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# Implications of coastal darkening for contaminant transport, bioavailability and trophic transfer in northern coastal waters

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## Coastal Darkening

Increased transport of terrestrially-derived organic material (tOM) from catchments to rivers and lakes is leading to *browning* of northern freshwaters (Creed *et al.,* 2018). Although the same environmental drivers can be expected to lead to widespread increases in tOM and subsequent *darkening* in adjacent coastal systems (Dupont & Aksnes, 2013), this has received considerably less attention, and long-term monitoring data for detecting these changes are scarce. This is despite expectations that darkening is likely to have a range of implications for coastal biogeochemistry and food-web ecology.

tOM is accompanied by other catchment-derived materials including contaminants, inorganic particles, and nutrients. Together, these *terrestrial inputs* can affect cycling and food-web accumulation of contaminants, particularly methyl mercury (MeHg) and persistent organic pollutants (POPs). Due to the human and ecosystem health risks associated with these contaminants, and to ensure informed policy decisions, there is need for better understanding of the drivers of darkening and their direct and indirect impacts on the *transport, bioavailability,* and *trophic transfer* of contaminants in northern coastal waters.

**Increased transport**

In northern regions, rising temperatures are leading to greener catchments and the acceleration of the hydrological cycle, both of which enhance the transport of sediments (particularly relevant in Arctic glaciated catchments) and tOM (which can be significant in boreal regions recovering from acidification) from catchments to adjacent surface waters. Hg and organic contaminants have a strong affinity for particles and dissolved tOM (Ripszam *et al.,* 2015). Thus, the flux of inorganic sediments and tOM facilitates the transport of contaminants from catchment soils to rivers, fjords and coastlines, directly influencing contaminant concentrations in surface waters and sediments and potentially increasing exposure of coastal biota.

Northern tundra and boreal soils contain more than twice as much Hg as the ocean, atmosphere and other soils combined (Schuster *et al.,* 2018). Snow, permafrost, sea-ice and glaciers also represent potentially important storage pools for POPs and Hg. Despite the potential for these pools of Hg and POPs to be mobilized through melting, permafrost slumps, and coastal erosion, pool sizes and susceptibility to mobilization and transport to surface waters remain poorly characterized. In addition, little is known regarding the fate of tOM and associated contaminants once they reach the marine environment. Contaminants bound to inorganic particles may settle out quickly in nearshore waters (e.g. in river estuaries and coastal lagoons) or be transported offshore if associated with finer particles or dissolved tOM. Changes in salinity and availability of iron and other metals can also drive increased contaminant flocculation and sedimentation. Research is needed to determine climate-sensitivity and the magnitude of future tOM and contaminant mobilization and transport as well as their fate in coastal waters.

**Reduced bioavailability**

Higher aqueous contaminant concentrations following increased inputs might lead to increased exposure for local biota, but uptake in coastal food webs also depends on contaminant bioavailability, which is determined by speciation (for Hg) and degree of sorption to inorganic particles and dissolved and particulate OM (for Hg and POPs). The bioavailable and highly neurotoxic organic methyl Hg (MeHg) is produced by inorganic Hg methylation by sulfur and iron reducing bacteria as well as in the water column in association with carbon remineralization. Degradation of MeHg can occur biotically (microbial demethylation via oxidative pathways) and abiotically (photodemethylation). Coastal darkening could potentially alter these processes by reducing light penetration, shifting microbial species composition and changing the fraction of freely dissolved elemental Hg available for methylation. In contrast, provision of tOM as a substrate for bacterial methylation may lead to increased concentrations of MeHg in darker waters. However, despite higher total concentrations, the tendency for all forms of Hg as well as POPs to bind to inorganic particles and tOM is likely to lead to reduced concentrations in the freely dissolved phase with increased tOM-loading in coastal waters.

**Changing food-web structure**

The movement of contaminants into and through coastal food webs is linked to basal food sources and energy flow pathways, both of which can be expected to change with darkening waters. Humic-rich tOM is generally considered to be refractory, and of limited bioavailability for lower trophic levels. However, recent studies have challenged this traditional view, with evidence of efficient bacterial utilization of tOM as well as coastal food-web reliance on terrestrial carbon sources. More extensive coastal darkening, which both attenuates light needed for photosynthesis and provides a substrate (tOM) for bacterial production, has the potential to increase the importance of heterotrophic food sources to higher trophic level organisms through the microbial loop.

A shift towards a microbial-based food web can lead to higher concentrations of biomagnifying contaminants in consumer organisms, since microbial food webs have additional trophic transfers compared to phytoplankton-based food webs, thus increasing the effective trophic level of consumers (Jonsson *et al*., 2017). At the same time, microbial food-sources have lower nutritional value, lacking essential fatty acids like docosahexaenoic acid (DHA) and eicosapentaenoic acid (EPA), suggesting that darkening could lead to reduced food-quality and trophic efficiency in coastal food webs.

Furthermore, additional effects of tOM-inputs, including higher sedimentation rates, can lead to changes in benthic and pelagic community composition due to changes in food availability and physicochemical conditions. A changing light environment is also expected to have implications for visual predators that may be unable to select for their preferred food-choices. These types of shifts in species composition, behavior, and trophic interactions are poorly understood, yet may be key to understanding the contamination of affected coastal food webs.

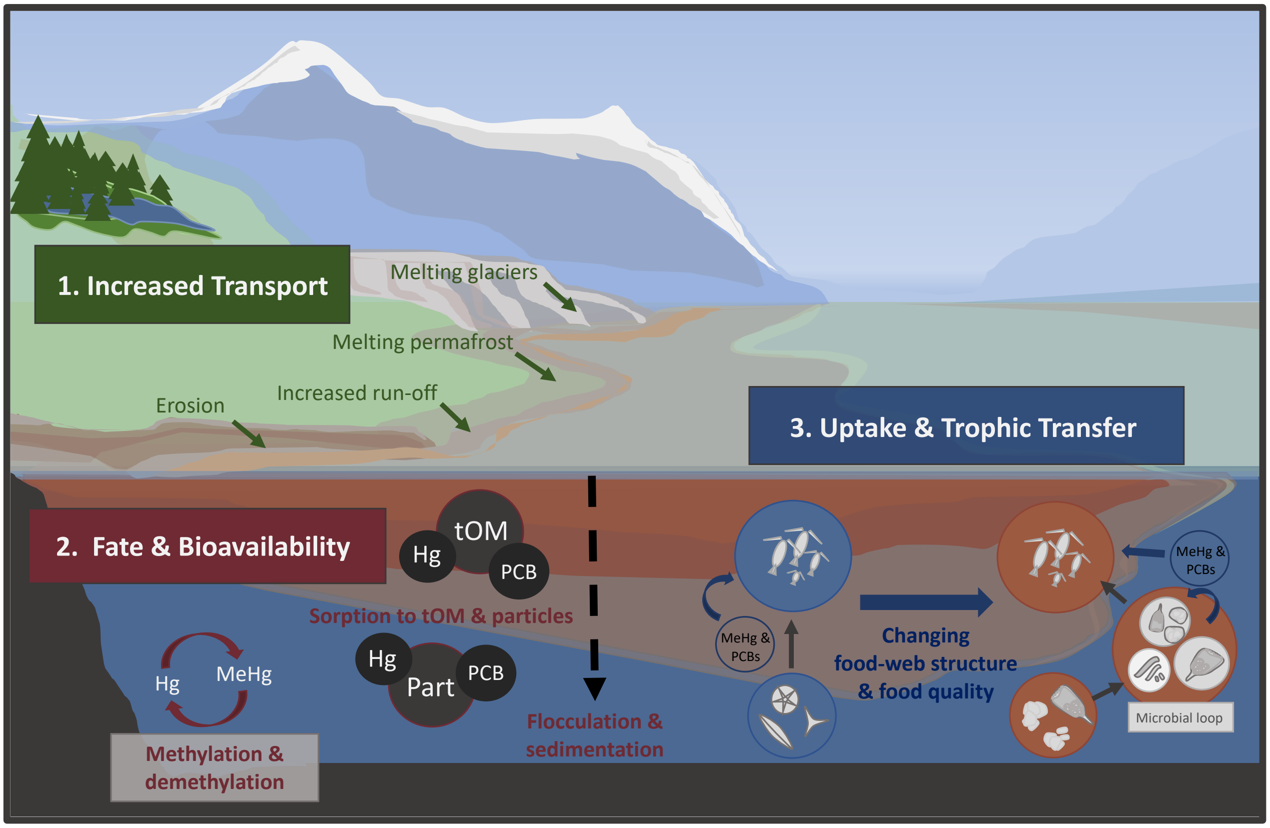


Figure 1. Local impacts of coastal darkening on contaminant (1) transport, (2) fate and bioavailability and (3) uptake and trophic transfer in northern regions.

**Research Needs**

Mirroring observations in freshwater systems, northern coastlines are darkening and the implications for contamination of coastal fauna depend on a complex set of often contradictory processes (figure 1). Considering the social and economic importance of our coastal zones, there is a strong need for knowledge on the extent of coastal darkening, and the potential physicochemical and ecological implications of darker waters. Understanding the impacts of darkening on transport, bioavailability and food-web accumulation of contaminants will require a combination of observational, experimental and modelling approaches, ideally along spatial, seasonal and latitudinal gradients.

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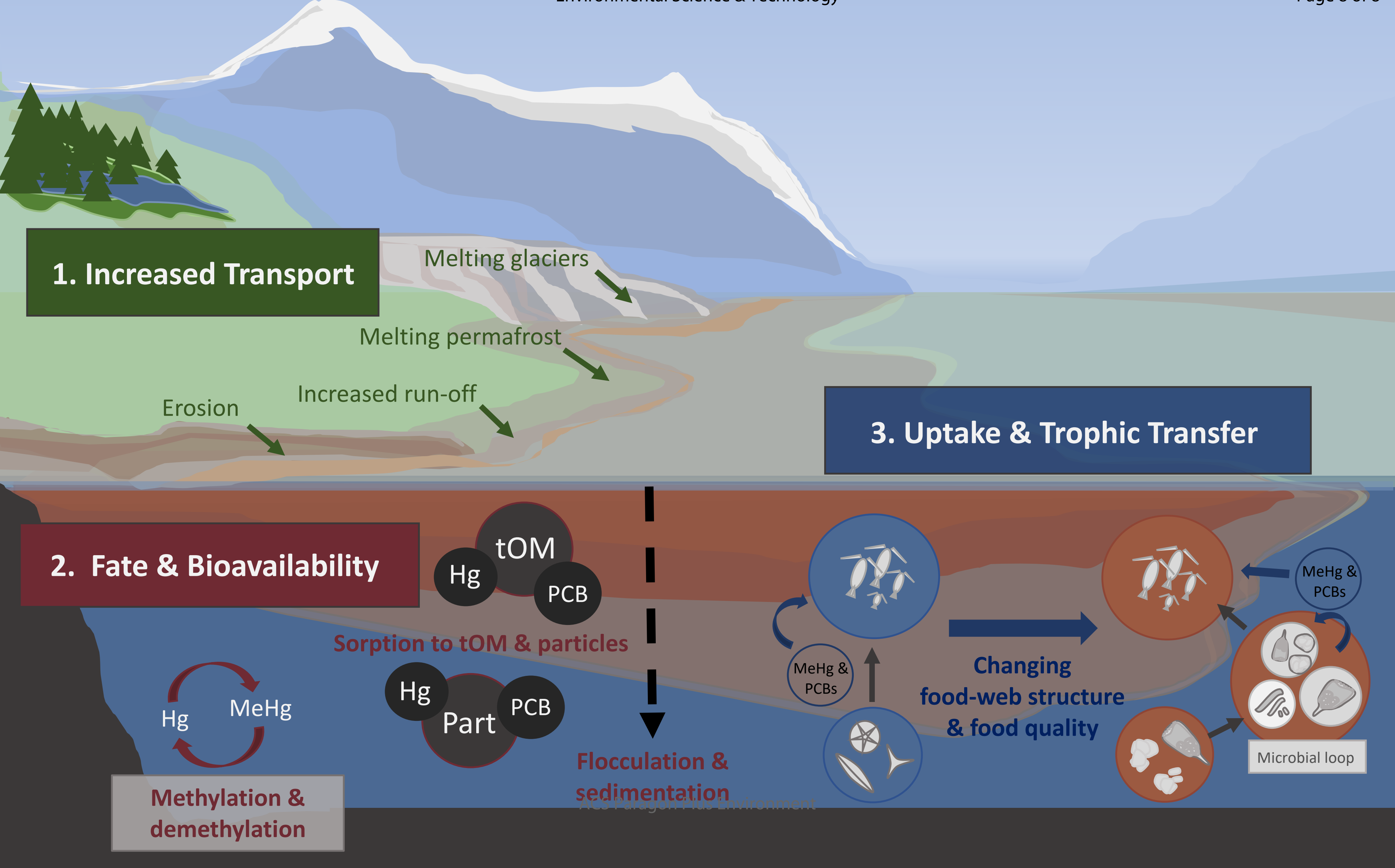
1. Increased Transport
Melting glaciers
Melting permafrost
Increased run-off
Erosion
3. Uptake & Trophic Transfer
2. Fate & Bioavailability
tOM
Hg
PCB
Sorption to tOM & particles
Hg
MeHg
Hg
Part
PCB
Flocculation & sedimentation
Methylation & demethylation
MeHg & PCBs
MeHg & PCBs
Changing food-web structure & food quality
Microbial loop