Final

Long-Term Environmental Monitoring Program

2022–2023 Summary Report

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ACRONYMS AND ABBREVIATIONS

ANS	Alaska North Slope
BWTF	Ballast Water Treatment Facility
EPA	U.S. Environmental Protection Agency
EVOS	Exxon Valdez Oil Spill
LTEMP	Long-Term Environmental Monitoring Program
NOAA	National Oceanic and Atmospheric Administration
PAHs	Polycyclic aromatic hydrocarbons
PPB (or ng/g)	Parts Per Billion
PWSRCAC	Prince William Sound Regional Citizens' Advisory Council
UV	Ultraviolet

ABSTRACT

To understand the environmental impact, fate, and source of hydrocarbons related to the operations of Alyeska Pipeline Service Company's Valdez Marine Terminal, hydrocarbon concentrations were monitored in sediments, in intertidal Pacific blue mussels, and in the water via passive sampling devices. In the 2022 and 2023 results, we see low levels of petroleum (petrogenic) hydrocarbons in sediments at the terminal that can be attributed to terminal operations. Passive water sampling devices and Pacific blue mussels from all sampled locations had low levels of toxic hydrocarbons. Sediment and mussels sampled from sites away from the terminal in Port Valdez contained more combustion (pyrogenic) related compounds than detected at the terminal. In 2022, mussels from the Valdez Small Boat Harbor had the highest levels of hydrocarbons, likely due to frequent small spills and heavy human activity not forensically attributed to terminal operations. In 2023, higher polycyclic aromatic hydrocarbons (PAH) levels were found in some mussel samples at Knowles Head in northeastern Prince William Sound than those in the harbor. Other mussel sites sampled in 2023 as part of the expanded sampling regime included Disk Island, Zaikof Bay, a new site in outer Zaikof Bay, Sleepy Bay, and Sheep Bay. Generally, the expanded sampling sites had comparable PAH levels to annual sampling sites (e.g., Gold Creek and sites near the terminal) with low potential hydrocarbon ecotoxicity for organisms.

In 2022 and 2023, the hydrocarbons detected by the Long-Term Environmental Monitoring Program sampling, and determined to be from the terminal and tankers, posed low potential ecotoxicological risk. Since 1993, hydrocarbon concentrations are generally low with localized spikes corresponding with spill events like the April 2020 oil spill at the terminal. Following an all-time low in the mid-2010s, hydrocarbon concentrations detected in sediments and mussels have slowly increased across all sites but are still below any threshold for adverse effects on aquatic life. Prince William Sound-wide trends in these hydrocarbon concentrations may be influenced by environmental factors such as increased freshwater input, glacial melt, and warming ocean temperatures. We recommend that future monitoring efforts maintain the current three-matrix design and attempt to preserve, economize, and modernize aspects of Prince William Sound Regional Citizens' Advisory Council's Long-Term Environmental Monitoring Program.

1. INTRODUCTION

The Long-Term Environmental Monitoring Program (LTEMP), managed by the Prince William Sound Regional Citizens' Advisory Council (PWSRCAC), is in its 30th year of monitoring hydrocarbons in the wake of the Exxon Valdez oil spill. Through LTEMP, we are able to determine the source of hydrocarbons and the potential adverse effects on the ecosystem from Alyeska Pipeline Service Company's Valdez Marine Terminal (terminal) and tanker activity. These data have been insightful in understanding the influence of terminal and non-terminal sources of hydrocarbons and environmental factors on hydrocarbon dynamics across Prince William Sound and the Gulf of Alaska.

Hydrocarbons are an extremely diverse group of compounds that make up the bulk of petroleum products like crude oil, fuel, and various maritime products like hydraulic and motor oil. However, hydrocarbons are also readily created by marine and terrestrial plants, locked up in organic sediments and rocks, and produced by combustion. Hydrocarbons in the environment undergo processes called weathering, which includes dissolution, evaporation, ultraviolet (UV) degradation, and microbial degradation. These change the physical and chemical properties of the released oil. Polycyclic aromatic hydrocarbons (PAHs) are a group of hydrocarbons in oil with varying numbers of benzene rings that are relatively resistant to degradation and toxic to living organisms. This group of chemicals tends to adsorb rapidly on suspended materials and sediments, and accumulate in biological tissues once released into the marine environment.

PAHs, as a group, are comprised of hundreds of compounds, each with its own degree of toxicity, and their mixtures can exhibit a wide range of toxicities. Specific hydrocarbons, patterns, and diagnostic compounds (i.e., chemical biomarkers) aid in the identification of specific hydrocarbon sources and are indicative of their weathering history (e.g., degree of weathering, degradation, dissolution). PAH profiles are used to identify petrogenic (of crude oil origin) or pyrogenic (of combustion origin), based on well-established pattern changes (e.g., on the ratio of parent and alkylated compounds). Chemical biomarkers, comprising the hopanes, steranes, terpenes, triaromatic, and monoaromatic steroids, are much more resistant to degrading in the environment and thus used to confirm sources (e.g., between different crude oils) even when the PAH patterns are heavily weathered. Saturated hydrocarbons (n-alkanes) are used to identify naturally occurring plant hydrocarbons and determine the degree of weathering and biodegradation.

While many aquatic organisms like fish can metabolize PAHs, marine invertebrates, such as Pacific blue mussels, are less able to efficiently metabolize these compounds, remain sedentary in a fixed location, filter particles from their immediate surroundings, and therefore serve as efficient natural samplers and indicators of overall environmental PAH exposure (Neff and Burns 1996). Toxic responses to PAHs in aquatic organisms include inhibiting reproduction, developmental effects, tissue damage, cellular stress, oxidative stress, damage to genetic material, and mortality. While the body of knowledge on the adverse effects of petroleum exposure is immense, specifics regarding PAH mixtures, exposure routes, duration and magnitude, species and life stages exposed, and other environmental factors that may act synergistically on organisms, challenge the predictive ability of any hydrocarbon study and necessitate the continued monitoring efforts of LTEMP.

The ubiquity of hydrocarbons in the environment complicates tracing sources, understanding ecotoxic thresholds, and following dynamics over time and space. Environmental samples, like sediments, can accumulate multiple hydrocarbon sources over time, resulting in a mixed or unresolved profile. Organisms such as blue mussels can accumulate, eliminate, or alter hydrocarbon compounds, which complicates the task of identifying the sources. Passive sampling devices are specifically designed to complement the biological and toxicological interpretations by measuring just the dissolved compounds available to aquatic organisms (the bioavailable fraction) but are not well suited for hydrocarbon forensics. Sources investigated for the present study are those associated with terminal operations, including Alaska North Slope (ANS) crude oil (which is pumped through the trans-Alaska pipeline and is loaded into tankers at the terminal), effluent from the Ballast Water Treatment Facility (BWTF) at the terminal, and samples from recent spills at the terminal.

The following study presents the 2022 and 2023 results from the LTEMP and aims to determine:

- The extent, if any, that the terminal and associated tankers' hydrocarbon fingerprint is present in 2022 and 2023 samples with varying ranges from the terminal.
- The potential ecotoxicological risk posed by the measured hydrocarbon contribution from the terminal and tankers.
- The historical trends, ecotoxicological risk, and hydrocarbon fingerprint from mussels collected from extended sampling sites across greater Prince William Sound in 2023.
- Other factors (e.g., environmental or anthropogenic) that may be influencing hydrocarbon presence and composition in 2022 and 2023 samples, and the ecotoxicological relevance of these results.
- Recommendations for future monitoring of petroleum hydrocarbons at the terminal and in Prince William Sound.

2. RESULTS AND DISCUSSION

Sediment, passive sampling device, and Pacific blue mussel tissue samples were collected in June of 2022 and 2023 from annual and quinquennial expanded sampling at LTEMP monitoring stations in Port Valdez and greater Prince William Sound. The sampling program investigated three matrices: sediment, Pacific blue mussels, and water quality. For 2022 and 2023 sediments were sampled at two sites –Alyeska's Valdez Marine Terminal and Gold Creek (Figure 1). In 2022, Pacific blue mussel samples were taken from four sites around the Port of Valdez with a focus on the terminal – Saw Island, Jackson Point, Gold Creek, and Valdez Small Boat Harbor entrance (RED - a site that is chemically different from the ANS terminal source signature and currently acts as a high human use, non-ANS reference site). In 2023, mussels were collected from the four standard sites in Port Valdez in addition to the quinquennial expanded sampling sites of Knowles Head, Disk Island, Zaikof Bay, a new station in outer Zaikof Bay, Sleepy Bay, and Sheep Bay. Three Gulf of Alaska stations (i.e., Aialik Bay, Windy Bay, and Shuyak Harbor) planned to be included in the five-year survey will instead be sampled in 2024 due to weather preventing sampling in 2023. Water was sampled with passive sampling devices at three sites in 2022 — Gold Creek, Jackson Point, and Saw Island. In 2023 passive sampling devices were again deployed at Gold Creek, Jackson Point, and Saw Island, and additional devices were deployed at Knowles Head and Disk Island; the Knowles Head devices could not be relocated due to a severed line and were not retrieved.

Samples were analyzed for PAHs, saturated hydrocarbons, and geochemical petroleum biomarkers using advanced analytical techniques at the NewFields (2022) and Alpha Analytical Laboratory (2023) in Mansfield, Massachusetts (sediments and tissues), and the Oregon State University Food Safety and Environmental Stewardship lab in Corvallis, Oregon (passive sampler, PAHs only). These are the same laboratories that have participated in the LTEMP effort for the last eight years. Briefly, the results continue to be of acceptable precision and accuracy and can be compared to previous years' data. Physical characteristics of sediments were also reported in laboratory results though not presented herein.

Many compounds, especially in the mussel tissues, were below or near the analytical methods detection limit or were not detected in the sample. Sediment and mussel tissue concentrations are plotted and discussed as a sum of multiple PAHs (sum PAH) either by dry weight or wet weight where appropriate. Passive sampling device concentrations have been converted by the analytical lab into the dissolved-phase water concentration, C-free concentration. By converting the concentration units, comparisons can be made across other studies, areas, and ecotoxicological effect thresholds. Concentrations below the method level of detection threshold were provided by the lab as an estimate. These estimated concentrations were plotted on PAH profile figures and included in sum calculations; compounds that were not detected in a sample or were biased by laboratory issues were not included in the sum calculations. Forensic interpretation was done using analyte profile pattern comparisons for likely petroleum sources (i.e., ANS crude, a sample of the April 2020 oil spill at the terminal, and a spring 2017 effluent sample from the BWTF) for PAH, geochemical petroleum biomarkers, and saturated hydrocarbons in sediment sample. Blue mussels and passive sampling devices tentative forensic assertions were made by qualitative ratios of parent to alkylated compounds and low and high molecular weight PAH compounds. Analytical results and calculations for all samples and all analytes, pattern profiles, forensic ratios, and laboratory blanks are presented in the Technical Summary (Owl Ridge 2023) to support the assertions made in this summary report.

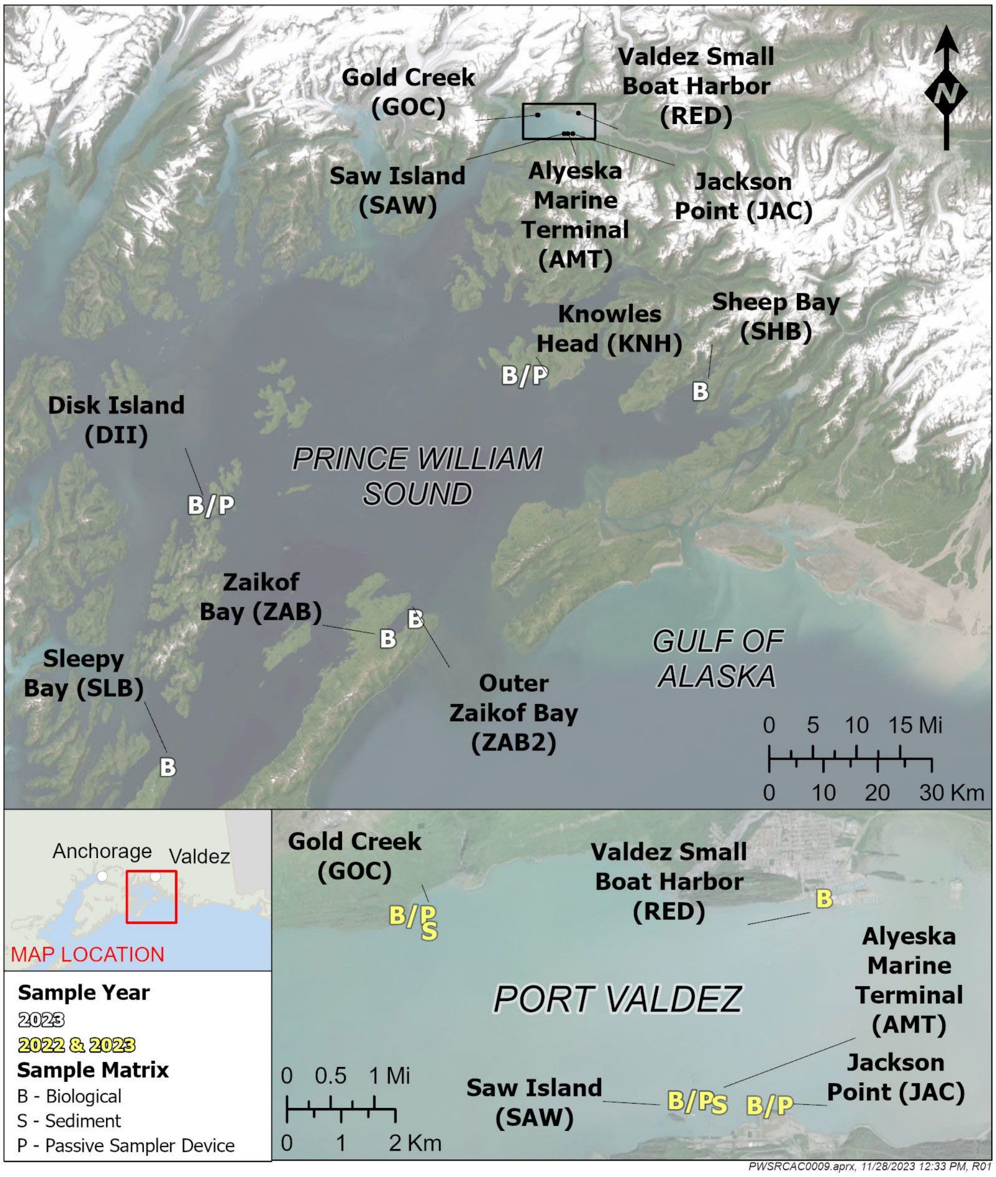

Figure 1. Map of 2022 and 2023 LTEMP sites for sediment (S), Pacific blue mussels (B), and passive sampling devices (P).

2.1. Sediments

Hydrocarbons were detected in all sediments sampled at the terminal and Gold Creek sites in the low parts per billion range (ppb or ng/g). One (1) ng/g or 1 ppb can be visualized as the concentration of 50 drops in an Olympic-sized swimming pool. In 2022, the highest sum (∑) PAH concentrations were found in the Gold Creek sediment (17.6±22.0 ng/g dry weight), while in 2023 the highest concentrations were found in the terminal sediments (41.1±5.7 ng/g dry weight) (Figure 2 and Figure 3). Naphthalenes and alkylated fluorenes, phenanthrenes, fluoranthenes/pyrenes, and chrysenes made up the bulk of PAHs at Gold Creek in 2022 and 2023 (see Figure 4 for 2023 results). At the terminal, similar compounds made up the bulk of detectable PAHs for both years but with great contribution from naphthobenzothiophenes. For comparison, PAH concentrations across both Port Valdez sites are lower than those reported in Norwegian fjords, Novia Scotia small boat harbors, and the Baltic Sea (Oen et al. 2006; Davis et al. 2018; Pikkarainen 2010). Present Port Valdez concentrations were more similar to those reported from sediments of Cook Inlet and St. Paul Island, Alaska (Nesvacil et al. 2016).

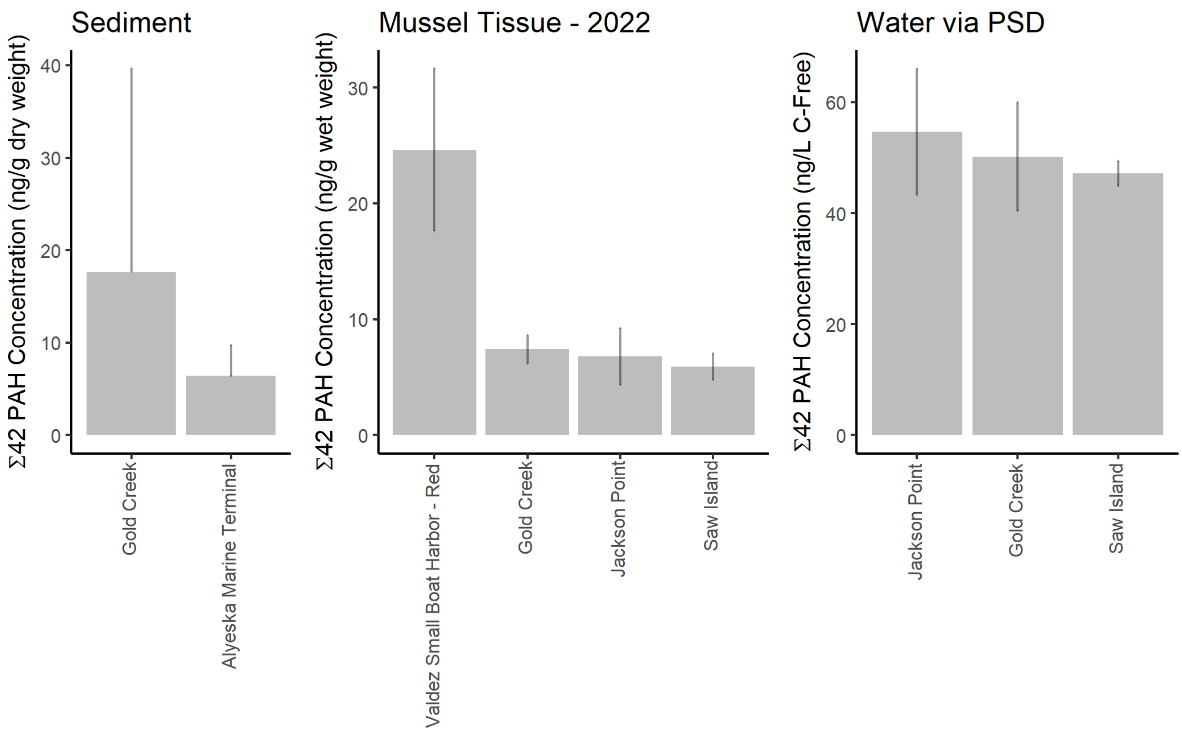

Figure 2. ∑ PAH concentrations for 2022 sediments, Pacific blue mussel tissues, and water sampled via passive sampling devices by site plotted at the mean ± 1 standard deviation. Due to large deviation between replicate samples, standard deviation was plotted only in the positive direction for sediment samples. Note difference in units between matrices (i.e., parts per billion for sediments and mussel tissues and parts per trillion for passive sampling devices).

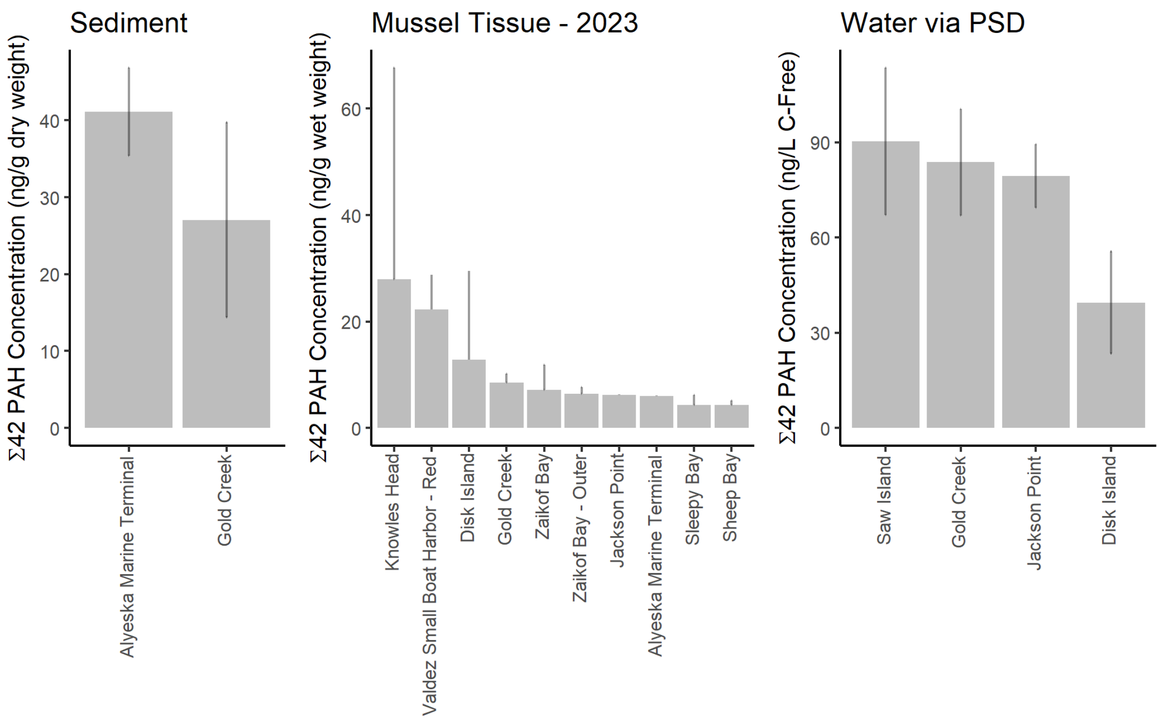

Figure 3. ∑ PAH concentrations for 2023 sediments, Pacific blue mussel tissues, and water sampled via passive sampling devices by site plotted at the mean ± 1 standard deviation. Due to a large deviation between replicate samples, standard deviation was plotted only in the positive direction for mussel samples. Note difference in units between matrices (i.e., parts per billion for sediments and mussel tissues and parts per trillion for passive sampling devices).

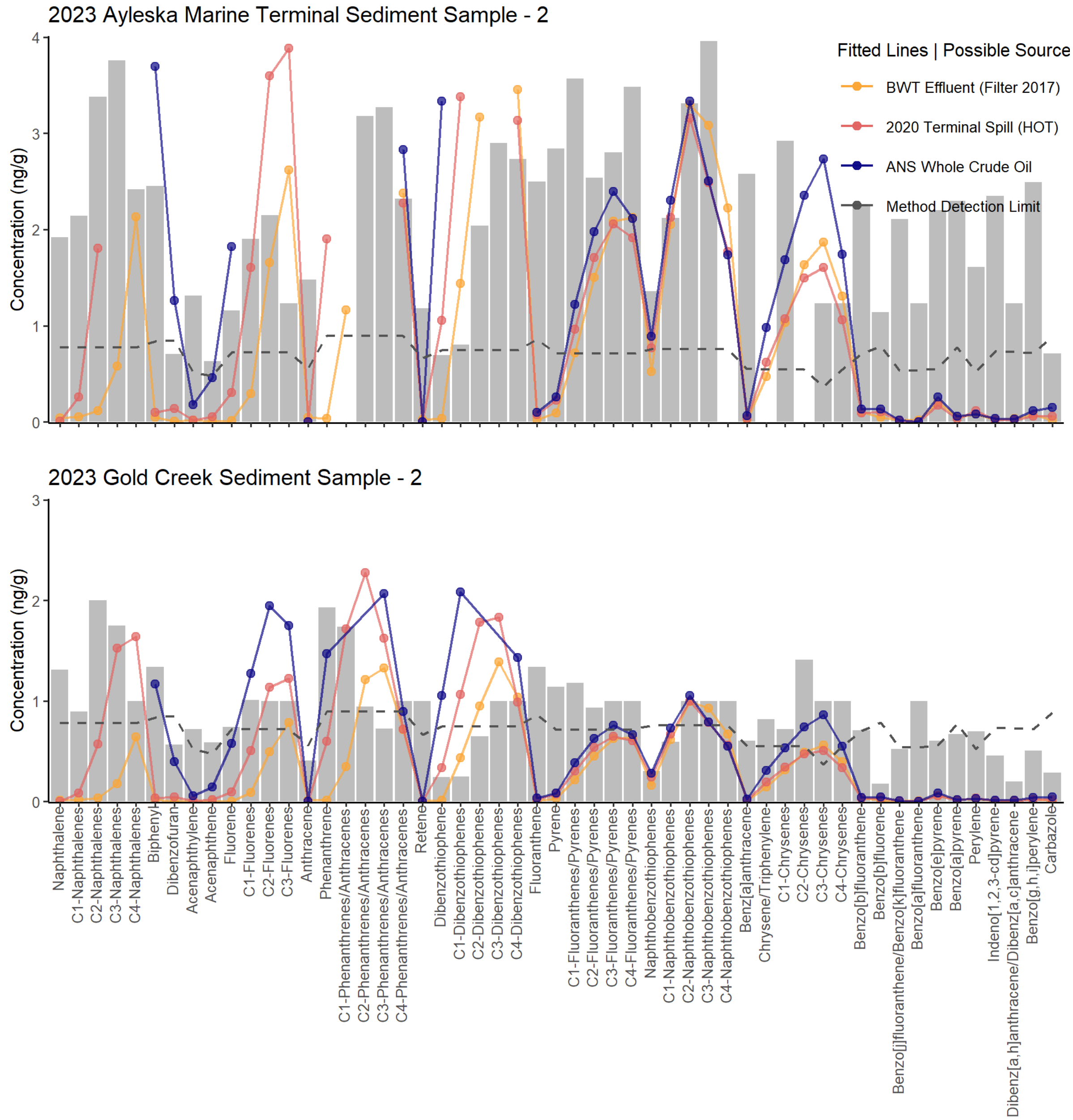

Figure 4. 2023 PAH profiles from sediments sampled at the terminal and Gold Creek site. Each plot displays a representative sample from the three replicates analyzed (note difference in y-axis scale). Possible Alaska North Slope Crude related source profiles are super imposed as different colored lines. A dashed, dark line indicated the analyte specific method detection limit.

2.1.1. Ecotoxicological Interpretation

In 2022 and 2023, individual and ∑ PAH concentrations in sediment at the terminal and Gold Creek sites pose little to no acute or chronic risk for marine organisms with concentrations of individual compounds and sums 1% or less than the U.S. Environmental Protection Agency (EPA) sediment quality PAH benchmarks for aquatic life (EPA 2016). While benthic communities adapted to the cold and sediment-rich waters of Port Valdez may not be adequately represented in these EPA benchmarks, past monitoring efforts around the terminal have indicated little to no change in the benthic community with varying PAH concentrations (Shaw and Blanchard 2021). The total organic carbon concentration in the sediment is low (0.4–0.6%), which indicates higher bioavailability of PAHs to marine organisms. High molecular weight PAHs are detected in sediments but concentrations of this group do not exceed any protective benchmarks nor are these compounds generally present in oil. Known carcinogenic PAHs are present in low concentrations at both sites.

2.1.2. Site-Specific Source Identification

Using PAH and biomarker profiles, the source of the hydrocarbons in the 2022 and 2023 terminal sediments is determined to be mostly petrogenic and derived from ANS crude oil. Biomarker patterns closely match those of previous oil spills at the terminal in 2017 and April 2020 (Payne and Driskell 2021) and particulate-phase oil in the effluent from the BWTF (Payne and Driskell 2018). The diagnostic biomarkers confirm ANS crude oil as the source. Two other patterns are also seen including a water-washing weathering of fluorenes and pyrogenic indicative phenanthrene/anthracene ratios. Accumulation of higher molecular weight alkylated PAHs, likely from local combustion sources, indicates residuals of prior PAH inputs inefficiently degraded over time, especially in 2023 samples. Saturated hydrocarbons in the terminal sediment reveal strong microbial degradation and weathering of the hydrocarbons leaving the higher molecular weight compounds (and in some cases, terrestrial plant wax compounds).

At Gold Creek, chemical biomarkers were sparse compared to those at the terminal, still petrogenic biomarker traces confirm the oil signal as a distant source. However, the PAH patterns are mixed petrogenic and pyrogenic. Gold Creek sediments are moderately weathered with a near complete loss of saturated hydrocarbons, except those contributed by terrestrial plants. In summary, relatively low hydrocarbon concentrations in the terminal sediments are linked to the terminal activities and incidents (BWTF effluent, spills, and combustion) with residues that have undergone environmental degradation and accumulated over time. Gold Creek sediments show mixed pyrogenic and lower petrogenic sources with a greater degree of weathering.

2.1.3. Historical Perspective

Hydrocarbon concentrations have varied widely throughout the LTEMP monitoring period from 1993 to the present (Figure 5). The highest sediment PAH concentrations were measured in the early 2000s at nearly 36 times the present concentrations. Since 2005, hydrocarbon concentrations have remained low with an all-time low seen in the mid-2010s. Since the low, a gradual increase in PAHs has been measured in sediments at the terminal and Gold Creek (Figure 5B). Terminal sediments have generally contained higher, more variable PAH loads than Gold Creek although considerable overlap in PAH concentration ranges between the two stations has persisted since 2008.

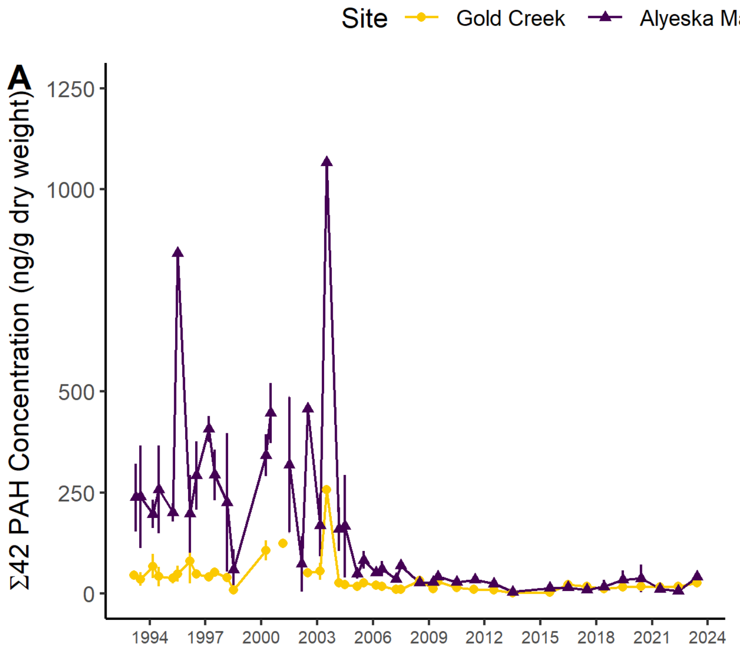

Figure 5. Sum 42 PAH concentrations in sediments (A) over the entire duration of the LTEMP and (B) since 2005 when concentrations have remained relatively low. Note the difference in scale. Colors and shapes indicate sampling site; mean values ± 1 standard deviation are plotted for each sampling event.

2.2. Pacific Blue Mussels

PAHs were detected in Pacific blue mussels (*Mytilus trossulus*) at low to moderate concentrations at all sites (Figure 2 and Figure 3). In 2022, the highest PAH concentrations were found at the Valdez Small Boat Harbor entrance, a non-ANS positive control site at the red harbor navigation light (range 18.4–32.2 ng/g wet weight; Figure 6). PAH concentrations in 2022 were similar at Gold Creek, Saw Island, and Jackson Point (range 4.7–9.5 ng/g wet weight). In 2023, mussels were collected from ten sites around the terminal, Port Valdez, and greater Prince William Sound (Figure 7). Samples were intended from the North Gulf coast of Alaska but these were not collected in 2023 due to inclement weather. The highest (and lowest) PAH levels were seen in mussels from Knowles Head in northeastern Prince William Sound (range 2.8–73.8 ng/g wet weight) although variability between replicates was high. Other relatively high PAH levels were found in mussels from the Valdez Small Boat Harbor and Disk Island.

Phenanthrene was the most abundant PAH at sites in 2022 except for the harbor where larger PAHs were more prevalent (Figure 6). In 2023, higher molecular weight PAHs were found in some replicates from Disk Island, Knowles Head, and the Valdez Small Boat Harbor, while Naphthalene and Phenanthrene were most prevalent at other Port Valdez sites, Sleepy Bay, and other Zaikof Bay sites.

The 2022 and 2023 mussel tissue PAH concentrations in Port Valdez are comparable to those found in relatively pristine locations in national parks and forests around southcentral and southeast Alaska and well below the high concentrations (>1000 ng/g dry weight (138 ng/g wet weight when using mean conversion factor from LTEMP mussel data)) found in the harbor at Skagway, Alaska

(Rider 2020). Only mussels from the Valdez Small Boat Harbor exceeded National Oceanic and Atmospheric Administration's (NOAA) national long-term monitoring status "Low Concentration" range (0–173 ng/g dry weight (0–24 ng/g wet weight)). Like the Valdez Small Boat Harbor location, fluoranthene was also the most abundant PAH in mussels in a Norwegian fjord with moderate human activity where sum PAH concentrations were otherwise comparable to this study (Schøyen et al. 2017). Mussel tissue PAH concentrations were comparable to those measured in pelagic zooplankton in Valdez Arm (Carls et al. 2006) and to mussels caged two kilometers or greater from an oil rig in the North Sea (Sundt et al. 2011).

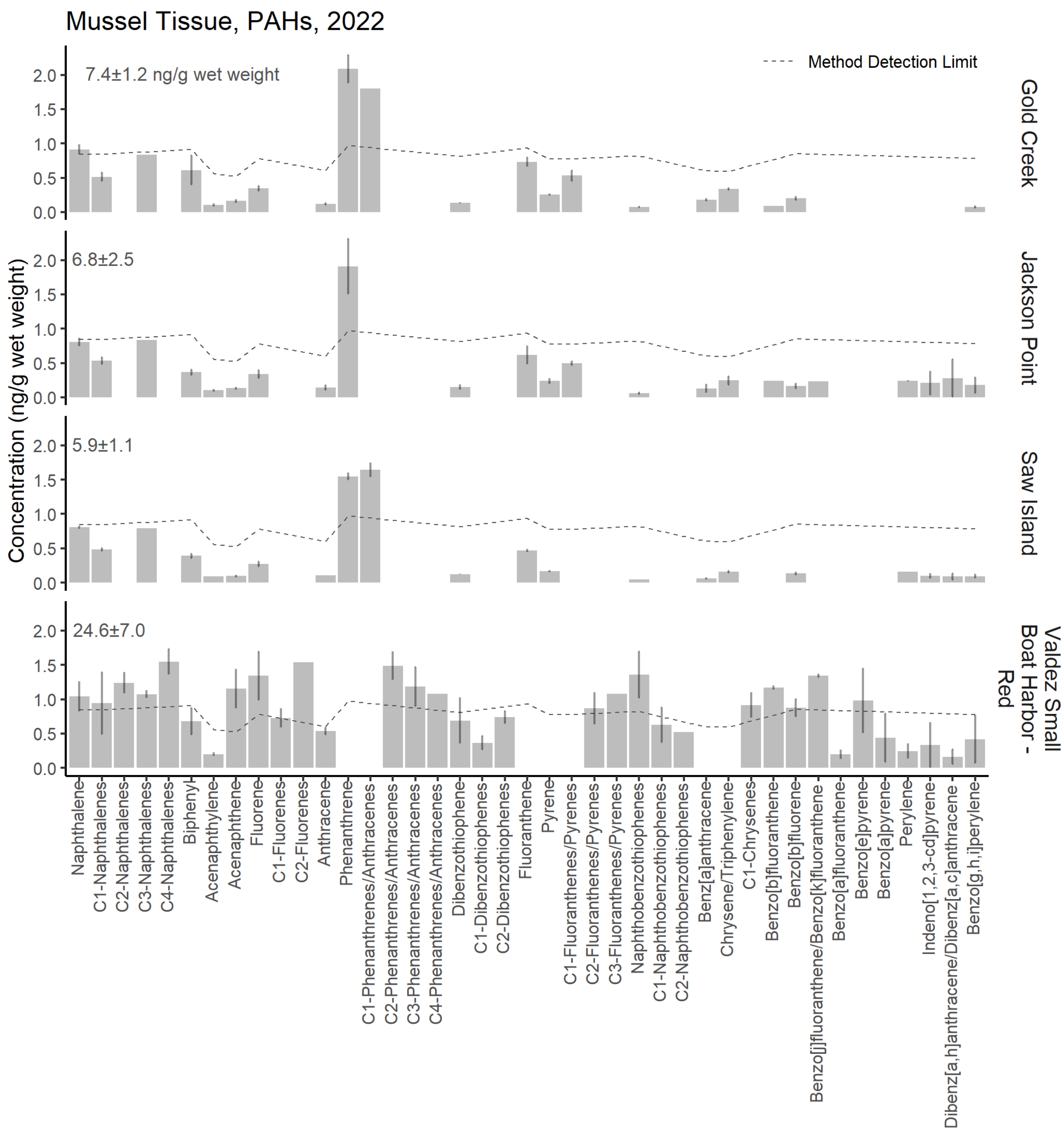

Figure 6. PAH profiles from Pacific blue mussels sampled at four sites in Port Valdez in 2022. Values represent mean ± 1 standard deviation and sum 42 PAH values are displayed in the upper left of each profile. The dashed line represents the PAH specific method detection limit.

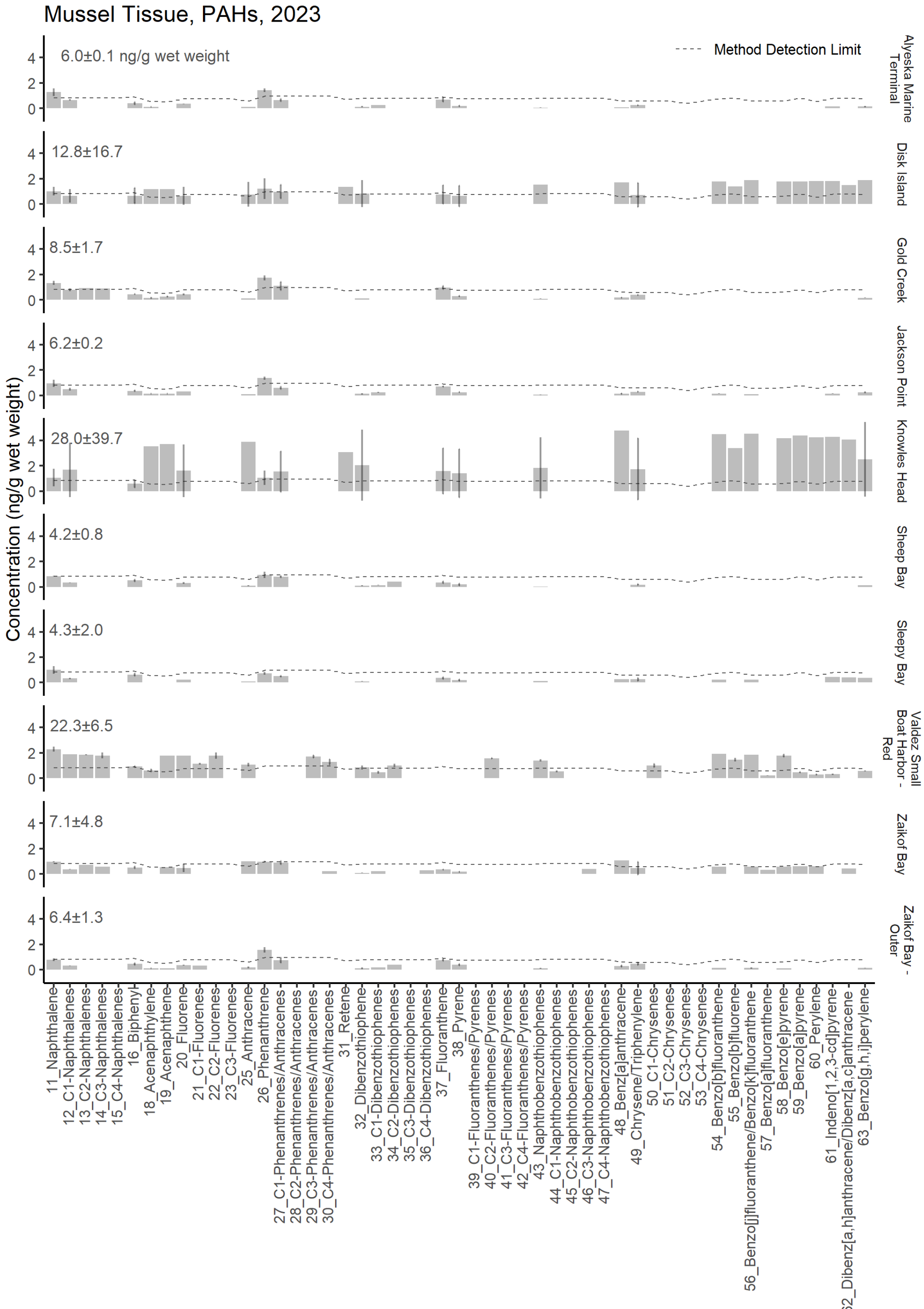

Figure 7. 2023 PAH profiles from Pacific blue mussels sampled at ten sites in Port Valdez and Prince William Sound. Values represent mean ± 1 standard deviation and sum 42 PAH values are displayed in the upper left of each profile. The dashed line represents the PAH specific method detection limit.

2.2.1. Ecotoxicological Interpretations

At the 2022 and 2023 tissue concentrations, no adverse biological effects are predicted. Considering the behavior of larger PAHs to adhere to lipids, mussel tissue concentrations are likely higher in the winter and early spring, before Pacific blue mussel spawning events (i.e., lipid-rich eggs will carry away significant amounts of PAHs). In this case, the post-spawning June sampling may represent a PAH accumulation low over the annual cycle.

Similar mussel tissue concentrations did not elicit early warning signs for genotoxicity or cellular toxicity in laboratory and field studies (Hylland et al. 2008; Sundt et al. 2011). At tissue PAH concentrations two orders of magnitude greater, laboratory studies observed reduced body size and greater cellular stress but no significant differences in gamete development in fuel-oil-exposed mussels (Ruiz et al. 2014).

Mussels accumulate more than just hydrocarbons. Across Prince William Sound and the North Gulf Coast, elevated concentrations of many metals and legacy pollutants are found locally in Pacific blue mussels (Rider 2020). While some of these concentrations are directly related to local past and present anthropogenic sources (e.g., mining, chemical storage, shipping, accidents and spills, and human activities), long-range transport of chemicals is likely a contributing factor. The potential for adverse effects on aquatic organisms from the combined stressors either through contaminant mixtures and/or environmental stressors should be highlighted but any further assertion as to the degree of injury would be speculative.

2.2.2. Site-Specific Source Identification

As tissue hydrocarbon concentrations and chemical compositions are driven by the bioavailability of compounds, environmental conditions, and physiological, cellular, and molecular processes in the mussels, which govern exposure, uptake, metabolism, and elimination, source identification analysis should be performed with caution.

In 2022, Gold Creek, Jackson Point, and Saw Island mussels exhibited similar PAH profiles with very few petroleum biomarkers detected. Saturated hydrocarbon in these samples reveal a higher relative presence of lighter saturated hydrocarbons compared to 2021 and 2023 which indicate a larger contribution of marine biogenic origin hydrocarbons (e.g., n-C15, n-C17, and pristane). The PAH profile at the harbor shows a greater contribution of pyrogenic sources with a lesser pyrogenic signature at sites around the terminal (i.e., Saw Island and Jackson Point). Gold Creek had so few PAHs detected but can tentatively be assessed as more petrogenic in origin whereas the other sites are more mixed source in origin. The ratio of n-C17/Pristane was greater than one at the Valdez Small Boat Harbor indicating a less biodegraded hydrocarbon source. At the other Port Valdez sites this ratio was less than one and thus reveals greater biodegradation.

In 2023, many sites exhibited detectable presence of higher molecular weight PAHs, indicative of bioavailable pyrogenic PAH and/or selective accumulation and retention of these compounds. Very few petroleum biomarkers were seen in the Knowles Head, Sheep Bay, and Sleepy Bay samples, thus exposure of these mussels to petroleum compounds is likely very low. At Disk Island and Knowles Head, high molecular weight PAHs were observed at relatively high concentrations in a single replicate. In both instances these high levels were not supported by the presence of petroleum biomarkers indicating a specific source. Similar patterns and sources attributed were seen in Port Valdez sites in 2023 as in 2022.

2.2.3. Historical Perspective

Historical trends in Pacific blue mussel tissue PAH concentrations are variable, reflecting known oil spill incidents in 2004 at Gold Creek, and 2017 and April 2020 spills at the terminal, and mirroring high concentrations found in sediments pre-2005 (Figure 8). Within the larger trend, PAH variability and mean tissue concentrations have stabilized since ~2010 in the absence of known spills (Figure 8B). In non-spill conditions, mussel tissue concentrations have remained below < 1,000 ng/g wet weight, indicating the mussels are likely not under PAH exposure-induced stress. However, high values have been recorded following spill incidents (e.g., 244,000 ng/g wet weight after the April 2020 terminal spill, not shown), a value likely to induce adverse effects at the molecular to the individual level for organisms (Figure 8A). Expanded sampling stations (e.g., Disk Island, Knowles Head, Sheep Bay, Sleepy Bay, and Zaikof Bay) show less variability in recent years, likely due to them being less exposed to recent spill events and the bias of less frequent sampling. Overall, 2022 and 2023 represent years with one of the lowest PAH concentrations found in mussels in LTEMP's 30-year history. However, this should be interpreted with caution as analytical methods are at the lower limits of detection and as such many compounds are considered an estimation in sum calculations.

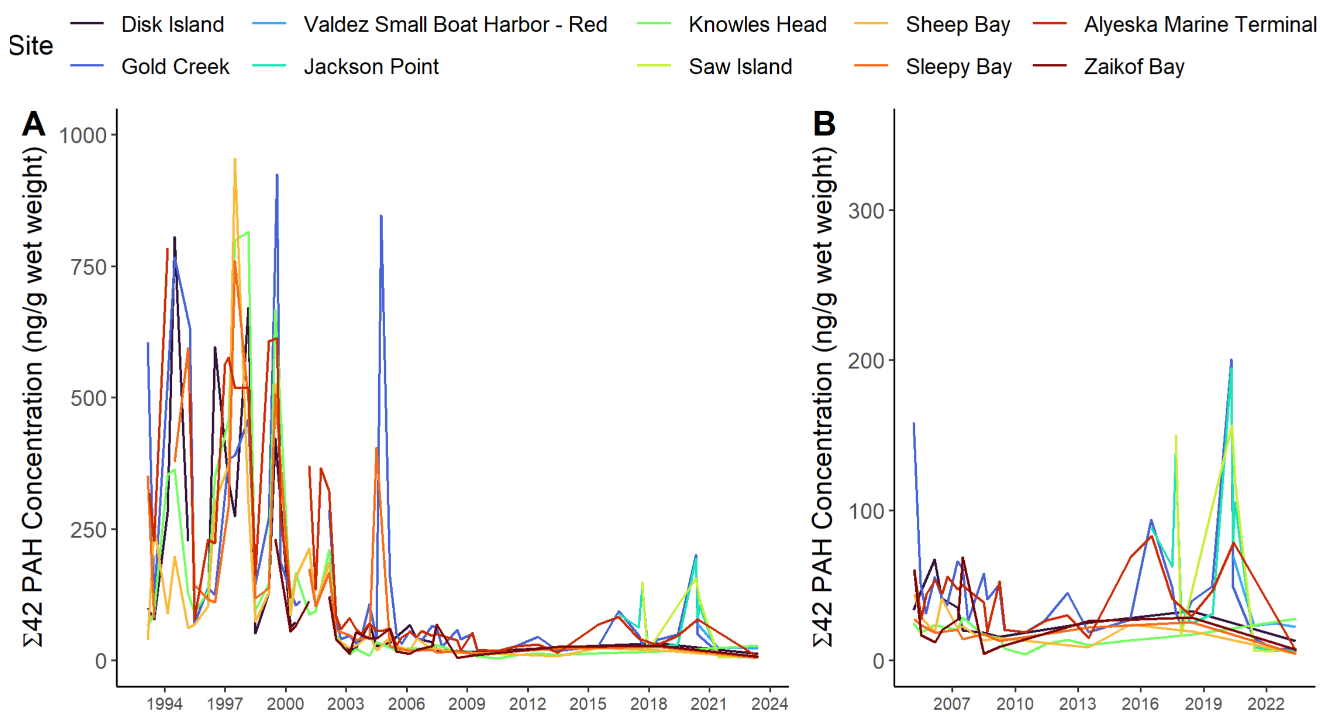

Figure 8. Total PAH concentrations in Pacific blue mussel tissue (A) over the entire duration of the LTEMP; note concentrations > 1000 ng/g wet weight (i.e., known spill events) were removed for clarity even though max post spill concentration >200 000 ng/g wet weight, and (B) over the last 18 years and excluding concentrations >350 ng/g wet weight for clarity. Colors indicate sampling site and mean values are plotted for each sampling event.

The range of the 2022 and 2023 PAH concentrations in Port Valdez mussel tissues is within the historical range of locations with limited human use and not oiled during the Exxon Valdez oil spill (Boehm et al. 2004).

2.3. Water sampled via Passive Sampling Devices

Hydrocarbons were found at low concentrations in water sampled via passive sampling devices in 2022, at sites in Port Valdez (47–54 ng/L sum 42 PAHs) (Figure 2) and in Port Valdez and greater Prince William Sound in 2023 (38–83 ng/L sum 42 PAHs) (Figure 3). These concentrations represent the dissolved constituents (C-free) and are not traditional total water concentrations, but in this report the passive sampling device C-free concentrations are used as a proxy for water concentrations of PAHs. In 2022, the highest relative passive sampling device-derived water concentrations were measured at Jackson Point (54±11 ng/L) closely followed by Gold Creek (49±9 ng/L) and Saw Island (47±2 ng/L). In 2023, Port Valdez trends were reversed with Saw Island reporting the highest relative PAH concentration (84±19 ng/L) followed by Gold Creek (81±17 ng/L), Jackson Point (78±10 ng/L) and the extended sampling site in central western Prince William Sound, Disk Island, (38±16 ng/L). A passive sampling device was deployed at Knowles Head in 2023, but could not be located for retrieval.

In both years, dissolved and heavily water-washed naphthalenes made up the majority of the PAH bulk across all samples and sites (see Figure 9 for 2023 PAH profile). Smaller, 2–3 ring PAHs made up 99% of the sum concentrations, indicative of the more readily water-soluble fraction. Other PAHs that were detected at lower concentrations at all sites were fluorenes, fluoranthenes, dibenzothiophenes, phenanthrenes, and anthracenes. Concentrations of alkylated compounds were greater than those of parent compounds at Disk Island indicating a water-washed oil source, evaporative transfer of dissolved compounds into the atmosphere, or weathering of a surface oil film before it was entrained into near-surface water and dissolved to an appreciable extent. At Port Valdez sites a petrogenic pattern was seen in parent and alkylated fluorenes. While direct comparison of the passive sampling data to other environmental hydrocarbon studies is challenging due to methodological differences, present dissolved PAH concentrations from the passive sampling devices are comparable to water concentrations at unoiled sites and sites with medium human activity around Prince William Sound (Short et al. 2008; Lindeberg et al. 2017). The present passive sampling device-derived water concentrations in Port Valdez and at Disk Island were all at least two to three orders of magnitude below published water quality standards and below those of polluted areas across the United States (EPA 2002).

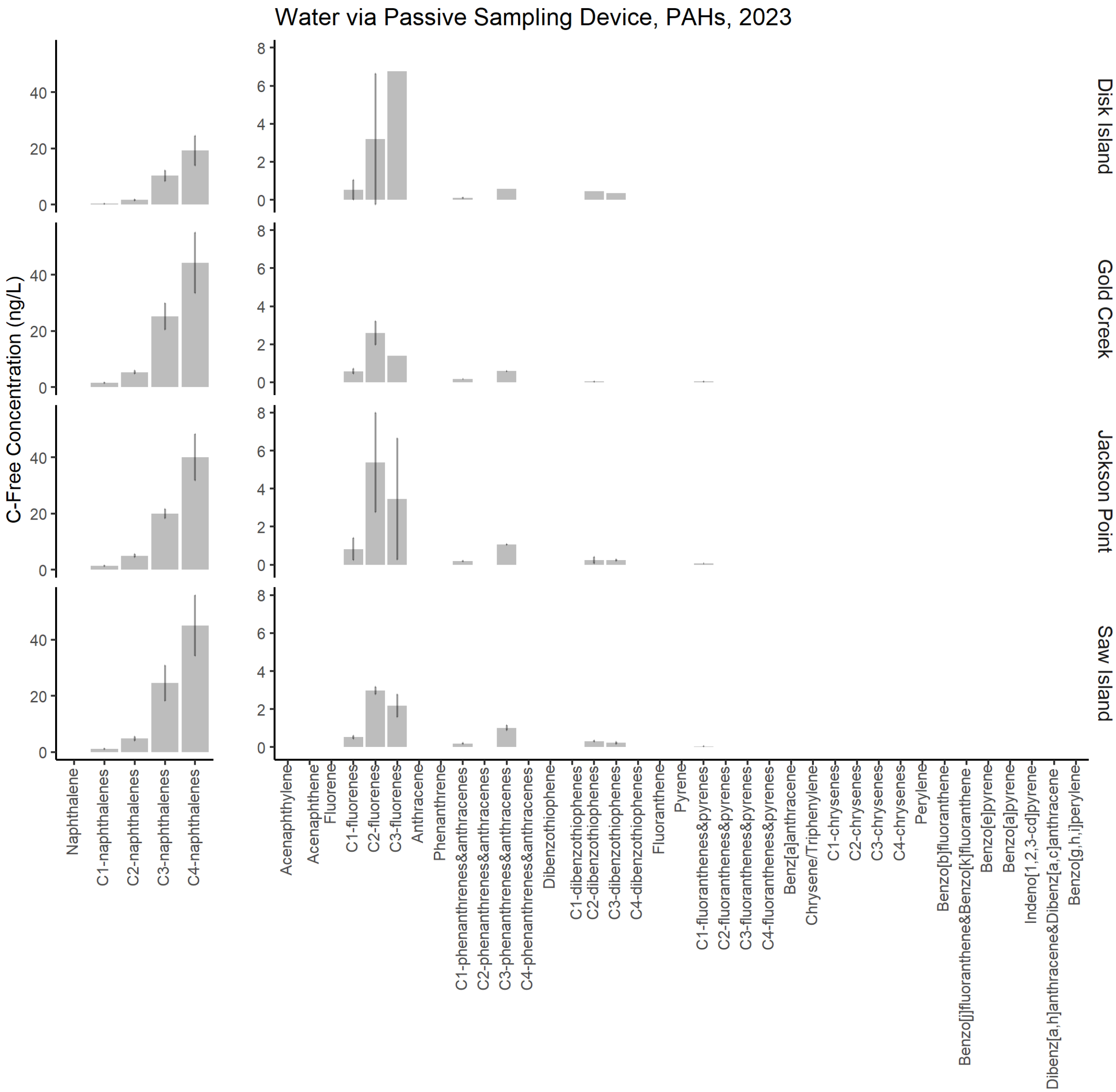

Figure 9. PAH Profiles in 2023 water sampled via passive sampling devices placed at Gold Creek, Jackson Point, and Saw Island. Values represent mean ± standard deviation for the three replicates.

2.3.1. Ecotoxicological Interpretations

Concentrations reported in the Port Valdez passive sampling device-derived water concentrations are below those reported to cause adverse effects even in the most sensitive of life stages for marine organisms. The 2022 and 2023 PAH concentrations in the parts per trillion range (i.e., one drop in 20 Olympic-sized swimming pools) are an order of magnitude lower than those reported to cause developmental and delayed effects in herring and salmon early life stages (Incardona et al. 2015), although no analytical lower limit measured from water or tissues has been identified for developmental cardiac effects in herring (Incardona et al. 2023). Studies on Arctic cod embryos, a Bering Sea species not present in Prince William Sound, report malformations and reduced survival at concentrations similar to those measured by the passive samplers; however, the analytic methods and exposure PAH composition differs with the Arctic cod study using whole crude oil (Bender et al. 2021). Naphthalene, while present at greater concentrations than other PAHs, is of low toxicological concern at present concentrations and is not a carcinogen.

2.3.2. Site-Specific Source Identification

Though not the focus of the passive sampling device, which measures the dissolved and bioavailable fraction (C-free concentrations) in the water, PAH profiles can be used conservatively for source identification and forensic analysis. One striking observation is the large naphthalene peak with ascending alkylation, indicative of a water-washed and weathered petrogenic source present in all samples. Similar patterns are seen in the fluorenes in all 2022 samples; however, the pattern is more petrogenic in 2023 at Gold Creek and Saw Island.

2.3.3. Historical Perspective

PAH concentrations in passive samplers have remained low since the 2016 inclusion of passive sampling device-derived water concentrations into LTEMP (Figure 10). A peak in PAH levels is seen at the terminal adjacent site, Jackson Point, following the 2020 terminal spill. Passive sampler PAH profiles over time have also remained consistent with high naphthalene spikes dominating PAH profiles as noted in previous LTEMP reports (Payne and Driskell 2021).

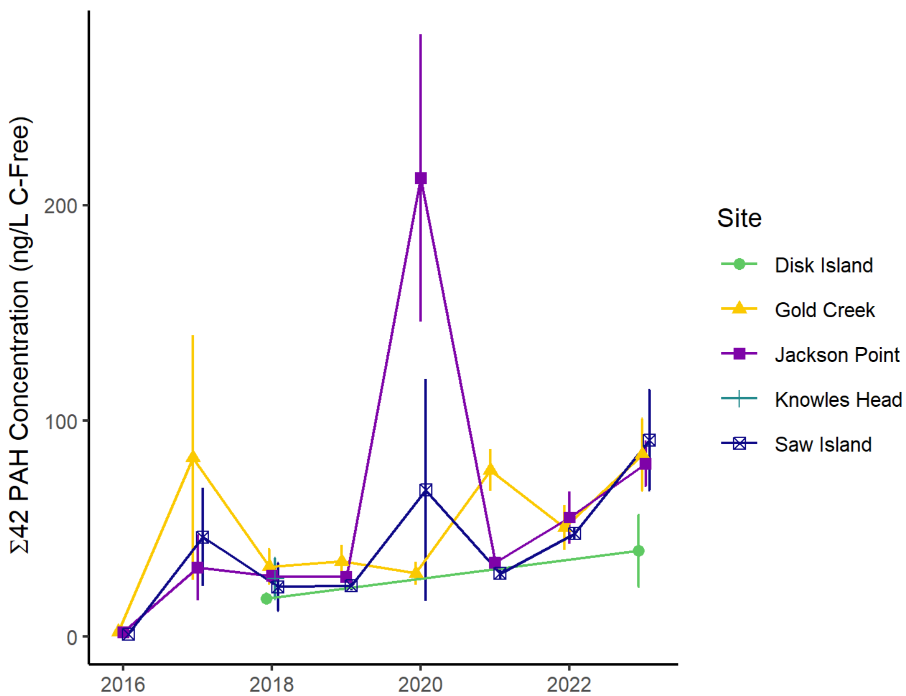

Figure 10. Sum 42 PAH concentrations in passive sampling device-derived water concentrations at five sites for 2016–2023. Sites are distinguished by color and shape and plotted by mean ± 1 standard deviation. Note that 2016 values only include parent PAHs, no alkylated PAHs were quantified in 2016.

2.4. Holistic Interpretation

In 2022, we saw agreement on low-level PAHs at similar concentrations across the three standard LTEMP stations in Port Valdez (i.e., Gold Creek, Saw Island, and Jackson Point). Mussel PAH levels found at the Valdez Small Boat Harbor were higher than other stations but could not be confirmed by sediment or passive sampler results as these samples were not taken. In 2023, the standard LTEMP stations in Port Valdez reported similar PAH concentrations and similarities to one another as in 2022. Surprisingly, the expanded LTEMP stations of Knowles Head and Disk Island had average PAH concentrations more similar to the Valdez Small Boat Harbor. Other expanded LTEMP mussel sites of the Zaikof Bay (both inner and outer), Sleepy Bay, and Sheep Bay had low PAH concentrations similar to those around the terminal. The passive sampling device deployed and retrieved at Disk Island did not corroborate the relative increased abundance of PAHs found in mussels at Disk Island but rather reported a concentration of water-soluble PAHs below that of the Port Valdez sites. Both mussels and passive sampling devices from Disk Island had considerable variability between replicates compared to other sites so the ranking of hydrocarbon contaminant between sites should be done with caution. Even greater variability between replicates was seen in the Knowles Head mussel samples which may indicate a difference in the sampling for these expanded efforts may have impacted sample agreement (e.g., holding time, sample quality, cross contamination procedures). However, both locations from the remote site of Zaikof Bay had relatively good agreement so other factors may contribute to the variability (e.g., site specific heterogeneity in mussel community or habitat).

Looking across time both sediments and mussel PAH concentrations have varied over time (Figure 11) with both matrices experiencing peaks and troughs in PAH concentrations. Relatively low R-squared values, which reflect the amount of variation in the data explained by the 3rd order polynomial log transformed model, are expected for this type of environmental chemistry data, however these values indicate that other factors besides time likely influence PAH concentrations (e.g., environmental changes in Port Valdez such as increased glacial melt/freshwater runoff (Campbell 2018), recent spills). Although sampling locations for sediments and mussels are not identical in all years, more recent PAH peaks are seen in mussels compared to sediments. This is likely due to the shorter response time mussels have to spill events, something highlighted in LTEMP adjacent studies (e.g., Bowen et al. 2021) which investigated the transcriptomic response of mussels exposed to the April 2020 spill at the terminal.

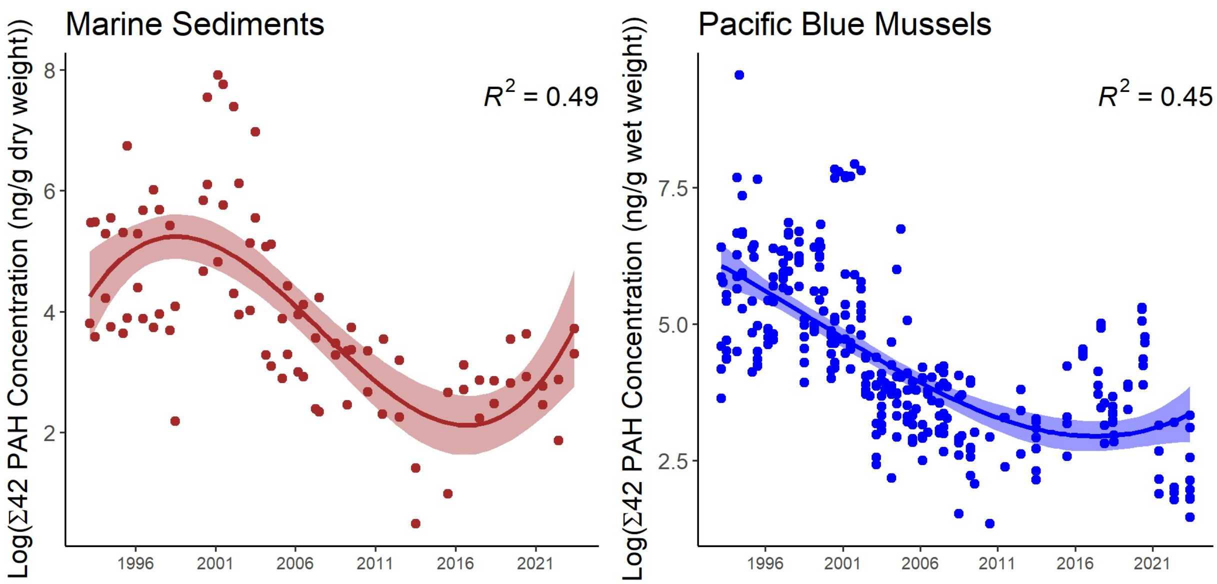

Figure 11. Polynomial trend line (3rd order) with standard error trend line fit to log transformed marine sediment (left) and Pacific Blue mussel tissue (right) PAH levels since 1993 for the sites sampled in 2023.

The forensic agreement between 2022 and 2023 samples is consistent with the mixed source petrogenic signal closer to the terminal and pyrogenic signal of stations further away. Again, string pyrogenic and mixed sources contribute to blue mussel hydrocarbons profiles at the Valdez Small Boat Harbor. As blue mussel tissues do not provide robust forensic data (e.g., few biomarkers of detection) interpretation of the expanded LTEMP sampling locations is limited. In a recent published study by Short and Maselko (2023) analyzing intertidal sediment oil samples from 2006 in western Prince William Sound, including Disk Island, crude oil from Exxon Valdez oil spill (EVOS) was determined to be the primary PAH contributor even when considering historical and ongoing human activities (e.g., mining, logging, fish processing, and fish hatcheries), and natural disasters such as the hydrocarbon pollution resulting from the 1964 earth quake and subsequent tank ruptures, and past forest fires.

The ecotoxicological risk to organisms from the hydrocarbon levels present in the sediments, mussel tissue, and dissolved in the water from 2022 and 2023 was low. Previous work focusing on how low levels of hydrocarbon exposure can influence ecologically and commercially important fish species in Prince William Sound has found profound effects on heart development (Incardona et al. 2021). In fact, recent herring research reveals that analytical chemistry with detection levels in the sub parts per billion level (ng/g) is not sensitive enough to distinguish between exposure and background concentrations in water or embryo tissue even when crude oil-induced effects on heart development and PAH-induced enzymatic response were detected (Incardona et al. 2023). Rather enzymatic induction related to nominal crude oil exposure (e.g., CYP1A induction) is directly related to cardiac deformities in herring and may provide a more sensitive assessment of injury at the low end of PAH exposure levels (Incardona et al. 2023). Targeted laboratory experiments have yet to confirm the link between early life stage oil exposure and sensitivity to pathogens later in life, which is the latest ecotoxicological hypothesis for the post-EVOS herring collapse (Whitehead et al. nd).

Current herring dynamics research has shifted focus away from hydrocarbon-induced direct effects and on to how ocean climate, freshwater input, and changes in timing of spawning have influenced survival of herring (Dias et al. 2022). Recent survey results indicate that herring may be rebounding with strong age classes observed in 2021 (Pegau et al. 2023).

3. FUTURE PERSPECTIVE

Recent work done by Harsha and Podgorski (2023) on hydrocarbon oxidation products and heavy metals in the BWTF and effluent has highlighted the presence and potential environmental risk of compounds not captured by the current LTEMP monitoring scheme. This work also argues that the assumption that stormwater and runoff from the terminal is "uncontaminated," is a finding supported from LTEMP sampling of sediments and blue mussels in the absence of spill events. Specifically, assessing the risk of toxic effects from the bioaccumulation of heavy metals, zinc, and arsenic, from BWTF effluent not removed in the filtration and biodegradation process has not been carried out in LTEMP or Alyeska pollution discharge permitting (i.e., APDES) monitoring (Shaw and Blanchard, 2021). In fact, the recent 2019 ADEC report cites that the biggest water quality concerns from the terminal BWTF effluent is zinc, total aromatic hydrocarbons, and whole effluent toxicity (ADEC 2019).

Heavy metal monitoring is routinely done in other petroleum and hydrocarbon monitoring efforts including in forensic studies in marine sediments and offshore petroleum industry monitoring efforts although typically focusing on mercury, lead, cadmium, and barium (e.g., Norwegian Environmental Agency, 2020).

Frequent reanalysis of LTEMP's aims and methodology is necessary to maintain the utility of such a powerful monitoring program even in its 30th year. While maintaining the integrity of the program with the three matrix approaches, efforts must be taken to ensure that future monitoring and reporting is conducted in a manner that guarantees comparability to previous analysis. The following represents a list of potential additions, subtractions, and alterations in methodology that could be considered for future LTEMP cycles.

1. Alter forensic analysis from its current and recent historical qualitative profile analysis to a quantitative statistical analysis using multidimensional scaling to allow greater comparative power over time, space, and between studies.
2. Place a passive sampling device at the Valdez Small Boat Harbor to allow for direct comparability for mussels sampled from this site.
3. Work with existing laboratories to expand analytical power to include emerging contaminants of environmental concern (e.g., PFAS, per- and polyfluoroalkyl substances, or the magnitude of the unresolved complex mixture which may include oxygenated products).
4. Perform a comprehensive evaluation of LTEMP in light of international environmental marine monitoring standards for planning, implementation, analysis, and reporting while still tailoring LTEMP to the needs of PWSRCAC.

5. Execute a "rat hunt" to explore the utility of the current and past LTEMP analyte and sampling regime. For example, assessing if running full hydrocarbon forensic analysis on blue mussels is necessary as a high frequency of geochemical biomarkers analytes are not detectable and therefore not useful in forensic analysis.
6. Investigate the potential to include additional biological information to reduce potential variability between biological samples including assessing spawning status, size, and condition in Pacific blue mussels.
7. Expand biological sampling. (1) Include PAH analysis in liver and bile of wild caught resident fish species (e.g., sculpin); (2) expand BWTF effluent testing as whole effluent testing reveals concerning toxicity (suggestion by Harsha and Podgorski 2023); and (3) include hydrocarbon specific biomarkers of PAH exposure and injury with mussel sampling.
8. Consider all phases of LTEMP in the current era of rapid environmental change, demand for scientific transparency, and environmental justice.

At this point in time many options mentioned above have not been fully investigated as this would require additional analysis. This list is intended for discussion purposes amongst the PWSRCAC Scientific Advisory Committee. Modernizing LTEMP could involve inclusion of biosensors for real time monitoring as was suggested by Harsha and Podgorski (2023) in their work on the hydrocarbon oxidation products in the BWTF effluent (Gavrilas et al. 2022) or remote sensing environmental monitoring of oil pollution using satellites, an emerging technique for remote areas with rapid environmental change and human activity (Sizov et al. 2014).

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4. CONCLUSION

In the 30th year of the LTEMP run by PWSRCAC, two years of data were analyzed for the concentration, source, and potential ecotoxicological effects of hydrocarbons in marine subtidal sediments, Pacific blue mussels, and dissolved in the nearshore waters via passive sampling devices. The hydrocarbon fingerprints in the 2022 and 2023 samples vary by site with those at or near the Valdez Marine Terminal revealing ANS crude and its associated products (i.e., BWTF effluent) as the primary source for hydrocarbons. Hydrocarbons found in Pacific blue mussels from Gold Creek, Knowles Head, Disk Island, Sheep Bay, Sleepy Bay, Zaikof Bay, and the Valdez Small Boat Harbor cannot be linked directly to the terminal operations although these samples revealed a mix of sources. Low potential environmental and toxicological risk is posed by hydrocarbons contributed by the terminal and tankers in 2022 and 2023. Surprisingly, concentrations of toxic hydrocarbons were similar at the remote sites of Knowles Head and Disk Island and the Valdez Small Boat Harbor, a site of high human activity and potential chronic petroleum pollution. Passive sampling devices continue to report low levels of bioavailable hydrocarbons in the water column with higher concentration within Port Valdez compared to the remote, historically EVOS oiled site of Disk Island. Since 1993, hydrocarbon concentrations in Prince William Sound are generally low with localized spikes corresponding with spill events like the April 2020 oil spill at the terminal. Following an all-time low in the mid-2010s, hydrocarbon concentrations in sediments and mussels have slowly increased across all sites but are still below any threshold for adverse effects on aquatic life. Several suggestions have been made to expand, economize, and modernize LTEMP.

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