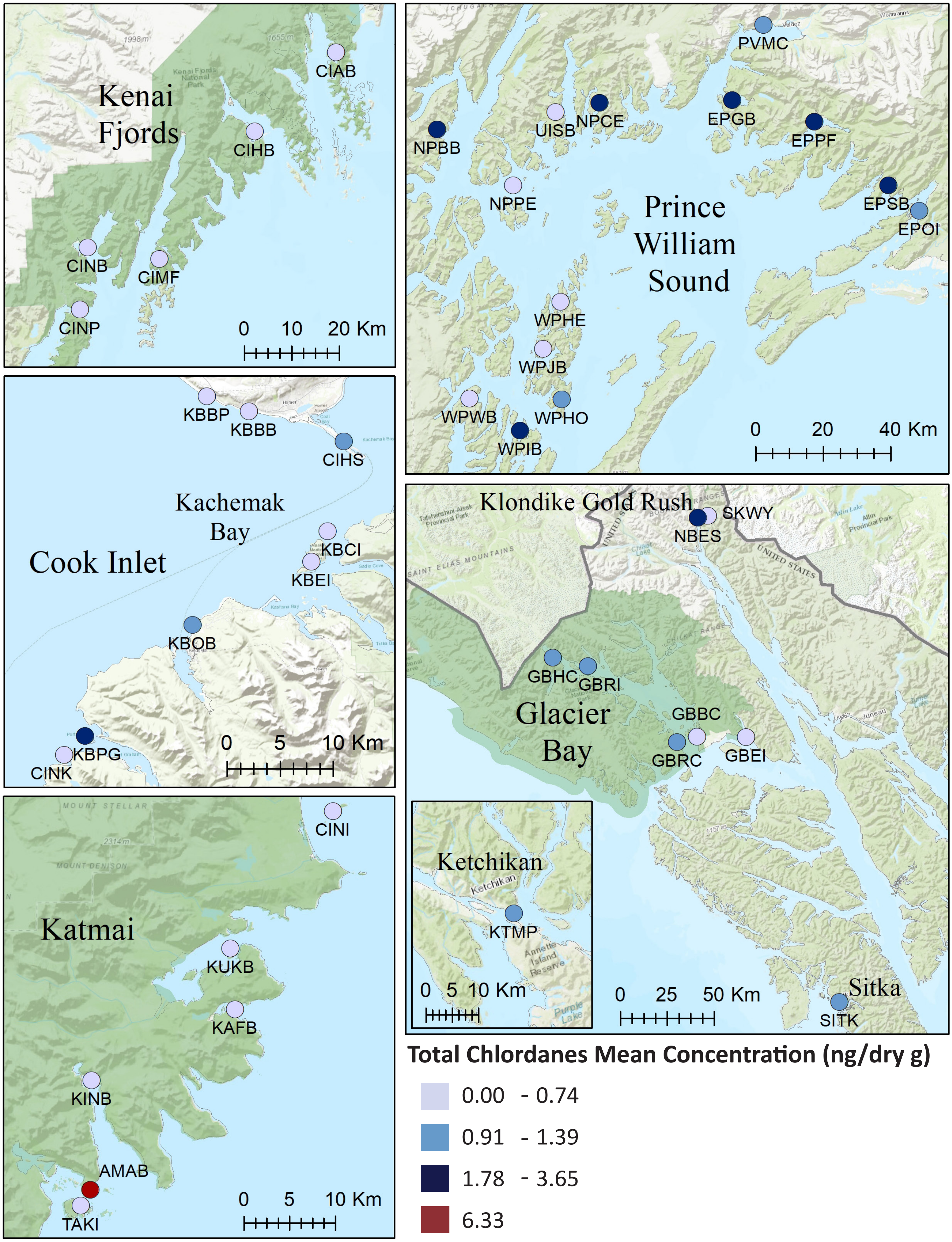

Figure 19. Mean concentration of Total Chlordanes in mussel tissue at each site.

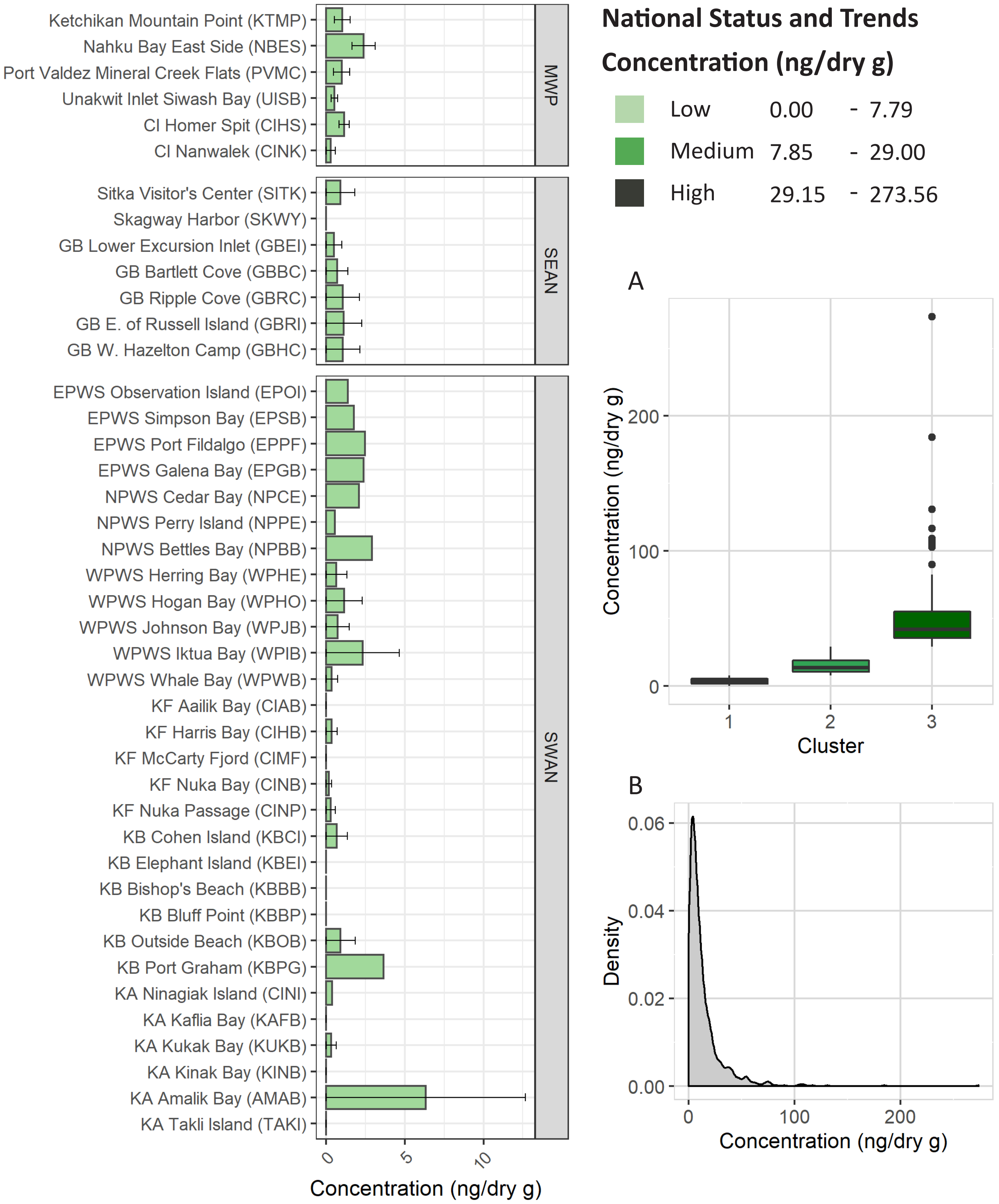

Figure 20. Bar plot showing the mean concentrations of Total Chlordanes in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total Chlordanes

- Concentration range: 0.00 - 12.65 ng/dry g
- Highest mean concentration: 6.33 ng/dry g (Katmai Amalik Bay - AMAB)

Chlordanes were detected at 33 of the 42 sites (Figure 19, Figure 20). Although nearly ubiquitous, Total Chlordane concentrations were relatively low. The highest individual year concentration (12.65 ng/dry g, 2007) (Appendix 1) as well as the highest mean concentration (6.33 ng/dry g) were detected at AMAB, but this site stood out as an outlier within the Alaska data (Figure 19). All of the mean site concentrations were within the NS&T low group and the maximum 2007 concentration found at AMAB was the only sample to fall within the NS&T medium cluster. There were no site based trends in the Total Chlordanes data. The highest concentration of chlordane in wet weight was 0.0019 ppm, well below the US FDA safety level for shellfish of 0.3 ppm wet weight.

Blue mussels in Nanwalek, AK. Credit: NOAA

Total Chlorobenzenes

CHEMICAL DESCRIPTION

Chlorobenzenes belong to the family of organic halogen compounds and are widely used as degreasers, chemical intermediates and solvents for pesticide formulations, adhesives, paints, polishes, dyes and drugs. For example, pentachloroanisole comes from the biomethylation of pentachlorophenol, a chemical used as a general biocide, fungicide, bactericide, herbicide, molluscicide, algaecide and insecticide by agriculture and other industries including textiles, paints, oil drilling and forestry (Canada, 2012). Although chlorobenzenes are not banned, due to regulations their production has decreased by 60% since the peak in 1960, due primarily to regulations on DDT in the manufacture of which it was used (ATSDR, 1990).

There is inadequate evidence to classify chlorobenzenes as carcinogens, however, animal studies indicate that livers, kidneys and the central nervous system are affected by exposure to chlorobenzenes (ATSDR, 1990). Chlorobenzenes typically rapidly evaporate into the air or are broken down by bacteria and are not considered to build up in the food chain.

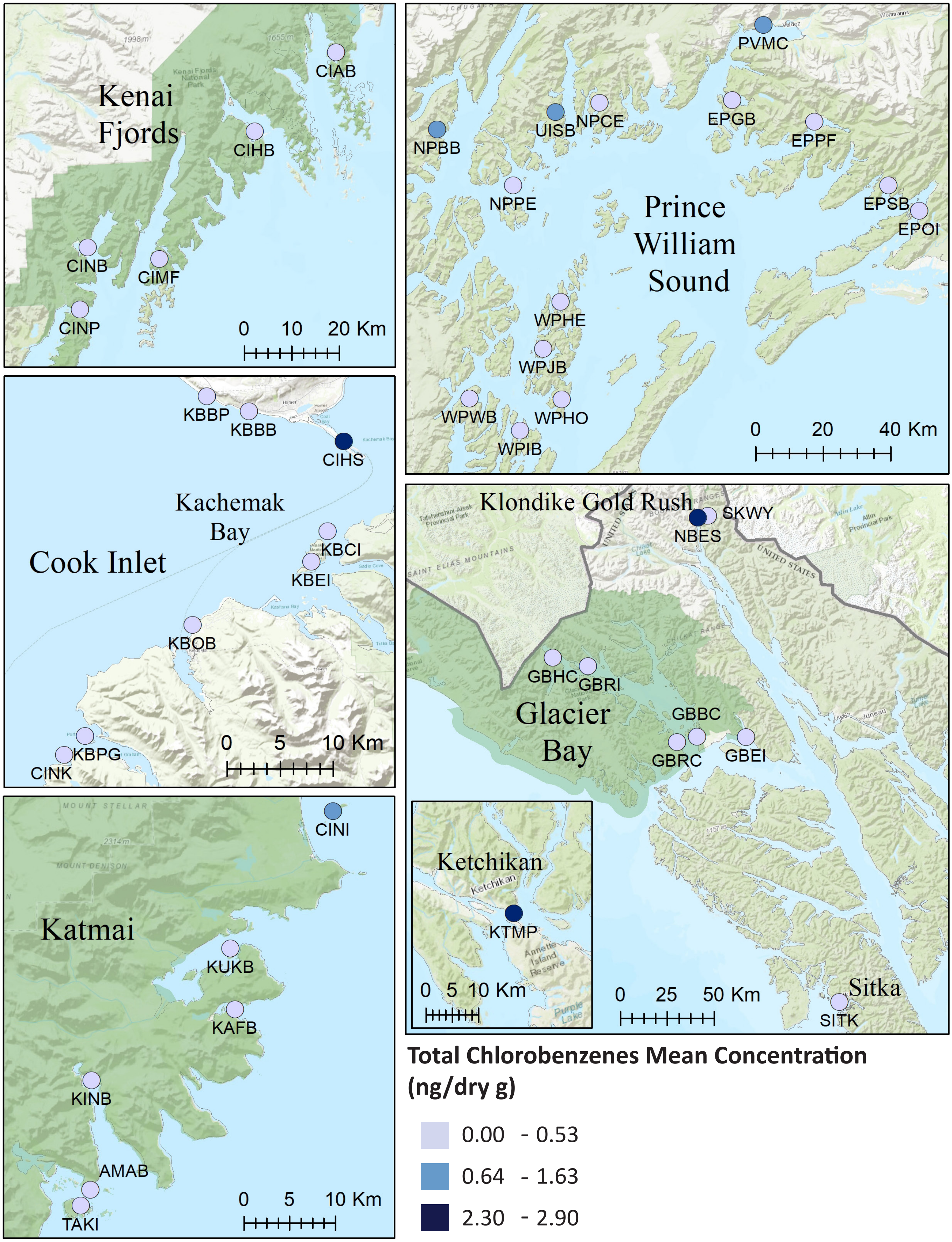

Figure 21. Mean concentration of Total Chlorobenzenes in mussel tissue at each site.

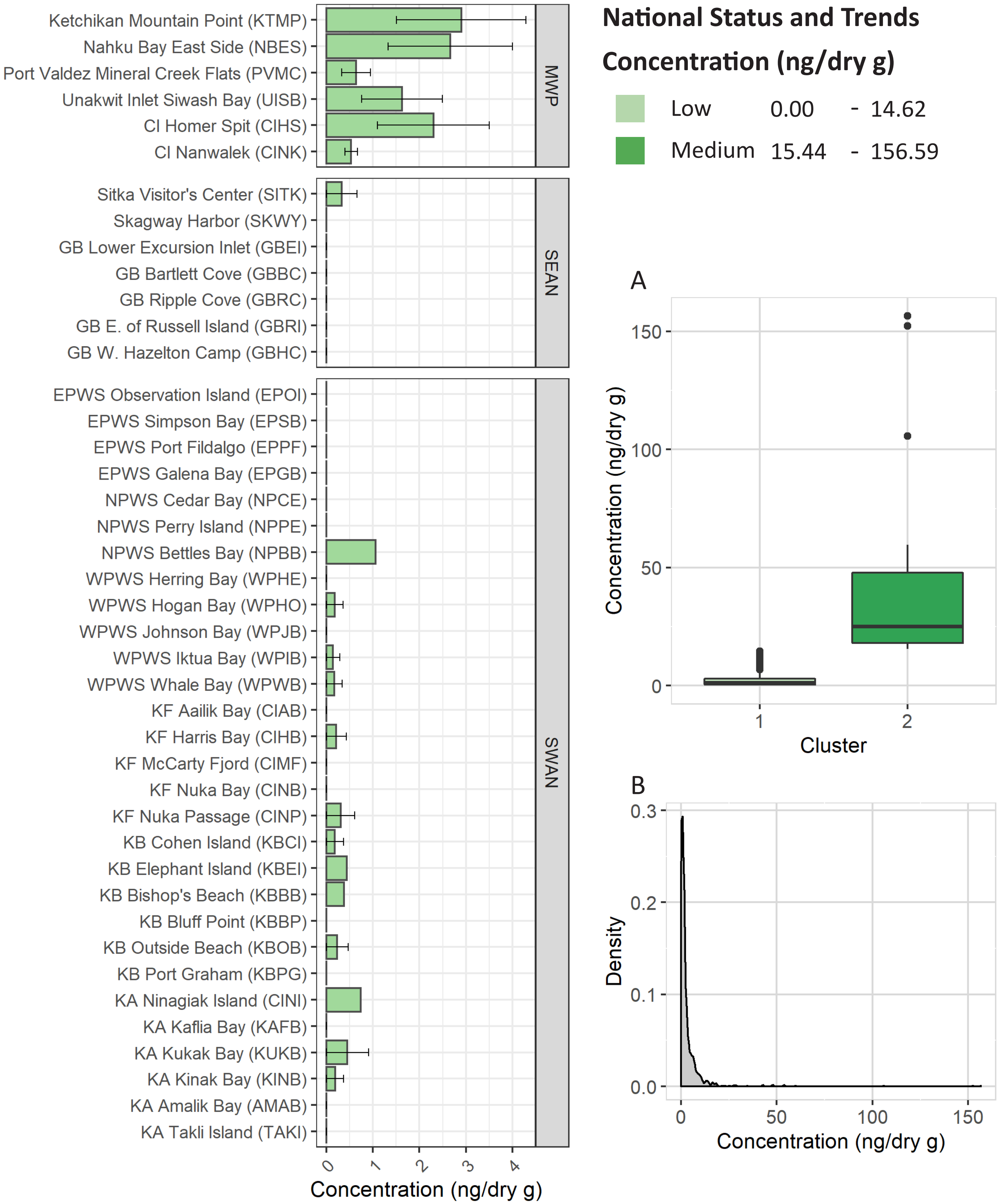

Figure 22. Bar plot showing the mean concentrations of Total Chlorobenzenes in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total Chlorobenzenes

- Concentration range: 0.00 - 10.73 ng/dry g
- Highest mean concentration: 2.90 ng/dry g (Ketchikan Mountain Point - KTMP)

Chlorobenzenes were detected at 20 of the 42 sites (Figure 21, Figure 22). There was a decreasing site trend for Total Chlorobenzenes at Cook Inlet Homer Spit (CIHS) (rho = -0.82, p = 0.02), Nahku Bay East Site (NBES) (rho = -0.92, p = 0.001), and Port Valdez Mineral Creek Flats (PVMC) (rho = -0.93, p = 0.008). The predominance of MWP sites among the highest site mean concentrations is a result of higher concentrations detected in the late 1990s and early 2000s (Appendix 1). Even so, all of the individual year concentations from this report were within the NS&T low group and therefore all of the mean site concentrations for Total Chlorobenzenes were as well. Total Chlorobenzenes concentrations in samples before 2000 were mainly comprised of 1,2,3,4-Tetrachlorobenzene and 1,2,4,5-Tetrachlorobenzene, however, since 2001 Hexachlorobenzene and Pentachloroanisol have been predominate and since 2009 only Hexachlorobenzene has been detected.

Total DDTs

CHEMICAL DESCRIPTION

Total DDTs (dichlorodiphenyltrichloroethane) is the sum of six compounds comprised of ortho and para forms of DDT and its transformation products DDE and DDD, the latter being the most predominant form found in the environment. Technical DDT, the insecticide, was composed of up to 14 compounds, of which 65-80% was the active ingredient, p,p’-DDT. The next major component, o,p’-DDT (15-21%), is nearly inactive as an insecticide. DDT was used worldwide as an insecticide for agricultural pests and mosquito control. Its use in the United States was banned in 1972, but it is still used in some countries today. Due to its persistence and hydrophobic nature, DDT bioaccumulates in organisms. Organochlorine pesticides are typically neurotoxins and DDT has been shown to interfere with the endocrine system (Rogan & Chen, 2005). DDT and its metabolite DDE were specifically linked to eggshell thinning in birds (Lincer, 1975). The US FDA has established a safety level of 5 ppm wet weight for DDT and DDE in all fish (US FDA, 2011).

DDT and its metabolites can be rapidly broken down by sunlight in the air, however in soil they are slowly broken down by microorganisms. These chemicals can bioaccumulate in the fatty tissue of animals (ATSDR, 2002b). Evaporation of DDT from soil followed by long distance transport results in its widespread global distribution, i.e. the "grasshopper" effect (Wania and Mackay, 1996). DDT that enters surface waters is subject to volatilization, adsorption to suspended particulates and sediment, and bioaccumulation. About half of the atmospheric DDT is adsorbed to particulates (Bidleman, 1988).

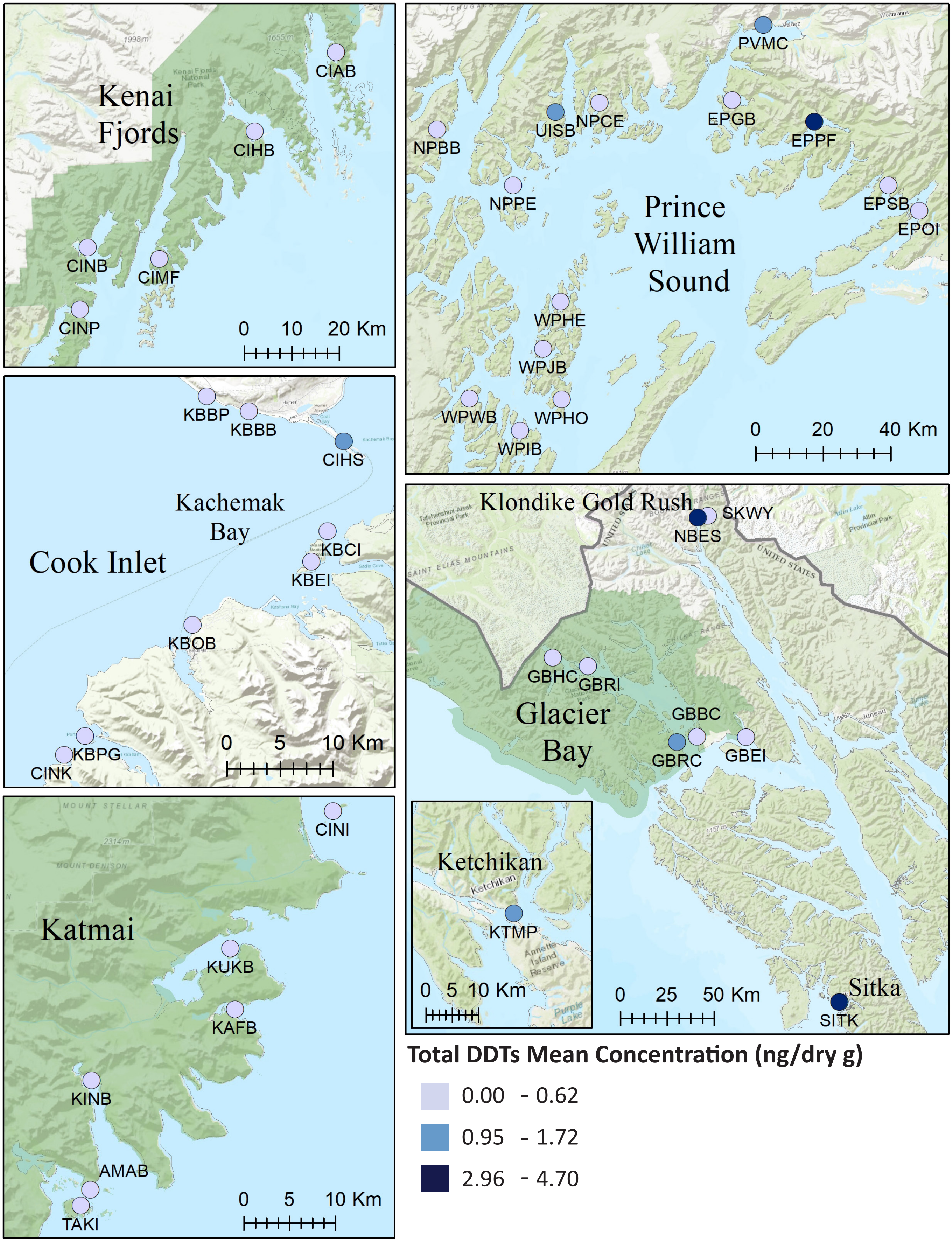

Figure 23. Mean concentration of Total DDTs in mussel tissue at each site.

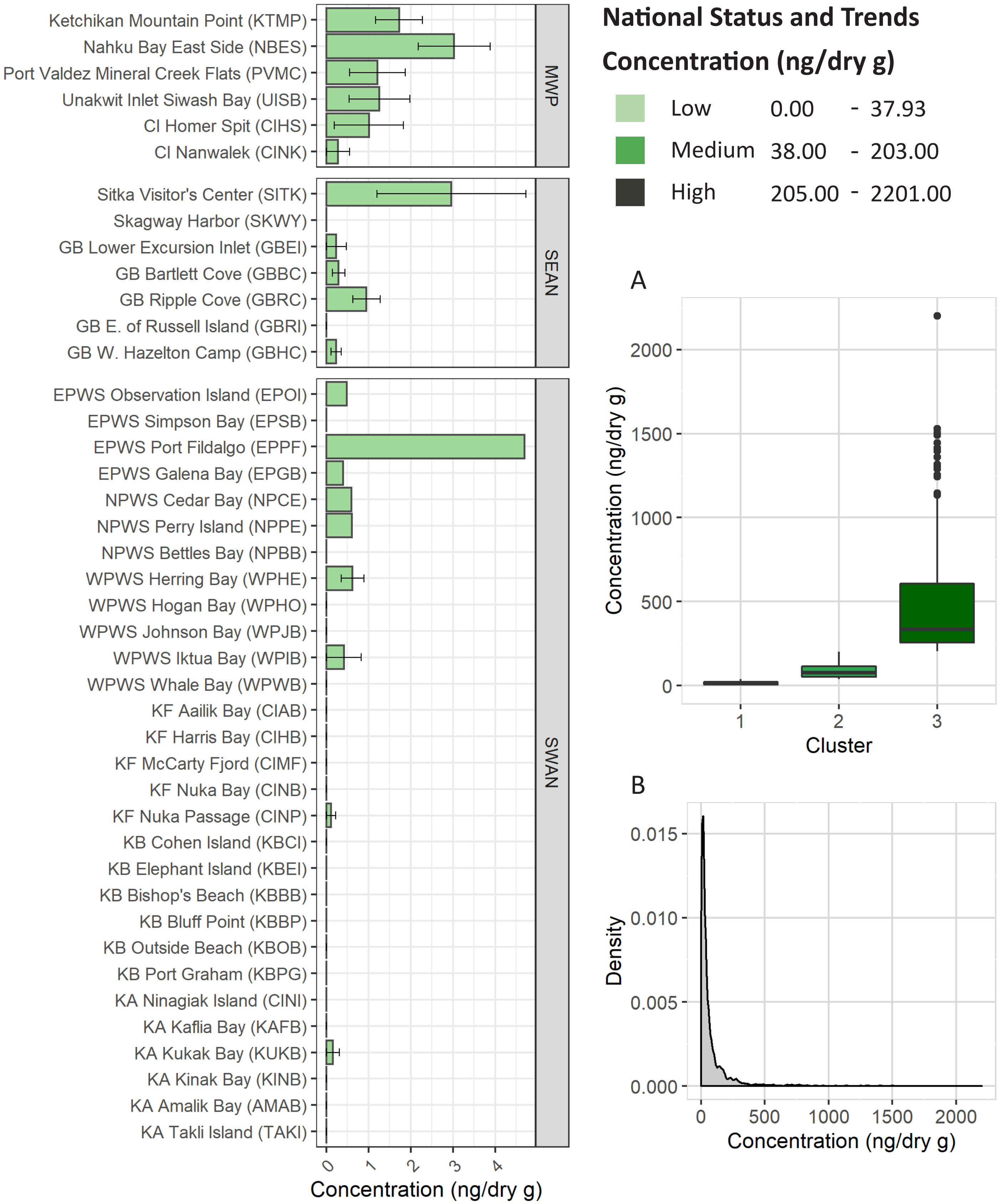

Figure 24. Bar plot showing the mean concentrations of Total DDTs in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total DDTs

- Concentration range: 0.00 - 9.27 ng/dry g
- Highest mean concentration: 4.70 ng/dry g (Eastern Prince William Sound Port Fidalgo - EPPF)

DDT and/or its breakdown products were were detected at 20 of the 42 sites (Figure 23, Figure 24). There were no site trends in Total DDT concentrations. All of the mean site concentrations fell within the NS&T low group as well as all of the individual year concentrations (Appendix 2). The highest concentration of Total DDT in wet weight was 0.00099 ppm which is well below the US FDA safety level of 5 ppm wet weight for all fish. Although the majority of the MWP samples had fresh inputs of o,p'- and p,p'-DDT prior to 2007, since that time only three sample have had detectable levels of the parent compounds: Nahku Bay East Site (NBES) in 2007 (o,p'-DDT), Eastern Prince William Sound Port Fidalgo (EPPF) in 2012 (o,p'-DDT) and Sitka Visitor's Center (SITK) in 2013 (o,p'-DDT and p,p'-DDT). However, in 2015 and 2017 only the transformation product p,p'-DDE was detected at SITK. Additionally, in only one of the 23 sites sampled in 2018 was any DDT or any of its breakdown products detected (Western Prince William Sound Herring Bay - WPHE; p,p'-DDE), suggesting that these contaminants are no longer widely present in the environment at detectable levels in mussels.

Total Dieldrins

CHEMICAL DESCRIPTION

In this document, Total Dieldrins is defined as the sum of three compounds: dieldrin, aldrin and endrin. Dieldrins were widely used as insecticides in the 1960s for the control of termites around buildings and general crop protection from insects. In 1970, all uses of dieldrins were banned based on concern that they could cause severe aquatic environmental change and and potential carcinogenicity (EPA, 1980). The ban was lifted in 1972 to allowed limited use of dieldrins, primarily for termite control. All uses of dieldrins were finally banned in 1989 (EPA, 1990).

The predominance of dieldrin in the environment can be explained by the degradation of aldrin to dieldrin in the environment by sunlight and bacteria. Additionally, aldrin rapidly changes to dieldrin in plants and animals. Dieldrins in water break down very slowly and once they enter the body, are stored in fat and leave the body very slowly. Exposure to dieldrins occurs through ingestion of contaminated water and food products, including fish and shellfish, and through inhalation of indoor air in buildings treated with these insecticides. Acute and long-term human exposures are associated with central nervous system intoxication (ATSDR, 2002a).

Because dieldrins can build up in the body, health effects can occur from long periods of exposure to smaller amounts. Aldrin and dieldrin are carcinogenic to animals and classified as likely human carcinogens. The US FDA has established a safety level of 0.3 ppm wet weight for aldrin and dieldrin in all fish (US FDA, 2011).

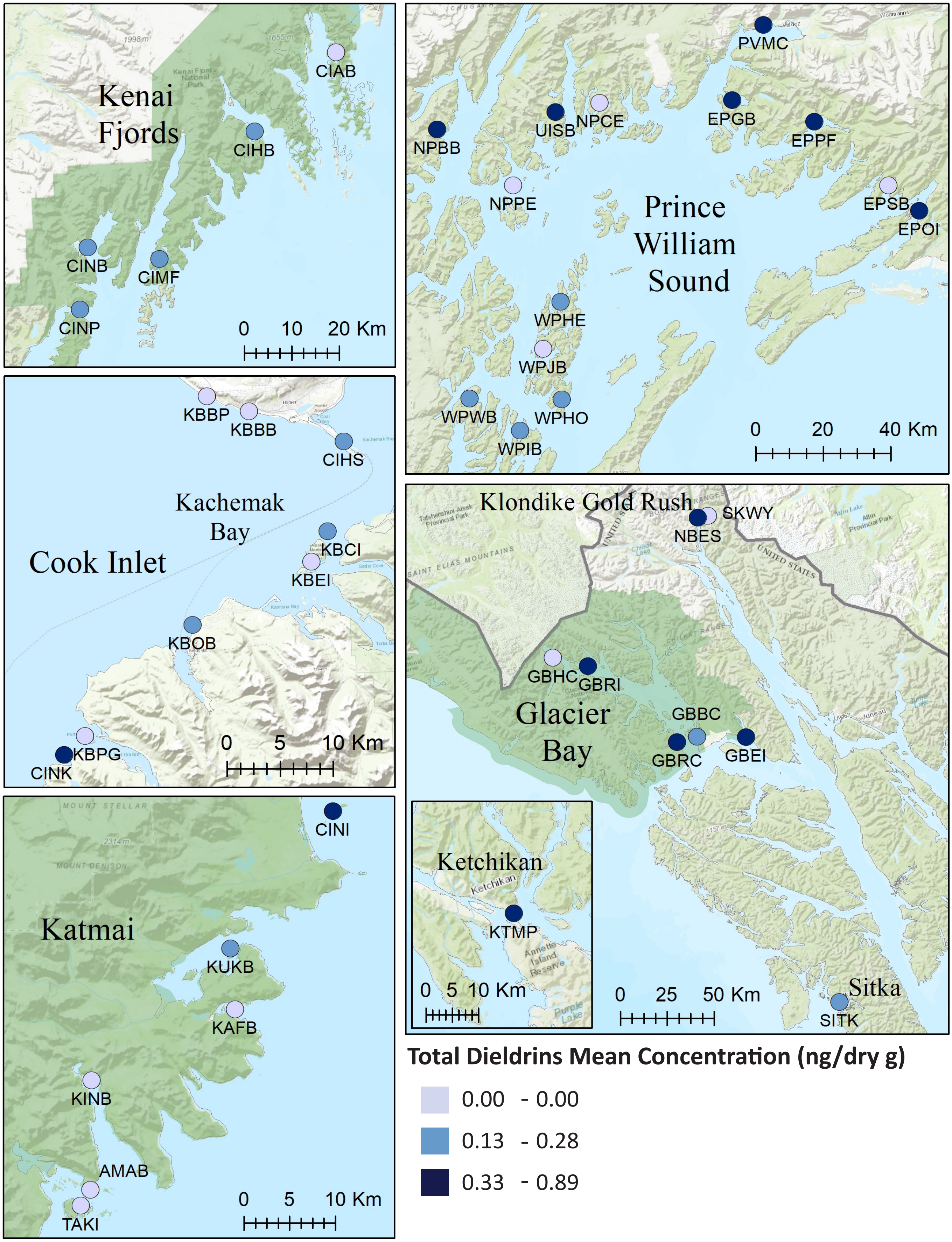

Figure 25. Mean concentration of Total Dieldrins in mussel tissue at each site.

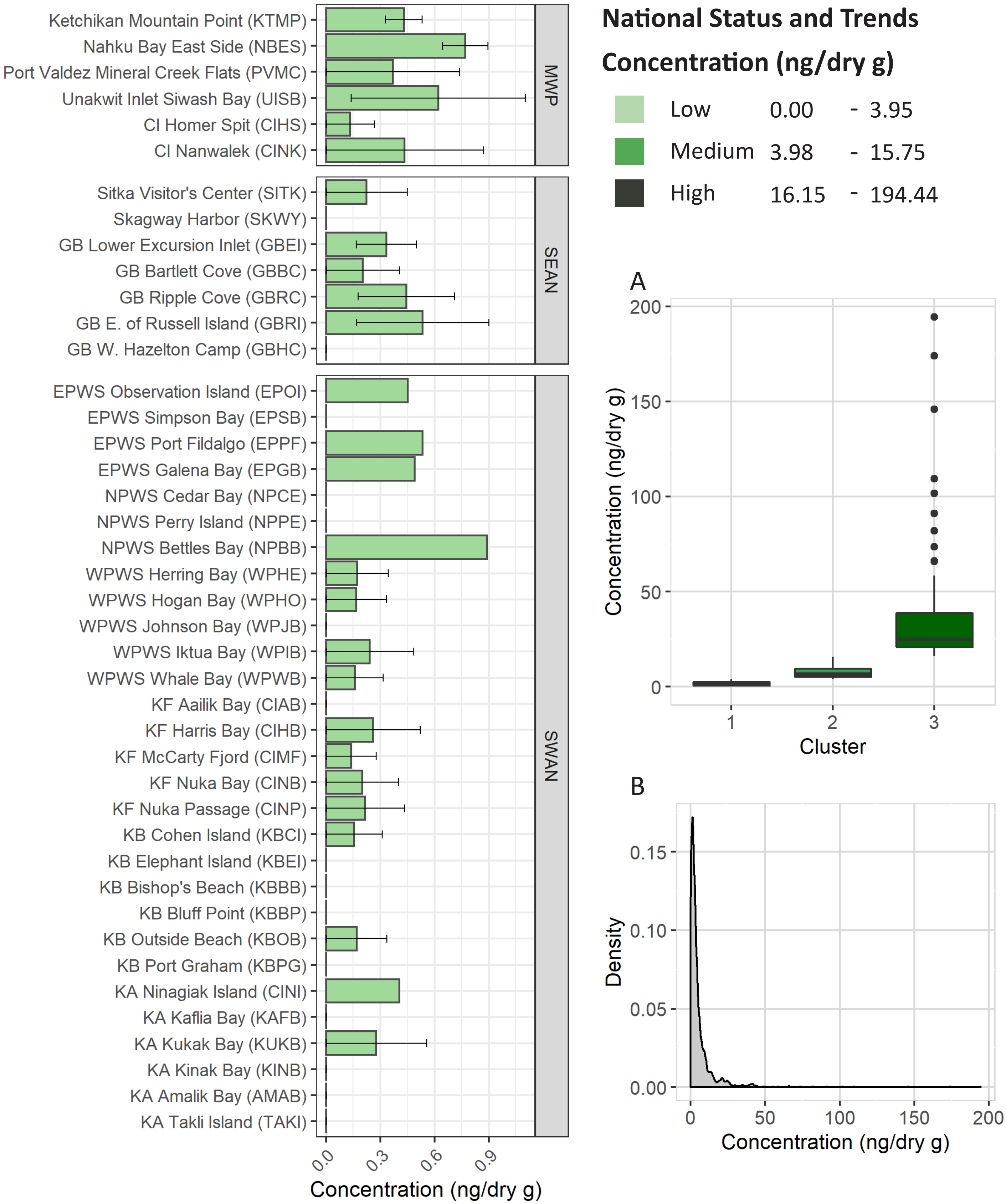

Figure 26. Bar plot showing the mean concentrations of Total Dieldrins in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total Dieldrins

- Concentration range: 0.00 - 4.43 ng/dry g
- Highest mean concentration: 0.89 ng/dry g (Northern Prince William Sound Bettles Bay - NPBB)

Total Dieldrins were detected at 27 of the 42 sites (Figure 25, Figure 26). There were no site trends in Total Dieldrins. The highest individual year concentration for Total Dieldrin was 4.43 ng/dry g at Unakwit Inlet Siwash Bay (UISB) (1999) and this was also the only individual year value that went above the NS&T low group (Appendix 2). All of the site mean concentrations were within the NS&T low group. The highest concentration of aldrin/dieldrin in wet weight was 0.00017 ppm which is far below the US FDA safety level of 0.3 ppm wet weight in all fish. With the exception of year 1999, in which both aldrin and dieldrin were detected, dieldrin was the only compound detected of the three compounds included in Total Dieldrins in this report. Total Dieldrins were not detected in any of the samples during the two most recent sampling years, 2017 and 2018, suggesting that these contaminants are no longer widely present in the environment at detectable levels in mussels.

A view of Cook Inlet, Alaska. Credit: NOAA

Total HCHs

CHEMICAL DESCRIPTION

Hexachlorocyclohexane (HCH) is a mixture of eight or more stereoisomers used as an insecticide to protect crops. Technical grade HCH, contains the alpha, beta, gamma, delta and epsilon forms of HCH. Almost all of the insecticidal properties are found in gamma-HCH which is used as an insecticide on fruit, vegetables and forest crops. It is also found in lotion, cream or shampoo as a prescription to treat head and body lice and scabies (ATSDR, 2005b).

All of the isomers are toxic to animals to varying degrees and are persistent in the environment. In sediments and water, HCH can be broken down into less toxic substance by algae, bacteria and fungi, however it is a slow process. HCH has been shown to accumulate in the fatty tissue of fish. The Department of Health and Human Services (DHHS), International Agency for Research on Cancer (IARC) and the EPA vary in their classification of HCH as a human carcinogen. However, technical HCH, alpha-HCH, and beta-HCH are listed by all three as at least possible human carcinogens (ATSDR, 2005b). In 2009, the Stockholm Convention on Persistent Organic Pollutants implemented an international ban on the use of lindane in agricultural but allowed a 5-year extension for its use in the treatment of head lice and scabies (UNEP, 2009). The US did not ratify the convention, however the EPA requested the voluntary cancellation of the last agricultural uses of lindane in 2006 (EPA, 2006). In 2015, based on a review of the most recent data on lindane, the IARC modified its classification from "probably carcinogenic to humans" to "known to cause human cancer" (IARC, 2015).

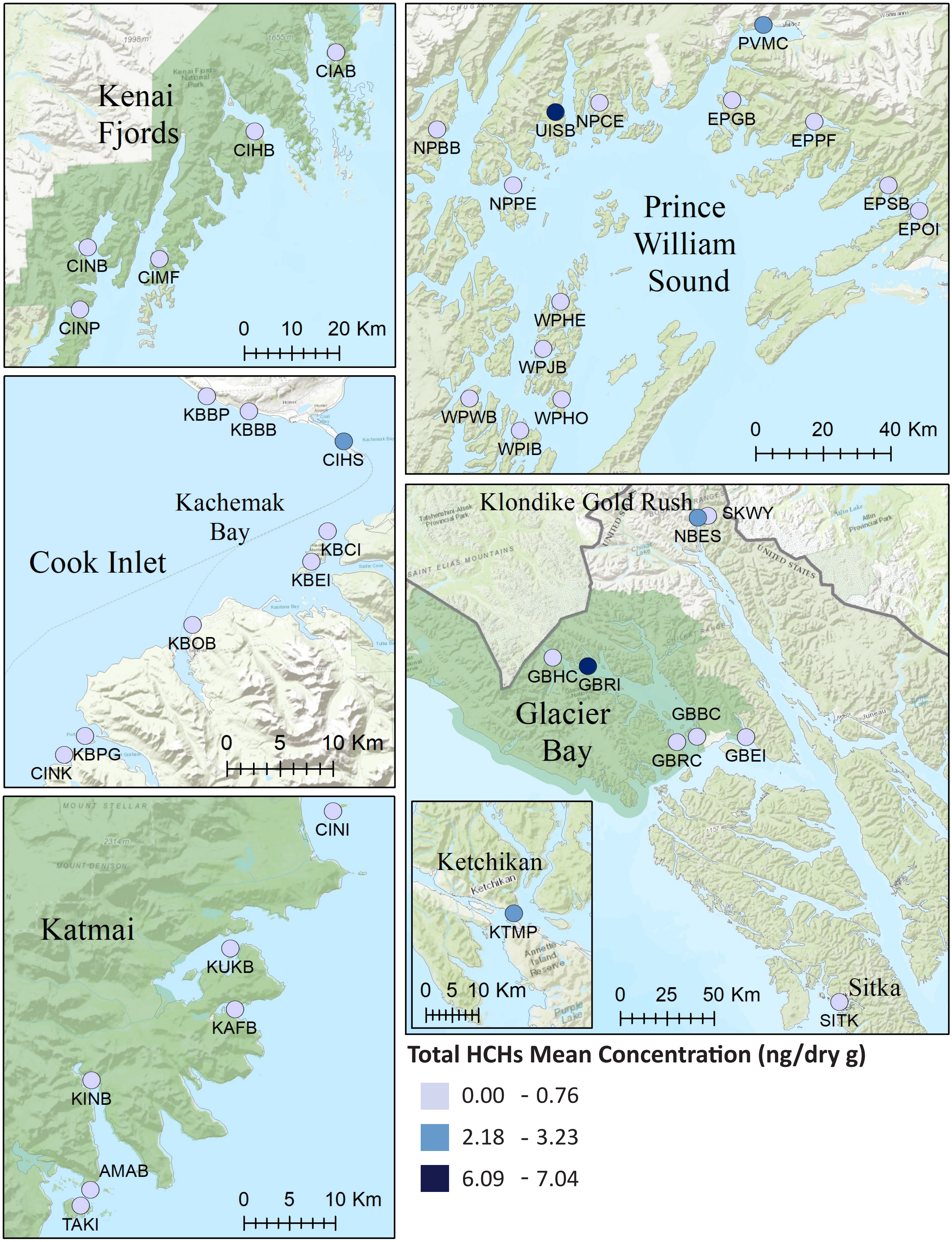

Figure 27. Mean concentration of Total HCHs in mussel tissue at each site.

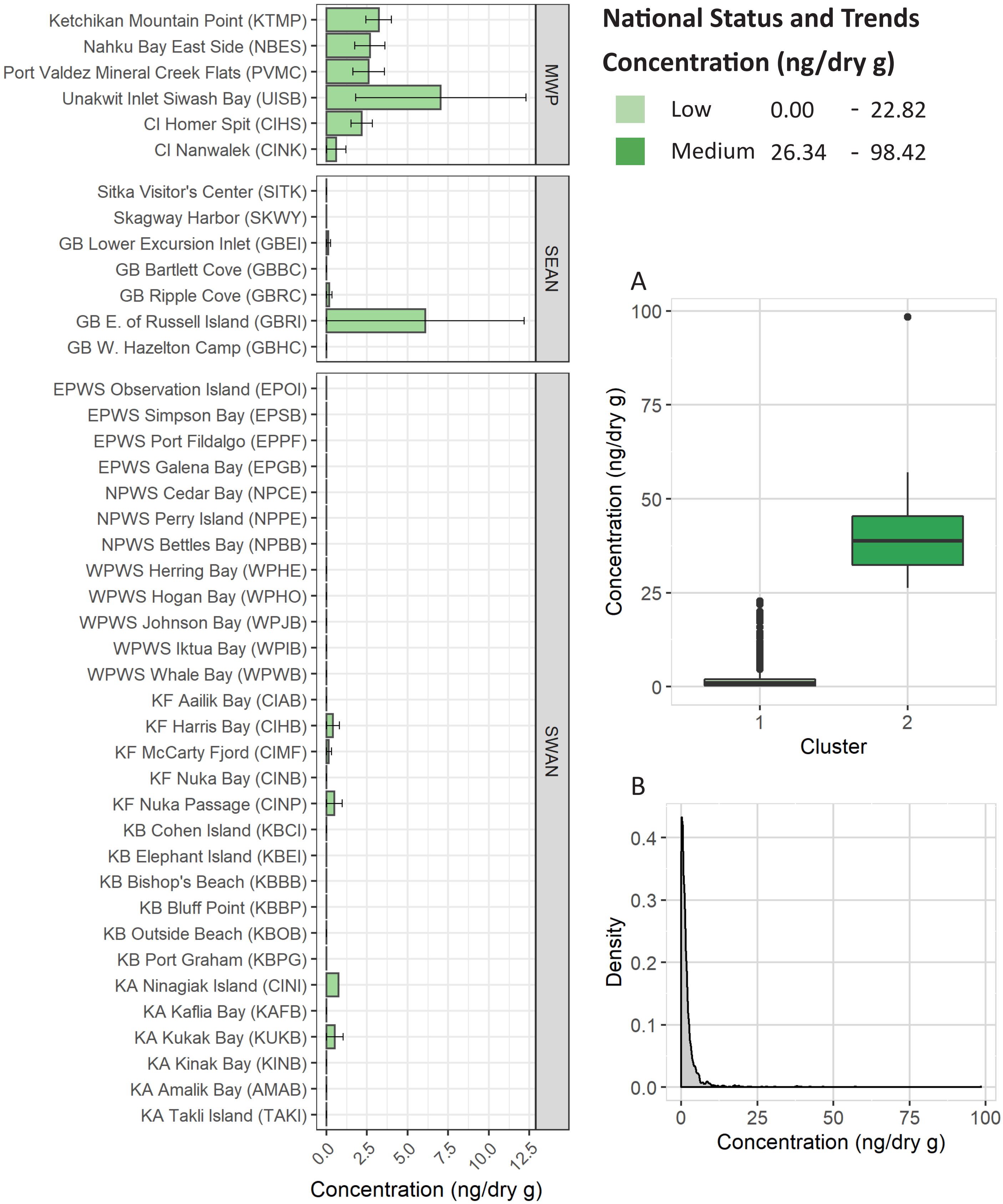

Figure 28. Bar plot showing the mean concentrations of Total HCHs in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total HCHs

- Concentration range: 0.00 - 38.28 ng/dry g
- Highest mean concentration: 7.04 ng/dry g (Unakwit Inlet Siwash Bay - UISB)

HCHs were detected at 14 of the 42 sites (Figure 27, Figure 28). The highest individual year concentration for Total HCHs was 38.28 ng/dry g at UISB (1999) (Appendix 2) and the highest mean site concentration (7.04 ng/dry g) was also at UISB. Similar to Total Dieldrins, the concentration at UISB in 1999 is the only individual year concentration that was within the NS&T medium group. All of the other individual year and site mean concentrations were in the NS&T low group. Despite the consistently low concentrations, there was a decreasing site trend for Total HCHs at Cook Inlet Homer Spit (CIHS) (rho = -0.71, p = 0.047), Nahku Bay East Site (NBES) (rho = -0.78, p =0.01), and Ketchikan Mountain Point (KTMP) (rho = -0.71, p = 0.047). Gamma-HCH was detected in the majority of samples in 2009 and prior, but since 2009 it was only detected at Glacier Bay E. of Russell Island (GBRI). In 2013, GBRI had the second highest individual year concentration (18.27 ng/dry g) which included gamma-HCH, but the Total HCHs was dominated by beta-HCH. No HCHs were detected in the most recent sampling years, 2015, 2017, and 2018, at any of the sites including GBRI, suggesting that these contaminants are no longer widely present in the environment at detectable levels in mussels.

Total PAHs

CHEMICAL DESCRIPTION

Polycyclic aromatic hydrocarbons (PAHs) are found in creosote, soot, petroleum, coal and tar, and are the only organic contaminants measured by the Mussel Watch Program that have natural sources (forest fires and volcanoes) in addition to anthropogenic sources (automobiles emissions, home heating, coal fired power plants). PAHs are formed from the fusing of benzene rings during the incomplete combustion of organic materials. PAHs can also enter the aquatic environment by means of discharge from industrial and wastewater treatments plants (ATSDR, 1995). The fate and transport of PAHs is variable and dependent on the physical properties of each individual compound. Most PAHs strongly associate with particles. High molecular weight (HMW) PAHs associate to a higher degree with particles relative to low molecular weight (LMW) PAHs (ATSDR, 1995). LMW PAHs predominate in petroleum products whereas HMW compounds are associated with combustion.

Made up of a suite of hundreds of compounds, PAHs exhibit a wide range of toxicities. While many aquatic organisms like fish can metabolize PAHs, marine invertebrates, such as oysters, are less able to efficiently metabolize them and as such can be better indicators of overall environmental exposure (Neff, 1985). The PAH contents of plants and animals may be much higher than PAH contents of soil or water in which they live (ATSDR, 1995). A number of the PAHs that bioaccumulate in aquatic and terrestrial organisms are toxic and some including benzo(a)pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-c,d)pyrene are likely carcinogens (ATSDR, 1995). Toxic responses to PAHs in aquatic organisms include reproduction inhibition, mutations, liver abnormalities and mortality. Exposure to aquatic organisms results from oil spills, boat exhaust and urban runoff. There is no US FDA recommended safety level for PAHs in fish and fish products.

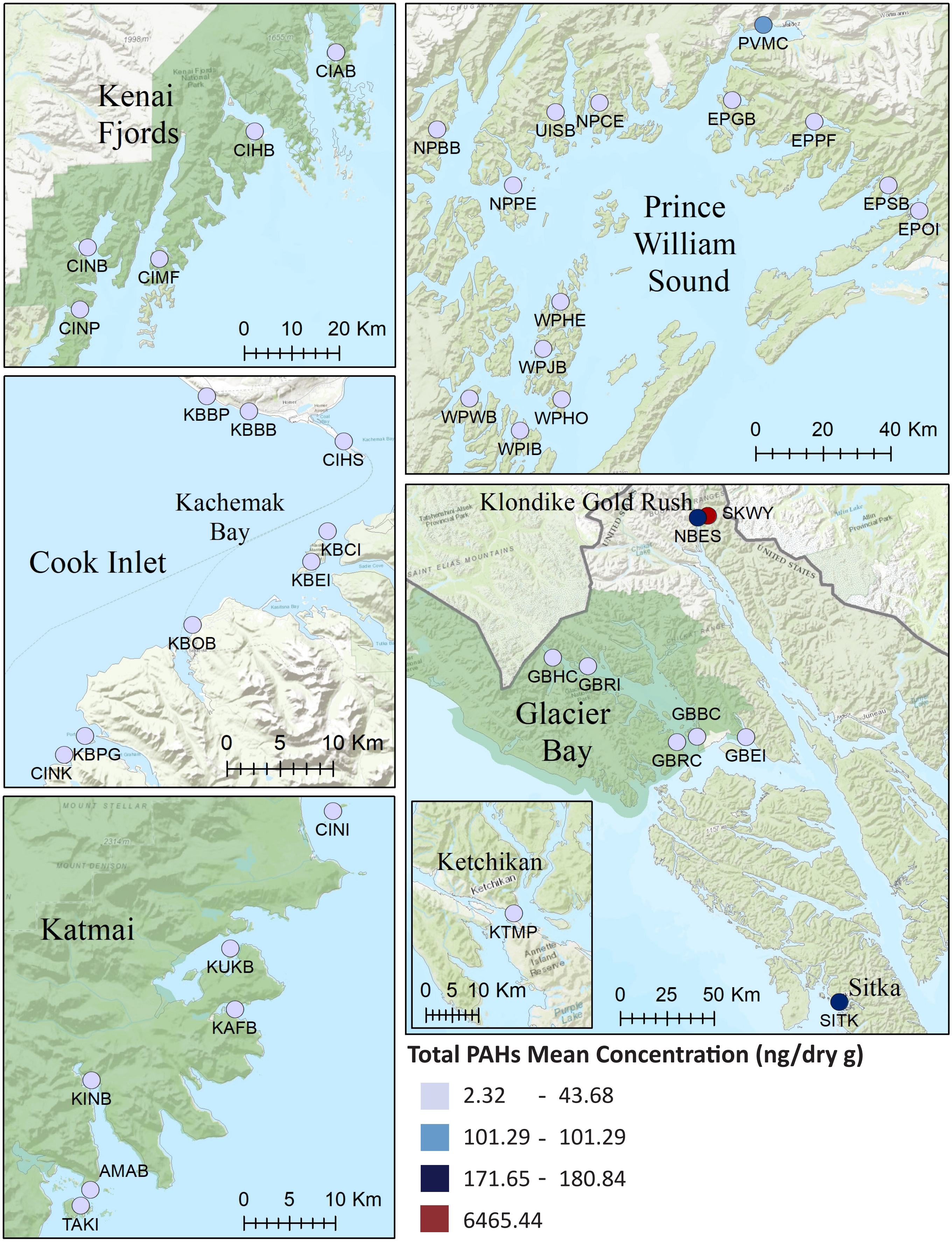

Figure 29. Mean concentration of Total PAHs in mussel tissue at each site.

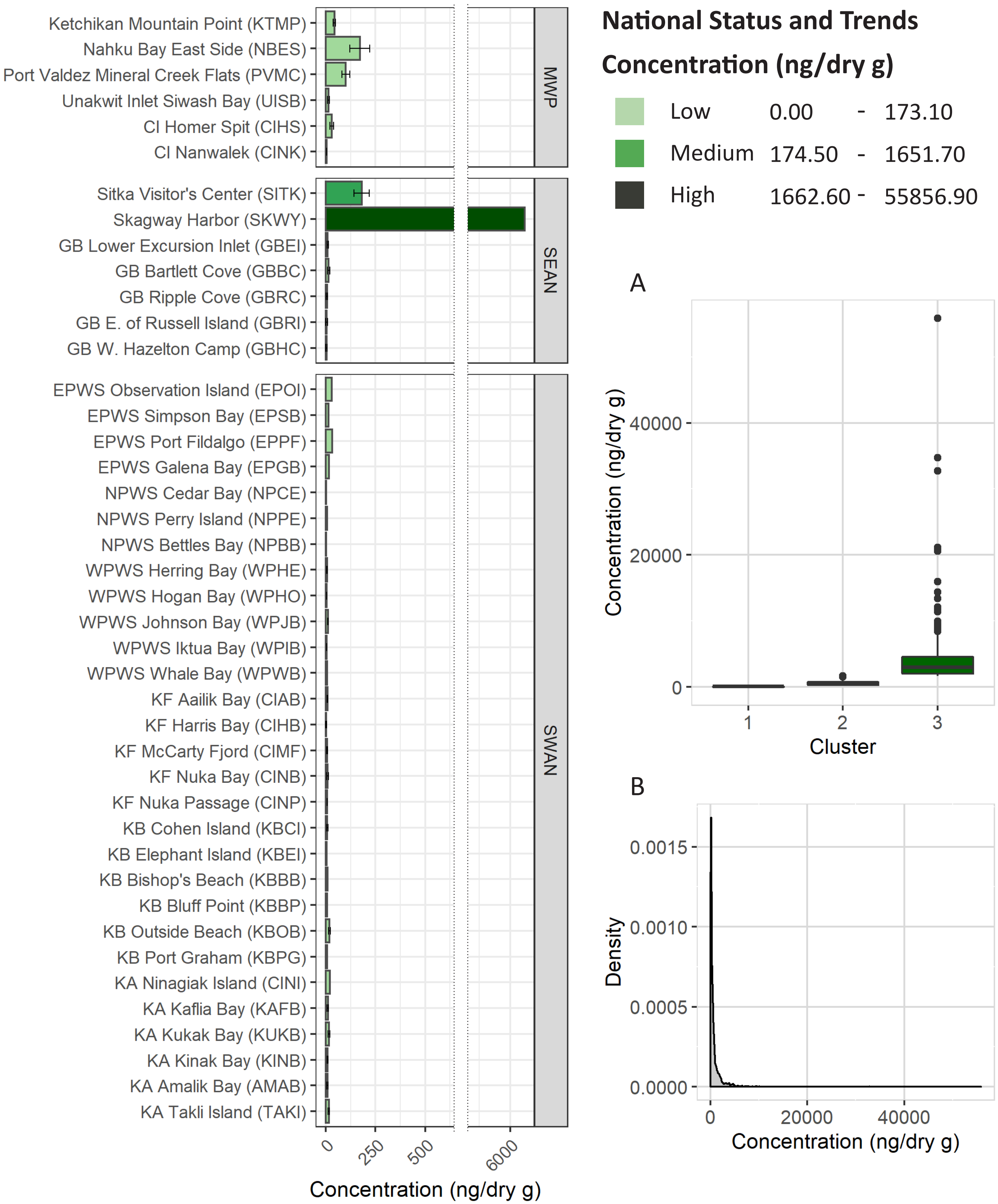

Figure 30. Bar plot showing the mean concentrations of Total PAHs in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total PAHs

- Concentration range: 0.00 - 6465.44 ng/dry g
- Highest mean concentration: 6465.44 ng/dry g (Skagway Harbor - SKWY)

PAHs were detected at all of the 42 sites analyzed (Figure 29, Figure 30). The highest individual year Total PAHs concentration (6465.44 ng/dry g) (Appendix 2) at SKWY was also the highest mean site concentration since SKWY was only sampled once in 2017. This site was a distinct outlier within the Alaska data and was the only Total PAH concentrations that was within the NS&T high group. However, the lack of replication at this site means that these results should be interpreted with caution. The second highest individual year concentration (555.57 ng/dry g, 1995) was at Nahku Bay East Site (NBES), a site adjacent to SKWY. Sitka Visitor's Center (SITK) was the only site whose mean site concentration was within the NS&T medium group. All of the remaining site means were within the NS&T low group. The 17 highest individual year Total PAH concentrations are either SKWY, SITK, NBES or Port Valdez Mineral Creek Flats (PVMC). The Total PAH composition, also known as the profile, for SKWY, SITK and NBES are very similar and appear to represent pyrogenic sources. The combination of the relatively elevated concentrations at these sites as well as the similar compositional patterns suggests that they are subject to anthropogenic contamination, possibly from local vessel traffic. There were no site trends associated with Total PAHs in Alaska.

Tallmon (2012) considered SKWY and Crescent Harbor, a site adjacent to SITK, to be "hot control" sites of relatively heavy human use. Similar to this study, Tallmon (2012) found that concentrations of PAHs were generally one to two orders of magnitude higher at these sites in comparison to sites within Glacier Bay. However, the relative difference between SKWY and Glacier Bay sites in 2017 was greater than that detected in 2007, which suggests that we should interpret these results conservatively and continue sampling at that site.

Total PBDEs

CHEMICAL DESCRIPTION

Brominated flame retardants (BFRs), such as polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls (PBBs), are a group of chemicals that are used in firefighting materials and in consumer and household products to reduce flammability. PBDEs and PBBs are a group of chemicals with 209 possible unique congeners. Commercially, three types of PBDE industrial mixtures were historically produced, the pentabromodiphenyl ether (penta-BDE), octabromodiphenyl ether (octa-BDE) and the decabromodiphenyl ether (deca-BDE) mixtures (EPA, 2009). As these products age and degrade, PBDEs leach into the environment in the form of less brominated PBDEs like tetra-, penta- and hexa-BDE.

PBDEs have become ubiquitous in the environment and are detected in materials that include household dust, human breast milk, sediment and wildlife (ATSDR, 2017). The less brominated PBDEs demonstrate high affinity for lipids and tend to bioaccumulate in animals and humans, while highly brominated PBDEs tend to absorb more onto sediment and soil. The toxicology of PBDEs is not well understood, but PBDEs have been associated with tumors, neurodevelopmental toxicity and thyroid hormone imbalance. Other PBDE congeners have hepatotoxic and mutagen effects. Still others may act as estrogen receptor agonists in vitro (Meerts et al., 2001). Due to ubiquitous distribution, persistence and potential for toxicity, the manufacturing of the 'penta' and 'octa' PBDEs mixtures have been phased out starting in 2004, and the deca mixture starting in 2013 (EPA, 2009; Schreder and La Guardia, 2014).

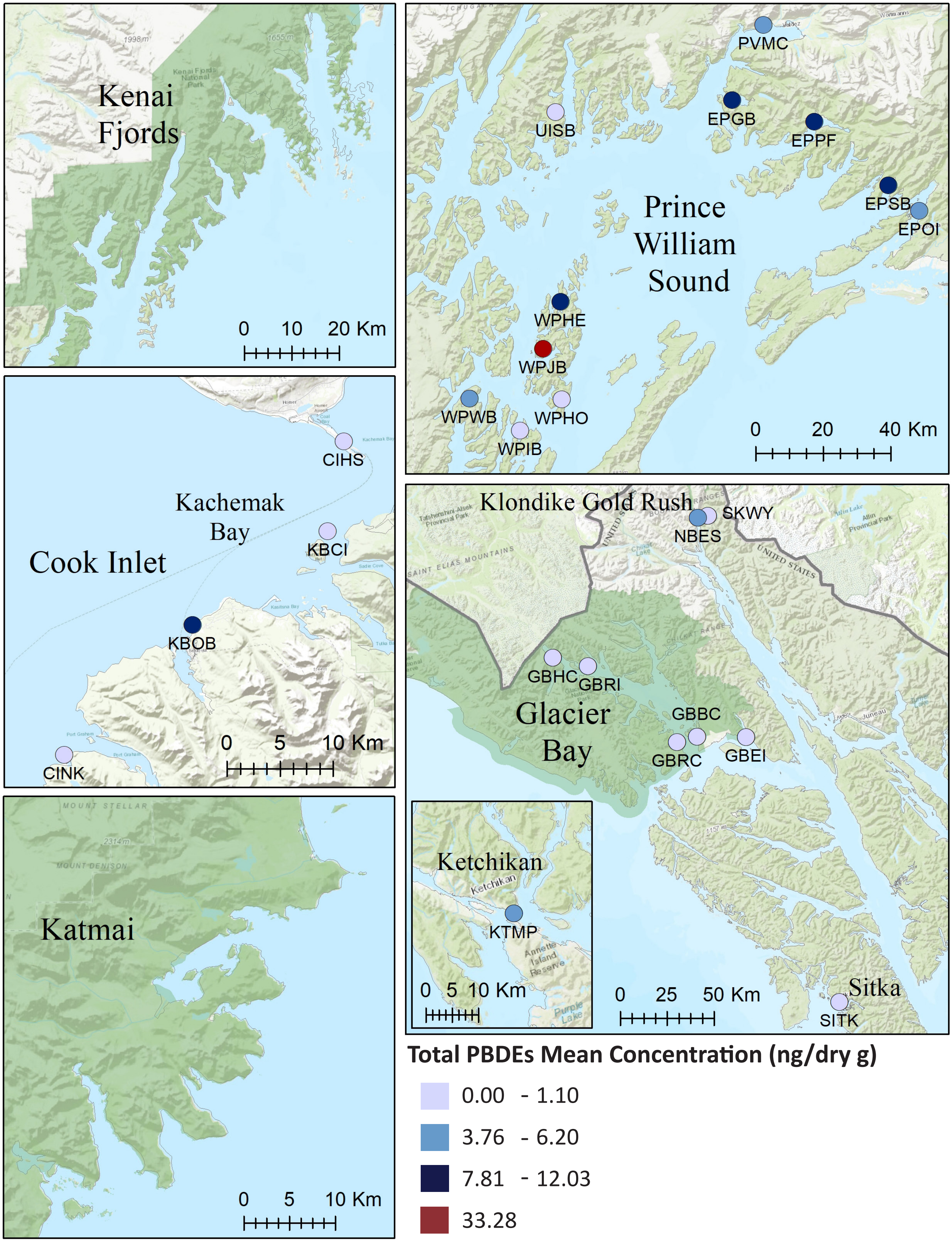

Figure 31. Mean concentration of Total PBDEs in mussel tissue at each site.

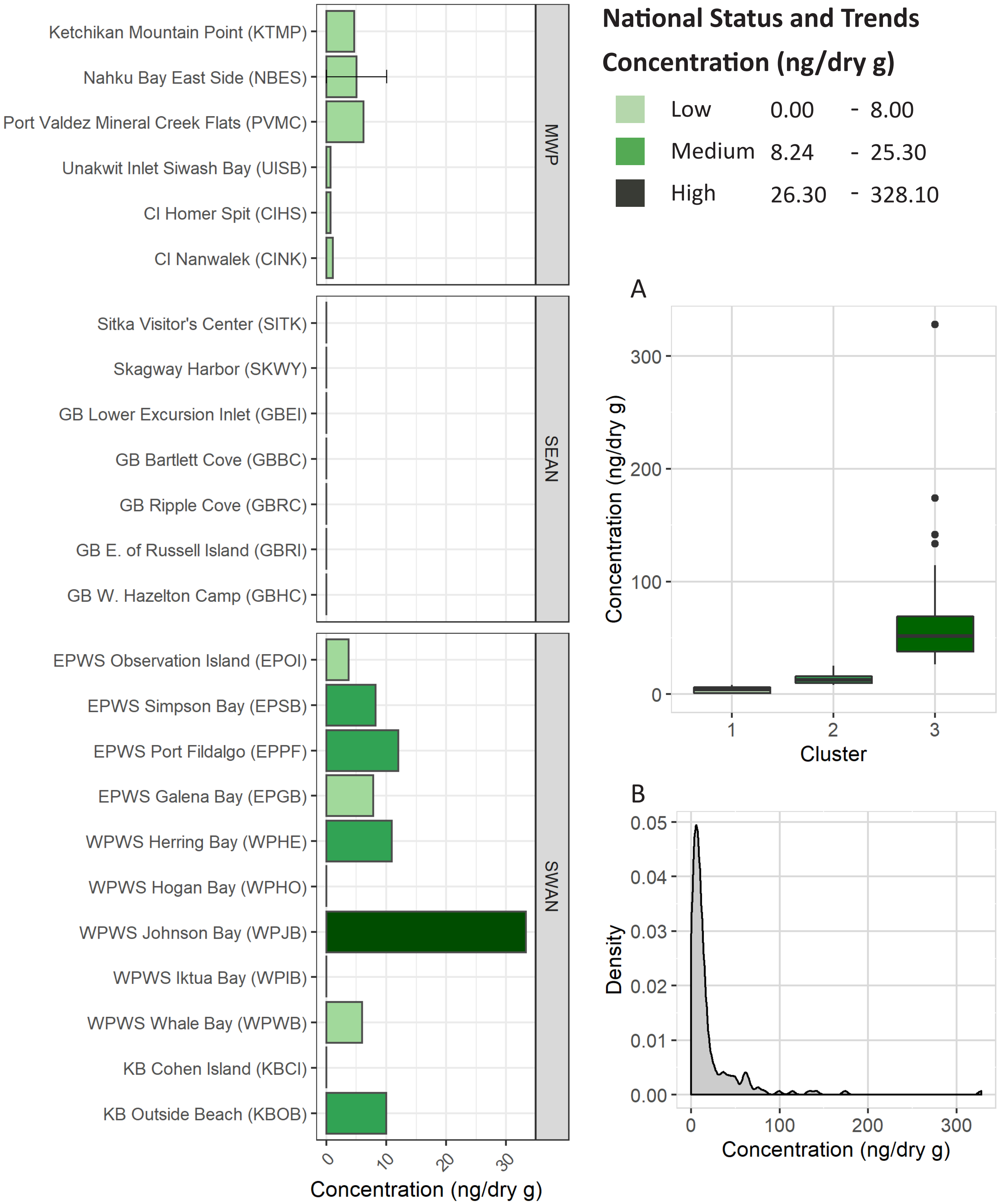

Figure 32. Bar plot showing the mean concentrations of Total PBDEs in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total PBDEs

- Concentration range: 0.00 - 33.28 ng/dry g
- Highest mean concentration: 33.28 ng/dry g (Western Prince William Sound Johnson Bay - WPJB)

PBDEs were detected at 14 of the 24 sites at which they were tested (Figure 31, Figure 32). There were not enough samples for any particular site to assess any site based trends. The highest individual year concentration (33.28 ng/dry g, 2012) (Appendix 2) and the highest site mean concentration (33.28 ng/dry g) were both at WPJB. This sample was an outlier within the Alaska samples and it was also the only concentration that was within the NS&T high group. The other site mean concentrations were within either the NS&T medium or low groups. None of the sites in SEAN had detectable levels of Total PBDEs. However, Tallmon (2012) did detect PBDEs in SEAN in 2007 and 2011. This suggests that PBDEs are no longer widely present in the environment at detectable levels in mussels in the SEAN area.

PBBs were not detected in any of the samples for which they were tested (Appendix 2).

Bald eagles in Homer Spit, Alaska. Credit: NOAA

Total PCBs

CHEMICAL DESCRIPTION

Polychlorinated Biphenyls (PCBs) are synthetic organic compounds that have been used in numerous applications including electrical transformers and capacitors, hydraulic and heat transfer fluids, pesticides and in paints. PCBs have a biphenyl ring structure (two benzene rings with a carbon to carbon bond) and a varying number (1-10) of chlorine atoms. There are 209 individual PCB compounds or congeners possible. PCBs were manufactured in the US between 1929 and 1977. In the US, a single manufacturer produced all PCBs and the commercial products were referred to as Aroclors, which are mixtures of PCB congeners. Approximately 65% of PCBs manufactured in the US were used in electrical applications (Eisler & Belisle, 1996). Although no longer manufactured in the US, ecosystem contamination by PCBs is widespread due to their environmental persistence and tendency to bioaccumulate. In water, small amounts of PCB may remained dissolved, but the majority adhere to fine sediment and organic particles and can can take years to degrade. Current pollution sources include volatilization from landfills, leaks from old electrical equipment, and dredging of contaminated sediments (WHO & IPCS, 1993).

PCBs readily accumulate in the tissues of organisms including filter feeders, fish, and marine mammals. They have been linked to many health issues including adversely affecting reproduction, growth, metabolism and survival in animals (Eisler & Belisle, 1996). PCBs are associated with skin ailments, neurological, and immunological responses and at high doses can decrease motor skills and cause memory loss. Other effects can include hepatotoxicity, immunotoxicity, neurotoxicity, low birth weight, and teratogenicity (Eisler & Belisle, 1996). Exposure to PCBs in fish has been linked to reduced growth, reproductive impairment, and vertebral abnormalities (Eisler & Belisle, 1996). PCBs have also been shown to cause cancer in laboratory animals and are likely carcinogens in humans (ATSDR, 2000). The main human exposure route for PCBs is through eating contaminated seafood and meats which is the reason for many consumption advisories. The US FDA safety level for PCBs in all fish (edible portion) is 2 ppm wet weight, irrespective of which mixture of PCBs is present at the residue (US FDA, 2011).

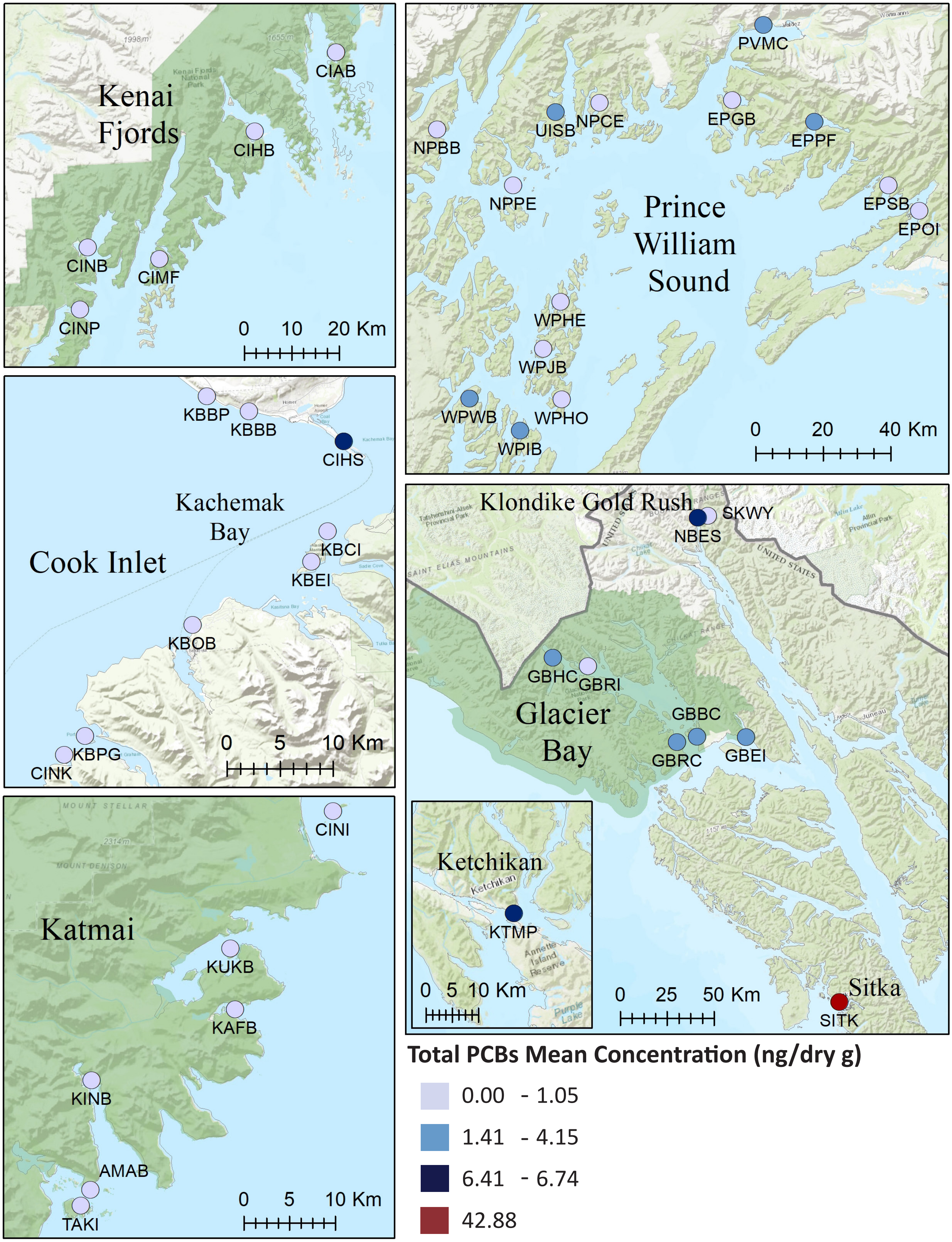

Figure 33. Mean concentration of Total PCBs in mussel tissue at each site.

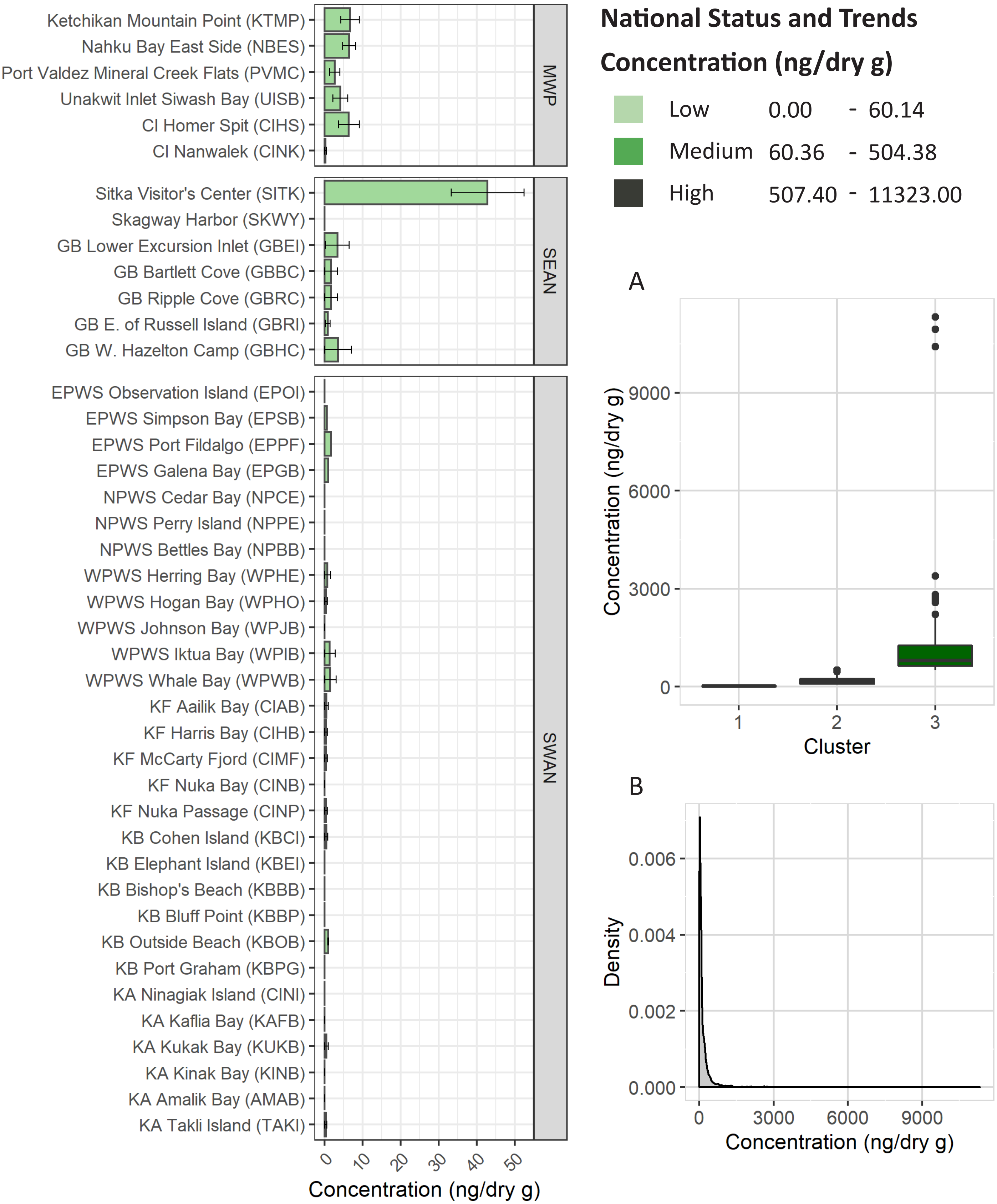

Figure 34. Bar plot showing the mean concentrations of Total PCBs in mussel tissue in Alaska (±SE) compared to the National Status and Trends (NS&T) clusters. Box and whisker plot (A) and density plot (B) of the national NS&T concentrations for comparison. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

Summary of Total PCBs

- Concentration range: 0.00 - 61.82 ng/dry g
- Highest mean concentration: 42.88 ng/dry g (Sitka Visitor's Center - SITK)

PCBs were detected in 27 of the 42 sites in which they were analyzed (Figure 33, Figure 34). The three highest individual year Total PCB concentrations (61.82 ng/dry g, 2013; 35.77 ng/dry g, 2015; 31.06 ng/dry g, 2017) all occured at SITK (Appendix 2). Not surprisingly, SITK was also the site with the highest mean site concentration and was an outlier within the Alaska data (Figure 33). Although the highest individual year concentration found at SITK was in the NS&T medium group, the individual year and site mean concentrations for this site and all other sites were still within the NS&T low group. Tallmon (2012) also found higher levels of Total PCBs at SITK from 2007-2011 compared to other sites sampled in Glacier Bay, but also noted that the concentrations were still low relative to NS&T concentrations. Although all of the concentrations at Nahku Bay East Site (NBES) were in the NS&T low group, there was a decreasing trend (rho = -0.83, p = 0.01) at this site. The highest wet weight concentration for Total PCBs was 0.0095 ppm which did not exceed the US FDA safety level of 2 ppm wet weight.

Summary

Cook Inlet, AK. Credit: NOAA

With the exception of Total PAHs and trace metals, which were detected at 100% of the sites, all of the other contaminants were detected at varying percentages of the total number of sites analyzed (Table 4). Chlordanes were present at 79% of the sites while Butyltins were only detected at 20% of the sites, showing that some contaminants are subject to region-wide influences while others are controlled by localized sources. In general, as is visible by comparing the Alaska data to the NS&T low, medium and high groupings, the majority of the concentrations in the study areas were at background levels (Figure 35). There were several concentration values considered as outliers within the study areas, but for many of the contaminant groups, such as Mercury, Total Butyltins, Total Chlordanes and Total PCBs, these high concentrations are within the NS&T low or medium groups and do not exceed US FDA regulations (Figure 35, Figure 36). Only concentrations of Copper at site Kachemak Bay Bluff Point, Total PAHs at Skagway Harbor and Total PBDEs at Western Prince William Sound Johnson Bay were both considered as outliers and within the NS&T high group. Even at sites where a decreasing trend in contaminant concentrations was observed (Table 5), the highest concentrations were within the NS&T low groups. This speaks to the relatively low background concentrations in this region and the utility of nationwide comparisons and regulatory guidelines to provide perspective to local concentrations.

The relatively high concentrations of cadmium, copper, and nickel in comparison to the NS&T national groups could be a combination of natural inputs and anthropogenic sources. Every site mean cadmium and copper concentration fell within the NS&T medium and high groups. The highest mean concentrations of cadmium were detected mainly in Glacier Bay and Western Prince William Sound. Elevated concentrations of copper were detected at individual sites throughout the study areas but consistently in Glacier Bay and Kachemak Bay. The majority of nickel site mean concentrations were within the medium NS&T grouping, however a few higher concentrations were detected throughout the study area with the highest concentrations detected at sites GBRI and GBHC in Glacier Bay. The natural exposure and weathering of rocks in southern Alaska, as well at the characteristically high elemental concentration in these rocks, can contibute to elevated background concentrations of these metals. This was highlighted in the limited concentration range of the arsenic samples and the low site-specific variability, which suggests no particular point source of contamination but rather a low background level of contamination due to general sources such as atmospheric deposition and natural crustal rock erosion. However, some of the elements were both more variable among sites and had similar spatial distributions suggesting that concentration levels may be related to site characteristics. In particular, lead, copper and nickel were more variable both spatially and/or temporally than arsenic or cadmium. They also seem to be affected by the same environmental or anthropogenic sources because many of the sites with the highest concentrations of lead were also the sites with the highest concentrations of copper and nickel, such as sites KBBP, CINB, NPBB in SWAN. In SEAN, the concentrations of copper and lead had similar patterns with higher values at SITK and GBBC. This suggests site specific influences for these contaminants in addition to ubiquitous natural sources.

Table 4. Frequency of detection (percent of total) of contaminant groups in mussel tissue from SE and SW Alaska networks.

Contaminant Group	Number of sites with detects	Number of sites analyzed	Detection frequency
Arsenic (As)	42	42	100%
Cadmium (Cd)	42	42	100%
Copper (Cu)	42	42	100%
Lead (Pb)	42	42	100%
Mercury (Hg)	42	42	100%
Nickel (Ni)	42	42	100%
Total Butyltins	8	41	20%
Total Chlordanes	33	42	79%
Total Chlorobenzenes	20	42	48%
Total DDTs	20	42	48%
Total Dieldrins	27	42	64%
Total HCHs	14	42	33%
Total PAHs	42	42	100%
Total PBDEs	14	24	58%
Total PCBs	27	42	64%

NATIONAL COMPARISON

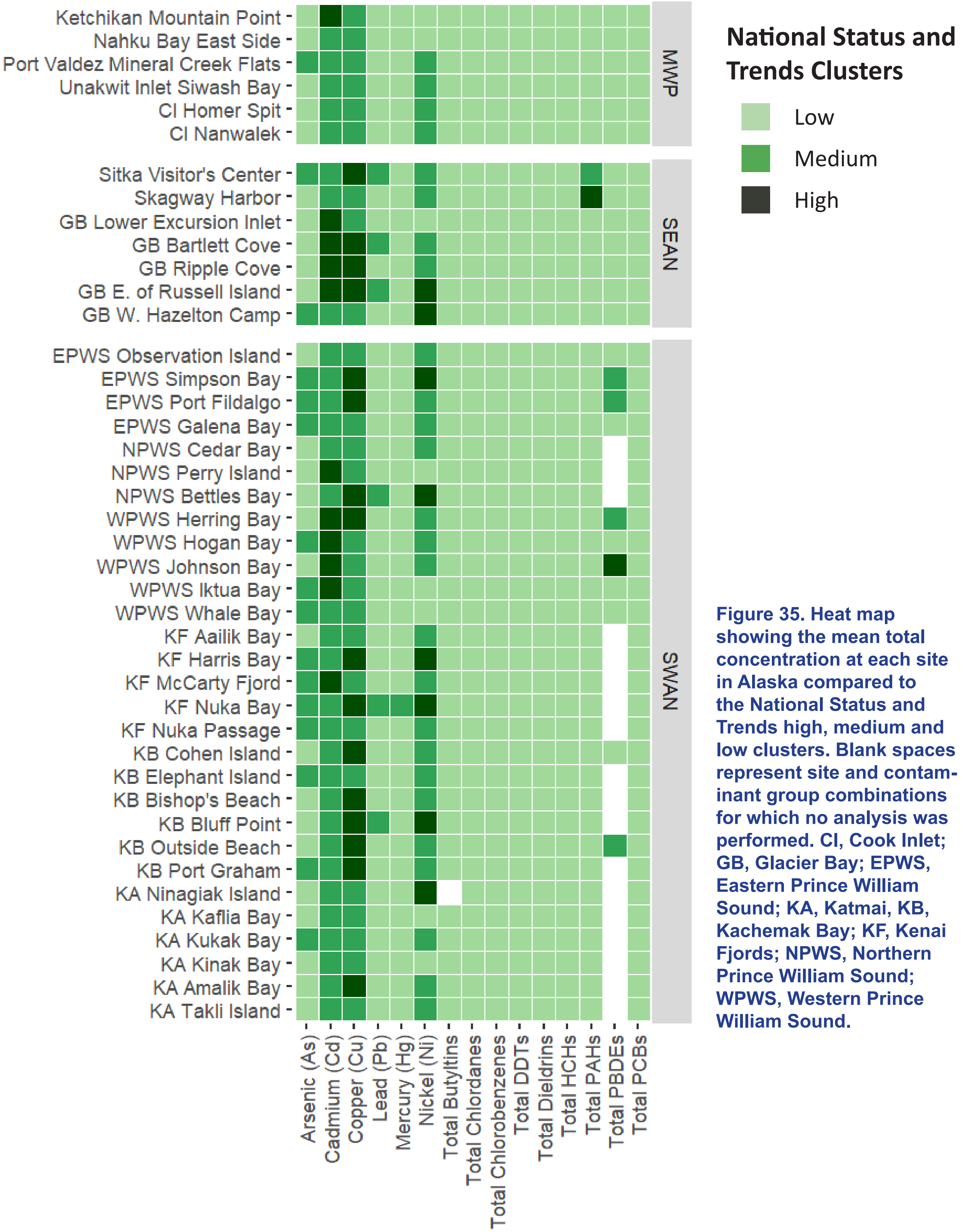

Figure 35. Heat map showing the mean total concentration at each site in Alaska compared to the National Status and Trends high, medium and low clusters. Blank spaces represent site and contaminant group combinations for which no analysis was performed. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound.

ALASKA SUMMARY

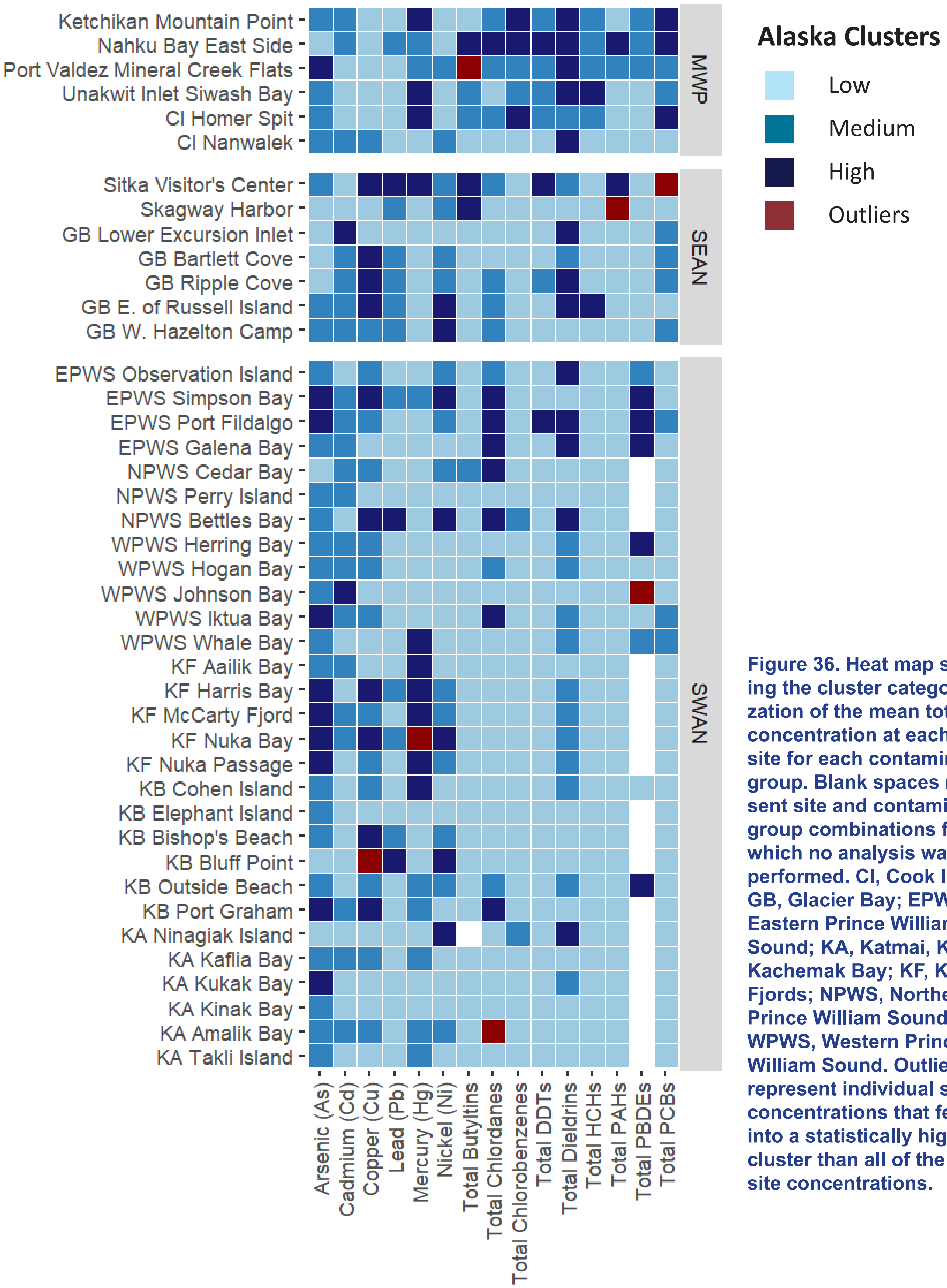

Figure 36. Heat map showing the cluster categorization of the mean total concentration at each site for each contaminant group. Blank spaces represent site and contaminant group combinations for which no analysis was performed. CI, Cook Inlet; GB, Glacier Bay; EPWS, Eastern Prince William Sound; KA, Katmai, KB, Kachemak Bay; KF, Kenai Fjords; NPWS, Northern Prince William Sound; WPWS, Western Prince William Sound. Outliers represent individual site concentrations that fell into a statistically higher cluster than all of the other site concentrations.

SUMMARY

Table 5. Summary of site specific trends for sites with at least seven samples. "D" indicates a decreasing trend and blank indicates no trend.

Contaminant Group	CIHS	KTMP	NBES	PVMC	UISB
Arsenic (As)			D		
Cadmium (Cd)					
Copper (Cu)					
Lead (Pb)					
Mercury (Hg)					
Nickel (Ni)					
Total Butyltins					
Total Chlordanes					
Total Chlorobenzenes	D		D	D	
Total DDTs					
Total Dieldrins					
Total HCHs	D	D	D		
Total PAHs					
Total PCBs			D		

Despite the low concentrations for many of the contaminant groups, composition and variation can help to provide insight into the nature of the contamination in the study areas. Some contaminant groups have decreased in concentration over time but most of them have shown no discernible trend (Table 5). Many contaminants have remained at background levels making further significant reductions unlikely. The decreasing trend in concentrations of Total Chlorobenzenes at sites CIHS, NBES and PVMC, Total HCHs at CIHS, KTMP and NBES, and Total PCBs at NBES demonstrates the importance of long standing monitoring programs and provide a point of reference for current sampling. These trends allow us to observe the effects of regulations on the concentrations of these contaminants in the environment and to put into perspective any recently detected concentrations. Sample concentrations and compositions for Total DDT, Total Dieldrins and Total HCHs in the most recent sampling years suggests that these contaminants are no longer widely present in the environment at detectable levels. DDT parent compounds were only detected in three samples since 2007 and in none of the samples since 2013. Dieldrins were not detected in any of the samples from 2017 and 2018, and HCHs were not detected in 2015, 2017 or 2018. Total Butyltin concentrations were low compared to the NS&T national concentrations, but the presence of the parent compound TBT in recent years at SITK and SKWY indicates that fresh sources of butyltin are still entering this environment, probably through vessel traffic at these sites. Tallmon (2012) did not find TBT at SITK in 2007, 2009 or 2011, however it was present at the neighboring Crescent Harbor site, indicating a source in that area. Additionally, the similar Total PAH compositions and higher concentrations at SITK, SKWY and NBES suggest that these sites are receiving anthropogenic sources of PAH contamination. This is consistent with the findings of Tallmon (2012) and reflects the heavy boat use in these areas. This type of information can help to anticipate sources of contamination and guide future sampling.

The low concentrations of most contaminants detected in the study areas when compared to nationwide NS&T data, as well as the high concentration of Total PAHs detected at SKWY, support the need for consistent monitoring programs that can contextualize any one particular area or result. All monitoring programs are subject to limitations due to funding and resources, and monitoring plans should be created with longevity and consistency in mind. Intensive initial monitoring efforts can serve as a baseline from which more limited and less resource intensive monitoring efforts can continue. A successful monitoring program must both achieve an understanding of background contamination levels and capture the variability and range of possible contamination. Existing contaminant data from Alaska, including this report, has provided an understanding of general background contamination including range, trends and variability. Further monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale (i.e. through periodic sampling and biennial sampling at select sites) but focus on areas of interest that could give further insight on range and variation (see supplemental material). For example, further sampling at Skagway Harbor could help to clarify the relatively high Total PAH concentration that was detected. Additionally, sampling when significant landscape changes occur could provide information on the effects of both manmade and natural transformations. Through an understanding of both the temporal and spatial variations, a monitoring program can serve its purpose of assessing potential contaminant exposure.

Prince William Sound, AK. Credit: NOAA

Bald eagles in Nanwalek, AK. Credit: NOAA

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Appendix 1. Yearly contaminant group totals for each site for Arsenic, Cadmium, Copper, Lead, Mercury, and Nickel (µg/dry g), Total Butyltins (ng Sn/dry g), Total Chlordanes and Total Chlorobenzenes (ng/dry g).

Map Code	Site Name	Group	Year	Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Lead (Pb)	Mercury (Hg)	Nickel (Ni)	Total Butyltins	Total Chlordanes	Total Chlorobenzenes
CIHS	CI Homer Spit	MWP	1995	10.86	1.94	7.80	1.18	0.11	2.10	0.00	1.49	NA
CIHS	CI Homer Spit	MWP	1997	10.40	1.72	15.60	0.95	0.13	1.70	2.40	1.47	4.68
CIHS	CI Homer Spit	MWP	1999	8.55	1.37	7.60	0.54	0.13	1.58	13.26	2.63	8.54
CIHS	CI Homer Spit	MWP	2001	8.08	1.78	6.64	0.59	0.09	1.48	0.00	0.38	0.48
CIHS	CI Homer Spit	MWP	2003	10.70	2.33	10.50	1.30	0.09	2.55	0.83	0.00	1.50
CIHS	CI Homer Spit	MWP	2005	12.10	1.66	10.30	1.29	0.12	3.42	0.00	1.45	0.48
CIHS	CI Homer Spit	MWP	2007	9.22	1.57	9.01	0.84	0.11	1.99	0.00	1.83	0.00
CIHS	CI Homer Spit	MWP	2018	13.19	2.03	83.60	3.43	0.04	12.88	0.00	0.00	0.46
CINK	CI Nanwalek	MWP	2007	9.21	3.67	6.91	0.34	0.05	1.67	0.00	0.59	0.67
CINK	CI Nanwalek	MWP	2018	11.72	5.11	48.12	1.08	0.05	6.80	0.00	0.00	0.40
KTMP	Ketchikan Mountain Point	MWP	1995	11.71	5.00	7.36	0.37	0.06	1.18	0.00	0.33	NA
KTMP	Ketchikan Mountain Point	MWP	1997	8.30	3.61	13.80	0.22	0.04	0.70	0.80	1.11	4.11
KTMP	Ketchikan Mountain Point	MWP	1999	11.50	3.62	6.38	0.26	0.22	1.45	0.16	4.32	10.73
KTMP	Ketchikan Mountain Point	MWP	2001	12.00	5.39	6.98	0.34	0.08	1.10	0.00	0.00	0.49
KTMP	Ketchikan Mountain Point	MWP	2003	9.00	4.64	7.56	0.26	0.12	0.83	0.00	0.00	2.19
KTMP	Ketchikan Mountain Point	MWP	2005	11.40	7.12	7.03	0.59	0.07	1.20	0.00	0.30	0.98
KTMP	Ketchikan Mountain Point	MWP	2007	8.67	5.11	5.86	0.38	0.07	1.23	0.00	1.38	0.99
KTMP	Ketchikan Mountain Point	MWP	2009	NA	NA	NA	NA	NA	NA	0.00	0.76	0.80
NBES	Nahku Bay East Side	MWP	1995	9.95	3.14	6.86	1.94	0.06	1.01	0.00	6.40	NA
NBES	Nahku Bay East Side	MWP	1997	10.10	4.06	12.60	3.10	0.09	1.50	27.00	1.53	6.60
NBES	Nahku Bay East Side	MWP	1999	8.79	4.02	7.03	1.22	0.15	1.51	2.58	0.87	10.39
NBES	Nahku Bay East Side	MWP	2001	9.77	4.38	5.41	0.85	0.05	0.00	19.78	2.06	0.97
NBES	Nahku Bay East Side	MWP	2003	8.24	4.23	6.71	1.29	0.05	1.29	0.00	0.83	1.89
NBES	Nahku Bay East Side	MWP	2005	9.15	5.42	6.00	2.14	0.10	2.00	0.00	4.08	0.61
NBES	Nahku Bay East Side	MWP	2007	7.69	7.04	8.18	1.37	0.09	1.13	0.00	4.99	0.85
NBES	Nahku Bay East Side	MWP	2009	NA	NA	NA	NA	NA	NA	0.00	0.79	0.00
NBES	Nahku Bay East Side	MWP	2017	7.41	2.34	51.90	3.49	0.05	3.35	0.00	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	1986	12.67	2.83	10.67	1.47	0.06	2.63	NA	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1987	13.33	2.47	12.00	1.43	0.06	1.63	NA	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1988	12.00	2.67	13.67	2.10	0.07	5.00	NA	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1990	12.62	2.42	7.70	1.43	0.00	3.30	0.00	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1991	13.00	2.00	12.67	1.27	0.08	2.50	107.00	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1992	11.23	2.44	10.36	1.51	0.09	2.66	36.25	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1993	13.00	2.67	8.35	1.10	0.08	1.66	0.00	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1995	10.43	2.32	10.52	1.19	0.08	2.72	0.00	0.88	NA
PVMC	Port Valdez Mineral Creek	MWP	1997	9.30	2.41	16.40	1.30	0.09	2.60	17.00	0.26	0.96
PVMC	Port Valdez Mineral Creek	MWP	1999	11.40	2.89	11.50	2.21	0.11	7.90	27.03	1.04	2.00
PVMC	Port Valdez Mineral Creek	MWP	2001	8.79	3.03	6.05	1.53	0.07	2.86	15.27	0.00	0.58
PVMC	Port Valdez Mineral Creek	MWP	2003	17.30	3.97	7.71	0.95	0.07	2.26	1.05	0.00	0.32
PVMC	Port Valdez Mineral Creek	MWP	2005	12.10	3.45	26.90	2.98	0.09	8.93	0.00	3.94	0.00
PVMC	Port Valdez Mineral Creek	MWP	2007	16.70	4.51	10.80	1.79	0.10	9.97	0.00	0.85	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1986	16.00	4.63	9.70	1.00	0.09	2.20	NA	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1987	11.00	3.47	11.33	1.30	0.09	2.77	NA	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1988	11.67	2.67	9.87	1.01	0.11	1.90	NA	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1990	10.61	2.70	9.07	0.88	0.04	2.24	0.00	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1991	11.00	3.00	8.67	0.75	0.08	1.53	0.00	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1992	9.71	2.34	8.09	0.70	0.10	1.99	11.40	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1993	8.41	2.87	6.68	0.56	0.10	1.16	0.00	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1995	8.33	2.96	11.83	2.22	0.11	4.79	0.00	0.51	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1997	10.70	3.61	15.50	1.60	0.10	2.90	0.00	0.00	5.75
UISB	Unakwit Inlet Siwash Bay	MWP	1999	10.40	2.38	22.00	1.42	0.10	3.10	0.00	0.83	2.14
UISB	Unakwit Inlet Siwash Bay	MWP	2001	11.40	3.65	8.47	0.87	0.12	1.96	0.00	0.00	0.49
UISB	Unakwit Inlet Siwash Bay	MWP	2003	10.30	3.38	7.99	0.59	0.07	1.69	0.00	0.21	0.30

Appendix 1. (Continued)

Map Code	Site Name	Group	Year	Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Lead (Pb)	Mercury (Hg)	Nickel (Ni)	Total Butyltins	Total Chlordanes	Total Chlorobenzenes
UISB	Unakwit Inlet Siwash Bay	MWP	2005	12.00	2.61	33.30	1.97	0.11	7.43	0.00	0.75	0.44
UISB	Unakwit Inlet Siwash Bay	MWP	2007	11.70	4.30	16.70	1.15	0.10	2.38	0.00	1.50	0.64
GBBC	GB Bartlett Cove	SEAN	2013	9.32	6.27	28.60	1.49	0.06	4.37	0.00	2.10	0.00
GBBC	GB Bartlett Cove	SEAN	2015	9.07	4.69	18.00	0.97	0.05	6.34	0.00	0.00	0.00
GBBC	GB Bartlett Cove	SEAN	2017	9.90	3.68	134.00	6.61	0.05	4.94	0.00	0.00	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2013	10.30	6.46	24.20	0.90	0.04	2.77	0.00	1.53	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2015	8.53	6.00	11.00	0.53	0.04	1.40	0.00	0.00	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2017	10.30	6.30	35.00	1.42	0.04	1.64	0.00	0.00	0.00
GBHC	GB W. Hazelton Camp	SEAN	2013	8.09	3.52	11.70	0.53	0.02	2.65	0.00	3.23	0.00
GBHC	GB W. Hazelton Camp	SEAN	2015	13.00	4.62	35.90	2.25	0.04	25.80	0.00	0.00	0.00
GBHC	GB W. Hazelton Camp	SEAN	2017	13.30	4.22	68.60	4.05	0.05	6.09	0.00	0.00	0.00
GBRC	GB Ripple Cove	SEAN	2013	8.31	4.44	77.30	3.46	0.04	6.18	0.00	3.20	0.00
GBRC	GB Ripple Cove	SEAN	2015	8.41	5.03	59.70	2.41	0.04	4.88	0.00	0.00	0.00
GBRC	GB Ripple Cove	SEAN	2017	11.60	5.41	19.60	1.17	0.08	6.46	0.00	0.00	0.00
GBRI	GB E. of Russell Island	SEAN	2013	12.60	4.09	72.50	3.59	0.04	21.40	0.00	3.39	0.00
GBRI	GB E. of Russell Island	SEAN	2015	9.23	5.33	27.10	1.38	0.05	8.55	0.00	0.00	0.00
GBRI	GB E. of Russell Island	SEAN	2017	11.50	4.96	57.60	3.02	0.06	4.64	0.00	0.00	0.00
SITK	Sitka Visitor's Center	SEAN	2013	10.20	1.98	167.00	7.62	0.11	7.84	18.71	2.74	0.00
SITK	Sitka Visitor's Center	SEAN	2015	13.70	4.55	9.68	1.54	0.16	2.51	0.00	0.00	0.99
SITK	Sitka Visitor's Center	SEAN	2017	10.60	2.29	42.60	1.71	0.11	5.01	11.17	0.00	0.00
SKWY	Skagway Harbor	SEAN	2017	6.56	1.90	24.30	2.02	0.05	4.41	6.95	0.00	0.00
AMAB	KA Amalik Bay	SWAN	2007	9.65	3.27	9.04	0.24	0.06	5.79	NA	12.65	0.00
AMAB	KA Amalik Bay	SWAN	2018	12.72	4.44	80.02	2.78	0.08	4.44	0.00	0.00	0.00
CIAB	KF Aailik Bay	SWAN	2007	10.90	4.42	32.90	0.95	0.08	4.32	NA	0.00	0.00
CIAB	KF Aailik Bay	SWAN	2018	10.06	3.83	10.30	0.47	0.09	2.47	0.00	0.00	0.00
CIHB	KF Harris Bay	SWAN	2007	12.60	2.98	13.90	0.72	0.14	8.38	NA	0.72	0.43
CIHB	KF Harris Bay	SWAN	2018	10.94	2.30	85.74	2.97	0.08	4.72	0.00	0.00	0.00
CIMF	KF McCarty Fjord	SWAN	2007	13.40	5.18	11.10	0.66	0.13	7.18	NA	0.00	0.00
CIMF	KF McCarty Fjord	SWAN	2018	11.05	3.90	45.00	1.23	0.08	3.46	0.00	0.00	0.00
CINB	KF Nuka Bay	SWAN	2007	12.90	4.28	16.30	1.14	0.18	8.94	NA	0.35	0.00
CINB	KF Nuka Bay	SWAN	2018	13.82	3.94	87.77	3.97	0.15	9.67	0.00	0.00	0.00
CINI	KA Ninagiak Island	SWAN	2007	8.76	2.53	10.30	0.28	0.06	7.66	NA	0.38	0.74
CINP	KF Nuka Passage	SWAN	2007	10.60	3.50	10.40	0.77	0.12	4.63	NA	0.59	0.62
CINP	KF Nuka Passage	SWAN	2018	13.25	3.26	51.77	1.81	0.10	5.20	0.00	0.00	0.00
EPGB	EPWS Galena Bay	SWAN	2012	11.50	4.43	16.90	0.69	0.06	2.99	0.00	2.39	0.00
EPOI	EPWS Observation Island	SWAN	2012	11.10	2.23	35.30	1.48	0.04	6.01	0.00	1.39	0.00
EPPF	EPWS Port Fildalgo	SWAN	2012	11.80	3.95	38.80	1.57	0.05	6.36	0.00	2.47	0.00
EPSB	EPWS Simpson Bay	SWAN	2012	12.10	4.04	52.80	2.24	0.07	9.37	0.00	1.78	0.00
KAFB	KA Kaflia Bay	SWAN	2007	11.80	6.38	11.60	0.42	0.09	1.57	NA	0.00	0.00
KAFB	KA Kaflia Bay	SWAN	2018	9.49	2.14	55.98	2.29	0.07	2.97	0.00	0.00	0.00
KBBB	KB Bishop's Beach	SWAN	2018	10.90	1.84	63.99	2.22	0.03	4.08	0.00	0.00	0.38
KBBP	KB Bluff Point	SWAN	2018	9.58	1.71	133.80	4.26	0.04	9.54	0.00	0.00	0.00
KBCI	KB Cohen Island	SWAN	2012	9.49	3.09	26.70	0.79	0.06	3.01	0.00	1.37	0.00
KBCI	KB Cohen Island	SWAN	2018	12.09	3.61	51.49	1.70	0.11	3.07	0.00	0.00	0.37
KBEI	KB Elephant Island	SWAN	2018	11.41	3.66	14.43	0.43	0.04	3.02	0.00	0.00	0.45
KBOB	KB Outside Beach	SWAN	2012	9.84	3.75	20.20	0.75	0.07	3.80	0.00	1.86	0.00
KBOB	KB Outside Beach	SWAN	2018	11.28	2.50	65.43	2.02	0.08	6.19	0.00	0.00	0.47
KBPG	KB Port Graham	SWAN	2018	12.38	4.07	82.17	1.25	0.07	3.67	0.00	3.65	0.00
KINB	KA Kinak Bay	SWAN	2007	8.75	4.89	7.64	0.21	0.05	1.06	NA	0.00	0.38
KINB	KA Kinak Bay	SWAN	2018	11.54	2.15	26.07	0.57	0.05	3.13	0.00	0.00	0.00
KUKB	KA Kukak Bay	SWAN	2007	12.10	3.11	9.49	0.29	0.07	3.07	NA	0.67	0.91
KUKB	KA Kukak Bay	SWAN	2018	11.38	2.46	18.52	0.50	0.06	2.86	0.00	0.00	0.00
NPBB	NPWS Bettles Bay	SWAN	2013	10.90	3.15	70.00	3.67	0.06	8.35	0.00	2.93	1.06

Appendix 1. (Continued)

Map Code	Site Name	Group	Year	Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Lead (Pb)	Mercury (Hg)	Nickel (Ni)	Total Butyltins	Total Chlordanes	Total Chlorobenzenes
NPCE	NPWS Cedar Bay	SWAN	2013	9.80	3.92	28.40	1.13	0.05	4.41	3.11	2.08	0.00
NPPE	NPWS Perry Island	SWAN	2013	10.00	5.40	6.38	0.57	0.05	1.56	0.00	0.56	0.00
TAKI	KA Takli Island	SWAN	2007	11.00	4.45	8.00	0.24	0.08	2.73	NA	0.00	0.00
TAKI	KA Takli Island	SWAN	2018	10.58	2.26	22.40	0.47	0.06	2.87	0.00	0.00	0.00
WPHE	WPWS Herring Bay	SWAN	2012	10.40	5.98	65.30	1.95	0.06	4.12	0.00	1.34	0.00
WPHE	WPWS Herring Bay	SWAN	2018	10.06	3.50	18.90	0.49	0.05	2.05	0.00	0.00	0.00
WPHO	WPWS Hogan Bay	SWAN	2012	10.90	5.78	23.90	0.83	0.04	3.59	0.00	2.29	0.00
WPHO	WPWS Hogan Bay	SWAN	2018	12.11	4.91	44.87	0.83	0.09	3.29	0.00	0.00	0.36
WPIB	WPWS Iktua Bay	SWAN	2012	11.70	5.00	19.20	0.56	0.05	1.68	0.00	4.65	0.00
WPIB	WPWS Iktua Bay	SWAN	2018	12.34	5.23	45.78	1.41	0.05	2.48	0.00	0.00	0.29
WPJB	WPWS Johnson Bay	SWAN	2012	9.67	8.34	20.90	0.57	0.07	2.95	0.00	1.47	0.00
WPJB	WPWS Johnson Bay	SWAN	2018	10.99	5.59	22.77	0.60	0.05	2.26	0.00	0.00	0.00
WPWB	WPWS Whale Bay	SWAN	2012	11.50	4.03	7.69	0.67	0.10	1.85	0.00	0.75	0.00
WPWB	WPWS Whale Bay	SWAN	2018	11.50	2.68	32.79	0.86	0.08	2.62	0.00	0.00	0.35

Appendix 2. Yearly contaminant group totals (ng/dry g) for each site for Total DDTs, Total Dieldrins, Total HCHs, Total PAHs, Total PBDEs, Total PBBs, Total PCBs, Total Endosulfans, Chlorpyrifos, and Mirex.

Map Code	Site Name	Group	Year	Total DDTs	Total Dieldrins	Total HCHs	Total PAHs	Total PBDEs	Total PBBs	Total PCBs	Total Endosulfans	Chlorpyrifos	Mirex
CIHS	CI Homer Spit	MWP	1995	0.00	0.00	4.46	8.64	NA	NA	NA	NA	NA	NA
CIHS	CI Homer Spit	MWP	1997	0.00	0.00	1.53	32.40	NA	NA	15.72	NA	0.00	0.00
CIHS	CI Homer Spit	MWP	1999	6.60	1.06	5.27	88.34	NA	NA	15.29	NA	0.00	0.00
CIHS	CI Homer Spit	MWP	2001	0.00	0.00	2.47	30.70	NA	NA	0.67	0.00	0.00	0.00
CIHS	CI Homer Spit	MWP	2003	1.47	0.00	0.26	16.00	NA	NA	0.00	0.00	0.00	0.00
CIHS	CI Homer Spit	MWP	2005	0.00	0.00	2.26	25.20	NA	NA	10.48	0.00	0.00	0.00
CIHS	CI Homer Spit	MWP	2007	0.00	0.00	1.20	24.50	0.70	0.00	2.69	0.00	0.00	0.00
CIHS	CI Homer Spit	MWP	2018	0.00	0.00	0.00	17.22	NA	NA	0.00	0.00	0.00	0.00
CINK	CI Nanwalek	MWP	2007	0.55	0.87	1.22	5.20	1.10	0.00	0.51	0.00	0.00	0.00
CINK	CI Nanwalek	MWP	2018	0.00	0.00	0.00	5.05	NA	NA	0.00	0.00	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	1995	0.00	0.00	4.24	53.18	NA	NA	NA	NA	NA	NA
KTMP	Ketchikan Mountain Point	MWP	1997	1.86	0.81	5.32	61.70	NA	NA	9.10	NA	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	1999	4.66	0.40	6.98	29.84	NA	NA	19.24	NA	2.48	0.00
KTMP	Ketchikan Mountain Point	MWP	2001	0.82	0.00	2.12	15.60	NA	NA	0.00	0.00	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	2003	3.49	0.55	0.68	63.40	NA	NA	8.33	0.00	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	2005	1.31	0.58	3.78	30.70	NA	NA	2.59	0.00	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	2007	0.62	0.51	1.37	49.30	4.70	0.00	4.71	0.00	0.00	0.00
KTMP	Ketchikan Mountain Point	MWP	2009	1.05	0.59	1.32	45.70	NA	NA	3.22	0.00	0.00	0.00
NBES	Nahku Bay East Side	MWP	1995	5.11	NA	6.45	555.57	NA	NA	NA	NA	NA	NA
NBES	Nahku Bay East Side	MWP	1997	2.28	0.98	1.70	183.00	NA	NA	11.76	NA	0.00	0.00
NBES	Nahku Bay East Side	MWP	1999	3.54	0.88	8.31	150.06	NA	NA	14.85	NA	5.02	0.00
NBES	Nahku Bay East Side	MWP	2001	2.30	0.85	2.43	62.70	NA	NA	4.97	0.00	0.73	0.00
NBES	Nahku Bay East Side	MWP	2003	8.69	1.07	1.54	154.30	NA	NA	7.03	0.00	0.00	0.00
NBES	Nahku Bay East Side	MWP	2005	1.82	0.98	1.15	114.30	NA	NA	5.83	0.00	0.00	0.00
NBES	Nahku Bay East Side	MWP	2007	2.38	0.91	1.72	114.40	10.10	0.00	5.77	0.00	0.00	0.00
NBES	Nahku Bay East Side	MWP	2009	1.14	0.49	0.87	68.80	NA	NA	1.78	0.00	0.00	0.00
NBES	Nahku Bay East Side	MWP	2017	0.00	0.00	0.00	141.69	0.00	0.00	0.00	0.00	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	1986	3.38	NA	NA	NA	NA	NA	NA	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1987	0.67	NA	NA	NA	NA	NA	NA	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1988	0.00	NA	NA	NA	NA	NA	0.00	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1990	0.00	NA	NA	28.53	NA	NA	0.00	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1991	1.30	0.00	NA	193.64	NA	NA	3.14	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1992	0.00	0.00	NA	130.09	NA	NA	0.00	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1993	0.00	0.00	NA	215.98	NA	NA	0.00	NA	NA	0.00
PVMC	Port Valdez Mineral Creek	MWP	1995	0.00	0.00	6.14	77.72	NA	NA	NA	NA	NA	NA
PVMC	Port Valdez Mineral Creek	MWP	1997	0.00	0.00	3.74	45.40	NA	NA	2.20	NA	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	1999	9.16	3.70	5.83	103.71	NA	NA	15.54	NA	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	2001	0.00	0.00	0.93	19.20	NA	NA	1.86	0.00	0.70	0.00
PVMC	Port Valdez Mineral Creek	MWP	2003	0.72	0.00	0.31	48.60	NA	NA	0.00	0.00	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	2005	1.18	0.00	0.37	136.10	NA	NA	4.43	0.00	0.00	0.00
PVMC	Port Valdez Mineral Creek	MWP	2007	0.57	0.00	0.98	115.20	6.20	0.00	2.95	0.00	0.00	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1986	5.27	NA	NA	NA	NA	NA	NA	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1987	1.33	NA	NA	NA	NA	NA	NA	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1988	0.00	NA	NA	NA	NA	NA	11.43	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1990	0.00	NA	NA	0.00	NA	NA	0.60	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1991	1.13	0.00	NA	0.00	NA	NA	0.00	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1992	0.00	0.00	NA	12.71	NA	NA	0.00	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1993	0.00	0.00	NA	16.47	NA	NA	5.63	NA	NA	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1995	0.00	NA	5.12	50.18	NA	NA	NA	NA	NA	NA
UISB	Unakwit Inlet Siwash Bay	MWP	1997	0.00	0.00	1.59	30.80	NA	NA	3.33	NA	0.00	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	1999	9.27	4.43	38.28	9.85	NA	NA	20.76	NA	0.00	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	2001	0.00	0.00	1.97	3.20	NA	NA	0.59	0.00	0.00	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	2003	0.00	0.19	0.25	14.80	NA	NA	0.00	0.00	0.00	0.00

Appendix 2. (Continued)

Map Code	Site Name	Group	Year	Total DDTs	Total Dieldrins	Total HCHs	Total PAHs	Total PBDEs	Total PBBs	Total PCBs	Total Endosulfans	Chlorpyrifos	Mirex
UISB	Unakwit Inlet Siwash Bay	MWP	2005	0.38	0.56	1.14	16.90	NA	NA	1.81	0.00	0.00	0.00
UISB	Unakwit Inlet Siwash Bay	MWP	2007	0.24	0.41	0.94	3.70	0.70	0.00	1.55	0.00	0.00	0.00
GBBC	GB Bartlett Cove	SEAN	2013	0.00	0.61	0.00	11.70	NA	NA	5.13	0.00	0.00	0.00
GBBC	GB Bartlett Cove	SEAN	2015	0.51	0.00	0.00	6.63	NA	NA	0.00	0.00	0.00	0.00
GBBC	GB Bartlett Cove	SEAN	2017	0.37	0.00	0.00	27.59	0.00	0.00	0.00	0.00	0.00	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2013	0.71	0.50	0.39	10.76	NA	NA	9.65	0.00	0.00	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2015	0.00	0.50	0.00	4.79	NA	NA	0.00	0.00	0.00	0.00
GBEI	GB Lower Excursion Inlet	SEAN	2017	0.00	0.00	0.00	16.95	0.00	0.00	0.50	0.00	0.00	0.00
GBHC	GB W. Hazelton Camp	SEAN	2013	0.33	0.00	0.00	0.00	NA	NA	10.60	0.00	0.00	0.00
GBHC	GB W. Hazelton Camp	SEAN	2015	0.38	0.00	0.00	3.45	NA	NA	0.00	0.00	0.00	0.00
GBHC	GB W. Hazelton Camp	SEAN	2017	0.00	0.00	0.00	10.31	0.00	0.00	0.00	0.00	0.00	0.00
GBRC	GB Ripple Cove	SEAN	2013	1.57	0.92	0.53	7.97	NA	NA	5.14	0.00	1.82	0.00
GBRC	GB Ripple Cove	SEAN	2015	0.83	0.41	0.00	0.00	NA	NA	0.00	0.00	0.00	0.00
GBRC	GB Ripple Cove	SEAN	2017	0.46	0.00	0.00	12.36	0.00	0.00	0.00	0.00	0.00	0.00
GBRI	GB E. of Russell Island	SEAN	2013	0.00	1.23	18.27	0.00	NA	NA	1.97	0.00	0.00	0.00
GBRI	GB E. of Russell Island	SEAN	2015	0.00	0.37	0.00	0.00	NA	NA	0.00	0.00	0.00	0.00
GBRI	GB E. of Russell Island	SEAN	2017	0.00	0.00	0.00	16.02	0.00	0.00	0.59	0.00	0.00	0.00
SITK	Sitka Visitor's Center	SEAN	2013	6.46	0.67	0.00	191.84	NA	NA	61.82	0.00	0.00	0.00
SITK	Sitka Visitor's Center	SEAN	2015	0.82	0.00	0.00	108.36	NA	NA	35.77	0.00	0.00	0.00
SITK	Sitka Visitor's Center	SEAN	2017	1.61	0.00	0.00	242.32	0.00	0.00	31.06	0.00	0.00	0.00
SKWY	Skagway Harbor	SEAN	2017	0.00	0.00	0.00	6465.44	0.00	0.00	0.00	0.00	0.00	0.00
AMAB	KA Amalik Bay	SWAN	2007	0.00	0.00	0.00	13.10	NA	NA	0.00	0.00	0.00	0.00
AMAB	KA Amalik Bay	SWAN	2018	0.00	0.00	0.00	6.19	NA	NA	0.00	0.00	0.00	0.00
CIAB	KF Aailik Bay	SWAN	2007	0.00	0.00	0.00	10.70	NA	NA	0.98	0.00	0.00	0.00
CIAB	KF Aailik Bay	SWAN	2018	0.00	0.00	0.00	6.69	NA	NA	0.00	0.00	0.00	0.00
CIHB	KF Harris Bay	SWAN	2007	0.00	0.52	0.80	0.00	NA	NA	0.77	0.00	0.00	0.00
CIHB	KF Harris Bay	SWAN	2018	0.00	0.00	0.00	5.17	NA	NA	0.00	0.00	0.00	0.00
CIMF	KF McCarty Fjord	SWAN	2007	0.00	0.28	0.32	9.80	NA	NA	0.74	0.00	0.00	0.00
CIMF	KF McCarty Fjord	SWAN	2018	0.00	0.00	0.00	4.71	NA	NA	0.00	0.00	0.00	0.00
CINB	KF Nuka Bay	SWAN	2007	0.00	0.40	0.00	13.30	NA	NA	0.00	0.00	0.00	0.00
CINB	KF Nuka Bay	SWAN	2018	0.00	0.00	0.00	5.04	NA	NA	0.00	0.00	0.00	0.00
CINI	KA Ninagiak Island	SWAN	2007	0.00	0.40	0.76	21.90	NA	NA	0.00	0.00	0.00	0.00
CINP	KF Nuka Passage	SWAN	2007	0.22	0.43	0.96	7.30	NA	NA	0.74	0.00	0.00	0.00
CINP	KF Nuka Passage	SWAN	2018	0.00	0.00	0.00	8.50	NA	NA	0.00	0.00	0.00	0.00
EPGB	EPWS Galena Bay	SWAN	2012	0.40	0.49	0.00	17.24	7.81	0.00	1.01	0.00	0.00	0.00
EPOI	EPWS Observation Island	SWAN	2012	0.48	0.45	0.00	31.06	3.76	0.00	0.00	0.00	0.00	0.00
EPPF	EPWS Port Fildalgo	SWAN	2012	4.70	0.53	0.00	32.90	12.03	0.00	1.79	0.00	0.00	0.00
EPSB	EPWS Simpson Bay	SWAN	2012	0.00	0.00	0.00	14.32	8.23	0.00	0.68	0.00	0.00	0.00
KAFB	KA Kaflia Bay	SWAN	2007	0.00	0.00	0.00	15.00	NA	NA	0.00	0.00	0.00	0.00
KAFB	KA Kaflia Bay	SWAN	2018	0.00	0.00	0.00	7.66	NA	NA	0.00	0.00	0.00	0.00
KBBB	KB Bishop's Beach	SWAN	2018	0.00	0.00	0.00	9.26	NA	NA	0.00	0.00	0.00	0.00
KBBP	KB Bluff Point	SWAN	2018	0.00	0.00	0.00	7.95	NA	NA	0.00	0.00	0.00	0.00
KBCI	KB Cohen Island	SWAN	2012	0.00	0.31	0.00	10.99	0.00	0.00	0.94	0.00	0.00	0.00
KBCI	KB Cohen Island	SWAN	2018	0.00	0.00	0.00	4.46	NA	NA	0.00	0.00	0.00	0.00
KBEI	KB Elephant Island	SWAN	2018	0.00	0.00	0.00	5.78	NA	NA	0.00	0.00	0.00	0.00
KBOB	KB Outside Beach	SWAN	2012	0.00	0.34	0.00	23.88	10.04	0.00	1.16	0.00	0.00	0.00
KBOB	KB Outside Beach	SWAN	2018	0.00	0.00	0.00	14.92	NA	NA	0.93	0.00	0.00	0.00
KBPG	KB Port Graham	SWAN	2018	0.00	0.00	0.00	6.32	NA	NA	0.00	0.00	0.00	0.00
KINB	KA Kinak Bay	SWAN	2007	0.00	0.00	0.00	12.60	NA	NA	0.00	0.00	0.00	0.00
KINB	KA Kinak Bay	SWAN	2018	0.00	0.00	0.00	6.75	NA	NA	0.00	0.00	0.00	0.00
KUKB	KA Kukak Bay	SWAN	2007	0.31	0.56	1.05	21.50	NA	NA	1.02	0.00	0.00	0.00
KUKB	KA Kukak Bay	SWAN	2018	0.00	0.00	0.00	11.71	NA	NA	0.00	0.00	0.00	0.00
NPBB	NPWS Bettles Bay	SWAN	2013	0.00	0.89	0.00	2.32	NA	NA	0.00	0.00	0.00	0.00

Appendix 2. (Continued)

Map Code	Site Name	Group	Year	Total DDTs	Total Dieldrins	Total HCHs	Total PAHs	Total PBDEs	Total PBBs	Total PCBs	Total Endosulfans	Chlorpyrifos	Mirex
NPCE	NPWS Cedar Bay	SWAN	2013	0.60	0.00	0.00	3.37	NA	NA	0.00	0.00	0.00	0.00
NPPE	NPWS Perry Island	SWAN	2013	0.61	0.00	0.00	6.47	NA	NA	0.00	0.00	0.00	0.00
TAKI	KA Takli Island	SWAN	2007	0.00	0.00	0.00	13.20	NA	NA	0.67	0.00	0.00	0.00
TAKI	KA Takli Island	SWAN	2018	0.00	0.00	0.00	19.02	NA	NA	0.00	0.00	0.00	0.00
WPHE	WPWS Herring Bay	SWAN	2012	0.89	0.34	0.00	4.40	10.97	0.00	1.59	0.00	0.00	0.00
WPHE	WPWS Herring Bay	SWAN	2018	0.35	0.00	0.00	8.49	NA	NA	0.00	0.00	0.00	0.00
WPHO	WPWS Hogan Bay	SWAN	2012	0.00	0.33	0.00	5.02	0.00	0.00	0.79	0.00	0.00	0.00
WPHO	WPWS Hogan Bay	SWAN	2018	0.00	0.00	0.00	5.44	NA	NA	0.00	0.00	0.00	0.00
WPIB	WPWS Iktua Bay	SWAN	2012	0.83	0.48	0.00	4.17	0.00	0.00	2.83	0.00	0.00	0.00
WPIB	WPWS Iktua Bay	SWAN	2018	0.00	0.00	0.00	5.42	NA	NA	0.00	0.00	0.00	0.00
WPJB	WPWS Johnson Bay	SWAN	2012	0.00	0.00	0.00	12.37	33.28	0.00	0.00	0.00	0.00	0.00
WPJB	WPWS Johnson Bay	SWAN	2018	0.00	0.00	0.00	10.75	NA	NA	0.00	0.00	0.00	0.00
WPWB	WPWS Whale Bay	SWAN	2012	0.00	0.32	0.00	NA	5.99	0.00	3.03	0.00	0.00	0.00
WPWB	WPWS Whale Bay	SWAN	2018	0.00	0.00	0.00	7.87	NA	NA	0.00	0.00	0.00	0.00

Supplemental Material

SAMPLING FREQUENCY RECOMMENDATIONS

INTRODUCTION

As part of the objectives outlined by the interagency agreement between the United States Department of the Interior (DOI), National Park Service (NPS), Southeast Alaska Network (SEAN) and Southwest Alaska Network (SWAN), and the United States Department of Commerce (DOC), National Oceanic and Atmospheric Administration (NOAA), National Centers for Coastal Ocean Science (NCCOS), the National Status and Trends (NS&T) program agreed to "recommend sampling frequency at long-term sites and potential need for sampling new sites in Arctic parklands that will experience more frequent vessel traffic in the future".

Some specific follow-up questions were as follows:

> 1) *Given the findings to date, is there support for shifting to a 4 or 5 year sampling frequency for all or a subset of the contaminant classes? One consideration on my end is how the longer period among samples might affect the utility of our data for establishing a suitable baseline in the event of a spill for the purposes of damage assessment (NRDA). Of course, there are funding and workload implications to take into account as well. I think we would primarily be asking for your perspective based on the variability among years for key analytes.*
>
> 2) *Is there merit to sampling every two years, but only analyzing samples every four years, sending the intervening sample to an archive such as the NIST Marine ESB in SC? I see a few potential advantages to this approach, particularly if there is little variation among our prior samples. First, there are potential cost and workload savings to our program. Next, we don't always know what analytes are of concern at the time of sampling. Archived samples could be analyzed at a future date to confirm the presence of emerging contaminants of concern, or for trend analysis if we need additional power. Of course, archived samples could also be analyzed in the event of a spill to gain a more current baseline.*
>
> – Michael Bower, pertaining to SEAN

In this document, we attempt to address this objective and these questions by examining the variability in the existing data, and thus making an informed recommendation for future monitoring activities in the Alaska region. Additionally, we provide guidance on archiving opportunities and requirements but emphasize the importance of making a programmatic decision due to the resources required for long-term archiving.

RECOMMENDATIONS

Optimal sampling frequency determination for a contaminant monitoring program could take into account a number of factors including variability in existing data, knowledge about existing and possible contamination risks at sites, funding and resources available for long-term monitoring, etc. Herein, we attempt to make a sampling frequency recommendation for Southeast Alaska Network (SEAN) based on existing monitoring data for the Alaska region. The report 'A Synthesis of Ten Years of Chemical Contaminant Monitoring Data in National Park Service - Southeast and Southwest Alaska Networks' summarizes the monitoring data collected by National Park Service and NOAA's Mussel Watch Program (MWP). These monitoring programs/entities have very different spatial and temporal coverage; nevertheless, there is valuable information in a decade of monitoring data that can be examined to inform future sampling frequency.

For the data included in this report, Mussel Watch Program has long-term monitoring data from 1986-2018 albeit from a very small number of sites Southwest Alaska network (SWAN) has almost four times more sites than Mussel Watch but with more recent data from 2007-2018, sampled at 5-11 year intervals. SEAN has roughly the same number of sites as Mussel Watch with data starting from 2013 and sampled thereafter at 2-year intervals until 2017. Since SEAN only has 3 years of data that we could analyze, we leveraged the data from SWAN and Mussel Watch from the region to make an informed decision for future sampling frequency for SEAN. We examined the data across these programs using box and whisker plot for each contaminant, the objective of which was solely to observe whether the data were randomly spread across years. The randomness in the years around the median line of the box plot among each program suggest that variability of the concentrations measured are randomly spread for almost all the contaminants (Figures 1-15). In other words, we did not observe any temporal pattern in the contaminants data across three programs that represent a wide range of monitoring data, both temporally and spatially. Further, a linear model analysis of data to examine contaminant concentration differences between programs was conducted. Linear model estimates indicate that despite temporal (MWP: 1986-2018; SEAN: 2013-2017; SWAN: 2007-2018) and spatial sampling (MWP-6; SEAN-7; SWAN-29) differences, the majority of the contaminants showed no differences (ND) in mean estimates among programs (Table 1). Given these results, we opine that SEAN can switch to sampling on a 4/5 year frequency and it will not negatively affect future chemical concentration analyses.

Additionally, comparison of Alaska monitoring data to the low, medium and high groupings of National Status and Trends data revealed that the majority of the concentrations in Alaska fell within the low group and therefore can be considered to be at background levels (Figure 35 from report). Those concentrations that were in the highest clusters were mainly metals (cadmium, copper and nickel). Given that mussels can regulate the uptake of metals and the fact that metals occur naturally, the observed high concentrations do not warrant specialized monitoring. However, PAHs and PBDEs were not uniformly high, rather only at very few select sites. Organic contaminants that have current and ongoing sources such as PAH and more recently banned PBDEs (compared to legacy contaminants) may require further scrutiny through consistent monitoring at high-risk sites. Depending on the funding and resources, SEAN can perhaps conduct targeted biennial sampling for PAHs and PBDEs or collect and archive samples for retrospective analyses as needed.

The scientific merits beyond preserving samples for routine analyses of core analytes are that archiving may provide other valuable services including the opportunities: (1) to measure "new" contaminants that were overlooked or unknown during the real-time monitoring, (2) to use improved analytical methods, and (3) to verify previous results. Depending on the goals set forth by SEAN and availability of funds, different archiving techniques may be utilized to preserve sample integrity for later use. These may range from a laboratory technique to preserve homogenized or whole samples for two to five years, to a longer-term archiving, which may be more involved. For example, SEAN could make programmatic decisions for:

1) A short-term archiving approach working with laboratories to preserve samples in -20 to -70^{o}C freezers equipped with adequate monitoring systems.

2) A long-term archiving approach based on a cryogenic storage technique that uses a special liquid nitrogen vapor phase freezer like the one used by the National Institute of Standard and Technology (NIST) for the now ceased Mussel Watch's Specimen Banking Program.

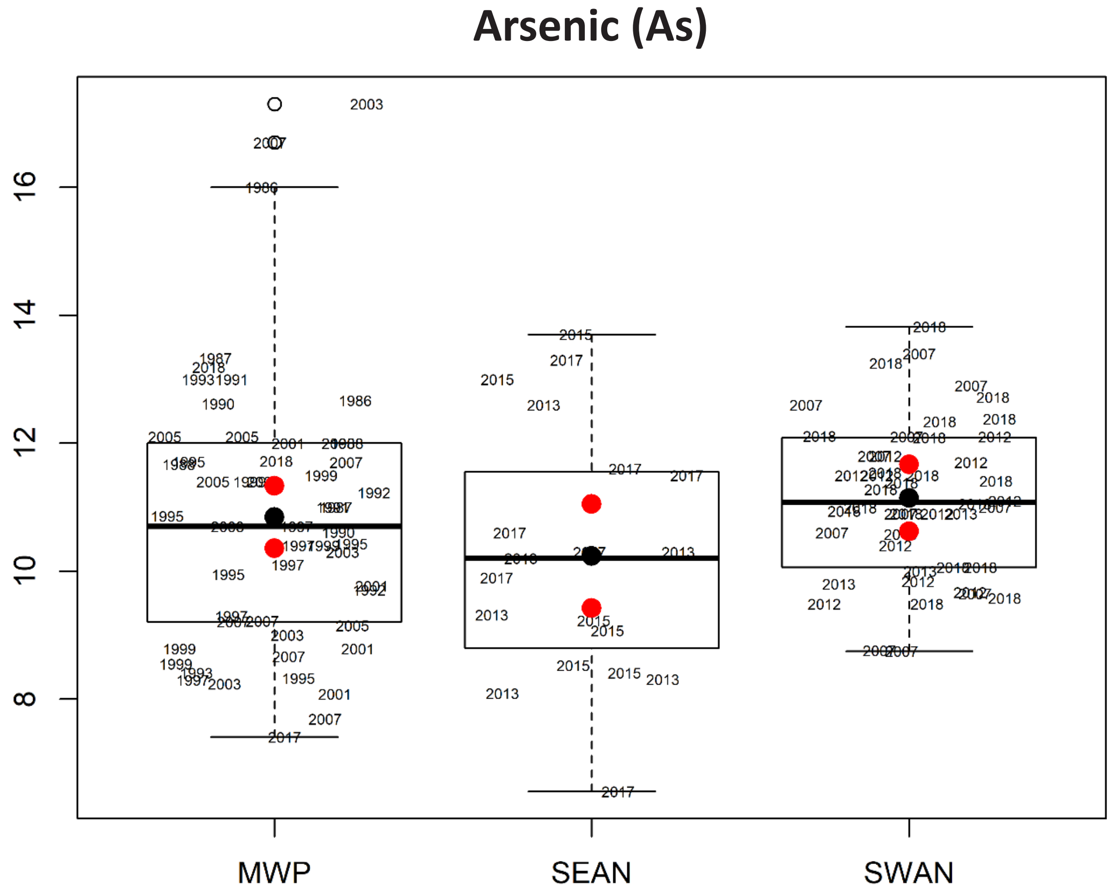
SUPPLEMENTAL MATERIAL

Table 1. Multiple Comparisons of chemical contaminant concentration estimates among programs. Non-overlapping 95 % confidence intervals indicate significant difference (lower or higher) among programs. For example, the mean estimate for Copper in MWP is significantly lower than SEAN. Overlapping 95 % confidence intervals indicate no significant difference (ND) among programs.

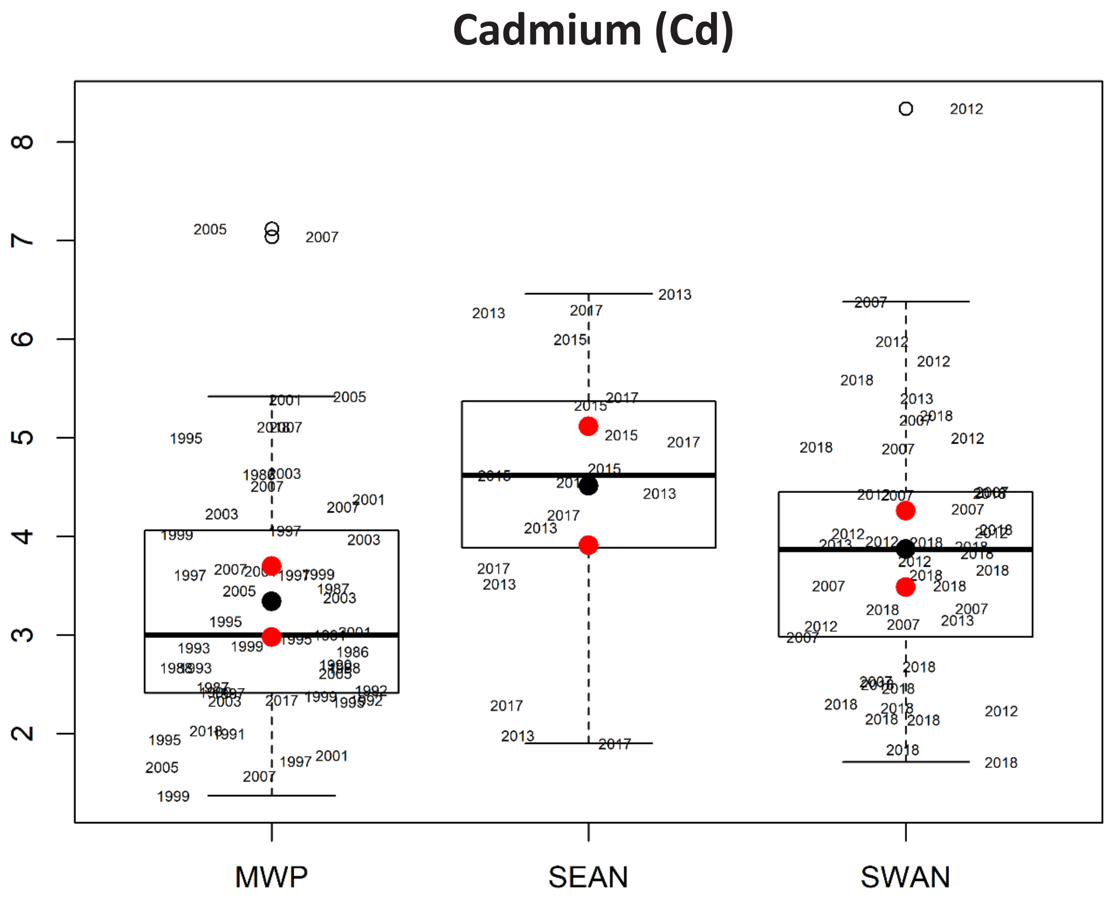
	MWP vs SEAN	MWP vs SWAN	SEAN vs SWAN	Figure
Arsenic	ND	ND	ND	1
Cadmium	ND	ND	ND	2
Copper	lower	lower	ND	3
Lead	lower	ND	higher	4
Mercury	higher	ND	ND	5
Nickel	lower	ND	ND	6
Total Butyltins	ND	ND	ND	7
Total Chlordanes	ND	ND	ND	8
Total Chlorobenzenes	higher	higher	ND	9
Total DDTs	ND	ND	ND	10
Total Dieldrins	ND	ND	ND	11
Total HCHs	ND	ND	ND	12
Total PAHs	ND	ND	ND	13
Total PBDEs	ND	ND	ND	14
Total PBBs	ND	ND	ND	15

Figures 1-15. Boxplot of contaminant data from each monitoring program in Alaska region (MWP: 1986-2018; SEAN: 2013-2017; SWAN: 2007-2018) with different spatial resolution (MWP-6 sites; SEAN-7 sites; SWAN-29 sites). Black points (•) identify the linear model mean estimate. Red points (•) identify the lower and upper 95% confidence intervals.

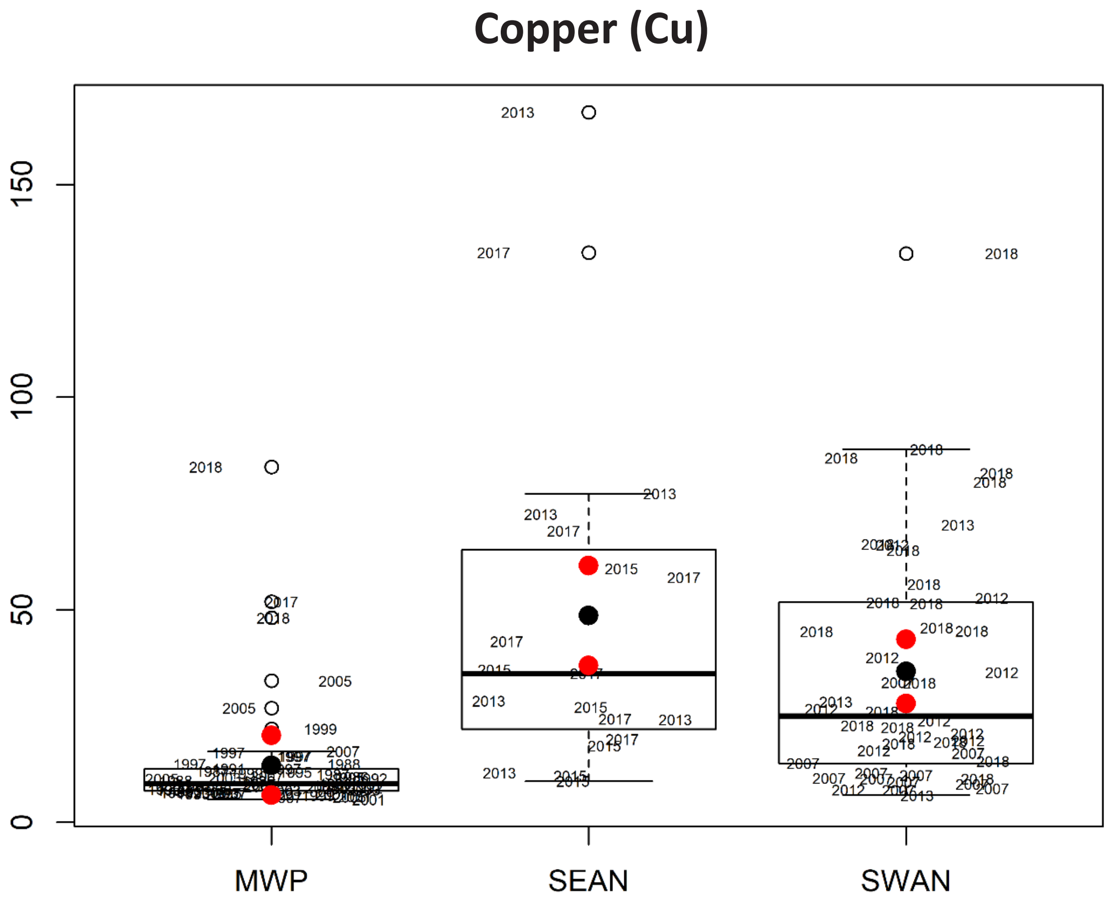
1)



2)

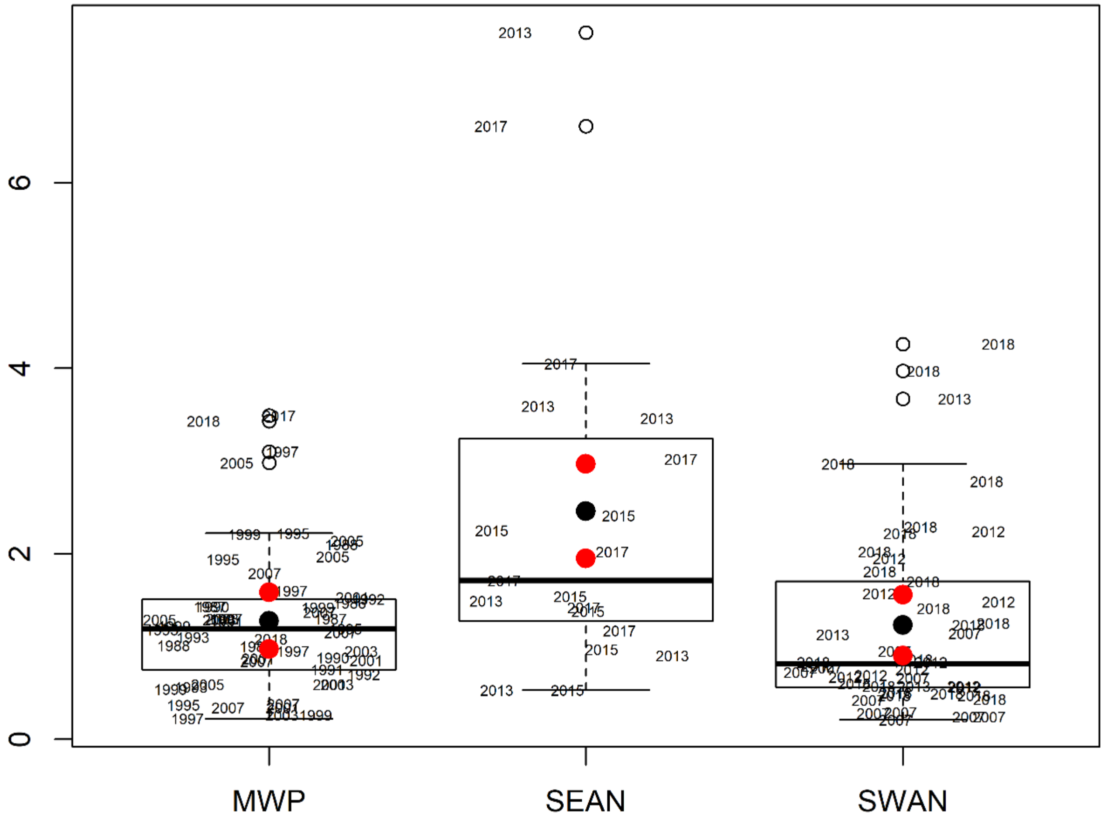

Cadmium (Cd)
8
7
6
5
4
3
2
MWP
SEAN
SWAN
2012
2005
2007
2013
2017
2015
1995
1999
2001
2003
2018

3)


Copper (Cu)
150
100
50
0
MWP
SEAN
SWAN
2013
2017
2018
2015
2012
2005
1999
1997
2007
2001

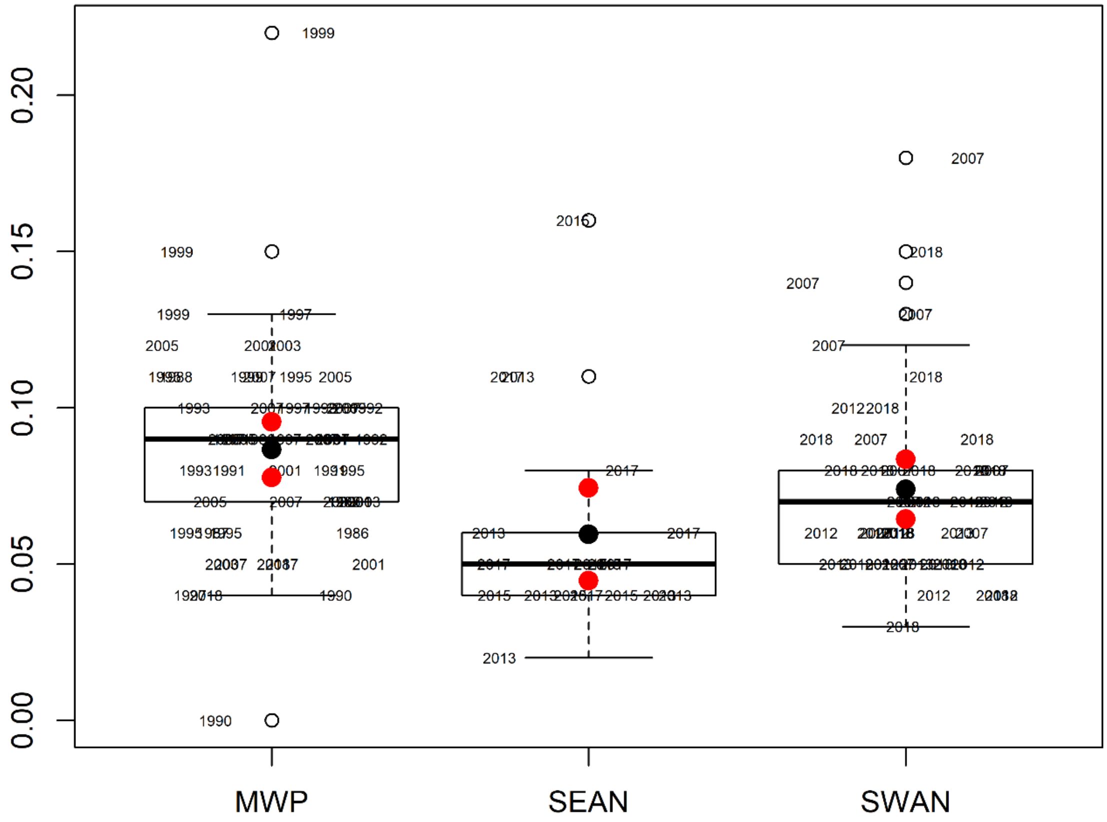
4)

Lead (Pb)



5)

Mercury (Hg)



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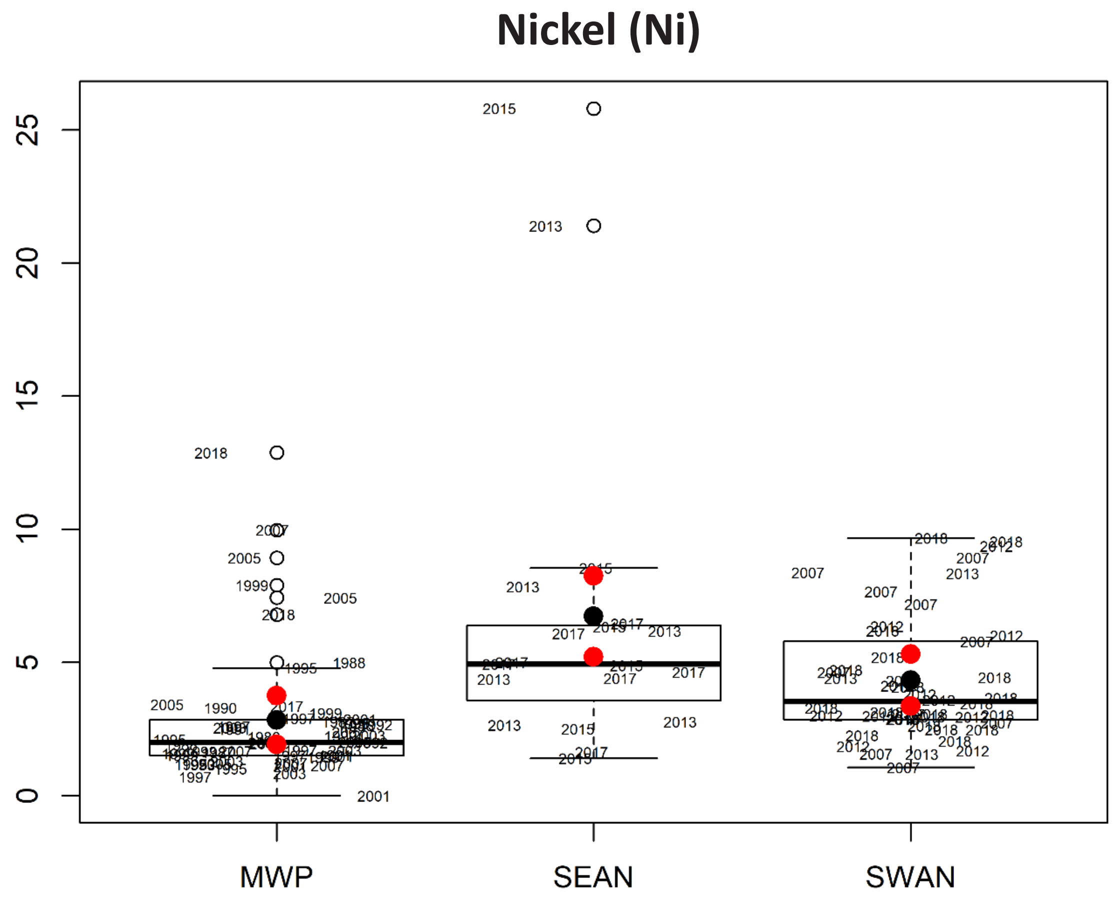
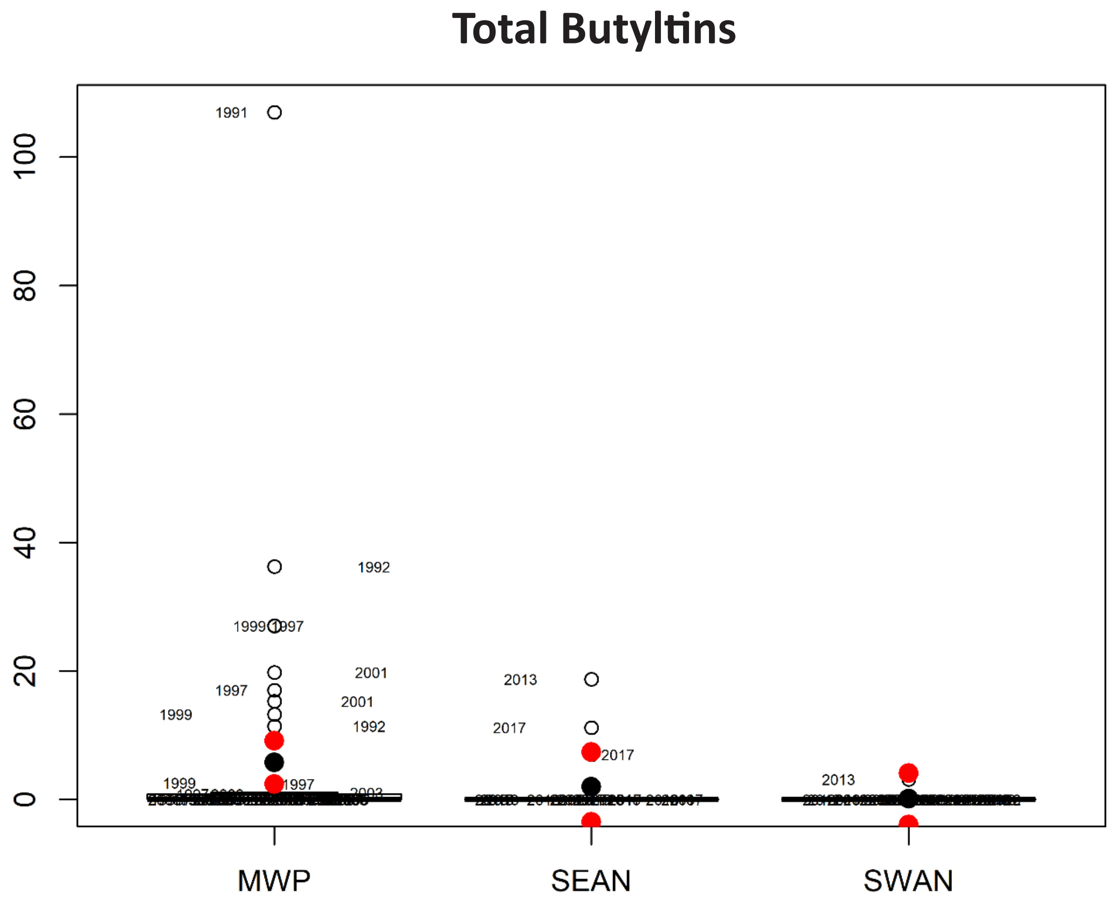
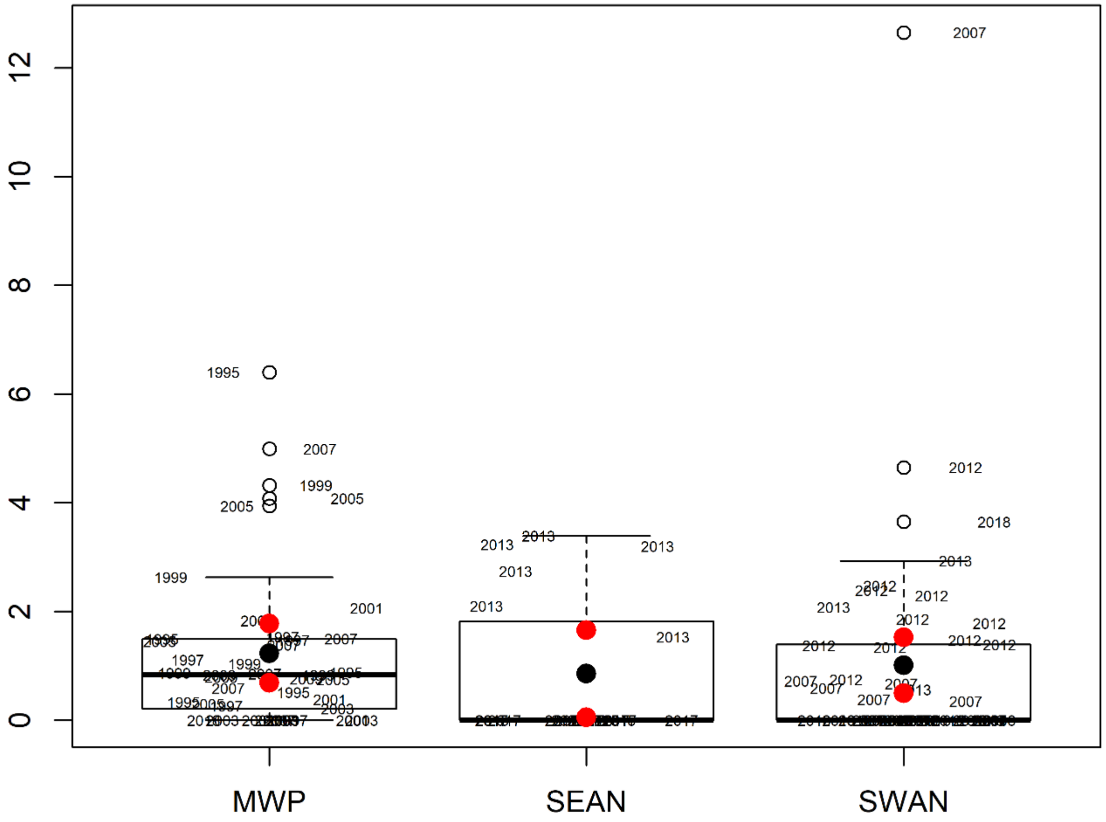

Nickel (Ni)
25
20
15
10
5
0
2015
2013
2018
2007
2005
1999
2005
2018
1995
1988
2001
MWP
SEAN
SWAN

7)


Total Butyltins
100
80
60
40
20
0
1991
1992
1999
1997
2001
2001
1992
1997
1999
2013
2017
2017
2013
MWP
SEAN
SWAN

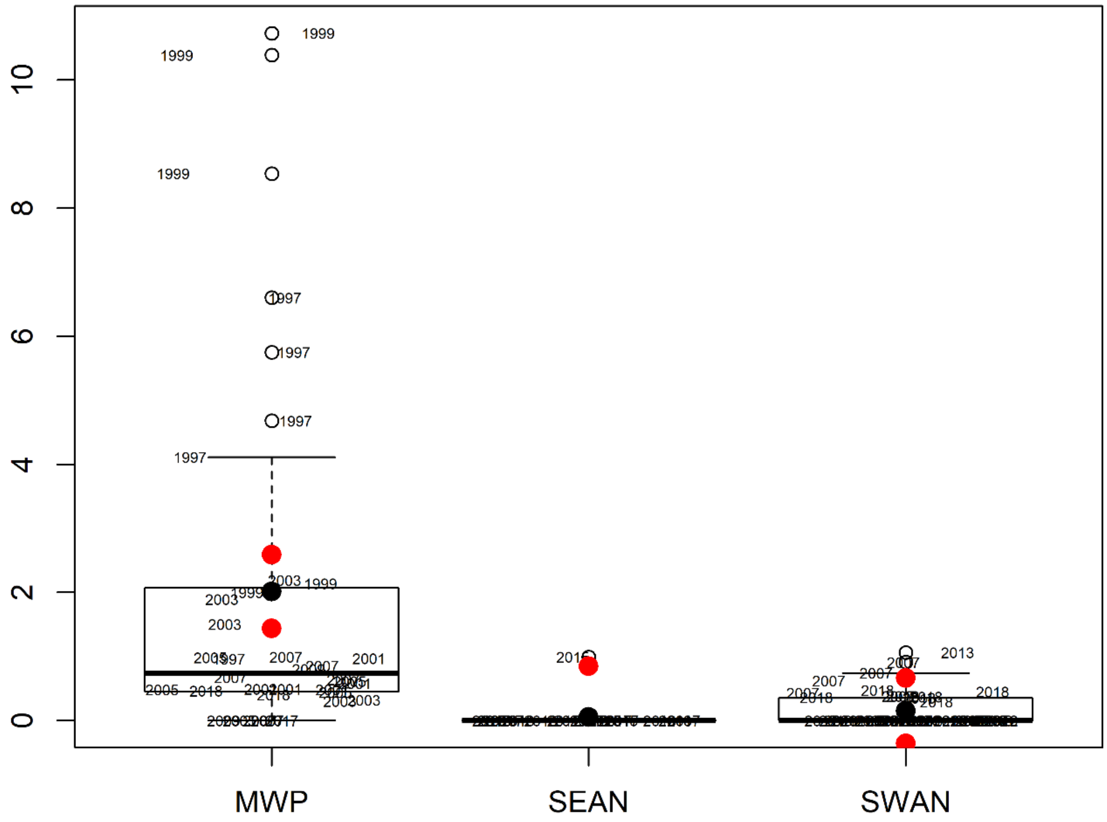
8)

Total Chlordanes



9)

Total Chlorobenzenes



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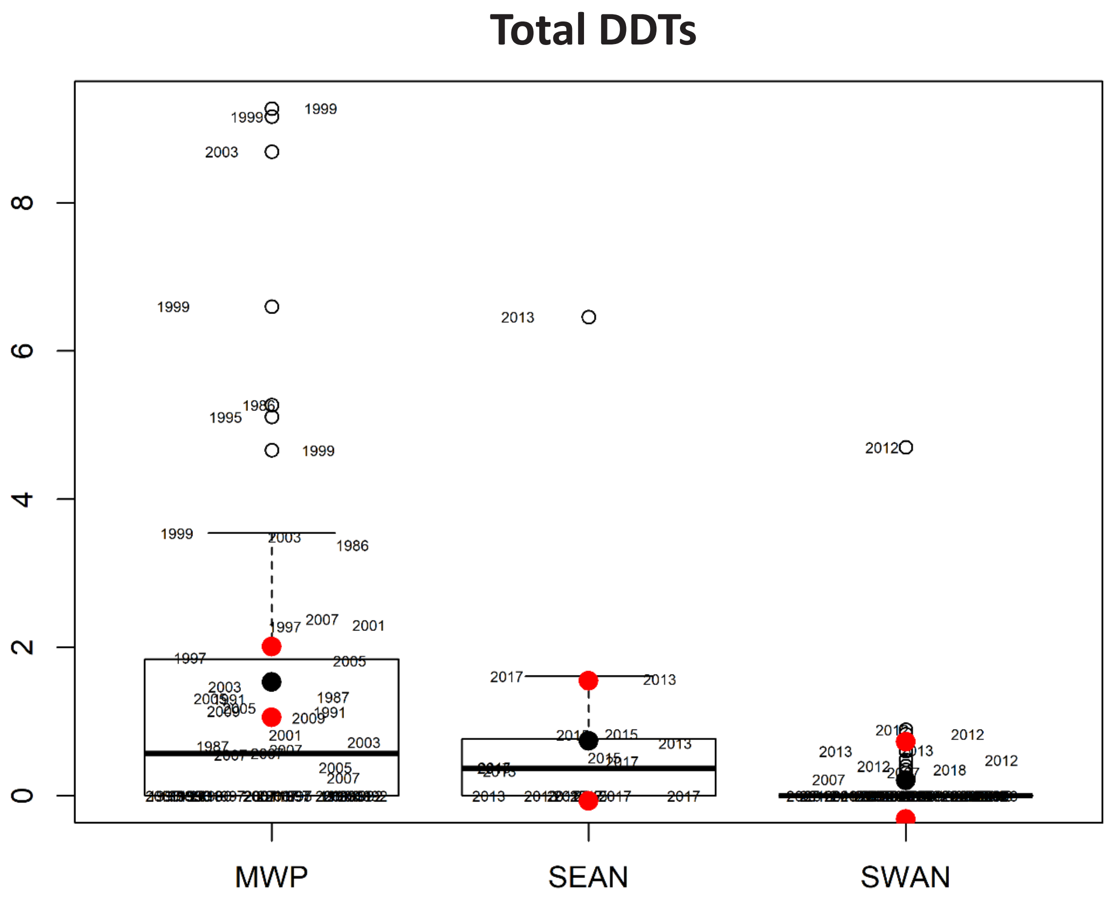
Total DDTs
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2
4
6
8
1999
1999
2003
1999
1995
1986
1999
1999
2003
1986
1997
2007
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2005
2003
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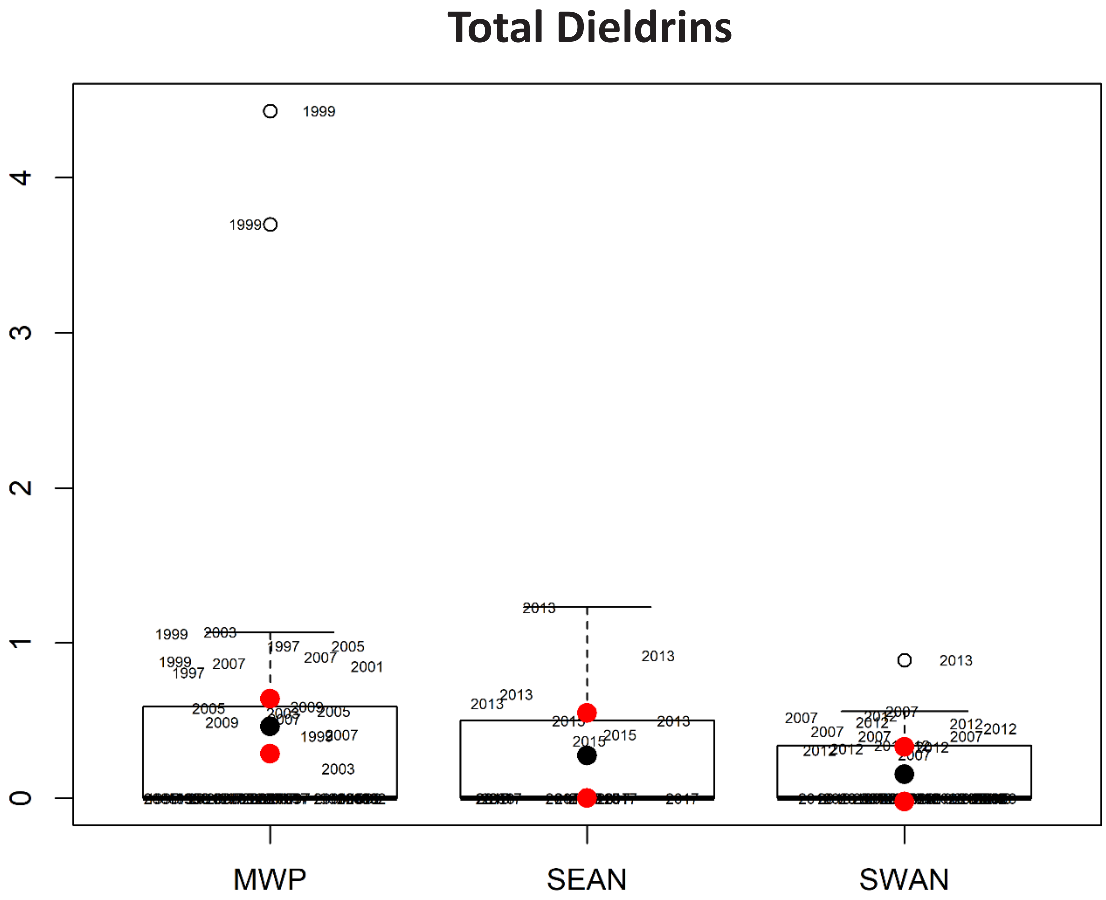
MWP
SEAN
SWAN

11)


Total Dieldrins
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1
2
3
4
1999
1999
1999
2003
1997
2005
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1997
2007
2007
2001
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2003
2007
2009
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2012
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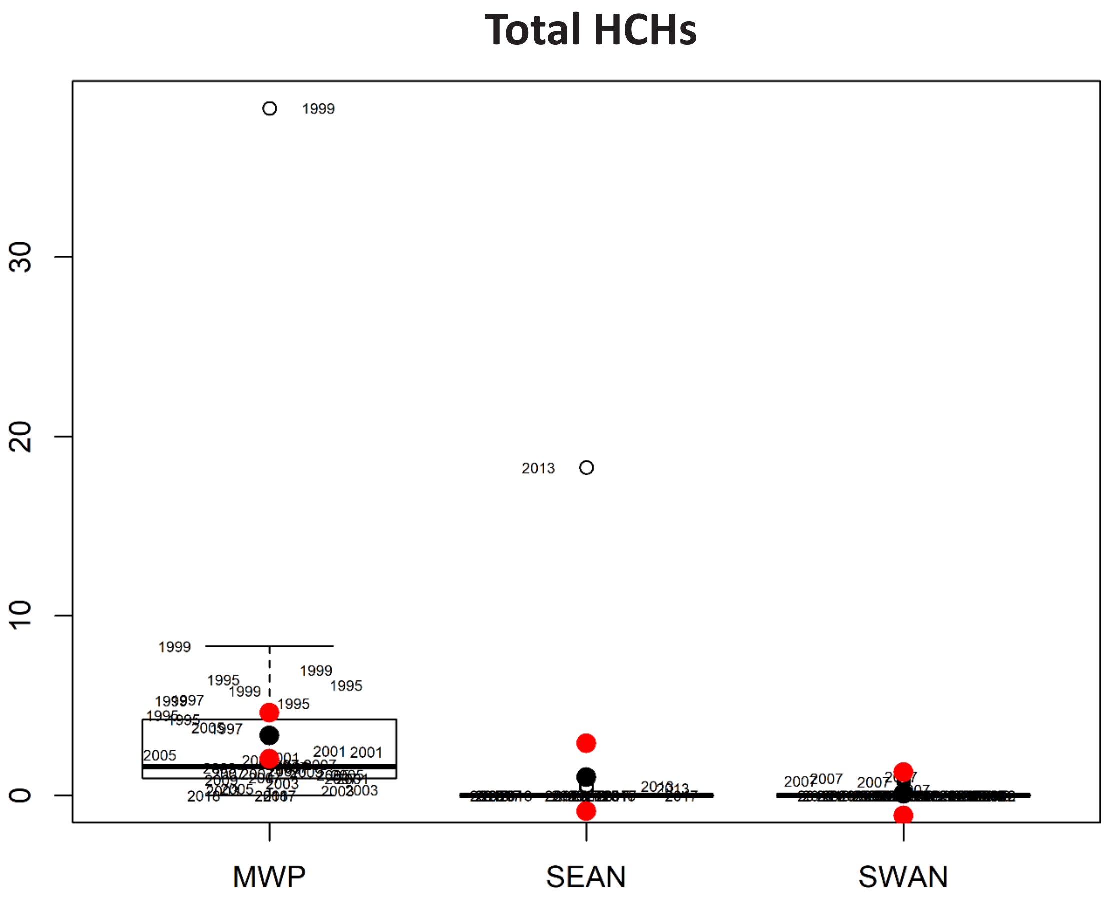
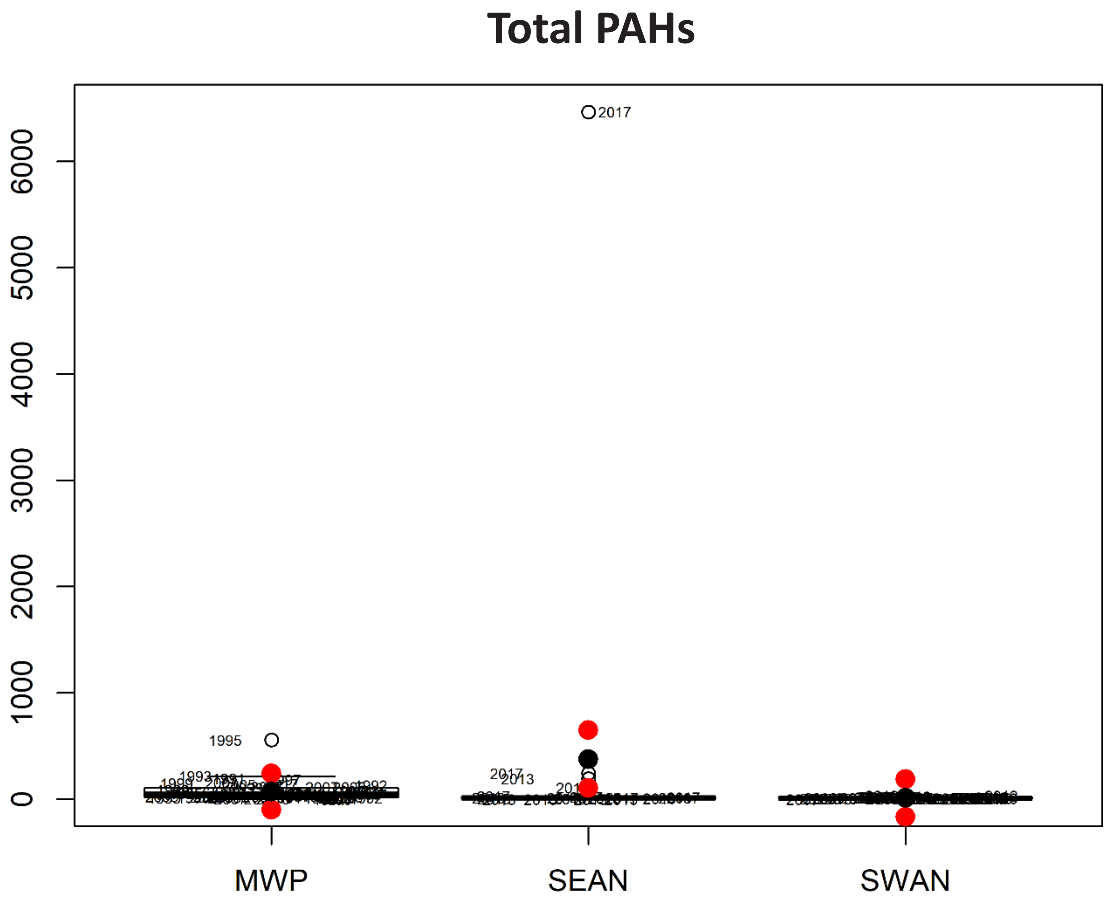
2012
2012
2012
2007
MWP
SEAN
SWAN

12)

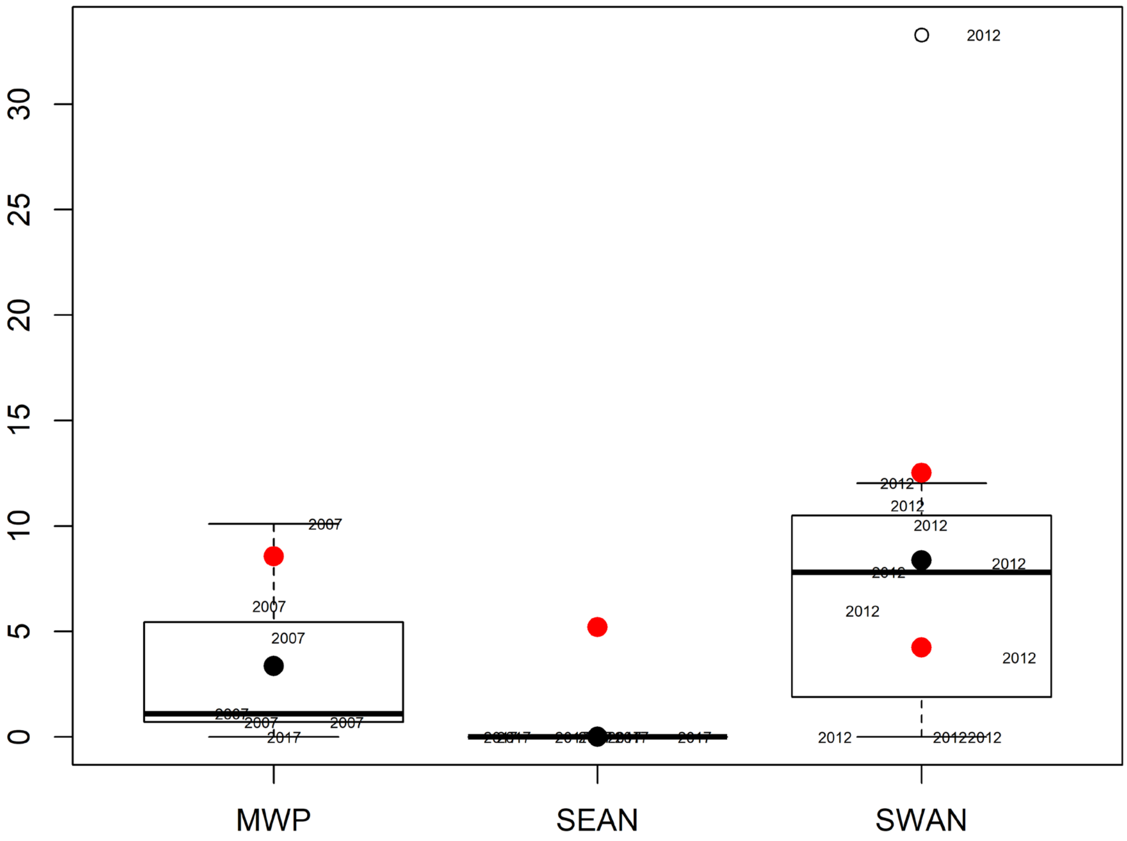

Total HCHs
0
10
20
30
MWP
SEAN
SWAN
1999
2013
1999
1995
1999
1999
1995
1995
2005
2001 2001
2003
2007
2007
2007

13)


Total PAHs
0
1000
2000
3000
4000
5000
6000
MWP
SEAN
SWAN
2017
1995
1993
2017
2013

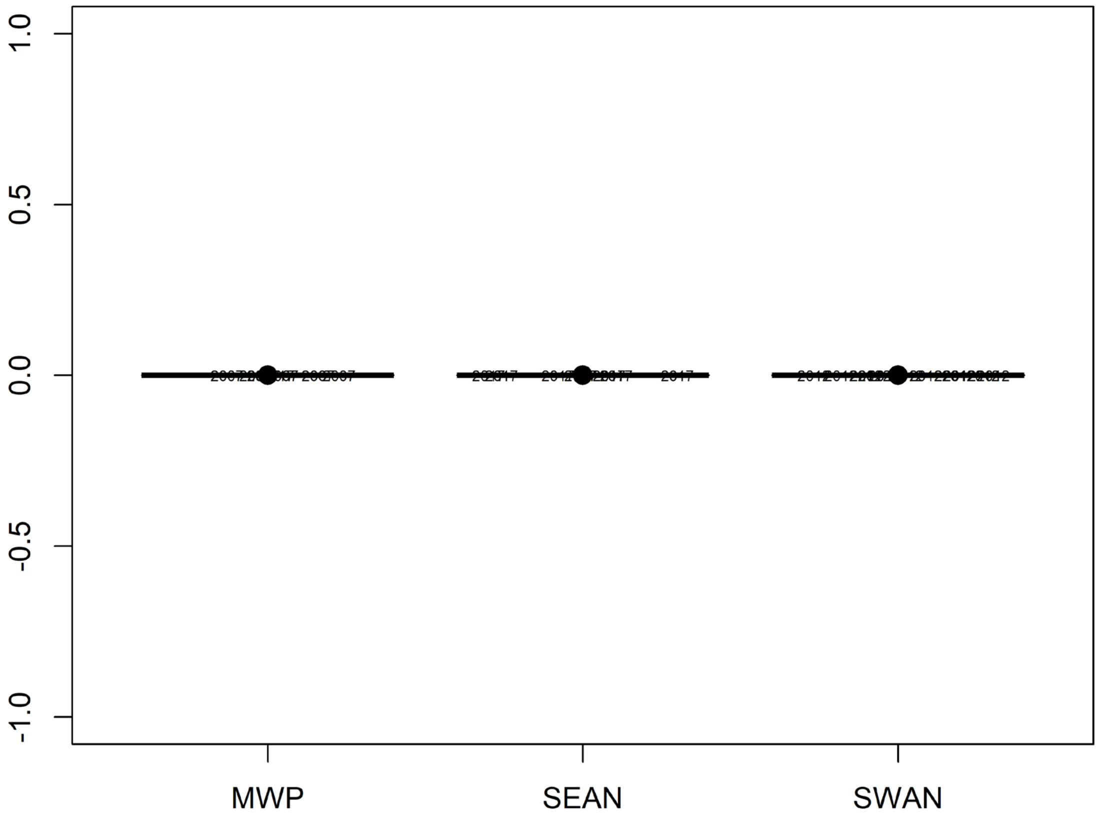
14)

Total PBDEs



15)

Total PBBs



U.S. Department of Commerce

Wilbur L. Ross, Jr., *Secretary*

National Oceanic and Atmospheric Administration

Neil Jacobs, *Acting Under Secretary of Commerce for Oceans and Atmosphere*

National Ocean Service

Nicole LeBoeuf, *Acting Assistant Administrator for National Ocean Service*

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