Marine environmental contaminant issues in the North Pacific: What are the dangers and how do we identify them?

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Introduction

A little over a decade ago, Waldichuk (1990) reviewed the state of industrial and domestic pollution of the North Pacific and concluded that interfaces (*e.g.*, air-water, water-sediment, shorelines) and coastal areas, especially those surrounded by dense population and industry, were most at risk. His list of critical contaminants – hydrocarbons and polynuclear aromatic hydrocarbons (PAHs), organochlorine compounds, metals, radionuclides, and persistent solids – remains valid today.

Although toxic effects of contaminants have long been known, it was only during the past decade that we have learned the myriad ways trace quantities of chemicals can produce subtle disruption to endocrine systems every bit as threatening to aquatic animals as outright toxicity (Colborn *et al.* 1996). For this reason, Waldichuk's priority contaminant issues – halogenated hydrocarbons and sewage – were well chosen. During the past decade, expanded industry and increasing coastal populations have escalated the pressure on productive marginal seas (Fig. 1) – the very regions that are being counted on to provide even more protein for future populations. For example, in 1998 an estimated two-thirds of the world's population (3.6 billion) lived within 60 km of the coast (UNESCO 1998), and total population is increasing at about 77 million per year (*U.S.* Census Bureau 2002). In U.S.A., over 50% of the population lives in the narrow coastal fringe and that population is increasing by about 1.3 million per year (Culliton 1998).

In addition to chemical contaminants, these coastal seas are under onslaught from enhanced nutrient and sediment loadings, climate change, over-fishing, habitat disruption and the introduction of exotic species. For the most intensively utilized enclosed seas of East Asia, *e.g.*, the South China Sea, projections are indeed grim (Morton and Blackmore 2001).

Here we discuss the major threats human activities present to North Pacific marine ecosystems with chemical contamination as a central theme. We discuss briefly concurrent issues of climate change, disruption of CNP (carbon, nitrogen, phosphorus) cycles, and predation, because these factors confound chemical contamination, both in terms of its effects and in the way chemical contaminants pass through marine systems. It is not our intention to present a comprehensive review of the literature. Rather, we highlight the contaminant issues facing the North Pacific, drawing suitable examples for the most part from literature of the last decade. Finally, we propose the sorts of observations and research required to mobilize society to reverse its present course, which if unchecked, will lead to the yet further widespread destruction of coastal ecosystems.

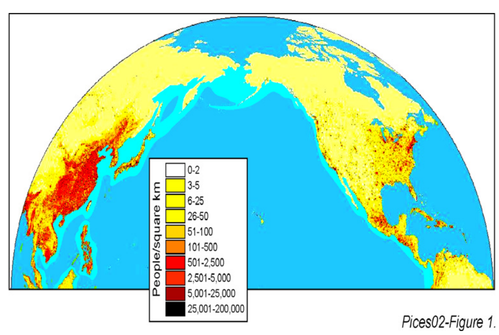

Fig. 1 The North Pacific Ocean, showing the density of human population near coastal seas and the importance of coastal seas for primary production, which, by inference, also represents secondary production.

Pressures on North Pacific marine ecosystems

Climate change

Climate change poses several kinds of risk, including temperature rise through greenhouse gas forcing (IPCC 1995), alteration of the hydrological cycle (Dynesius and Nilsson 1994; Vörösmarty *et al.* 2001), increased exposure from ultraviolet radiation (Weatherhead and Morseth, 1998) and sea-level rise (Ledley *et al.* 1999). These sorts of change, which imply significant effects on aquatic systems and humans, are often difficult to detect at their early stages due to natural variability at time scales from years to decades to centuries (see, for example, Francis *et al.* 1998; Hare and Mantua 2000).

Air temperature in the northern hemisphere has been anomalously high during the past decade (IPCC 1995), as have been the heat content and surface temperature of the North Pacific Ocean (Levitus *et al.* 2000; Ma *et al.* 1995). However, on both sides of the North Pacific, temperature anomalies are associated with El Niño – for example, sea-surface temperatures in northeastern China are lower during summer in El Niño years (Li 1989), whereas those in the eastern North Pacific are higher (Whitney and Freeland 1999). Similarly, the intrusion of Oyashio waters usually causes abnormally cold summers in the northern areas of Honshu, Japan, which, together with the import of nutrients, influences coastal fisheries (Sawada and Hayakawa 1997; Sekine 1996); and atmospheric warming and cooling drive short-term variation in sea surface temperature in the Japan/East Sea (Chu *et al.* 1998). Sub-decadal signals in ocean temperature like these complicate the determination of any temperature trend associated with greenhouse gas (GHG) warming. Ocean warming and ocean-atmosphere disturbance can cause the large-scale re-distribution of species (see, for example, Di Tullio and Laws 1991; Karl *et al.* 1995; Saar 2000; Schell 2000). Anadromous fish may be exceptionally vulnerable to temperature change because threshold temperatures in rivers, once passed, may eliminate spawning. Model projections warn that temperature may increase sufficiently in the Fraser River within a few decades to put at risk the world's largest wild salmon runs (Morrison *et al.* 2002).

Sea level rise (SLR) threatens all coastlines, but especially those with low gradient, poorly-bonded soils, high human population and land subsidence – conditions that frequently converge in deltas. Records suggest that a SLR of perhaps 10-30 cm has occurred during the past century (Anonymous 2000; Wang 1998) and a further 10-25 cm SLR is projected to occur over the next century. Due to overpumping of groundwater and overloading by construction on deltas, the rate of relative SLR is even higher in critical locations like Tianjin on the old Yellow River Delta (24.5-50.0 mm/yr), the modern river delta (4.5-5.5 mm/yr), and the Shanghai area of the Yangtze River mouth (6.5-11.0 mm/yr) (Wang 1996). Assuming a SLR of 30-100 cm in the next century and accounting for land subsidence, Liu *et al.* (1999) estimate that the coastline of the Bohai Sea will retreat by 50-70 km over the next century, involving a marine transgression of 10,000-11,500 km^2, and perhaps as much as 16,000 km^2 if storm surges are taken into account (Zhang and Wang 1994).

The "aliasing" inherent in natural variability at decadal or longer time scales presents what is probably the greatest challenge to detecting recent trends in the ocean produced by human activities. During the last decade, regime shifts have been recognized as a pervasive manifestation of relatively abrupt physical and biological alterations to the upper Pacific Ocean (Hare and Mantua 2000). For example, a restructuring of the mixed-layer depth in the mid to late 1970s (Fig. 2a) (Freeland *et al.* 1997) must have been accompanied by altered nutrient cycling with 'bottom-up' consequences for the ecosystem. At about the same time, it appears that anadromous fish recruitment was affected, probably due to changes in marine survival (Fig. 2b) (Welch *et al.* 2000). It has recently been recognized that, starting with nutrient supply, a large-scale ecosystem restructuring has occurred in the Bering Sea. The associated change in organic carbon cycling was recorded by Bowhead whale baleen (Fig. 3) (Schell 2000) and other wide-spread systematic changes in food-web dynamics (Hunt *et al.* 1999; Niebauer 1998; Rugh *et al.* 1999; Saar

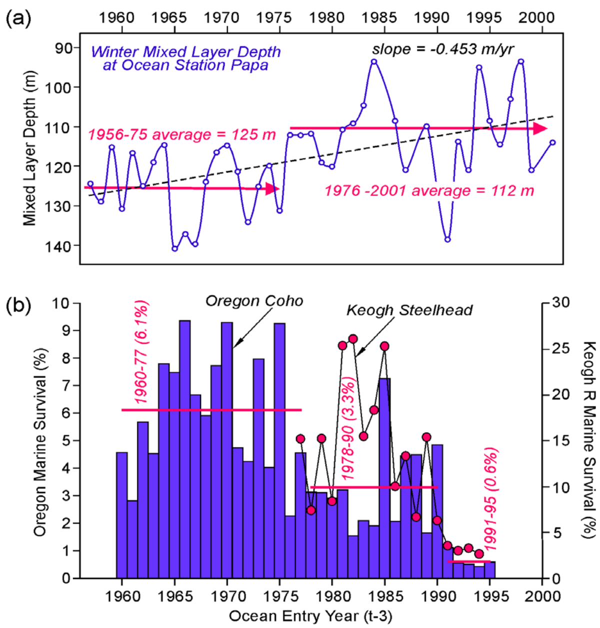

Fig. 2 a) The shallowing of the surface mixed layer at Ocean Station P observed after the regime shift of the mid-1970s (Freeland *et al.* 1997). b) Changes in survival at sea of Oregon and Keogh steelhead between 1960 and 1995, attributed partly to ocean climate conditions (Welch *et al.* 2000).

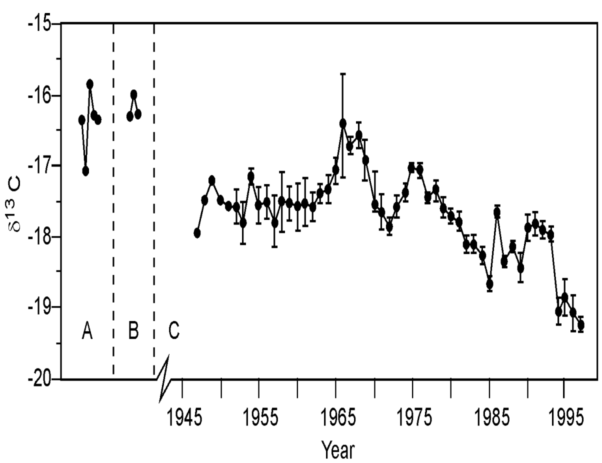

Fig. 3 The change in carbon cycling in the Bering Sea as recorded by δ^{13}C in bowhead whale baleen (Schell 2000).

2000; Stabeno and Overland 2001; Stockwell *et al.* 2000). The wide variety of physical and biological pathways implicated in regime shifts (Hare and Mantua 2000), has significant consequences for the transport and processing of contaminants both regionally and locally – especially for contaminants that concentrate and biomagnify in food-webs (*e.g.*, Hg and organochlorines).

Predation by humans

Over the past decade there has been growing concern that ocean trophic structure can be affected by commercial fisheries. Selective extraction of fish – referred to by Pauly *et al.* (1998; 2001) as 'fishing down the food web' – may lead to a global-scale reduction in marine trophic levels (Fig. 4a), which exerts its influence from the top down (Parsons 1996) (Fig. 4b).

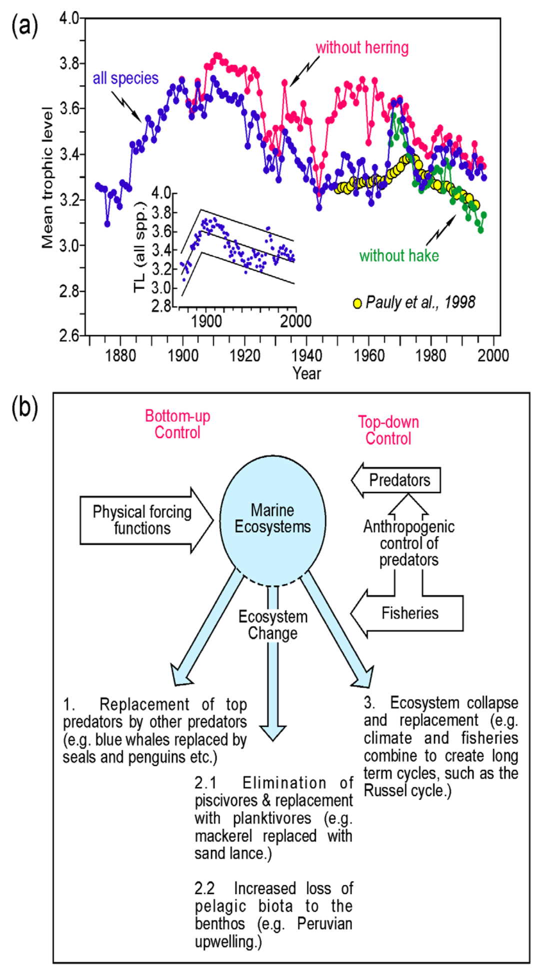

Fig. 4 a) The change in mean trophic level for the Canadian west coast between 1873 and 1996 (Pauly *et. al*. 2001). b) A schemetic showing how trophic structure in aquatic systems can be altered from the bottom-up or from the top-down (Parsons 1992).

All highly-prized species are vulnerable to this impact, but with increasing fishing pressure and dwindling stocks, less desirable species or smaller individuals become targets of commercial and private fisheries, and often, governments prolong un-economical practises which eventually lead to the demise of the resource (Ludwig *et al.* 1993). Destructive fishing methods (blast and cyanide fishing) widely practiced in Asian shelf waters (Morton and Blackmore 2001) exacerbate the problem of over-fishing and undermine the potential for recovery. Driftnet and other "ghost" fisheries have well-known, but perhaps poorly-quantified, effects on non-target species (Dayton 1998).

It is clear that fishing and contamination both provide stresses to coastal ecosystems, but the alteration of trophic structure – either from the top down by fishing, or the bottom up by climate change and coastal eutrophication – has special significance to chemicals that biomagnify (*e.g.*, Hg and organochlorines).

Exotic species

Intentional and unintentional release of non-native species plague coastal seas and freshwater systems. The partial list of introduced species for the Northeast Pacific (Table 1) illustrates the extent of the impact and demonstrates that, as in the case of commercial harvesting, aquatic trophic structure can be altered at almost every level.

Harmful algal blooms

Harmful algal blooms (HABs) pre-date human encroachment on marine systems. However, there is concern that the incidence and severity of HABs have increased due to human activities such as coastal eutrophication and contaminant loading, and global change such as warming (see for example Goldberg 1995; Morton and Blackmore 2001; Waldichuk 1990). HABs can present a risk to fish (*e.g.*, Heterosigma Khan *et al.* 1997) or to humans (*e.g.*, PSP, amnesic poisoning: Horner and Postel 1993; IOC 2000) and no corner of the North Pacific may be considered immune from them (Horner *et al.* 1997; Konovalova 1993; Morton and Blackmore 2001). Although HAB-forming algae are widespread, they may in some circumstances be classified as "exotics" since ship's ballast water can transport them (Hallegraeff 1998). In the context of contaminants, HABs can be considered either as a point of leverage where anthropogenic nutrient and trace metal loadings promote a process that produces toxic compounds, or as exotic species with the potential to alter trophic structure either by direct insertion into the food web or by removal of a trophic component through selective toxicity.

Sediment discharge into coastal water

Some 1 billion tonnes of fine sediments are supplied annually to the eastern coastline of China, brought mainly by the Yellow River from the Loess Plateau as a result of soil erosion from human activities since historical times (Wang 1996). Asian rivers have especially been affected by human activities, modern sediment loads being perhaps five times those prior to the development of agriculture (GESAMP 1993). The consequence of these higher loadings is that affected coastal areas may become overly productive and either hypoxic or anoxic (Goolsby 2000). Enhanced sediment fluxes also provide the means to scavenge and bury particle-reactive contaminants in deltas and on the adjacent continental shelves. Recent damming of the Yellow River, however, appears to have reversed the historical trend (Yang *et al.* 1998), with sediment supply now dwindling. The withdrawal of sediment loading alters the balance between sediment supply, wave re-suspension, and coastal transport, with the potential consequence of accelerating the loss of deltaic areas already threatened by sea-level rise.

Chemical contamination

Hydrocarbons and polynuclear aromatic hydrocarbons

Combustion and petrogenesis are the two major sources of hydrocarbons in the environment, and both can occur either naturally or through human activities (Yunker *et al.* 2000). There have not been any major oil spills in the North Pacific since the *Exxon Valdez* incident in 1989, but the effects of that spill were devastating. Marine organisms

Table 1 Non-native aquatic species present in Washington and British Columbia (source Anonymous, 2001).

Fish	Invertebrates	Aquatic Plants
American shad *Alosa sapidissima*	Varnish clam *Nuttallia obscurata*	Japanese weed *Sargassum muticum*
Grass carp *Ctenopharyngodon idella*	Manila clam *Tapes philippinarum*	Japanese eel grass *Zostera japonica Lomentaria hakodatensis*
Striped bass *Morone saxatilis*	Asian clam *Corbicula fluminea*	Purple Loosestrife *Lythrum salicaria*
Common carp Cyprinus carpio	Soft-shell clam *Mya arenaria*	Brazilian Elodea *Egeria densa*
Goldfish Carassuis auratus	Japanese trapezium *Trapeziium liratum*	Parrotfeather Milfoil *Myriophyllum aquaticum*
Largemouth bass *Micropterus salmoides*	Japanese little neck clam *Venerupis philippinarum*	Fanwort *Cabomba caroliniana*
Smallmouth bass *Micropterus dolomieui*	Pacific oyster *Crassostrea gigas*	Eurasian Watermilfoil *Myriophyllum spicatum*
Bluegill, Green sunfish *Lepomis* spp.	Eastern oyster *Crassostrea virginica*	Hydrilla *Hydrilla verticillata*
Black Crappie *Pomoxis* spp.	Japanese or green mussel *Musculista senhousia*	Spartina/Cordgrasses Spartina alterniflora, anglica, patens
Walleye *Stizostedion vitreum*	Slipper shell *Crepidula fornicata*	Yellow Iris *Iris pseudacorus*
Yellow Perch *Perca flavescens*	Mud snail *Nassariuis obsoletus/Ilyanassa obsoleta*	Agar weed *Gelidium vagum*
Channel Catfish *Ictalurus* spp.	Eastern oyster drill *Urosalpinx cinerea*	
Flathead Catfish *Pylodictis olivaris*	Japanese oyster *drill Ceratostoma inornatum*	
Black Catfish Brown Bullhead *Ictalurus* spp.	Red beard sponge *Microciona prolifera*	
Northern Pike *Esox* spp.	Boring sponge *Cliona* spp.	
Atlantic salmon *Salmo salar*	Bowerbank's halichondria *Halichondria bowerbanki*	
Brown trout *Salmo trutta*	Asian copepod *Pseudodiaptomus inopinus*	
	Bivalve intestinal copepod *Mytilicola orientalis*	
	Mud worm *Polydora ligni*	
	Wood-boring gribble *Limnoria tripunctata*	
	Shipworm *Teredo navalis*	
	Green crab *Carcinus maenas*	

from barnacles to seals were killed, including about 250,000 sea birds (Piatt and Anderson 1996). Long-term effects include depressed populations and the lowered reproductive success of most of the oiled species, although in many cases it is difficult to distinguish between effects of the oil spill and those of decadal-scale environmental change. For example, the unusually low flow of the Alaskan Coastal Current in the years following the spill may have contributed partly to the low murre population during that time (Piatt and Anderson 1996), and the number of seals killed by the spill is disputed, due to limited observations over their natural range (Hoover-Miller *et al.* 2001). A definite link has been made in one case: Brown pelicans that had been oiled, cleaned and released were marked and compared over the course of several years with marked control birds (Anderson *et al.* 1996). The oiled birds disappeared much more quickly than control birds, and they failed to reproduce, whereas the controls continued to behave normally.

Lowered reproductive success of animals that have been exposed to oil is not surprising, given that PAHs are known endocrine-disrupters (Carls *et al.* 1999). Oil from the *Exxon Valdez* spill remained under the stones and mussel beds of nearby beaches five years after the spill (Spies *et al.* 1996), although the sediments of the intertidal zone had lost their toxicity to oysters and amphipods after two years (Wolfe *et al.* 1996).

Increasing pressure to find oil on continental shelves will probably increase the risk of hydrocarbon pollution to the North Pacific: Canada (British Columbia), the U.S.A. (California), Republic of Korea and Japan have all indicated that they intend either to begin or to expand exploration on the continental shelves of the Pacific, and drilling already occurs off Alaska and California and in the East China Sea. The environmental risks posed by offshore exploration and production are well known. They include the loss of hydrocarbons to the environment, smothering of benthos, sediment anoxia, destruction of benthic habitat, and the use of explosives (Patin 1999). Oil released from offshore operations may contain other harmful components like the endocrine-disrupting alkylphenols (Lye 2000). The generally high seismic activity of the Pacific Rim may further enhance the risk of spills (for comments regarding the South China Sea, see Zhang 1994).

Despite the high media and public interest in catastrophic oil releases, the predominant sources of hydrocarbons to coastal seas are either land based (*via* rivers) or derive from intense shipping activity as exemplified by studies in Peter the Great Bay (Nemirovskaya 1999), around Vladivostock on the Russian coast of the Sea of Japan (Tkalin 1992), and the Georgia Basin of the British Columbia coast (Yunker *et al.* 2000).

Halogenated hydrocarbons

Organochlorine compounds (OCs) have been released to the global environment in a number of ways, including industrial applications (*e.g.*, PCB), incineration (*e.g.*, dioxins, furans), chlorination in pulp mills (dioxins, furans, PCBs) and pesticide application (*e.g.*, DDT, HCH, chlordane). As a result, for any coastal sea in the North Pacific there will be a long-range, global source component for these compounds which is then augmented to a lesser or greater degree by local sources, either through the air or through runoff. Waldichuk (1990) noted that winds in the North Pacific would tend to transport volatile contaminants from Asia eastward to North America. Recent work has clarified this general transport scheme and provided further evidence of its efficacy in spreading volatile and particulate contaminants from Asia across the ocean to North America (Fig. 5) (Bailey *et al.* 2000; Jaffe *et al.* 1997; Li *et al.* 2002; Macdonald *et al.* 2000a; Wilkening *et al.* 2000).

Despite bans or restrictions during the 1970s and 1980s in most of the countries surrounding the North Pacific, PCBs, DDT and other organochlorine pesticides remain in soils and in aquatic environments. In the latter, they biomagnify to especially high concentrations in apex feeders such as marine mammals (Muir *et al.* 1999; Ross *et al.* 2000). In the early years following bans, the concentrations of PCBs and DDT decreased rapidly in the Pacific Ocean (Waldichuk 1990), but that seems no longer to be universally true. For example, between the late 1970s and early 1990s, there has been no trend in PCB concentration in particulate and dissolved fractions of San Francisco estuary water (Jarman *et al.* 1996). According to Iwata *et al.* (1994a), the concentrations of DDT, PCBs, HCH and HCB (hexachlorobenzene) have not been decreasing rapidly in the Bering Sea, because atmospheric deposition exceeds the sedimentation rate. However, decreased atmospheric concentrations of HCH following the elimination of technical HCH use in China and India during the 1980s and 1990s (Li *et al.* 2002), have reversed the net exchange of α-HCH, such that the Bering Sea has now become a source to the atmosphere (Jantunen and Bidleman 1995). The long-range atmospheric and/or oceanic transport of HCHs together with large changes in emissions have made them (*i.e.* α-, γ-, β-HCH) useful tracers of transport processes in the Bering Sea and into the Arctic Ocean (Li *et al.* 2002; Rice and Shigaev 1997).

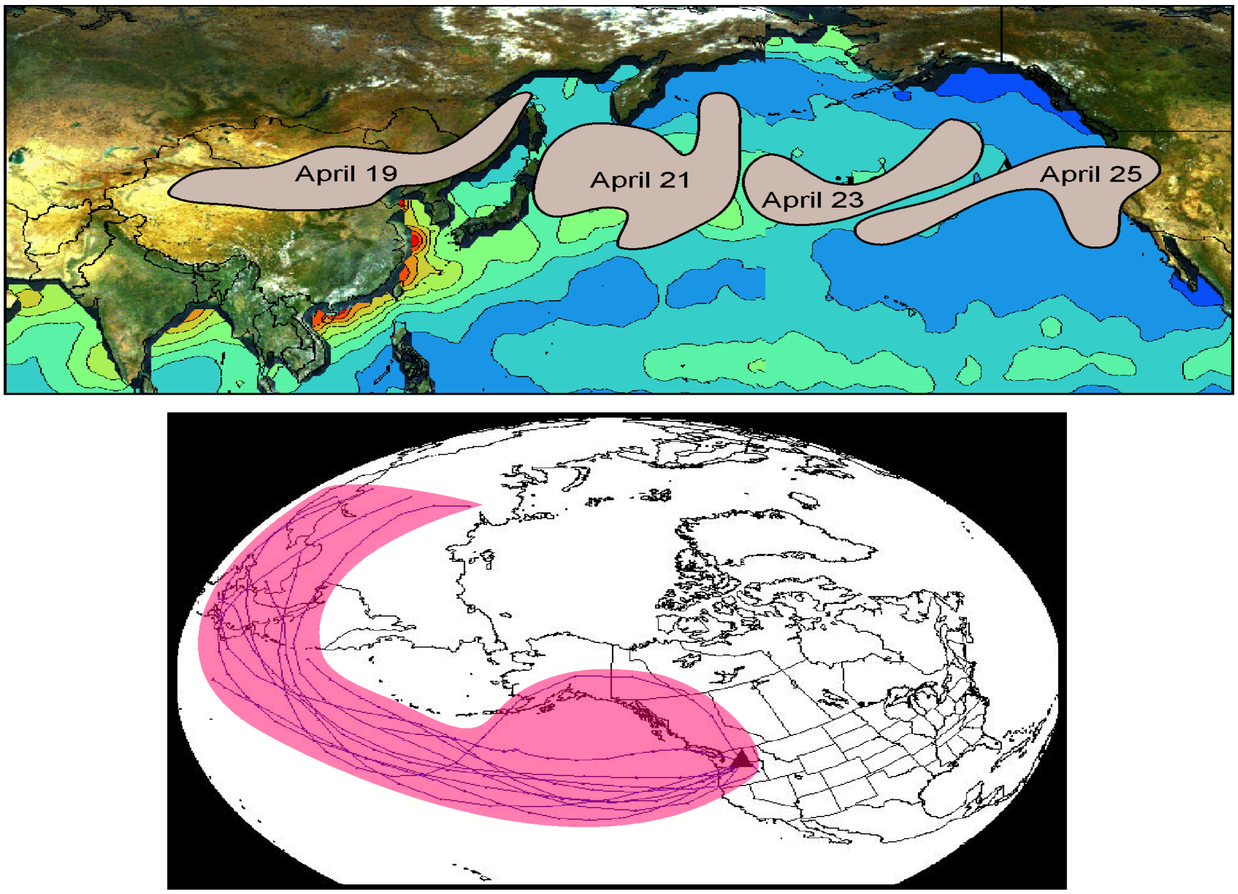

Fig. 5 Trans-Pacific atmospheric transport from Asia to North America as shown by a) dust from the Gobi Desert (Wilkening *et al.* 2000), and b) back trajectories from an air monitoring site in Canada's Yukon Territory (Bailey 2000 # 179).

The Bering Sea illustrates that long-range transport together with physical and biological processes (Chernyak *et al.* 1996; Hoekstra *et al.* 2002; Sokolova *et al.* 1995) can produce significant concentrations of pesticides in apex feeders far from local sources. Furthermore, the animals themselves may then become significant vectors of OC contaminant transport as exemplified by anadromous fish in Alaska (Ewald *et al.* 1998).

Local sources of OCs support high sediment concentrations in some locations. For example, DDT and HCH in the sediments of Peter the Great Bay probably reflect a continuing local source of those contaminants near Vladivostok (Tkalin *et al.* 2000), and high concentrations of DDT, DDD and DDE in the sediments of Lianyungang Harbour in China suggests continuing use of these compounds in local agriculture (Zhu and Tkalin 1994). There appear to be a number of local sources of DDT in some areas of the South China Sea (probably local industry or illegal dumping), as evidenced by variability in the ratio of DDE/ ΣDDT in the sediments (Morton and Blackmore 2001).

Marine mammals are particularly vulnerable to OCs due to biomagnification. Whales (Aono *et al.* 1997; Hayteas and Duffield 2000; Jarman *et al.* 1996; Ross *et al.* 2000), dolphins (Jarman *et al.* 1996; Minh *et al.* 2000), porpoises (Jarman *et al.* 1996; Minh *et al.* 2000; Zhou *et al.* 1993), seals (Nakata *et al.* 1998), sea lions (Lee *et al.* 1996) and humans (Morton and Blackmore 2001) are all contaminated. The degree of contamination and specific pollutants in each case depend on geographic location and trophic level.

Temporal trends in the concentration of organochlorines in marine mammals also vary among species and with organochlorine type. In Minke whales, the concentration of DDT is decreasing, while PCB concentration is not, suggesting a continuing source of them to the North Pacific (Aono *et al.* 1997). The concentration of PCBs and other organochlorines in Killer whales varies with age and gender (Ross *et al.* 2000) and, off the coast of British Columbia and California, is generally higher than in dolphins and porpoises (Jarman *et al.* 1996). British Columbia Killer whales (and seals) exhibit high concentrations of dioxins and furans (Jarman *et al.* 1996; Ross *et al.* 2000), but for these compounds, local sources (pulp mills) have clearly made a substantial contribution (Bright *et al.* 1999; Macdonald *et al.* 1992). Elimination of chlorine bleach and pentachlorophenol- (PCP) contaminated feed stock after 1987 has led to substantial declines in PCDD/F concentrations in sediments and crabs (Yunker and Cretney 1996) and in seals (Fig. 6). Source controls, which have all but eliminated the pulp mill PCDD/Fs, however, have made no inroads on the PCBs which derive predominantly from other sources – local, regional and global (Addison and Ross 2000).

Juvenile Pacific salmon accumulate immunosuppressive OCs as they develop in estuaries (Arkoosh *et al.* 1998), which may make them especially susceptible to the pathogens common in these environments. Sea birds are similarly affected. At Port Alberni, on the west coast of Vancouver Island, Canada, fish-eating grebe and seaduck were heavily contaminated with dioxins and furans from a nearby pulp mill (Elliott and Martin 1998). Those compounds also present the main pollution threat to Marbled murrelets along the British Columbia, Washington and California coasts. Eggshell thinning due to organochlorine pesticides is no longer considered a threat to seabirds off California (Pyle *et al.* 1999), and in herons the threat is mainly restricted to those that live near agricultural areas (Speich *et al.* 1992). Amphipods, sea urchins, bioluminescent microbes (Long 2000) and squid (Shibata, pers. comm.; Sato *et al.* 2000) also accumulate OCs although toxic effects are as yet unclear. Oysters off Taiwan are so contaminated with DDT that there is a high lifetime risk of cancer for people who consume them (Han *et al.* 2000). The concentrations of HCH and PCB ($Cl_{5\text{-}9}$) in squid livers correlate well with those in nearby sea water, lagged by 1-2 years, suggesting a reasonably dynamic equilibrium rather than progressive accumulation with age (Sato *et al.* 2000).

Metals

The waters off Hong Kong (Parsons 1998) and the sediments of the Japan/East Sea (Shulkin and Bogdanova 1998; Vaschenko *et al.* 1999), the South China Sea (Morton and Blackmore 2001)

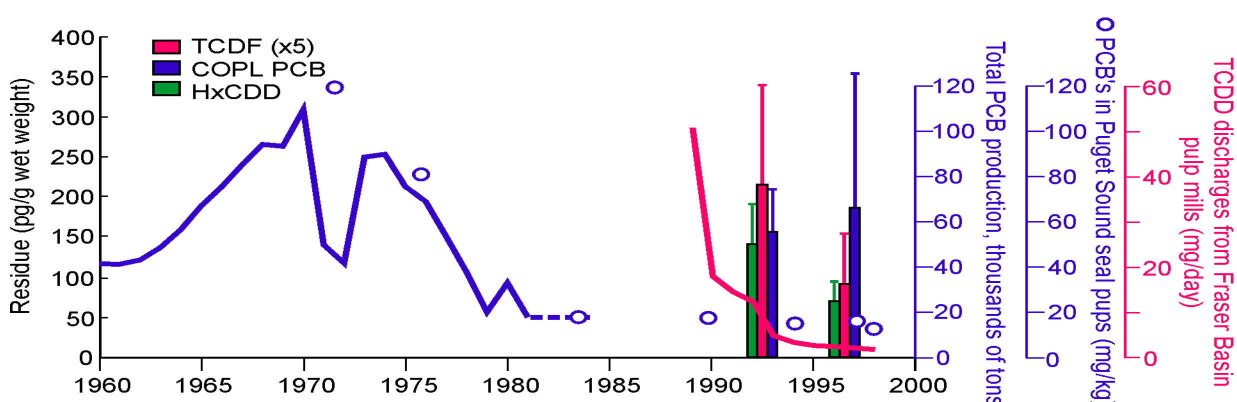

Fig. 6 An example of trend data from the west coast of North America showing PCB production (solid blue line) and TCDF discharge from Fraser basin pulp mills (solid red line) compared with residues in Harbour seals (Addison and Ross 2000) and in Harbour seal pups (blue circles, Calambokidis *et al.* 1991).

and the Yellow Sea (Shao *et al.* 1995; Yu *et al.* 1996; Zhang *et al.* 1998) are highly contaminated with metals, especially near farmed shrimp ponds (Cui *et al.* 1997), and the problem is increasing. Long-range transport of some metals (Pb: Lin *et al.* 2000; Cd: Patterson and Duce 1991*)* from Asia has been observed in the North Pacific, and some particulate trace metals cross the shelf from the East China Sea to enrich the intermediate layer of the Kuroshio Current (Hung and Chan 1998). But the main effect of metal pollution remains close to the source. A pronounced increase in anthropogenic lead loading to the Yangtze River during the 1980s and 1990s has been inferred from sediment cores collected from the East China Sea continental shelf (Huh and Chen 1999). These trends probably reflect the rapid economic growth and the lack of waste control in China. Contaminant metals in the marginal seas derive mainly from untreated sewage and industrial wastes (Parsons 1998; Shao *et al.* 1995) that either washes off the shore or enters rivers. Resuspension (Fichet *et al.* 1998) and deposition of dissolved and particulate matter by rain (Gao *et al.* 1997; Zhang *et al.* 1999; Zhang *et al.* 1992) are also major sources of metal pollution in Asian marginal seas.

As a consequence of the sediment and water pollution, much of the marine life in Asian marginal seas exhibits metal contamination. In Zhifu Bay in the Yellow Sea, for example, increased benthic pollution between 1986 and 1998 caused a change in dominant species from non-pollutant-resistant echinoderms to pollutant-resistant polychaetes (Zhang *et al.* 1998). Oysters (Cheung and Wong 1992; Han *et al.* 2000), scallops (Vaschenko *et al.* 1999) and fish (Parsons 1998) are also contaminated.

The shelves of the northwest coast of North America appear to be almost pristine compared to Asia (Macdonald and Pedersen 1991; Naidu *et al.* 1997). However, metal contamination can certainly be identified in enclosed embayments (Flegal and Sañudo-Wilhelmy 1993; Macdonald and Crecelius 1994; Paulson *et al.* 1993; Sañudo-Wilhelmy and Flegal 1992).

Of the metals, mercury and tributyltin (TBT) cause particular concern due to their toxicity and endocrine-disrupting characteristics. Furthermore, mercury biomagnifies by factors of 1000-3000 from particulate organic matter to apex predators (Atwell *et al.* 1998), and its rate of cycling in the global environment appears to have increased by perhaps a factor of three since pre-industrial times (Mason *et al.* 1994). Mercury, therefore, provides a problem not unlike that of the OCs, in that the upper ocean has globally-enhanced mercury concentrations (Mason *et al.* 1994), which are then augmented locally (usually from land). Furthermore, enhanced global cycling together with biomagnification can create biotransport vectors (Zhang *et al.* 2001) as was shown for OCs (Ewald *et al.* 1998).

Symptoms of Minimata disease were detected during the 1980s in fishermen who relied heavily on fish from the Songhua River in China, a river that had been polluted by mercury in the 1970s (Gao *et al.* 1991). The intake of methyl mercury was estimated to be between 0.17-0.34 mg/day and the average mercury contents of the hair and urine were 13-58 and 10-33 times higher than normal, respectively. In the Japan/East Sea the concentration of mercury is increasing in water, sediments and the tissues of molluscs (Luchsheva 1995). In the most affected area in Alekseev Bight in Peter the Great Bay, the concentration of mercury in molluscs exceeds pollution guidelines. Indo-Pacific humpbacked dolphins off the coast of Hong Kong also contain dangerous levels of mercury (Parsons 1998). On the eastern side of the Pacific, problems arose during the 1960s, but the sources of mercury have since been controlled (Waldichuk 1990). This is reflected in the sedimentary records of the Strait of Georgia and Puget Sound, Washington (Macdonald and Crecelius 1994). The common sources of mercury (*e.g.*, dental and medical offices, light industry (Nriagu and Pacyna 1988)) imply that municipal outfalls are probably important local conduits for this metal to coastal environments.

Tributyltin is prevalent in the sediments, water and biota in the North Pacific and the South China Sea (Iwata *et al.* 1994b; Morton and Blackmore 2001) but it is the manifestation of imposex in shellfish at extraordinarily low TBT concentrations (< 0.5 ng l^{-1}, Ronis and Mason 1996) that has engendered the greatest concern in the literature.

In 1989, the use of TBT to prevent biofouling on hulls was restricted to ships > 25 m long; TBT is now found mainly in heavily-used ports, especially those with dry-dock facilities (Evans *et al.* 1995; Morton and Blackmore 2001). In two such areas - the Strait of Malacca and Tokyo Bay - the concentration of TBT in seawater is high enough to cause imposex in gastropods and damage to other marine life (Hashimoto *et al.* 1998). In areas with less ship traffic, TBT restrictions have been successful at reducing imposex in gastropods and shell-thickening in oysters (Evans *et al.* 1995).

In British Columbia's coastal waters there is some evidence that the gastropod population in the Strait of Georgia is recovering since TBT restrictions have been implemented (Tester *et al.* 1996). However, imposex in female whelks continues near Victoria, and in Vancouver Harbour there are still no animals to study. Although TBT is highly toxic, its use persists on large ships (and probably illegally on many smaller boats) because of its effectiveness and the tremendous saving in fuel that it allows (Morton and Blackmore 2001). A related compound, triphenyltin (TPT), has been detected in water and mussels from Osaka Bay, but levels appear to have declined between 1989 and 1996 (Harino *et al.* 1999).

Other bioactive metals may threaten marine life (Bruland *et al.* 1991; Waldichuk 1990). Manganese and copper have been reported in snow geese off British Columbia and California (Hui *et al.* 1998). Manganese can cause neurological damage in seabirds and copper can cause anemia (Hui *et al.* 1998). The birds that feed off agricultural land in California are more contaminated than those that feed on British Columbia's pastures and marshes, probably because agricultural fungicides and fertilizers contain both metals (Hui *et al.* 1998). Edible seaweed in British Columbia and Japan is contaminated with arsenic, but the human health risk is unknown, because its bioavailability in seaweed has not been determined (van Netten *et al.* 2000).

The Bering Sea is less polluted with metals than are Asian marginal seas and the coast of North America. In contrast to the Yangtze River that feeds the East China Sea, the Anadyr River, the second-largest to flow into the Bering Sea, is not measurably contaminated with either metals or radionuclides (Alexander and Windom 1999). The concentrations of zinc, copper, cadmium and lead in Bering Sea fish (pollock, hake, whiting and mackerel) are low (Polak-Juszczak and Domagala 1993) as are the concentrations of such metals in sediments (Naidu *et al.* 1997).

Placer mining, tailings disposal and the collection of polymetallic nodules from the deep sea are likely to be sources of contaminant metals into the future. Placer gold mining in Norton Sound in the northeastern Bering Sea from 1986 to 1990 appears not to have affected the concentration of metals in King crabs because they were only in the area in the winter, which was the off-season for mining (Jewett 1999; Jewett *et al.* 1999; Jewett and Naidu 2000).

The Rudnayu River discharges mining wastes into the Japan/East Sea from Russia contaminating coastal sediments with lead, cadmium, copper and zinc in a 25 km long plume southward of the river's estuary (Shulkin and Bogdanova 1998). Disposal of metal-rich mine tailings in coastal fiords of British Columbia creates a combined impact from smothering and metal contamination, which may persist for decades due to instability of sub-sea tailings deposits (Burd *et al.* 2000).

The technology for mining polymetallic nodules and crusts in the Pacific Ocean has advanced sufficiently to allow serious prospecting for large-scale mining by Japan (Nakao 1995) and China (Xu *et al.* 1994). Deep-sea mining of nodules would bring with it the risks of physical disruption to benthic habitats, spills of toxic leaching fluids and smothering by sediment plumes and degradable organic matter (Ahnert and Borowski 2000).

Radionuclides

Waldichuk's (1990) conclusion, that artificial radionuclides from atmospheric weapons testing posed little risk to marine environments in 1990, can be repeated with the comment that radio-decay will have further reduced inventories of the predominant radioactive contributors (^{137}Cs, ^{90}Sr –

$t_{½}$ ~30 years) by 20% over the past decade. However, in the early 1990s, it was revealed that the former Soviet Union had disposed of liquid and solid radioactive wastes at a number of sites including the Northwest Pacific (Yablokov 2001). Extensive studies during the 1990s concluded that, despite the size of the releases both in the Arctic and North Pacific, there was actually little radiological risk (Layton *et al.* 1997). For example, Hong *et al.* (1999b) reported that the concentration of $^{239,\ 240}Pu$ in zooplankton in the Bering Sea was similar to that of zooplankton found in the rest of the Pacific Ocean and represented long-range transport of radionuclides. In the Sea of Okhotsk, as of 1995, most of the ^{90}Sr, ^{137}Cs and $^{138,139,140}Pu$ was still in the water column (Pettersson *et al.* 2000). The concentration of these elements was consistent with previous measurements, but the total inventory in water and sediments represented more radionuclides than expected from global fallout (Pettersson *et al.* 2000). Measurements of $^{239,\ 240}Pu$ and ^{137}Cs in fish, shellfish, cephalopods, crustaceans and algae in the Japan/East Sea and off the Pacific coast of Japan showed no evidence of pollution from dumping by Russia or the former U.S.S.R. (Yamada *et al.* 1999), even immediately after 14GBq of liquid radioactive waste was dumped into the Japan/East Sea in October 1993 (Hong *et al.* 1999a). The ratio of $^{239}Pu/^{240}Pu$ in the sediments was consistent with global fallout (Yamada *et al.* 1999).

Persistent solids

Waldichuk (1990) reported that entanglement by plastic driftnets, other fishing gear and other plastic objects, such as grocery bags, was estimated to be responsible for killing two million sea birds and 100,000 marine mammals each year. Entanglement was considered to be a particularly significant problem for endangered species. There have not been many studies on the prevalence and effect of plastics in the North Pacific in the last ten years, but the research that is available supports the seriousness of the problem and demonstrates that plastics affect different species to different degrees. Sea birds (Blight and Burger 1997; Robards *et al.* 1995) are particularly strongly affected, since they tend either to ingest the plastic or become entangled by it. Benthic communities can be smothered by the plastics, which are slow to break down (Uneputty and Evans 1997). California sea lions, however, although many of them do become entangled in plastic, are seven times more likely to be shot than entangled, according to data from a rehabilitation centre in California (Goldstein *et al.* 1999).

Domestic pollution

Domestic pollution consists of sewage and some industrial wastes that end up in the municipal treatment system (from hospitals, dentists, photographic processors and other industries). Many of the industrial wastes are toxic, and some bioaccumulate or biomagnify. Nutrients from sewage can cause eutrophication, bacterial pollution and harmful algal blooms, whereas other components are known to disrupt endocrine processes (Goldberg 1995; Kramer and Giesy 1995; Shang *et al.* 1999). Waldichuk (1990) described sewage-related problems in coastal British Columbia and commented that the situation was worse in Asia, where there was a much larger human population. The impact of sewage discharge is site-specific, depending on, among other things, cumulative loadings, rate of coastal flushing and mechanism of discharge (*e.g.*, deep, shallow, diffuse). In western North America, untreated and secondarily-treated sewage is still discharged to coastal waters by some cities (*e.g.*, Victoria and Vancouver) (Thomson *et al.* 1995), but upgrades are proceeding in many areas, and it seems likely that the impact of municipal outfalls on shallow coastal waters has been declining despite population increases. Widely-distributed poorly-maintained septic systems continue to contaminate shorelines in many places, however.

In the Asian marginal seas, domestic pollution is especially severe: less than 10% of China's domestic and industrial waste is treated before it flows into rivers or the ocean (Morton and Blackmore 2001). The degree of nutrient pollution and eutrophication varies geographically (Ma *et al.* 1997). In the Japan/East Sea, between 1982 and 1995, domestic pollution of water and sediment increased, changing the availability of a substrate for barnacle larvae to settle on and causing an increased mortality of young barnacles and decreased growth rate where the temperature had risen above 18°C and dissolved oxygen concentrations were critically low (Silina and Ovsyannikova 2000).

In Tokyo Bay, organic pollution is so severe that benthic organisms decline in summer when a thermocline is formed in the water column (Hisano and Hayase 1991). Over a period of 15 and 18 years respectively, Hirota (1979) and others (Anakubo and Murano 1991; Nishida, 1985; Nomura and Murano 1992; Uye 1994) recorded that for the Seto Inland Sea and Tokyo Bay, Japan, as eutrophication problems grew, there were zooplankton community structure shifts from a calanoid copepod to a cyclopoid-dominated one. In Tokyo Bay, the copepod community became dominated by *Oithona davisae*. These authors also recorded a shift in phytoplankton community structure towards small dinoflagellates and diatoms. Pollution thus seems to favour dinoflagellate feeders, such as *O. davisae*. Furthermore, the anoxic bottom-water formed in Tokyo Bay from organic enrichment and stratification acts selectively to advantage or disadvantage plankton life cycles. Copepod eggs that are spawned freely into the water column may sink onto the seabed where they are adversely affected by oxygen-deficient water, resulting in heavy recruitment loss. Inseminated *O. davisae*, however, which carries its eggs in egg sacs, can complete its life cycle by avoiding anoxic habitats. Recruitment of egg-carrying copepods would thus be favoured and *O. davisae* comes to dominate the resident community. Formation of oxygen deficient bottom water might also be detrimental to copepods with no flexible vertical distribution. For example, male *Parvocalanus crassiroustris* and species of Acartia remain in deeper waters, especially late in the day (Ueda 1987).

In the Yellow Sea the concentration of inorganic phosphorus is increasing (Ma *et al.* 1997), and eutrophication is thought to be responsible for more frequent HABs (Jiao and Guo 1996; You *et al.* 1994). In the East China Sea, human deaths have resulted from ingestion of toxic bivalves and gastropods; the HABs responsible for the toxicity of the shellfish are thought to have been caused by eutrophication in combination with physical processes, including coastal upwelling and climate events (Chen and Gu 1993).

Twenty to fifty percent of the "new" nitrogen in the Yellow Sea comes from atmospheric deposition and groundwater (Paerl 1997). Urban and agricultural discharges to groundwater are increasing (Paerl 1997), and rain over the Yellow Sea has a high concentration of nutrients from air pollution (Zhang *et al.* 1999). Groundwater and rain bypass the estuarine filters and can cause eutrophication and HABs at a considerable distance from the source. Atmospheric deposition of nitrate varies seasonally, with higher concentrations in the winter, when there is less precipitation (Zhang and Liu 1994); the episodicity of the high atmospheric delivery of nutrients corresponds with HABs in the nearby Pacific Ocean (Zhang 1994; Zhang and Liu 1994).

Due to increasing population and a relatively small land base, Korean bays have become sinks for a variety of domestic and industrial wastes. In Chinhae Bay, oxygen deficient conditions due to organic pollution perturbated the resident marine benthic communities in 1989 (Lim and Hong 1994; Yang 1991). In the early 1990s, Shihwa Lake was formed by impounding a marine bay on the west coast of Korea with a 12.7 km long barrier. The bay, which became stratified by salt and temperature, then went eutrophic and the sea bed became anoxic. Sea bed levels of nutrients and industrial wastes increased and macrobenthic diversity collapsed with blooms of *Polydora ligni* and *Capitella capitata* in winter (Lee and Cha 1997).

Aquaculture, a source of organic carbon, nutrients, and industrial chemicals (antifoulants, pharmaceuticals, contaminants in feedstock), is an expanding industry. Although total amounts of materials from any one operation may be small, there is the potential for impacts close to the operation and, with sufficient density in poorly-flushed coastal waters, there could be regional impacts. For example, fish culture in meshed cages in a bay in southern Japan resulted in an azoic sea bed with summer defaunation followed by recolonization the following spring (Tsutsumi 1995). Molluscs were progressively replaced by polychaetes as the dominant macrobenthos below the cages.

Components of a warning system

The multiple stresses briefly reviewed here provide an enormous and increasing challenge to North Pacific coasts and shelves. These stresses do not operate independently but, rather, interact with one another in a manner that varies among locations (Fig. 7). If we survey the Pacific Rim, we see that the Asian coasts are most immediately threatened on a large scale by over-fishing, destructive fishing practices, nutrient loadings and inputs of contaminants from large populations undergoing industrial transition (Morton and Blackmore 2001). To the far north, local sources dwindle in importance and climate change and long-range transport of contaminants become leading causes for concern (Alexander and Windom 1999; Rice and Shigaev 1997; Shaporenko 1997; Vaschenko 2000). Finally, for the temperate west coast of North America, climate change, over-fishing and long-range contaminant transport remain important issues, with contaminant loadings to enclosed seas (Strait of Georgia, Puget Sound, San Francisco Bay) assuming high local profiles (see for example, Macdonald and Crecelius 1994; Parsons 1996; Ross *et al.* 2000; Sañudo-Wilhelmy and Flegal 1992).

The challenge that ocean scientists must meet if we are to avert the demise of coastal ecosystems is: (1) to produce observations that forewarn us (trends); (2) to understand ocean processes sufficiently to associate ecosystem response with cause (human or natural) and; (3) to assign the order of importance of stresses put upon coastal seas by human activities. Clearly, for this scientific effort to be of benefit it must be translated into action, for example, either to reduce or to eliminate contaminants at local, regional and international scales. One strategy widely promoted to conserve biological resources is the development of a network of Marine Protected Areas (MPAs). If carefully chosen, MPAs provide undisputed benefits for conservation (Roberts *et al*. 2001). However, they allow us little room for complacency as they provide no protection against coastal eutrophication, chemical contamination, introduction of exotic species, over-harvesting of free-ranging biota, or climate change – the

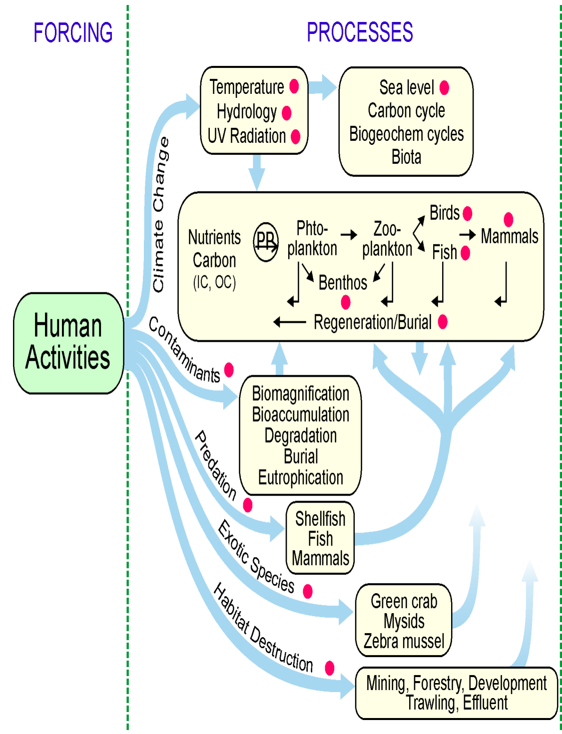

Fig. 7 A schematic diagram illustrating how human activities may affect marine ecosystems at multiple points. Points at which the marine systems are monitored for contaminants (dots) may be influenced by other confounding factors.

majority of the stresses that threaten our oceans (Fig. 7). To recognize and prioritize threats to coasts from these disparate stressors will require coherent observations and research, which we suggest should include at least the following elements.

Box models, other models and case studies

Box models are especially useful in enclosed seas where inputs and outputs can in principle be tightly constrained, but they may also be applied to open shelves (Chen *et al*, 2002; Johannessen *et al.* 2002; Liu *et al.* 2000). Beginning with water, salt, and nutrients (Gordon *et al.* 1996), budgets can be scaled up to include sediments, organic carbon and contaminants. These budgets then allow a preliminary assessment of human loadings compared with fluxes and budgets in the undisturbed system. From such an assessment, an estimation can be made of the likely scale (local, regional) of impact, and human loadings can be ranked to allow for a logical approach to mitigation. An example from the Seto Sea (Fig. 8) illustrates that human loadings dominate the zinc and copper budgets and that most of the contaminant load of these metals ends up in its sediments. Box models provide a schematic understanding, which can help to validate the output of more sophisticated ones.

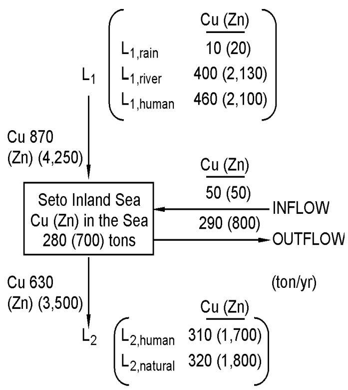

Fig. 8 An example of the application of a box model to an enclosed sea. Mass balances are given for copper and zinc in the Seto Island Sea (tons/yr). L1 identifies load into the sea, L2 identifies load into sediment, and zinc loadings are given in parentheses (Hoshika *et al.* 1988).

Box models provide a solid foundation upon which to build case studies (Macdonald *et al.* 2000b). Case studies can be applied to a relatively constrained environmental impact such as the disposal of mine tailings to a coastal fjord (Ellis *et al.* 1995), to a specific chemical like PCB, HCH or toxaphene (Macdonald *et al.* 2000b) and may provide the basis to initiate appropriate environmental action (Lindstrom and Renescu 1994).

Time-series observations

The observation of change is one of the most powerful means to initiate action. The difficulty, however, is to recognize it early enough to avert irreversible change, and to be able to draw clear inference from observations to causes so that appropriate action can be taken. As shown in Figure 7 (dots), time series can be assembled at many points of the ocean system. However, the meaning of such time series varies from point to point and, in many cases, multiple components of change act simultaneously, so that a simple observation (declining PCB levels with time) can be produced by more than one factor (*e.g.*, reduced global emission, reduced local emission, change of atmospheric or ocean pathway through regime shift, changes to the food web structure (top-down or bottom-up)). Generally, time series have been collected *ad hoc* without worrying about confounding factors or comparability with other time series. It is time for the scientific community to develop coherent, intercomparable time series of sufficient sophistication to guide administrators toward appropriate action.

Sediment cores

Sediments provide well-recognized archives of the history of particle reactive contaminants and, as such, will remain a key resource to understanding current loadings of contaminants in the context of pre-industrial loadings (see, for example, Huh and Chen 1999; Macdonald and Crecelius 1994). Finney *et al.* (2000) have demonstrated elegantly that in certain circumstances, sediment can record both the forcing (anadromous fish return) and effects (lake eutrophication), allowing a more secure inference of how climate change and human predation work together to affect fish escapement. Such insights are not available in the instrumental observation record. This study certainly points the way to more powerful application of sediment cores to sort out combined stresses; for example, the findings of Finney *et al.* (2000) could be further expanded to consider the effects of fish on lake contaminants and, potentially, the effects of contaminants on fish (see, for example, Ewald *et al.* 1998; Zhang *et al.* 2001).

Monitoring components of the food web

A food web provides an enormous scope for monitoring, from filter feeders (Beliaeff *et al.* 1997) to apex predators (Addison and Smith 1998; Ross *et al.* 2000) to HABs (Yanagi 1988). Presently, time series data for any component of the food web in the North Pacific are extremely rare, and where there is such information, it often comprises few time series points, several or more years apart, and well after contaminants began to be released into the environment (for example see Fig. 6). It is now recognized that contaminant burdens recorded by aquatic animals depend on their life histories and cycles they may exhibit (age, sex, size, season, prey). With research, many of these factors can be taken into account through, for example, sampling strategy. However, the food web itself is a dynamic system (Fig. 7) subject to alteration in a number of ways, as discussed earlier.

The problem with monitoring individual components in the food web, therefore, is that a change in contaminant burden with time may have non-unique causes. For example, a shift in a single trophic level produced by over fishing or eutrophication can produce a change in mercury concentration by a factor of 10 (Fig. 9). The same problem exists for the organochlorines, which also biomagnify. In the latter context, a particularly apt example was provided in the Great Lakes where the invasion of an exotic species, the zebra mussel (*Dreissena polymporpha*), led to a fundamental change in lake trophic structure and, presumably, to contaminant pathways (Morrison *et al.* 1998; Whittle *et al.* 2000). Given the varied pressures on the aquatic food webs of the North Pacific reviewed here, it seems clear that we need to institute a monitoring programme that incorporates all trophic levels. Furthermore, support data (stable isotope composition) must be assembled to help interpret changes in trophic level together with changes in contaminant burdens.

Sample archives

Tissue archives provide a safety net for ongoing monitoring. We cannot hope to anticipate all future chemicals, nor can we predict accurately the sorts of changes that might occur in our ecosystems. We can be sure, however, that new and better techniques will be developed with time to apply to problems of chemical contamination and ecosystem change. For example, the change in trophic structure due to zebra mussel invasion of the Great Lakes would not have been identified without such archives (Kiriluk *et al.* 1999), nor would the relationship between this change and

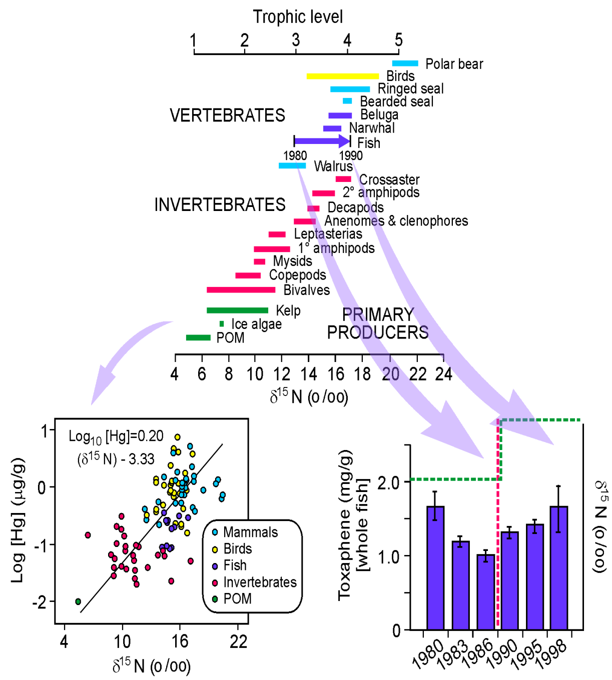

Fig. 9 A schematic diagram showing trophic organization of a marine food web based on $\delta^{15}N$ measurements (top panel, Hobson and Welch 1992), and how biomagnification increases mercury concentration as trophic level is increased (bottom left, Atwell *et al.* 1998). Alteration of the food web resulting in, for example, a change in the trophic level of fish can accordingly alter contaminant burdens observed in time series (after Whittle *et al.* 2000).

contaminant burdens. It is astonishing to note that the tissue archive applied to understanding what had occurred in this system was maintained unofficially with soft funding. Recognizing the importance of tissue archives, we should institutionalize immediately the collection, cataloguing and storage of appropriate samples.

Ecological indicators

Monitoring environmental quality using chemical measurements tends to be prejudiced either toward chemical analyses for which we have developed skill, or toward chemicals known to cause environmental problems (the usual suspects). As a consequence, one detects only those chemicals that have been sought, and unexpected chemicals are likely to go unrecognized. Biological measurements are required, therefore, to alert us to the presence of unidentified chemicals that require the development of new analytical methods (the research to isolate and identify domoic acid, following shellfish poisoning of humans on Canada's East Coast provides an excellent example (Addison and Stewart 1989)). Although marine pollution is often presented as a chemical problem, our ultimate interest is not in the chemicals themselves, nor of their burdens in environmental media. Rather, we would like to be able to relate chemical loadings to harmful effects on the structure and functioning of ecosystems (Addison 1996). To do this requires the development of ecological indicators, the science of which is in its infancy. The difficulty we face is that many of the relatively simple and affordable measurements (*e.g.*, PCB burden in seal blubber) cannot be related confidently to animal health, and even less so to population health (Fig. 10), even though we suspect that certain clinical toxic thresholds may have been exceeded. On the other hand, monitoring community structure and relating changes to chemical and other stresses is not only beyond our present understanding but also beyond our financial means. A crucial task remains therefore, to develop indicators that exhibit reliability, robustness and specificity but which also are affordable.

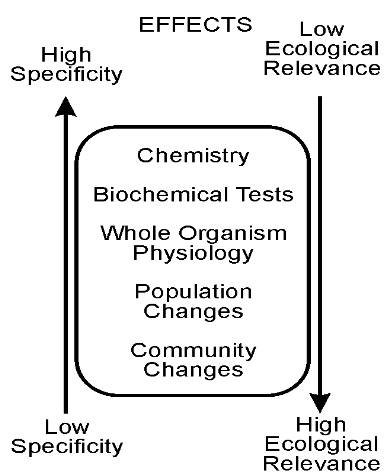

Fig. 10 A schematic diagram relating the cost of ecosystem monitoring with the complexity and relevance of the measurements (modified from Addison 1996).

Conclusions

Like Waldichuk (1990), we conclude that continental shelves and near shore areas of the North Pacific are under the greatest stress from chemical contaminants. Increasing population and industrialization will increase that stress. With either their restriction or elimination (PCBs, OC pesticides), global cycling of some of these contaminants have decreased; however, it is likely that they will continue to cause concern for some time (Ross *et al.* 2000). And we will discover new chemical problems to replace old ones (*e.g.*, see Betts 2001; Kramer and Giesy 1995; Paasivirta 1998).

The problem we face is not just chemical contamination, but assault on coastal systems from multiple stressors. Presently, we lack coherent observational networks, reliable inventories for contaminants, and an understanding of processes that would unequivocally distinguish real threats from perceived threats. Given the degree of concern that pervades much of the literature cited here, it is surprising that the scientific and political communities of the North Pacific have not collaborated to conduct a regional assessment. The Arctic Monitoring and Assessment Programme (AMAP 1998) provides an apt example where international hurdles were overcome to produce a well-founded, science-based review that led to action. We therefore suggest that the highest priority for PICES should be to produce, within the next five years, an International North Pacific Assessment Programme.

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