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Effects of Ultraviolet Radiation and Contaminant-related Stressors on Arctic Freshwater Ecosystems

Climate change is likely to act as a multiple stressor, leading to cumulative and/or synergistic impacts on aquatic systems. Projected increases in temperature and corresponding alterations in precipitation regimes will enhance contaminant influxes to aquatic systems, and independently increase the susceptibility of aquatic organisms to contaminant exposure and effects. The consequences for the biota will in most cases be additive (cumulative) and multiplicative (synergistic). The overall result will be higher contaminant loads and biomagnification in aquatic ecosystems. Changes in stratospheric ozone and corresponding ultraviolet radiation regimes are also expected to produce cumulative and/or synergistic effects on aquatic ecosystem structure and function. Reduced ice cover is likely to have a much greater effect on underwater UV radiation exposure than the projected levels of stratospheric ozone depletion. A major increase in UV radiation levels will cause enhanced damage to organisms (biomolecular, cellular, and physiological damage, and alterations in species composition). Allocations of energy and resources by aquatic biota to UV radiation protection will increase, probably decreasing trophic-level productivity. Elemental fluxes will increase via photochemical pathways.

INTRODUCTION

Changes in climate and ultraviolet radiation regimes in the Arctic will have far-reaching impacts, affecting aquatic species of varying trophic levels, the physical environment that makes up their habitat, the chemical properties of that environment, and the processes that act on and within freshwater ecosystems. Interactions of climate-related variables such as temperature and precipitation with freshwater ecosystems are highly complex and can be propagated through the ecosystem in ways that are not readily projectable. This reduces our ability to accurately forecast specific effects of climate and ultraviolet-radiation change upon freshwater systems. This is particularly the case when dealing with threshold responses, i.e., those that produce step-wise and/or non-linear effects. Moreover, our ability to project impacts is further hampered by our still relatively poor understanding of Arctic freshwater systems and their basic inter-relationships with climatic and other global environmental stressors such as persistent organic pollutants

(POPs), metals, and radionuclides which have been widely distributed into northern freshwater ecosystems through long-range atmospheric transport (1). In addition, other atmospheric-related environmental stressors such as alterations in UV radiation regimes will also have profound effects on freshwater ecosystems. Hence, climate change is very likely to act cumulatively and/or synergistically with other stressors to affect physical, chemical, and biological aspects of aquatic ecosystems.

The following sections discuss the projected synergistic effects and risks of UV radiation and contaminant-related stressors (in particular POPs and Hg) on the structure and function of freshwater ecosystems as a result of projected climate change in the Arctic. A broader treatment of projected alterations in stratospheric ozone and ultraviolet radiation in the Arctic is found in (2). (1, 3, 4, 5, 6) provide general treatments of the effects and potential risks of contaminant-related stressors on northern ecosystems. (7) further discusses the potential effects of UV radiation and contaminant stressors on freshwater fish populations and related fisheries, while (8) address responses to increased UV radiation in arctic tundra and polar desert ecosystems and (9) for marine systems.

ULTRAVIOLET RADIATION EFFECTS ON FRESHWATER ECOSYSTEMS

Ultraviolet radiation is the most photochemically reactive wavelength of solar energy reaching the surface of the earth, and has a broad range of effects on aquatic biogeochemistry, biota, and ecosystems. As a result of anthropogenic impacts on the atmosphere of the earth, UV radiation exposure in arctic environments is changing substantially. Although anthropogenic emissions of ozone-depleting substances have declined since the ratification of the Montreal Protocol and its amendments, future levels of ozone and UV radiation in the Arctic are uncertain, depending not only on continued compliance with the Protocol and changes in legislation, but also on climate change effects on temperatures and trace gases (also see 2, 10, 11). In the subsequent sections we provide an overview of how underwater UV radiation exposure is linked to climate, followed by a discussion of general principles concerning UV radiation impacts in aquatic ecosystems (including natural protection mechanisms) and a systematic analysis of potential UV radiation impacts on arctic freshwater habitats.

Box 1 Implications of changing snow and ice cover for ultraviolet radiation exposure

The warming northern climate is prolonging open water conditions. The loss of UV radiation-attenuating snow and ice earlier in the season, when water temperatures are still low but UV irradiances are maximal, is likely to be especially stressful for aquatic biota. As shown below, white ice (ice with air inclusions) has a strong attenuating effect on PAR (visible light) and an even greater effect on UV-A and UV-B radiation. This snowclearing experiment on Hudson Bay showed that only 2 cm of snow reduced the below-ice exposure to UV radiation and to PAR by about a factor of three, with slightly greater effects at the shorter wavelengths.

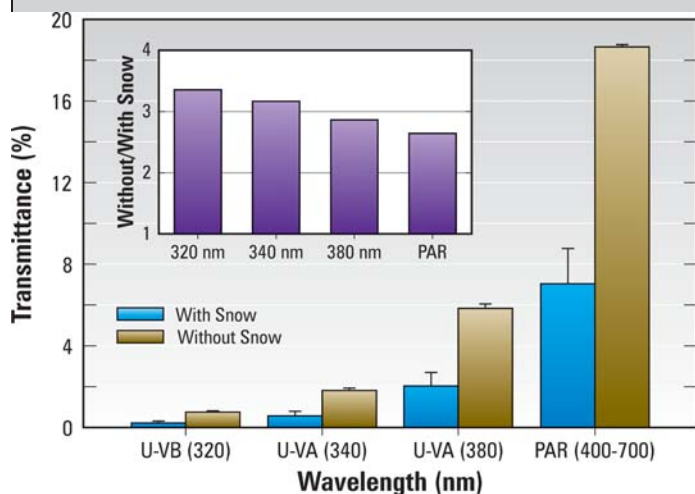


Figure 1. The UV radiation-attenuating effect of ice and snow over the plume of the Great Whale River, in Hudson Bay, Northern Québec, immediately offshore from the river mouth (April 1999). The ice was 1 m thick with white ice at its surface, and the snow was 2 cm thick. The percentage transmittance values are for the penetration of each wavelength into the water beneath before and after the removal of the snow. Inset shows the ratio of transmittance through the ice in each wavelength after (without snow) relative to before (with snow) the 2 cm of snow was cleared from the ice (adapted from 12).

Climate effects on underwater ultraviolet radiation exposure

To understand the overall impact of changes in UV radiation levels, the synergistic and antagonistic processes resulting from climate change have to be considered since they have the potential to modify the underwater UV radiation regime and consequently the stress on aquatic organisms. Climate change is very likely to be accompanied by shifts in biological UV radiation exposure in arctic river, lake, and wetland environments via three mechanisms (12): changes in stratospheric ozone levels, changes in snow- and ice-cover duration, and changes in the colored materials dissolved in natural waters that act as sunscreens against UV radiation.

Although it is projected that the downward trends in ozone levels are likely to reverse in the near future as a consequence of reduced anthropogenic emissions of chlorofluorocarbons (CFCs) and related compounds, some of the longer-lived ozone-depleting substances are still accumulating in the stratosphere and climate change is likely to prolong the effects of depletion. Temperature increases in the troposphere are projected to be accompanied by temperature decreases in the lower stratosphere, and there is already some evidence of this effect in the polar regions. Temperature decreases in the lower

stratosphere are very likely to increase the frequency and extent of polar stratospheric clouds (PSCs) that catalyze CFC–ozone reactions, and result in a strengthening of the polar vortex, which in turn is likely to lead to longer-lasting conditions for ozone depletion (13). The minimum winter temperatures in the arctic stratosphere are very close to the threshold for the formation of PSCs and the chlorine reactions that lead to ozone loss, and the Arctic remains vulnerable to large-scale ozone depletion (14). It is also possible that temperature increases could lead to increased zonal flow at midlatitudes causing the polar vortex to be more stable, again favoring ozone depletion and a delay in the eventual recovery of the ozone layer (15). Furthermore, as greenhouse gas concentrations increase, the tropical tropopause is very likely to become warmer, resulting in the transport of more water vapor into the stratosphere, which in turn is likely to lead to the formation of PSCs at higher temperatures (16). Therefore, the ozone in the arctic stratosphere would be at greater risk of depletion (see 2 for more detailed discussion).

The underwater UV radiation environment changes dramatically with a decrease in snow- and ice-cover duration, especially if this occurs during periods of greatest UV radiation flux and ozone depletion. Analyses of the effects of melting arctic sea and lake ice show that this process results in order-of-magnitude increases in biological UV radiation exposure that greatly exceed those caused by moderate ozone depletion (Box 1); (12). Lake and river ice are relatively transparent to UV radiation because of CDOM exclusion from the ice during freeze-up (17). Small changes in snow cover and white ice, however, can radically influence the below-ice UV radiation levels in arctic waters (18).

In arctic aquatic environments, variations in suspended particulates, and especially CDOM, affect transmission of UV radiation (see Box 2). These variations can be more important than ozone depletion in determining the UV radiation exposure in the water column of freshwater systems.

In some areas of the Arctic, climate change is very likely to be accompanied by increased vegetation, a concomitant increase in CDOM loading (30), and reduced exposure to underwater UV radiation. These positive effects are likely, however, to be offset by reduced availability of PAR (28, 31, 32). Marked south–north gradients in present-day CDOM concentrations in arctic waters are associated with the latitudinal distribution of terrestrial vegetation. Colored dissolved organic matter loading of freshwater systems is less pronounced at higher latitudes. Lakes in the tundra and polar-desert biomes contain low amounts of these materials; small variations in CDOM concentration in these systems can cause major changes in underwater UV radiation exposure (21, 33). Freshwaters in northern Scandinavia are low in dissolved carbon similar to water bodies in North America; the median DOC concentration for 25 lakes above the treeline in Finnish Lapland was 18 mg C/l (34). Although acid precipitation enhances underwater UV radiation levels by reducing DOC concentrations in the water, increased thawing of permafrost with climate change is very likely to increase soil runoff and levels of DOC (or CDOM) in arctic freshwater systems. This is very likely to be accompanied by an increase in water turbidity, which will probably not only decrease PAR penetration but also increase the relative proportion of UV radiation, thereby hindering repair processes in aquatic organisms that are stimulated by longer wavelengths. Increased physical turbulence is also likely to expose planktonic organisms to unfavorable irradiance conditions (e.g., exposure to high levels of surface UV radiation and PAR), the effects of which are likely to be especially severe for species that cannot migrate.

Box 2

Colored dissolved organic matter: the natural sunscreen in arctic lakes and rivers

Colored dissolved organic matter (CDOM) is composed of humic and fulvic materials (average to low molecular weight) that are derived from terrestrial soils, vegetation, and microbial activities, and is known to be an effective protective screen against UV radiation for freshwater biota (e.g., plankton, 19; amphibians, 20). These compounds absorb UV-A and UV-B radiation and short-wavelength visible light, and in high concentrations such as in arctic rivers they stain the water yellow or brown. Colored dissolved organic matter is now known to be the primary attenuator of underwater UV radiation in subarctic and high-arctic lakes (21); Toolik Lake, Alaska (22); arctic ponds (23); and arctic coastal seas

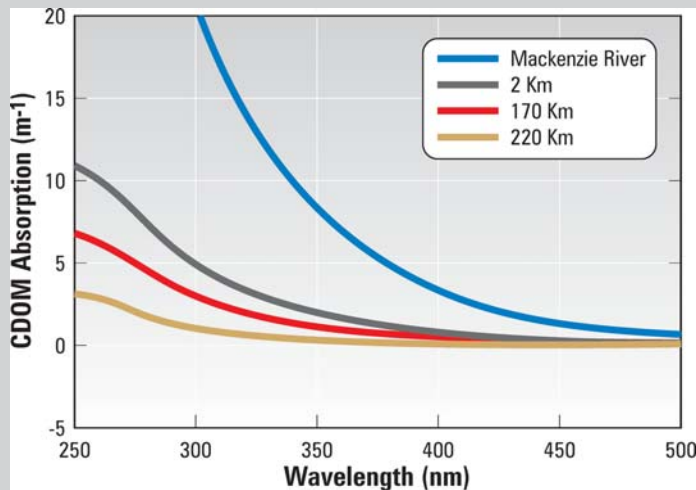


Figure 2. Ultraviolet (<400 nm) and blue-light (400–500 nm) radiation absorption by CDOM in the Mackenzie River (Inuvik, October 2002). The lower curves are for surface samples near the same date in the Beaufort Sea showing the CDOM influence at 2, 170, and 220 km offshore from the mouth of the river (29).

influenced by river inflows (24, 12). The concentrations of CDOM in natural waters are influenced by pH (acidification can cause a severe decline; 25), catchment morphology, runoff, and the type and extent of terrestrial vegetation. The latter aspects are especially dependent on climate.

The paleoecological record has been helpful in examining past impacts of climate on biological underwater UV radiation exposure, specifically by using fossil diatoms in lake sediments as quantitative indicators of variations in CDOM. This record also underscores the large regional differences in the magnitude and direction of change in underwater UV radiation levels (26; 27). Shifts in vegetation and hydrology caused by warming or cooling trends are very likely to affect the quantity of CDOM exported from catchments to their receiving waters, in turn affecting underwater UV radiation levels. For example, an analysis of the past underwater climate (paleo-optics) of subarctic treeline lakes indicated that recent Holocene cooling (from about 3500 years ago to the present) was accompanied by a southward retreat of the treeline and a large decrease in CDOM concentrations in lake waters. This decreasing CDOM resulted in an increase in biological UV radiation exposure that was two orders of magnitude greater than that associated with moderate (30%) ozone depletion (28). (27) reported large decreases in biological UV radiation exposure (starting about 3000 years BP) in a coastal lake in subarctic Québec that were associated with the establishment of terrestrial vegetation in its catchment.

Ultraviolet radiation effects on aquatic biota and ecosystems

The effects of UV radiation in the aquatic environment range from molecular to whole-ecosystem. Photobiological damage includes the direct effects of UV radiation in which photons are absorbed by biological molecules such as nucleic acids and proteins that then undergo photochemical alteration. An alternative damage pathway is via the interaction of UV radiation and organic compounds or other photosensitizing agents to produce reactive oxygen species such as superoxide and hydroxyl radicals. These can diffuse away from the site of production and cause oxidative damage to enzymes, lipid membranes, and other cellular constituents.

Aquatic biota have four main lines of defense against UV radiation damage: escape, screening, quenching (chemical inactivation), and repair. The net stress imposed by the UV radiation environment reflects the energetic costs of protection and repair in addition to the rate of photochemical degradation or alteration of cellular components (35). These defenses are well illustrated by arctic zooplankton (see Box 3), but despite this protection these organisms remain vulnerable to ambient UV radiation levels (36), particularly in the cold, shallow, CDOM-poor waters that characterize many arctic lakes and ponds.

Changes in underwater UV radiation exposure are likely to directly affect the species composition of aquatic biota at each trophic level, as well as cause effects that cascade throughout the benthic (e.g., 48) or pelagic (e.g., 49) food webs and the coupling between them. Some trophic responses are likely to be “bottom-up effects” in that UV radiation exposure reduces the quantity or quality of prey and thereby reduces food supply to the next level of consumer organisms. This could occur, for example, via shifts towards inedible or less digestible algal species (50; 51) or by reducing the nutritional value of food

organisms (52). The effects of variations in UV-B radiation on the quality of phytoplankton photosynthetic products have received little attention except for studies at the pigment level (e.g., 47, 53). Any alteration in the biochemical composition of primary producers is likely to change the nutritional value of food consumed by grazers (thus influencing energy flow throughout the food web) as well as restrict the production of photoprotective compounds against UV radiation. Short-term exposure to enhanced UV-B radiation levels in phytoplankton populations of various lakes in the Canadian High Arctic influenced the allocation of newly fixed carbon into the major macromolecular classes (46). Generally, synthesis of both protein and polysaccharides was inhibited by enhanced UV-B radiation levels, and the photosynthate would remain or accumulate in the pool of low molecular weight compounds. Lipid synthesis was insensitive to UV-B radiation levels and represented the most conservative and uniform class, accounting for about 20% of total carbon fixed.

Overall, these results were similar to those observed for Lake Ontario (54). However, the various classes of lipid may respond differently to variations in UV-B radiation levels. For example, exposure to UV radiation influenced fatty acid composition in algal cultures (55; 56). Other studies observed that the effect of UV-B radiation on the major lipid classes is species-specific (57).

Higher trophic levels are dependent on phytoplankton either directly as food or indirectly via trophic cascades. Inhibition of growth and cell division in phytoplankton will most often affect the food quality of these cells by placing stoichiometric constraints on the grazer (58). (59) also showed that water hardness could be a major determinant of susceptibility to UV radiation damage among calcium-demanding species such as *Daphnia*. They suggested that calcium, which is an important

Box 3

Ultraviolet radiation protection and recovery mechanisms in arctic freshwaters

Aquatic organisms have varying abilities to counter the effects of UV radiation. Photoprotective and repair processes are particularly important in preventing and reversing UV radiation damage to photosynthetic mechanisms. A range of potential repair processes is stimulated by longer wavelengths to counteract the damaging effects of UV radiation. The relative importance of repair versus protection will vary depending on specific conditions and the physiological characteristics of the species assemblage (32). Organisms living in arctic lakes have evolved several strategies to cope with UV radiation, which play an important role in shallow and highly UV-transparent arctic lakes and ponds. Some species of algae and zooplankton have an ability to reduce their exposure to UV radiation by vertical migration, which may be a response to high intensities of UV radiation (37). (38, 39) provided further evidence that organisms avoid highly irradiated areas by escaping the brightly lit surface zone.

In addition to avoidance, aquatic organisms can escape from UV radiation by reducing the effective radiation that penetrates the cell. A number of UV-protecting compounds have been described; the three major types are carotenoids, mycosporine-like amino acids (MAAs), and melanin. The photoprotective properties of carotenoids are mainly associated with antioxidant mechanisms and inhibition of free radicals, as opposed to direct UV radiation screening (40). Carotenoids absorb wavelengths in the visible light spectrum and do not therefore provide direct protection from UV radiation. Mycosporine-like amino acids have absorption maxima ranging from 310 to 360 nm within the UV wavelength range. They are present in alpine phytoplankton and zooplankton (41; 42) and also occur in arctic freshwater organisms although there is no research on this. Animals are unable to synthesize MAAs and carotenoids, and must therefore acquire these compounds from their diet.

Cladocera and fish produce melanin, with absorption maxima between 250 and 350 nm. Melanin acts by absorbing radiation before it enters the body tissues, and its synthesis seems to be a direct response to UV radiation (43). Melanin zooplankton are typically found in clear arctic waters

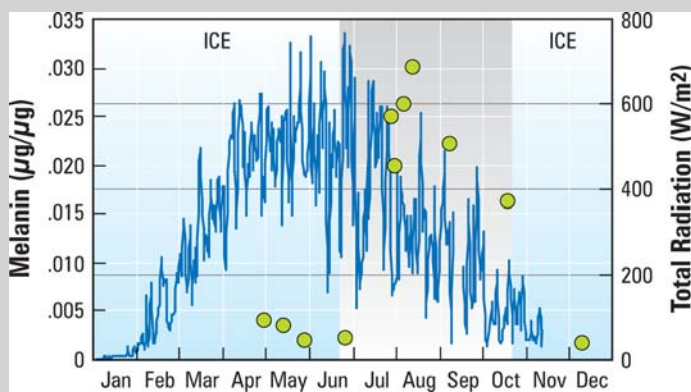


Figure 3. Changes over time in solar radiation (solid line) and the rise in the UV-screening pigment melanin (data points) in the zooplankton *Daphnia umbra* in Lake Saanajärvi, northern Finland, immediately after ice-out (adapted from 23).

where the absorbance of UV radiation is low, and in shallow ponds where high DOC levels may not provide enough protection from UV radiation (34, 44). It has also been shown that melanin synthesis followed the annual variation in UV radiation levels (i.e., synthesis peaked at the time of maximum underwater UV irradiance, see figure). Aquatic organisms can also repair damage from UV radiation by nucleotide excision repair or by photoreactivation mechanisms, such as photoenzymatic repair (45).

Brief exposure to UV radiation triggers only the initial UV radiation stress response. However, responses over long periods show that organisms can acclimate to the UV radiation stress and/or recover growth rates with the development of photoprotective strategies (e.g. the synthesis of photoprotective compounds). A long-term enclosure experiment conducted in a high-arctic lake on Ellesmere Island (Nunavut, Canada) showed an initial decrease in phytoplankton productivity with enhanced UV-B radiation exposure, with recovery after 19 days (46). Long-term acclimation to and recovery from increased levels of UV radiation were also observed in a cultured marine diatom (47). Antecedent light conditions, temperature, nutrient availability, and/or variations between species are all factors that can affect acclimation of organisms to high intensities of UV-B radiation (47).

element for invertebrates with calcified exoskeletons, in low concentrations (low-pH lakes, acidification) could reduce the stress tolerance of organisms. Although several studies of freshwater invertebrate species have reported increased mortality in response to increased UV radiation levels, especially zooplankton (23; 45; 60; 61; 62; 63), the variation in UV radiation tolerance is high among species and life stages (45). In general, small zooplankton (small rotifers) are considered to have a high UV radiation tolerance, while large species vary in their tolerance both among and within species. (45) found that cladocerans had the lowest UV radiation tolerance and exhibited high variability among species. *Daphnia* was one of the most sensitive groups of organisms, while adult calanoid and cyclopoid copepods had high UV radiation tolerances. In a comparison of lakes across a successional gradient of catchment vegetation and thus CDOM content, three zooplankton species (*Asplanchna priodonta*, *Ceriodaphnia quadrangula*, and *Bosmina longirostris*) were absent from low-CDOM, UV-transparent waters, and perished when transplanted from a CDOM-rich lake in the series and held at 0.5 m depth under full UV radiation exposure in a clear lake. In contrast, two species that avoided high UV radiation exposure in the near-surface waters (*Daphnia pulicaria* and *Cyclops scutifer*, a highly UV-tolerant species) occurred in even the clearest lakes (38). Morphotypic and biochemical differences among populations of a given species may also play an important role. Pigmented clones of *Daphnia* were more tolerant of UV radiation than transparent

clones (64), and pigmentation appears to increase in response to increased UV radiation exposure (34; Box 3). Studies of the effects of natural and enhanced UV radiation levels on fish are rare, but laboratory experiments have shown that high-latitude species of trout have sunburns, increased fungal infections, and higher mortality when exposed to increased dosages of UV radiation (65).

Other trophic responses are likely to be top-down effects, in which some species are released from grazing pressure or predation by UV inhibition of the consumers and thereby achieve higher population densities (48). This complex combination of direct and indirect effects makes any future shifts in aquatic ecosystem structure extremely difficult to project. In addition, the deleterious effects of UV-B radiation at the community level are difficult to assess since they are generally species-specific. For example, 66 showed that the responses of the planktonic microbial communities in Greenland ponds to ambient UV-B radiation levels varied greatly between species, especially rotifers and ciliates.

Multiple factors seem to affect amphibians negatively. These factors include both site-specific, local effects (e.g., pesticide deposition, habitat destruction, and disease) as well as global effects (e.g., increased UV-B radiation exposure and climate change; e.g., 67). Amphibians have been the focus of special interest at temperate latitudes because of the recent widespread decline in many frog populations and the recognized value of these organisms as sensitive indicators of environmental change.

Although many amphibians can be relatively resistant to UV-B radiation, it can cause deformities, delays in development, behavioral responses, physiological stress, and death in frogs. The rise in UV-B radiation levels associated with stratospheric ozone depletion has been widely promoted as one of several hypotheses to account for their decline (68), and references therein). However, the effects are controversial and in many habitats where the frogs are declining, the animals are well protected by CDOM (Box 2). A small number of frog species occur in the subarctic and the Arctic, including the common frog (*Rana temporaria*) and the wood frog (*Rana sylvatica* – North America), with distributions extending north of the Arctic Circle. Contrary to expectation, however, these populations may experience higher UV radiation exposures under natural conditions relative to temperate regions, and therefore be more pre-adapted, because of lower concentrations of UV-screening CDOM in high-latitude waters (e.g., 20) and life-cycle characteristics (phenology: higher UV-B radiation doses during the breeding season at higher latitudes; 69). There is considerable variation in UV radiation tolerance between amphibian strains and species; for example, a latitudinal comparison in northern Sweden found that *R. temporaria* embryos were relatively tolerant of UV-B radiation, with no clear latitudinal differences (70). The positive and negative effects of climate change on arctic habitats (e.g., duration of open water, extent of wetlands) are likely to have much greater impacts on amphibians than changes in UV radiation exposure.

Impacts on physical and chemical attributes

The large arctic rivers are relatively protected from UV radiation exposure because of their high CDOM content (24). Conversely, natural and increased UV radiation levels are likely to be important for photochemical loss of carbon from these systems. For example, there is evidence that the duration of ice-free conditions has increased in the Mackenzie River, Canada (71) and the River Tornio, Finland. The resultant increased exposure to UV radiation is likely to favor increased annual rates of UV degradation of riverine DOC, with possible impacts on the inshore coastal waters that receive these inputs.

In addition to having low CDOM concentrations and resultant deep penetration of UV radiation, many arctic lakes, ponds, and wetlands are shallow systems. The mean measured depth for more than 900 lakes in northern Finland was less than 5 m (72), and was about 5 m for 31 lakes in the vicinity of Tuktoyaktuk (Northwest Territories; 73) and 46 lakes on Ellesmere Island (Nunavut; 74). Consequently, all functional groups, including the benthos, are often exposed to UV radiation throughout the entire water column. In addition, many aquatic species stay in the offshore pelagic zone. Even species that are more benthic or littoral are protected minimally by macrophytes, as arctic waters, especially those in barren catchments, often contain little aquatic vegetation.

Changes in UV radiation exposure in these northern ecosystems are amplified by the low CDOM concentrations. Most have concentrations of DOC below 4 mg DOC/l, the threshold below which there are marked changes in UV radiation penetration through the water column, and in the ratio of wavelengths controlling the damage–repair balance, with only minor changes in CDOM (21).

The initial impacts of climate change are likely to be associated with the loss of permanent ice covers in the far northern lakes: this appears to have already recently taken place in the Canadian High Arctic (17). These effects are likely to be amplified by prolonged open-water conditions in lakes and ponds. However, other physical changes in these environments (e.g., wind-induced mixing) are likely to have greater perturba-

tion effects than those associated with increased UV radiation exposure. Although increases in CDOM are very likely to mitigate the effects of increased UV radiation levels, decreases in PAR are very likely to hamper photosynthesis. Furthermore, increased turbidity associated with thawing permafrost is likely to further reduce the exposure of organisms to damaging UV radiation (for turbidity effects on UV radiation, see 75).

The photochemical effects of increased UV radiation levels are also likely to influence the toxicity of contaminants (see also contaminant section below). Mercury (Hg) is the principal toxic chemical of concern in the Arctic and elsewhere. Methyl mercury (MeHg) is the most toxic form and the only form that biomagnifies in food chains. It was recently shown that: ultraviolet radiation exposure photoreduces divalent mercury (Hg²⁺, the soluble form in lakes) to elemental mercury (Hg⁰, the form that can volatilize from lakes; 76); ultraviolet radiation can also influence photooxidation (the conversion of Hg⁰ to Hg²⁺; 77); the formation of MeHg in arctic wetlands is very sensitive to temperature; ultraviolet radiation photodegrades MeHg; and, most of the Hg in recently fallen snow moves back to the atmosphere within a few days of exposure to solar radiation (77).

Not only is photochemical reduction of Hg important in the Arctic, but microbial reduction and oxidation may also occur as shown previously in temperate lake waters. Microbial oxidation is turned on by a hydrogen peroxide-dependent enzyme likely triggered by photochemical production of hydrogen peroxide (78). The interactions between UV radiation, temperature, and pH can alter Hg mobilization and speciation, and regulate the levels of Hg in organisms at the base of the food web. Photochemical events during spring are likely to be especially sensitive to rising UV-B radiation levels. Large quantities of Hg are photochemically oxidized and precipitate out of the arctic atmosphere with the first sunlight each spring, resulting in a rapid rise in Hg concentrations in snow; 24-hour variations in these atmospheric Hg-depletion events correlate with fluctuations in UV-B irradiance (79). Increasing spring levels of UV radiation due to stratospheric ozone depletion would probably enhance this so-called “mercury sunrise” phenomenon.

Wetlands and peatlands are rich in CDOM and the aquatic biota are therefore well protected from UV radiation exposure.

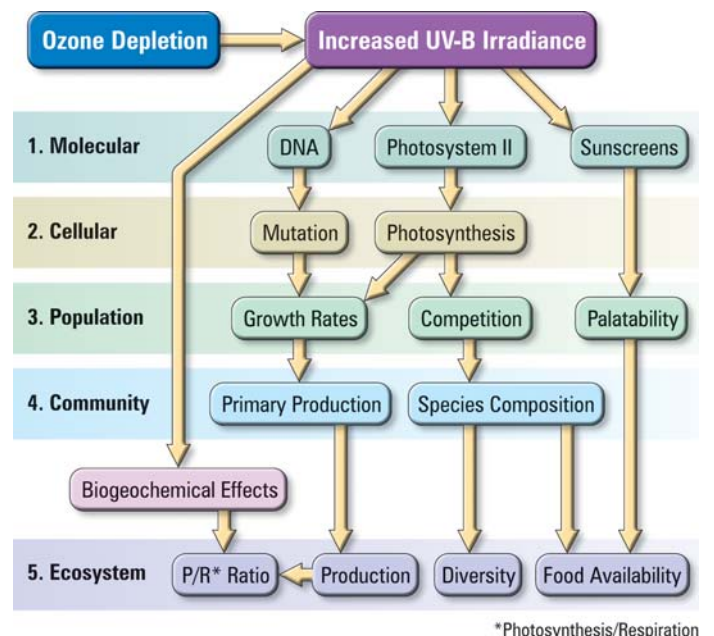


Figure 4. Ultraviolet radiation is the most photochemically reactive wavelength in the solar spectrum and has a wide range of effects, from molecular to whole ecosystem (19).

Early loss of snow and ice, however, is likely to increase exposure during a critical growth phase. Photochemical processes may be especially active in these shallow waters, and this mechanism of CDOM loss is very likely to accelerate with temperature increases (snow-cover loss) and ozone depletion.

Impacts on biotic attributes

Mild increases in UV radiation levels are likely to stimulate biological processes via photochemical release of low molecular weight organic carbon substrates and nutrients. More severe increases are likely to cause damage and/or a shift toward UV-tolerant species with a potential loss of diversity or other unique ecosystem attributes (Figure. 4).

Rivers and streams. Benthic mats and films are a common feature of highlatitude streams as well as many ponds, lakes, and wetlands, and are often dominated by cyanobacteria, especially the nitrogen-fixing genus *Nostoc* and filamentous species of the order Oscillatoriales (80). These communities commonly occur in shallow water systems where UV radiation exposure is likely to be high. Ultraviolet radiation has a broad range of deleterious effects on benthic cyanobacteria including on their pigment content, nitrogenase activity, photosynthesis, and respiration (81; 82). Much of the literature, however, reports experiments conducted under unrealistically high UV radiation dosages provided by artificial lamps, and many of the effects are likely to be much less apparent or absent in natural ecosystems, even under conditions of severe ozone depletion (35). Periphyton and benthic invertebrates are well protected given their avoidance and sunscreen capacities. Zooplankton and phytoplankton communities are well developed in large arctic rivers (e.g., 83), however, they are generally protected by high CDOM concentrations in these waters.

Lakes, ponds, and wetlands. In the Arctic, lake organisms have to cope with low nutrient conditions and/or low food availability, low temperatures, and short growing seasons (3–5 months). The UV radiation damage–repair balance may be especially sensitive to features of the arctic freshwater environment. Most of these ecosystems are oligotrophic and phytoplankton are therefore commonly limited by nutrient supply (84) in addition to low temperatures (83). As a result, the photosynthetic rates per unit biomass tend to be extremely low, even in comparison with other low temperature systems such as sea ice, polar oceans, and low-temperature cultures (85). Because of the low temperatures and low nutrients, phytoplankton photosynthetic rates are extremely low in Canadian high-arctic lakes (usually less than $1.5 \mu\text{g C}/\mu\text{g Chl-a/hr}$) and phytoplankton perform very poorly under high-light regimes (46, 86; 87). There have been few explicit tests of the effect of temperature on UV radiation damage of planktonic systems (83). However, since enzymatic processes are temperature-dependent (whereas damage induction is not), the slow metabolic rates of northern phytoplankton are likely to have a direct effect on the net stress imposed by increased UV radiation exposure by reducing all cellular processes including the rate of repair of photochemical damage. In the antarctic waters, for example, low temperatures drastically reduce repair to the extent that algal cells failed to show any photosynthetic recovery for at least five hours after UV radiation exposure (88). The low nutrient conditions that characterize northern lakes are likely to further compound this effect by reducing the availability of elemental resources for building enzyme systems involved in the functioning of the cell, including the repair of UV radiation damage, and are also likely to limit the investment in photoprotective mechanisms. Moreover, the lower temperatures probably reduce the affinity of the phytoplankton cells for

nutrient uptake by membrane transport processes (89), thereby increasing nutrient limitation.

The paleoclimatic record has also provided insights into the possible effects of past climate change on UV radiation exposure in aquatic ecosystems. For instance, analyses of fossil diatom assemblages in northern and alpine lake sediments have indicated that variations in underwater UV irradiance during the Holocene had major impacts on algal community structure and productivity (28, 90). Paleo-optical studies from subarctic lakes have revealed large fluctuations in biologically damaging underwater UV irradiance over the last 6000 years, accompanied by pronounced shifts in algal species composition and changes in the balance between benthic and pelagic primary producers (28). (91) observed similar effects in low-CDOM mountain lakes.

There are only a few studies of temperature-dependent UV radiation damage to zooplankton (92). In general, it is assumed that low temperatures would slow down the UV radiation damage repair mechanisms such as DNA repair and detoxification of reactive oxygen species. However, contrary to expectations, (92) found that reduced temperatures increased survival among UV irradiated *Daphnia*. They argued that although repair mechanisms are slower in the cold, UV-triggered activation processes (such as reactive oxygen species metabolism and lipid peroxidation) also slow down with decreasing temperature, thereby increasing *Daphnia* survival.

At ambient levels, UV-B radiation can contribute up to 43% of the photoinhibition of photosystem II function in phytoplankton populations of Canadian high-arctic lakes (as measured by both *in vivo* and dichlorophenylmethyl urea-enhanced fluorescence) as well as decreasing (by up to 40%) phytoplankton productivity rates near the water surface (93). The smallest size fraction (i.e., picoplankton: $0.2\text{--}2 \mu\text{m}$) usually represents more than 50% of total phytoplankton productivity in high-arctic lakes (93). Oligotrophic conditions tend to select for small cells with a high surface-to-volume ratio that favors nutrient transport at low substrate concentrations. Small cells are especially sensitive to UV radiation because they have high illuminated surface-to-volume ratios, little self-shading, and low effectiveness of screening pigments (94; 95). Even the production of UV radiation sunscreens is unlikely to confer much protection given the short path length in these cells (96), and studies of a variety of organisms have shown that enhanced UV radiation exposure inhibits the growth of larger cells less than that of smaller cells (94). (97) evaluated the size dependence of UV radiation effects on photosynthesis in subarctic lakes with a series of short-term photosynthetic experiments, which showed that, in contrast to expectations, the smaller cells were more resistant to UV radiation than larger cells. This smaller cell fraction was dominated by cyanobacteria, a group known to have a broad range of effective UV-protective mechanisms (80). (98) also found low UV radiation susceptibility in a picocyanobacteria-dominated phytoplankton assemblage from a clear lake in southern Canada. On the other hand, short-term experiments in several high-arctic lakes showed that the relative contribution of picoplankton ($0.2\text{--}2 \mu\text{m}$) to phytoplankton production generally decreased with increased UV-B radiation exposure while the larger cells ($>20 \mu\text{m}$) were more UV-B radiation tolerant and their contribution to productivity usually increased after UV-B radiation exposure. Arctic lake experiments at Spitsbergen also indicated a greater sensitivity of the picocyanobacteria relative to larger colonial species to UV-B radiation exposure (51). A study by (99) showed that for marine tropical plankton, UV-B-induced DNA damage was not significantly different between two size classes ($0.2\text{--}0.8 \mu\text{m}$ and $0.8\text{--}10 \mu\text{m}$). Given the variability in results between studies, other aspects such as species-specific sensitivity, repair capac-

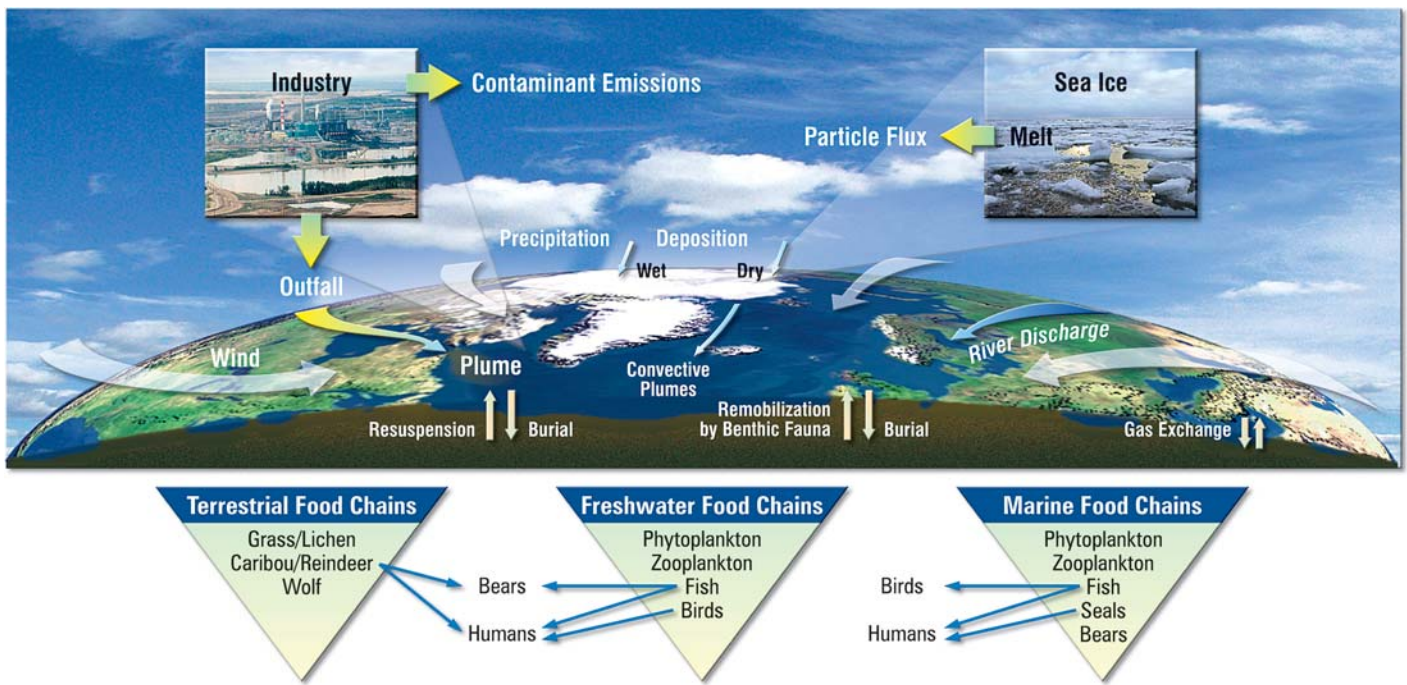


Figure 5. Contaminant pathways, transfers, and exchanges in freshwater systems: land-atmosphere-water interactions (adapted from 3).

ities, or cell morphology might be more important than cell size (99).

The level of photoinhibition by UV radiation in phytoplankton can be modified by many factors that influence the extent of exposure within the water column. For instance, vertical mixing can affect the time and duration of phytoplankton exposure to UV radiation and diminish or aggravate projected inhibition of photosynthesis obtained under simulated conditions (continuous UV radiation exposures). In a well-mixed water column, the planktonic community can seek refuge from UV-B radiation, and photo-repair in the deeper portion of the column. However, the formation of near-surface thermoclines caused by high solar irradiance, calm winds, and solar heating of the surface water can retain the phytoplankton under high irradiances for longer periods of times (100) and result in UV radiation damages that can exceed what can be repaired (101). During the spring and summer in the Arctic, climatic conditions (e.g., clouds, rain, snow, fog, and wind) that change tremendously from day-to-day can affect the amount of UV radiation exposure as well as the ratios of UV radiation to longer wavelengths reaching the surface of the earth. In the coastal areas of northern Norway, variation in cloudiness was demonstrated to influence UV radiation levels. The relative amount of UV-A and UV-B radiation to PAR increased during periods of heavy cloud cover (102) because clouds reflect and return radiation (103).

Ultraviolet radiation may impair the transfer of carbon from the microbial food web to higher trophic levels, including zooplankton and fish. However, increased photochemical activity associated with UV radiation also has the potential to stimulate some heterotrophic species by causing the breakdown of high molecular weight organic compounds into a more available form (104; 105; 106; 107) that can then be taken up for bacterial and protist growth. A study using large enclosures in a high arctic lake with high levels of dissolved organic compounds showed that after long-term exposure to enhanced UV-B radiation, heterotrophic bacterial production and zooplankton density increased, which may have resulted from an increase in nutrient availability caused by photodegradation of organic compounds and the stimulation of heterotrophic pathways (46).

Some wetland biota such as amphibians are known to be highly sensitive to UV radiation, although sensitivity varies greatly among populations, and other factors such as climate effects on habitat extent are likely to have much greater impacts on northern species. A variety of complex responses have been observed to date in wetland plants. For example, UV-B radiation effects on the growth of high-latitude mosses appear to be a function of water supply as well as species. Field irradiation (UV-B radiation levels increased 15%) of *Sphagnum fuscum* caused a 20% reduction in growth, however, growth of the other moss species increased by up to 31% with the enhanced UV-B radiation. This stimulatory effect, however, ceased or was reversed under conditions of reduced water supply (108).

GLOBAL CHANGE AND CONTAMINANTS

During the past 50 years, persistent organic pollutants (POPs), metals, and radionuclides have been widely distributed into northern freshwater ecosystems by long-range atmospheric transport (1, 109). Within some catchments, deposition from the atmosphere may be augmented locally by industry or agriculture (3) or bio-transport (6; 110; 111). Figure 5 illustrates contaminant pathways, transfers, and exchanges in freshwater systems. This section briefly discusses how projected global change might alter these pathways, focusing especially on POPs and Hg because they have the greatest potential for change in risks to freshwater ecosystems as a result of climate change (112, 113).

Contaminant pathways and arctic freshwater ecosystems

There are two components of long-range transport pathways: transport to arctic freshwater catchments, and processes within the catchments (Figure 6). Transports and transfers within each of these components are altered by climate change manifested in such things as windfields, precipitation (amount, timing, form), snow cover, permafrost, extreme events, UV radiation exposure, the hydrological cycle, ice cover, the organic carbon cycle, and food webs, and are very likely to result in enhanced bioaccumulation of contaminants (e.g., 114).

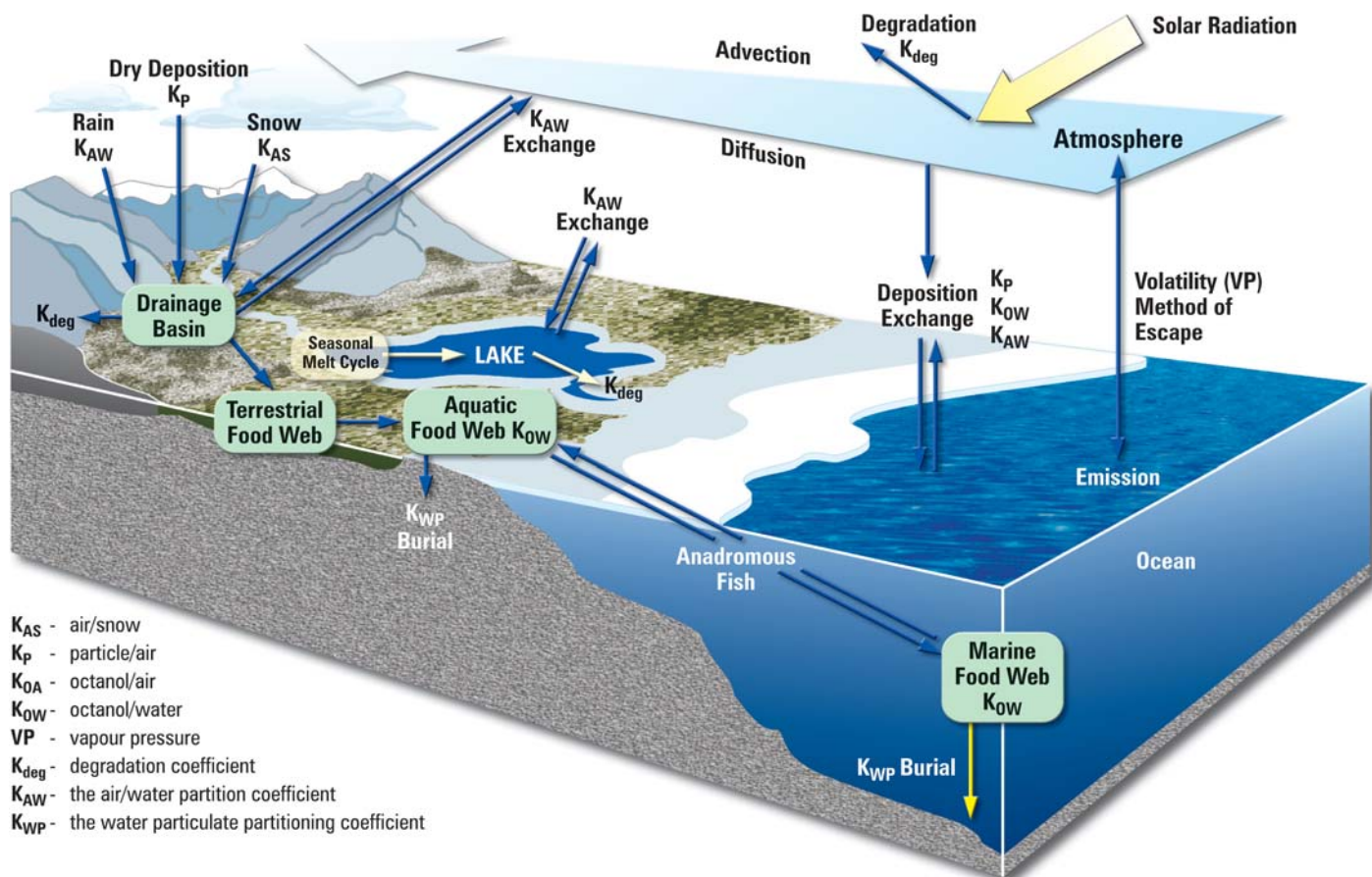


Figure 6. Processes involved in transporting POPs to the Arctic and depositing them into terrestrial ecosystems. Transport, deposition, and exchange can occur anywhere along the transport pathway. Contaminants can also be transported within aquatic and terrestrial food chains. Climate change can alter the physical couplings between the systems (e.g., by changing rain or snowfall patterns), or alter the biological couplings by changing trophic structure or migratory pathways.

Before describing specifically how global change may alter contaminant pathways, it is important to understand how contaminants become concentrated in the environment. (115) suggest that there are two distinct concentrating processes,

which they term solvent switching and solvent depletion. Solvent switching can, for example, lead spontaneously to concentration amplification of hexachlorocyclohexanes (HCHs) in water because HCH partitions strongly out of air (116), or

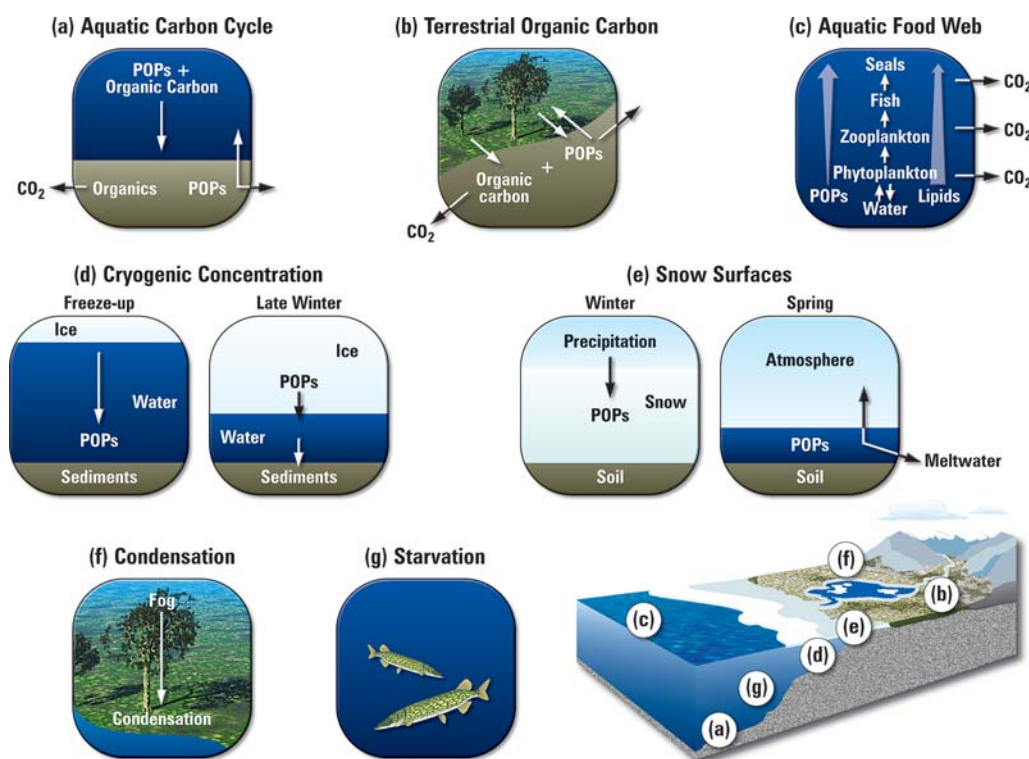


Figure 7. In the illustrated solvent-depleting processes, POPs are concentrated beyond thermodynamic equilibrium through the removal of solvent by organic carbon metabolism in (a) aquatic and (b) terrestrial carbon cycles, by (c) inefficient lipid transfer in aquatic food webs, by (d) exclusion into a dwindling layer of water during the growth of ice, by (e) the loss of snow surfaces during aging or melting, by (f) the loss of surface area through condensation of fog into water droplets or onto surfaces, and (g) through loss of lipid pools during periods of starvation.

high concentrations of PCBs in phytoplankton due to strong partitioning out of water and into lipids (117). Solvent depletion involves a reduction in the mass of solvent in which the contaminant is held, a process that can lead to fugacity amplification (i.e., POP concentrations exceeding thermodynamic equilibrium with the surrounding media). Examples include inefficient fat transfers in aquatic food webs (i.e., biomagnification, 118), the loss of organic carbon in settling particles or during sediment diagenesis (119; 120), the decrease of snow surface area as crystals become more compact during aging or the entire loss of snow surface during melting (115; 121), or cryogenic concentration during the formation of ice (113; Figure 7) Many of the consequences of climate change for the solvent-switching processes are relatively easy to project and model because the effect of temperature on partition coefficients is known. For example, for contaminants that presently are saturated in arctic surface waters, increased temperatures will generally lead to net gas evasion (122) and the ocean is very likely to become a net source of those contaminants to the atmosphere. (123) concluded that, with temperature increases, the risk hexachlorobenzene (HCB) presents to aquatic biota is likely to decrease slightly because HCB will partition less into water (123). The solvent-depleting processes; however, provide a much greater challenge to projection and have not yet been incorporated realistically into models.

Persistent organic pollutants in arctic catchments

The freeze, melt, and hydrological cycles and the organic carbon cycles of arctic lakes are likely to provide sensitive sentinels of change. As discussed in more detail in the previous sections and (124) probable changes as a result of projected temperature increases include reduced thermal contrast between winter and spring; reduced duration of snowmelt (later freeze-up, earlier melting); reduced ice formation; increased annual precipitation; thawing of permafrost producing a deeper active layer, enhanced soil erosion, mobilization of organic carbon, and reduced pond areas owing to drainage; more frequent extreme weather events; changes in catchment vegetation (i.e., more leaf-bearing plants); changes in nutrient availability; warming of lakes; and an increase in the frequency of wildfires (e.g., 25, 125, 126, 127). For some lakes, permafrost degradation together with reduced ice cover is very likely to result in enhanced nutrient and organic carbon loadings and higher productivity. Conversely, if dry summer conditions produce extensive fires, affected lakes are very likely to receive reduced spring melt, fewer nutrients from the catchment, reductions in productivity (25), and higher burdens of combustion PAHs.

Most arctic lakes receive their contaminant burdens from the atmosphere, with the catchment acting as a receptor through snow, rain, and dry deposition especially during winter, and a conveyor through snow and ice melt and runoff in spring (e.g., see 1, 120). This section describes the stages from contaminant release to its final emergence in top freshwater predators (Figure 6), noting in particular those components of the pathway likely to be altered as a result of climate change.

Upon release, contaminants are transported through the atmosphere either as gases or adsorbed onto particles. During atmospheric transport, washout and air-surface exchange remove some of the contaminant to the surface where it may become permanently sequestered or re-volatilized as a result of, for example, seasonal heating cycles, eventually arriving in the Arctic via a number of "hops" (109). Accordingly, POPs undergo hemispheric-scale chromatography, with surfaces (soil, water, vegetation) providing the stationary phase and the atmosphere providing the moving phase. Global temperature increases will generally accelerate this cycling. Processes that are

effective at capturing contaminants in arctic drainage basins (e.g., strong partitioning onto particles, into precipitation, into vegetation) are also effective at removing contaminants during transport. For example, (116) suggested that air-water partitioning alone restricts the entry of beta-hexachlorocyclohexane (b-HCH) into the Arctic by removing it to surfaces by rain and air-sea exchange. In the case of b-HCH, and in the case of contaminants that partition strongly onto particles (e.g., many PAHs, dichloro-diphenyltrichloroethane (DDT), and highly chlorinated PCBs), changes in rainfall patterns (amount and location) are very likely to alter the efficiency of transport to arctic locations and capture within the Arctic. Heavy metals provide an instructive example that will likewise apply to many of the POPs. Presently, less than 20% of particulate metal entering the Arctic is captured there (128). Since the five ACIA-designated models project that the Arctic will become a "wetter" place, 129, see also 130; 131), the capture of particulates and contaminants that partition strongly into water is likely to significantly increase by a factor that could more than offset efforts to reduce global emissions.

Because much of the contaminant delivery to the Arctic occurs during late winter as "arctic haze" or as "brown snow" events (132; 133), it is clear that sequestering by snow is an important process. Hence, careful consideration must be given to any changes in arctic snow conditions. Newly formed snow has a large surface area (as much as 0.4 m²/g, 134) that scavenges both particulate and gaseous POPs, eventually sequestering them into the snowpack (135). Precipitation form (snow, rain, fog) is therefore important and, considering the seasonal modulation in atmospheric concentrations of contaminants (1, 136; 137), so is timing. For example, snowfall during a period of arctic haze would be much more important for transferring contaminants to the ground than at other times of the year.

As snow ages or melts, its surface area to volume ratio decreases, resulting in the removal of the solvent that captured the POPs (e.g., 138). (115) estimated that this process could lead to fugacity amplification of ~2000 times that of the air – clearly an enormous thermodynamic forcing. Depending on the exact circumstances under which snow loses its surface area, the POPs will be vaporized back to the air or partitioned into particles, soil, vegetation, or meltwater. Changes in the frequency and timing of snowfall, unusual events like freezing rain, or the rate and timing of snowmelt are likely to effect large changes in the proportion of POPs that enter the arctic hydrological cycle.

Terrestrial organic carbon in soils and vegetation has a large capacity to store many POPs (139), with PCBs, DDT, HCH, and chlorobenzenes figuring prominently (6). (140) have shown that forests "pump" organochlorines from the atmosphere into foliage and subsequently to long-term soil reservoirs. Accordingly, increased proportions of leaf-bearing plants in arctic catchments will enhance this "pump". Increased metabolism of soil organic carbon owing to temperature increases, changes in soil moisture, or reduced snow cover will force POPs associated with soil organic carbon to redistribute, probably into groundwater or meltwater.

Climate variation results in the storage of contaminants in perennial snow and ice or in soils, vegetation, and delta/estuarine sediments during periods of cold climate (years to decades). These stored contaminants may subsequently be released during a period of warming and, although this process may not be sustainable, it is likely to produce episodes of high contaminant loadings into water (141, 142). During permafrost degradation, a shift toward dendritic drainage patterns (e.g., 143) allows a more complete transport of contaminants into ponds and lakes and possibly re-mobilizes contaminants stored in soils. Simultaneously, the reduction of pond areas owing to

drainage channels in permafrost (125) is likely to enhance contaminant transport into the remaining surface water. After POPs enter the hydrological cycle through the mechanisms discussed, a proportion of them will be stored in lakes and lake sediments. Evidence from a limited number of studies (1, 144; 145) suggests that meltwater currently enters arctic lakes when they are thermally stratified beneath an ice cover. Therefore, much of the annual snowmelt traverses under the ice to exit at the outflow carrying its contaminant burden; that is, arctic lakes are not efficient at capturing POPs entering via streamflow. It is probable that many of the previously described alterations in freshwater systems induced by climate change (such as reduced ice cover, increased mixing and primary production, and increased loading of organic carbon and sediment from the contributing catchments) will also enhance contaminant capture in lakes (e.g., see 125).

Because most arctic lakes tend to be oligotrophic, only a small proportion of POPs is transported by vertical flux of organic particles and buried in sediments (e.g., 144; 146; 147). A second solvent-depletion process occurs due to organic carbon metabolism during particle settling and within bottom sediments (Figure 7a). The loss of organic carbon can provide exceptionally strong thermodynamic forcing to drive the POPs off solid phases and into sediment pore water, where they may diffuse into bottom waters or partition into benthos (115, 148; 149). An increase in the vigor of the organic carbon cycle (e.g., increased primary production, organic carbon loadings, and microbial activity) will enhance this thermodynamic pump. Cryogenic concentration (Figure 7d) is likely to work together with organic carbon metabolism in sediments during winter to produce exceptionally high concentrations of POPs in bottom water. Although relatively poorly studied, contaminants are believed to be excluded from ice as it forms. For shallow water that freezes nearly to the bottom, dissolved contaminants are likely to be forced into a very small volume of remaining water and the resultant high contaminant concentrations will promote partitioning into remaining organic material including sediment surfaces, benthos, plankton, and larger animals. As noted previously, such under-ice zones are often a critical winter refuge for biota (e.g., 150). It is likely that a general reduction in the depth of ice formed during warmer winters will reduce cryogenic concentration. However, cryogenic concentration interacts with water levels, which are likely to decrease during permafrost degradation (125).

The transfer of lipid-soluble POPs upward in aquatic food webs is one of the most important routes of exposure to apex feeders, including humans. In this solvent-depleting process, much of the lipid is metabolized at each trophic level while the organochlorines are retained. This results in higher trophic levels exhibiting organic carbon biomagnification factors of 3 to 100 in their lipids, and a net bioaccumulation of 107 to 109 times higher than in the water (112, 118, 151; 152). Changes in aquatic trophic structure either through alteration of the number of food web steps or the size distribution of predatory fish will likewise change contaminant burdens. With climate change, wide-ranging shifts in zoogeographic distributions have the potential to affect every step in freshwater food chains (125, 126).

There are several other ways that global change can alter contaminant pathways in arctic aquatic ecosystems. As noted in 114, recent evidence suggests that salmon migrations undergo large, climate-related variation (153, 154) and that Pacific salmon may respond to change by invading arctic rivers (155). Given that these salmon biomagnify and bioaccumulate contaminants in the Pacific Ocean, they are an important means of contaminant transport into particular arctic catchments. In specific lakes, fish may supply more POPs than atmospheric deposition (110). Similarly, bird migrations that

change in location and intensity have the potential to concentrate, transport, and deposit contaminants in particular catchments (6, 125, 151). For example, detailed studies of Lake Ellasjoen, Norway, found that seabirds can serve as important biological pathways carrying contaminants (in this case POPs) from marine to freshwater environments (4). Climate change or human intervention is also very likely to lead to the introduction of exotic species to the Arctic. Although probably not a risk to arctic lakes, the invasion of the Great Lakes by the zebra mussel (*Dreissena polymorpha*) provides an instructive example of just how disruptive an exotic species can be to organic carbon and POP cycles (156, 157; 158).

Changes within arctic catchments that cause apex feeders (e.g., humans, bears, mink, birds) to switch their diet from aquatic to terrestrial food sources or vice versa have a large potential to alter contaminant exposure. Whereas arctic aquatic food webs exhibit endemic contamination from biomagnifying chemicals, arctic land-based food webs are among the cleanest in the world (6; 159). Dietary changes are forced by fluctuations in the populations of prey species or by changes in access to the species due to early ice melt or permafrost degradation (e.g., 160; 161).

As conditions more suitable for domestic crops develop, agriculture or silviculture within arctic drainage basins and associated chemical use is likely to expand. Demographic shifts and population increases in northern regions could possibly lead to increased local release of contaminants. South of the Arctic, global temperature increases and alteration of hydrological cycles will probably result in insect and other pest outbreaks (e.g., West Nile virus or malaria), provoking the re-introduction of banned pesticides (122). Finally, contaminants in dumps or sumps presently contained by permafrost are very likely to be released by permafrost degradation (162).

Increased fluxes of PAHs are likely to result from the erosion of peat-rich soils (163) or drying trends leading to an increase in wildfires, and are likely to have a greater impact on small rivers that presently receive most of their PAHs from combustion sources (164).

Mercury in arctic catchments

Mercury exhibits a natural global cycle that has been enhanced by human activities such as coal burning, soft and ferrous metal smelting, cement production, and municipal waste with the consequence that two to three times as much Hg is now cycling through the atmosphere and surface waters than was before the rise of industry (165). (166) estimated that worldwide anthropogenic Hg emissions totaled 2235×10^3 kg in 1995, with fossil fuel consumption contributing over half of that. This value may be compared to the 2500×10^3 kg/yr estimate of natural emissions (167). The largest emitter of Hg from fossil fuel consumption is China (495×10^3 kg in 1995), which is directly upwind from the Bering Sea, Alaska, and the western Arctic. In comparison, Russia released about 54×10^3 kg in 1995.

Many of the concentrating processes discussed for POPs (Figure 7) apply equally to Hg. However, atmospheric Hg depletion events (MDEs) after polar sunrise provide a unique, climate-sensitive pathway to Hg deposition in arctic catchments (Figure 8, 1; 79; 168; 169; 170). The process requires snow surfaces, solar radiation, and the presence of sea salts (bromides and chlorides). Although MDEs are initiated over the ocean, and especially over the marginal seas where halides are more available from frost flowers (crystals of ice that form directly from the vapor phase, often associated with new sea ice, which can become very salty by channeling brine upward from the ice) or first-year ice (171), atmospheric advection can subsequently deposit reactive Hg in arctic catchments (170). As with POPs, Hg can

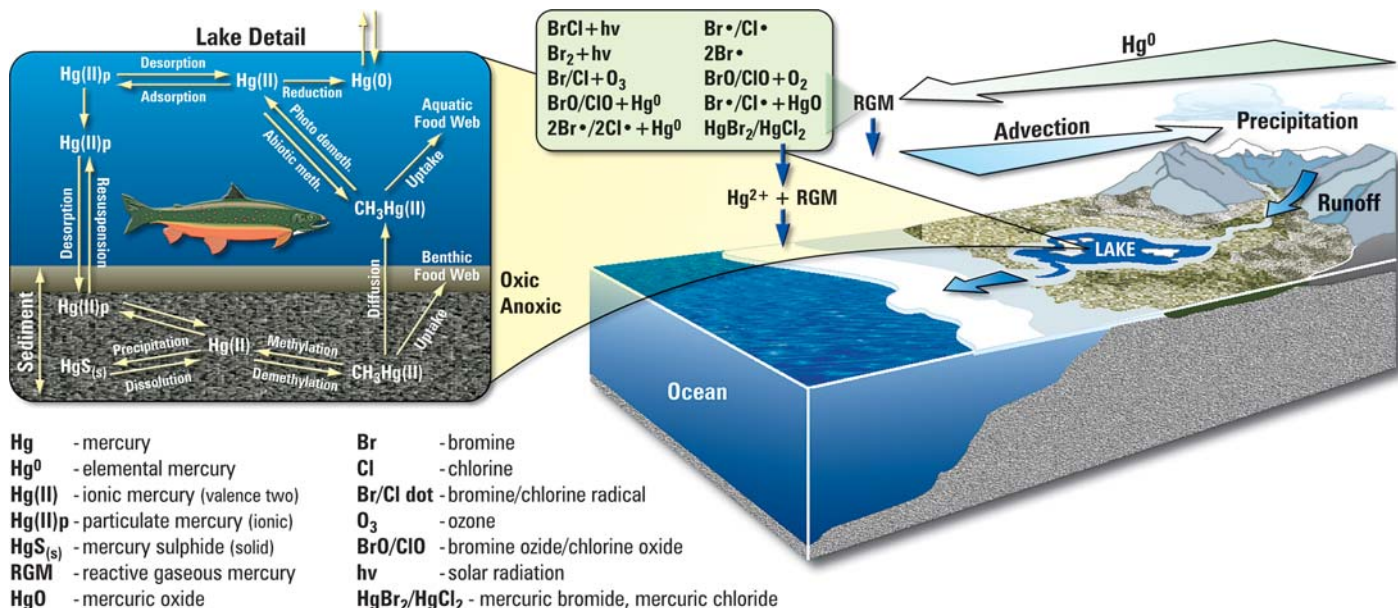


Figure 8. Production of particulate and reactive gaseous mercury over the ocean after polar sunrise (right side) and the advection of reactive and bioavailable forms of mercury into catchments where it is deposited. After deposition, the mercury enters lakes through meltwater and is then subject to reduction and methylation (meth.) processes. Methyl mercury ($CH_3Hg(II)$) is the most toxic form.

Table 1 – Environmental factors affecting mercury concentration in aquatic top predators.

Projected Change	Effect on mercury concentration in predatory fish	Reference
Flooding of soil	Increase	Bodaly and Johnston
Increased primary production	Reduce	Pickhardt et al.
Increased number of trophic levels	Increase	Kidd et al.
Shift toward larger fish	Increase	Sherwood et al.
Reduced lake size	Increase	Bodaly et al.
Increased anadromous fish migration	Increase	Zhang et al.

be transferred and concentrated during snow aging and melting, such that a large pulse of Hg is released to terrestrial and freshwater environments during spring melt (e.g., see 5; 144; 172). (173) showed that Hg deposited through the MDE mechanism is in a form that can readily be taken up by biota. Once Hg enters the hydrological cycle, it can be concentrated and transferred through the carbon cycle and food webs, both of which are vulnerable to change. In addition, the efficiency of arctic lakes in capturing Hg is very likely to be altered by changes in the timing of freshet, ice melt, and productivity (1, 144; 151; 174).

Once Hg has been deposited into arctic catchments, a number of processes may lead to elevated concentrations in old, predatory fish (Table 1). The coupling between Hg deposition on surfaces and its entry into lakes is likely to be enhanced by projected changes in the hydrological and organic carbon cycles (144; 172). Apex feeders are most vulnerable to any change in the Hg cycle considering that biomagnification factors are 250 to 3000 (174; 175; 176; 177). Because MeHg presents a far greater health hazard than inorganic or elemental Hg, methylation is a crucial process upon which climate change operates. Wetlands and wetland sediments are net producers of MeHg (178; 179), and Hg observed in fish from small lakes appears to correlate with the amount of watershed occupied by wetlands (180). Flooding of terrestrial landscapes has the well-known consequence of releasing Hg from submerged soils (181). Therefore, alteration of wetland distribution or area in the

Arctic resulting from thawing permafrost (see the discussion on carbon dynamics in 182) is very likely to release Hg, which will be more serious if arctic soils contain an inventory of contaminant Hg accumulated during the past century or two.

CONCLUSIONS

Climate change impacts related to alterations in UV radiation regimes and climate-contaminant interactions resulting from global change are projected to have significant synergistic and/or cumulative effects on the structure and function of Arctic freshwater ecosystems. Reduced ice cover is likely to have a much greater effect on underwater UV radiation exposure than the projected levels of stratospheric ozone depletion. A major increase in UV radiation levels will cause enhanced damage to organisms (biomolecular, cellular, and physiological damage, and alterations in species composition). Allocations of energy and resources by aquatic biota to UV radiation protection will increase, probably decreasing related trophic-level productivity. Elemental fluxes are also expected to increase via photochemical pathways.

Increases in temperature and precipitation are also very likely to increase contaminant capture in the Arctic. Projected increases in temperature and changes in the timing and magnitude of precipitation will affect the deposition of contaminants at high latitudes and will accelerate rates of contaminant transfer. Climate scenarios currently project a “wetter” Arctic, increasing the probability of wet deposition of contaminants such as heavy metals and persistent organic pollutants. Episodic releases of high contaminant loadings from perennial snow and ice are very likely to increase. Enhanced permafrost degradation is also expected to enhance the mobilization of contaminants and lower water levels will amplify the impacts of contamination on high-latitude freshwater bodies. Arctic lakes are therefore very likely to become more prominent contaminant sinks. Contaminant capture in lakes will increase with reduced lake-ice cover (decreased stratification), increased mixing and primary production, and greater organic carbon and sediment loading.

The nature and magnitude of contaminant transfer in aquatic food webs are also likely to change. Changes in aquatic

trophic structure and zoogeographic distributions will alter biomagnification of contaminants, including persistent organic pollutants and mercury, and potentially affect freshwater food webs, especially top-level predatory fish (e.g., lake trout) that are sought by all types of fisheries.

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