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Toxic chemicals and their impacts in the St. Lawrence Estuary and Saguenay Fjord, Quebec, Canada: from a chemical to an ecosystem-based risk management

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Abstract

The St. Lawrence Estuary (SLE) and Saguenay Fjord (SF), Quebec, Canada, have received world-wide attention in the early 1980s when high concentrations of contaminants and high prevalence of lesions including neoplasia, hermaphrodism and infection by opportunistic agents were reported in beluga whales (*Delphinapterus leucas*). Both persistent organic pollutants (POPs) such as PCBs mainly originating from the upstream industrialized sectors of the Great Lakes and the upper St. Lawrence River, and local contamination by polycyclic aromatic hydrocarbons (PAHs) and mercury have been incriminated. The release of these chemicals has been successfully reduced through environmental regulations and restoration of contaminated areas. Since 1970s, declines in mercury, PAHs and PCBs have been observed in sediments and in biota. However, organisms remain exposed to complex mixtures of contaminants including regulated persistent compounds remaining in the ecosystem and newer compounds which also have the potential of causing deleterious effects. Interactions between toxic chemicals and other environmental stressors may increase the risk of deleterious impacts. New concerns include: chronic inputs of tributyltin from ship transportation associated with reproductive/immune disturbances, interaction between nutritional condition and POPs, increasing concentrations of brominated flame retardants in beluga tissues and inputs of agricultural chemicals and nutrients from SLE tributaries. Several fish populations historically used as preys by the beluga are declining as a consequence of multiple anthropogenic factors. Moreover hypoxic area in the bottom of the SLE is increasing and could act as an additional stressor. An ecosystem-based approach is being developed to pursue the protection the SLE/SF ecosystems facing multiple stressors and variable environmental conditions.

Keywords: St. Lawrence Estuary, Saguenay Fjord, toxic chemicals, beluga whale

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The St. Lawrence Estuary and Saguenay Fjord ecosystem

The St. Lawrence River Estuary (SLE), Quebec, Canada is one of North America's major estuary (Fig.1). It receives most of its freshwater inputs from the St. Lawrence River, draining the Great Lakes, and from the Saguenay Fjord (SF). The starting point of the Laurentian Trough, a 350 m deep channel that crosses the Gulf of St. Lawrence, is located near the confluence of the SF and the SLE. At this site, rapid depth changes and tidally-induced cold water upwelling promote nutrient circulation, biological diversity and high productivity. The SLE and SF provide a feeding habitat for several cetacean and phocid species including the SLE beluga whale (*Delphinapterus leucas*), a permanent resident of this area and a threatened species (COSEWIC, 2004). In 1998, the SF-SLE Marine Park (http://www.pc.gc.ca/amnc-nmca/qc/saguenay/default.asp) was created to protect a section of the SLE and SF. The SLE marine protected area (MPA) is under development to extend the boundaries of the protected zone and to ensure the long-term conservation and protection of marine mammals, their habitats and food resources through integrated management of the ecosystem (http://www.qc.dfo-mpo.gc.ca/ ZPMEstuaire /fr/proj.asp) (Fig.1).

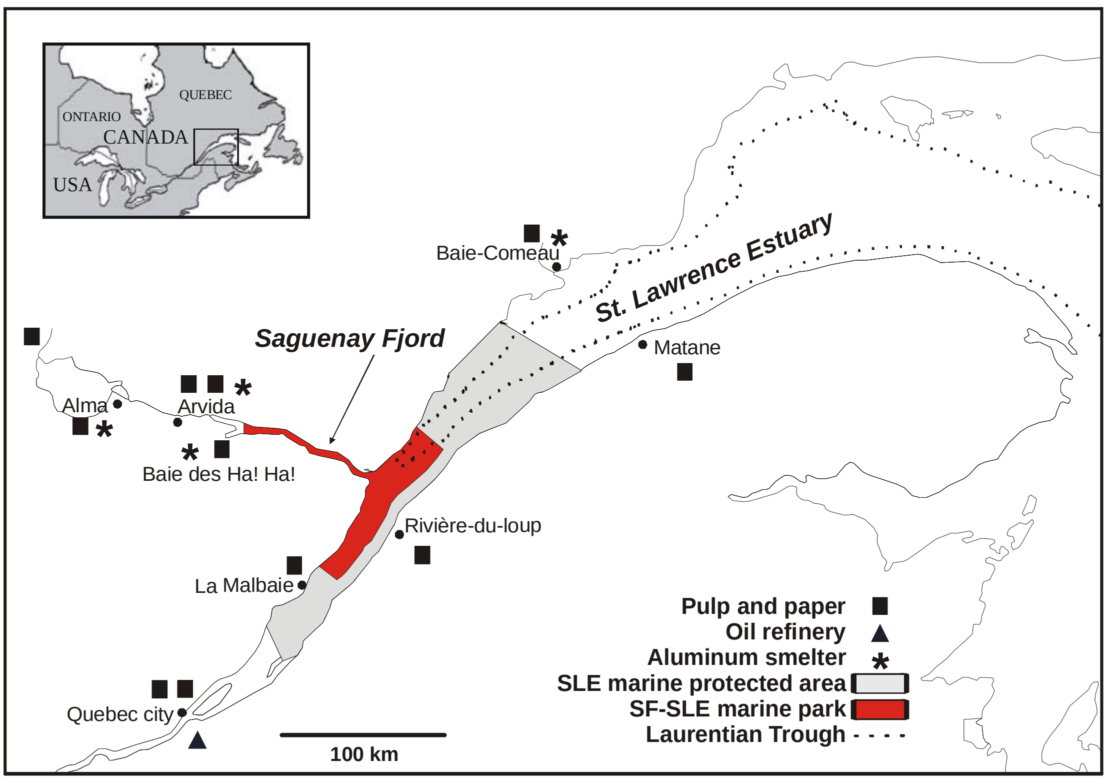

Fig. 1. Map of the St. Lawrence Estuary (SLE) and Saguenay Fjord (SF) indicating the main local industrial sources of contaminants, and the boundaries of the SF-SLE marine park and of the proposed SLE marine protected area (adapted from White and Frank, 1997, and from http://www.qc.dfo-mpo.gc.ca/ ZPMEstuaire/fr/ proj .asp).

One important threat to SLE marine mammals is chemical contamination of their habitats and diets. The SLE receives relatively high loads of contaminants through fluvial and atmospheric inputs, originating from the industrialized sectors the Great Lakes and the upper St. Lawrence River (Lebeuf and Nunes, 2005). In addition, the Laurentian Trough is the first significant zone of permanent accumulation of fine-grained sediments and associated contaminants downstream of Lake Ontario (d'Anglejan, 1990). Local sources of industrial pollution include several pulp and paper mills and aluminium smelters, an oil refinery, runoffs and effluents from agricultural and urban areas (White and Frank, 1997). Several harbours and marinas, and shipping traffic are other sources of pollution (Viglino *et al.*, 2006).

Chemical-oriented management

In the 1970s, commercial fisheries were restricted in the SF because of mercury (Hg) contamination of shrimp and fish containing concentrations greater than 10 µg g^{-1} ww. The main source of Hg was a chloralkali plant located in Arvida, at the head of the SF operating since 1946. Following regulation of the release of Hg (1974) and the closure of the plant (1978), concentrations of Hg declined markedly in shrimp and are now below the Canadian human consumption guideline (0.5 µg g^{-1} ww) (Cossa, 1990; Gobeil *et al.*, 1998). However, Hg persists in deep sediments and fishing restrictions were maintained to prevent re-suspension of the contaminated sediments. In the 1980s, large concentrations of persistent, bioaccumulative and toxic (PBTs) organic chemicals were observed in the tissues of dead stranded beluga whales collected on the shores of the SLE. Up to 756 µg Hg g^{-1} dw were measured in their liver (Wagemann *et al.*, 1990) and 300 µg g^{-1} lw PCBs (Aroclor 1254) in their blubber (Martineau *et al.*, 1987). Exposure of the SLE beluga population to contaminants was identified as a possible cause of its failure to recover from overhunting interrupted in the 1950's (Hammill *et al*., in press). Furthermore, stranded beluga whales exhibited unusual lesions potentially related to toxic chemicals including gastrointestinal tumors affecting 27 % of the stranded animals (Martineau *et al.* 2002), one hermaphrodite (De Guise *et al.*, 1994) and one pseudo-hermaphrodite. Infections with opportunistic agents were suggestive of immunosuppression (De Guise *et al.*, 1995). The finding of benzo[*a*]pyrene DNA adducts in SLE beluga liver indicated that they were exposed to genotoxic polycyclic aromatic hydrocarbons (PAHs) (Shugart *et al.*, 1990). PAHs released by an aluminium smelter located in Arvida contaminating sediments and benthic prey, were proposed as a contributing factor for the development of neoplasia. Other factors could be involved in the pathogenesis of neoplasia such as organochlorine chemicals (e.g PCBs) or infectious agents acting as tumor promoters or the presence of unidentified natural genotoxic substances in the diets (Martineau *et al.*, 2002). The Arvida aluminium smelter has markedly reduced its emission of PAHs since the 1980s and a decline in the concentrations of PAHs was observed in the SF sediments (Smith and Levy, 1990). There was also an indication of reduced exposure of a prey fish species to PAHs, with a 4-fold reduction in the hepatic DNA adducts in SLE Atlantic tomcod (*Microgadus tomcod*) from 1991 to 2000 (Wirgin *et al.*, 1994; Couillard *et al.*, 2005). Concentrations of PCBs and of organochlorine pesticides (OCPs) have decreased by up to a factor of two between 1987 and 2004 in blubber of SLE beluga (Fig 2), presumably in response to regulation of use and release and to reduced inputs of these compounds in their habitat (Lebeuf *et al.*, 2007). Furthermore, a deluge of rain in the SF has covered the upper north harm of the Fjord and Baie des Ha!Ha! with a layer of new and low contaminated sediments reducing the risk of transfer of Hg, PAHs, PCBs and OCPs to the foodweb (Pelletier *et al.*, 1999; Lebeuf *et al.*, 2003).

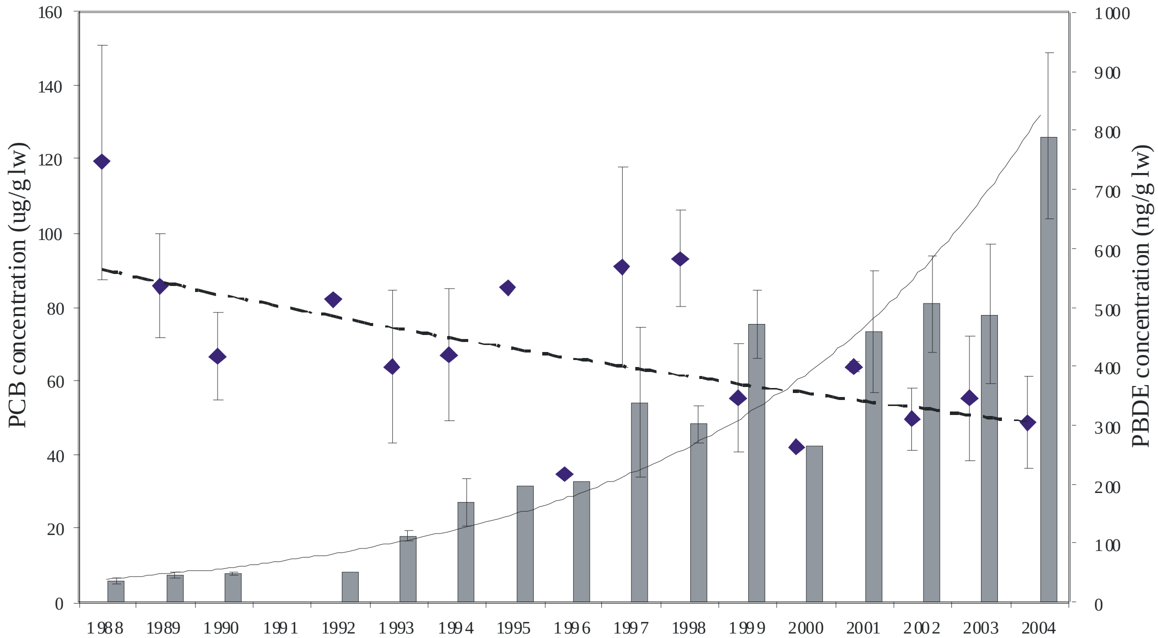

Fig. 2. Temporal trends of PCBs (diamonds and dotted line) and PBDEs (bars and full line) in blubber of male beluga whales from the SLE between 1988 and 2004 (adapted from Lebeuf *et al.*, 2004, 2007, and personal communication).

Although environmental regulations have led to significant declines of the concentrations of several PBTs in the SLE ecosystem, aquatic species remain exposed to complex mixtures of contaminants. Regulated persistent compounds are remaining in the system and newer products such as pharmaceutical agents, brominated and perfluorinated compounds have the potential of causing deleterious effects alone or in combination with other stressors. Concentrations of the flame retardants polybrominated diphenyl ether (PBDE) have increased exponentially from 1988 to 1999 in the blubber of beluga whale and are still increasing (Fig 2; Lebeuf *et al.*, 2004 and personal com.). These compounds could potentially impair endocrine and neurological functions in aquatic species. Tributyltin (TBT), an antifouling agent still used on large-size boats (until 2008), is accumulating in sediments of the SF (Viglino *et al.*, 2006) and has been measured in the liver of SLE beluga (up to 2.1 mg kg^{-1} dw; St.-Louis, 2000). This endocrine disruptive compound has been associated with imposex in whelks (*Buccinum undatum*) in the SLE and SF and with altered reproduction and male bias sex ratio in clams (*Mya arenaria*) at the mouth of the SF and in harbours on the south shore of the SLE (Gagné *et al*., 2003; Siah *et al.*, 2003; Viglino *et al.*, 2006). Little is known on the inputs of pesticides, nonylphenols, pharmaceutical compounds in the SLE ecosystem. The potential impacts of these compounds on neuroendocrine function of aquatic species are causes for concern, particularly for anadromous/catadromous fish species in which delayed effects on salinity adaptation have been demonstrated after spike exposure to environmentally realistic concentrations (Fairchild *et al.*, 1999; Waring and Moore, 2004).

The need for an ecosystem-based management

As a consequence of human activities, the variability of environmental conditions is increasing and environmental changes can interact with toxic chemicals and increase the risk of deleterious impacts without new inputs of chemicals (Couillard *et al.*, 2007a, b). First, a zone of hypoxia is developing in deep waters of the Laurentian through in the SLE (Gilbert *et al.*, 2005). Hypoxia could alter the bioavailability of toxic chemicals through changes in their chemical and biological transformation pathways. For example, anoxic sediment can favour the conversion of Hg to methylHg and its transfer to the food chain (Debruyn *et al.*, 2006). Hypoxia can also increase ventilation rate and uptake of chemicals, interact with the Ah receptor pathway involved in the toxicity of dioxin-like chemicals, or alter the food web structure and the spatial distribution of prey species and therefore affect the dietary exposure of top predators such as beluga to PBTs (Prasch *et al.*, 2003; Debruyn *et al.*, 2006; Couillard *et al*., 2007a, b). Secondly, changes in water temperature and habitat are increasing the duration and severity of the seasonal fasting period in high latitude fish species, such as cod which are preyed upon by beluga. Fasting can lead to redistribution and changes in concentrations of PBTs in target tissues and increase the risk of toxic effects (Jörgensen *et al*., 2006). In the SLE, large-size emaciated tomcod sampled in spring at the end of their winter fasting period, had higher concentrations of PCBs in their liver and inhibition of a cytochrome P4501A activity, indicative of hepatocellular injury (Couillard *et al.*, 2004 and 2005). Lastly, more frequent introduction of pathogens as a result of global trade, tourism and climate change could interact with immunotoxic contaminants in the SLE, a great risk for the isolated SLE beluga population. As an example, exposure of mussels (*Mytilus edulis*) to TBT (ng L^{-1}) increases their susceptibility to infectious diseases and their mortality rate (St-Jean *et al*., 2002a, b).

In the SLE and SF, aquatic organisms are still exposed to a wide variety of potentially toxic chemicals and changing environmental conditions can markedly increase the risk of deleterious impacts. Thus, a chemical-based approach to risk management centered on the management of single sources of contaminants with no consideration of possible interactions with other stressors could lead to a marked underestimation of the risk and to inadequate protection of marine aquatic life in the SLE and SF. Management of the risks from toxic chemicals should not be limited to the development and application of regulations on the release of individual chemicals into the aquatic environment; it must also include management of the exposure and the vulnerability of aquatic species populations to toxic chemicals through habitat, ocean and fisheries. For example, to reduce the risk associated with immunotoxic PBTs in beluga, regulation of the release and uses of these substances is mandatory but the introduction of novel pathogens in the SLE should also be prevented, and the effects of habitat degradation and of urban and agricultural pollution on prey species should be managed (Fig.3; Couillard *et al*., 2007a, b). Environmental monitoring of exposure and effects of toxic chemicals should be integrated with monitoring of chemical, physical and biological stressors. A better understanding of the interactions between toxic chemicals and environmental factors is a fundamental requirement for an efficient management and protection of the SLE ecosystem. New multidisciplinary and integrative research programs are in development for that purpose.

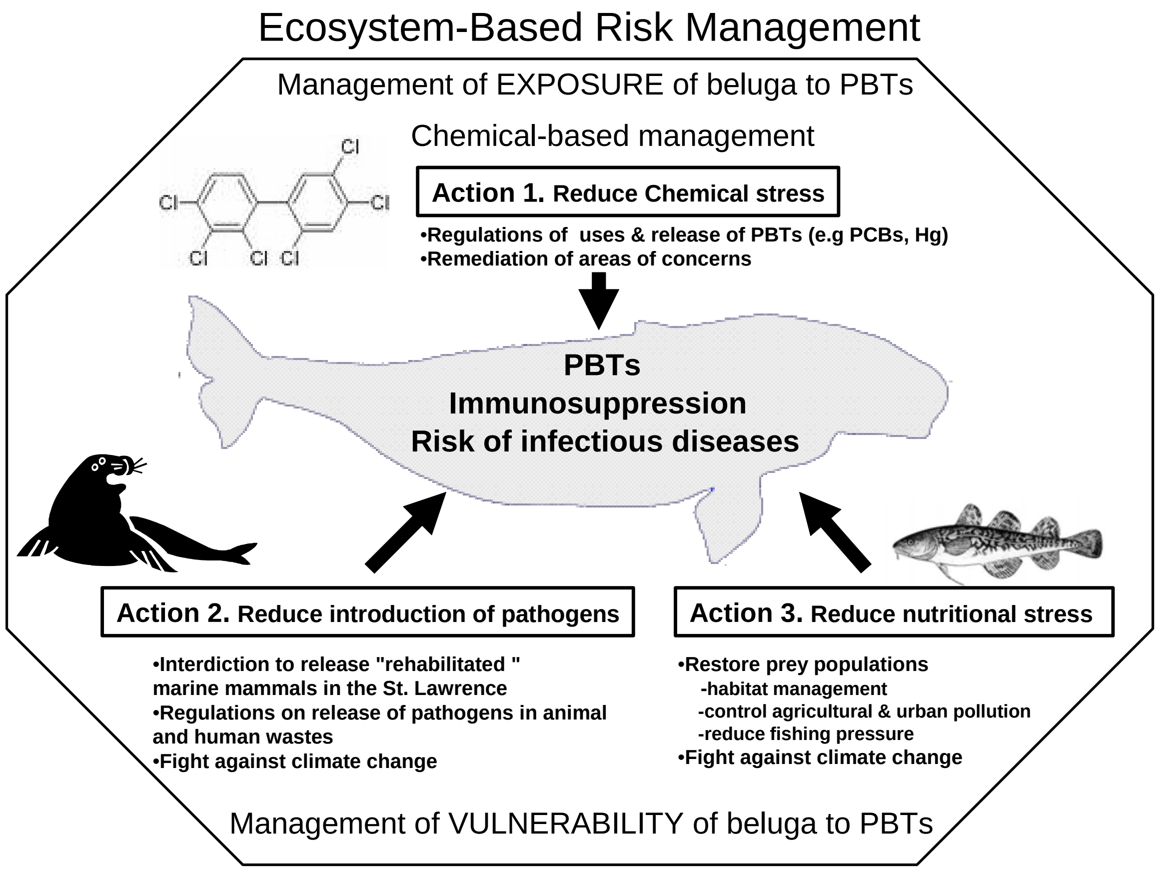

Fig.3. Different actions to reduce the risk related to exposure of beluga to persistent bioaccumulative toxic compounds in the St. Lawrence Estuary. Adapted from Couillard *et al.* 2007b.

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