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Northern Australia, whither the mercury?

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Biomagnification of mercury (Hg) leads to high concentrations in fish and subsequent health risks for fish-eaters. Despite the global concern afforded to Hg over the past 40 years, little attention has been paid to this toxic heavy metal in Australia in general, and tropical northern Australia in particular. This review examines past Hg research in Australia and explores seven hypotheses as to why so little research and monitoring has been conducted in northern rivers and estuaries. We rule out the possibility that fishing intensity (an indicator of potential Hg exposure in humans) is lower in Australia than in other countries with more intensive Hg research programs. Instead, we hypothesise that low atmospheric deposition, owing to prevailing wind direction and few local point sources, coupled with highly productive waterbodies, contributes to low Hg bioaccumulation and hence the reduced interest in measuring Hg. Outstanding questions regarding Hg in northern Australia include the assessment of atmospheric deposition rates of Hg, the trophic level and growth and food consumption rates of consumers such as large-bodied fishes, linkages between fire regimes and Hg availability, and the capacity for *in situ* Hg methylation in tropical systems.

Additional keywords: fishing intensity, atmospheric deposition, fire, productivity, trophic level

Introduction

Mercury (Hg) contamination of aquatic systems is a global problem (Mergler *et al*. 2007) that can threaten the health of fish-eating humans and wildlife. These threats can subsequently lead to closures of recreational and commercial fisheries with concomitant socioeconomic impacts (Swain *et al*. 2007). The cycling and fate of Hg in the environment is complex and sources often difficult to identify. Hg enters aquatic systems via atmospheric deposition, weathering and biotic transport (Boening 2000; Blais 2005). Some fraction then becomes methylated to form methyl Hg (meHg) that biomagnifies through food chains to reach potentially toxic concentrations in top predators (Cabana and Rasmussen 1994; Watras *et al*. 1998). Hg methylation is enhanced under acidic and anoxic conditions and is associated with the presence of sulfate-reducing bacteria (Gilmour *et al*. 1992); as a result, high concentrations may occur in fishes and other aquatic biota even in areas that are far removed from point sources (Håkanson *et al*. 1988).

Public concern about Hg contamination and subsequent bureaucratic intervention is globally asymmetric. Currently, in the United States, Canada and the Scandinavian countries, a large proportion of lakes and rivers are under some form of government Hg advisory aimed at limiting consumption of fish of a particular species or size by high-risk groups such as young children and women who are pregnant or of a child-bearing age (Håkanson *et al*. 1988; Cunningham *et al*. 1994). In Australia, there are currently no advisories regarding contaminants in recreationally or traditionally caught fish. However, prior research on Hg concentrations in human hair showed that residents of Sydney and Darwin had concentrations that were

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comparable to those in humans in Canada and the United States (Airey 1983). This suggests that at least moderate doses of Hg are potentially being consumed by Australian residents.
Despite the lack of research and monitoring, awareness of Hg as a health issue is apparent in Australia. Guidelines pertaining to fish consumption are available from government agencies; however, they focus specifically on store-bought food (Food Standards Australia New Zealand 2004). Canned tuna tends to be one of the few fish items consumed on a weekly or even daily basis. An acceptable concentration of 1.0 μg g^{-1} is set as the limit for the sale of commercial species such as shark (subclass Elasmobranchii) (Lyle 1984), tuna (*Thunnus* spp.) and barramundi (*Lates calcarifer*), with lower limits imposed on other species. This concentration is similar to that recommended by institutions such as Health Canada (0.5 μg g^{-1} for most species, 1.0 μg g^{-1} for snake mackerel (*Gempylus serpens*), marlin (Family: Istiophoridae), orange roughy (*Hoplostethus atlanticus*), tuna, shark and swordfish (*Xiphias gladius*); Health Canada 2004) and higher than that imposed by the United States Environmental Protection Agency (0.3 μg g^{-1} triggers an advisory, USEPA 2001). Risk assessments for seafood in Australia, however, have indicated that Hg in fish ranks low in terms of risk relative to potential causes of acute human fatalities such as ciguatera, viruses in shellfish (oysters), and algal biotoxins (Sumner and Ross 2002). This could therefore underestimate the long-term risk resulting from Hg exposure.

The purpose of the present review is to advance seven hypotheses as to why so little research and monitoring of Hg has been conducted in Australia in general, and northern Australia in particular, relative to other Western countries with similar populations and resources for scientific inquiry (e.g. Canada). Although we do not rule out the possibility that Hg is indeed high in parts of Australia, we explore several features of the northern Australian landscape that could lend itself to low Hg availability (low Hg deposition, few point sources, high aquatic productivity). The review focuses mainly on riverine and estuarine ecosystems, with some reference to coastal waters where appropriate. We define northern Australia as the region that includes both wet (high annual precipitation) and wet–dry (monsoonal summer rains) tropics extending from Broome in the west (17.95S, 122.23E) to Townsville in the east (19.26S, 146.82E).

Possible explanations for the lack of concern about Hg in Australia

Fishing intensity is lower in Australia than in other countries

Consumption of fish is the primary route of exposure to Hg for most of the global human population (Mergler *et al*. 2007). Therefore, research and monitoring of Hg should be proportional to the importance of fish as a source of protein in a given society. Because of the fact that Australian freshwater resources are generally quite scarce, it would be fair to assume that fishing intensity might be similarly lower than in countries where water resources are richer (e.g. Canada). However, parts of Australia's north do see extensive consumption of freshwater resources by recreational users and Aboriginal people. Fishing is named as the most important reason for tourists visiting communities (Karumba and Normanton) in the southern Gulf of Carpentaria (Stoeckl *et al*. 2006), and recreational and commercial landings of coastal northern species such as Talang queenfish (*Scomberoides commersonianus*) continue to increase (Griffiths *et al*. 2006).

Recreational fishing statistics for Australia and Canada are summarised in Cooke and Cowx (2006) and show that on a per capita basis, fishing intensity is essentially equal between the two countries, with 3.6 million anglers per annum spending 47.9 million days fishing in Canada (with a population ~30 million) and 3.4 million anglers per annum spending 20.6 million days fishing in Australia (with a population ~20 million). These two countries could therefore be considered roughly equivalent, yet a search of listed journal articles (ISI Web of Science) using the search terms ‘Australia + mercury + fish’ yields only 51 citations, whereas a search with terms ‘Canada + mercury + fish’ yields 435 citations. Clearly, there is a mismatch between the effort of the angling community in catching fish in Australia and the effort of scientists in determining whether Hg is a problem.

We have not looked hard enough

Given the above, the simplest and most plausible explanation for the lack of attention to Hg in Australia in general and northern Australia in particular is simply a lack of data. Many of Australia’s northern rivers are located far from large urban centres and therefore are under-studied relative to major drainages in south-eastern Australia such as the Murray–Darling (e.g. Gehrke *et al*. 1995). Much of the little Hg research that has been conducted in Australia has focussed on marine sites in heavily industrialised areas such as Port Philip Bay in Victoria (Phillips *et al*. 1992), the southern coast of New South Wales (Gibbs and Miskiewicz 1995) and the Derwent Estuary in Tasmania (Plaschke *et al*. 1997). These case studies were likely initiated in response to public concern surrounding the discharge of waste containing Hg and other metals to receiving waters, and will be discussed in more detail below.

In Australia, population growth in the north, combined with increased water demands in the south, has led to development pressures for the northern region’s fresh water (Hamilton and Gehrke 2005). It is expected, therefore, that at some point, an evaluation of the threats to ecosystem and human health associated with development will be warranted. Furthermore, Indigenous groups in the north rely on the harvesting of bush foods (Meehan 1991) far more than individuals of European ancestry, and thus are more likely to suffer the effects of poorly managed resources. A repeat of past tragedies in other Indigenous communities (e.g. Grassy Narrows Indian Reserve on the Wabigoon River, Ontario, Canada; Troyer 1977) could therefore be avoided by a proper screening program for contaminants such as Hg.

It is possible that a survey of Hg in the biota of the region will find that concentrations are at or below background levels. If so, it will become necessary to explain why this might be. The remaining five hypotheses deal specifically with features of northern Australia that may (or may not) be responsible for low Hg bioaccumulation in riverine and estuarine systems.

Atmospheric deposition of Hg is low

Mercury concentrations in fish are related to deposition of Hg. In a large-scale survey, Hammerschmidt and Fitzgerald (2006) found a linear relationship between US state-averaged Hg deposition and Hg concentrations in smallmouth bass (*Micropterus dolomieu*). Similarly, studies at the Experimental Lakes Area in central Canada have shown that the aerial addition of isotopically labelled Hg results in its rapid incorporation into the food web, causing fish Hg concentrations to respond linearly to the amount applied (Harris *et al*. 2007). These and other studies suggest that, broadly speaking, Hg in fish will depend to a large degree on the amount of Hg entering aquatic food webs via deposition. Therefore, rates of Hg deposition will partly determine concentrations in fish in northern Australia.

Since a large fraction of certain Hg species (i.e. Hg^{2+}) that are emitted is deposited in the near vicinity of its source (Schroeder and Munthe 1998; Lindberg *et al*. 2007), there are established relationships between total emissions and deposition for a given geographic region (Table 1). Ryaboshapko *et al*. (2007) found that for the countries United Kingdom, Italy and Poland, although a large fraction of emitted Hg left the country of origin, there remained links between emissions and deposition, with Poland having higher emission and deposition. Likewise, concentrations of Hg in rainwater in areas of China affected by coal combustion (accounting for Hg emissions of 8.3 t in 1998; Feng *et al*. 2002) and Hg mining are in the range of 33 ng L^{-1}, leading to high estimated wet deposition rates of 39 µg m^{-2} $year^{-1}$ (Feng *et al*. 2002). Mercury deposition in Norway decreases from south to north, corresponding to a gradient in industrial emissions from continental Europe through to sparsely inhabited northern regions (Iverfeldt 1991).

Deposition rates will also be related in part to hydrology. Wetter areas typically receive higher deposition, as in Japan where deposition ranges from 6 to 18 µg m^{-2} $year^{-1}$, with the highest rates associated with high annual rainfall (Sakata and Marumoto 2005). Likewise, the highest wet deposition in North America occurs in the south-east of the continent where the rainfall is highest; deposition approximates 15–20 µg m^{-2} $year^{-1}$ (National Atmospheric Deposition Program 2007), as opposed to areas in the West where it is typically 5 µg m^{-2} $year^{-1}$. In other areas in the US and Sweden removed from urban centres, the deposition rate ranges from 8 to 14 µg m^{-2} $year^{-1}$ (summarised in Schroeder and Munthe 1998). Therefore, a combination of rainfall amounts and proximity to emission sources will determine deposition rates.

Low estimates of deposition across Australia are to be expected because of relatively low anthropogenic emissions of Hg to the atmosphere (Nelson 2007), combined with geographic position on the globe and low annual rainfall. Measurements of atmospheric deposition of Hg in Australia have never been formally reported, yet all signs point to low deposition rates, particularly in the north. Although Australia uses coal to generate the majority of its electricity (~77%; Mukherjee *et al*. 2008), and coal combustion is perhaps the single largest source responsible for anthropogenic Hg emissions (Mukherjee *et al*. 2008), the Australian population (and hence demand for electricity) is small enough that the pool of Hg emitted is correspondingly spread out over a large-enough area that it likely has little effect on remote areas. In northern Australia, one potential major Hg emission source is a base-metal mine at Mount Isa (20.74S, 139.50E), located at the southern edge of the tropics. It emits ~1100 kg of Hg per year (National Pollutant Inventory, www.npi.gov.au, accessed 15-August 2009) and smaller emission sources have been shown to lead to elevated Hg concentrations in the surrounding environment (e.g. Jardine *et al*. 2009).

Given the strong seasonality of precipitation in much of northern Australia, both wet and dry deposition of Hg are likely to be important. During monsoonal rains from December to March, wet deposition will likely predominate, whereas in the dry months of May–October, dry deposition could play an important role, especially Hg in the particulate phase bound to dust particles that are carried across the continent (McTainsh 1989). Although difficult to measure, dry deposition has been shown to account for 50% or more of total Hg deposition to various landscapes (Lindberg *et al*. 2007).

Asia contributes approximately half of all global anthropogenic Hg emissions (Jaffe and Strode 2008). Some of the Hg emitted in Asia is transported across the Pacific to north-western North America, yet large-scale atmospheric patterns preclude any deposition to Australia of Asian-origin Hg (Jaffe and Strode 2008). This, combined with recent estimates that have placed Oceania as having the lowest emission rates (only 25–35 t $year^{-1}$) of all major continental areas (Pirrone *et al*. 1996; Nelson 2007), likely results in low Hg deposition to the Australian landscape, although this awaits confirmation via empirical measurements.

There are few point sources of Hg to the aquatic environment

Although atmospheric deposition is important in determining the amount of Hg available to aquatic food webs and ultimately to fish, direct discharges to the aquatic environment can also be important (Harada 1995). Areas around the world where point sources of Hg exist consistently show higher concentrations of Hg in hair and blood of resident humans. For example, in the Amazon Basin, alluvial gold mining that involves amalgamation with elemental Hg has led to elevated levels of methyl Hg in fish in the river and in the hair of residents (Akagi *et al*. 1995).

The Amazonian example could possibly apply in northern Australia. In many northern Australia rivers, there are rich deposits of gold and other metals, and current high world mineral prices (particularly gold) have led to the reworking of sediments previously considered to be unprofitable. Rivers such as the Palmer (a tributary of the Mitchell River, northern Queensland) had an initial gold rush in the mid- to late 1800s, similar in timing to Reedy Creek and the Lerderberg River in Victoria, and the Gympie area in Queensland. These latter areas maintain elevated Hg concentrations to the current era in soils, sediments, water and in some cases fish (Bycroft *et al*. 1982; Dhindsa *et al*. 2003; Churchill *et al*. 2004). Gold rushes in these areas occurred at a time when Hg was used as an amalgam and therefore applied directly to sediment slurries (Dominique *et al*. 2007). These Hg-rich sediments could pose a major threat if they are resuspended into the water column and transported downstream (Telmer *et al*. 2006).

In southern Australia, areas with other known point-source discharges to rivers and estuaries have also shown elevated concentrations of Hg in fish. In the Derwent Estuary, Tasmania, the operation of a zinc refinery and paper mill resulted in direct Hg discharges to the receiving environment and waterborne concentrations as high as 16 μg L^{-1} in the 1970s (Plaschke *et al*. 1997). Although concentrations in fish were high at the time (1 μg g^{-1} in sand flathead *Platycephalus bassenis* in 1974; Dix *et al*. 1975), they have since declined (0.5 μg g^{-1} in sand flathead in 1983; Langlois *et al*. 1987), and much of the current Hg is bound to suspended solids (Plaschke *et al*. 1997), possibly rendering it unavailable for uptake to the food web.

Port Philip Bay in Victoria followed a similar trajectory to the Derwent Estuary through time, with concentrations of Hg and other metals elevated in the 1970s and reductions occurring over the past 30 years.

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A large fraction of the metals were being delivered to the Bay by the Yarra River that flows through Melbourne, where they were deposited to sediments under low flow conditions (Fabris *et al*. 1999). Between 1975 and 1990, concentrations in sand flathead decreased from an average of 0.50 μg g^{-1} (Walker 1982) to 0.23 μg g^{-1} (Fabris *et al*. 1992), possibly in response to the diversion of industrial wastes to the city's sewage system. The most recent survey has found concentrations to be unchanged from 1990 levels at 0.21 μg g^{-1}, and Port Philip Bay currently has heavy metal concentrations in consumer species (flathead, abalone, lobster and snapper) that are below consumption guidelines and not significantly different from those in other coastal Victorian waters (Fabris *et al*. 2006).

The southern coast of New South Wales has also seen elevated Hg concentrations as a result of industrial activities. In Lake Macquarie, local sources such as coal-fired power stations, smelters and mining have caused elevated sediment Hg concentrations (Roach 2005), yet Hg in fish (yellowfin bream, *Acanthopagrus australis*, silverbiddy, *Gerres subfasciatus*, trumpeter whiting, *Sillago maculata*, and two gobies) is not higher than in reference locations (Roach *et al*. 2008), possibly because of the interacting effects of deposited selenium (Belzile *et al*. 2006). Sewage outfalls near Sydney, however, have contributed to higher than average Hg concentrations in fish (McLean *et al*. 1991). Concentrations in tarwhine (*Rhabdosargus sarba*) collected near the Malabar sewage outfall, the recipient of several industrial wastes, were found to be higher than recommended consumption guidelines (>0.5 μg g^{-1}) (Gibbs and Miskiewicz 1995). Sediments in this area also have Hg concentrations that are higher than background levels (Schneider and Davey 1995).

Another less explored potential source of Hg is via runoff from sugarcane fields. Historically, Hg fungicides were used to treat cane. This was recognised as a possible source of contamination of municipal drinking-water supplies in the Burdekin River (Brodie *et al*. 1984), and more recently as a threat to the health of the Great Barrier Reef, where Hg concentrations in surface sediments have increased 3-fold relative to pre-1850 levels (Haynes and Michalek-Wagner 2000). However, concentrations in fish residing in areas offshore from sugarcane production (e.g. Townsville, Queensland) are not higher than expected (Denton and Breck 1981).

Natural geologic sources of Hg may also lead to localised areas with concentrations in sediment and biota higher than those in the background. Biotite-rich granite formations have been linked to high Hg concentrations in fish and water of Kejimkujik National Park in eastern Canada (Page and Murphy 2003), and cinnabar deposits worldwide are known to contain associated Hg (Hylander and Meili 2003; Zhang *et al*. 2004). Large-scale studies of links between geological features and Hg in higher-order consumers such as fish have yielded results that are suggestive but not conclusive (Lockhart *et al*. 2005*a*).

All of the examples above illustrate that the most likely locations for high Hg concentrations in fish are in areas with known point sources, yet the presence of high concentrations in water and sediment does not guarantee high concentrations in the biota. Given these conditions, the most probable locations in northern Australia where Hg may be of concern are in systems affected by gold mining and geological sources of Hg, whereas other locations are unlikely to have fish with elevated concentrations.

Repeated landscape burning removes stored mercury

At first glance, it would appear logical that the practice of controlled landscape burning by Aboriginal people in northern Australia (Haynes 1991) could result in lower Hg availability to aquatic systems. Burning releases more than 90% of stored carbon and Hg to the atmosphere (Mailman and Bodaly 2005), thereby making it unavailable for methylation after flooding (see next section). However, a recent study suggests that the opposite may be true; burning may in fact increase Hg concentrations in biota.

Kelly *et al*. (2006) reported increased Hg concentrations in consumers in a lake in the Rocky Mountains, USA, following a forest fire. They proposed the following two mechanisms for the higher Hg concentrations after fire: (*1*) enhanced productivity, leading to a lengthening of the food chain because of greater consumption of fish and zooplankton and (*2*) increased delivery of inorganic and methyl Hg to the lake by streams that drained the lake catchment. Fire resulted in significant increases in dissolved nutrients (nitrogen and phosphorus), conductivity, chlorophyll *a* concentrations, zooplankton density and fish growth rates, and caused dietary changes in fish, the two key changes being a switch from detritivorous *Hyalella* to zooplanktivorous *Mysis* as a major dietary component and an increased consumption of rainbow trout (Kelly *et al*. 2006). All of these changes resulted in higher Hg in fish muscle than was the case before the fire period.

Other studies on the relationship between fire and Hg have found similar results. Amirbahman *et al.* (2004) demonstrated higher methyl Hg concentrations in upper soil horizons of a burned catchment compared with an unburned catchment. They attributed this difference to a more rapid cycling of organic matter (and hence methyl Hg production) in the burned catchment. Garcia and Carignan (2005) found that Hg concentrations in fish were highest in lakes with partially burnt catchments whereas fully burnt lakes showed no difference compared with reference lakes. These studies suggest that the effect of fire on Hg cycling will occur via indirect pathways, and that the location of the burning relative to waterbodies will be important in determining whether resultant Hg and nutrients bound to particles are deposited onto water surfaces.

In northern Australia, where as much as 30% of the landscape can be burned every year (Vigilante *et al.* 2004), there will likely be some relationship between the frequency of burning and Hg availability; however, studies are required to determine the exact nature of the relationship. Recent work by Packham *et al.* (2009) estimated total emissions of Hg from biomass burning of 129 t per year for the entire Australian continent, with tropical savannas contributing ~60% of the total. Clearly, this is a large number and could represent a significant source of Hg to aquatic ecosystems in northern Australia. However, given that the emission and deposition of Hg resulting from burning represents an annual or semi-annual cycle in most of northern Australia (as opposed to a net addition that would be expected when, for example, coal is extracted and burned), it is possible that concentrations in aquatic systems are in an equilibrium that has been achieved over several thousand years of burning.

Conditions for methylation are less prevalent

Concentrations of Hg in sediments and surface waters in Australia are generally at the lower end of the range reported for other locations globally. Serena and Pettigrove (2005) reported Hg concentrations of 0.01–0.50 μg g^{-1} in creeks near Melbourne, whereas Lottermoser (1998) found concentrations below detection (<0.08 μg g^{-1}) in the Richmond River, northern New South Wales, and concentrations as high as 0.39 μg g^{-1} near industrial sites in Newcastle, New South Wales. Despite the presence of industry at many of these sites, the concentrations are lower than those observed in more heavily contaminated regions of the world (summarised in Ullrich *et al*. 2001). For surface waters in northern New South Wales, running waters had Hg concentrations ranging from 5 to 7 ng L^{-1} (Shah *et al*. 2007), whereas in two reservoirs in Tasmania (Lake Gordon and Lake Pedder), surface waters had Hg concentrations ~2 ng L^{-1}, comparable to remote lakes in North America and Europe (Bowles *et al*. 2003*a*, 2003*b*). In the latter systems, methyl Hg concentrations were relatively high (~0.3 ng L^{-1}), likely owing to a high proportion of wetlands in the catchment, because wetlands are known to be net producers of methyl Hg (Rudd 1995). This illustrates that despite low Hg concentrations expected in remote locations such as northern Australia, the major determinant of Hg risk to consumers will be the amount of methyl Hg that is available. Methyl Hg is the toxic form of Hg that is also more efficiently transferred up the food chain. Therefore, the presence of conditions known to promote Hg methylation will more likely contribute to the Hg risk in northern Australia.

Mercury methylation in aquatic environments occurs via several biotic pathways, most notably by sulfur-reducing bacteria (Gilmour *et al*. 1992); other pathways such as iron reduction by bacteria have recently been discovered (Fleming *et al*. 2006). There are also several abiotic pathways for Hg methylation, most involving humic matter (Weber 1993). MeHg production is greatest in lake sediments and wetlands under anoxic conditions (Rudd 1995). Hypoxic and anoxic waters are common in isolated waterbodies in northern Australia, particularly those with large infestations of weeds such as water hyacinthe (*Eichhornia crassipes*, Perna and Burrows 2005). Other contributions to meHg production come from the water column and mucus and intestines of fish. Methylation occurs largely at interfaces, including the sediment–water interface, the anoxic–oxic interface, and on leaf surfaces following flooding (Ullrich *et al*. 2001). Typical variables known to enhance Hg methylation include low pH, low salinity, a reducing environment and a pool of decomposable organic matter (Ullrich *et al*. 2001). Methylation is also inhibited under high sulfide concentrations (Benoit *et al*. 1999).

Some features of the northern Australian landscape may lend themselves to enhanced Hg methylation. Seasonal hydrology is such that one or more large flood pulses pass through in a distinct annual wet–dry cycle (Kingston 1991). These flood pulses could stimulate methyl Hg production in the soil and on the surfaces of terrestrial detritus. They also yield high sulfate concentrations and low pH (Hart and McKelvie 1986), conditions amenable to methyl Hg production. Furthermore, large macrophyte mats that colonise temporarily wetted surfaces and permanent and semi-permanent lagoons may be important sites for methylation, as has been observed for Amazonian systems (Guimarães *et al*. 2000). Low pH (<6) is relatively common in floodplain waters, yet high sulfate concentrations tend to be rare outside of lowland

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areas influenced by seawater (e.g. Mitchell River, T. D. J., unpubl. data). This suggests that generalities may be difficult to predict, because each waterhole may have unique characteristics that either enhance or limit Hg methylation.

Wetlands are known to be hotspots for Hg methylation, and biotic Hg concentrations are often correlated with both the proportion of wetlands in a given catchment or dissolved organic carbon concentrations (DOC) (Rencz *et al*. 2003); the latter is considered a surrogate for the influence of wetlands. The spatial extent of wetlands across northern Australia and the length of time this land remains wetted in the annual wet–dry cycle remains relatively unknown. Finlayson *et al*. (2005) estimated up to 98 700 km^2 of surface area could qualify as wetlands; however, this includes many areas that receive a very brief flood each year and thus would contribute little to meHg production. The little information that exists on the concentrations of DOC in northern Australian waters suggests that concentrations are low, less than 10 mg L^{-1} (Hart and McKelvie 1986; Ford *et al*. 2005; Hogan *et al*. 2005).

Aquatic systems are productive

Productivity of aquatic systems can play a role in determining the uptake of contaminants such as Hg. Tropical streams and rivers are an order of magnitude more productive than temperate streams and rivers (Gross *et al*. 1988; Davies *et al*. 2007). This enhanced productivity could lead to lower Hg concentrations via the following two mechanisms: (*1*) a shortening of food chains because of increased omnivory (feeding on more than one trophic level) and (*2*) growth dilution of Hg by consumers. These two mechanisms will be examined separately.

Food chains are short

Long food chains have higher concentrations of contaminants in top predators (Kidd *et al*. 1995). This is due to the effects of biomagnification, wherein a contaminant such as Hg increases with each step in the food chain. The relationship between a contaminant and food chain position is now most commonly evaluated with stable nitrogen isotopes (Jardine *et al*. 2006), which accurately reveals omnivory. Omnivory can dampen the relationship between the presumed trophic level and contaminant concentration. Whereas traditional models typically described simple, linear food chains with predicted high concentrations in top predators (e.g. temperate lakes, Rasmussen *et al*. 1990), later assessments in these same systems with stable isotopes found considerable omnivory in top predators, leading to lower concentrations than would be expected on the basis of discrete trophic-level classifications (Cabana and Rasmussen 1994; Vander Zanden and Rasmussen 1996).

Food chain length and the presence of omnivory continue to generate debate in the ecological literature (Thompson *et al*. 2007). There is not yet any clear evidence whether more productive systems sustain shorter or longer food chains, owing to the often confounding effect of ecosystem size (Vander Zanden *et al*. 1999; Post *et al*. 2000). Theoretically, productivity could increase food chain length because more energy available at the base of the food web could overcome inefficiencies in energy transfer to higher-order predators

(Hairston and Hairston 1993). However, an argument can also be made for shorter food chains in more productive environments, because of a greater abundance of food at low trophic levels. Layman *et al.* (2005) showed that in tropical river food webs, large-bodied predators often exploited low trophic level prey, and there was no relationship between the predator body size and corresponding trophic level. They attributed this to the presence of large herbivorous prey fish that achieved their large body size by feeding on a variety of resources, including nuts/seeds and benthic flora. Similarly, Lewis *et al.* (2001) found that fish of the Orinoco floodplain more commonly fed on trophic levels 2 (herbivory) and 3 rather than trophic level 4 (piscivory), suggesting compressed food chains. Many northern Australian freshwater fish species exhibit opportunistic diets, consisting of a combination of plants, invertebrates and fish (e.g. northern Queensland; Pusey *et al*. 2004). This includes some of the larger species that are favoured in recreational and traditional fisheries. Whereas freshwater longtom (*Strongylura kreffti*) is almost exclusively piscivorous, most species feed on a mix of crustaceans, insects and occasionally fish. For example, large black bream (*Hephaestus fuliginosus*) had a maximum of 10% of its diet from fish in larger individuals whereas on average (all size classes) only 3.4% of its diet was composed of fish (Pusey *et al*. 2004). Saratoga (*Scleropages jardinii*) and tarpon (*Megalops cyprinoides*) also exhibit mixed diets of invertebrates and small fish (Coates 1987) as does sleepy cod (*Oxyeleotris lincolatus*) (Pusey *et al*. 2004). The diet of fork-tailed catfish varies with species, with *Arius midgleyi* the only species that is piscivorous and salmon catfish (*Neoarius leptaspis*) feeding on a mix of food sources, including fish (Allen 2002). Bony bream (*Nematalosa erebi*) and some other catfish species primarily consume detritus (Pusey *et al*. 2004; Sternberg *et al*. 2008) and can be prey species for piscivorous fish such as barramundi (Davis 1985), which would leave barramundi only two trophic links away from primary producers. All of these examples illustrate that food chains in northern Australia are unlikely to be linear, but rather a branched network with much omnivory and correspondingly low trophic levels, even for presumed higher-order predators. Examining available literature lends some credence to the low trophic level occupied by northern Australian fish (Table 2) as predicted by Douglas *et al.* (2005). Iconic species in North America that are often the focus of Hg and organochlorine research are largemouth bass (*Micropterus salmoides*), smallmouth bass (*Micropterus dolomieu*), lake trout (*Salvelinus namaycush*), walleye (*Sander vitreus*) and northern pike (*Esox lucius*). The maximum trophic level of these species in lakes is often greater than 4.0, indicating complete piscivory (Vander Zanden and Rasmussen 1997; Post *et al*. 2000) (Table 2). By using the same techniques for calculating the trophic level as in the North American studies, barramundi and fork-tailed catfish from floodplain rivers in northern Australia were found to have trophic levels ranging from 3.0 to 3.5, albeit in a limited dataset (Douglas *et al*. 2005) (Table 2). For barramundi, the proportion of fish in the diet increases strongly with increasing body size, with adults having up to two-thirds of their diet composed of fish (Davis 1985; Pusey *et al*. 2004), likely explaining the linear increase in Hg with increasing body size (Jones *et al*. 2005). Whether this results in trophic levels (TLs) and Hg concentrations equivalent to North American fish species is yet to be determined.

Growth dilution occurs

Growth dilution is a phenomenon whereby the rapid addition of new tissue dilutes the concentration of a given contaminant. This can occur at various trophic levels, including algae (Hill and Larsen 2005), invertebrates (Karimi *et al*. 2007) and fish (Simoneau *et al*. 2005). Efficient conversion of food to biomass is responsible for growth dilution in fish, although it can be modulated by energy costs associated with obtaining food and other energy-demanding activities (e.g. reproduction) (Trudel and Rasmussen 2006).

The question remains whether tropical Australian fish grow faster than their temperate counterparts. One commonly fished species in Canada, lake trout (*Salvelinus namaycush*), is a slow grower with a long life-span (Fig. 1). Commonly, harvested lake trouts from boreal lakes are >10 years old. A 10-kg lake trout will have a minimum age of ~15 years (Ferreri and Taylor 1996). Walleye (*Sander vitreus*) similarly grows slowly, and differences in growth rates among populations in Canada have contributed to differences in Hg concentrations (Simoneau *et al*. 2005). Growth rates in those species can be contrasted with that of barramundi which is reported to reach body sizes of 350 mm (~0.3 kg) by the age of 1, 750 mm (~3 kg) by the age of 4, and 1075 mm (~8 kg) by the age of 10 (Pusey *et al*. 2004).

Whether the apparently superior growth of barramundi is due to better assimilation efficiency, more abundant food resources, a longer growing season, or simply species-specific constraints is as yet unknown. Ultimately, food conversion efficiency, rather than growth rate, is the best measure of evaluating the likelihood of growth dilution (Trudel and Rasmussen 2006). Indeed, barramundi shows a relatively high growth efficiency (>40%) over the temperature range 27–35°C (Katersky and Carter 2007) in controlled conditions. Estimates of growth efficiency for lake-trout populations, meanwhile, range from 3 to 24% (Pazzia *et al*. 2002). However, comparing these data is questionable because the former data come from laboratory conditions where feeding was on a formulated diet to satiation whereas the latter data are averaged over an annual cycle and using energy values of natural prey. Tropical fish that live at higher ambient temperatures have correspondingly higher resting metabolic rates than do temperate fish (Clarke and Johnston 1999). These higher metabolic rates likely require higher food consumption rates and, thus, could lead to higher contaminant exposure despite more rapid growth on an annual basis. Unravelling the relationship between food consumption, growth and Hg accumulation will require laboratory-based trials for both tropical and temperate species.

Where might we see high concentrations and effects?

The hypotheses advanced above, all point to a low likelihood of Hg contamination of aquatic food webs in northern Australia. However, there will undoubtedly be considerable variability in Hg concentrations among different consumer groups in the north.

Given the dominant non-piscivorous dietary habits of fish species in northern Australia, exposure to Hg for human populations consuming these fish may be minimal. Most major Hg contamination episodes in humans occur as the result of populations relying on locally available foods. This is the case for the Amazon basin, where families of fishermen in goldmine-affected areas have Hg concentrations that are as high as those of the gold miners themselves (Akagi *et al*. 1995), owing to consumption of fish such as piraiba (*Brachyplatystoma filamentosum*), a predatory catfish. It is also an issue in First Nations groups in Canada (Van Oostdam *et al*. 1999), where country food includes marine mammals and freshwater fish that often sit atop long food chains (Hobson and Welch 1992; Campbell *et al*. 2005) and have correspondingly high Hg concentrations (Lockhart *et al*. 2005*b*). In the Lake Murray region, Papua New Guinea, barramundi is the only species that consistently has individuals with Hg concentrations higher than the recommended consumption guideline (tuna and mackerel do not) (Sorentino 1979); this occurs despite an apparent lack of anthropogenic or natural geologic sources as Hg concentrations in water are low (total Hg = 1.5 ng L^{-1}, meHg = 0.07 ng L^{-1}; Bowles *et al*. 2002). Rather, a high bioaccumulation rate from water to phytoplankton (meHg in seston = 0.015 µg g^{-1}), coupled with higher than average biomagnification power in the food web leads to the observed high concentrations in barramundi (Bowles *et al*. 2001). The barramundi is an important source of protein for residents, leading to high Hg concentrations in the local population (Abe *et al*. 1995). All of these situations have occurred because of a high level of subsistence on locally available foods, with Hg concentrations that may exceed those of foods currently consumed by Indigenous peoples in northern Australia.

Similar to the fish species described above, other traditional foods consumed by humans may have low Hg concentrations as a result of the dietary habits of these target species. Magpie geese (*Anseranas semipalmata*) and wallabies (Family Macropodidae) are strictly herbivores (Whitehead and Tschirner 1992), whereas turtles eat a mix of plants, invertebrates and occasionally fish and carrion (Georges and Kennett 1989). An exception may be filesnakes (*Acrochordus arafurae*) that are piscivorous, feeding mainly on catfish and sleepy cod (Madsen and Shine 2000) and thus are likely to have relatively high Hg concentrations. Filesnakes, however, are unlikely to feature prominently in the modern Aboriginal diet. These observations suggest that exposure to Hg in Aboriginal populations may indeed be low.

There are some birds with a purely piscivorous diet, including pelicans, cormorants, darters, storks and egrets, all of which occur in northern Australia (Morton and Brennan 1991). Piscivorous birds have been shown to have high Hg concentrations (Odsjo *et al*. 2004), and in some cases this has led to health impairments such as those observed with common loons in eastern Canada (Burgess and Meyer 2008). During dry-down of floodplains after the wet season, large aggregations of fish become trapped in residual waterholes, making them susceptible to high levels of predation by waterbirds. Under these circumstances, birds could be ingesting a large dose of Hg, depending on the species on which they are foraging. The most abundant species, however, are likely to be rainbowfish (*Melanotaenia* spp.), glassfish (*Ambassis* spp.), spangled perch (*Leiopotherapon unicolor*) and barred grunters (*Amniataba percoides*) (Bishop *et al*. 1995), all lower trophic level feeders with correspondingly low Hg concentrations.

Estuarine crocodiles (*Crocodylus porosus*) are perhaps the most likely candidates for Hg exposure because of their predatory nature and long life-spans; yet their trophic level may not be exceedingly high because their diet includes herbivorous mammals, including cattle, according to anecdotal reports. In cases where crocodile diet is composed largely of waterbirds that feed on fish, exposure may be high. However, Jeffree *et* *al*. (2001) reported Hg concentrations in flesh and osteoderms for *C. porosus* from the Alligator Rivers region, Northern Territory, that were below the detection limit (0.05 µg g^{-1}). Likewise, freshwater crocodiles (*C. johnstoni*) from the Lynd River, northern Queensland, also had Hg concentrations below the detection limit (0.01 µg g^{-1} dry weight) in osteoderms (Jeffree *et al*. 2005). The diet of *C. johnstoni* is highly variable, consisting of an assortment of invertebrates and fish, with rare consumption of birds and mammals by larger individuals (Tucker *et al*. 1996). These patterns of low Hg concentrations in top predators are surprising and require further investigation. There have been documented cases of high Hg concentrations in crocodiles in other locations globally; however, health effects are rare (Rainwater *et al*. 2002).

Future research

All signs point to a low likelihood of considerable Hg exposure for humans and wildlife in northern Australia. However, without empirical data this assertion is entirely speculative. A conceptual model (Fig. 2) illustrates the hypothesised links between ecosystem components and potential determinants of Hg concentrations. An initial assessment of Hg concentrations in iconic species such as barramundi across the region would be an important first step in determining whether indeed there is a potential Hg issue in the north. Earlier work by Jones *et al*. (2005) showed that concentrations in barramundi could exceed WHO guidelines, but only at large body sizes. Large piscivores should therefore be targeted for studies on Hg, and sampling should include sites that are potentially affected by mining activities. Other commonly consumed aquatic food items, including prawns, crabs and other fish species, should also be analysed.

Another major priority for future work on Hg in Australia is an estimate of atmospheric deposition of Hg, both in remote locations (e.g. the north) and more heavily populated urban locations (e.g. the south-east). Given the links between Hg deposition and concentrations in fish (Hammerschmidt and Fitzgerald 2006; Harris *et al*. 2007), these measurements would help determine the likelihood of high concentrations in aquatic systems. Studies at the landscape scale should be concentrated on the relationship among fire, nutrients, productivity and the prevalence of conditions that are ideal for Hg methylation. Work in aquatic systems should focus on food chain length and growth rates and food conversion efficiency of consumers. All of these measures will help determine the relative risk of Hg exposure in northern Australia compared with other locations globally.

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Table 1. Estimates of atmospheric wet deposition of mercury in various locations around the globe

Location	Deposition (μg m^{-2} $year^{-1}$)	Conditions	Reference
Guizhou Province, China	39	Mercury-mining area with high coal combustion	Feng *et al*. (2002)
Eastern United States	5–22	Numerous point sources; high population density	NADP (2007)
Western United States	2–9	Low population density	NADP (2007)
Sweden	10–14		Schroeder and Munthe (1998)
Norway	3–35	Decreasing from south to north	Iverfeldt (1991)
Japan	6–18	Highest in wettest areas	Sakata and Marumoto (2005)
Canadian Arctic	5–18	Estimated from sediments	Hermanson (1998)
Australia	n.a.		

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Table 2. Trophic level of predatory fishes in temperate regions compared with estimates for predatory fishes of northern Australia

All values are calculated with stable nitrogen isotopes, using the formula $TL_{fish} = (\delta^{15}N_{fish} - \delta^{15}N_{primary\ consumer})/3.4 + 2$. Numbers in italics indicate approximate values taken from figures in original articles

Species	System	Body size	Trophic level (mean, max.)	Reference
Temperate predatory fishes				
Lake trout, *Salvelinus namaycush*	Lake Ontario	n.a.	*4.9*, n.a.	Kiriluk *et al*. (1995)
Lake trout, *Salvelinus namaycush*	Laurentian Great Lakes	460–730 mm	n.a., *5.3*	Post *et al*. (2000)
Lake trout, *Salvelinus namaycush*	Boreal lakes	n.a.	n.a., 4.6	Vander Zanden and Rasmussen (1996)
Smallmouth bass, *Micropterus dolomieu*	Boreal lakes	n.a.	4.02, 4.73	Vander Zanden *et al*. (1997)
Largemouth bass, *Micropterus salmoides*	Boreal lakes	n.a.	4.08, 4.41	Vander Zanden *et al*. (1997)
Largemouth bass, *Micropterus salmoides*	Laurentian Great Lakes	250–440 mm	n.a., *4.5*	Post *et al*. (2000)
Northern pike, *Esox lucius*	Laurentian Great Lakes	450–840 mm	n.a., *4.0*	Post *et al*. (2000)
Northern pike, *Esox lucius*	Boreal lakes	n.a.	3.87, 4.51	Vander Zanden *et al*. (1997)
Walleye, *Sander vitreus*	Laurentian Great Lakes	300–700 mm	n.a., *4.3*	Post *et al*. (2000)
Walleye, *Sander vitreus*	Boreal lakes	n.a.	4.40, 4.86	Vander Zanden *et al*. (1997)
Walleye, *Sander vitreus*	Lake Champlain	350–499 mm	4.18, 4.45	Overman and Parrish (2001)
Walleye, *Sander vitreus*	Lake Champlain	500–549 mm	4.33, 4.95	Overman and Parrish (2001)

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Walleye, *Sander vitreus*	Lake Champlain	550–599 mm	4.68, 4.99	Overman and Parrish (2001)
Walleye, *Sander vitreus*	Lake Champlain	>600 mm	4.66, 4.93	Overman and Parrish (2001)
Northern Australia predatory fishes				
Saratoga, *Scleropages jardini*	Magela Creek	n.a.	3.39, n.a.	Douglas *et al*. (2005)
Fork-tailed catfish, *Arius leptasis*	Magela Creek	n.a.	3.57, n.a.	Douglas *et al*. (2005)
Fork-tailed catfish, *Arius midgleyi*	Ord River	n.a.	2.82, n.a.	Douglas *et al*. (2005)
Barramundi, *Lates calcarifer*	Magela Creek	n.a.	3.54, n.a.	Douglas *et al*. (2005)
Barramundi, *Lates calcarifer*	Ord River	n.a.	3.12, n.a.	Douglas *et al*. (2005)

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Fig. 1. Estimated growth trajectories for two temperate fish species (lake trout, *Salvelinus namaycush*, and walleye, *Sander vitreus*) and one tropical fish species (barramundi, *Lates calcarifer*). Weight-at-age estimates are adapted from Ferreri and Taylor (1996) for lake trout, Simoneau *et al*. (2005) for walleye and Pusey *et al*. (2004) for barramundi.

Fig. 2. Conceptual model of mercury (Hg) cycling in northern Australia. Boxes represent compartments where Hg can be stored and measured. Circles represent factors that can increase (+) or decrease (–) Hg in a given compartment. Solid arrows represent the flow of Hg through the system and hatched arrows show locations of effects.

Fig. 1

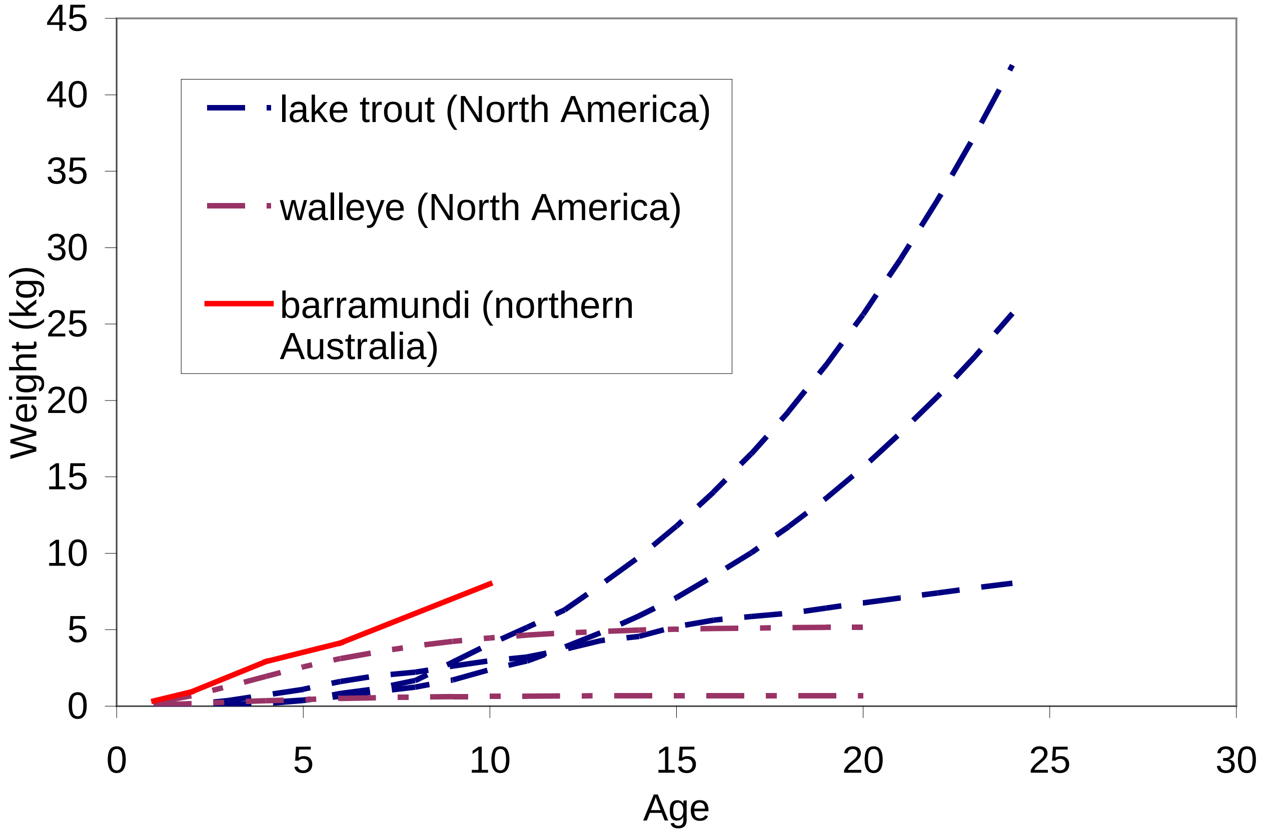
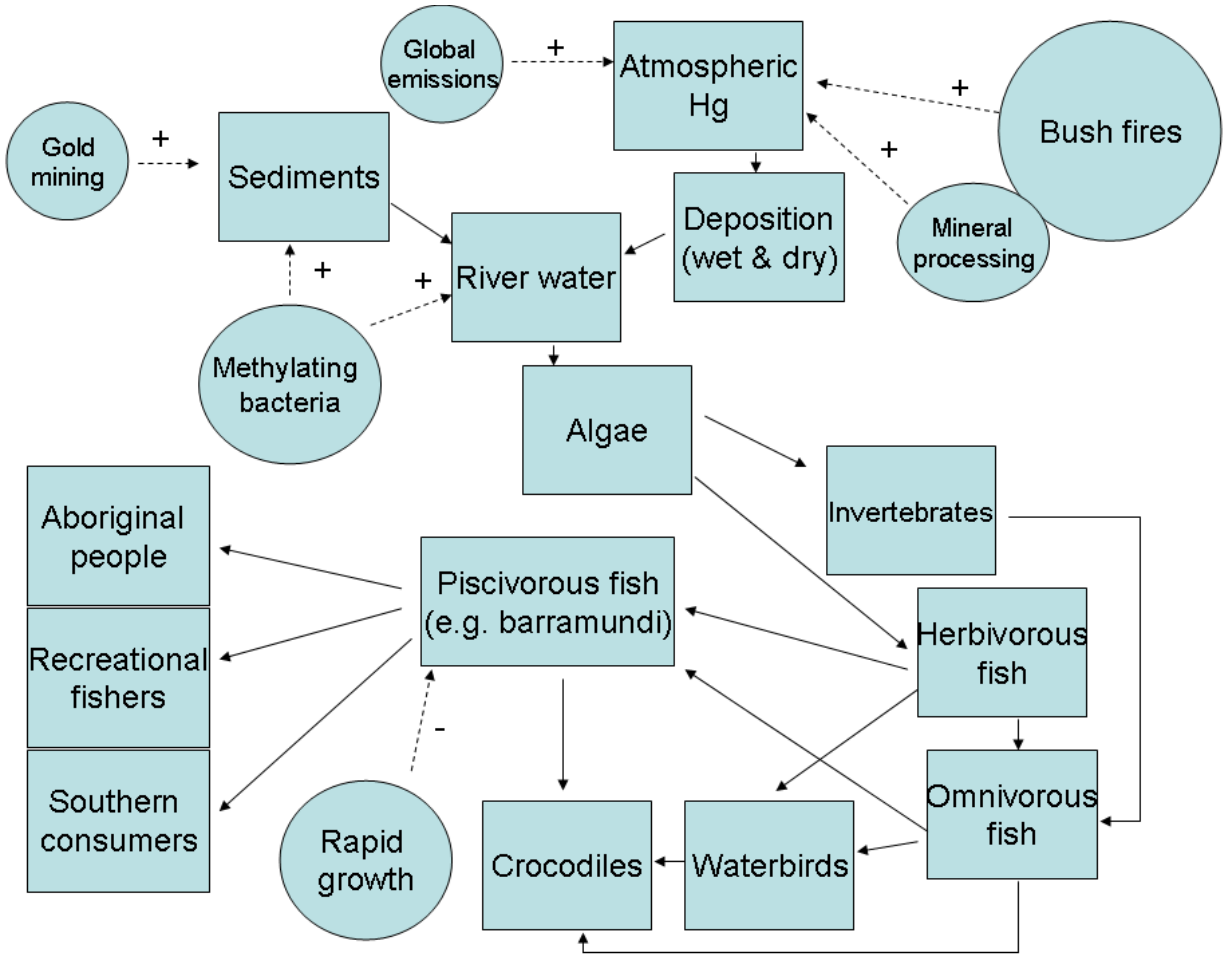

45
40
35
30
25
20
15
10
5
0
Weight (kg)
lake trout (North America)
walleye (North America)
barramundi (northern Australia)
0
5
10
15
20
25
30
Age

Fig. 2


Global emissions
+
Atmospheric Hg
+
Bush fires
Gold mining
+
Sediments
+
Mineral processing
Deposition (wet & dry)
River water
+
+
Methylating bacteria
Algae
Invertebrates
Aboriginal people
Piscivorous fish (e.g. barramundi)
Herbivorous fish
Recreational fishers
-
Southern consumers
Omnivorous fish
Rapid growth
Crocodiles
Waterbirds