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Assessing lead sources in fishes of the northeast Pacific Ocean

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ABSTRACT

Industrial lead (Pb) emissions have changed oceanic Pb concentrations and isotopic compositions significantly over the last century. Asian industrial emissions are currently the dominant Pb sources in the northwest and central Pacific Ocean. This study investigated major Pb sources in the northeast Pacific Ocean (inland, coastal, and open ocean), where no comprehensive data exist currently. We measured Pb concentrations and isotopic compositions of a variety of shellfish and fish collected in British Columbia (BC) and used them for Pb source apportionment applications in the marine environment. We identified a clear trend in Pb isotopic compositions, from higher $^{206}Pb/^{207}Pb$ in inland fish to lower $^{206}Pb/^{207}Pb$ in open ocean fish, with coastal fish possessing mid-range values. These results indicate that natural sources of Pb dominate freshwater ecosystems in the central BC coast, whereas anthropogenic Pb sources prevail in western North America and the northeast Pacific. Pacific herring that forage along BC coast exhibit Pb isotopic composition that deviates toward the Chinese Pb regression line. We estimate an Asian origin for about one third of their total Pb. This study shows that Pb isotopes in aquatic organisms are valuable tools for assessing Pb sources across coastal and offshore regions of the northeast Pacific, with great potential for identifying primary foraging areas of marine organisms.

1. Introduction

Changes in the types and magnitudes of global industrial lead (Pb) emissions have altered Pb concentrations and isotopic compositions in the environment over the last century. Emissions from leaded gasoline used to be the dominant source of the total global Pb in the environment but have drastically decreased due to its nearly complete phase-out over the past four decades (Eisenreich et al., 1986; Wu and Boyle, 1997; Boyle et al., 2014). In parallel, rapid industrialization in Asia, accompanied by intense coal combustion and metal smelting, has released a large amount of Pb into the atmosphere (Li et al., 2012; Flegal et al., 2013; Zurbrick et al., 2017). Diminishing impacts from leaded gasoline combined with growing Pb emissions of Asian industrial activities have been recorded in a variety of environmental samples, including seawater (Wu and Boyle, 1997; Boyle et al., 2014), ice and snow (Gross et al., 2012; Bory et al., 2014), corals (Inoue and Tanimizu, 2008), and sediments (Chen et al., 2016; Hosono et al., 2016).

A number of studies have observed marked Asian Pb contributions in various regions of the Northern Hemisphere (Osterberg et al., 2008; Díaz-Somoano et al., 2009; Ewing et al., 2010; Gallon et al., 2011; Gross et al., 2012; Li et al., 2012; Zurbrick et al., 2017; Asher et al., 2018). Bory et al. (2014) reported growing impacts of anthropogenic Pb from Chinese aerosols and dust deposited on Greenland snow since the 1990s. Intergovernmental Oceanographic Commission cruise data and global Pb emission estimates indicate that present-day anthropogenic Pb in surface and near-surface waters of the western and central Pacific Ocean is likely dominated by emissions from East Asia; mostly from China, Russia, and Japan (Gallon et al., 2011; Zurbrick et al., 2017). The Pb concentrations and isotopic compositions of ice cores and airborne particulate samples collected in western North America suggest that Chinese industrial aerosols have undergone long-range transport via mid-latitude westerlies and contributed 29–46 % of total atmospheric Pb deposition during the peak seasons of Asian impacts, i.e., spring and summer (Osterberg et al., 2008; Ewing et al., 2010; Gross et al., 2012; Asher et al., 2018).

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Lead isotopes are a powerful tool for tracking sources of contamination in the environment (Patterson and Settle, 1987; Komárek et al., 2008; Cheng and Hu, 2010). The relative abundance of Pb isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb) varies systematically with geological setting and the time since geologic formation (e.g., age of the bedrock, ore body) (Faure and Mensing, 2005; Weis, 2017). Different environmental Pb sources are frequently distinguished by their characteristic isotope ratios (Komárek et al., 2008; Cheng and Hu, 2010). Lead enters marine food webs through primary producers and undergoes negligible isotopic fractionation within the biosphere. Therefore, the environmental source(s) of Pb are the determinant of isotopic composition in marine species, rather than physical, chemical, and biological processes (Flegal et al., 1987; Michaels and Flegal, 1990; Smith et al., 1990). Consequently, Pb isotope ratios in shellfish and fish are expected to reflect the Pb isotopic composition of the ambient water, sediment, and diet (Prosi, 1989). Previous studies have shown that the Pb isotopic composition of biological samples can provide a fingerprint for sources of Pb (Spencer et al., 2000; Ip et al., 2005).

Scarce Pb data exist for seawater and marine biota from the northeast Pacific Ocean. Substantial inputs of Pb from Asian dusts and aerosols have been observed in western North America (Osterberg et al., 2008; Ewing et al., 2010; Gross et al., 2012; Asher et al., 2018). In this study, we address the following research questions: What are the dominant sources of present-day Pb in the northeast Pacific Ocean? To what extent do Pb emissions from Asian industries, principally China, account for the Pb found in this region? We investigate the relative importance of various Pb sources across inland, coastal, and the open ocean waters of the northeast Pacific. We use a variety of fish and bivalves from British Columbia (BC), Canada and report Pb isotope and concentration data for these species. Juvenile and returning adult Pacific salmon reflect the Pb isotopic composition of their freshwater and open ocean habitats, respectively, and coastal fish (herring) and shellfish (mussels, and clams) act as bioindicators for Pb exposure in coastal waters.

2. Materials and methods

2.1. Sample collection and processing

Fish and bivalve species were collected from diverse habitats across >800 km of coastline in British Columbia, Canada (Table 1, Fig. 1). All samples were stored frozen after collection. Four species of returning adult Pacific salmon (chinook, sockeye, chum, pink) were collected in 2014 and 2018 from Rivers Inlet on the central coast of BC. Juvenile sockeye salmon (fry) were captured with beach seines in 2015 in Owikeno Lake, a large glacial lake that is surrounded by coastal volcanic mountains and empties into Rivers Inlet via the Wannock River. Pacific herring were collected from five sites spanning Strait of Georgia and central and north BC coast. Spring pre-spawning and spawning herring aggregations were sampled in 2016 from purse seine surveys (Haida Bay; Denman Island) conducted by Fisheries and Oceans Canada and with cast nets by independent research surveys (other sites). Mussel samples were handpicked from Third Beach on Calvert Island on the central coast of BC in 2018. Clams were collected from four sites near Quadra Island in the northern Strait of Georgia in 2007. The Strait of Georgia is a semi-enclosed estuarine sea between the BC mainland and Vancouver Island and has different oceanographic conditions from other BC coastal environments that connect directly to open ocean (Ianson et al., 2016). Hereafter, we call other coastal regions including the west coast of Vancouver Island and the central and north BC coast as coastal ocean to distinguish them from Strait of Georgia (Fig. 1).

The Pb concentrations of fish muscle tissue have direct implications for human health and trophic transfer along the food chain but are generally much lower than other biological tissues in fish (e.g., gill, liver, intestine, hard tissues) (Jezierska and Witeska, 2006). Limited data have been reported for Pb levels in the muscle of marine fish, which is partly due to analytical challenges with low concentration levels. At the Department of Earth, Ocean and Atmospheric Sciences at the University of British Columbia (UBC), we subsampled fish muscle tissue (salmon and herring) with an acid-cleaned ceramic knife and freeze-dried the tissue prior to digestion and chemical analysis. Juvenile salmon samples were too small to fillet muscle tissue so were freeze-dried and digested as whole fish samples. Bivalve samples were shelled and prepared for analysis as individual samples, rather than pooled. After freeze-drying, all subsequent sample preparation and Pb concentration and isotopic analyses were performed in Class 1000 (or better) clean laboratories (with Class 100 laminar flow hoods) in the Pacific Centre for Isotopic and Geochemical Research at UBC.

2.2. Lead concentration and isotope analysis

Approximately 0.2 g of freeze-dried fish tissue was digested in 10 mL 14 M HNO_3 using a laboratory microwave system (CEM Corporation, Matthews, NC, USA). The digests were completely evaporated and then reconstituted in a mixed solution of 0.5 % HCl, 1.5 % HNO_3 and 10 ng/g indium (added as an internal standard). Following established sample preparation and analytical procedures (Smith et al., 2019), the Pb concentration and isotopic composition of the solution for all samples but clams were

Table 1
Sample collection locations and predominant feeding habitats of fish and shellfish species used in this study.

Common name (*Scientific name*)	Sample Size	Capture Locations	Feeding Habitats	Mean Pb (1 SD) (ng/g dry weight)	
Sockeye Salmon Fry[a] (*Oncorhynchus nerka*)	10	Owikeno lake	Freshwater Inland	230.8	(162.1)
Manila clams (*Venerupis philippinarum*)	8	Quadra Island	Strait of Georgia	33.7	(24.9)
Pacific Oyster[b] (*Crassostrea gigas*)	3	Desolation Sound	Strait of Georgia	89.3	(41.0)
Pacific Oyster[b] (*Crassostrea gigas*)	8	Barkley Sound	Coastal ocean	126.8	(48.9)
Blue Mussel (*Mytilus edulis*)	10	Calvert Island	Coastal ocean	208.6	(61.5)
Pacific Herring (*Clupea pallasii*)	3	Denman Island	Coastal ocean	10.6	(2.8)
Pacific Herring (*Clupea pallasii*)	4	Haida Bay	Coastal ocean	38.8	(10.8)
Pacific Herring (*Clupea pallasii*)	7	Kitasu Bay	Coastal ocean	34.5	(22.0)
Pacific Herring (*Clupea pallasii*)	2	Kwakshua Channel	Coastal ocean	23.9	(17.0)
Pacific Herring (*Clupea pallasii*)	4	Smith Inlet	Coastal ocean	31.2	(15.4)
Chinook Salmon (*Oncorhynchus tshawytscha*)	3	Rivers Inlet	Open ocean	17.7	(3.3)
Chum Salmon (*Oncorhynchus keta*)	2	Rivers Inlet	Open ocean	21.1	(13.3)
Pink Salmon (*Oncorhynchus gorbuscha*)	3	Rivers Inlet	Open ocean	9.3	(4.4)
Sockeye Salmon (*Oncorhynchus nerka*)	9	Rivers Inlet	Open ocean	8.5	(4.3)

[a] Whole-body digestion.
[b] Data from Shiel et al. (2012).

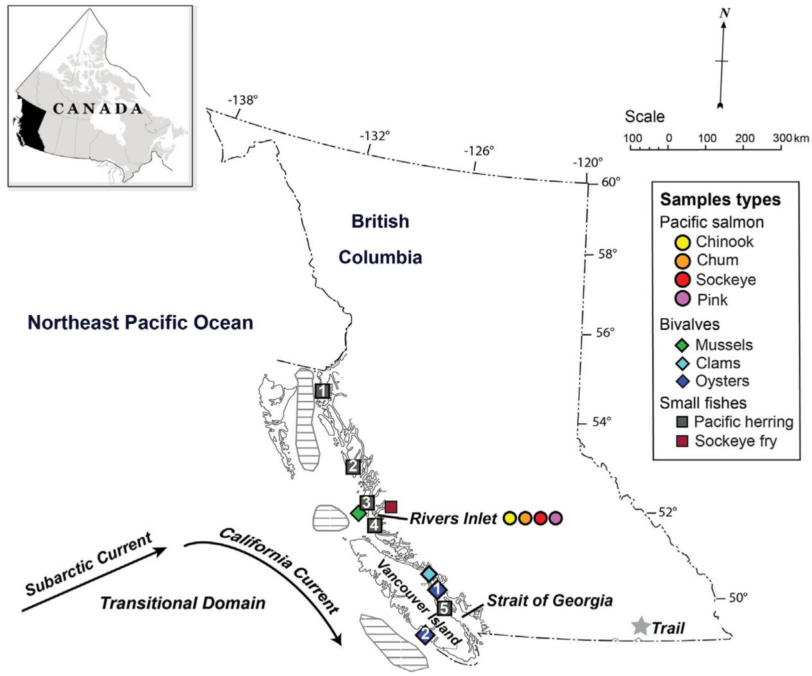

Fig. 1. Sample locations of fish and shellfish collected along the BC coast. All Pacific salmon are adult salmon that returned to Rivers Inlet to spawn. Sockeye fry are the juvenile salmon that lived in Owikeno Lake, upstream of Rivers Inlet. The oyster samples were collected in southern BC (1) Mainland and (2) Vancouver Island and data were reported by Shiel et al. (2012). Pacific herring samples were collected from five sites along the BC coast from north to south (1) Haida Bay, (2) Kitasu Bay, (3) Kwakshua channel, (4) Smith Inlet, (5) Denman Island. The striped areas and transitional domain signify the major feeding areas of Pacific herring and Rivers Inlet sockeye salmon, respectively (Espinasse et al., 2018; Fisheries and Oceans Canada, 2019). The grey star signifies the approximate location of Trail Teck Operations, a large Canadian metal and mining company.

analyzed using an Agilent 7700x quadrupole inductively coupled plasma mass spectrometer (ICP-MS, Agilent Technologies, Santa Clara, CA, USA) and a Nu AttoM high-resolution (sector-field) inductively coupled plasma mass spectrometer (HR-ICP-MS, Nu Instruments Ltd., UK), respectively. Clam samples were prepared and measured in the same way as oyster samples in a previous study and details regarding sample preparation and analytical methods can be found in Shiel et al. (2012). Briefly, ELEMENT2 HR-ICP-MS (Thermo Finnigan, Germany) and Nu Plasma multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS, Nu Instruments Ltd., UK) analyzed Pb concentration and isotopic composition of clam samples, respectively (Shiel et al., 2012).

There are no marine biological standard reference materials (SRMs) currently commercially available that are certified for Pb isotopic composition. We used TORT-3 (Lobster Hepatopancreas Reference Material, National Research Council Canada) as our in-house reference material because it is certified for Pb concentrations and is typically used in studies that focus on trace metals in marine fauna. We found that TORT-3, among other SRMs (e.g., DORM-4 fish protein; SRM 1566b oyster tissue), behaved most similarly to our fish and shellfish samples during sample digestion. TORT-3 was prepared and analyzed in the same manner as samples. Precision of Pb concentrations, estimated by replicate analysis of TORT-3 and fish samples, was better than 9 % (n = 6) and 10 % (n = 4), respectively. For Pb isotope analysis by HR-ICP-MS, each sample was bracketed by NIST SRM 981 for mass discrimination correction, using the standard-sample bracketing (SSB) correction method. For a given batch, the samples and bracketing standards were all prepared so the final concentration of Pb matched within 15 % for all analytes. We are currently building a Pb isotope database for SRM TORT-3 and use it as an in-house reference. Characterization of TORT-3 by both HR-ICP-MS and MC-ICP-MS yields consistent Pb isotope results ($^{206}Pb/^{207}Pb = 1.165 \pm 0.005$ (2SD), $^{208}Pb/^{206}Pb = 2.090 \pm 0.008$ from HR-ICP-MS (n = 9); $^{206}Pb/^{207}Pb = 1.164 \pm 0.001$, $^{208}Pb/^{206}Pb = 2.090 \pm 0.002$ from MC-ICP-MS (n = 3)).

3. Calculations

We estimated the fraction of Pb contributed by Asian sources in each sample using methods modified from previous studies (Ewing et al., 2010; Asher et al., 2018). As Chinese source ores and coals have characteristic higher $^{208}Pb/^{206}Pb$ due to higher Th/U in source rocks (Bollhöfer and Rosman, 2001; Bi et al., 2017), Asian Pb influence can be quantified using the vertical divergence of $^{208}Pb/^{206}Pb$ ratios from the local BC Pb isotopic array without Asian Pb influence. Here we use the leaded gasoline line to represent historical BC Pb isotope characteristics given its dominant impacts on the Pb isotopic signatures found in North America (Bollhöfer and Rosman, 2001). The fraction of Asian influence (F_{Asia}) is calculated as the ratio of Δ^{208}Pb Obs/Δ^{208}Pb Asia (Eq. (3)), where Δ^{208}Pb signifies the offset between the expected BC $^{208}Pb/^{206}Pb$ ratio and either the observed or the expected Asian $^{208}Pb/^{206}Pb$ ratio for a given $^{206}Pb/^{207}Pb$ ratio (Eqs. (1) and (2) and Fig. 2) (Ewing et al., 2010).

$$\Delta^{208}\text{Pb Obs} = {}^{208}\text{Pb}/{}^{206}\text{Pb}_{\text{Observed}} - {}^{208}\text{Pb}/{}^{206}\text{Pb}_{\text{BC}} \quad (1)$$

$$\Delta^{208}\text{Pb Asia} = {}^{208}\text{Pb}/{}^{206}\text{Pb}_{\text{Asia}} - {}^{208}\text{Pb}/{}^{206}\text{Pb}_{\text{BC}} \quad (2)$$

$$F_{\text{Asia}}\,(\%) = \Delta^{208}\text{Pb Obs} \,/\, \Delta^{208}\text{Pb Asia} * 100 \quad (3)$$

For local and regional context, we compare the Pb data from this study with environmental samples previously collected in the North Pacific Ocean and west coast of North America (California and northward). The Pb isotopic signatures in the ocean have

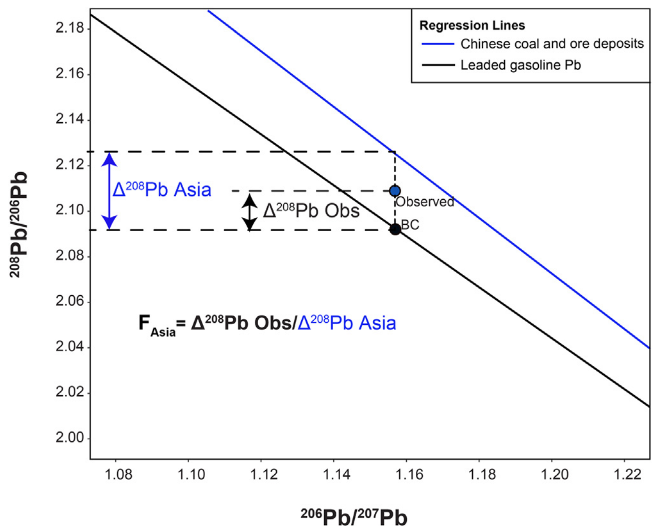

Fig. 2. Illustration of the calculation method for Asian Pb as a fraction of total Pb. The leaded gasoline line connects two major ore deposits (Australian Broken Hill and US Mississippi Valley) previously used for producing tetraethyllead used worldwide (Sangster et al., 2000; Soto-Jimenez et al., 2006).

changed substantially with time due to bans on leaded gasoline and rapidly growing industries in Asia. As the residence time of Pb in North Pacific surface water is relatively short (6–20 months) (Tsunogai and Nozaki, 1971; Gallon et al., 2006; Boyle et al., 2014), we limited our comparison to recent data in the literature (2000-present day). Differences in isotopic compositions and the extent of Asian Pb influence among fish from different feeding habitats were examined using ANOVA and Tukey's Honest Significant Difference (HSD) test. All statistical analyses were performed using R version 3.1.1 (R Core Team, 2014).

4. Results

4.1. Lead concentrations in fish and shellfish

Lead concentrations in muscle tissue of adult Pacific salmon (11.7 ± 7.0 ng/g dry weight) are higher than fresh albacore tuna muscle (1.5 ng/g, assuming 80 % moisture content) measured by Settle and Patterson (1980), which could be explained by a variety of factors including different Pb contamination levels in seawater between present day and four decades ago and distinct life histories and habitats across species. Lead concentrations in Pacific salmon are nearly 3 and 11 times lower than those of Pacific herring (30.1 ± 17.7 ng/g dry weight) and bivalves (125.1 ± 84.7 ng/g dry weight). This is consistent with prior findings that biomagnification of Pb generally does not occur during trophic transfer and inverse relationship between trophic levels and Pb concentrations in muscle tissue has been observed in laboratory and field research (Michaels and Flegal, 1990; Suedel et al., 1994; Cardwell et al., 2013). The average species-specific Pb concentrations by capture locations can be found in Table 1 and detailed Pb concentrations and isotopic compositions of each sample can be viewed in Table S1. Due to the whole-body digestion involving calcareous skeletal materials where Pb is concentrated relative to muscle tissue, the Pb concentrations of sockeye fry should not be compared directly with the muscle tissue of other fish.

The differences among Pb concentrations of salmon, herring, and bivalves largely result from distinct exposure pathways and physiological characteristics of individual species. For example, the primary exposure route of Pb for finfish appears to be the direct uptake of aqueous Pb^{2+} across the gills and dietary Pb intake can be readily excreted (Spry and Wiener, 1991; Jezierska and Witeska, 2006). In contrast, besides aqueous Pb intake, suspended particulate matters adsorbing much higher levels of Pb than ambient water concentrations also contributes considerably to the body burden of Pb in filter-feeding organisms such as bivalves (Prosi, 1989). This explains the much higher Pb tissue concentrations found in bivalves than finfish.

4.2. Lead isotopic compositions of marine biota vary with feeding habitat

Lead isotopic compositions of fish and shellfish that feed in lake, coastal, and open ocean waters are notably different, exhibiting a clear trend of Pb isotope ratios varying from highest ^{206}Pb/^{207}Pb in Owikeno Lake to lowest ^{206}Pb/^{207}Pb in the open ocean northeast Pacific (Fig. 3; see Fig. 1 for capture locations). Along with previously published local geological and environmental samples, the Pb isotopic composition of samples collected in this study form the BC Pb regression line (R^2 = 0.95, Fig. 3). Some of our Pacific herring results deviate from the BC Pb regression line and we address this anomaly in the discussion section. Sockeye salmon fry have not left Owikeno Lake since hatching and their Pb isotopic compositions mostly overlap those of BC volcanic rocks and Cascadia basin sediments (Mullen and Weis, 2013; Carpentier et al., 2014). This indicates that the sockeye salmon fry were primarily exposed to natural sources of Pb and that anthropogenic sources had minimal contribution to their Pb burden.

Bivalves (clams, mussels, and oysters) are filter feeders living on or near the seafloor and they obtain metals from both seawater and suspended solids including suspended sedimentary particles (Prosi, 1989; Gagnon and Fisher, 1997). Prior studies have found that Pb concentration and isotopic compositions in bivalves are more likely determined by sediment than seawater (Flegal et al., 1987; Wang et al., 1996; Shulkin et al., 2003; Dang et al., 2015). Bivalve samples collected in south, central BC coast, and Strait of Georgia display a wide range of Pb isotope ratios (^{206}Pb/^{207}Pb: 1.15–1.18), which overlap those of other environmental samples (road dust, lichen, river particulate matter) collected from southern BC where most of the BC human population and industries are located (Fig. 3). We infer from our data that diverse Pb sources, probably from southern BC, have contributed to the Pb content of suspended particles in southern and central BC coastal waters and Strait of Georgia, and ultimately lead to the variability of the Pb compositions in bivalves.

In contrast to the observed variation of Pb isotopic compositions in bivalves, Pacific herring exhibit smaller variation in ^{206}Pb/^{207}Pb (range: 1.16–1.17) and are not correlated with capture locations (Fig. S1). Herring is a coastal pelagic fish species that forage along BC coast and spawn in bays and estuaries (Fig. 1) (Hay et al., 2009; Therriault et al., 2009), and therefore their Pb isotopic compositions are expected to reflect those of seawater and be minimally influenced by sediment. The observed limited Pb isotopic variation in herring indicates a relatively homogenous Pb source in BC coastal ocean seawater and/or their migratory behaviors have likely helped integrate Pb exposures from various coastal locations (Fisheries and Oceans Canada, 2019).

Adult Pacific salmon exhibit distinct Pb isotopic compositions from those of inland and coastal fish (Fig. 3). Before returning to freshwater to spawn, the Rivers Inlet sockeye salmon typically spend 2–3 years foraging and obtain 90 % of their final adult body weight in offshore waters of the northeast Pacific Ocean, primarily in the easterly flowing subarctic current of the transitional domain (Fig. 1) (Groot and Margolis, 1991; Ishida et al., 1998; Espinasse et al., 2018). Therefore, these adult sockeye

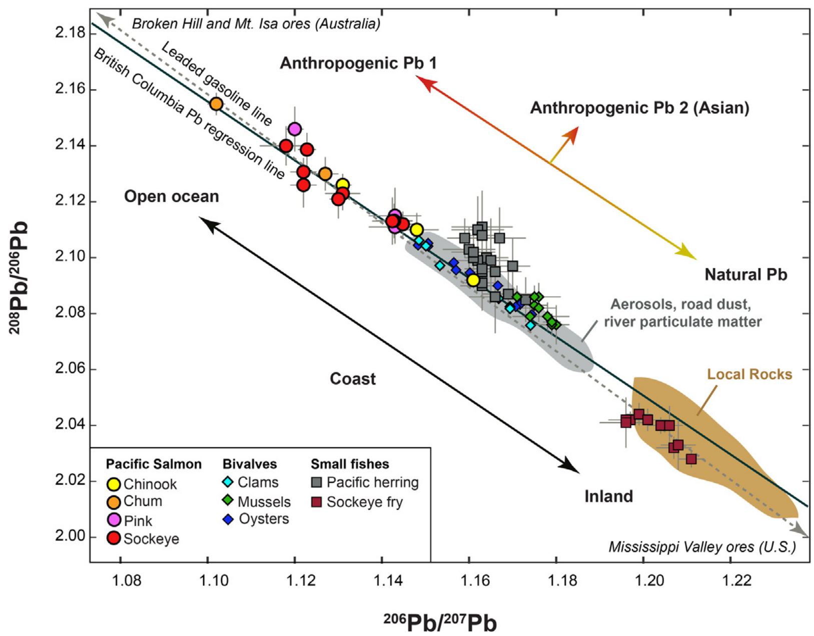

Fig. 3. Lead isotopic composition of all analyzed fish and shellfish from this study, with local geologic and environmental context. The solid line illustrates the Pb regression lines between local lithology, including the coast plutonic complex (Cui and Russell, 1995), Garibaldi volcanic belt (Mullen and Weis, 2013), Pacific sediment (Carpentier et al., 2014), Sullivan mine ore (Sangster et al., 2000), and a variety of environmental samples collected in BC, including fish and shellfish (this study), lichens (Simonetti et al., 2003), road dust (Preciado et al., 2007), BC oysters (Shiel et al., 2012), seawater (McAlister, 2015), and Fraser river particulate matter (Snauffer et al., 2010). The dashed line represents the leaded gasoline curve connecting two major ore deposits previously used for producing tetraethyllead used worldwide (Sangster et al., 2000; Soto-Jimenez et al., 2006). Error bars denote analytical uncertainty (2SE). Clams and oysters were analyzed by MC-ICP-MS and their error bars are smaller than the symbol size.

salmon would have accumulated most of their Pb from open ocean habitats and their Pb isotope ratios indicate that the open ocean seawater has a lower ^{206}Pb/^{207}Pb ratio compared to that of inland and coastal waters (Fig. 3). Adult chum and pink salmon from Rivers Inlet also mature in the offshore northeast Pacific and they show similar Pb isotopic compositions to those of sockeye salmon (Groot and Margolis, 1991; Ishida et al., 1998), confirming that lower ^{206}Pb/^{207}Pb compositions exist in the northeast Pacific Ocean. Unlike the other salmon species, the Pb isotopic compositions of Chinook salmon overlap with both oceanic and coastal fish (Fig. 3). This can be explained by Chinook salmon's behavioral differences in migration — some complete

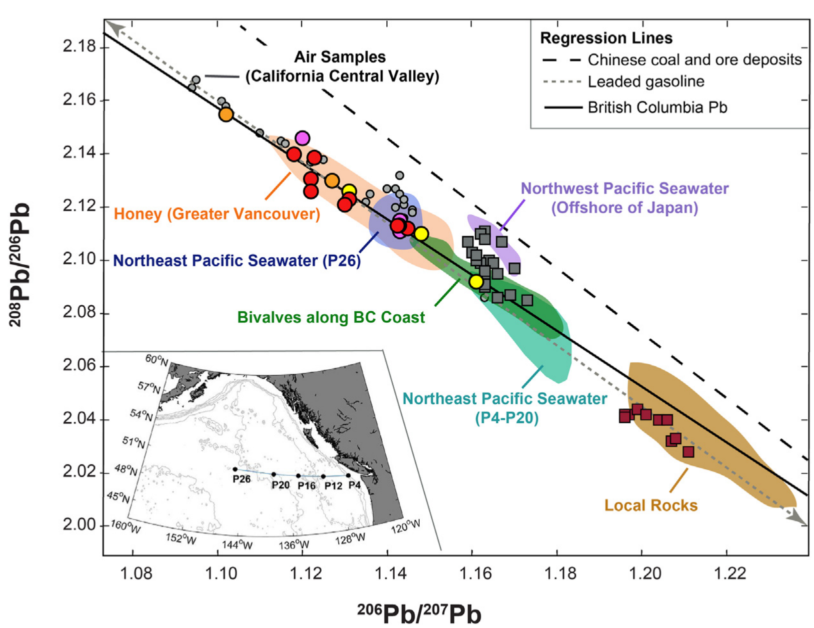

Fig. 4. Comparison of Pb isotopic composition between fish and shellfish collected from the BC coast and relevant environmental data from western North America and the northern Pacific Ocean. Data for seawater in the northwest Pacific (Zurbrick et al., 2017), northeast Pacific (McAlister, 2015), air samples from central California (Ewing et al., 2010), and honey samples from the Greater Vancouver Regional District (Smith et al., 2019) are also illustrated. Figure legend is the same as Fig. 3. The regression line of Chinese coal and ore deposit is from data compiled by Bi et al. (2017) and Cheng and Hu (2010). The selected sampling locations within the Line P transect in the northeast Pacific Ocean are depicted in the bottom left (Schuback, 2016).

extensive offshore oceanic migration while others spend most of their ocean life in coastal waters (Groot and Margolis, 1991). Our results therefore imply the great potential of Pb isotopes to distinguish different habitat use among fish stocks.

5. Discussion

5.1. Input of anthropogenic Pb to the northeast Pacific Ocean

Given that natural Pb sources in southern BC (Garibaldi volcanic belt; Cascadia basin sediments) and Alaska (Aleutians) exhibit high $^{206}Pb/^{207}Pb$ and low $^{208}Pb/^{206}Pb$ ratios (Fig. 3) (Kay et al., 1978; Mullen and Weis, 2013; Carpentier et al., 2014), the Pb isotopic compositions of Pacific salmon imply that there must be some anthropogenic Pb sources with low $^{206}Pb/^{207}Pb$ and high $^{208}Pb/^{206}Pb$ values dominating in the northeast Pacific Ocean. Recent studies reported Pb isotopic composition of honey samples from the Greater Vancouver Regional District, BC and air samples from cities in California's Central Valley also resemble those of Pacific salmon (Fig. 4) (Ewing et al., 2010; Smith et al., 2019). We therefore suggest that similar anthropogenic Pb sources prevail in western North America and the northeast Pacific Ocean. We further explore this anthropogenic Pb source by examining the relationship between $^{208}Pb/^{206}Pb$ values and the inverse of Pb concentration (1/[Pb]) in Pacific salmon (Fig. 5a). We find a two-end member mixing model can explain the variability in the Pb content and isotopic compositions in all salmon samples except for two Chinook samples. The higher Pb concentrations in these salmon are potentially a result of more influence from Pb source(s) with $^{208}Pb/^{206}Pb$ near 2.14 in the open ocean (black star in Fig. 5a).

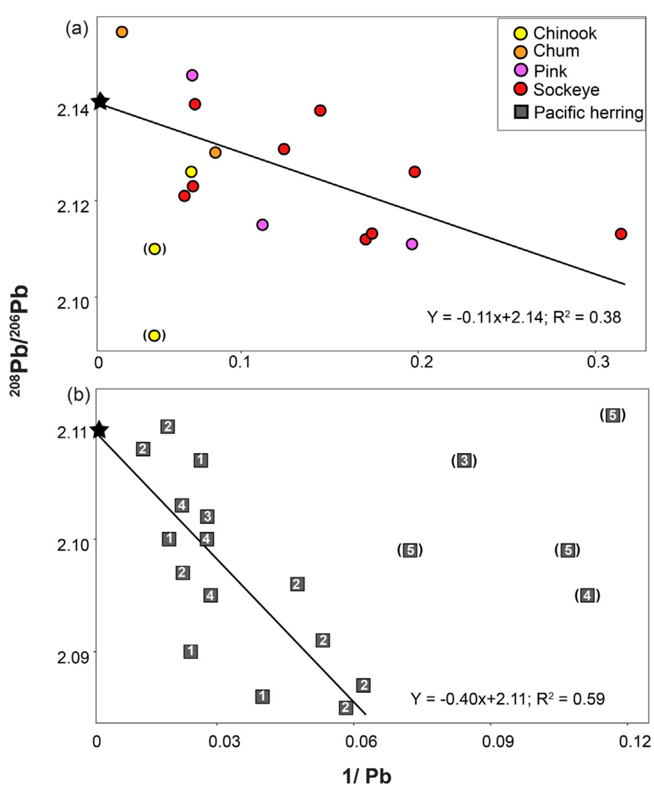

Fig. 5. Lead isotope ratio $^{208}Pb/^{206}Pb$ vs. 1/Pb concentration (ng/g dry weight)$^{-1}$ for (a) Pacific salmon and (b) Pacific herring. The stars represent the Pb pollution sources predicted based on linear regression. Samples in parentheses are not considered for linear regression (see explanation in text). The numbers in square symbols in (b) illustrate the capture location of each herring sample, as described in Fig. 1.

The BC Pb regression line based on local geological and environmental samples (y =-1.05 x+ 3.31) is very similar to the world legacy gasoline Pb line resulting from the addition of tetraethyl Pb (TEL) as an antiknock agent in gasoline in the late 20th century (y=-1.13 + 3.40) in Fig. 3. The majority of fish samples in this study align well along the legacy gasoline Pb line, suggesting that they are likely impacted by Pb sources resembling legacy gasoline Pb (Australian type and US Mississippi Valley type ores). The Pb end-member in the northeast Pacific Ocean reflected by salmon samples points to Australian-type Pb ore compositions (Mt. Isa and Broken Hill).

Some regional emissions of Pb ultimately sourced from Mt. Isa ore deposit may explain the observed Pb isotopic composition in salmon from the northeast Pacific. Prior studies attributed anthropogenic Pb in BC to the smelting and refining Trail facility of Teck Metals Ltd in southern BC (Fig. 1) and the use of aviation gasoline by small, propeller aircraft, which contains high levels of TEL (Simonetti et al., 2003; Shiel et al., 2012). The Trail facility has the largest on-site release of Pb to the atmosphere in BC (>700 kg/year during 2010–2014 and >1500 kg/year pre-2010) (Government of Canada, 2019). The Pb used for TEL production and for smelting at Trail is largely from Mt. Isa ore deposit after BC Sullivan mine was closed in 2001 (Shiel et al., 2012), characterized by a distinctly low $^{206}Pb/^{207}Pb$ ratio (≈1.04) (Sangster et al., 2000). Additional Pb isotopic measurements that assess major regional industrial sectors (e.g., fuel combustion, smelting, wastewater) are needed to further identify specific emission sources contributing Pb to the northeast Pacific Ocean and their evolution over time.

5.2. Distinct Pb sources in coastal waters from open ocean

The differences of Pb isotopic compositions observed in coastal samples (bivalves and herring) and adult salmon imply that the dominant Pb sources in coastal waters have lower $^{208}Pb/^{206}Pb$ than that of the open ocean and Australian-type Pb inputs (Fig. 3). Consistently, the predicted dominant Pb source for coastal fish (herring) has a lower $^{208}Pb/^{206}Pb$ value (2.11) than that derived from the majority of salmon samples (2.14) (black stars in Fig. 5). The two Chinook samples that do not follow the general pattern of Pb source attribution for other salmon have Pb isotopic compositions overlapping those of coastal fish and shellfish in Fig. 3. We infer that the same coastal Pb sources likely dominate Pb exposure in these Chinook and result in their lower $^{208}Pb/^{206}Pb$ values and higher Pb concentrations relative to those of other salmon samples (Fig. 5a).

Based on the literature values of Pb isotope ratios in other environmental samples, we postulate that the Pb in coastal waters are from urban runoff and river flows into coastal ocean and/or the coastal upwelling that brings deeper waters with legacy Asian and North American industrial lead ($^{206}Pb/^{207}Pb$ = 1.16–1.21; $^{208}Pb/^{206}Pb$ = 2.02–2.12) to surface waters (Flegal et al., 1986; McAlister, 2015). It is worth noting that five herring samples collected from the southern and central BC coast have the lowest Pb concentrations among all herring samples and fall off the mixing line derived from more contaminated herring (Fig. 5b). Although aqueous Pb concentration is the determining factor for Pb uptake, Pb concentrations in fish are also affected by many environmental, ecological, and physiological characteristics of individual species (Prosi, 1989). Our data reveal that it is challenging to use Pb isotopes in biological materials for tracking contamination sources when the Pb exposure is very low.

5.3. Influences of Chinese Pb in coastal and open ocean waters

Chinese coal, ore deposits, and aerosols have a distinct Pb isotopic fingerprint characterized by enrichment in ^{208}Pb, a radioactive decay product of ^{232}Th (Bollhöfer and Rosman, 2001; Bi et al., 2017). A number of North American studies have shown that environmental samples influenced by Chinese emissions tend to deviate from the local Pb isotopic mixing line toward the Chinese Pb regression line (Ewing et al., 2010; Christensen et al., 2015; Asher et al., 2018). Strong influence of Chinese Pb emissions in other East Asian countries such as Japan and South Korea is well known and previous studies concluded that Chinese emissions are the dominant source of Pb input to surface waters of the northwest and central Pacific Ocean (Wu et al., 2010; Gallon et al., 2011; Zurbrick et al., 2017). This is confirmed by Pb isotopic compositions of the surface waters of the Kuroshio current and the western subarctic gyre located offshore of Japan, plotting nearer to the Chinese Pb regression line (purple shade in Fig. 4) (Zurbrick et al., 2017). In contrast, Pb isotopic ratios of surface waters collected along line P in the northeast Pacific Ocean align well with the BC Pb mixing line and show little deviation toward the Chinese Pb line (green and blue shades in Fig. 4) (McAlister, 2015). The difference in seawater Pb isotope ratios between the northwest and northeast Pacific Ocean highlights the difference in predominant Pb sources between these geographic regions and indicates lower contribution of Pb from Chinese emissions in the northeast Pacific relative to the northwest Pacific.

Fig. 6 shows the species-specific fractions of Asian Pb influence derived from the vertical divergence of their ^{208}Pb/^{206}Pb ratios from the legacy gasoline line (see method in Fig. 2). We find that Asian Pb contributes significantly higher input (31 % of total Pb) in BC coastal ocean as reflected in the herring and central coast shellfish than other ecological zones (4 % in the open ocean, 5 % in northern Strait of Georgia, and minimal in inland freshwater ecosystems) (Figs. 6 and S2). Similar elevated Asian Pb impact was found in the sediment of lower Alberni Inlet and Barkley Sound on the west coast of Vancouver Island relative to the upper Alberni Inlet directly connected and influenced by upstream terrestrial/freshwater ecosystems (Ikehata, 2013). Determining the exact cause for this geographic discrepancy in Asian Pb impact will require further research. Here we provide two scenarios that may lead to the elevated Asian Pb fractions in coastal ocean observed in this study.

In the first scenario, heavier precipitation along the BC coast as an effect of the coastal mountain range may result in transporting higher amounts of Pb from the atmosphere to coastal ocean

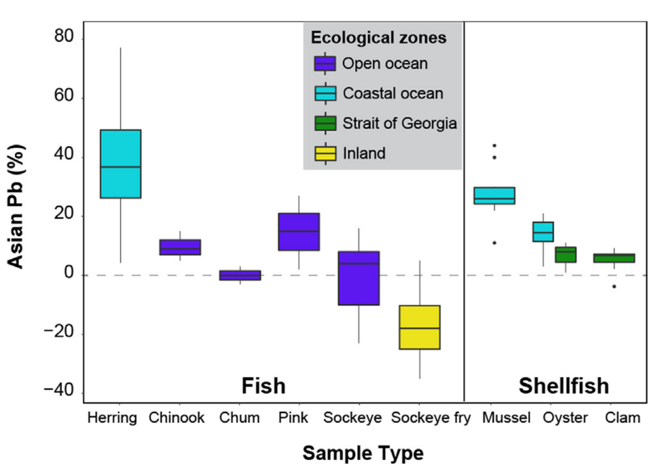

Fig. 6. The fractions of Asian Pb influence in each species used in this study, estimated by the deviation toward the Chinese Pb line from the leaded gasoline Pb line (Fig. 2). Box and whiskers show interquartile and 1.5 times the interquartile, respectively. Estimated negative values of Asian Pb contribution results from the uncertainty associated with the regression slopes and analytical uncertainty of fish measurements and they are not significant from zero (see Fig. 3).

through wet deposition, compared to the open ocean or inland BC. Most Pb in the ocean originates from atmospheric deposition and surface water Pb concentrations tend to follow atmospheric deposition fluxes of the previous few years (Boyle et al., 2014). Castello and Shelton (2004) estimated that the spatial distribution of total winter precipitation on the Pacific coast of BC and Washington rapidly decreases from 60 cm on the coast to 10 cm in open ocean within ~500 km. Several studies estimated trans-Pacific influence of Asian aerosols and dust account for an average of ~46 % of Pb along the coast of California during peak seasons (spring-summer) of Asian dust influence on North America (Ewing et al., 2010; Asher et al., 2018). Pacific herring in this study would reflect an accumulation of Pb from all seasons over 3–5 years (Ware, 1985), and suggest annual average Asian Pb input (39 % of total Pb) along the BC coast.

An alternative explanation involves anthropogenic source(s) that occur predominantly in the coastal ocean but not in the open ocean or inland. We consider emissions from heavy ship traffic in BC coastal ocean as the most plausible source (Johannessen and Macdonald, 2009). Central and northern BC have little industrial activities and small human populations (<0.5 people per sq. kilometer), therefore, the direct release of Pb to the oceans from BC inland activities is likely minimal (Statistics Canada, 2019). The BC coast serves 25 routes for frequent ferries within BC and additional routes from Washington, USA. It is also the inevitable cruise route to Alaska, which hosts about 500 voyages per year (Cruiselines International Association, 2019). Oceangoing traffic could release a considerable amount of Pb into the air and seawater given the high Pb content of these fuels (e.g., diesel and heavy fuel oil) (Akpoveta and Osakwe, 2014; Bi et al., 2017). The lower Asian Pb fractions observed in bivalves from the northern Strait of Georgia, relative to those from coastal ocean, could be a result of its restricted seawater and sediment exchange with coastal and open ocean (Ianson et al., 2016) (Fig. 6). Future research that investigates the Pb isotope compositions of fuels used for various kinds of marine transport along BC coast is required to elucidate the Pb contribution of marine traffic in the northeast Pacific coastal waters.

6. Conclusions

This study takes the initial step to establish the Pb isotope landscape of the northeast Pacific Ocean and shows a clear trend in Pb isotopic compositions, from highest ^{206}Pb/^{207}Pb in inland fish to lowest ^{206}Pb/^{207}Pb in open ocean fish. We conclude that some Australian-type Pb end-member source(s) with low ^{206}Pb/^{207}Pb and high ^{208}Pb/^{206}Pb values largely contribute to the Pb in the northeast Pacific whereas natural sources of Pb dominate freshwater ecosystems in the central BC coast. We estimate that Chinese-sourced Pb contributes to about 31 % of total Pb in coastal species but it has minimal input to the Pb in the freshwater lakes of central BC and open ocean. We propose heavy marine traffic and precipitation in coastal ocean as the most plausible explanations and thus themes for future research.

The distinct Pb isotopic compositions across species primarily foraging in inland, coastal, and offshore regions indicate great potential for using Pb isotopes to distinguish among habitats (e.g., coastal vs. open ocean) of migratory species in the northeast Pacific Ocean. To date, very few studies have explored the use of Pb isotopes for distinguishing stocks of marine organisms (fish, cephalopods, mammals) and identifying their nursery or feeding grounds (Outridge and Stewart, 1999; Spencer et al., 2000; Raimundo et al., 2009). Our results show the promise of Pb isotopes to discern two different behavioral patterns (offshore oceanic vs. coastal migration) within Chinook stocks. With further efforts to develop the Pb isotope landscape in the northeast Pacific and BC coast (e.g., GEOTRACES cruise GP15 in 2018, seasonal line

P cruises by UBC), we expect that Pb isotopes will become an effective tool for answering critical ecological and environmental questions in the region such as identifying pollution sources and foraging hotspots for species that have tremendous ecological and economic importance.

Supporting information

Additional two figures and all data produced in this study are available in supporting information.

Declaration of Competing Interest

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.ancene.2019.100234.

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