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Final Report

Trace Metals and Hydrocarbons in Sediments of Elson Lagoon (Barrow, Northwest Arctic Alaska) as Related to the Prudhoe Bay Industrial Region

by

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Abstract

A one-year study (2001–2002) in Elson Lagoon consisted of determining the concentrations of total mercury (THg) and methylmercury (MeHg) in five van Veen grab gross sediment samples from the lagoon, and a suite of heavy metals (arsenic, barium, cadmium, chromium, copper, lead, manganese, nickel, tin, vanadium and zinc) in the mud fraction (<63 µm size) of the sediments, as well as from stratigraphic sections of a gravity core sample. Additionally, hydrocarbons (normal and isoprenoid alkanes, triterpanes, steranes and polycyclic aromatic hydrocarbons [PAHs]) were analyzed in three of the five gross sediments. The geochronology of the core was established by ^{210}Pb- and ^{137}Cs-based methods.

The concentration levels of all the trace metals in the lagoon are generally below or similar to those in shales and unpolluted nearshore marine sediments. Encompassing the past ~70 years, stratigraphic samples show a net significant increase ($p < 0.05$) up the core in Ba, Cr and Mn accompanied by a significant ($p < 0.05$) down core increase in THg, and no significant change in the other metals. The up core increase in the three metals may be explained in terms of three possibilities: 1) the lagoon is getting contaminated with the metals, 2) a reflection of up core stratigraphic changes in lithology and/or organic matter, or 3) diagenetic upward mobilization of the metals in solution from anoxic sediments and subsequent oxidative precipitation at the surface. The down core increase in THg may be due to increased concentration of Hg by methylation in progressively anoxic sediment layers.

Long et al. [1995] defined the guideline values effects range–low (ERL) and effects range–median (ERM), representing possible-effects ranges, pertaining to concentrations of trace metals and organics in sediments, for the occurrence of adverse effects on biota at varying frequencies. ERL and ERM were determined for nine trace metals and several categories of organic compounds. The concentrations of eight metals in Elson Lagoon sediments were compared with corresponding metals in Long et al.'s study (their ninth metal, silver, was not included in this study). The comparison indicates that the concentrations of As, Cr, Cu and Ni in Elson Lagoon sediments are at levels such that adverse effects on biota would occasionally occur, whereas the concentrations of Cd, Pb, Zn and THg are below such levels. However, we suggest that the conclusions, based on the extrapolation of Long et al.'s [1995] guidelines to Elson Lagoon, must be verified by detailed site-specific investigations, including assessment of the proportion of a total metal in sediment that may be available for bioaccumulation and studies involving cause & effects relating to individual metals on resident organisms of the lagoon.

Trace metal data in mud from the urban-impacted Elson Lagoon were compared with those from the oil-related industrialized region of Colville Delta–Prudhoe Bay. By comparison, the Elson Lagoon muds have significantly higher concentrations ($p < 0.05$) of Cr, Ni, V and THg, with no significant regional difference in other metals. A possibility for the higher concentrations of the four metals in Elson Lagoon is a greater input of natural or anthropogenic metals from local sources. Alternate possibilities are a relatively lower rate of sedimentation in Elson Lagoon resulting in lower dilution of metals in sediments, or local focusing in Elson Lagoon of anthropogenic metal contaminants derived from Eurasia via long-distance atmospheric transport and subsequent deposition in precipitating snow and rain. The latter explanation is consistent

with the report of Snyder-Conn et al. [1997] and Garbarino et al. [2002] of relatively higher concentrations of several metals in Elson Lagoon snow samples than samples from the Colville Delta–Prudhoe Bay region, under similar snow and rain accumulation rates in both regions. A lack of significant ($p < 0.05$) correlation between all of the trace metals and clay percent in mud either suggests that the sediment granulometry is not an important factor partitioning the metals, or it is an artifact of the limited samples analyzed.

The overall molecular compositions of alkanes and polycyclic aromatic hydrocarbons, which are very similar to those reported in our previous study for sediments of the Colville Delta–Prudhoe Bay region, are characteristic of biogenic origin and very little petroleum input is reflected in their composition. Preliminary qualitative analyses indicate the presence of small amounts of coprostanol, a sewage sterol, and coprostanone, but no linear alkylbenzenes, a sewage tracer from household laundry. This suggests that sewage input in the sediments, if at all present, may not be important. Among a selected group of the PAHs examined, in context of Long et al.'s [1995] guidelines, the concentration of 2-methylnaphthalene in only one of the three samples (EL-3) of Elson Lagoon sediments is between ERL and ERM threshold levels wherein adverse effects on biota would occasionally occur. As suggested for the trace metals, these conclusions must be clarified by site-specific studies on resident organisms.

In summary, our study indicates that Elson Lagoon (as well as the Colville Delta–Prudhoe Bay nearshore of the North Slope of Alaska) has remained generally a relatively clean environment as far as sediment trace metals and hydrocarbons are concerned, despite the minor increase in some metals subsequent to accelerated anthropogenic activities in the lagoon during the past 30 years. The database on Elson Lagoon should be considered a baseline for monitoring inorganic and organic chemical contaminants in the lagoon region, an urban-impacted section of the North Slope of Alaska. These data will be useful for efforts relating to ecological risk assessment of the study area in the context of contaminant inputs.

Background and Relevance to Framework Issues

In recent years, there has been great concern about the Arctic marine environment, especially relating to the presence of anthropogenic contaminants and their possible biological effects. Until recently the Alaskan Arctic seas were considered generally free of contaminants. However, during the past decade organic and inorganic contaminants have found their way into the above so-called 'pristine' environment [Barrie et al 1992; Chernyak et al. 1996; Naidu et al. 1997, 2001]. Several possible sources could supply anthropogenic contaminants to the Beaufort Sea nearshore:

1. Local petroleum-related developmental activities including drilling operations [Snyder-Conn et al. 1997; Naidu et al. 2001].
2. Discharge of municipal litter from rural villages (e.g., Barrow) where adequate sewage and waste disposal and treatment facilities do not exist.

3. Debris deposited on the beaches and tundra adjacent to Elson Lagoon, at the west end of the Beaufort Sea coast (Figure 1), during the oil and gas exploration of the 1940s as well as the construction of the U.S. Air Force DEW (distant early warning) Line in the 1950s. Some of this debris was dispersed during occasional storms and washovers into Elson Lagoon, which could result in possible metal contamination.

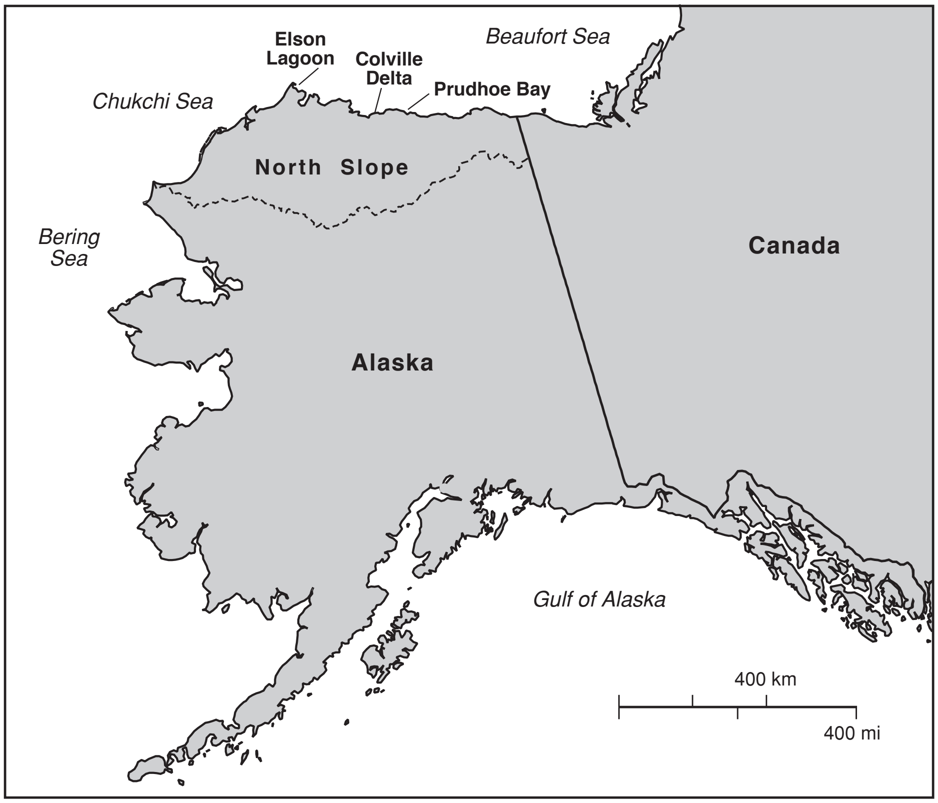

Figure 1. Map showing the North Slope of Alaska, Elson Lagoon, and the Colville River–Prudhoe Bay region.

4. There is also the possibility of long-range atmospheric and marine transport of anthropogenic contaminants to the Alaskan Arctic [Rahn and Lowenthal 1986; Barrie et al. 1992; Chernyak et al. 1996; AMAP 1997; Fitzgerald et al. 1998; Garbarino et al. 2002] especially from the highly polluted regions of Eurasia [Molnia and Taylor 1994]. Results of modeling exercises imply that northwest Alaska, including Elson Lagoon, is on the transport pathway of contaminants originating from Eurasia [Levno-Chythlook et al. 1999].

5. Several environmental factors unique to the Arctic may lead to biomagnification of toxic metals and organics. For example, the Arctic marine organisms which are lipid-rich are prone especially to accumulate lipophilic (fat loving) contaminants, and the relatively short/simple linear food chains in the Arctic can transfer contaminants readily from lower to higher trophic levels and result in their biomagnification in marine subsistence food sources [Ayotte et al. 1995; Mulvad et al.1996; Naidu et al. 2001].

6. Marine sediments, which act as an initial sink for particle-reactive trace metals and hydrocarbons [Förstner and Wittmann 1979; Chapman et al. 1998] could be a major source of contaminants to the benthos [Geffard et al. 2002; Lee and Wiberg 2002; Mountouris et al. 2002] and other organisms which have a close link with sediments [Long et al. 1995, 1998; Thompson et al. 1999; Valette-Silver 1999; Krantzberg et al. 2000].

For contaminant monitoring it is, therefore, important to record the regional concentrations of and understand the natural biogeochemical processes of trace metals and hydrocarbons in sediments. Such information is crucial to ecological risk assessment and management of marine areas with contaminant input. In this context, several investigations have reported the concentrations and partitioning of trace metals and hydrocarbons in sediments of the Alaskan Arctic seas [Naidu and Hood 1972; Shaw et al. 1979; Venkatesan and Kaplan 1982; Sweeney and Naidu 1989; Crecelius et al. 1991; Steinhauer and Boehm 1992; Snyder-Conn and Lubinski 1993; Naidu et al. 1997, 2001, 2003; Valette-Silver et al. 1999]. More recently, under a contract from CMI/MMS, historical changes in trace metals and hydrocarbons in sediments were reported for the Colville Delta–Prudhoe Bay region of the North Slope (Figure 1), with an objective to assess the impact on contaminant input resulting from oil-related developmental activities [Naidu et al. 2001].

By comparison, little environmental data are available for the Elson Lagoon region (Figure 2), [Hume et al. 1972; Schell 1975; Nummedal 1979; Walker 1991; Braddock and Gannon 2000]. In September 2000 Kelley and Naidu (unpublished) conducted a side-scan sonar survey of Elson Lagoon to identify the nature and extent of industrial and municipal debris. Brown et al. [in press] have reported the long-term erosion rates of the Elson Lagoon coast. The results of sea-ice–related research in Elson Lagoon and its environs, including preliminary data from our ongoing CMI study [Naidu and Kelley 2002; Naidu et al. 2003], appear in the proceedings of a recently concluded workshop [FRSGC/IARC 2002]. Lindberg et al. [2002] have monitored the photochemically-driven oxidation of gaseous mercury in the troposphere at Barrow. We believe that any extrapolation to Elson Lagoon of site-specific chemical data collected from the Colville Delta–Prudhoe Bay region [Venkatesan and Kaplan 1982; Sweeney and Naidu 1989; Crecelius et al. 1991; Steinhauer and Boehm 1992; Naidu et al. 2001]), as a benchmark for monitoring contaminants in that lagoon, will be of limited practical use. This is because the environmental settings and potential sources of contaminants in the above two regions are quite different. Elson Lagoon is essentially a large, shallow embayment with relatively little fluvial input of water and sediment, and sheltered from the open ocean by a continuous barrier spit (Figure 2). In contrast, the Colville Delta–Prudhoe Bay region, located to the east, is dominated by a lagoon-barrier chain, receives relatively large fluxes of particulates from rivers and high coastal erosion, and has greater access to the open ocean. Consequently, rates of deposition of sediment and associated contaminants in the latter region are likely to be higher and sediment reworking by ice gouging more pervasive. Additionally, the major potential source of contaminants to Elson Lagoon is of municipal origin (sewage and garbage litter disposal and community recreational activities), relatively higher input via long-distance atmospheric transport, and past defense-related activities (e.g., rocket firing experiments, road and airstrip building and maintenance, oil exploration, DEW Line construction) as opposed to the more easterly slope region where the dominant potential source of contaminants is presumably local ongoing petroleum-related activities.

One of the trends in petroleum exploration on the North Slope is progressively towards the west of Prudhoe Bay. It is quite possible that eventually Elson Lagoon and vicinity will be potential grounds for oil and gas exploration. It, therefore, seems timely to establish inorganic and organic chemical baselines to monitor contaminants in this environmentally-sensitive region. In fall 1999, with funding from the National Oceanic and Atmospheric Administration Office of Oceanic Resources Conservation and Assessment (NOAA/ORCA) and logistic support from the Barrow Arctic Science Consortium (BASC), Sathy Naidu, University of Alaska Fairbanks (UAF), collected samples of surficial sediments and gravity cores from five representative locations in Elson Lagoon (Figure 2, Table 1). Joan Braddock, also from UAF and under contract with CMI, investigated the microbial degradation potential of petroleum hydrocarbons in splits of the sediment samples. This present report is on the analyses of a suite of trace metals (As, Ba, Cd, Cr, Cu, Pb, Mn, Ni, Sn, V, Zn, and total and methyl mercury [THg and MeHg]) in the five surficial sediments and hydrocarbons (normal and isoprenoid alkanes, triterpenoids, steranes, and polycyclic aromatic hydrocarbons [PAHs]) in three of the five surficial sediments collected from Elson Lagoon in 1999. We also report stratigraphic variations in the metals in one core sample collected from the lagoon, the geochronology of the core, and historical changes in the metals consequent to increased urbanization, since 1970, of the community of Barrow lying adjacent to the lagoon (Figure 2). The data collected in this study, which are the first of their kind for Elson Lagoon, will provide a chemical baseline for an environmentally-sensitive region of the North Slope. A premise in this study has been that comparison of the data of Elson Lagoon with those we collected earlier from the Colville Delta–Prudhoe Bay region [Naidu et al. 2001] will help to set a basis for differentiating sources of contaminants derived from urban-impacted and petroleum-related activities for the North Slope nearshore region.

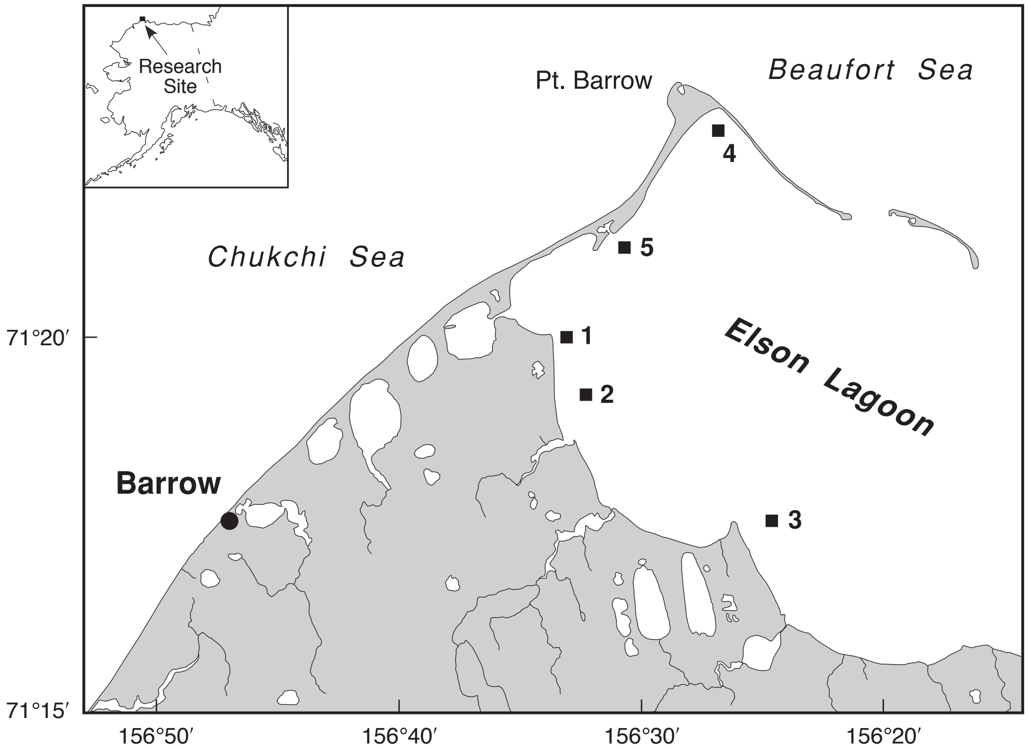

Figure 2. Sample sites in Elson Lagoon.

Table 1. Locations of sediment samples, water depth of locations, and contents (%) of solids, sand, silt and clay in the sediments [after Braddock and Gannon 2000].

Station	Latitude N	Longitude W	Water Depth (m)	Solids (%)	Sand (%)	Silt (%)	Clay (%)
EL-1	71° 20.00′	156° 33.58′	1.5	79	92.5	1.25	6.25
EL-2	71° 19.18′	156° 32.47′	1.8	64.6	55	20.75	16.25
EL-3	71° 17.53′	156° 24.71′	3.7	49.6	54	10	36
EL-4	71° 22.82′	156° 26.78′	2.8	97.8	3.75	43.75	52
EL-5	71° 21.18′	156° 30.89′	2.5	77.9	46.25	27.5	26.25

Study Objectives/Hypotheses

1. To determine the concentrations of a suite of trace metals (As, Ba, Cd, Cr, Cu, Pb, Mn, Ni, Sn, V, Zn) in the mud fraction (<63 µm size) of contemporary sediments, and THg, MeHg and hydrocarbons (normal and isoprenoid alkanes, triterpenoids and steranes, and PAHs) in gross surficial sediments of Elson Lagoon.

2. To analyze historical changes in concentrations of the trace metals, based on stratigraphic changes in the metals in a dated sediment core.

3. To develop a geochronology for the sediment core, using ^{210}Pb- and ^{137}Cs-based dating methods and AMS (accelerator mass spectrometry) ^{14}C dating of calcareous fossils.

4. To compare the sediment trace metals and hydrocarbons of Elson Lagoon with those of the Colville Delta–Prudhoe Bay region of the North Slope, with a motive to identify differences in the nature and concentrations of contaminants derived from urban-impacted and petroleum-related industrial activities.

5. To examine concentrations of the metals and hydrocarbons in the context of thresholds of varying rates of occurrence of adverse effects on marine organisms that live in and close to sediments [Long et al. 1995].

One of our hypotheses is that there have been no significant historical changes in sediment trace metal concentrations in Elson Lagoon during the past 30 years, despite increased anthropogenic activities resulting from urban development in Barrow and its impact on the lagoon. The second hypothesis is that there are no significant differences in the concentrations of sediment trace metals and the distribution pattern of hydrocarbons of urban-impacted Elson Lagoon and the Colville Delta–Prudhoe Bay region, which is impacted by petroleum-related activities.

Materials and Methods

Samples and analytical methods

In this study the sediments analyzed consist of five van Veen grab samples and one of the three gravity cores collected in fall 1999 in Elson Lagoon at selected locations (Figure 2, Table 1). Each of the grab samples was split into three subsamples and transferred into I-Chem glass jars. The core sample was sectioned at 1-cm intervals from the core top, and each of the sections was split into three subsamples and placed in pre-baked I-Chem jars. All the above sediment samples were stored frozen until analysis. One split of the five grab samples and sections of one core (core EL-3) were shipped frozen to Frontier Geosciences Inc., Seattle for analysis of trace metals, and splits of three grab samples (EL-1, EL-2 and EL-3) were sent to M. Indira Venkatesan (University of California, Los Angeles) for analysis of hydrocarbons. Because of limited funds received from CMI it was not possible to analyze the metal content in the entire core and the hydrocarbons in two (EL-4 and EL-5) of the three grab samples collected. The metal analyses were restricted to 1-cm intervals of continuous sections from the core top to 9 cm below and one section taken at the bottom of the core at the19-cm depth. Presumably, the trace metal data on the basal sediment section (about 70 years of age based on ^{210}Pb geochronology, see results section) provide a baseline for metal concentrations prior to intense urbanization of Elson Lagoon.

The trace metals and hydrocarbons were analyzed according to the methods followed in the previously funded CMI/MMS projects on sediments collected elsewhere along the North Slope nearshore [Crecelius et al. 1991; Steinhauer and Boehm 1992; Naidu et al. 2001]. As in previous studies, the mud fraction, consisting of the silt plus clay sizes (<63 μm) of each of the sediment samples, was taken for the metal analyses. By using this fraction the effect of varying granulometry on the trace metal contents between the samples was minimized [Daskalakis and O'Connor 1995; Naidu et al. 2001]. Further, in this fraction, which is generally enriched in clay minerals and organics, the particle-reactive metals are usually concentrated by ion exchange, adsorption and clay–metal–organic complex formation. The analyses on Elson Lagoon muds will have an additional use—that of comparing metal data with samples collected on the same size sediments in previous studies from other parts of the North Slope and Chukchi Sea nearshore [Naidu et al. 2001 and references therein]. Consequently, the mud fraction is a more desirable sediment component for monitoring metal pollution.

For separation of the mud fraction, a 5-g split of a thawed grab or core subsample was taken and suspended in deionized distilled water and homogenized. The suspension was sieved through a 230-mesh nylon screen, and the separated mud particles were then freeze-dried and powdered using an agate mortar and pestle. It is possible that during the process of separating the mud fraction by wet sieving the gross sediments may have hydrolyzed some metals, or desorbed them from the sediment (or both). If this occurred then some metal portions were lost to the water that was subsequently siphoned and discarded. It is assumed that the fraction of metals thus lost, and excluded from the analysis, was negligible.

A 0.5-g aliquot of the dried powder was taken into a 140-ml Teflon bomb and digested in 15 ml of a high-purity concentrated 7-ml-HF+15-ml-HNO_3+3-ml-HCl acid mixture in a conventional oven at 130 °C for 12 hours. After cooling to room temperature, the digest was diluted to 100 ml with water processed by a Milli-Q Ultrapure Water Purification System, and the diluted sample was returned to its original bomb. The resulting solution was dried and then again dissolved in 10% HNO_3 and made up to 20 ml by adding deionized distilled water. From this final solution Ba, Cd, Cr, Cu, Pb, Mn, Ni, Sn, V, and Zn were analyzed using either an inductively coupled plasma mass spectrometer (PerkinElmer Elan 6100 quadrupole ICP-MS) or by graphite furnace atomic absorption spectrometry (GF-AAS). Arsenic was analyzed in a separate split of the dry mud with a PSA Excalibur automated hydride generation atomic fluorescence spectrometry (AFS) system, using EPA method 1632.

Total mercury and the more toxic methyl mercury were analyzed on the whole sediment rather than the mud fraction, to avoid any possible loss of the highly volatile Hg from the gross sediment during the sieving process. Total Hg was analyzed on 0.01–1.0 ml aliquots from the solution retained in the bomb, using $SnCl_2$ reduction, dual gold amalgamation pre-concentration, and cold-vapor atomic absorption spectrometry (CVAAS). Methyl mercury was analyzed on leachates of an acidic bromide–methylene chloride extraction of sediment, using aqueous phase ethylation, followed by purging onto Carbotrap adsorbent resin, isothermal GC (gas chromatograph) separation, and cold-vapor atomic fluorescence spectrometry (CVAF). Further details on the methods of the THg and MeHg analyses are described in Bloom [1989, 1992] and Bloom and Fitzgerald [1988].

The QA/QC (quality assurance/quality control) protocol prescribed by the U.S. Environmental Protection Agency (EPA) for metal analysis was followed. It consisted of determining analytical precision through replicate runs, checking analytical accuracy via analyses of certified standard reference materials (1632-b, coal; NIST-2710, soil; NRCC DORM-2, dogfish tissue [for MeHg only]; and NIST-600, bauxite), analyzing spiked sample and reagent blanks, and using ultrapure reagents. The estimated method detection limits, MDL, (with three standard deviations, SD, of the blanks) for the trace metals, and the QA/QC results are shown in Table 2. There is close agreement between the analytical results for the reference materials and their certified values. It should be noted that the concentrations of MeHg in a few samples are below the MDL (i.e., 0.036 ng g^{-1}) and, therefore, the results for these samples should be considered close to the MDL, at best. As in our previous CMI-funded study, the trace metal analyses were subcontracted to Frontier Geosciences Inc., Seattle, which in 1997 participated successfully in the NOAA/NIST (National Institute of Standards and Testing) round robin interlaboratory exercise NOAA/11 for certification of QA/QC. This exercise was conducted by the National Research Council of Canada (NRCC). The Frontier Geosciences rating was good to excellent.

The analyses of hydrocarbons were on gross sediments, according to well established methods [Venkatesan et al. 1987; Venkatesan 1994] and as described in detail in our previous study on Colville Delta–Prudhoe Bay region sediments [Naidu et al. 2001]. After solvent extraction and GC separation into individual compounds, the alkanes were quantified using a flame ionization detector. Tricyclic di- and pentacyclic triterpenoids and PAHs were measured by GC/mass spectrometry. As in our last CMI-funded investigation [Naidu et al. 2001] 24 PAHs were analyzed, as well as six additional PAHs, which are routinely analyzed by NOAA/NS&T (National Status and Trends Program). To maintain continuity and high standards, the hydrocarbon analysis was subcontracted to M. Indira Venkatesan (University of California, Los Angeles), who first in 1992 and then in 1999 successfully participated in interlaboratory round-robin exercises conducted by NIST/NOAA-NS&T, for the QA/QC program.

The geochronology (sediment accumulation rate) of a sediment core (EL-3) from Elson Lagoon was established by the age–depth model using ^{210}Pb and ^{137}Cs dating [Weiss and Naidu 1986; Baskaran and Naidu 1995; Naidu et al. 1999]. The activities of the two radionuclides in stratigraphic sections of core EL-3 (Figure 2) were measured by high resolution gamma ray mass spectrometry. The radionuclide analysis was subcontracted to Mark Baskaran, Wayne State University, Detroit. The 1-cm stratigraphic sections from the core were microscopically examined in search of calcareous fossil shell/test samples for the purpose of AMS ^{14}C dating. However, no such samples were to be found in any of the core sections.

Grain size distributions of the grab samples [Braddock and Gannon 2000] were used to explain elemental and hydrocarbon distributions in the study area.

Table 2. Results of QA/QC analysis.

Trace Metals ($\mu g\ g^{-1}$ dry weight basis [MeHg $ng\ g^{-1}$])													
Sample	**As**	**Ba**	**Cd**	**Cr**	**Cu**	**Pb**	**Mn**	**Ni**	**Sn**	**V**	**Zn**	**Thg**	**MeHg ($ng\ g^{-1}$)**
blank–1	0.0	0.1	–0.01	0.2	–0.13	0.2	0.17	0.03	0.6	0.1	–0.16	0.0009	0.002
blank–2	–0.3	0.3	0.02	0.1	–0.09	0.2	0.21	0.04	0.7	0.1	–0.09	0.0007	–0.005
blank–3	0.0	0.6	–0.02	0.2	–0.14	0.1	0.21	0.08	0.5	0.1	–0.11	0.0011	0.002
blank–4	0.2	0.1	–0.01	0.3	0.01	0.1	0.18	0.18	0.6	0.1	–0.19	0.0007	NP
blank–5	–0.4	0.1	–0.01	0.0	–0.14	0.1	0.14	0.03	0.4	–0.2	–0.13	0.0011	NP
blank–6	–0.2	0.1	0.01	0.0	–0.06	0.2	0.22	0.05	0.6	0.0	–0.17	0.0014	NP
blank–7	–0.1	0.5	0.04	0.1	–0.14	0.3	0.18	0.06	0.4	–0.2	–0.14	0.0013	NP
blank–8	–0.3	0.1	0.04	–0.1	–0.10	0.3	0.12	0.06	0.7	–0.2	–0.10	0.0006	NP
mean	–0.1	0.2	0.01	0.1	–0.10	0.2	0.18	0.07	0.6	0.0	–0.14	0.0010	0.008
estimated MDL	0.6	0.5	0.07	0.4	0.16	0.3	0.10	0.15	0.4	0.4	0.11	0.0009	0.036
EL-5 rep 1 (2.0–3.0 cm for MeHg only)	16.8	500	0.213	83.3	26.1	14.7	283	39.7	2.1	129	102	0.0578	0.008
EL-5 rep 2 (2.0–3.0 cm for MeHg only)	16.6	487	0.162	85.7	26.4	14.3	273	39.5	2.2	128	99	0.0504	0.001
mean	16.7	494	0.187	84.5	26.3	14.5	278	39.6	2.1	129	100	0.0541	0.004
RPD (%)	1.1	2.7	27.1	2.9	1.1	2.9	3.6	0.5	4.1	1.0	3.2	13.7	158.9*
matrix spike level	218	*not spkd*	218	218	218	218	218	218	218	218	218	2.18	5.590
EL-5 + MS (2.0–3.0 cm + MS for MeHg only)	217	517	193	282	228	221	477	242	206	326	297	2.27	5.639
% recovery	91.8	*not spkd*	88.3	90.7	92.4	94.5	91.2	92.5	93.5	90.3	90.3	101.8	100.8
matrix spike dup level	216	*not spkd*	216	216	216	216	216	216	216	216	216	2.17	5.730
EL-5 + MSD (2.0–3.0 cm + MS for MeHg only)	207	504	187	281	225	216	477	239	199	324	283	2.09	6.091
% recovery	88.0	*not spkd*	86.4	91.1	91.9	93.2	92.2	92.2	91.0	90.3	84.7	94.0	106.2
mean recovery	89.9	*not spkd*	87.4	90.9	92.2	93.8	91.7	92.5	92.3	90.3	87.5	97.9	103.5
RPD (%)	4.1	*not spkd*	2.1	0.5	0.5	1.5	1.0	0.7	2.7	0.0	6.4	8.0	5.2
1632-b (coal) (DORM-2 [dogfish] for MeHg only)	3.26	60.7	0.053	9.9	6.30	3.85	11.47	6.34	0.4	13.0	12.17	0.0767	1.889
true value	3.72	67.5	0.057	*not cert*	6.28	3.67	12.40	6.10	*not cert*	14.0	11.89	0.0780	2.240
% recovery	87.7	90.0	93.3	*not cert*	100.3	104.9	92.5	103.9	*not cert*	92.9	102.4	98.3	84.3
NIST-2710 (soil) (mL spike for MeHg only)	549	623	19.0	27.6	2710	5165	9561	13.3	6.0	67.9	5991	31.1	0.046
true value	626.0	707	21.8	*not cert*	2950	5532	10100	14.3	*not cert*	76.6	6952	32.6	0.050
% recovery	87.7	88.1	87.1	*not cert*	91.9	93.4	94.7	93.3	*not cert*	88.6	86.2	95.5	92.0
NIST-600 (bauxite)	16.6	55.5	0.04	132	14.0	17.3	106	4.26	7.8	334	24.2	0.0794	NP
true value	*not cert*	*not cert*	*not cert*	164	*not cert*	*not cert*	101	*not cert*	*not cert*	336	24	0.0790	NP
recovery	*not cert*	*not cert*	*not cert*	80.3	*not cert*	*not cert*	105.3	*not cert*	*not cert*	99.5	100.7	100.5	NP

*anomalous data

MDL = method detection limit, RPD (%) = relative percent difference, MS = matrix spike, MSD = matrix spike duplicate, NP = not performed

Statistical analysis consisted of determining correlation coefficients (r-values) between the metal concentrations and clay percent in mud, which were calculated by prorating the proportion of silt plus clay in the gross sample at 100%, (Table 1). The statistical analysis also included a comparison of mean concentrations of the individual metals in sediments of Elson Lagoon and of Colville Delta–Prudhoe Bay [Naidu et al. 2001]. The Student-t test and null hypothesis [Zar 1984] were considered in the latter analysis.

The significance of up core stratigraphic net variation in metal concentrations was verified using regression analysis. In this analysis the data on the basal core section were also considered. The correlation coefficient (r-value) and Student-t values were considered significant if $p < 0.05$.

Results

The percents of solids, sand, silt and clay in the individual sediment samples are included in Table 1. Table 3 shows the concentrations (on dry weight basis) of the trace metals in the mud fraction (<63 µm size) of the five sediment samples of Elson Lagoon. With the exception of Cr and Mn, the concentrations of each of the metals in four out of five samples (excepion EL-1) are similar. This is reflected in the overall relatively low standard deviation (SD) for all metals except Cr and Mn (Table 4). The stratigraphic variations of the metal concentrations are also shown in Table 3 and illustrated in Figure 3. Regression analysis indicates that there are significant net increases ($p < 0.05$) up core in Cr, Mn and Ba, and an up core decrease in THg, accompanied by no significant stratigraphic variations in the remaining metals. It should be noted that the determination of the up core net trends is based on all metal data from the core, including those from the disjointed core section, but with no data from core sections in between the upper 9 cm of the core and the core bottom section. The ^{210}Pb- and ^{137}Cs-based linear sediment accumulation rates for Elson Lagoon are 0.27 cm yr^{-1} and 0.23 cm yr^{-1}, respectively, and the mass accumulation rate based on the ^{210}Pb method is 0.34 g cm^{-2} yr^{-1} as estimated by subcontractor Mark Baskaran at Wayne State University. Therefore, the stratigraphic data on the 19-cm long core, EL-3, encompasses time-series variations in metals during the past 70–83 years.

Table 3. Trace metals in freeze-dried sediments of Elson Lagoon grab samples (EL-1 to EL-5) and core sample EL-3. R-values are indicated for regression analysis on down-the-core net metal stratigraphic changes for core EL-3.

Trace Metals (μg g^{-1} dry weight basis [MeHg ng g^{-1}])														
Sample		**As**	**Ba**	**Cd**	**Cr**	**Cu**	**Pb**	**Mn**	**Ni**	**Sn**	**V**	**Zn**	**THg**	**MeHg (ng g^{-1})**
Grab	**EL-1**	31.4	559	0.171	188	36.7	25.6	1,388	52.3	3.1	211	146	0.0654	0.012
	EL-2	17.6	631	0.195	134	35.7	19.3	406	51.6	2.5	186	141	0.0639	0.054
	EL-3	14.8	520	0.176	91.3	31.2	15.5	320	45.3	2.1	147	114	0.0978	0.133
	EL-4	21.9	531	0.143	96.9	35.0	18.1	341	50.6	2.5	168	126	0.0908	0.066
	EL-5	18.4	525	0.160	94.1	33.1	16.8	331	47.9	2.3	157	120	0.0541	0.047
Core EL-3	**0–1 cm**	23.3	553	0.859	203	35.5	24.7	1,144	50.1	2.5	178	131	0.0630	0.002
	1–2 cm	19.1	562	0.515	358	30.1	16.8	921	75.3	3.8	156	108	0.0519	0.008
	2–3 cm	22.8	777	0.234	486	30.9	19.7	914	47.2	6.5	184	126	0.0530	0.008
	3–4 cm	15.6	683	0.288	245	26.3	16.1	382	36.6	3.2	158	111	0.0529	0.001
	4–5 cm	11.2	542	0.101	190	27.0	13.9	259	43.0	2.1	147	107	0.0532	0.037
	5–6 cm	12.5	568	0.112	136	30.3	15.7	270	45.4	3.4	164	124	0.0544	0.068
	6–7 cm	14.3	532	0.178	130	29.6	16.7	263	45.4	3.0	174	122	0.0575	0.081
	7–8 cm	17.0	579	0.225	198	28.7	16.7	271	43.0	2.7	178	122	0.0525	0.035
	8–9 cm	23.2	561	0.269	171	34.3	22.5	258	50.8	7.8	212	137	0.0638	0.032
	18–19 cm	22.1	432	0.224	99	26.9	20.0	197	39.3	2.2	166	115	0.0735	0.013
	r-value	0.14	–0.61	–0.44	–0.58	–0.35	0.06	–0.67	–0.42	–0.12	0.13	0.00	0.73	0.15

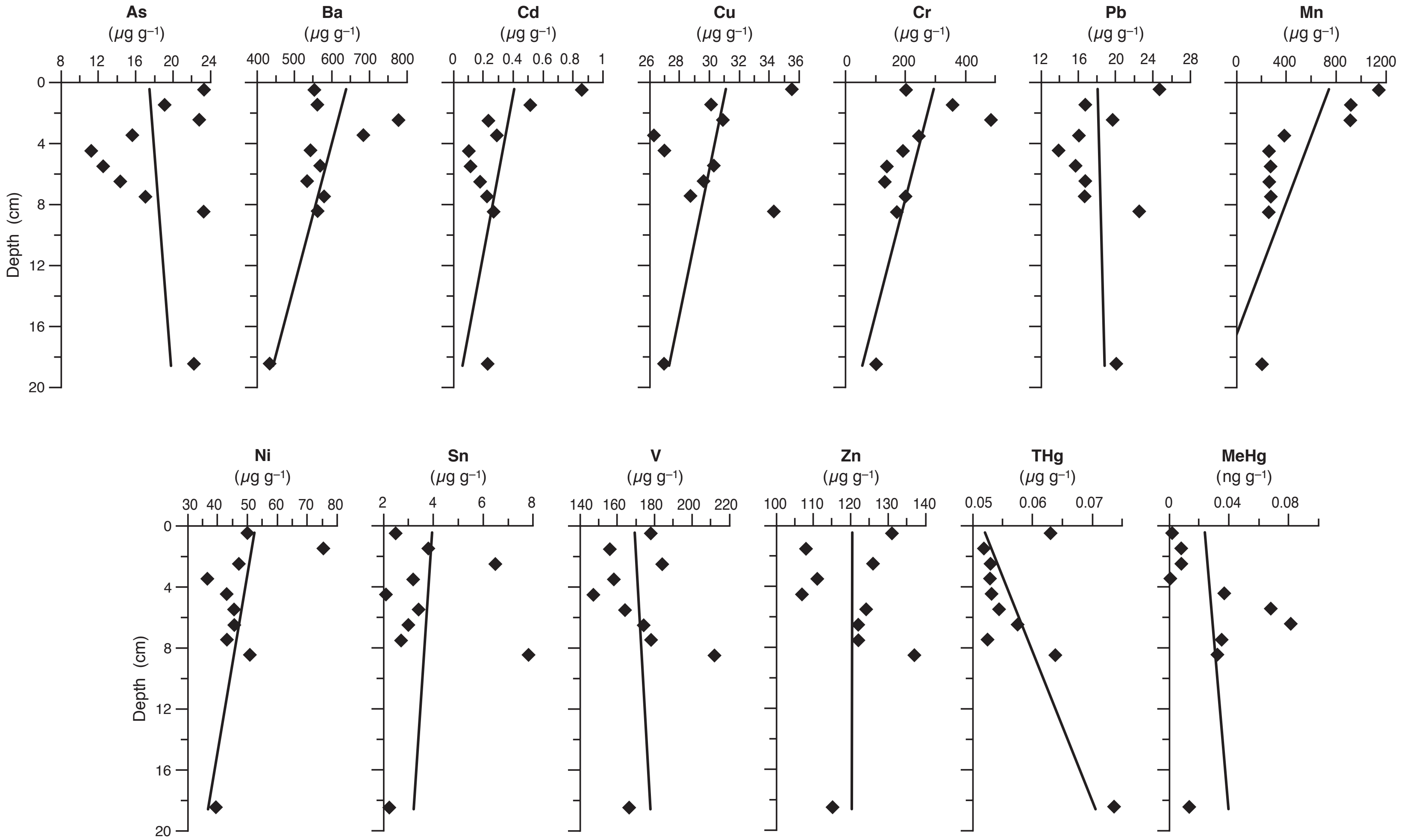

Figure 3. Stratigraphic variations of trace metals in core EL-3. Refer to Table 3 for raw data.

Table 4 compares the mean concentrations in muds of trace metals from the urban-impacted Elson Lagoon with those we collected from the oil-related industrialized region of Colville Delta–Prudhoe Bay [Naidu et al. 2001]. By comparison, the Elson Lagoon muds have relatively higher mean concentrations ($p < 0.05$) of Cr, Ni, V and THg accompanied by no significant regional differences in the means of the remaining metals.

Table 4. Comparison of the mean concentrations of trace metals in the mud fraction (<63 µm size) of sediments from two nearshore/lagoon regions of the North Slope of Alaska. The concentration of THg is in ng g^{-1} whereas that of other elements is in µg g^{-1}.

	As	**Ba**	**Cd**	**Cu**	**Cr**	**Pb**	**Mn**	**Ni**	**V**	**Zn**	**THg**
Elson Lagoon											
Mean	21	553	0.169	34	121	19	557	50	174	129	74
Std. Dev.	6.4	46	0.019	2.2	41.4	3.9	466	2.9	25	13.7	19
Colville Delta–Prudhoe Bay											
Mean	16	569	0.264	27	63	15	540	34	117	96	17
Std. Dev.	7	114	0.115	7	13	6	337	7	23	18	7

Table 5 shows correlation coefficient values between metals. For example, it is clear that V covaries with Cu, Pb, Ni, Sn, and Zn; and Ba with Cr; whereas there is no correlation between THg or MeHg with any of the other metals or clay percent. It is to be noted that no significant correlation was found between any of the metals and the clay percent in mud.

Table 5. Correlations between trace elements, and between elements and clay % of muds from Elson Lagoon. (bold type = significant correlation for n = 5 at $p < 0.05 = 0.878$; other correlations are in gray type)

	As	**Ba**	**Cd**	**Cr**	**Cu**	**Pb**	**Mn**	**Ni**	**Sn**	**V**	**Zn**	**THg**	**MeHg**	**Clay %**
As	**1.00**													
Ba	0.05	**1.00**												
Cd	–0.22	0.74	**1.00**											
Cr	–0.29	**0.92**	0.66	**1.00**										
Cu	0.77	0.60	0.06	0.35	**1.00**									
Pb	**0.93**	0.37	0.14	0.00	0.85	**1.00**								
Mn	**0.92**	0.14	0.10	–0.24	0.65	**0.95**	**1.00**							
Ni	0.71	0.64	0.08	0.43	**1.00**	0.80	0.58	**1.00**						
Sn	**0.96**	0.31	0.00	–0.03	**0.89**	**0.99**	**0.92**	0.84	**1.00**					
V	0.84	0.58	0.27	0.24	**0.93**	**0.97**	0.86	**0.90**	**0.96**	**1.00**				
Zn	0.71	0.74	0.39	0.45	**0.94**	**0.89**	0.73	**0.93**	0.87	**0.98**	**1.00**			
THg	–0.28	–0.42	–0.25	–0.30	–0.44	–0.39	–0.30	–0.44	–0.38	–0.45	–0.48	**1.00**		
MeHg	–0.78	–0.36	0.07	–0.12	–0.85	–0.79	–0.66	–0.83	–0.83	–0.81	–0.78	0.77	**1.00**	
Clay %	0.58	–0.36	–0.03	–0.64	0.04	0.52	0.73	–0.05	0.46	0.33	0.15	0.31	–0.01	**1.00**

Table 6 lists the concentrations (ng g^{-1}) of total and individual homologs of *n*-alkanes and ratios of selected alkanes for Elson Lagoon and sample 6G from the previous Colville River–Prudhoe Bay study [Naidu et al. 2001]. The Elson Lagoon gas chromatograms showed a trimodal distribution with maxima at *n*-C17, *n*-C23 and *n*-C27, and with alkanes >*n*-C25 predominating in the samples. The odd/even ratios in all the samples are greater than one. Among the triterpanes 27(17β), 29ββ and diploptene are the major components (Figure 4). The former two hopanes are the most dominant in EL-1 and EL-2 and diploptene is in EL-3. 29αβ and 30αβ hopanes are present in smaller amounts relative to biogenic hopanes. Extended hopanes with >*n*-C31 are present only in trace quantities and only the R isomer is detected.

Table 6. Concentrations of *n*-alkanes in Elson Lagoon sediments and in sample 6G from the Colville River Delta [Naidu et al. 2001].

	Sample ID			
	Elson Lagoon			**Colville Delta**
***n*-alkane (ng g^{-1} dry)**	**EL-1**	**EL-2**	**EL-3**	**6G***
n-C10	41	21	164	66
n-C11	5	30	234	110
n-C12	25	122	604	115
n-C13	4	38	232	150
n-C14	3	29	203	99
n-C15	3	39	246	160
n-C16	3	33	177	123
n-C17	11**	67	336	204
Pr	3	27	135	99
n-C18	7	55	273	138
Ph	2	17	66	44
n-C19	15	115	612	242
n-C20	15	109	598	129
n-C21	43	366	2163	574
n-C22	35	260	1477	422
n-C23	101	826	5482	1179
n-C24	33	253	1443	395
n-C25	75	668	4312	1221
n-C26	20	158	932	300
n-C27	98	1020	6691	2053
n-C28	16	101	598	205
n-C29	74	809	5097	1529
n-C30	8	66	1086	175
n-C31	61	611	3692	1417
n-C32	3	27	126	203
n-C33	18	178	969	457
n-C34	7	33	149	34
n-C35	4	20	108	72
n-C36	1	nd	9	7
Total *n*-alkanes	725	6054	38017	11778
Σ C12–C19	70	499	2684	1230
Σ C20–C33	598	5450	34668	10259
pr/ph	1.07	1.59	2.05	2
odd/even***	1.92	2.97	3.00	4

* n = 3

** three most dominant homologs >*n*-C-14 for Elson Lagoon are underscored for each sample
*** summed from *n*-C15 to *n*-C36

Qualitatively, the thermally mature ($\alpha\beta$ hopanes) biomarker content in the samples follows the order: EL-3 > EL-2 > EL-1. Steranes are about 10× lower than triterpanes. The alkane and PAH contents in the three samples decrease in the following order: EL-3 > EL-2 > EL-1, with EL-3 containing the maximum concentrations of identifiable components.

Table 7 shows the concentrations of polycyclic aromatic hydrocarbons in Elson Lagoon sediments and sample 6G from the Colville River Delta [Naidu et al. 2001]. For Elson Lagoon the PAH composition is dominated by the homologous series of naphthalenes and phenanthrenes. With the exception of naphthalene all other parent PAHs dominate over their methylated homologs. Compared to the lower molecular weight PAHs, 4- and 5-ring PAHs are relatively less, perylene is always the most dominant PAH, and chrysene/triphenylene is the next most dominant PAH in the samples analyzed. Perylene is the most dominant PAH of all the parent PAHs in two of the three samples and second most dominant in the third. Preliminary and qualitative analysis indicates the presence of small amounts of coprostanol and coprostanone in all the samples, but no linear alkylbenzenes (LABs) were found. However, the identification of fecal sterols needs to be confirmed by GC/MS, especially in view of the large amounts of biogenic sterols in the samples, which are present at levels of 10 to 60× fecal sterols.

Comparison of the composition of the *n*-alkanes and PAHs in the Elson Lagoon sediments (Tables 6 and 7) with those from Colville Delta–Prudhoe Bay [Naidu et al. 2001] shows some minor differences. Sample EL-3 has nearly three times greater total *n*-alkanes than sample 6G from the Colville River Delta, which has a maximum level of all the samples analyzed by us in the previous CMI study encompassing the Colville Delta–Prudhoe Bay region. Similarly, sample EL-3 from Elson Lagoon has nearly twice the amount of total PAHs compared to sample 6G from the Colville Delta, which again has the maximum PAH content of all the samples analyzed from Colville Delta–Prudhoe Bay. Except for these differences, the remaining two Elson Lagoon sediments (EL-1 and EL-2) have comparable levels of total *n*-alkanes and PAHs, and a similar distribution pattern of triterpenoids (Figure 3) to the Colville Delta–Prudhoe Bay sediments.

Table 7. Polycyclic aromatic hydrocarbons (PAHs) in Elson Lagoon sediments and sample 6G from the Colville River Delta [Naidu et al. 2001].

	Sample ID			
	Elson Lagoon			**Colville**
PAH (ng g^{-1} dry)	**EL-1**	**EL-2**	**EL-3**	**6G***
naphthalene	nd	10.84	76.22	16.72
C1-naphthalenes	nd	56.08	353.71	94.54
2-methylnaphthalene	nd	28.97	186.30	51.04
1-methylnaphthalene	nd	27.16	167.74	43.63
C2-naphthalenes	nd	50.47	293.95	168.91
2,6-dimethylnaphthalene**	nd	25.94	108.73	18.19
C3-naphthalenes	nd	41.92	222.12	129.70
2,3,5-trimethylnaphthalene	nd	10.86	44.45	22.17
C4-naphthalenes	nd	11.33	58.46	52.71
biphenyl	nd	9.68	56.40	14.47
acenaphthylene	nd	2.82	16.01	nd
acenaphthene	nd	0.96	5.10	1.39
fluorene	nd	6.91	34.23	9.04
2-methylfluorene	nd	15.69	56.89	8.13
C1-fluorenes	nd	7.64	31.69	30.39
C2-fluorenes	nd	11.91	42.43	39.30
C3-fluorenes	nd	13.65	36.62	70.26
phenanthrene	2.84	39.07	205.58	50.46
1-methylphenanthrene	nd	24.50	106.35	24.44
anthracene	nd	1.21	5.25	22.96
C1-phenanthrenes/anthracenes	6.81	41.59	120.79	160.85
C2-phenanthrenes/anthracenes	nd	42.57	117.03	132.00
3,6-dimethylphenanthrene	nd	18.63	17.05	5.89
C3-phenanthrenes/anthracenes	nd	22.16	72.97	103.10
C4-phenanthrenes/anthracenes	nd	nd	nd	12.68
2,3-benzofluorene	nd	nd	62.64	8.38
1,1′-binaphthalene	nd	nd	nd	nd
dibenzothiophene#	nd	3.52	38.38	nd
C1-dibenzothiophenes#	nd	nd	6.02	2.59
C2-dibenzothiophenes#	nd	nd	nd	0.55
C3-dibenzothiophenes#	nd	nd	nd	nd
C4-dibenzothiophenes#	nd	nd	nd	nd
fluoranthene	nd	6.82	29.36	12.10
pyrene	nd	9.65	45.38	14.80
C1-fluoranthenes/pyrenes	nd	13.94	65.56	34.43
C2-fluoranthenes/pyrenes	nd	14.59	48.38	32.84
C3-fluoranthenes/pyrenes	nd	nd	24.19	13.82
C4-fluoranthenes/pyrenes	nd	nd	nd	10.53
benz(a)anthracene	nd	3.10	18.20	3.43

Table 7. continued

	Sample ID			
	Elson Lagoon			Colville
PAH (ng g^{-1} dry)	EL-1	EL-2	EL-3	6G*
chrysene/triphenylene	nd	28.27	117.18	27.19
C1-chrysenes/triphenylenes	1.02	17.46	69.37	57.81
C2-chrysenes/triphenylenes	nd	8.15	40.41	64.98
C3-chrysenes/triphenylenes	nd	nd	nd	50.02
C4-chrysenes/triphenylenes	nd	nd	nd	nd
benzo(k)fluoranthene	1.36	14.47	50.20	1.94
benzo(b)fluoranthene	0.93	nd	4.27	17.44
benzo(e)pyrene	1.88	21.00	99.52	21.78
benzo(a)pyrene	nd	3.64	22.40	4.86
9,10-diphenylanthracene	nd	nd	nd	nd
perylene	2.08	50.86	258.41	166.74
indeno(1,2,3-cd)pyrene	nd	nd	19.94	8.20
dibenz(a,h)anthracene	nd	nd	nd	5.47
picene	nd	nd	nd	23.70
benzo(ghi)perylene	nd	22.33	110.56	28.60
anthanthrene	nd	nd	nd	1.18
coronene	nd	21.81	89.89	14.45
1,2,4,5-dibenzopyrene	nd	29.03	128.35	26.87
C1-C20H12 aromatics	nd	14.37	92.86	70.27
C2-C20H12 aromatics	nd	nd	35.27	44.78
C3-C20H12 aromatics	nd	nd	nd	33.46
C4-C20H12 aromatics	nd	nd	nd	nd
sum naphthalenes (N)	nd	170.63	1004.46	480.77
sum fluorenes (F)	nd	40.11	144.98	148.98
sum phenanthrenes/anthracenes (PA)	9.65	146.59	521.62	482.05
sum dibenzothiophenes (D)	nd	3.52	44.40	3.14
sum fluoranthenes/pyrenes (FP)	nd	45.00	212.86	118.54
sum chrysenes (C)	1.02	53.88	226.95	200.19
sum C20H12 aromatics (C20)	6.26	104.33	562.94	361.13
sum 4,5 P-PAH	6.26	137.80	644.93	
sum 4,5 PAH (4,5 PAH)	7.28	191.94	892.83	534.59
sum PAH(t-PAH)	16.93	653.80	3225.30	1912.67
N/PA	nd	1.16	1.93	1.00
N/perylene	nd	3.35	3.89	2.88
F/perylene	nd	0.79	0.56	0.89
PA/perylene	4.63	2.88	2.02	2.90
FP/perylene	nd	0.88	0.82	0.70
t-PAH/perylene	8.12	12.85	12.48	11.42

nd: not detected, below MDL * n = 3 ** coelutes with another isomer

\# very low recovery because of activated cu treatment for sulfur removal

% recovery of some methylated homologs assumed to be the same as that of methylated phenanthrenes

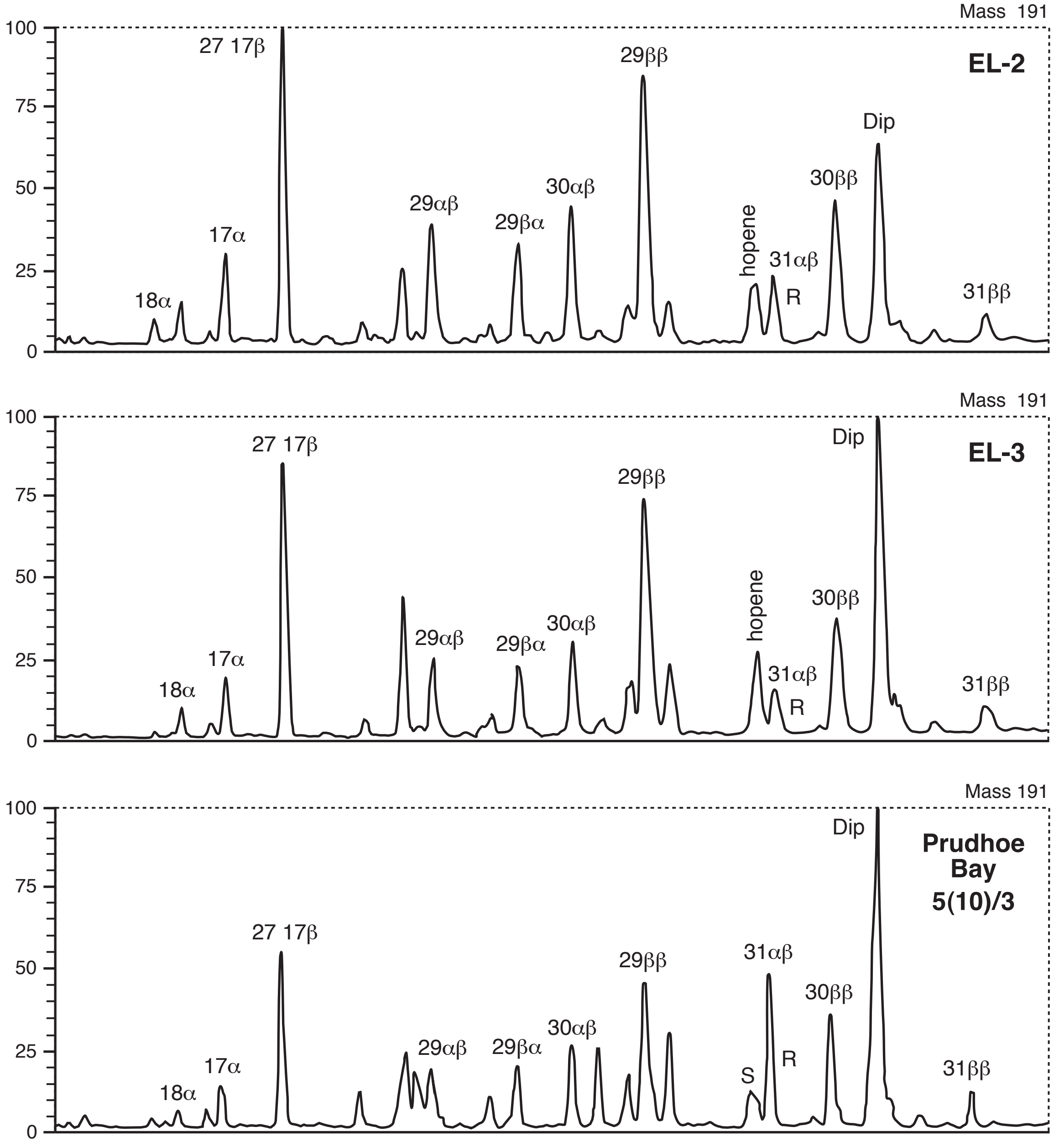

Figure 4. Typical mass chromatogram from GC/MS analysis of triterpenoid distribution in sediments from Elson Lagoon (EL-2 and EL-3) and Prudhoe Bay.

Discussion

Trace metal studies

The mean concentrations of all metals in the Elson Lagoon muds (Table 3) are at or below those from unpolluted argillaceous marine sediments of the world (including the Arctic Ocean), natural baseline levels of regions now presumed polluted (based on data in core sections representing the pre-industrial age), and mean concentrations in shale which represent argillaceous rocks with natural metal values [Förstner and Wittman 1979 and references therein; Bruland et al. 1974; Salomons and Förstner 1984; Naidu et al. 2001]. However, regression analysis of the stratigraphy of the metals in core EL-3, in conjunction with the geochronology of the core, indicates significant ($p < 0.05$) net increases in concentrations of Ba, Cr and Mn in Elson Lagoon during the past ~70 years (Table 3, Figure 3). In contrast, a significant up core net decrease in THg is found, but no statistically significant (at $p < 0.05$) stratigraphic changes in concentrations of other metals, including MeHg, are detected. It should be noted that in the regression analysis, to assess net stratigraphic trends in the metal variations, data from the disjointed basal section of core EL-3 were also taken into account. It could be argued that the regression analysis incorporating data from the basal (18–19 cm) core section in the absence of data from the 10–18 cm core section would give extra emphasis to the basal section and, therefore, the analysis may be invalid. However, we contend that by including data from the basal section the net stratigraphic trends provide a better means to assess long-term temporal changes in the context of the presumed baseline provided by the basal core section. It would have been, of course, desirable to include metal data from the entire core, but limited funds precluded this. Taking the above into account, the up core increase in Ba, Cr and Mn may either suggest that the lagoon is getting contaminated in the above metals, or the trend may be a manifestation of up core stratigraphic changes in lithology and/or organic matter in the mud fraction, which generally control the abundance of particle-reactive metals, such as the ones we have analyzed, by processes of adsorption and/or organic chelation. Alternatively, the up core increase in Cr and Mn could be due to diagenetic upward mobilization in solution from subsurface anoxic layers and subsequent co-precipitation of the metals in oxic surficial sediments. Because of limited funds we were not able to verify the possible control of lithology and diagenesis on the stratigraphic changes in the metal concentrations. If the reason is recent contamination, we do not then have knowledge of the possible sources for the metals. It is to be noted that Elson Lagoon and its vicinity are a popular place for recreation (snow machining, fishing, hunting, boating, camping) for Barrow residents, and in the past the lagoon has been a site for municipal garbage disposal (including machine parts), and accumulation of debris from rocket firing by the U.S. Navy. The region has also been impacted by oil and gas exploration. It is quite possible that any or all of these activities could have contributed to contaminant input. We recommend that cautionary measures, such as avoidance of litter disposal into the lagoon, must be enforced to prevent any further increase in metal concentration in the environment.

The down core net increase in THg presumably reflects higher methylation of Hg in subsurface anoxic sediment layers, as suggested by the presence of increasing levels of MeHg from the upper 4 cm composite of the core (representing possibly oxic layers) to about 7 cm down the core (Figure 2, Table 3). Results of regression analysis of the stratigraphic variations in MeHg values, including data from the upper 7 cm core, indicate that the regression is significant at the 5% level

(P = 0.0069). It is interesting to note that a similar down core increase in MeHg was also reported for the Colville Delta–Prudhoe Bay area [Naidu et al. 2001], and also suggested that the down core increase in MeHg was likely due to greater methylation of Hg in subsurface anoxic sediments. Further detailed investigations will be necessary to better understand the processes of methylation in sediments from Elson Lagoon and Colville Delta–Prudhoe Bay.

One of our stated objectives in this study was to compare the mean concentrations of trace metals and hydrocarbons in sediments of Elson Lagoon and those of the Colville Delta–Prudhoe Bay region, with a motive to identify differences in the nature of and concentrations of contaminants in sediments derived from urban-impacted and petroleum-related industrial activities. Elson Lagoon sediments have been taken as a representative of the former activities, whereas the Colville Delta–Prudhoe Bay sediments have been considered to represent deposits influenced by petroleum-related industrial activities. As indicated (Table 4), there are significantly higher ($p < 0.05$) mean concentrations of Cr, Ni, V and THg in mud samples of the urban-impacted Elson Lagoon than in muds of the nearshore of the petroleum-related industrial area of Colville Delta–Prudhoe Bay region [Naidu et al. 2001]. The reason for these differences is unknown. Some of the possible factors contributing to this disparity are relatively higher input of metals into Elson Lagoon from confined natural terrestrial sources from the hinterland, anthropogenic local contributions, and/or regional differences in mass sediment accumulation rates (MSAR). The MSAR in the Colville Delta–Prudhoe Bay region is 0.77 g cm^{-2} yr^{-1} [Naidu et al. 2001] as opposed to 0.34 g cm^{-2} yr^{-1} in Elson Lagoon. The higher rate would tend to dilute the concentrations of metals in sediments and, thus, could account for differences in the metal levels in the above two regions. Alternatively, a relatively higher atmospheric deposition of anthropogenic metals (metals derived from human or human-related activities) into Elson Lagoon than the Colville Delta–Prudhoe Bay, via long-distance transport from Eurasia [Barrie et al. 1992; Rahn and Lowenthal 1986], may result in regional differences in sediment metal concentrations. The Eurasian region of China and Russia (Siberia) is highly industrialized and populated, and the environment there is widely contaminated with a variety of organic and inorganic chemicals [AMAP 1997]. The above premise, relating to regional differences in metal input from long distance transport, is supported by the presence generally of a west to east concentration gradient of metals in snow along the North Slope coast of Alaska. It is shown that relatively higher concentrations of Cr, Ni, V and THg and several other metals, as well as a few anthropogenic organics, exist in freshly precipitated snow from Elson Lagoon in the west through the Colville Delta–Prudhoe Bay region to Demarcation Bay in the east [Snyder-Conn et al. 1997; Garbarino et al. 2002]. The above authors have shown that snow is a significant medium in the Alaskan Arctic for sequestering and entraining atmospheric-borne metals, with subsequent co-precipitation in the lagoon. As there is no marked difference in the annual depositional rate of precipitation between Elson Lagoon (snow: 74.17 cm yr^{-1}; rain: 11.40 cm yr^{-1}) and Colville Delta–Prudhoe Bay (snow: 82.55 cm yr^{-1}; rain: 9.12 cm yr^{-1}) (Theodore Fathauer, National Weather Service, pers. comm.), it is possible that over the years higher metal fluxes tied to the depositional snow result in greater concentrations of metals in the muds of Elson Lagoon. Consistent with the above premise is the contaminant transport model developed by Levno-Chythlook et al. [1999], which has implications for the trans-boundary transport pathway of airborne contaminants from Eurasia to northern Alaska. The model suggests that there will be a focusing of air-borne contaminants at Elson Lagoon compared to other coastal regions in the east of the Alaskan North Slope.

In this study the use of the correlation coefficient analysis (Table 5), to get insight into geochemical partitioning of the metals, appears tenuous, at best. It would seem that in Elson Lagoon the amounts of the clay fraction in the muds have a relatively minor role in the sequestering of metals, as suggested by a lack of positive correlation between all of the metals and clay percent (prorated for mud based on gross sediment data in Table 1). This finding is inconsistent with that for mud samples from the Colville Delta–Prudhoe Bay region, where strong positive correlations between several metals and clay percent are noted [Naidu et al. 2001]. Almost invariably, clay is a major sequester of particle-reactive metals (such as the ones we have investigated in this study), by adsorption and/or metal–clay–organic complex formation. Perhaps the oddity in the case of Elson Lagoon can be explained as an artifact imposed by the limited number of samples (n = 5) that were analyzed. In contrast, 62 samples were analyzed from the Colville Delta–Prudhoe Bay area [Naidu et al. 2001].

We have evaluated the mean concentrations of selected heavy metals in Elson Lagoon sediments to determine their potential for adverse effects on resident benthic and demersal organisms, following the guidelines proposed by Long et al. [1995]. Long et al., using a broad database consisting of equilibrium-partitioning modeling, laboratory bioassays and field studies, have developed "two guideline values (an effects range–low, and an effects range–median)". Further, "the two values defined concentration ranges that were: (1) rarely, (2) occasionally, or (3) frequently associated with adverse effects". Comparison of the mean concentrations of heavy metals from our study with the above ERL and ERM and the three ranges for the selected metals listed in Long et al. 1995 (Table 8), indicates that the mean concentrations of As, Cr, Cu and Ni in Elson Lagoon muds fall into the ERL−ERM category, whereas mean concentrations of Cd, Pb, Hg, and Zn are below ERL. This implies that the concentrations of As, Cr, Cu and Ni in Elson Lagoon sediments are at or above the threshold level where adverse effects on biota would occasionally occur. However, whether the extrapolation of the guidelines suggested by Long et al. is relevant or applicable to Elson Lagoon is a question that needs to be further explored with detailed site-specific cause & effect investigations on resident organisms. Nevertheless, the above assessments suggest that measures must be taken to minimize the anthropogenic input of the above four metals of concern.

Table 8. Comparison of the mean concentrations of selected trace metals in Elson Lagoon sediments with ERL and ERM guideline values, and percent incidence of biological effects in concentration ranges defined by the two values as reported by Long et al. [1995].

	THIS STUDY	DATA FROM LONG ET AL. [1995]				
		Guidelines		Percent (ratios) incidence of effects*		
	Elson Lagoon ng g^{-1} dry wt	ERL ppb dry wt	ERM	<ERL	ERL–ERM	>ERM
As	21	8.2	70	5.0 (2/40)	11.1 (8/73)	63.0 (17/27)
Cd	0.17	1.2	9.6	6.6 (7/106)	36.6 (32/87)	65.7 (44/67)
Cr	121	81	370	2.9 (3/102)	21.1 (15/71)	95.0 (19/20)
Cu	34	34	270	9.4 (6/64)	29.1 (32/110)	83.7 (36/43)
Pb	19	46.7	218	8.0 (7/87)	35.8 (29/81)	90.2 (37/41)
Ni	50	20.9	51.6	1.9 (1/54)	16.7 (8/48)	16.9 (10/59)
Zn	129	150	410	6.1 (6/99)	47.0 (31/66)	69.8 (37/53)
THg	0.07	0.15	0.71	8.3 (4/48)	23.5 (16/68)	42.3 (22/52)

* Number of data entries within each concentration range in which biological effects were observed divided by the total number of entries within each range.

Hydrocarbon studies

The alkane and PAH contents in the three gross sediment samples decrease in the order EL-3 > EL-2 > EL-1 (Tables 6 and 7). This pattern of decrease is consistent with the sediment granulometry, reflected in the content of mud (<63 µm size), which is EL-3 > EL-2 > EL-1 (Table 1). The mud fraction is the most reactive fraction of the total sediment for hydrocarbon adsorption and/or organic–inorganic complex formation.

The composition of hydrocarbons documented in our study provides clues to their source, biogenic or petrogenic, as enumerated below.

Alkanes: The gas chromatogram is trimodal with maxima at *n*-C17, *n*-C23 and *n*-C27, and alkanes >*n*-C25 predominate in the samples. The *n*-C17 is characteristic of aquatic algae and the *n*-C23 maximum in the unresolved complex mixture of alkanes (which is illustrated as a broad hump in the GC chromatogram) reflect microbial degradation. Normal alkanes dominant in odd carbon alkanes ≥ *n*-C25 are derived from higher terrestrial plants. This is also documented by the odd/even ratio, which is much greater than one. The *n*-C20 and *n*-C21 olefins identified are derived from plankton and bacteria.

Triterpanes: These are mostly biogenic, and 27(17β), 29ββ, 30ββ and diploptene are the four major components (Figure 4). The former two hopanes are the most dominant in EL-1 and EL-2 and diploptene in EL-3. Qualitatively, thermally mature (αβ hopanes) biomarker content in the samples follows the order: EL-3 > EL-2 > EL-1 and these hopanes are most probably derived from peat and/or coal.

Steranes: These are about 10× lower than triterpanes. Their profile is very different from that of oil and most probably reflects diagenetic origin.

PAHs: As mentioned, PAH composition (Table 7) is dominated by the homologous series of naphthalenes and phenanthrenes. With the exception of naphthalene, all other PAHs dominate over their methylated homologs. This suggests the general absence of significant petroleum input in the sediments. Compared to the lower molecular weight PAHs, 4- and 5-ring PAHs are relatively less, perylene is always the most dominant PAH, and chrysene/triphenylene is the next most dominant PAH in this suite. Perylene is the most dominant in the EL-3 sample, consistent with its origin in Alaskan peats similar to that found in coastal sediments of Alaska.

The qualitative analysis on sewage organic tracers in PAH and sterol fractions has allowed some tentative deductions. Linear alkylbenzenes (LABs), which are sewage tracers derived from household laundry, were not detected upon examination of the qualitative analysis of the GC/MS run of the PAH fractions in any of the three sediment samples. However, coprostanol, a sewage sterol, and coprostanone were found in small amounts in all the three samples. Their identification needs to be confirmed by GC/MS, especially in view of the large amounts of biogenic sterols in the samples, which are present at levels of 10 to 60× fecal sterols. A detailed investigation of these tracers was out of the scope of the CMI-funded project. Nonetheless, the very low relative proportions of fecal sterols would indicate that sewage input in the sediments, if at all present, may not be important.

Among a selected group of the PAHs considered, in the context of Long et al.'s [1995] guidelines for identifying hydrocarbon pollution in terms of effect range–low and effect range–median (refer to the related discussion above on trace metals), the mean concentration of 2-methylnaphthalene and the EL-3 total PAH are between the ERL and ERM, indicating that they are at or above the threshold level where adverse effects on biota would occasionally occur (Table 9). As suggested for the trace metals, this conclusion must be clarified by site-specific studies on resident organisms.

Therefore, the normal and cyclic alkane distribution is characteristic of biogenic origin in all of the three samples and very little petroleum and sewage input is reflected in their composition. Additionally, the overall profiles of alkanes and PAHs are similar to those of inner shelf sediments from the Beaufort Sea [Naidu et al. 2001 and Figure 2 for comparison with Figure 4 in the present report].

Table 9. Comparison of the mean concentrations of selected hydrocarbons in Elson Lagoon sediments with ERL and ERM guideline values◊, and percent incidence of biological effects in concentration ranges defined by the two values as reported by Long et al. [1995].

	THIS STUDY	DATA FROM LONG ET AL. [1995]				
		Guidelines		Percent (ratios) incidence of effects*		
	Elson Lagoon ng g^{-1} dry wt	ERL ppb dry wt	ERM	<ERL	ERL–ERM	>ERM
naphthalene	29.0	160	2100	16.0 (4/25)	41.0 (16/39)	88.9 (24/27)
2-methylnaphthalene◊	71.8	70	670	12.5 (2/16)	73.3 (11/15)	100 (15/15)
acenaphthylene	6.3	44	640	14.3 (1/7)	17.9 (5/28)	100 (9/9)
acenaphthene	2.0	16	500	20.0 (3/15)	32.4 (11/34)	84.2 (16/19)
fluorene	13.7	19	540	27.3 (3/11)	36.5 (19/52)	86.7 (26/30)
phenanthrene	82.5	240	1500	18.5 (5/27)	46.2 (18/39)	90.3 (28/31)
anthracene	2.2	85.3	1100	25.0 (4/16)	44.2 (19/43)	85.2 (23/27)
fluoranthene	12.1	600	5100	20.6 (7/34)	63.6 (28/44)	92.3 (36/39)
pyrene	18.3	665	2600	17.2 (5/29)	53.1 (17/32)	87.5 (28/32)
benz(a)anthracene	7.1	261	1600	21.1 (4/19)	43.8 (14/32)	92.6 (25/27)
chrysene/triphenylene#	48.5	384	2800	19.0 (4/21)	45.0 (18/40)	88.5 (23/26)
benzo(a)pyrene	8.7	430	1600	10.3 (3/29)	63.0 (17/27)	80.0 (24/30)
dibenz(a,h)anthracene	nd	63.4	260	11.5 (3/26)	54.5 (12/22)	66.7 (16/24)
Sum PAH◊	1298.7	4022	44792	14.3 (3/21)	36.1 (13/36)	85.0 (17/20)

◊ 2-methylnaphthalene has the only mean concentration between ERL and ERM. The EL-3 total PAH and 2-methylnaphthalene (3225.3 and 76.2 ng g^{-1}, respectively) are between ERL and ERM. Low and high molecular weight PAHs are not defined in Long et al., and are, therefore, not computed here. Total PAH may be different for the same reason.

* Number of data entries within each concentration range in which biological effects were observed divided by the total number of entries within each range.

Reported as chrysene in the Long et al. study.

nd = not detected

Conclusions

The analyses from Elson Lagoon of eleven trace metals in mud fractions of five grab sediment samples and stratigraphic sections of a core, and total mercury and methylmercury in gross grab sediment samples show mean concentrations that are generally similar or below levels that are reported for unpolluted marine nearshore sediments and shales. By comparison, the mean concentrations of Cr, Ni and V in mud and of THg in gross sediments of Elson Lagoon are significantly higher than in similar deposits from the Colville Delta–Prudhoe Bay region. These differences may be due either to relatively higher natural or anthropogenic metal input to the lagoon, or to greater depositional flux of anthropogenic metals that are associated with snow and derived from Eurasia via long-distance atmospheric transport. This is consistent with the general west to east decrease in the concentrations and fluxes of Cr, Ni and V and several other metals in coastal snow as reported in the literature for the North Slope of Alaska. Stratigraphic variations in metals indicate significant increases up core in Ba, Cr and Mn during the past ~70 years, which suggest minor contamination recently in the above metals. In contrast, a net increase down core is noted in THg, which may be related to greater methylation of Hg in progressively intense anoxic sediment layers down the core. The mean concentrations of eight metals in Elson Lagoon muds were compared with those examined in Long et al.'s [1995] analysis of guideline values for the incidence of adverse biological effects. The comparison indicates that the concentrations of As, Cr, Cu and Ni in Elson Lagoon sediments are at levels where adverse effects on biota would occasionally occur, whereas the concentrations of Cd, Pb, Hg and Zn are below such levels. However, we suggest that the conclusions, based on the extrapolation of the Elson Lagoon data in context of Long et al.'s [1995] guidelines, must be verified by detailed site-specific investigations involving cause & effect studies on resident organisms.

The hydrocarbon concentrations in gross sediments are also relatively low and are comparable to those found in our previous investigation on coastal sediments from the Colville Delta–Prudhoe Bay region and by others from the Beaufort Sea nearshore. The origin of the hydrocarbons is essentially biogenic, with no significant input from natural or anthropogenic petroleum products. Among the PAHs examined, the concentration of 2-methylnaphthelene in one sample is between Long et al.'s [1995] effective range–low and effective range–median guidelines, with an expectation of occasional adverse effects on resident organisms.

In summary, Elson Lagoon, like the Colville Delta–Prudhoe Bay nearshore, has remained a relatively clean environment despite the slight increase recently in some metal levels subsequent to accelerated anthropogenic activities there during the past 30 years. The data from this investigation should serve as a baseline for monitoring chemical contaminants in Elson Lagoon.

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Acronyms and Abbreviations

AFS	atomic fluorescence spectrometry
AMAP	Arctic Monitoring and Assessment Programme
AMS	accelerator mass spectrometry
BASC	Barrow Arctic Science Consortium
CVAAS	cold-vapor atomic absorption spectrometry
CVAF	cold-vapor fluorescence spectrometry
ERL	effect range–low
ERM	effect range–median
EPA	Environmental Protection Agency
FRSGC	Frontier Research System for Global Change
GC	gas chromatograph
GC/MS	gas chromatograph/mass spectrometer
GF-AAS	graphite furnace atomic absorption spectrometry
IARC	International Arctic Research Center
ICP-MS	inductively coupled plasma mass spectrometer
LAB	linear alkylbenzene
MDL	method detection limit
MeHg	methyl mercury
MSAR	mass sediment accumulation rate
NIST	National Institute of Standards and Technology
NOAA	National Oceanic and Atmospheric Administration
NRCC	National Research Council of Canada
NRCC DORM-2	dogfish muscle tissue (a standard reference material)
NS&T	National Status and Trends Program (NOAA)
ORCA	Office of Ocean Resources Conservation and Assessment (NOAA)
PAH	polycyclic aromatic hydrocarbon
QA/QC	quality assurance/quality control
THg	total mercury

Study Products

The concentrations and depositional fluxes of the trace metals and hydrocarbons are available in readily accessible electronic format through the Minerals Management Service. Please refer to the tables attached showing results of the statistical analysis, station-wise specific sediment trace metal and hydrocarbon data, which are also available in computer-aided graphic or tabulated form.

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