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Contaminants at Arctic Formerly Used Defense Sites

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Key Words: Formerly Used Defense Sites (FUDS); Arctic Contaminants; Remediation; Global Transport; Persistent Organic pollutants (POPs); Contaminant Redistribution.

Abstract

This study was conducted in order to determine if the source of contaminants at formerly used defense sites (FUDS) in Alaska were deposited as a result of military occupancy or from long-distance transport. This determination largely influences whether remediation will occur, and, if so, to what extent. For this reason, plant samples (rinsed and unrinsed) and sediment cores were collected at military and remote sites on St. Lawrence Island (SLI) and Norton Sound, Alaska during the summers of 2002, 2006, and 2007 and analyzed for persistent organic pollutants. On St. Lawrence Island sediment core samples were collected at the Northeast Cape FUDS, also a traditional fishing/hunting camp, and were sectioned and analyzed for concentrations of Polychlorinated biphenyl (PCB) congeners, Mirex, Dichlorodiphenyldichloroethylene (DDE), Hexachlorobenzene (HCB), Mercury and Cesium-137 (^{137}Cs). Differences in the total concentrations and distributions of PCB congeners, Mirex, DDE and Mercury in sediment cores and in plants collected from the two SLI and three Norton Sound mainland formerly used defense sites indicate the majority of the contaminants found can be temporally related to releases during military occupancy and subsequent redistribution of contaminants. Contaminants in plant samples at the SLI sites are elevated relative to the two remote sites located on St. Lawrence Island and the three mainland Norton Sound FUDS at Elim, Unalakleet, and Wales. The concentrations, lateral and vertical distribution of the total PCBs, and congener-specific differences in sediments and plants readily differentiate locally derived from globally transported contaminants. The relative contaminant concentrations in sediment cores and between rinsed and unrinsed plants collected from the NEC FUDS indicate contaminants were remobilized and redistributed during recent site remediation activities.

Key Words: Formerly Used Defense Sites; FUDS; Arctic Contaminants; Remediation, Global Transport, Persistent Organic Pollutants, (POPs), Contaminant Redistribution.

Introduction

Formerly used defense sites (FUDS) include properties previously owned, leased, possessed or operated by the U. S. Department of Defense (DoD) in the states and six U.S. territories. The majority of the sites were created during the Cold War era. The larger bases required the development and maintenance of extensive infrastructure and often, complete communities were created and maintained including living quarters, roads, runways, communication equipment, surveillance, electrical generation, fuel storage, recreation, airfields and solid and human waste management facilities.

There are approximately 600 FUDS in Alaska, many in proximity to Native communities and traditional fishing and hunting grounds. Chlorinated and non-chlorinated solvents, herbicides/pesticides, trace metals, containers of human wastes, chemical warfare agents, unexploded ordnance (UXO) and other toxic materials were not only used during active site operations, but were left behind when the posts were abandoned. Because of the remote locations of many of the Alaskan FUDS, the DoD policy on remediation has relied heavily on natural attenuation, long term monitoring and institutional controls (ICs) including fences and signs to minimize or prevent humans and animals from coming in contact with contaminants or explosive materials. Because many of the Alaska FUDS are located in remote areas, their use and therefore remedial standards and objectives are often less stringent than sites located in more populated regions.

Global Transport and Accumulation of Contaminants

The Arctic is recognized as a hemispheric sink for persistent organic pollutants (POPs) due to air and oceanic currents that redistribute a broad range of contaminants from the industrialized and agricultural regions of the lower latitudes. This flux and accumulation is enhanced by the physical and biological characteristics of the Arctic that favor the accumulation and retention of POPs (Wania and Mackay, 1993 a and b). Dated lake sediment cores collected from six, mid-continent, Canada lakes contained total PCB concentrations ranging from non-detect to a maximum of about 0.75 ug/kg. The same six lakes had maximum hexachlorobenzene (HCB) concentrations of about 1.5 ug/kg in the post 1970 dated core sections (Muir, et al, 1996; Lockhart, 1994).

In sub-arctic lakes, the onset of PCB introductions corresponded to the 1940s whereas arctic lakes were not impacted until the 1960s (Jensen, et al, 1997). Total PCB concentrations in four Yukon lakes ranged from about 3 ug/kg to about 30 ug/kg in core sections dated from 1941 to 1992 with the maximum concentrations in the more recent, near surface, sediments (Muir et al., 1994b).

During the period 1955-1988, 63 Distant Early Warning (DEW Line) sites were constructed and operated across the northern regions of Canada and Alaska. The DEW Line was designed to serve as a warning system against aircraft or missile attacks. During the time these facilities were in operation, it is estimated that more than 30 tons of PCBs were shipped to the sites. More than 3500 soil and 1600 plant samples were collected in proximity to recently operated and older, abandoned sites (Reimer, et al, 1991, 1993a, 1993b, 1993c, 1994; Dodd and Reimer, 1992, Dushenko and Reimer, 1994; Grundy, et al. 1994). Significant differences in PCB concentrations in the DEW Line stations relative to remote areas

have also been documented. Soils sampled from the east coast DEW Line regions had total PCB concentrations to 320 000 ug/kg (Jensen, et al, eds., 1997). Soil PCB concentrations at main DEW Line sites ranged up to 1 000 ug/kg whereas remote site soils collected at locations a minimum of 20 km from a DEW Line facility contained PCB concentrations of about 1.0 ug/kg (Fletcher, 1990).

A variety of plants are collected, preserved and eaten by Norton Sound Alaskans and in order to assess the role played by plants in accumulating FUDS-related contaminants, samples were collected at Gambell, the NEC on SLI and the three mainland Norton Sound FUDS communities. Plants are known to accumulate a range of organic and inorganic contaminants. Blankenship, et al. (2005) assessed the accumulation of PCBs in the terrestrial plants and animals at a Superfund site in Michigan where soils contained concentrations of 6.5 mg/kg dry weight, lower chlorinated congeners were selectively accumulated by the terrestrial plants. Zeeb, et al; (2006) conducted controlled greenhouse studies to determine the potential for phytoremediation of PCBs (Aroclor 1260) and found relationships to concentrations of PCB in soils and relative uptake by roots and shoots in eight plant species.

Norton Sound FUDS

The FUDS sites at Gambell, Elim, Wales and Unalakleet were established and operated at or near existing Alaska Native communities. The NEC FUDs was constructed at the site of an important traditional fishing and hunting camp for the Yupik people of St. Lawrence Island. The military hired Savoonga residents during the construction and operational phase of the NEC FUDS. Remediation contractors also employed Savoonga residents during the site characterization and removal of contaminants during the last approximate 40 years.

The Study Areas

St. Lawrence Island is located in the Bering Sea about 215 kilometers west, southwest of Nome, Alaska (Figure 1). The island is about 200 kilometers long and 50-65 kilometers wide and home to two Siberian Yup'ik communities of 600-700 inhabitants. Both St Lawrence Island communities rely heavily on traditional foods including whale, walrus, seal, fish and a range of plants and berries.

The community of Gambell is located on a large cobble spit at the northwest end of St. Lawrence Island, about 65 kilometers from the Siberian mainland (Figure 2). The community was an involuntary host to a military installation that operated for about twenty years beginning in the early 1950s. Plant sampling was conducted at Gambell during the July 2002 and 2006 field seasons. Sediment core sampling was not conducted at Gambell due to the nature of the cobble substrate. The Gambell community and the location of the former military base are situated on a cobble beach complex and upland bedrock on the shores of the Bering Sea. Gambell residents served as volunteer participants in Cold War activities including area surveillance and reporting of Russian movements and activities.

Savoonga was not occupied by the military, although the facility established at the NEC was used primarily by Savoonga residents as a hunting/fishing camp and residents also worked at the FUDS during

the time it was operational and have participated with the more recent site characterization and remedial activities.

The NEC FUDS is located about 90 kilometers south-southeast of Savoonga (Figure 2). The Air Force acquired the strategically located NEC site in 1952 and operated the facility as a surveillance station as part of the Cold War North American Air Defense Command from 1952-1972. Beginning in 1982, the Navy used the area as a White Alice communications site. Contamination at the NEC derived from large volume fuel spills, releases of solvents, trace metals, asbestos, hexachlorobenzene (HCB), DDE, mirex and PCBs are contaminants of concern in the surface and groundwater, soils, sediments and plants.

Geologically and physiologically, the NEC and Gambell differ significantly. Gambell is situated on a cobble beach whereas the NEC transitions from igneous highlands to the south of the former base to tundra lowlands bordering the Bering Sea.

The primary surface drainage at the NEC is the Suqitughneq River (Suqi), a small stream that originates in the highlands bordering the NEC to the south. North of the uplands, the river is incised in the tundra soils and ranges in width from about 0.5-3 meters and reaches a maximum depth of about one meter immediately north (downgradient) of the Main Operations Complex (MOC), which served as the center of the bases' activities. The river discharges to the two kilometer long estuary located about two kilometers south of the MOC. A cobble and sand beach at the mouth of the estuary intermittently restricts open flow to the Bering Sea (Figure 3). The river and estuary were favored fishing grounds by Savoonga residents prior to the military occupancy.

Thirty-four individual sites at the NEC FUDS have been distinguished primarily on former uses or differentiated ecosystems. The Suqi River originates from the Kinipaghulghat Mountain, flows north through the MOC and joins the eastern branch of the Suqi River. The MOC area included living quarters, electrical generating facilities, fuel storage, supply and maintenance centers, and other support structures. The upland regions of the NEC FUDS housed radar and observation facilities that were connected to the lowland area by a cable tramway. The NEC FUDS also included a White Alice communications center located just south and up gradient of the MOC.

The Main Drainage of the Suqi River is to the Suqi Estuary that connects to the Bering Sea, approximately two kilometers south of the MOC. Supplies and personnel accessed the NEC by air at a runway constructed north of the MOC and barges that off loaded on the beach landing. Near the mouth of the Suqi River, the flow measured in August 2002 was about 0.3 cubic meters per second. The Suqi Estuary is a long shallow body of water located down gradient of the MOC drainage. Depending on flow conditions and season, the estuary is partially cut off from the Bering Sea by a cobble beach.

Three sediment core sampling locations at the NEC (Figure 3) were selected in an attempt to gain information on the relative impact resulting from the use of the area by the military during the 1950s-1980s. Sediment cores were collected from within the Suqi drainage immediately down gradient of the MOC, from the Suqi Estuary and from a site located adjacent to a waste disposal site (Site 7, Figure 3) created by the military for disposal of refuse and waste materials including drums of petroleum products

including PCB-contaminated materials. The MOC drainage received runoff from the upland areas of the Northeast Cape including the White Alice communication facility and a tramway leading to a radar/communications complex located on top of the Kinipaghulghat Mountain.

Three Suqi Estuary core samples were collected about 1.0 kilometer north of the mouth of the Suqi River, midway along the length of the estuary. During the summer period, it is likely the mouth of the estuary is separated from the Bering Sea by a cobble beach. Formation of the cobble beach is subject to flow conditions of the Suqi River and estuary and periodically it opens to the Bering Sea. Three core samples were collected at a water depth of about one meter and all within about a meter from one another. The three estuary cores were collected near one another because it was the only location that was deep enough to collect sediment cores. The floor of the estuary was composed of medium to large boulders with finer-grained sediments between the boulders. Three two meter long, 6.3 cm diameter CAB tube samplers were driven to a depth of about 55 cm at the estuary sampling site. One of the three cores was sectioned and analyzed for congener-specific PCBs, mirex, HCB and DDE. Cesium-137 activity analysis was conducted on a second sectioned core, and trace metal analysis on the third.

SLI Remote sites. In order to determine the concentrations of PCBs in areas historically distant from military activity and/or human uses, sediment cores were collected during the 2006 field season from two remote locations on SLI near Savoonga including a dormant volcano (Atuk) lake located about 20 kilometers south, southeast of Savoonga and a Bering Sea lagoon site (Collier Lagoon) located 15 kilometers west of Savoonga (Figure 2). Sediment cores collected from the remote sites near Savoonga extended to a depth of about 15 cm and utilized the same coring and analytical procedures used for the NEC sediment cores.

Plant Samples. Edible plant samples were collected from five Alaskan mainland, Norton Sound communities. The Gambell and NEC plant samples were collected during the 2002 field season. Similar plant samples were collected from the three Norton Sound FUDS communities during the 2006 and 2007 field seasons. The SLI plant samples collected during 2002 were analyzed “as collected” and after being thoroughly rinsed with distilled/de-ionized water. All other plant samples were analyzed without rinsing and denoted un-rinsed.

Analytical Procedures. The Institute for Health & Environment, at the University at Albany (SUNY), a New York State certified PCB laboratory, analyzed the core sections and plants for PCBs, HCB, mirex and DDE. The Department of Earth and Atmospheric Sciences at the University at Albany analyzed the core sections for select trace metals and the Skidaway Institute of Oceanography conducted the ^{137}Cs analysis of the estuary core sections.

Congener-specific PCBs, Hexachlorobenzene, DDE and Mirex. Cores were received frozen at the Albany laboratory. Two and one-half centimeter core sections were extracted by adding 10 ml of hexane and sonicated using a micro-tip sonic disruptor horn and then extracted twice more with the extracts being pooled. The pooled extracts were concentrated to ~1 ml using a Labonco N-Vap. The concentrated extract was cleaned up using a 4% deactivated florisil (10mm X 300mm) eluted with 50 ml of hexane.

The eluent was concentrated to 1.0 ml, followed by addition of internal standard (CB 104) prior to analysis. Analysis was performed by dual-column gas chromatography (GC) with electron capture detection (ECD) on a Hewlett-Packard 6890 gas chromatograph with dual micro-ECDs and independent auto samplers. High-resolution, congener-specific analysis of 83 individual PCB congeners, 18 congeners as pairs or triplets, hexachlorobenzene, 4, 4'-DDE and mirex were quantified following the same procedures. Separation was accomplished using a J&W 30m DB-5 0.25mm diameter X 0.25µm film thickness and a RESTEK 30m Apiezon-L 0.20mm X 0.15µm. The total PCB reported is the sum of the individual congeners. Prior to the sampling and analysis conducted as a part of this study, the COE was unaware DDE, mirex and HCB were present in the sediments and plants within the Norton Sound region.

Mercury. Total mercury was analyzed by Cold Vapor Atomic Absorption Spectroscopy (CVAAS) on a Leeman Labs Hydra® atomic absorption automated mercury analyzer following EPA method 245.1. Samples were dried at 50°C overnight and hand crushed in an agate mortar and pestle. Calibration standards (0, 1, 5, and 20 ppb Hg solutions) and 0.1 g (dry wt.) sample aliquots were digested in aqua regia and $KMnO_4$. Excess $KMnO_4$ was reduced with $NH_4OH{\cdot}HCl$-NaCl. Dissolved Hg was reduced to Hg vapor with $SnCl_2$ and absorption was measured at 286 nm. Duplicate and laboratory fortified matrix (spike) samples were measured to estimate precision and analyte recovery, respectively. Relative precision ranged from 1.0-15%. Analyte recoveries range from 98-109%. NIST Standard Reference Materials 2709 (San Joaquin Soil) and 2711 (Montana Soil) were measured to assess method accuracy.

^{137}Cs. The activity of ^{137}Cs was determined following the technique of Alexander, et al. (1991). Sediments were dried, ground by hand with a ceramic mortar and pestle and sealed in 30 ml polypropylene jars. Activities were determined by quantifying the 661.6 KeV peak in each sample using an ORTEC LO-AX gamma detector and ORTEC Maestro software. The detector was efficiency calibrated using the NBS Columbia River Standard. ^{137}Cs is an impulse tracer with a peak input in 1963 and first appearance in 1954.

Discussion

The total PCB concentrations in the NEC estuary core sediments ranged from 3 µg/kg to 31 µg/kg, with the highest concentrations in the upper portions of the core extending from the surface to eight to nine centimeters (Figure 4). Below nine centimeters, the total PCB concentrations in the estuary core are less than 5 µg/kg. The PCB chlorine to biphenyl ratios increased with depth and was greater than 5.3 in the Suqi Estuary. It has been well documented that the more chlorinated congeners are less volatile and soluble (Chiarenzelli, et al, 1998, 2001) and are therefore less mobile.

The PCB concentrations in the sediments within the upper nine centimeters in the core collected immediately down gradient of the MOC were greater than 550 ug/kg (Figure 4). At depths below nine centimeters, the total PCB concentrations in the MOC core decrease rapidly. Although less concentrated, the landfill sediment core PCB concentration vertical profile followed a trend similar to the MOC core.

The ^{137}Cs activities are superimposed on the Suqi Estuary PCB sediment data in Figure 4. The ^{137}Cs profile in the estuary core indicates sediments deposited within the upper six to seven centimeters correspond with the time of military occupancy. These data indicate the PCB concentrations in the upper seven centimeters of the estuary core were deposited since about 1954 indicating an increase in the PCB concentrations in the estuary sediments since the early to mid-1950s. Below the 7.6 cm interval the total PCB concentrations averaged about 5 µg/kg whereas the shallower sediments are greater than 10 µg/kg.

The MOC core sample was collected from a shallow depositional pond within the main drainage of the Suqi River. This sampling site is immediately down gradient of the center of activities at the NEC FUDS. The most elevated PCB concentrations were found in the near surface sediments (Figure 4) of the core sample. The upper 10 cm of sediment contained a total PCB concentration of greater than 550 µg/kg. With depth, the PCB concentrations decreased abruptly to less than 35 µg/kg. This abrupt change in the MOC core likely represents the transition from more recently deposited sediments transported from the MOC and up gradient areas of the NEC site.

A contaminant trend similar to the PCB profile is evident in the estuary and MOC sediment mirex concentrations. The MOC core sediments contained about 15 µg/kg in the near surface sediments. Mirex concentrations decrease with depth and the concentrations in the MOC core are significantly elevated relative to the estuary and landfill core samples. In the estuary core, there is a gradual increase in the mirex concentrations from about 28 cm to the surface sediments (Figure 4). The mirex concentration increases from about 0.6 µg/kg at a depth of 6 cm and increases to concentrations greater than 1.7 µg/kg in the near surface sediments.

Although the mirex concentrations in the landfill core (Site 7) are less well defined, there is a general increase upwards. This site served as a solid waste landfill for the area. The decreasing concentrations with depth in the landfill core suggest the source of the mirex to the ephemeral pond waters adjacent to the landfill occurred during more recent times. Because there are no external drainages to the pond where the core was collected, the adjacent landfill is the likely source of the mirex.

The DDE concentrations in the MOC core showed a similar profile (Figure 4) to the PCB and mirex. The uppermost 10 cm of sediments contained concentrations of about 6 µg/kg. The deeper core intervals had DDE concentrations that were below 1.0 µg/kg. The DDE profile in the estuary and landfill cores followed a similar trend with progressively decreasing concentrations with depth and is about one-tenth the DDE concentration of the MOC core. As can be seen from Figure 4, the concentrations in these two core samples decreased abruptly from the near surface sediments to a depth of 10 centimeters.

Mercury concentrations collected from the MOC and estuary cores are similar to the PCB, mirex and DDE profiles. The most elevated concentrations were found in the near surface MOC core sediments. As can be seen from Figure 4, the mercury concentrations in the MOC sample decreased abruptly from 140 to 180 µg/kg at a depth of about 9 cm to <50 µg/kg at a depth of 14 cm. The deepest core segment contained about 30 µg/kg mercury. The estuary sediment core mercury concentrations increased from

about 15 µg/kg in the near surface sediments to about 58 ug/kg at a depth of 6-7 centimeters. Mercury concentrations gradually decrease to about 17 ug/kg at a depth of 15 cm. Based on the ^{137}Cs profile of the estuary core, the maximum mercury concentration occurred at about the time of military occupancy of the NEC site. The mercury concentrations in the deeper estuary core sediments may be indicative of atmospheric deposition as they are similar to total mercury profiles in remote lakes in the Canadian Arctic and Eastern North America, in which modern fluxes are 1.1 to 7 times higher than pre-industrial (Lockhard,1998; Macdonald et al. 2000).

The significantly elevated mercury concentrations in the near surface MOC core suggests the primary source was from activities conducted at the up gradient regions of the NEC during the time the military occupied the site, as well as, from remedial activities conducted during more recent times. The concentrations of mercury in the landfill core increase with depth from about 23 ug/kg at the surface to more than 90 ug/kg below 21 cm. The mercury concentrations in the MOC and landfill cores were two to four times more elevated than found in the estuary core sections. These data indicate the MOC and the landfill are sources of mercury to the down gradient sediments including the Suqi Estuary and the ephemeral pond located adjacent to landfill Site 7.

The highest concentrations of PCBs, volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs) of samples collected by the COE contractors in the sediments and soils at the NEC were collected from the Suqi River drainage and floodplain immediately down gradient of the MOC (Montgomery Watson Harza, 2002). Maximum PCB concentrations reported in the sediments collected and analyzed by the COE contractors were about 22 mg/kg total PCBs. Soil concentrations collected near the confluence of the Suqi River and estuary were about 5.4 mg/kg. The elevated diesel range organics in the region of the MOC identified during the remedial Investigations is likely related to the accidental spill of more than 185 000 gallons of fuel oil when one of the four, above ground, 400 000 gallon storage tanks was reportedly ruptured by snow clearing equipment in 1969. The overall elevated contaminant concentrations of PCBs, DDE, mirex and mercury in the Main Operations Complex core sample are illustrated in Figure 4.

The highest PCB concentration in the Atuk Volcano lake core sediments was 14.8 µg/kg at the 0-3 cm interval. Within Collier Lagoon sediment intervals PCBs were below study detection limits (1 µg/kg). Mirex, DDE, and hexochlorobenzene were at non-detect concentrations at the two remote sediment core sampling sites on St. Lawrence Island. Mercury was not analyzed for the samples collected at the remote SLI sites

NEC Plant Analysis. As can be seen from Table 1, there are significantly higher concentrations of PCBs in the St. Lawrence Island plant samples relative to the three mainland Norton Sound FUDS samples. The highest total PCB concentrations were found in plant samples collected at the Northeast Cape and among these the most elevated concentrations were found in the samples collected near the MOC. Plant samples collected near the MOC by the COE's contractors as a part of the remedial investigations phase contained more than 9.0 mg/kg total PCBs (Montgomery Watson, Harza, 2002).

Rinsing plant samples collected from Gambell and NEC with distilled-deionized water reduced total PCB concentrations by more than 75% (40 to 10 ug/kg) relative to unrinsed plants, indicating PCB contaminated dust was deposited on the leaves and stems. The NEC plant substrate sampling sites were on tundra, while the Gambell sites were upland (igneous bedrock) and from cobble beach deposits. There was active remediation at the NEC at the time the plants were collected including building and slab demolition and relative high vehicular and heavy equipment traffic on the dirt roads, resulting in the redistribution of contaminated dust. Remedial activities involving demolition and production of dust contributed to the redistribution of contaminants at the NEC, indicating contaminants were being mobilized during site remediation. In addition to the remobilization of contaminants resulting from dust, Chiarenzelli, et al., 2004, described the potential for the volatile release of PCBs resulting from remedial dredging operations providing an additional pathway for the release of contaminants from impacted soils and sediments.

The maximum total PCB concentration in the Mainland Norton Sound plant samples was less than 2.0 µg/kg; most sample concentrations were less than 1.0 µg/kg. The predominant PCB congeners in the mainland FUDS plant samples were tetrachlorinated biphenyls, whereas the PCB concentrations of the SLI plant sampling sites were comprised of hexachlorobiphenyl and more chlorinated congeners.

Table 1.

Distinguishing Locally and Atmospherically Derived Contaminants

Several lines of reasoning and empirical observation were used to distinguish "local" from atmospherically derived contaminants or background levels at the Norton Sound FUDS including:

- Changes in contaminant concentration with time (temporal variations);
- Gradient studies (spatial variations);
- Comparison to background concentrations (baseline studies);
- Contaminant fingerprints (compositional variation and makeup); and
- Associated contaminants.

Concentration profiles. Of the three NEC FUDS and SLI remote locations, the Northeast Cape cores had the highest PCB concentrations. The most elevated concentrations were found in the near surface sediments of the MOC core. Concentrations in the NEC cores decreased with depth. Based on ^{137}Cs concentrations, PCBs were first deposited in the 1940's and that concentrations subsequently increased during the post-1940/50 timeframe to the present time. The increased flux of PCBs decades after their maximum use and production in the 1970's must be explained. One possible explanation is a lag time between contaminant use at low latitudes and atmospheric transport and eventual deposition at high latitudes (Wania and MacKay, 1996). Alternatively, the remobilization of --contaminants during remedial activities could explain the elevated concentrations in the more recent sediments.

Contaminant Gradients. Assuming sampling locations and media are similar, a small area receiving primarily atmospherically derived contaminants should show relatively small variations in contaminant concentrations. In the case of the NEC sediment cores, there are considerable differences between PCB concentrations immediately down gradient of the MOC relative to the Suqi Estuary core. PCB concentrations in a core sampled immediately down gradient of the MOC contained more than 550 µg/kg indicating the source of PCBs and other contaminants was up gradient of the MOC sampling site.

Background Concentrations. Background PCB soil and sediment concentrations in remote northern areas without nearby sources generally are within the low µg/kg range. We are unaware of background concentrations in areas unaffected by local sources in excess of a few µg/kg. Certainly the concentrations recorded in the MOC core (>550 µg/kg) cannot be considered background. In addition, concentration trends in vertical profiles indicate that substantial fluctuations and large increases in PCB concentrations have occurred at the Northeast Cape over the last 50 to 60 years.

Contaminant Fingerprinting. Within contaminant groups, particularly those in which substitutions result in large variations in molecular weight, individual congeners often have physiochemical properties whose values range over several orders of magnitude. For example, PCB congeners vary in their reported aqueous solubility by over eight orders of magnitude. In addition, vapor pressure, octanol-water and air-water partitioning coefficients (log (PL/Pa), KOW, and KAW) all show large ranges and have been utilized by Wania and MacKay (1996) to explain contaminant partitioning on a global scale. As a general rule, heavier PCBs, dioxins, dibenzofurans and chlorinated pesticides and benzene compounds with extensive chlorine substitution tend to be relatively immobile and are likely to remain near sources. Conversely, lighter congeners are more mobile and will migrate and condense.

One measure of the chlorination of an environmental mixture of PCBs, is the average chlorine per biphenyl ratio. This ratio can be compared to commercial formulations known as Aroclors. For example, Aroclors 1248, 1254, and 1260 have chlorine per biphenyl ratios of 4.0, 5.2, and 5.8, respectively. Air and water PCB concentrations tend to be lightly chlorinated (ca. 4.0), whereas soil and sediment are slightly more chlorinated. Fish tissue, blood, lipids, and breast milk samples are often highly chlorinated (ca. 6.0).

The upper part of each core from the NEC, where maximum PCB concentrations were measured, have chlorine per biphenyl ratios that range from 5.39 to 5.66, mixtures too heavily chlorinated to have been transported atmospherically. In addition, the patterns are dominated by congeners with five, six, and seven chlorines, generally lacking from air samples, but abundant in highly chlorinated Aroclors such as 1254 and 1260.

Associated Contaminants. Mirex, DDE, mercury, and other contaminants have similar time-concentration trends as those shown by PCBs. The discovery of mirex at the Northeast Cape was especially puzzling in that it was produced for a short period of time (ca. 1959-1972). Mirex presence is likely related to the use of the material as a flame retardant in goods brought to the island by the military. The insecticide DDT was used universally and DDE is a commonly reported breakdown product

that persists in the environment. Mercury concentrations and distribution tracks well with the temporal trends and distribution of other contaminants in the NEC cores.

Local or Far-Traveled Contaminants?

The data presented above suggests that PCB concentrations measured in cores at the Northeast Cape are above those reported in remote areas located on St. Lawrence Island. Based on ^{137}Cs dating of the single Suqi Estuary core sample, deposition of PCBs commenced about 1940-1950 and increased rapidly reaching maximum concentrations in the uppermost, recent sediments of the Suqi Estuary. In addition, the abundance of other contaminants, including mercury and mirex show trends similar to the PCB sediment profiles. Because the concentrations of PCBs, mirex, DDE and mercury exceed the atmospheric input at this remote locality, the most likely scenario is one in which contaminant deposition began with the initiation of military activity at the NEC site.

The relatively high concentrations of contaminants found in the uppermost sediment layers at the NEC are likely a reflection of recent remedial activities and normal sedimentological processes in the basin, including erosion, reworking and re-deposition of contaminated sediments and soils. These data suggest that the redistribution of contaminants by wind, water, and animals is likely to occur for extended periods at elevated fluxes. With time, contaminated soil and sediment will eventually contain only the most recalcitrant, immobile and most highly chlorinated congeners, as others are degraded or removed from the ecosystem, including volatile losses (Chiarenzelli et. al., 1996).

Summary and Conclusions

The increase from about 5 µg/kg total PCB concentration in the estuary sediment core section below about 7.5 cm to more than 30 µg/kg in the surface sediments, provides a measure of the contributions made by the military occupancy to the NEC environment. The military influence is also evident in the sediment core data collected from the shallow pond located immediately down gradient of the MOC. The near surface sediments in the MOC core are more than an order of magnitude more concentrated in PCBs than the underlying sediments. The relative concentrations of the MOC sediment concentrations of PCBs, mirex and mercury indicate proximity to the contaminant source(s). The upper 10 cm of the MOC sediment core are more than 15 times more concentrated in PCBs than the more distant Suqi Estuary sediments. The ^{137}Cs dated estuary core provides a measure of the relative concentrations of PCBs, mirex, DDE and mercury contributed during the time the NEC site was occupied by the military.

The MOC area of the Northeast Cape is also the likely source of mirex. The MOC sediment core mirex concentrations in the near surface sediments are about eight times more concentrated than the estuary sediment core concentrations. Similar trends are evident in the MOC sediment core mercury data. The near surface sediments mirror the mirex and PCB profiles reflected by the relative six-fold increase in the near surface sediments. The increased mercury concentrations occurred subsequent to 1954 (post-^{137}Cs first appearance), reaching an approximate six-fold maximum in the near surface sediments. Similar trends are evident in the eight-fold increase in the concentration of mirex in the upper segments of the sediment core (Figure 8). The near surface sediments collected from the Suqi Estuary, from the

shallow pond down gradient of the MOC and from the shallow, ephemeral pond adjacent to the Site 7 landfill, contain elevated concentrations of PCBs, mirex and mercury.

Relative comparison of the sediment cores collected from the Northeast Cape to the two remote locations sampled on St. Lawrence Island' demonstrated that the PCB, DDE and mirex concentrations are significantly elevated and attributable to military activity at the NEC FUDS and indicate that the deposition of contaminants from long-distance transportation is minimal. Based on the relative concentrations of contaminants in the sediment cores, it is likely the military occupancy contributed an eight-fold increase in the total PCBs, a four- to six-fold increase in the mirex and a six- to eight-fold increase in the mercury concentrations to the NEC sediments. Further PCB mixtures are composed predominantly of highly chlorinated congeners whose chlorination and chemical properties precludes long-distance transport. The elevated concentrations of contaminants in the upper core segments also indicate contaminants are being remobilized as a consequence of remediation and changes in river and flow within the surface drainages.

The elevated concentrations of PCBs in the NEC and Gambell plant samples relative to the concentrations at the three mainland Norton Sound FUDS provides further evidence that the source of PCBs, mirex and DDE are directly related to the military presence, rather than atmospheric sources. The rinsed relative to the unrinsed plant data from the NEC and Gambell also indicate contaminants are being remobilized as a consequence of site remediation.

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Table 1. Total PCB concentrations (µg/kg) of plant samples collected from five Norton Sound, Alaska FUDS communities during 2002, 2006 and 2007. NEC and Gambell plant samples results are unrinsed.

Location	No. Plants	PCB concentrations (Range)	PCB concentrations (average)
NEC	7	1 – 90.3	40.9
Gambell	10	3.9 – 17	10.0
Elim	9	<1	<1
Unalakleet	19	<1	<1
Wales	20	1 – 1.5	<1

(Scrudato)

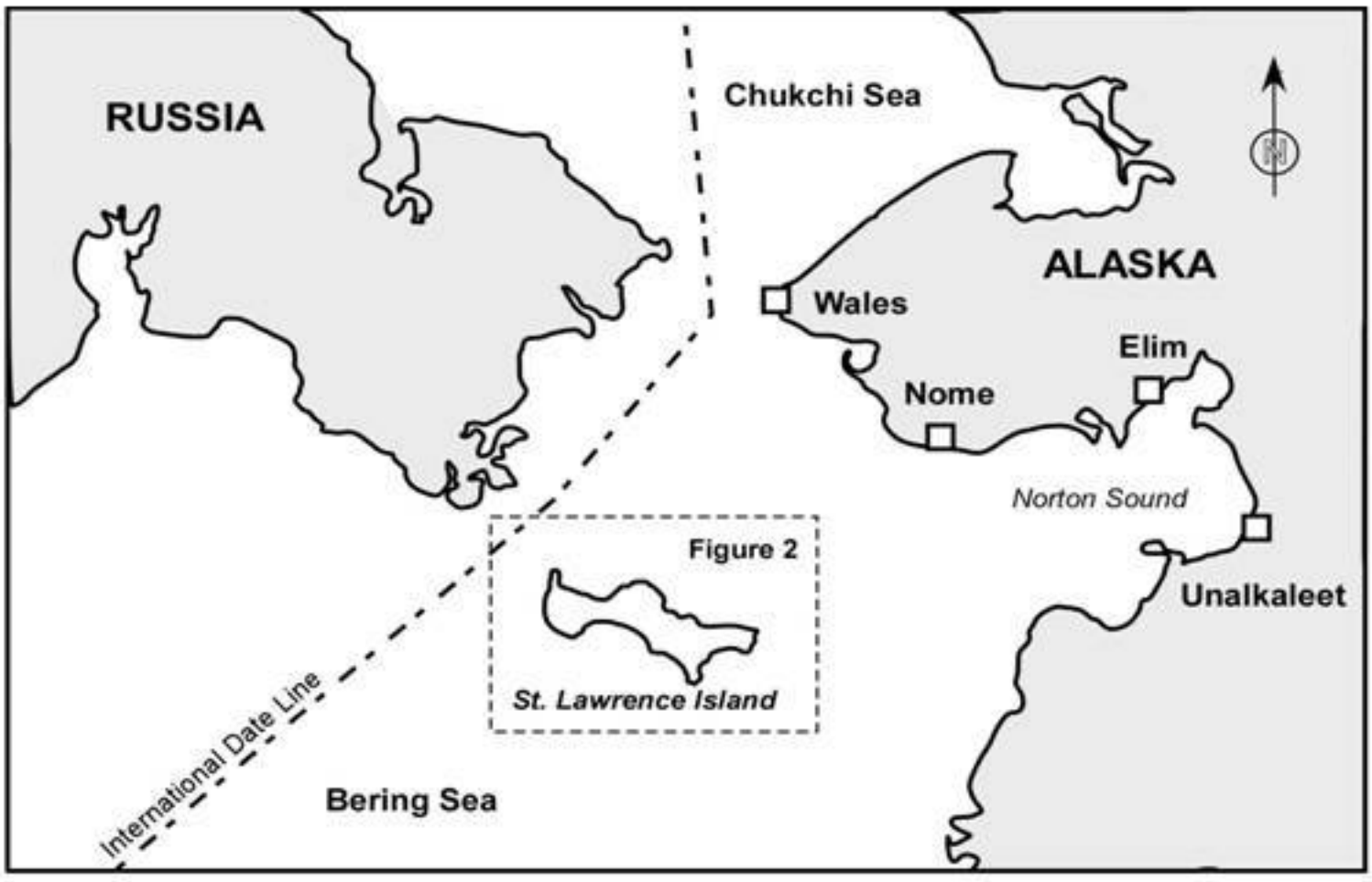
Figure Captions

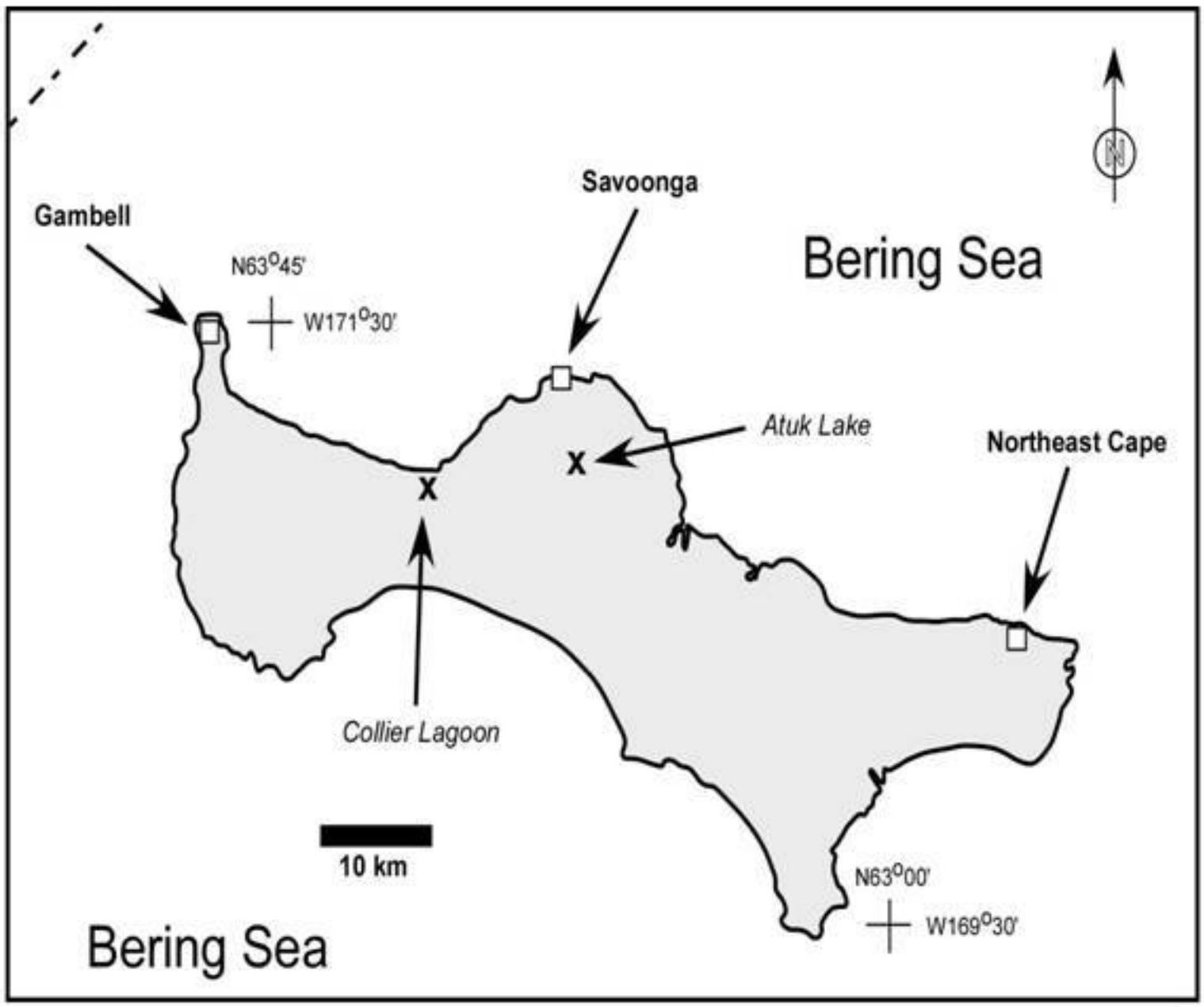
Figure 1. Locations of St. Lawrence Island FUDS at Northeast Cape (NEC), Gambell and the Mainland Norton Sound Alaska FUDS of Elim, Wales, Unalakleet.

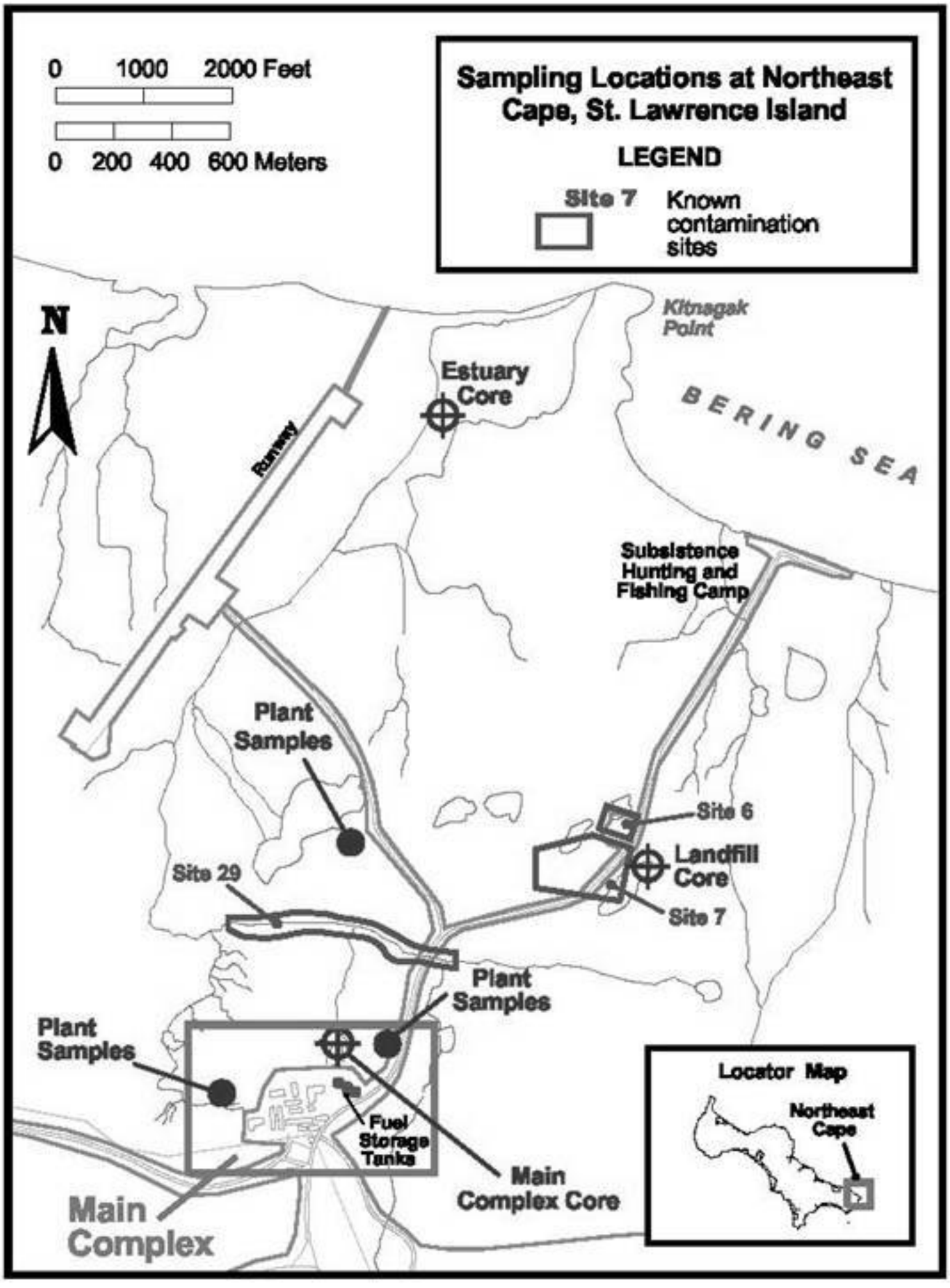
Figure 2. St. Lawrence Island communities of Gambell, Savoonga, the Northeast Cape and two remote sampling sites at Collier Lagoon and Atuk Volcano lake.

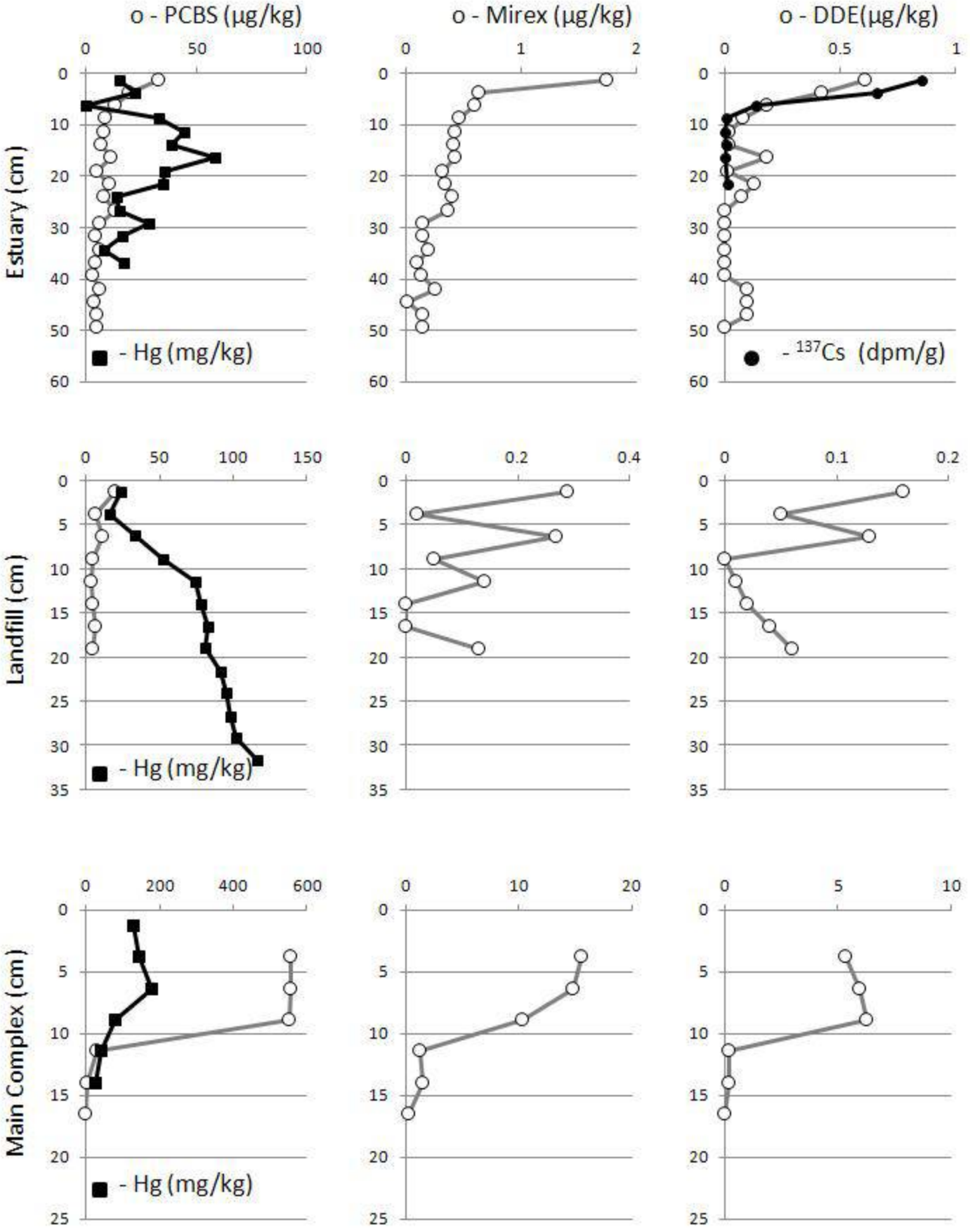
Figure 3. Northeast Cape (NEC) FUDS site on St. Lawrence Island showing locations of the three sediment core collection sites, plant collection sites, Main Complex location and Suqi River drainage system.

Figure 4. Comparison of PCB, Mirex, DDE, and Mercury distributions in the Suqi Estuary, Landfill 7, and the Main Complex sediment cores and the ^{137}Cs profile from the Suqi Estuary core at the Northeast Cape, St. Lawrence Island.


RUSSIA
Chukchi Sea
ALASKA
Wales
Elim
Nome
Norton Sound
Figure 2
Unalkaleet
St. Lawrence Island
International Date Line
Bering Sea


Gambell
N63°45'
W171°30'
Savoonga
Bering Sea
Atuk Lake
Northeast Cape
Collier Lagoon
10 km
N63°00'
W169°30'
Bering Sea


0 1000 2000 Feet
0 200 400 600 Meters
Sampling Locations at Northeast Cape, St. Lawrence Island
LEGEND
Site 7
Known contamination sites
N
Kitnagak Point
Estuary Core
BERING SEA
Runway
Subsistence Hunting and Fishing Camp
Plant Samples
Site 6
Landfill Core
Site 29
Site 7
Plant Samples
Plant Samples
Fuel Storage Tanks
Main Complex Core
Main Complex
Locator Map
Northeast Cape


o - PCBS (µg/kg)
o - Mirex (µg/kg)
o - DDE(µg/kg)
Estuary (cm)
- Hg (mg/kg)
- 137Cs (dpm/g)
Landfill (cm)
- Hg (mg/kg)
Main Complex (cm)
- Hg (mg/kg)