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Review

Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data

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Abstract

The Arctic has undergone dramatic change during the past decade. The observed changes include atmospheric sea-level pressure, wind fields, sea-ice drift, ice cover, length of melt season, change in precipitation patterns, change in hydrology and change in ocean currents and watermass distribution. It is likely that these primary changes have altered the carbon cycle and biological systems, but the difficulty of observing these together with sporadic, incomplete time series makes it difficult to evaluate what the changes have been. Because contaminants enter global systems and transport through air and water, the changes listed above will clearly alter contaminant pathways. Here, we review what is known about recent changes using the Arctic Oscillation as a proxy to help us understand the forms under which global change will be manifest in the Arctic. For Pb, Cd and Zn, the Arctic is likely to become a more effective trap because precipitation is likely to increase. In the case of Cd, the natural cycle in the ocean appears to have a much greater potential to alter exposure than do human releases of this metal. Mercury has an especially complex cycle in the Arctic including a unique scavenging process (mercury depletion events), biomagnifying foodwebs, and chemical transformations such as methylation. The observation that mercury seems to be increasing in a number of aquatic species whereas atmospheric gaseous mercury shows little sign of change suggests that factors related to change in the physical system (ice cover, permafrost degradation, organic carbon cycling) may be more important than human activities.

Organochlorine contaminants offer a surprising array of possibilities for changed pathways. To change in precipitation patterns can be added change in ice cover (air–water exchange), change in food webs either from the top down or from the bottom up (biomagnification), change in the organic carbon cycle and change in diets. Perhaps the most interesting possibility, presently difficult to predict, is combination of immune suppression together with expanding ranges of disease vectors. Finally,

Abbreviations: AO, Arctic Oscillation; DDT, dichlorodiphenyltrichloroethane; ENSO, El Niño–Southern Oscillation; GHG, greenhouse gases; HCB, hexachlorobenzene; HCH, hexachlorocyclohexane; HLC, Henry's Law Constant; IABP, International Arctic Buoy Programme; IPCC, Intergovernmental Panel on Climate Change; MDEs, mercury depletion events; NAM, Northern Hemisphere annular mode; NAO, North Atlantic Oscillation; OCPs, organochlorine pesticides; OCs, organochlorines; PAHs, polycyclic aromatic hydrocarbons; PBDE, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; PCDD/F, polychlorinated dibenzo-*p*-dioxin/furan; PCNs, polychlorinated naphthalenes; PDO, Pacific Decadal Oscillation; P−E, precipitation minus evaporation; PNA, Pacific North American atmospheric pattern; POPs, persistent organic pollutants; SAT, surface air temperature; SLP, sea-level pressure (atmospheric); TPD, transpolar drift.

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biotransport through migratory species is exceptionally vulnerable to changes in migration strength or in migration pathway—in the Arctic, change in the distribution of ice and temperature may already have caused such changes.

Hydrocarbons, which tend to impact surfaces, will be mostly affected by change in the ice climate (distribution and drift tracks). Perhaps the most dramatic changes will occur because our view of the Arctic Ocean will change as it becomes more amenable to transport, tourism and mineral exploration on the shelves. Radionuclides have tended not to produce a radiological problem in the Arctic; nevertheless one pathway, the ice, remains a risk because it can accrue, concentrate and transport radio-contaminated sediments. This pathway is sensitive to where ice is produced, what the transport pathways of ice are, and where ice is finally melted—all strong candidates for change during the coming century.

The changes that have already occurred in the Arctic and those that are projected to occur have an effect on contaminant time series including direct measurements (air, water, biota) or proxies (sediment cores, ice cores, archive material). Although these 'system' changes can alter the flux and concentrations at given sites in a number of obvious ways, they have been all but ignored in the interpretation of such time series. To understand properly what trends mean, especially in complex 'recorders' such as seals, walrus and polar bears, demands a more thorough approach to time series by collecting data in a number of media coherently. Presently, a major reservoir for contaminants and the one most directly connected to biological uptake in species at greatest risk–the ocean–practically lacks such time series.

Keywords: Arctic; Climate change; Contaminants; Mercury; Metals; Organochlorines; PAH; radionuclides

Contents

1. Introduction

In previous assessments of the Arctic, physical pathways, terrestrial and marine ecosystem uptake and human health have been discussed as separate and more or less independent components of the problem of contamination (AMAP, 1998; Braune et al., 1999; Macdonald et al., 2000a; Muir et al., 1999; Van Oostdam et al., 1999). Arctic systems are strongly interrelated making this arbitrary compartmentalization a difficult base upon which to produce a holistic discussion of why contaminants end up where they do, and how the environment conspires to put certain ecosystem components at risk far from any contaminating sources. Physical pathways, in particular, have been treated as a set of fixed compartments connected by exchanges between compartments (see, for example Fig. 2 in Macdonald et al., 2000a). This structure allows a coherent approach to discussing contaminant transfers from one compartment to another (e.g., atmosphere to ocean), an identification of where our most serious knowledge gaps lie, and a semi-quantitative assessment of progress made in understanding system components (e.g., rates for the various arrows and inventories for the various boxes). Accumulating evidence shows that polar environments are exceptionally sensitive to change and that some of these changes would have direct effects on processes (e.g., the role of snow in contaminant scavenging, the sensitivity of gas–particle partitioning to temperature, the importance of ice cover in air–sea exchange (see Shepson et al., 2003)). In contrast, the 'box and arrows' approach treats the Arctic as a rather rigid set of processes into which contaminants have entered, and implies that physical and chemical properties of the contaminants set the rules for where contaminants finally end up. How the Arctic's processes (e.g., hydrology, winds, currents, ice cover, organic carbon cycle, precipitation) might themselves be subject to change has been much less considered. And yet, we have learned not only that the physical structure of the system can change (e.g., ice cover can disappear), but that pathways can also change (e.g., diversion of Russian rivers into the Canada Basin, change in the carbon cycle) with the consequence that conclusions of earlier assessments may be inaccurate and, in some cases, wrong.

Here we depart from previous assessments by discussing contaminant pathways in the context of change in the Arctic and by seeking interactions that cross between physical and biological components of arctic systems. Two science communities have been vigorously conducting research in Arctic, one work-

ing on contaminants and the other on climate change. Communication between these two communities has been weak to the detriment of both.

Predicting how climate change will alter contaminant transport in the global environment poses an exceptional challenge. It requires a detailed knowledge of the physical and chemical properties of contaminants, for which much progress has been made during the past decade. It also requires a profound understanding of environmental pathways and how they might respond to change forced by, for example, alteration of the atmosphere's greenhouse gas (GHG) composition. We presently lack this depth of understanding. It is clear that greenhouse gases such as CO_2, CH_4, N_2O and chlorofluorocarbons (CFCs) and aerosols are being released to the Earth's atmosphere by human activities (IPCC, 1990, 1995, 2002), that the world's hydrological cycle is being massively altered by damming for power and irrigation (Dynesius and Nilsson, 1994), and that humans have altered the structure of the global marine food web (Myers and Worm, 2003; Pauly et al., 1998). Sufficient evidence has been assembled from paleorecords and much shorter instrumental observations to convince most–but not all–climate scientists that these kinds of disturbances will contribute to a variety of changes at the global scale if they have not already done so (e.g., see Gillett et al., 2003; IPCC, 1990, 2002; Parsons, 1996; Showstack, 2001).

As a component of the cryosphere with a large seasonal amplitude in ice and snow cover, the Arctic will be pivotal as a region sensitive to change (sentinel) and as an exporter of change to other parts of the world (amplifier) (Aagaard and Carmack, 1989; Vörösmarty et al., 2001; Walsh, 1995; Walsh and Crane, 1992). The 0 °C isotherm is an especially important threshold of change because shifts between liquid and solid water have immense consequences for physical and biological systems and for humans.

Two major difficulties face us in understanding change in the Arctic and projecting the future. Firstly, recent trends are difficult to detect and comprehend due to short, sparse instrumental records. This is especially true of the Arctic where any climate trend must be discriminated from an enormous seasonal amplitude in weather, and natural variation at time scales from annual to 5 years to centuries and longer (Fischer et al., 1998; McGhee, 1996; Polyakov and Johnson, 2000; Proshutinsky and Johnson, 1997; Stirling et al., 1999; Tremblay et al., 1997; Vanegas and Mysak, 2000; Wang and Ikeda, 2001). Secondly, our understanding of environmental processes in the Arctic is not sufficiently complete to allow a confident linking of primary changes (sea-level air pressure, air temperature, ice cover) with those of higher complexity but of much greater significance (e.g., thermohaline circulation, ecological structure and function, the hydrological cycle). Presently, these difficulties form an insurmountable hurdle to making confident projections of how, exactly, exposure of arctic biota to contaminants will be affected by global change. Nevertheless, sufficient evidence has accumulated during the past decade to assure us that contaminant pathways can change and will continue to change. Experience strongly warns us to expect the environment to deliver surprises (Macdonald et al., 2000a). One of those surprises is likely to be the abruptness of change—something for which the Arctic has recently shown unexpected predisposition (Alley et al., 2002; Dickson, 1999; Johnson and Marshall, 2002; Macdonald, 1996; Mysak, 2001; Rothrock et al., 1999). Other surprises lurk in the sometimes subtle and non-intuitive connections between global and regional pathways that put the Arctic at risk from contaminants in the first place (Fig. 1, AMAP, 1998; Macdonald et al., 2000a).

To project how global change may alter risks from contaminants in the Arctic, we build on the foundation developed in previous assessments (AMAP, 1998; Braune et al., 1999; Macdonald et al., 2000a; Muir et al., 1999; Van Oostdam et al., 1999), particularly those components addressing pathways. We might then proceed by considering model predictions that the globe will experience a mean global air temperature rise of 3 to 5 °C during the coming century (IPCC, 2002). Increased temperature will have direct effects on contaminants (enhanced volatility, more rapid degradation, altered partitioning between phases) and on the environment (loss of permafrost, change in the seasonal cycle of snow or ice). However, a general air-temperature projection like this prepares us very poorly for addressing the more important changes that will inevitably occur in the connections between the Arctic and regions to the south and in the detail of pathways within the Arctic. In the context of arctic ecosystems and humans who depend on them, it will not be temperature rise that

Fig. 1. The major physical pathways (wind, rivers and ocean currents) that transport contaminants to the Arctic.

counts but, rather, the consequent change in the timing of seasons and whether or not the temperature crosses the 0 °C isotherm such that ice, snow or permafrost disappear. During a contaminant's voyage to the Arctic, it may spend varying proportions of time in air, soil, water, ice, and in food webs or it may become degraded (Fig. 2). Each step along the path and every point of transfer can be altered by global change, which for a contaminant may mean dilution, concentration, bifurcation, shortcut or delay.

This synthesis is neither a review of change nor is it a review of contaminant pathways or of newly emer-

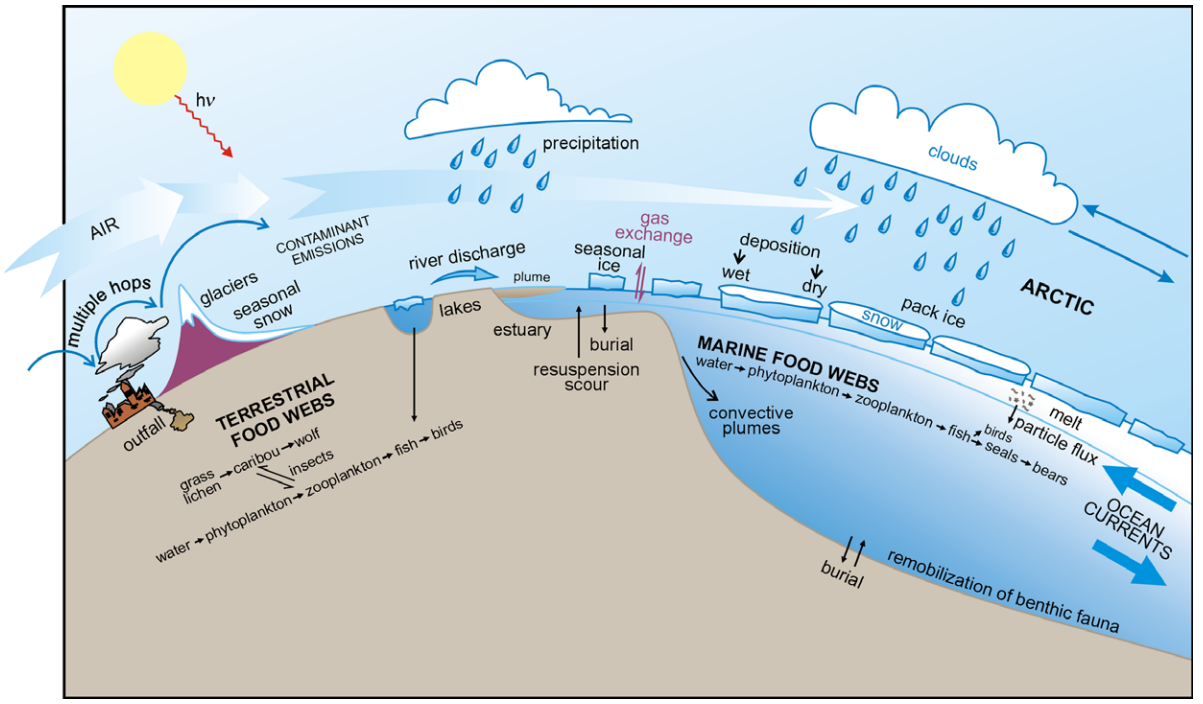

Fig. 2. A simplified schematic diagram showing how physical pathways deliver contaminants emitted from northern industrial regions to the Arctic where they may be concentrated in biota or removed through degradation and burial.

ging contaminants. These topics have been, or are being, thoroughly reviewed elsewhere (AMAP, 1998; IPCC, 1995, 2002; Macdonald et al., 2000a; Ruddiman, 2000). Rather, we seek here observations and projections of global change that appear most likely to play a significant role in the life history of contaminants headed for a destination in Canada's Arctic.

We first discuss change in *physical pathways*, emphasizing especially observations during the past decade. Recent dramatic changes, many of which can be related directly to variation in atmospheric pressure fields (popularly referred to as the Arctic Oscillation (AO); Wallace and Thompson, 2002), include winds, sea-ice drift and cover, ocean currents, precipitation and other environmental pathway components (Morison et al., 2000; Serreze et al., 2000). The contrast between high and low AO indices allows us to discuss with authority some of the ways in which the Arctic actually has changed. We discuss the likely consequences that these physical changes will have for *biological pathways*, noting that the organic carbon cycle and the food web are crucial pathway components for many contaminants (e.g., mercury, cadmium and most organochlorine compounds). We review briefly how *human responses* to global change are likely to alter contaminant pathways in the Arctic. A detailed review of the physical and biological pathway changes that have occurred or are likely to occur then puts us in a position to discuss consequences for each category of contaminant of concern in the Arctic (metals, organochlorines, radionuclides, hydrocarbons). Finally, we discuss the difficulty of interpreting time-series data given that contaminant trends observed in sediment, water, air, and biota collected from the Arctic may include variation related to contaminant emissions as well as variation related to change in pathways.

2. Long-term change in the Arctic

2.1. The distant past

The Arctic is not static. During the last 400,000 years, the earth has experienced four ice ages which have written records or partial records in glacial ice

accumulating in Antarctica (Petit et al., 1999) and in Greenland (Dansgaard et al., 1993; Sowers and Bender, 1995). The overall surface air-temperature change between glacial and interglacial periods is thought to have been about 12 °C, but perhaps more significant than temperature were the accompanying changes in continental ice masses, sea-ice climate and global ecosystems. In particular, sea-ice cover has proven to be a master variable in the equation of change. During the last glacial maximum, sea ice was locked within the Arctic and seasonal or perennial sea ice extended well south into the North Atlantic Ocean (Darby et al., 1997; de Vernal et al., 1993). The change from glacial to de-glacial to interglacial can be seen widely in arctic sediments both in sedimentation rate and in the amounts and sources of organic material deriving from primary production or land (see for example, Darby et al., 2001; Nørgaard-Petersen et al., 1998; Phillips and Grantz, 1997; Stein et al., 1994, 2001).

During the last glacial maximum, sea level dropped by about 120 m (Fairbanks, 1989). This exposed much of the Arctic Ocean's enormous continental shelves, forcing rivers to cut channels across them to enter the interior sea directly (Stein and Macdonald, 2004). Furthermore, the lower sea level cut the connection between the Arctic and Pacific Oceans and reduced or eliminated flow through most of the Canadian Archipelago channels. With sea-level rise, about 15,000 years ago the Bering land bridge was flooded (Hopkins, 1979) and then gradually submerged (Dyke et al., 1996b) allowing the Pacific Ocean access to the Arctic Ocean. This sequence together with inundation of the continental shelves and the reflooding of the Archipelago channels must have had enormous consequences for the oceanography and regional biogeography for the western Arctic and the Archipelago (Dunton, 1992; Dyke et al., 1996a,b; Héquette et al., 1995).

Although climate has been described as 'exceptionally stable' during the past 10,000 years (Dansgaard et al., 1993), it has actually continued to undergo substantial fluctuations. Indeed, it seems that very small shifts in temperature, perhaps only a degree or two, account for the so-called Medieval Warm Period (1100–1400 AD) and Little Ice Age (1450–1850 AD) that followed it (for the relevance of these terms see Bradley and Jones, 1993; Crowley and Lowery, 2000). Both of these minor and sporadic deviations in the temperature record had dramatic consequences for humans—especially those living on the margins of northern oceans (Alley et al., 2002; McGhee, 1996; Ogilvie and Junsson, 2000). During the past two centuries, small changes in ice and watermass distribution have continued to impact humans and ecosystems, sometimes leading to migration or abandonment of locations, but certainly requiring adaptation (McGhee, 1996; Miller et al., 2001; Vibe, 1967).

For most of the past 10,000 years (the Holocene), climate change was not accompanied by the added complexity of anthropogenic pollutants. However, over the past two millennia and especially during the past two centuries, arctic glacial ice has recorded the transient rise of virtually every contaminant emitted by human activities (Boutron et al., 1995, 1998; Gregor et al., 1995; Hong et al., 1994; Masclet and Hoyau, 1994; Rosman et al., 1997). Included in these contaminants are the greenhouse gases (GHGs) that force atmospheric temperature change (Petit et al., 1999) and it is the dramatic rise of these during the past several decades that make future projections based on past climates subject to such uncertainty.

2.2. The present and future

The 20th Century has been the warmest in the Arctic for the past 400 years (Fig. 3, Overpeck et al., 1997). The Intergovernmental Panel on Climate Change (IPCC) suggests that over the past century the global mean surface temperature has increased by about 0.3–0.6 °C, mostly attributable to human activities, and will probably further increase by 1.4 to 5.8 °C between 1990 and 2100 (Houghton et al., 1995; IPCC, 1995, 2002; Showstack, 2001). According to general climate models (Manabe et al., 1992; Mitchell et al., 1995; Zwiers, 2002), warming will be more pronounced in polar regions (Fig. 4), perhaps 5 °C or more near the pole and 2–3 °C around the margins of the Arctic Ocean, with a decreasing temperature contrast between poles and the equator. The observations, however, are not so clear; despite intervals of warming and cooling during the past century there appears little evidence to support enhanced temperature fluctuations in the Arctic (Polyakov et al., 2002b). For gases emitted to the

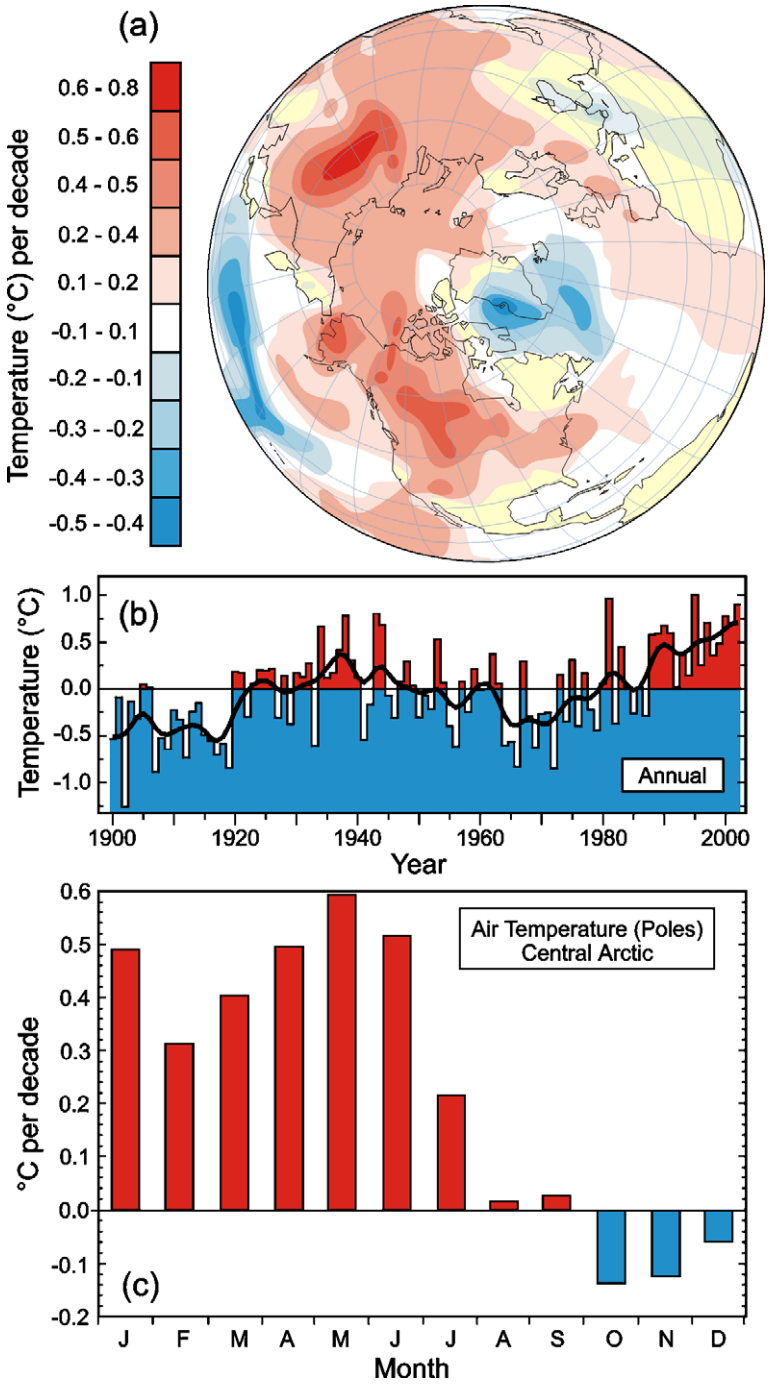

Fig. 3. Temperature trends for the Arctic showing (a) the annual surface temperature trends over the Northern Hemisphere expressed as rates of change for the period 1961–1990 (courtesy of the Climate Monitoring and Data Interpretation Division of the Atmospheric Environment Service, Stewart et al., 1998), (b) temperature anomalies (55–85°N) for 1900–2002 evaluated against the average for 1951–1980 showing the high temperatures of the late 1980s and 1990s are matched by equally high temperatures during the 1930s and 1940s (adapted from Serreze and Barry, in press; Serreze et al., 2000) and (c) the trend by month in surface air temperature of the central Arctic Ocean for the period 1979–1995 illustrating that recent warming is mainly a winter–spring phenomenon (adapted from Serreze et al., 2000).

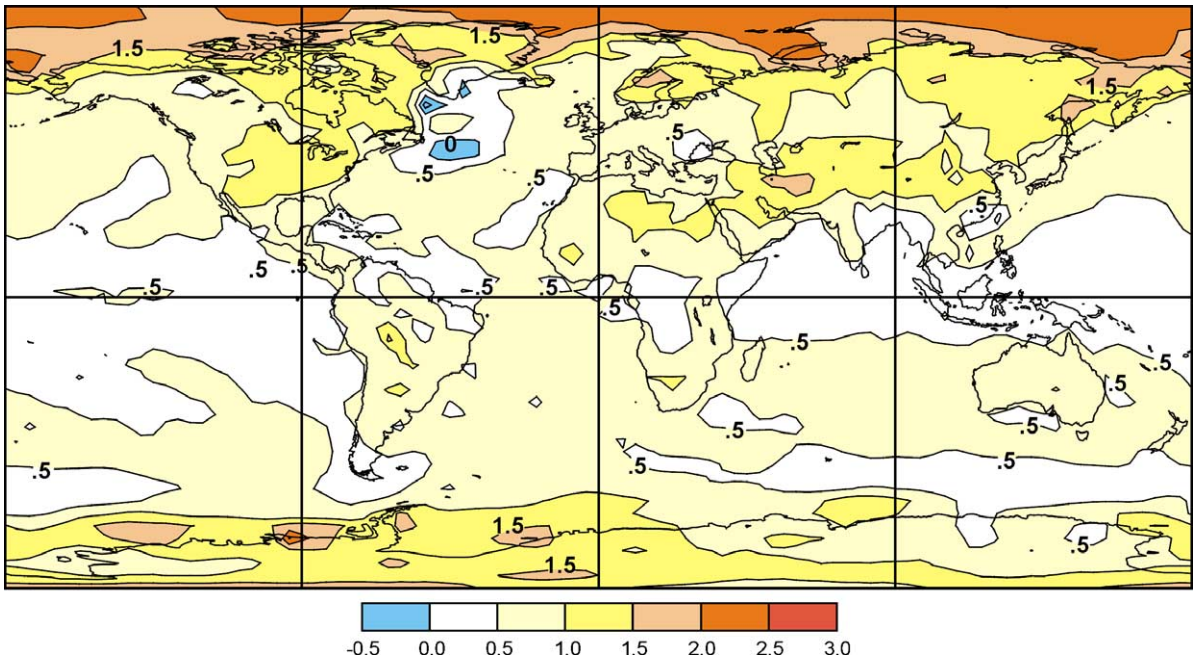

Fig. 4. Change in surface air temperature for 2020–2030 relative to 1990–2000 as projected by the Canadian Centre for Climate Modeling and Analysis CGCM2. Global warming is expected to have an uneven geographic distribution with the Arctic experiencing the highest projected warming (courtesy of CCCma and see Zwiers, 2002).

atmosphere, climate warming will increase inter-hemispheric exchange times, mixing times and mean transit times by perhaps 10% (Holzer and Boer, 2001). Furthermore, the greatest warming is likely to occur in autumn–winter due to delay in the onset of sea-ice cover (Manabe et al., 1992; Serreze et al., 2000). Continental interiors will become dryer and sea level will continue to rise, perhaps by a further 50 cm in addition to the estimated rise of 10–25 cm during the past century (Miller and Douglas, 2004; Proshutinsky et al., 2001; Serreze et al., 2000).

Models predict that after about 80 years of atmospheric CO_2 increase at 1% per year, precipitation will increase within the Arctic and subpolar regions to 0.5–1 m per year (Manabe et al., 1992), making the Arctic a considerably 'wetter' place. Over the past four decades sea-ice extent in the Arctic Ocean has decreased in summer by as much as 25% (Fig. 5a, Vinnikov et al., 1999). By the end of the 21st century, GHG forcing might produce an Arctic Ocean seasonally clear of ice (Fig. 5b, Flato and Boer, 2001).

Simulations based on GHG forcing predict that mean annual river discharge will increase by about 20% for the Yenisei, Lena and Mackenzie Rivers, but decrease by 12% for the Ob (Arora and Boer, 2001; Miller and Russell, 1992). Furthermore, the projection that high-latitude rivers will undergo marked changes in amplitude and seasonality of flow due to decreased snowfall and earlier spring melt (Arora and Boer, 2001) may already have some support in observations (Lammers et al., 2001). It is important to note that the Arctic contains major rivers (e.g., Lena, Yenesei, Ob, and Mackenzie Rivers), which are fed by inflow from basins south of the Arctic circle, and many smaller rivers wholly within the Arctic. Change for these two kinds of rivers is likely to be felt differently with perhaps some of the more dramatic changes happening in the truly arctic drainage basins where ice climate, seasonal snow cover and permafrost changes are all possible.

The coupling of the runoff cycle with northern lake hydrology is probably one of the points most sensitive to climate change (see for example, Vörösmarty et al., 2001) but understanding of the processes involved is not yet sufficient to make confident projections. If arctic lakes become more

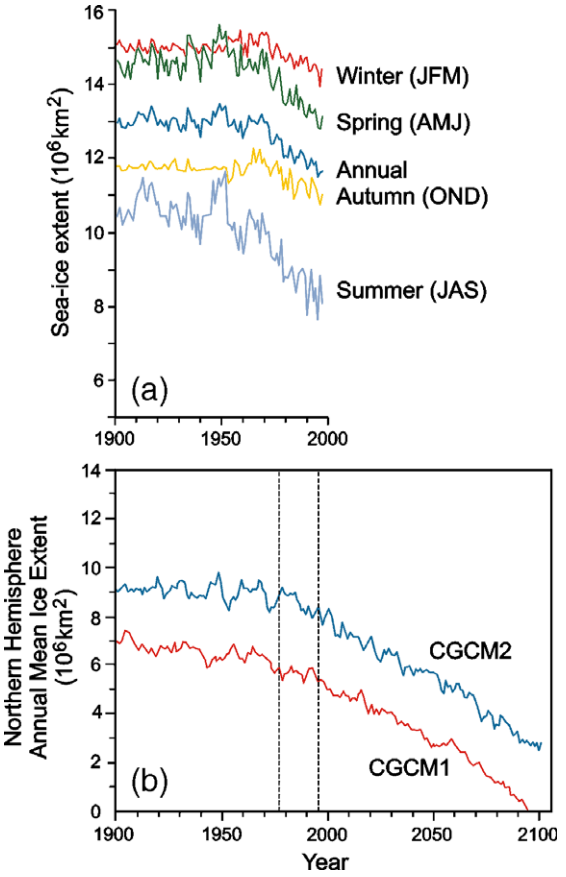

Fig. 5. (a) Time series from 1900 to 2000 of annual and seasonal sea-ice extent in the Northern Hemisphere (adapted from Vinnikov et al., 1999). (b) Model projections of annual mean sea-ice extent for the Northern Hemisphere as simulated by CGCM1 and CGCM2 where the latter model differs from the former in mixing parameterization (figure from Flato and Boer, 2001).

'temperate' in character, productivity will likely be enhanced due to less ice cover and more wind mixing, and there will be greater opportunity for runoff to mix into the lake during freshet further supporting a more vigorous aquatic food web. These changes will alter the efficiency with which lakes capture contaminants especially for chemicals that are presently only weakly captured (Diamond et al., 2003; Helm et al., 2002).

With primary changes like those listed above, we can expect an acceleration of permafrost melting which disrupts vegetation and enhances nutrient, organic carbon and sediment loading of rivers and lakes (Vörösmarty et al., 2001). The loss of sea ice in the marginal seas of the Arctic, particularly during the period of autumn storms, together with sea-level rise will promote further erosion of poorly bonded, low-gradient coasts.

3. Recent change in the Arctic and the Arctic Oscillation

3.1. The Arctic Oscillation

During the 1990s, a quiet revolution took place in our perception of the Arctic. Despite long-term evidence of cyclical change in northern biological populations and ice conditions (see for example, Bockstoce, 1986; Gudkovich, 1961; Vibe, 1967), the general view among many western physical scientists throughout the 1960s–1980s was that the Arctic was a relatively stable place (Macdonald, 1996). This view has been replaced by one of an Arctic where major shifts can occur in a very short time, forced primarily by natural variation in the atmospheric pressure field associated with the Northern-hemisphere Annular Mode (NAM).

The NAM, popularly referred to as the Arctic Oscillation (AO) (Wallace and Thompson, 2002), is a robust pattern of the surface manifestation of the strength of the polar vortex (for a very readable description, see Hodges, 2000). The AO correlates strongly (85–95%) with the more commonly used indicator of large-scale wind forcing, the North Atlantic Oscillation (NAO) (the NAO is the normalized gradient in sea-level air pressure between Iceland and the Azores—see for example, Deser, 2000; Dickson et al., 2000; Hurrell, 1995; Serreze et al., 2000). Here we will use the AO and NAO more or less interchangeably because they carry much the same information. We recognize, however, that in both cases the term 'oscillation' is a bit misleading because neither index exhibits quasi-periodic behaviour (Wallace and Thompson, 2002). The AO captures more of the hemispheric variability than does the NAO which is important because many of the recent changes associated with the AO have occurred in the Laptev, East Siberian, Chukchi and Beaufort Seas—a long way from the NAO's center of action (Thompson and Wallace, 1998). Furthermore, the Bering Sea and the Mackenzie Basin are both influenced to some degree

by atmospheric processes in the North Pacific (e.g., the Pacific Decadal Oscillation (PDO), and see Bjornsson et al., 1995; Niebauer and Day, 1989; Stabeno and Overland, 2001) whereas Baffin Bay ice climate appears to have an association with the Southern Oscillation (Newell, 1996) and the Canadian Archipelago and Hudson Bay probably respond to varied atmospheric forcings as yet not fully understood.

Although the AO is an important component of change in Arctic climate, it contains only 20% of the variance in the northern hemispheric atmospheric pressure field and much else can change in the atmospheric forcing. Because the AO is usually 'smoothed', it does not well represent events and short-term variations which are known to be important in the delivery of contaminants to the Arctic and may well also be important locally in forcing ice and surface water motion (see for example, Sherrell et al., 2000; Welch et al., 1991). It is important to note, therefore, that one of the projections of climate change is an increase in cyclone activity; extreme events may become a prominent component atmospheric transport in the coming century (an example may have been the earliest significant rain event on record (May 26, 1994) observed widely throughout the Canadian Archipelago and see Graham and Diaz, 2001; Lambert, 1995).

In about 1988–1989, the AO entered a positive phase of unprecedented strength (Fig. 6a,c). The pressure distribution pattern of the AO for winter and summer (Fig. 6b,d) shows that a positive shift is manifested by lower than average sea-level pressure (SLP) distributed somewhat symmetrically over the pole (Fig. 6b,d) and higher pressures over the North

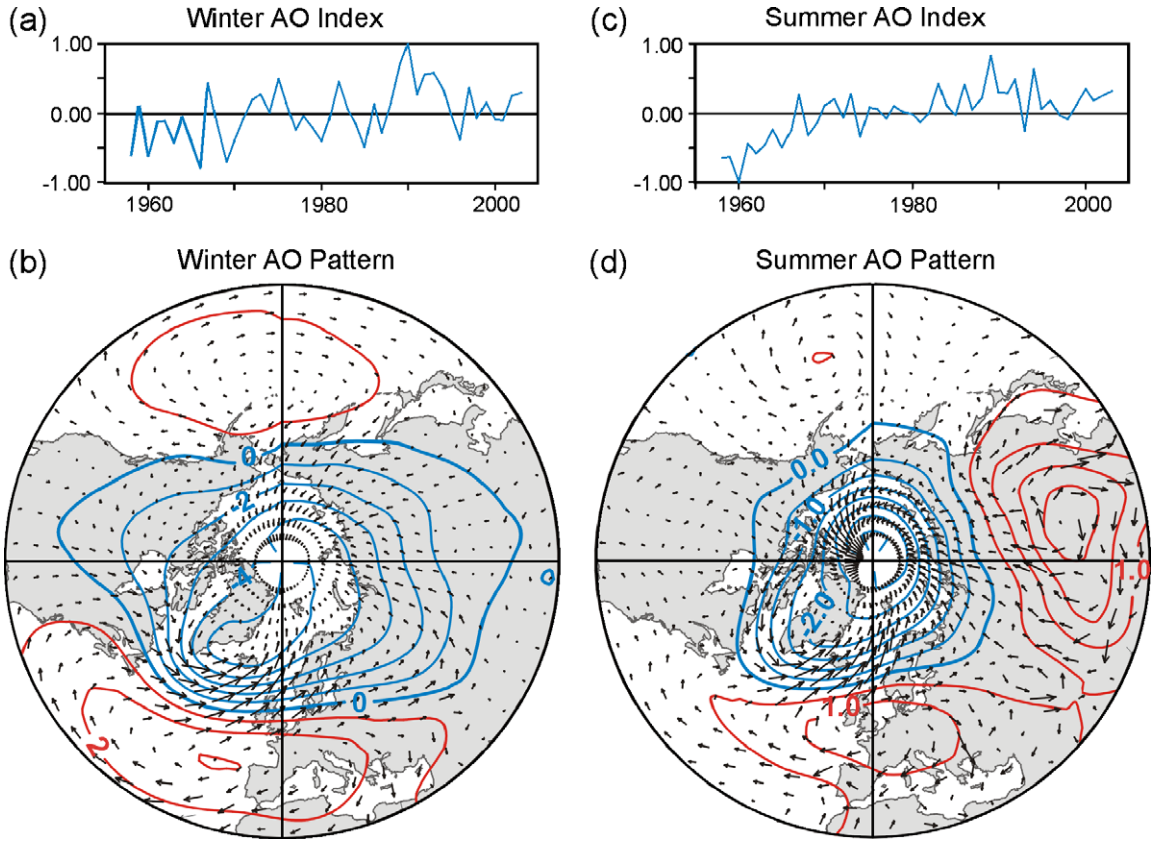

Fig. 6. (a) The Arctic Oscillation index between 1958 and 1998 in winter with (b) the winter pressure pattern associated with it and, (c) the Arctic Oscillation index between 1958 and 1998 in summer with (d) the summer AO pressure pattern associated with it. The summer and winter pressure patterns, which show the leading component of variance in the pressure-field time series, are multiplied by the respective AO index and added to the mean pressure field (along with the other variance components) to reproduce pressure fields with time.

Atlantic and North Pacific in winter and over Siberia and Europe in summer. As might be expected by examining the AO SLP pattern (Fig. 6b,d), when the AO index is strongly positive, conditions become more "cyclonic"—i.e., atmospheric circulation becomes more strongly counter-clockwise (Proshutinsky and Johnson, 1997; Serreze et al., 2000).

In discussing change it is important to distinguish between variability, which can occur at a variety of time scales (Fischer et al., 1998; Polyakov and Johnson, 2000) and trends caused by, for example, GHG warming. It has been argued that locking the AO into a positive position might actually be one way that a trend forced by GHGs can manifest itself in the Arctic (Shindell et al., 1999). Others, however, suggest that the extraordinary conditions of the 1990s were produced naturally by a reinforcing of short (5–7 years) and long (50–80 years) time-scale components of SLP variation (Polyakov and Johnson, 2000; Wang and Ikeda, 2001), and that GHG forcing will impact the mean property fields rather than alter the AO itself (Fyfe, 2002; Fyfe et al., 1999). Longer records of the NAO index (Fig. 7a) indeed suggest that there have been other periods of high AO index during the past 150 years (e.g., 1900–1914) but none so strong as that experienced during the early 1990s. During the late 1990s, the AO index decreased and the arctic system has to some degree begun to return to AO^- conditions (Björk et al., 2002; Boyd et al., 2002).

The contrast in conditions between the Arctic as we knew it in the 1960s, 1970s and 1980s (generally low AO index) and the Arctic it became in the early 1990s (exceptionally high AO index) provides an extraordinary opportunity to investigate how the Arctic might respond to climate change. Similarity between climate-change projections and AO-induced change suggests that contrasting the differences between AO^- and AO^+ states should provide insight into the likely effects of climate change forced by GHG emissions. Variation in SLP as recorded by the

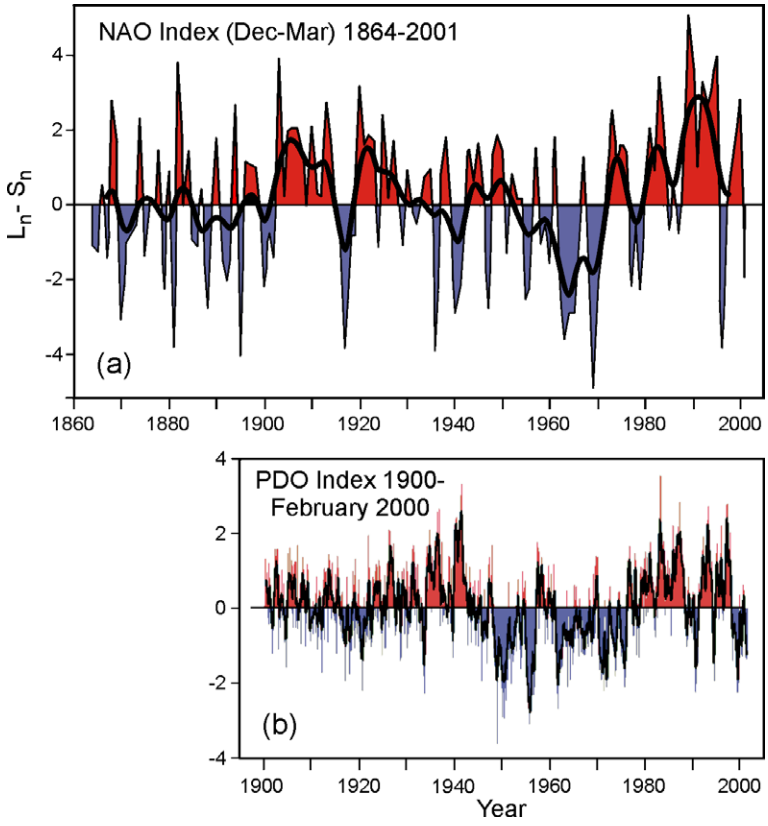

Fig. 7. The North Atlantic Oscillation (NAO) Index from 1860 to 2000 (source Hurrell, 2002).

AO index demonstrates that the Arctic exhibits at least two modes of behaviour (Morison et al., 2000; Proshutinsky and Johnson, 1997) and that these modes cascade from SLP into wind fields, ice drift patterns, watermass distributions, ice cover and probably many other environmental parameters.

The Arctic is to some degree constrained by overarching structures and processes in how it can respond to change. As illustrated in previous assessments, the Arctic Ocean is, and will remain, a 'mediterranean' sea much influenced by land–ocean interaction and with restricted exchange with other oceans (Fig. 1). Topography, bathymetry and global distribution of salinity in the ocean, require that water from the Pacific Ocean will predominantly flow *in* to the Arctic and the shallow sill at Bering Strait (50 m) guarantees only surface water will be involved in the exchange. Pacific water will remain above Atlantic Layer water which is denser. Deep basin water communicates predominantly with the Atlantic Ocean through the deep connection at Fram Strait. Ocean circulation within the Arctic is tightly tied to bathymetry through topographic steering of currents (Rudels et al., 1994) and these have a strong impact on the propagation of properties between basins and from basin to basin (McLaughlin et al., 2002). Considering these kinds of constraints, rapid change can occur in ocean current pathways or in the source or properties of the water carried by currents when, for example, fronts shift from one bathymetric feature to another (McLaughlin et al., 1996; Morison et al., 2000), when a given current strengthens or weakens (Dickson et al., 2000), when source-water composition alters (Smith et al., 1998; Swift et al., 1997), when relative strength of outflow varies between the Canadian Archipelago and Fram Strait (Macdonald, 1996), but not by reversal of flow in boundary currents or reversal of mean flow in at Bering Strait or out through the Archipelago.

Change associated with the NAM forces us to consider large-scale variability in the Arctic, to recognize in greater detail that physical pathways can change rapidly, and to assess potential effects of GHG emissions against this naturally variable background.

3.2. Winds

Winds transport contaminants directly to the Arctic by delivering volatile and semi-volatile chemicals from the south within a few days (Bailey et al., 2000; Barrie et al., 1998; Halsall et al., 1998; Hung et al., 2001; Stern et al., 1997). Winds also provide the primary forcing for ice and ocean (Mysak, 2001; Proshutinsky and Johnson, 1997) thereby indirectly affecting transport by these two media as well.

To understand how swings in the AO affect atmospheric circulation, we have constructed AO^+ and AO^- wind field/SLP maps for winter (Fig. 8a) and summer (Fig. 8b) by adding (AO^+) or subtracting (AO^-) patterns in Fig. 6b,d to the mean pattern for the period of record in the time series (Fig. 6a,c; 1958–1998). The changes discussed below can be considered generally as the difference between conditions during the 1960–1970s (low AO index) and during the early 1990s (high AO index) (see for example, Proshutinsky and Johnson, 1997).

In winter (Fig. 8a), the lower tropospheric circulation is dominated by high pressures over the continents and low pressures over the northern Pacific (Aleutian Low) and Atlantic Oceans (Icelandic Low). The Siberian High tends to force air on its western side into the Arctic acting as an effective atmospheric conduit from industrialized regions of Siberia and Eastern Europe into the high Arctic. The high-pressure ridge over North America then forces air southward giving a net transport out of Eurasia into the Arctic, across the Arctic and south over North America. The Icelandic Low produces westerly winds over the eastern North Atlantic and southerly winds over the Norwegian Sea providing a conduit for airborne contaminants from eastern North America and Europe to reach the Arctic rapidly. Finally, the Aleutian Low tends to steer air that has crossed the Pacific from Asia up into Alaska, the Yukon and the Bering Sea (Bailey et al., 2000; Li et al., 2002; Wilkening et al., 2000). During winter, these three routes into the Arctic—southerlies in the Norwegian Sea (40%), Eastern Europe/Siberia (15%), and Bering Sea (25%) account for about 80% of the annual south to north air transport (Iversen, 1996).

With a higher AO index (Fig. 8a left side), the Icelandic Low intensifies and extends farther into the Arctic across the Barents Sea and into the Kara and Laptev Seas (Johnson et al., 1999). This has the effect of increasing the wind transport east across the North Atlantic, across Southern Europe and up into the Norwegian Sea. During high NAO winters, westerlies

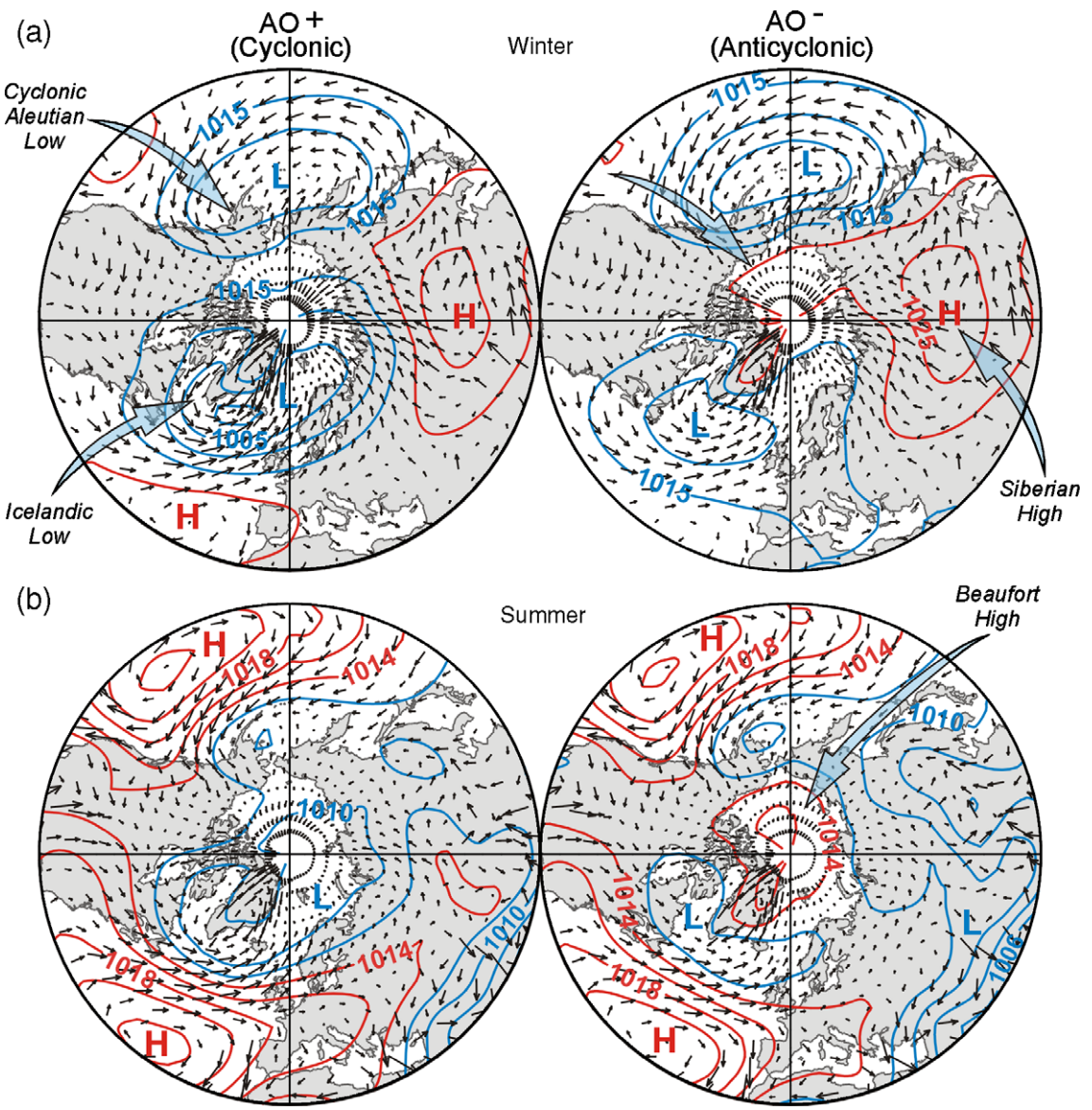

Fig. 8. Atmospheric pressure fields and wind stream lines for (a) winter with the Arctic Oscillation index strongly positive (left) and strongly negative (right) and (b) summer with the Arctic Oscillation index strongly positive (left) and strongly negative (right).

onto Europe may be as much as 8 m/s (~700 km/day) stronger (Hurrell, 1995). At the same time, strong northerly winds are to be found over the Labrador Sea (Mysak, 2001).

The extension of the Icelandic Low into the Arctic also implies an effect of the AO on the storm track. During the strong positive AO conditions of the early 1990s, there was a remarkable increase in the incidence of deep storms to around 15 per winter and such storms penetrated farther into the Arctic (Dickson et al., 2000; Maslanik et al., 1996; Semiletov et al., 2000). Increased cyclone activity increases poleward transport of heat and other properties carried by the air masses involved. Anomalous northward airflow over the Nordic Seas enhances the connection between industrial regions of North America and

Europe and the Arctic. At the same time, increased cyclones provide the means to transfer contaminants from atmosphere to surface through precipitation. Deep within the Arctic, the high SLP ridge that extends across the Canada Basin during AO^- conditions (the Beaufort High), disappears and withdraws into Russia (Johnson et al., 1999; Morison et al., 2000). Of note, the Pacific mean atmospheric pressure field and wind patterns appear to change little between strong positive and strong negative phases of the AO in winter. Penetration of air from the Pacific into the Arctic is hindered by the mountain barrier along the west coast of North America where intensive precipitation also provides a mechanism to remove contaminants and aerosols to the surface (Fig. 8a).

Summer pressure fields and air-flow patterns are markedly different from those of winter (compare Fig. 8a with Fig. 8b). In summer, the continental high-pressure cells disappear and the oceanic low pressure cells weaken with the result that northward transport from low latitudes weakens (Fig. 8b). According to Iversen (1996) summer accounts for only 20% of the annual south to north air transport (southerlies in the Norwegian Sea (10%), Eastern Europe/Siberia (5%), and Bering Sea (5%)). The streamlines show that winds provide a means to transport contaminants from industrialized North America and Europe to the North Atlantic but penetration into the Arctic weakens. In the North Pacific, there remain atmospheric pathways to move air masses into the Gulf of Alaska from the east coast of Asia (Fig. 8b). In particular, during AO^+ conditions the Beaufort High weakens or disappears (Fig. 8a,b), altering mean wind fields.

3.3. Surface air temperature

A strong trend of warming has been observed in the Arctic for the period from 1961 to 2000 (Fig. 3b). This warming, which has been especially evident over northwestern North America and Siberia, has been accompanied by cooling in northeastern Canada and Baffin Bay. An almost identical pattern of warming to that shown in Fig. 3a is produced by taking the difference between mean surface air temperatures on high AO index days and low AO index days (Wallace and Thompson, 2002). An extensive temperature record collected from drifting buoys, manned drifting stations and land stations, allows us to draw direct relationships between air surface temperature over sea and land in the Arctic and the changes in pressure field discussed above. Over the period 1979–1997, a trend of +1 °C per decade was found for winter surface air temperature (SAT) in the eastern Arctic Ocean, offset by a trend of −1 °C per decade in the western Arctic Ocean (Rigor et al., 2000). However, in spring almost the entire Arctic Ocean shows significant warming—as much as 2 °C per decade in the eastern Arctic where a trend toward lengthened melt season was also observed. The trend of increasing SAT over the ocean is matched by temperature increases over Arctic land masses of 2 °C per decade during winter and spring (Fig. 3a). Long records of temperature anomalies since 1900 (Fig. 3b) clearly show the warming trend since the 1970s but note also the similar episode of warming that occurred in the 1930–1940s. Taken together, the trends in surface air temperature over the central Arctic Ocean suggest that warming has occurred predominantly during January to July (Fig. 3c; Serreze et al., 2000). Over half of the changes in SAT for Alaska, Eurasia and the eastern Arctic Ocean have been ascribed to the AO but less than half in the western Arctic (see Dickson et al., 2000; Rigor et al., 2000; Serreze et al., 2000). The temperature changes associated with the AO are considered large enough to have an immediate effect on polar circulation (Morison et al., 2000).

3.4. Precipitation and runoff

Precipitation is a key pathway for contaminant transport (Fig. 2); rain or snow scavenge aerosols and gasses from the atmosphere to deposit them at the surface (Li et al., 2002; Macdonald et al., 2000a; Mackay and Wania, 1995; Wania and Halsall, 2003; Wania and Mackay, 1999). Scavenging by precipitation may presently be relatively weak due to the desert-like conditions of the Arctic. For example, mean precipitation for the Arctic Ocean is estimated at about 25.2 cm $year^{-1}$ and evaporation about 13.6 cm $year^{-1}$ for a net moisture flux to ground of 11.9 cm $year^{-1}$ (Barry and Serreze, 2000). The net precipitation over land in the arctic drainage basins is apparently greater as implied by runoff yield (precipitation minus evaporation (P−E)) being estimated at 21.2 cm $year^{-1}$ from the network of gauged discharge by rivers (Lammers et al., 2001).

The changes in winds (Fig. 8a,b) and temperature that occur with shifts in the AO imply that precipitation and evaporation within the Arctic will also be affected either in amount or seasonality (Serreze et al., 2000). It is difficult to assess with confidence the spatial or temporal variation of precipitation within the Arctic due to sparse networks and short time series. Nevertheless, records suggest that precipitation has been increasing over Canada's North by about 20% during the past 40 years (Serreze et al., 2000). The increase in southerly winds in the Norwegian Sea in winter and penetration of cyclones from the Atlantic into the Barents, Kara and Laptev Seas, when the AO (or NAO) index is high is reflected in increased moisture flux and precipitation during autumn and winter especially near 10°W–50°E (Fig. 9a,b, Dickson et al., 2000; Serreze et al., 2000; Walsh, 2000). The composite difference in precipitation (Fig. 9b), which may actually underestimate the change between index extremes (Dickson et al., 2000), shows an increase of up to 15 cm year^{-1} precipitation in the Norwegian–Greenland Sea atmosphere–ocean conduit to the Arctic when the NAO is strongly positive. The response over the central Arctic to changes in the AO/NAO index is clearly much less but it is likely that conditions there become wetter during index highs (Serreze et al., 2000). Overall, it is estimated that P−E north of 70°N is 36% higher during periods of high index compared to low index (Serreze et al., 1995). Over central and northern Canada, flux of moisture out of the Arctic increases when the AO/NAO is high, but toward the western Beaufort Sea and Chukchi Sea moisture flux into the Arctic again increases (red line on Fig. 9a).

Whether precipitation falls as snow or as rain, and how long snow covers surfaces are crucially important components of climate that control the interaction of contaminants with the hydrological cycle (Helm et al., 2002; Macdonald et al., 2002a; Wania, 1997).

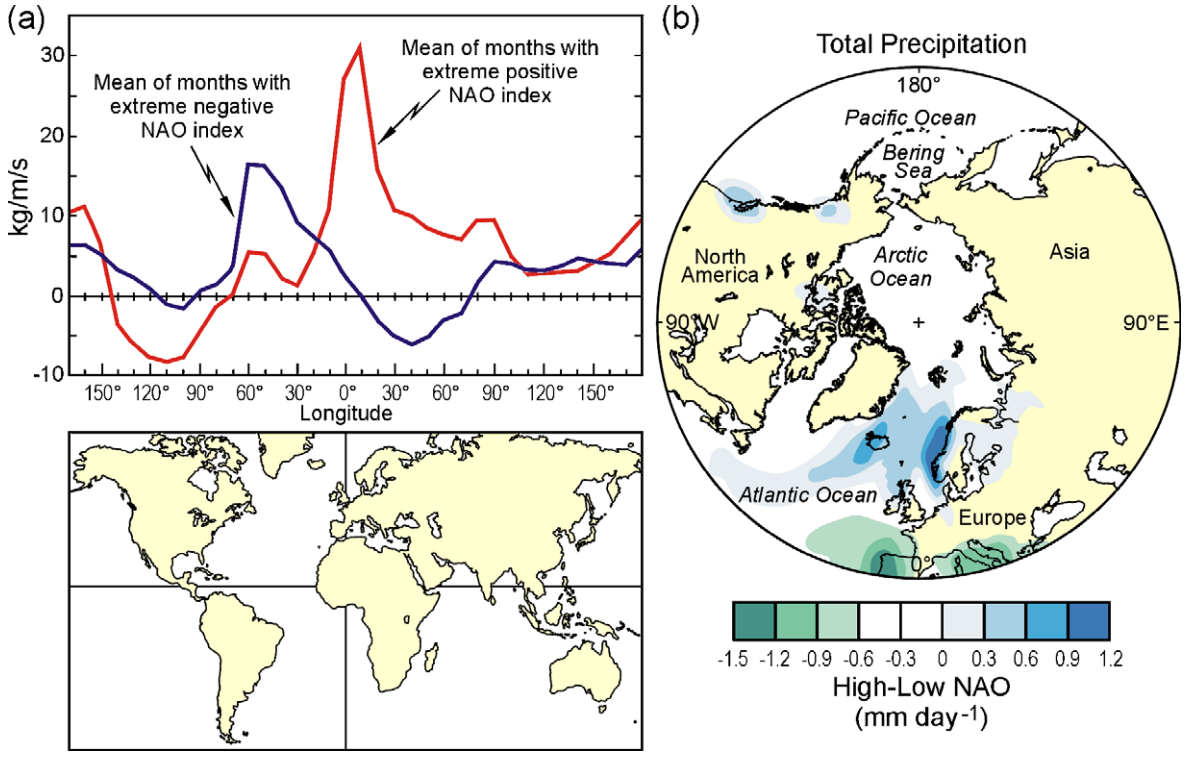

Fig. 9. The effect of the Arctic Oscillation on precipitation in the Arctic showing (a) vertically integrated meridional flux crossing 70°N in winter for low NAO conditions (blue) and high NAO conditions (red) and (b) the change in winter precipitation between low-index NAO and high-index NAO in mm per day (modified from Dickson et al., 2000). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Furthermore, the effect of a switch between snow and rain depends on the physical properties of the chemicals (Wania and Halsall, 2003) and, therefore, a change in the relative amounts of snow or rain may even have opposing effects for two different contaminants. Snow cover varies from a maximum of about 46×10^6 km^2 to as little as 4×10^6 km^2 (Serreze et al., 2000). The recent warming trends over arctic land masses (Fig. 3a) are paralleled by a decreasing trend in the average area covered by snow by about 2% (450,000 km^2) per decade between 1979 and 1999 (Armstrong and Brodzik, 2001). A correlation between the AO and snow cover in Eurasia for the period from 1972 to 1997 suggests that a change from minimum to maximum AO index is accompanied by a loss of about 4×10^6 km^2 of snow cover which could account for much of the trend described above (Vörösmarty et al., 2001). The snow-cover anomalies plotted by Armstrong and Brodzik (2001) show a downward step in about 1989 when the AO index sharply increased. From the late 1980s up to at least 1998 has been identified as a period of low snow cover for both Eurasia and North America with the largest changes occurring in spring–summer (Serreze et al., 2000) and, for Canada, there has been a decrease in snow depth, especially in spring, since 1946 (Brown and Goodison, 1996).

Precipitation minus evaporation integrated over a drainage basin should be equivalent to river discharge for the basin. Arctic rivers exhibit large interannual variation (Semiletov et al., 2000; Shiklomanov et al., 2000; Stewart, 2000) making it difficult to link river flow to precipitation or temperature trends or to climate variables like the AO. For example, Shiklomanov et al. (2000) suggested little change in mean annual discharge for arctic rivers between the 1920s and 1990s whereas Semiletov et al. (2000) found recent increases for several Eurasian rivers and Lammers et al. (2001) found evidence of increased winter discharge from rivers in Siberia and Alaska in the 1980s compared to the 1960s–1970s. Within Canada, the Mackenzie Basin has undergone an exceptional warming between 1961 and 1990 (Fig. 3a, Stewart et al., 1998); nevertheless, increased basin temperatures are not obviously recorded in this river's hydrology (Fig. 10a, Stewart, 2000) or in other arctic rivers (Shiklomanov et al., 2000). Instead, one sees evidence of 3- to 4-year periodicity in peak flow and alterations in the seasonal shape of the hydrograph with higher flows delayed well into August suggesting changes in the total annual discharge and its seasonality and possibly changes in the relative importance of the river's sub-drainage-basins. Such patterns appear to be only partially related to the AO as evidenced by significant correlations between runoff and precipitation for the Mackenzie Basin and variation in North Pacific storm tracks (Bjornsson et al., 1995). These correlations hint that trans-Pacific transport of airborne contaminants may be the dominant component of contaminant loading for northwestern Canada, something that is borne out by time-series data collected on the eastern side of the Pacific Ocean extending all the way from Tagish, in the Yukon (Bailey et al., 2000), to the continental United States (Jaffe et al., 2003). Hence, change related to atmospheric contaminant pathways for this region is more likely to come from the North Pacific, and it is possible that such change might be manifested as an alteration in the domains of influence for Pacific air masses versus Eurasian air masses.

The discharges for the Ob, Yenisei and Mackenzie Rivers appear to show a positive relationship with the North Pole pressure anomaly with a lag in discharge of about 0.5–0.7 years (Fig. 10b, Johnson et al., 1999) but such a relationship runs counter to the enhanced precipitation observed during AO$^+$ (cyclonic) conditions (Fig. 9). Even if all variation in arctic river discharge at the 4–5 year time scale is assigned to shifts in AO/NAO index, the maximum effect on annual flow would be about 5–15% which is within the range of interannual variability (for example see Johnson et al., 1999; Semiletov et al., 2000).

3.5. The ocean

3.5.1. Sea ice

Sea ice controls the exchange of heat and other properties between atmosphere and ocean and, together with snow cover, suspended particles and dissolved organic matter, determines the penetration of light into the sea. Ice also provides a surface for particle and snow deposition, a biological habitat above, beneath and within the ice and, when it melts in summer, stratification of the upper ocean.

During the 1990s, the science community recognized with some alarm that arctic sea ice had been

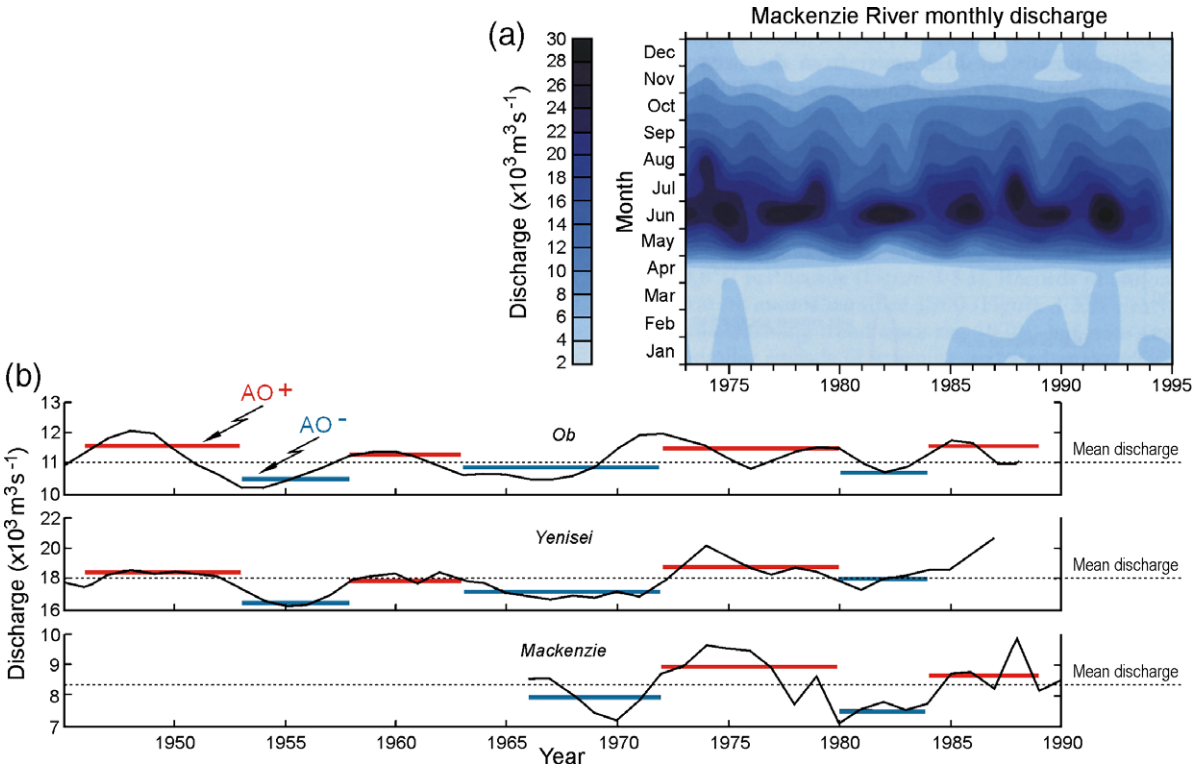

Fig. 10. (a) The monthly river discharge for the Mackenzie River from 1973 to 1995 inferred from observations at Arctic Red River and the Peel River (after Stewart, 2000) and (b) the relationship between the cyclonic (AO^+)/anticyclonic (AO^-) states and the 3-year smoothed monthly river discharge for the Ob, Yenisei and Mackenzie Rivers (after Johnson et al., 1999).

undergoing retreat over the past three decades. Observed changes include: a reduction in area covered by sea ice (Johannessen et al., 1999; Levi, 2000; Maslanik et al., 1996; Parkinson et al., 1999; Vinnikov et al., 1999), an increase in the length of the ice melt season (Rigor et al., 2002; Smith, 1998), a loss of multiyear ice (Johannessen and Miles, 2000), a general decrease in the thickness of ice over the central Arctic Ocean (Rothrock et al., 1999) and an increase of ice melt in the Beaufort Sea (Macdonald et al., 1999a; McPhee et al., 1998).

Analyses of satellite data from 1978 to 1987 revealed a decrease in arctic sea-ice area of about 2.4% per decade (Gloersen and Campbell, 1991). Subsequent analyses have revised that figure upward to 4% per decade for the period from 1987 to 1994 with an estimated average loss during the entire period (1978–1997) of 3% per decade which amounts to the disappearance of 0.3×10^6 km^2 per decade of sea ice (Cavalieri et al., 1997; Parkinson et al., 1999). Shelves of the Eastern Arctic contribute significantly to the estimated ice losses. Multi-year ice is apparently being lost at an even greater rate, estimated at 7% per decade, partly replaced by first-year ice (Johannessen and Miles, 2000).

The large seasonal amplitude in area covered by ice (Fig. 11) makes it difficult to assess trends. Furthermore, various authors have partitioned the Arctic differently to assess changes in ice cover or have compared different years and/or different seasons (Dickson et al., 2000; Johannessen and Miles, 2000; Maslanik et al., 1996, 1999; Parkinson et al., 1999). Despite these difficulties, the satellite data available since the late 1970s clearly imply a reduction of 2% per decade of total ice area in winter (Johannessen et al., 1999) and a significant shift in the marginal seas toward first-year ice which is easier to melt than multiyear ice because it is thinner and saltier. The total area of arctic sea ice, including the marginal seas, varies from about 13×10^6 km^2 in

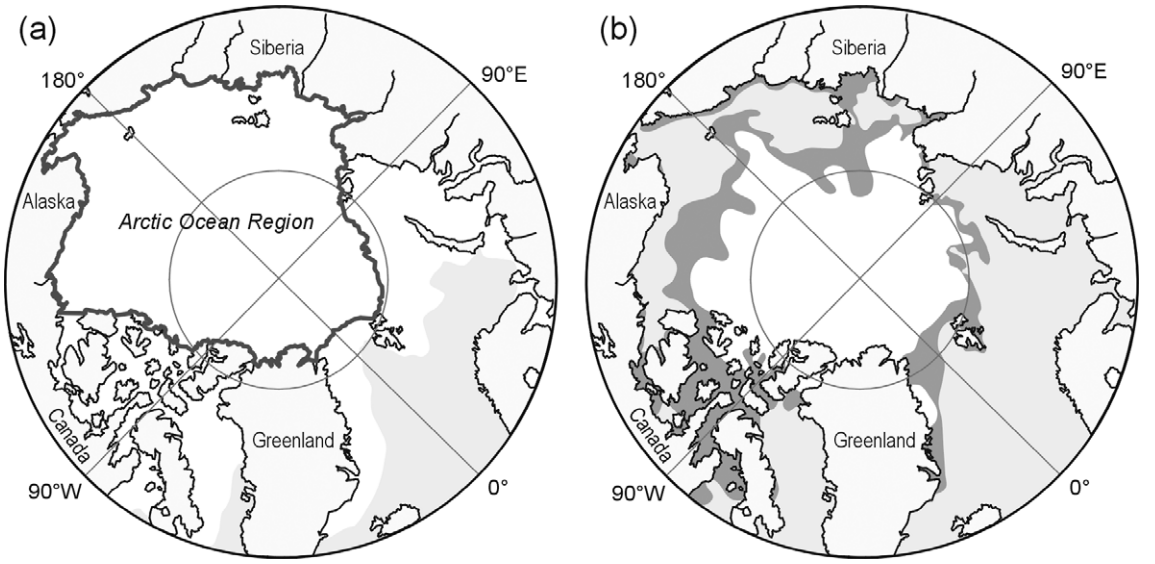

Fig. 11. Arctic sea ice cover in (a) winter maximum and (b) summer minimum as derived from satellite imagery (Johannessen and Miles, 2000). Grey areas indicate partial ice cover (greater than 15%). The dashed line in a) outlines the area defined as Arctic Ocean for the ice area trends given in Parkinson et al., 1999.

winter to 5×10^6 km^2 in summer, and has shrunk by about 0.6×10^6 km^2 between 1978 and 1997 (Johannessen and Miles, 2000). The Arctic Ocean component as defined by Parkinson et al. (1999) (Fig. 11), which is about 7×10^6 km^2 in area, began to exhibit a much stronger seasonal modulation in ice cover in about 1989 (Fig. 12a,b) with the East Siberian and Beaufort Seas experiencing anomalous areas of open water in late summer at various times during the 1990s (Maslanik et al., 1999; Parkinson et al., 1999; Rigor et al., 2002; Serreze et al., 1995). That the loss of sea-ice cover is predominantly a spring–summer phenomenon is clearly shown by seasonal monthly trends for which June to October show the greatest change (Fig. 12c, Serreze et al., 2000).

What part does the AO play in the variation of arctic sea-ice distribution? The trends in ice cover with time (Fig. 12a,b) suggest that the wholesale clearing of ice from shelves is a phenomenon of the 1990s, timed with (Russian Shelves) or slightly delayed from (Beaufort Shelves) the shift to strong positive AO index in 1989. In the Beaufort Sea, Macdonald et al. (1999a) used stable isotope data (δ^{18}O) collected from 1987 to 1997 to show that amounts of ice melt contained in the water column increased substantially at the same time the AO index increased in 1989. During such conditions, the cyclonic circulation leads to greater ice divergence, more new ice in leads, enhanced heat flux, and reduced ridging all of which imply thinning (Flato and Boer, 2001; Macdonald et al., 1999a; Rigor et al., 2002). Maslanik et al (1996) draw the connection between increased cyclones and increased poleward transport of heat, which are observed during AO$^+$ conditions, and the absence of ice in late summer over the Siberian shelves. Based on results of a coupled sea/ice/ocean model, Zhang et al. (2000) suggest that there is a strong correlation between sea-ice thinning and the AO (~80%) due to dynamical effects, and that the Eurasian and Canada Basins respond differently to the AO forcing. The removal of the supply of ice from the Beaufort to the East Siberian Sea when the index goes strongly positive (discussed below under sea-ice drift) results in depletion of thick ice in the Eastern Arctic Ocean but may enhance thick ice buildup to the north of the Archipelago. This point is important in light of the findings from repeat submarine surveys that ice thickness has decreased over the central Arctic by about 1.3 m between 1958–1976 and the 1990s (Rothrock et al., 1999; Wadhams, 1997, 2000). According to several models (Holloway and Sou, 2001; Polyakov and Johnson, 2000; Zhang et al.,

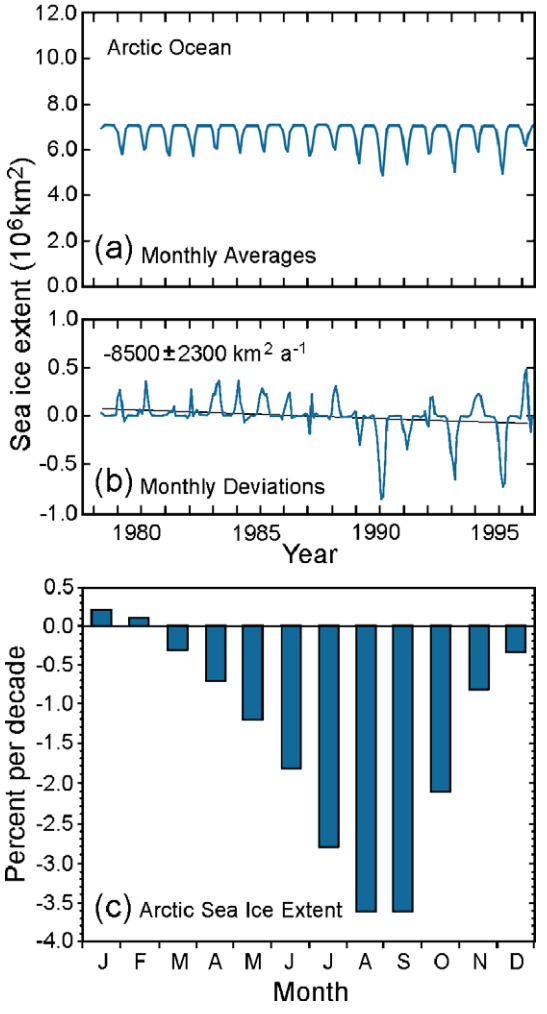

Fig. 12. Ice cover in the Arctic Ocean showing (a) Monthly averages in ice cover for the period from 1979 to 1996 for the Arctic Ocean (as defined by dashed line in Fig. 11a (after Parkinson et al., 1999)), and (b) the monthly deviations in ice cover for the same area showing the transition in 1990 to seasonally clear shelves (after Parkinson et al., 1999) and (c) the change in sea ice extent in % per decade (1979–1995) showing the ice loss to be predominantly a late winter–summer phenomenon (after Serreze et al., 2000).

2000), the submarine observations may have been conducted primarily in that part of the ocean that underwent thinning in response to a shift to high AO index. The conclusion of reduction of ice thickness, while valid for the domain of submarine measurements, is not necessarily true for the whole Arctic Ocean and there is an alternative hypothesis that ice thickness *distribution* changed but ice *volume* may not have changed in response to the AO. Recently, Yu et al. (in press) re-analyzed the submarine data and confirmed the loss of thick ice (>4 m) which they suggested was likely cyclical and at least partially thermally forced (i.e. ice melting is involved). The loss of ice cover between NAO^- and NAO^+ conditions is estimated at 590,000 km^2 in the Barents and Greenland Seas (Dickson et al., 2000) and if the remarkably open ice in the East Siberian Sea in 1990 and the Beaufort Sea in 1998 (Fig. 13) is a product of the high AO index of the early 1990s, then perhaps half as much again ice loss occurred over the Russian and North American Shelves due to AO forcing.

In light of the changes observed in ice cover during the 1990s it is worth noting that over a century ago the Pacific whaling fleet experienced similar dramatic changes in ice conditions in the western Arctic. Extraordinarily open water from 1861 to 1867 perhaps led to a complacency that resulted in the loss of 32 ships, crushed in the ice along the Alaska coast in 1871 (Bockstoce, 1986). One is reminded of the caution given by Polyakov et al. (2002a, 2003a,b, 2000) that short-term (decadal) oscillations may be misleading when they occur within a background of

Fig. 13. The contrast in ice cover between low AO^- index (1958) and high AO^+ index (1990s) (Dickson et al., 2000; Maslanik et al., 1999; Serreze et al., 1995).

low-frequency oscillations having periods of 60–80 years.

From data collected between 1979 and 1997, Rigor et al. (2000) determined that sea-ice melt begins in the marginal seas by the first week of June and advances rapidly to the pole in 2 weeks. Freezing begins at the pole on 16 August, returning to the marginal seas by late September for a total melt season length of about 58 days at the pole and 100 days toward the margin. Based on satellite data (SSMR and SSM/I) predominantly from the Beaufort Sea, Smith (1998) estimated that the length of the melting season has been increasing by about 5.3 days per decade during 1979–1996. In contrast, Rigor et al. (2000) found a shortening of the melt season in the western Arctic of 0.4 days per decade and an increase of about 2.6 days per decade in the Eastern Arctic.

Change in ice cover and its seasonality are especially important for contaminants like HCH, toxaphene and PCBs where air–sea exchange is a significant component of the Arctic Ocean budget (Li et al., 2002, 2004; Macdonald et al., 2000a). Furthermore, change in sea-ice cover, which alters light penetration and mixing, also alters primary production and carbon flux (Gobeil et al., 2001b) which then alters the vertical flux of particle reactive and bio-active contaminants from the ocean surface to depth. Recent work has shown that vertical flux of carbon-rich material has been very much neglected in accounting for POPs cycling in the ocean, and that the vertical export of POPs on sinking particles provides crucial forcing to air–sea exchange of POPs in temperate seas (Dachs et al., 2002). Clearly, potential changes in ice climate make the Arctic susceptible to change in organic carbon cycling (Carmack and Chapman, 2003; Stein and Macdonald, 2004) which then has the potential to alter air–sea exchange and vertical distribution of contaminants in the upper ocean. This latter would then feed back to the foodweb depending on vertical migrations and foraging of zooplankton plankton (Borgå et al., 2002).

General ice motion in the Arctic Ocean is organized into the Transpolar Drift (TPD) on the Eurasian Side of ocean and the Beaufort Gyre in the Canada Basin (Barrie et al., 1998). Although it has long been recognized that large-scale ice-drift patterns in the Arctic undergo change (Gudkovich, 1961), it was not until the International Arctic Buoy Programme (IABP) that sufficient data became available to map the ice drift in detail and thereby directly evaluate the role of the AO in changing ice drift trajectories. The IABP data from 1979 to 1998 suggest two characteristic modes of arctic ice motion, one during low index (AO^-) and the other during high index (AO^+) (Fig. 14a,b, Proshutinsky and Johnson, 1997; Rigor et al., 2002). The ice-motion scheme shown by drifting buoys is reasonably well corroborated by models investigating the influence of the atmospheric variability inherent in the AO (Maslowski et al., 2000; Polyakov and Johnson, 2000). There are two overarching differences between the two ice circulation modes; (1) during AO^- conditions (Fig. 14a), ice in the TPD tends to move directly from the Laptev Sea across the Eurasian Basin and out into the Greenland Sea whereas during strong AO^+ conditions (Fig. 14b), ice in the TPD takes a strong cyclonic diversion across the Lomonosov Ridge and into the Canada Basin (Mysak, 2001) and (2) during AO^+ conditions (Fig. 14b), the Beaufort Gyre shrinks back into the Beaufort Sea and becomes more disconnected from the rest of the Arctic Ocean exporting less ice to the East Siberian Sea and importing little ice from the region to the north of the Archipelago—a region known to contain the Arctic's thickest multi-year ice (Bourke and Garrett, 1987).

There are also changes in the time required for ice to transit the ocean (Fig. 15a,b, Rigor et al., 2002) and for the destinations of ice exported from shelves—i.e., connections between sources and sinks of ice. During winter under AO^+ conditions, there is an increase in ice advection away from the East Siberian and Laptev Sea coasts leading to the production of more new, thin ice in the coastal flaw leads (Fig. 16, Polyakov and Johnson, 2000; Rigor et al., 2002), a decrease in the advection of ice from the western Arctic into the eastern Arctic, possibly an increased advection of ice from the Arctic Ocean to the Barents Sea through the Spitzbergen–Franz Josef Land passage (Polyakov and Johnson, 2000), and an increase in the 900,000 km^2 $year^{-1}$ of ice advected out of the Arctic at Fram Strait (Morison et al., 2000; Rigor et al., 2002). Interestingly, increased ice export through Fram Strait can be produced by shifts to both negative and positive AO phases (Dickson et al., 2000).

Comparing the two modes of ice drift (Fig. 14a,b), it is apparent that during AO^- conditions the East

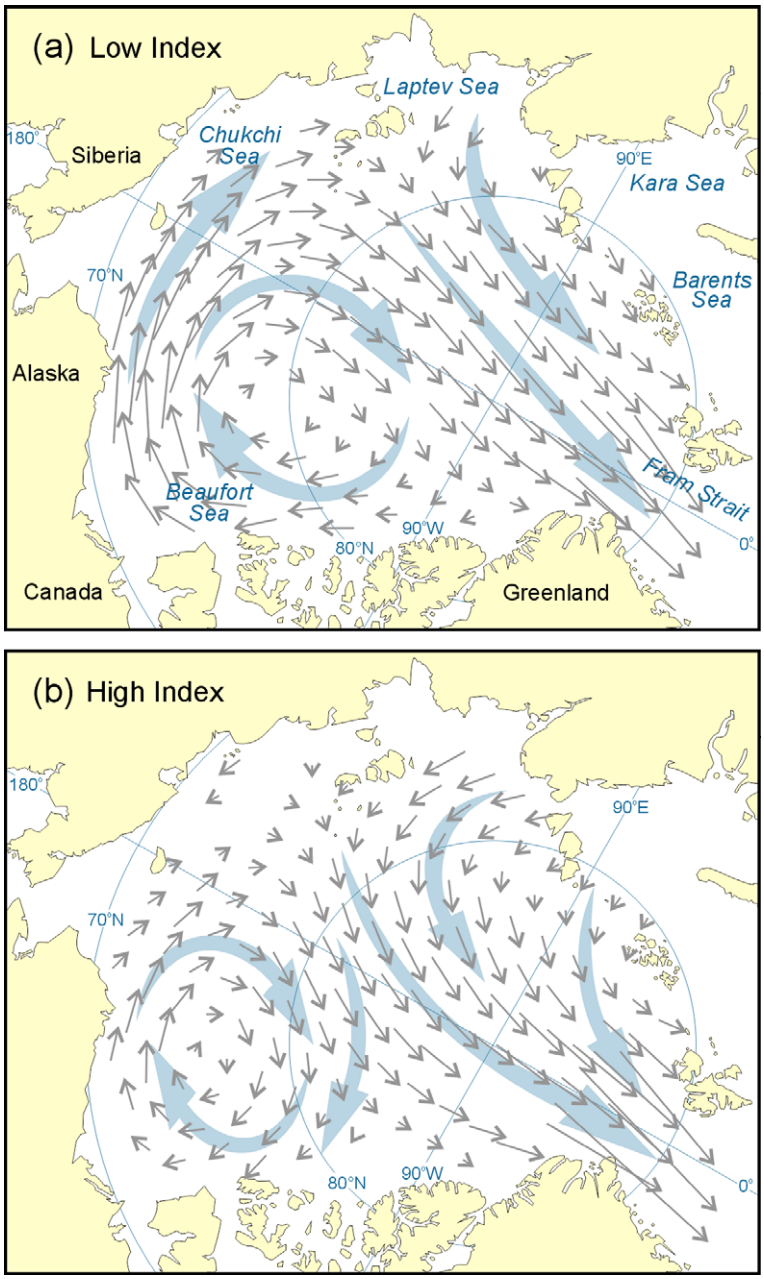

Fig. 14. Ice drift patterns for (a) years with low AO^- index (anticyclonic conditions) and (b) high AO^+ index (cyclonic conditions) (after Maslowski et al., 2000; Polyakov and Johnson, 2000; Rigor et al., 2002).

Siberian Sea imports much of its ice from the Beaufort Sea and that there is an efficient route to carry ice clockwise around the arctic margin of the East Siberian Sea and out toward Fram Strait. Under the strong AO^+ conditions of the early 1990s, the Beaufort Sea ice became more isolated whereas ice from the Kara, Laptev and East Siberian Seas was displaced into the central Arctic and toward the Archipelago. It is not

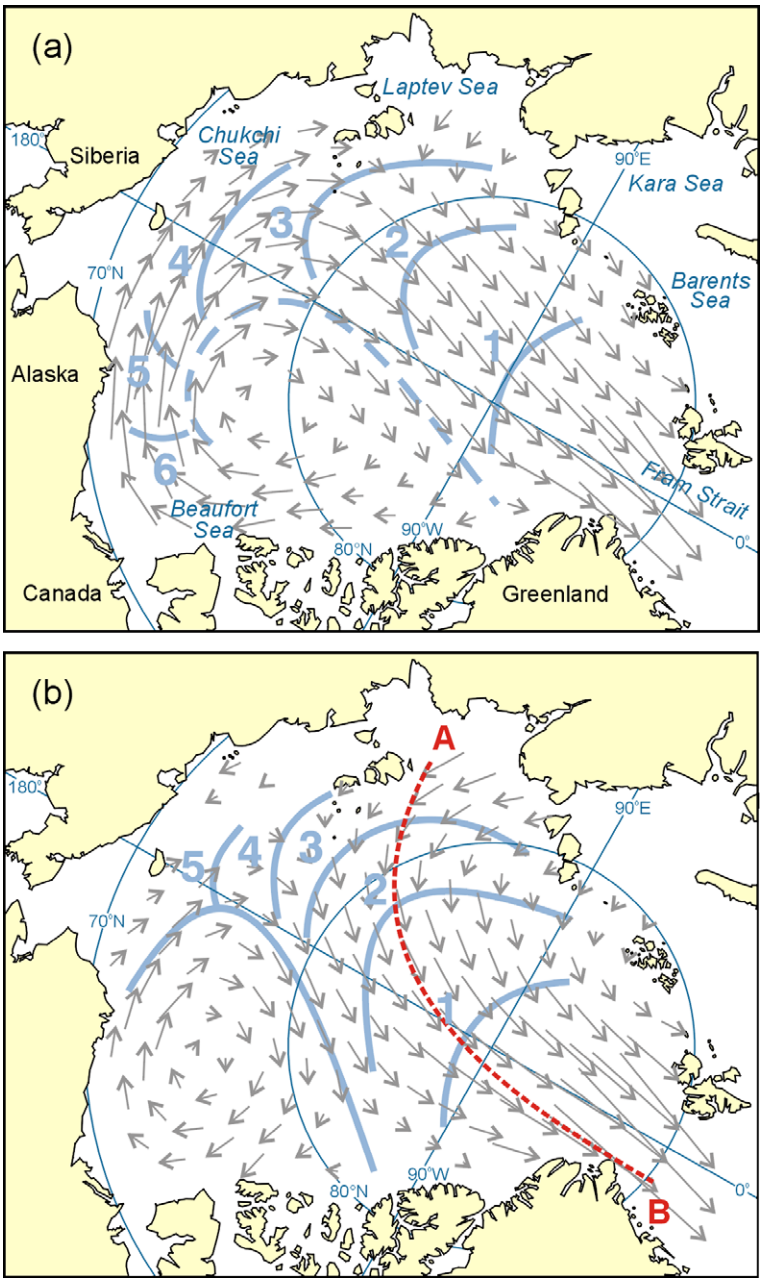

Fig. 15. Time taken in years for sea ice to reach Fram Strait during (a) low AO^- index conditions and (b) high AO^+ index conditions (after Rigor et al., 2002). Line A–B represents the transect used to describe change in sea ice during drift shown in Fig. 17.

clear from the IABP data how much ice from the Russian Shelves might transport into the Canadian Archipelago or the Beaufort Gyre under AO^+ conditions, but models (Maslowski et al., 2000; Polyakov and Johnson, 2000), palaeo-studies of Eurasian wood (Dyke et al., 1997; Tremblay et al., 1997) and sediment records (Darby et al., 2001) all suggest that such transport is likely and may at times be important.

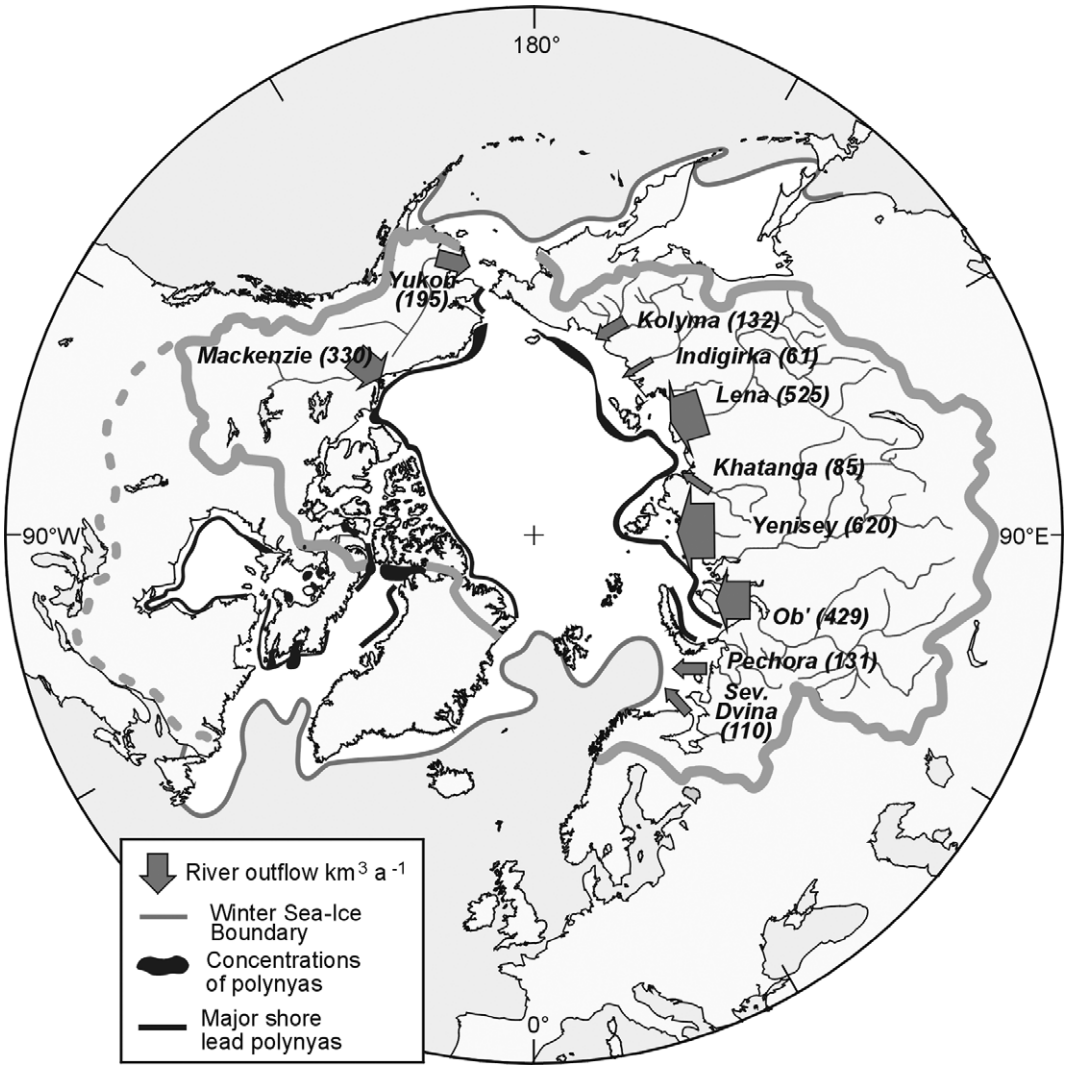

Fig. 16. The Arctic Ocean showing its predominant connections with the Atlantic and Pacific Oceans, the drainage basin, the runoff, the distribution of polynyas and the sea ice production.

Sea ice provides a rapid means to accumulate and transport contaminants long distances without dilution (Pfirman et al., 1995; Wadhams, 2000). The response in ice-drift trajectories to change in AO index (Rigor et al., 2002) therefore carries immense implication for the connectivity between contaminant source and sink regions for ice pathways within the Arctic Ocean.

Ice formation and drift provide an important means to transport coastal and continental shelf sediments to the interior ocean and out into the Greenland Sea (Barrie et al., 1998; Dethleff et al., 2000b; Nürnberg et al., 1994). Although all the shelves of the Arctic are implicated in this process, the Laptev Sea has proven so far to be the most efficient exporter of sediment-laden ice (Eicken et al., 1997, 2000; Reimnitz et al., 1992, 1993, 1994). This transport process involves several steps including (1) the delivery of sediment to the shelf from rivers or from coastal erosion where much of it may become trapped; (2) the incorporation of sediment into the ice, either through ice grounding or through suspension freezing in mid-shelf flaw polynyas; (3) the export of ice from the shelf to the

interior ocean; (4) the transport of ice across arctic basins potentially with some loss of sediment during transport and (5) the release of sediment at the location where the ice melts (Fig. 17). During transport, the ice 'weathers', ablating at the surface during summer and incorporating more ice on the bottom during winter, with the consequence that some of the sediment entrained over the shelf migrates to the surface of the ice (Barrie et al., 1998). Additionally, atmospheric particulates accumulate on the ice along the transport route and, therefore, increase or decrease in the time taken for ice to cross the Arctic Ocean (Fig. 15), respectively, increases or decreases the time available for accumulation of atmospheric aerosols and sediments at the ice surface. These factors have clear implications for biota living on, beneath or within the ice, but perhaps the environment most sensitive to change is the last. Krembs et al. (in press) discuss specifically the importance of chemical composition in brine pockets in ice-environments that are maintained in a liquid state suitable for organism survival at −50 °C temperatures by exclusion of brine and organic compounds when ice forms. This 'extreme habitat,' whose importance to the arctic carbon cycle and even to the physical properties of ice itself is only now being recognized, may be exceptionally vulnerable to contaminants accumulated on the ice and concentrated into the pockets in the same way that brine is.

Each step in the ice pathway can be altered by climate change. For example, fine river sediments (known to carry contaminants) become trapped in estuaries by the so-called 'marginal filter' (Lisitzin, 1995). Sea level rise, change in the ice climate or change in the river's hydrology alters the location of this filter. The process of suspension freezing might be enhanced by larger amounts of open water over shelves in the fall whereas more sediment might be lost from the ice during transport due to predominance of thin, first-year ice and augmented melting. Finally, the location at which ice melts and drops its particulate and dissolved loads can be changed. In a paper written prior to the 'discovery' of the Arctic Oscillation, Untersteiner (1988) suggested that the location of the sea-ice margin northwest of Spitsbergen is controlled dynamically by opposed advection

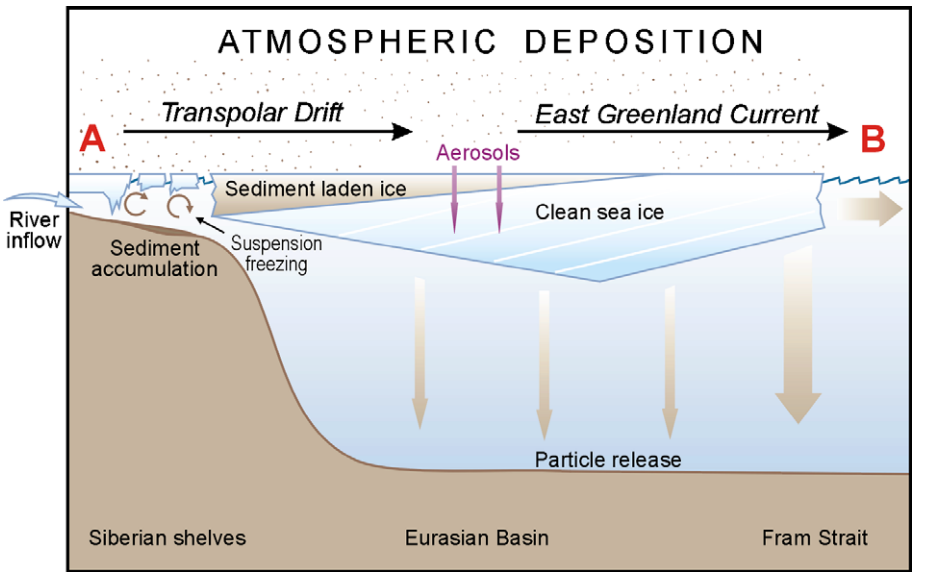

Fig. 17. A schematic diagram showing the accumulation and transport of sediments and contaminants by ice following the path marked A–B on Fig. 15 (modified from Lange and Pfirman, 1998). Sediments become incorporated in ice formed over shelves. Once exported to the interior ocean, the ice joins general ice circulation patterns (Figs. 14 and 15) which deliver much of the ice to the Greenland Sea in a time scale of two to six years. During its drift, the ice accumulates contaminants through surface deposition and it also weathers, ablating in summer and accumulating more ice on the bottom of floes in winter. Change in the location where ice melts or in the time taken to arrive there alters the contaminant pathway associated with ice transport.

of ice from the Arctic and warm seawater from the Atlantic. Untersteiner further proposed that most of the sea-ice melt water re-entered the Arctic Ocean forming a feedback cycle which supported Arctic Ocean stratification. Accordingly, there appears to be strong circumstantial evidence that under historical circulation patterns, the ice contaminant conveyor continuously deposited its load into the ocean near Spitsbergen where, coincidentally, some of the Arctic's highest POPs concentrations have been found in marine mammals (AMAP, 1998). Change in the dynamical balance between ice and heat will alter not only the location at which contaminants are released from the ice but also the balance between recirculation back into the Arctic Ocean and export into the North Atlantic.

There are no direct data on how the above components of the ice-transport pathway respond either individually or collectively to the AO; however, long-term sediment records (Darby et al., 2001), disequilibria in sediments (Gobeil et al., 2001b) and the distribution of sediments within the Arctic Ocean (Stein, 2000) suggest that climate forcing akin to the AO likely occurs.

3.5.2. Ocean currents and water properties

For ocean currents that deliver contaminants to arctic ecosystems, surface water is most important because it interacts most directly with biota and ecosystems. It is assumed that surface water pathways will to some extent reflect ice-drift trajectories (Morison et al., 2000), responding in like manner to the state of the AO (Fig. 14a,b) although there are actually very few data with which to evaluate this hypothesis due to the difficulty of making measurements. With a high AO index, water in the TPD makes a diversion into the Makarov Basin and the Beaufort Gyre contracts and retreats into Canada Basin. However, the AO produces other crucial changes in surface water not represented by ice drift. With the enhanced inflow and spreading of water in the Atlantic layer, a retreat of the cold halocline in the Eurasian Basin was also noted (Steele and Boyd, 1998). The halocline (Fig. 18) provides stratification between the Atlantic water layer and surface water thereby preventing or reducing the transfer of properties like heat or contaminants between deep and surface layers. The salinification of surface water in the Eurasian Basin noted by Steele and Boyd (1998), however, was not due to enhanced inflow from the Atlantic, which actually freshened slightly with the high AO/NAO index of the late 1980s, but rather to the diversion of river inflow at the margins of the Arctic Ocean.

Models (Fig. 19a, Dickson, 1999; Johnson and Polyakov, 2001; Maslowski et al., 1998) and geochemical measurements (Ekwurzel et al., 2001; Guay et al., 2001; Macdonald et al., 1999a, 2002b; Schlosser et al., 2002) show that with the high AO index of the late 1980s, river water entering the Laptev and Kara shelves was forced to the east rather than directly off the shelf and into the TPD. Under strong AO^{+} conditions, perhaps as much as 1000 km^3 $year^{-1}$ or more of runoff from the Lena, Ob and Yenesei rivers stopped entering the Eurasian Basin and entered, instead, the East Siberian Shelf and thence the Canadian Basin possibly to exit the Arctic Ocean via the Archipelago (Fig. 19b) (Morison et al., 2000). A consequence of this diversion was a reduction of stratification in the Eurasian Basin (Steele and Boyd, 1998) and an increase in stratification in the Canadian Basin (Macdonald et al., 1999a, 2002b). The drop in the AO index toward the end of the 1990s (Fig. 6) appears to have initiated a return to the former pathways for river water in the Eurasian Basin (Björk et al., 2002; Boyd et al., 2002) although it is not yet clear whether the ice cover will, likewise, respond to this change by returning to conditions similar to the 1960s and 1970s.

At the same time, toward the end of the 1980s, Atlantic surface water invaded the Makarov Basin, displacing water of Pacific origin from the top 200 m of the water column (McLaughlin et al., 1996): this represents an exceptionally rapid change of water source and properties for about 20% of the Arctic Ocean's area (Figs. 18 and 19). Although the total water inflow through Bering Strait (~0.8 Sv) does not appear to exhibit any AO-related modulation, there has been a general decline in the flow by about 15% since the early 1940s (Coachman and Aagaard, 1988; Roach et al., 1995) and the water may also have freshened due to runoff and precipitation in the Bering Sea (Weingartner, personal communication). The Pacific inflow is not uniform across Bering Strait including currents along the North American side (the Alaskan Coastal Current) and the Asian side (Anadyr

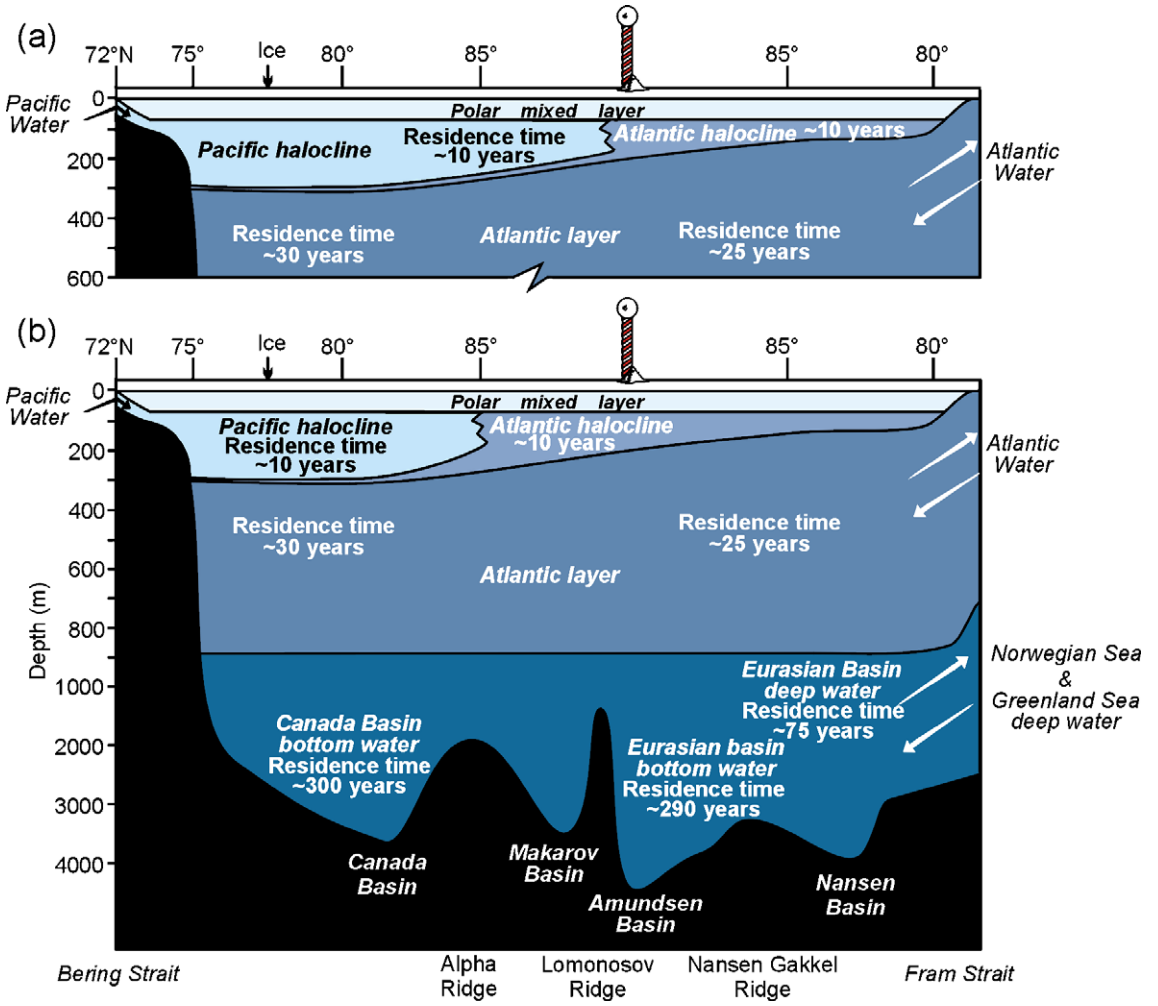

Fig. 18. The stratification of the Arctic Ocean showing the polar mixed layer, the Pacific and Atlantic domains of influence and the haloclines. The arrow at top center shows the displacement of the Atlantic Pacific front during the high Arctic Oscillation index of the early 1990s.

Current) each of which has very different geochemical properties and, very likely, different contaminant burdens (see for example, Li et al. (2002, 2004)). Once the water from these currents arrives in the Chukchi Sea, the AO may then provide a large-scale forcing that affects where the water then goes (Steele et al., 2004). Measurements made during SHEBA (Macdonald et al., 2001) imply that the inflowing waters from the Pacific Ocean almost certainly carry contaminant loadings that can be distinguished from surface water in the Canada Basin suggesting that AO modulation can affect the distribution of contaminants in the western Arctic. Regrettably, there are almost no data that can be used to infer contaminant links between the Pacific and Arctic Oceans.

Within the Arctic Ocean, the Atlantic layer has been shown to respond significantly to the state of the AO. Repeat hydrographic surveys of Arctic basins, commencing in 1987 (Aagaard et al., 1996; Anderson et al., 1989; Carmack et al., 1995; McLaughlin et al., 1996, 2002, 2004; Morison et al., 1998; Quadfasel et al., 1991; Swift et al., 1997), have revealed an Arctic Ocean in transition. The timing of the start of that transition in the late 1980s implicates the Arctic Oscillation (or North Atlantic Oscillation) as a major source of forcing that has altered

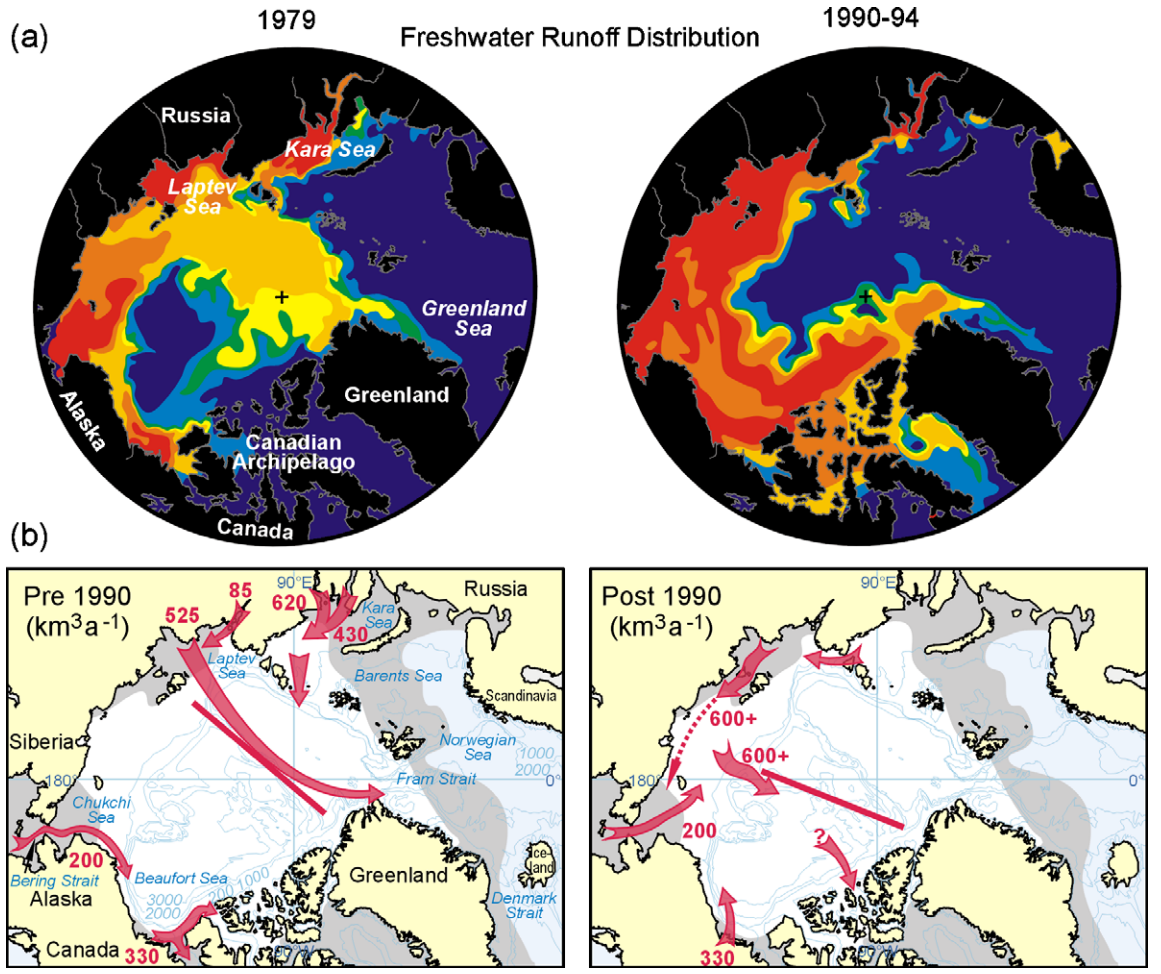

Fig. 19. (a) The change in river inflow pathways between low AO^- index (1979) and high AO^+ index year (1990–1994) (based on model results from W. Maslowski reproduced in Dickson, 1999) and (b) a schematic diagram showing the amounts and changes in pathways for river inflow to the Arctic Ocean under AO^- conditions (blue) and AO^+ conditions (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

connections between the Atlantic and the Arctic Oceans and changed the distribution of Atlantic water within the Arctic both in the surface layer, discussed above, and in the deeper Atlantic Layer water (Dickson, 1999; Macdonald, 1996). Ironically, some of the clearest evidence for these changes is seen in contaminant time series—in particular, the artificial radionuclides released from European sources to the eastern North Atlantic (Carmack et al., 1997; Smith et al., 1998).

A major change, starting in about 1989, was an intensification of flow from the Atlantic into the Arctic through Fram Strait and the Barents Sea in response to the shift to strong AO^+ or NAO^+ conditions (Fig. 20—for detailed reviews see Dickson et al., 2000; Morison et al., 2000; Serreze et al., 2000). The winds associated with AO^+ conditions (Fig. 8a,b) increased the rate of northward transport of surface water in the Norwegian Sea and produced warmer air temperatures which, together with the shorter transit times, contributed to warming by about 2.3 °C of the Atlantic water entering the Arctic (Swift et al., 1997). The Atlantic water also exhibited slightly decreased salinity (by 0.03–0.05) probably reflecting increased precipitation in the Nordic Seas during NAO^+ conditions (Fig. 9b).

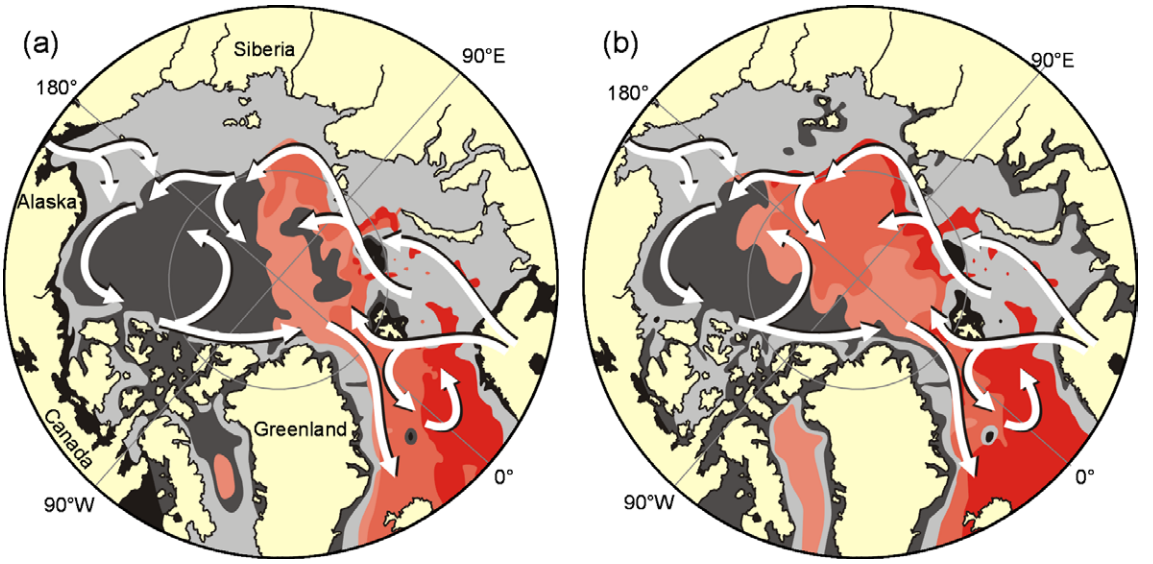

Fig. 20. The change in Atlantic water inflow to the Arctic and distribution within the Arctic produced by the exceptionally strong shift to AO^{+}/NAO^{+} in about 1989. The distribution of the Atlantic layer (see Fig. 18) is based on Hodges (2000), Maslowski et al. (2000), McLaughlin et al. (1996), and Morison et al. (1998, 2000). The Atlantic layer boundary currents, which are relatively fast, transport properties along basin margins at about 1–5 cm s^{-1} (300–1600 km $year^{-1}$; Woodgate et al., 2001).

Within the Arctic Ocean, the changes in the distribution and composition of the Atlantic layer water were spectacular when set against traditional perception of a quiet, steady-state ocean (see Figs. 18 and 20 for this layer's disposition in the water column). The front between Atlantic water and Pacific water, traditionally located over the Lomonsov Ridge, was forced over to the Alpha–Mendeleev Ridge (Fig. 18 and see McLaughlin et al., 1996; Morison et al., 2000). At the same time, the inflowing water could be detected in the Atlantic layer by an approximately 1.5 °C temperature rise above the climatological norm (Carmack et al., 1995). The changes in volume and composition of Atlantic water entering the Arctic Ocean through Fram Strait continue to cascade through the Arctic basins, first as change in properties along the boundaries (McLaughlin et al., 2002; Newton and Sotirin, 1997), then as change propagated into the basin interiors along surfaces of constant density (Carmack et al., 1997) (Fig. 18). Woodgate et al. (2001) estimated that in 1995–1996, the boundary flow over the southern margin of the Eurasian Basin was transporting 5±1 Sv (1 Sv=32,000 km^3 $year^{-1}$) at about 1–5 cm s^{-1} (300–1600 km $year^{-1}$). When water in the boundary current reached the Lomonosov Ridge, the flow split with about half entering the Canada Basin along its margin and half returning towards Fram Strait along the Lomonosov Ridge. The high NAO index of the late 1980s (Fig. 7a) also strengthened and warmed the inflowing Barents Sea Branch of Atlantic water, perhaps by as much as 25% since 1970 (Dickson et al., 2000), which probably led to a parallel warming and salinification of the Barents Sea (Zhang et al., 2000).

3.6. Adjacent polar seas and regions

3.6.1. The Canadian Archipelago

The Canadian Archipelago provides one of the important outlets for Arctic Ocean surface water (Fig. 1). Therefore, changes in Arctic Ocean surface water contaminant burdens or changes in the source of water flowing out through the Archipelago both have the potential to alter contaminant concentrations within the Archipelago's channels (Hargrave et al., 1997) and, indeed, much farther afield (Shen et al., 2004). There are few data with which to evaluate how seawater within the Archipelago channels responds to the Arctic Oscillation. However, changes

in distribution of surface water properties (Figs. 18 and 19) and ice drift trajectories (Fig. 14) in the Arctic Ocean itself together with non-uniform spatial distribution of properties including river water and contaminants (e.g., see Carmack et al., 1997; Guay and Falkner, 1997; Li et al., 2002; Macdonald et al., 1997) should alert us to the potential for upstream basin changes to be recorded as variable contaminant loadings in water flowing through the Archipelago. Furthermore, bowhead whale remains and driftwood on Archipelago shores suggest that ice-drift trajectories and/or ice cover have both varied greatly over time (Dyke et al., 1996b, 1997; Dyke and Savelle, 2000, 2001) implying that the Archipelago is vulnerable to rapid and dramatic change. Past changes like these in the ice climate have almost certainly affected humans living on the Arctic's margins (McGhee, 1996; Vibe, 1967).

3.6.2. Hudson Bay

Hudson Bay is a large, shallow semi-enclosed sea strongly influenced by seasonal runoff. The annual discharge (710 km^3 $year^{-1}$) is equivalent to a freshwater yield of about 65 cm (Prinsenberg, 1991). Presently, this sea exhibits a complete cryogenic cycle with summer (August–September) being ice free and winter fully ice covered. Climate models suggest that a doubling of CO_2 may lead to the virtual disappearance of ice from Hudson Bay thereby raising winter air temperatures and leading to the thawing of permafrost in adjacent land areas (Gough and Wolfe, 2001). These same models predict that the complete loss of ice will be preceded by years exhibiting earlier breakup and later freeze up. According to Stirling et al. (1999), some of these projected changes may already be occurring, putting considerable stress on the western Hudson Bay polar bear population.

The hydrological cycle of Hudson Bay has been strongly altered through immense damming projects in the drainage basin which have led to an increase in winter runoff to Hudson Bay of over 50% (Prinsenberg, 1991). Not only do such changes have impacts on stratification and hence nutrient cycling in this sea (Ingram et al., 1996), but also newly flooded reservoirs are well known for their secondary effect of releasing mercury to downstream aquatic environments (Bodaly and Johnston, 1992).

Hudson Bay, which is at the Arctic's southern margin, is clearly in the vanguard of cryospheric change and is, therefore, a region where it will be vital to collect time series. According to Fig. 3a, Hudson Bay lies on a divide between warming and cooling. Regional temperature maps and other evidence (Gilchrist and Robertson, 2000; Skinner et al., 1998) confirm that between 1950 and 1990, the western side has warmed at about the same rate as the eastern side has cooled. In agreement with this observation, bears on the eastern side of Hudson Bay do not show the same pattern of weight loss as the bears on the western side (Stirling, personal communication), further emphasizing the importance of this region as a laboratory to study detailed consequences of change by contrast.

3.6.3. Baffin Bay

Baffin Bay, Davis Strait and the Labrador Sea occupy a unique position in that they may not only receive contaminants from ice and water that exit the Arctic Ocean in the East Greenland Current but they may also receive contaminants from water and ice passing through the Archipelago (Fig. 1). Change, therefore, can be produced by variation of contaminant composition within either of these sources or by altering the relative strength of the sources and the strength of direct exchange with the atmosphere (see, for example, Shen et al., 2004). Furthermore, decadal-scale modulation very likely differs for the various sources, with the Arctic Oscillation perhaps influencing Archipelago throughflow or Fram Strait outflow whereas the ice cover in Baffin Bay is more closely associated with the Southern Oscillation (Newell, 1996). In agreement with spatial temperature patterns (Fig. 3a), whereas ice season has been getting shorter within the Arctic Ocean and its marginal seas, Davis Strait and the Labrador Sea have recently exhibited an increase in the length of ice season (Parkinson, 1992).

Long-period cycles in the ice climate of this region (50 years or more) have had dire consequences for both terrestrial and marine biological populations—including humans (Vibe, 1967). Like Hudson Bay, this region appears to be an important one to study in the context of contaminants (Fisk et al., 2001a; Shen et al., 2004), biogeographical variation (Johns et al., 2001) and the impact of change on humans (Woollett et al., 2000).

3.7. Lake and river ice

Arctic lakes and rivers likely provide sensitive sentinels of climate change in their freeze, melt and hydrological cycles (Vörösmarty et al., 2001). Whereas there appear to be no studies showing a relationship between freshwater ice cover and the AO, significant trends have been observed in ice cover during the past 150 to 286 years (Magnuson et al., 2000; Semiletov et al., 2000). There has been a mean delay of 5.8 days per century for freeze-up and a corresponding 6.5 days per century advance in breakup during the period between 1846 and 1995. This change in the freeze/melt cycle implies increasing temperatures of about 1.2 °C per century.

Most arctic lakes receive their contaminant burdens atmospherically with the catchment acting as a receptor through snow fall in winter, and a conveyor through runoff in spring. From a very limited set of studies, it appears that arctic lakes presently retain only a small fraction of the POPs input because the main runoff pulse, which precedes lake turnover and peak primary production, simply traverses the lake surface under the ice (Diamond et al., 2003; Helm et al., 2002; Macdonald et al., 2000a). For reasons that are as yet not clear, arctic lakes appear better able to capture Hg entering the lakes, possibly retaining as much as 50% of the Hg entering the lake (Diamond et al., 2003). With lakes exhibiting more temperate characteristics, the coupling of runoff with lake mixing and primary production will change, probably toward a greater capture of the inflowing contaminant burden. More open water together with enhanced vertical fluxes of organic carbon would strengthen surface water drawdown of POPs, hence supporting greater flux of POPs from air into lakes, in much the same way as described for the ocean (e.g., see Dachs et al., 2002).

Snow and ice offer special climate-sensitive mechanisms for concentrating and transporting contaminants (Macdonald et al., 2002a,c) because they provide surfaces for contaminant exchange (Hoff et al., 1995; Wania and Halsall, 2003). We can confidently state that contaminant delivery to watersheds and subsequent transfer into rivers and lakes will be acutely sensitive to the manner in which phase changes occur (melting/freezing) and the timing. Phase changes have the potential to produce diverging pathways—for example when snow melts, snow-bound contaminants may enter the water, exchange back into the atmosphere or attach to soil particles depending on exactly how melting occurs and how the water moves after it is melted. Quantitative measurements of contaminant–ice interactions in a variety of arctic settings are urgently required to determine the controls on these pathways which cannot be inferred from standard hydrological measurements.

3.8. Permafrost

Permafrost underlies about 25% of the land in the Northern Hemisphere including large areas of northern Canada (Fig. 21, Zhang et al., 1999) and under sediments of the continental shelves (not shown). Especially vulnerable to change are regions of discontinuous permafrost which, again, include large parts of northern Canada. The IPCC (2002) suggests that permafrost area could be reduced by 12–22% by the year 2100 with perhaps as much as half of the present-day Canadian permafrost disappearing.

The active soil layer in regions of permafrost is typically limited to the top 1 m where almost all of the biological processes occur. The loss of permafrost, therefore, alters these biological processes including the kind of vegetation that can grow, and changes the way soil interacts with the hydrological cycle (Osterkamp et al., 2000; Vörösmarty et al., 2001) both of which have consequences for contaminant transport. In particular, thawing frozen ground releases sediment, nutrients, and organic carbon which then enter ground water, rivers and lakes to impact biological cycles (see, for example, the studies done in the Mackenzie Basin (Cohen, 1997a) and elsewhere (Schindler, 1997; Schindler et al., 1996, 1997)). During permafrost destruction a shift toward dendritic drainage patterns (e.g., see McNamara et al., 1999) will allow a more complete transport of contaminants into ponds and lakes and possibly also re-mobilize contaminants archived in tundra soils. Simultaneously, reduced pond areas due to drainage channels in permafrost (Hinzman et al., submitted for publication) will likely enhance contaminant transport into remaining surface water.

The observed thawing trends in Alaska and Russia but not in northeastern Canada appear to match the

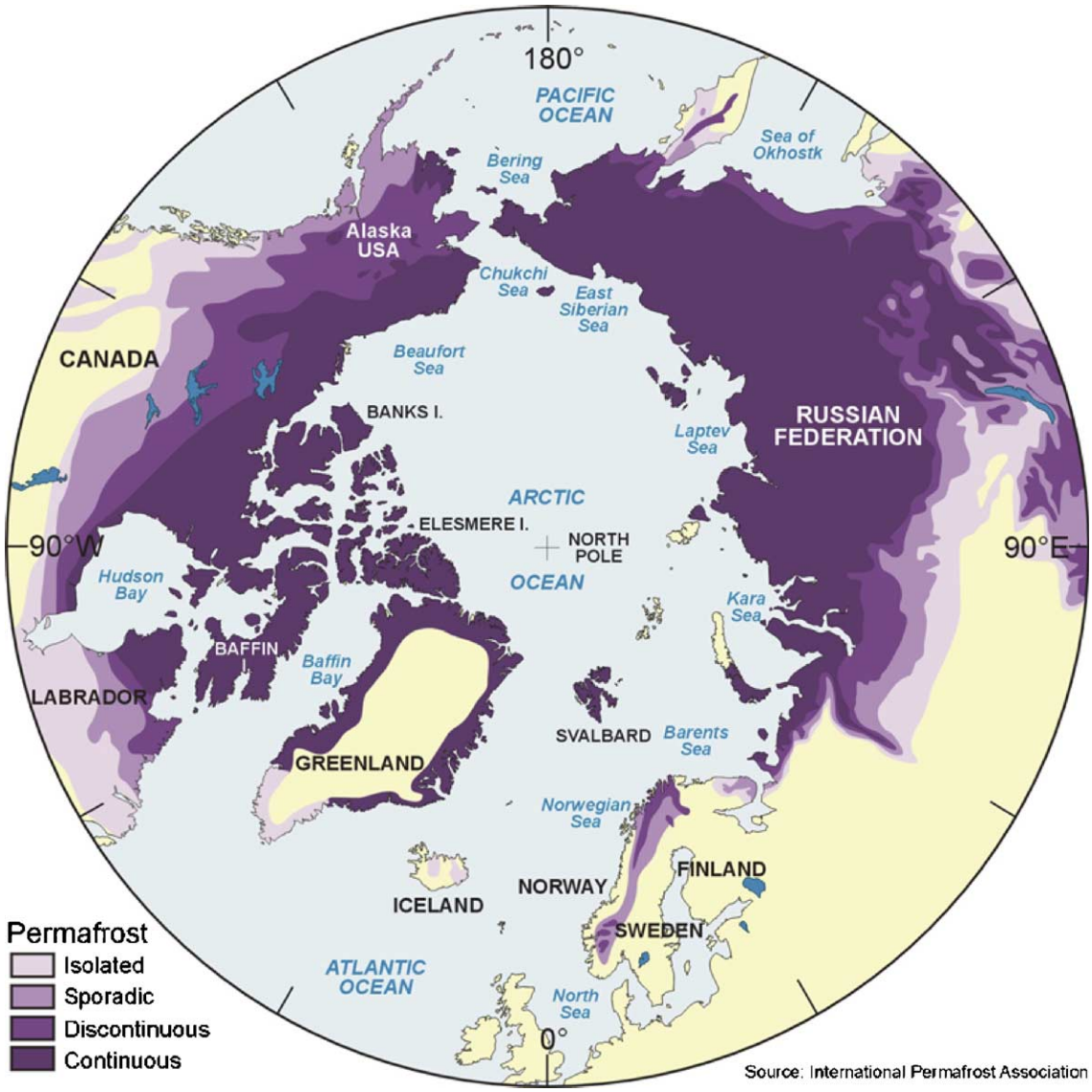

Fig. 21. The distribution of permafrost in northern landmasses. Discontinuous permafrost, found in large areas of Canada, Alaska and Russia, are particularly vulnerable to destruction with projected warming (source AMAP, 2002).

observed trends in SAT (see Fig. 3a and Rigor et al., 2000). Accelerated permafrost degradation during the 1990s can probably be ascribed at least partly to the AO (Morison et al., 2000) with, for example, the advection of warm air into the Russian Arctic during high AO index contributing to thawing in that region.

In addition to the biogeochemical pathway changes that will accompany permafrost degradation, there will also be the widespread problem of re-mobilization of contaminants (see, for example, conference proceedings dedicated to the issue of contaminants in frozen ground; Anonymous, 2001a,b). Historical disposal of waste substances has occurred in the form of sewage lagoons, dump sites at DEW line installations, solid waste dumps in small Arctic communities, mine tailings, and oil drilling sumps. A large component of the containment strategy for these sites is the permafrost itself. With permafrost degradation,

landfills can become washed directly into rivers or the ocean, or runoff can be leached into local groundwater. In locations like river deltas and coastal plains, low relief may provide a shortcut between such waste sites and drinking water.

3.9. Glacial ice

Most Arctic glaciers have experienced net loss in ice mass over the past few decades (Dowdeswell et al., 1997). The Greenland ice mass appears presently (1994–1999) to be decreasing, predominantly at lower elevations, at a rate of about 51 km^3/year (Krabill et al., 2000). Data also point clearly to loss of ice mass for small glaciers in the Arctic during the interval between 1961 and 1993 (Arendt et al., 2002; Dyurgerov and Meier, 1997; Serreze et al., 2000). Since about 1960, glacial melt back in the Canadian Arctic alone (Fig. 22) is estimated at over 800 km^3—about half of the melt back estimated for the whole Arctic (Dyurgerov and Meier, 1997). In conformance with the high AO index of the early 1990s, the loss of glacial ice mass in the Archipelago was exceptionally strong in the early 1990s amounting to 390 km^3 (Fig. 22).

Glaciers may act as long-term archives, sequestering and preserving airborne contaminants during peak emission years (1950–1970) later to release them during periods of melt back (Blais et al., 1998).

4. Biological responses to climate change

An exhaustive projection of the biological consequences of the kinds of physical changes projected for the Arctic is not feasible at this time, nor is it warranted for the purpose of identifying how biological changes might effect major change in contaminant pathways. Here, we highlight ecosystem changes that appear to have a strong potential to alter the exposure of arctic biota to contaminants or to alter their resilience to that exposure. We are confident that the kinds of changes discussed below have taken place, or will take place in the Arctic, but we are much less confident of their probable scope and timing. Our primary intent, therefore, is to provide examples of processes that ought to be included explicitly in models and to help focus future attention on biological connections of significance to contaminants.

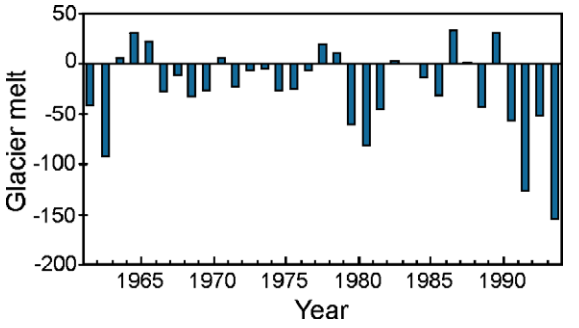

Fig. 22. The loss of glacial ice mass between 1961 and 1993 based on data compiled by Serreze et al. (2000) and Dyurgerov and Meier (1997).

Aquatic foodwebs in the Arctic exhibit endemic contamination from biomagnifying chemicals in contrast to terrestrial foodwebs which are found to be among the world's cleanest (de Wit et al., AMAP, 2005; DeMarch et al., 1998). Therefore, apex feeders that adapt to change by switching between land-based and aquatic foodwebs have a particularly large potential to change their exposure to contaminants like organochlorines and mercury. Humans probably provide the best example of such flexibility (e.g., see Vibe, 1967) but other animals (e.g., arctic foxes, grizzly bears and some birds) may also adjust diet to opportunity. Plasticity in diet, which might be considered an adaptive advantage for some species, implies large foodweb-mediated change in exposure to contaminants.

4.1. Terrestrial systems

Here, we use the term 'terrestrial systems' to refer to forests, grasslands, tundra, agricultural crops and soils but not freshwater aquatic systems. Surface–air exchange between airborne contaminants and terrestrial systems is important in the overall fate and long range transport of chemicals, especially for the semi-volatile chemicals which are split between the gaseous and condensed states. As a result of their high organic content, terrestrial components (e.g. soils, forests, grasslands) act as reservoirs for many POPs (Simonich and Hites, 1994) with PCBs, DDT, HCH and chlorobenzenes figuring prominently (de Wit et al., AMAP, 2005). Air–surface exchange of POPs into these phases is a dynamic process that controls air burdens of chemicals. Thus any change in the extent

of vegetative cover associated with global warming will have implications for contaminant fate and transport. Wania and McLachlan (2001) have shown that forests have a unique ability to mitigate atmospheric concentrations of OCs by "pumping" chemicals from the atmosphere into foliage and thence, through leaf litter, to a long-term reservoir in forest soil. This process is likely to be most important for OC compounds with log K_{OA} values of ~9–10 and log K_{AW} values ~−2 to −3 (where K_{OA} and K_{AW} are octanol–air and air–water partition coefficients—see Wania, 2001, 2003). Because these key properties are strongly temperature-dependent (discussed in greater detail under Section 6.3.4) even a small warming or cooling may dramatically alter the distribution of a contaminant between phases. Terrestrial vegetation also has an indirect impact on contaminants in its ability to alter snow accumulation and soil temperature (Sturm et al., 2001).

Arctic terrestrial populations have provided some of the clearest examples of large temporal cycles (Krebs et al., 2001; Predavec et al., 2001) and it is against this natural background that the effects of global change will have to be evaluated. Warmer winter temperatures promote the growth of woody shrubs and encourage the northward migration of the tree line (MacDonald et al., 1993; Serreze et al., 2000; Vörösmarty et al., 2001). Although the advance of the tree line might be expected to effect change very slowly in time scales measured in centuries (estimated at 100 km per °C warming (IPCC, 2002)), the particular sensitivity of tundra to water-table fluctuations and permafrost melt could produce widespread alteration in ground cover more rapidly with, for example, the replacement of tundra by vascular plants within decades (Gorham, 1991; Rouse et al., 1997; Weller and Lang, 1999). Gradual climate change can affect species distribution, population diet, abundance, morphology, behaviour and community structure (Easterling et al., 2000; Predavec et al., 2001). Although there appears to be no compelling evidence of recent large change in the Arctic's tundra, ecosystem models suggest that tundra may decrease to one third its present size (Everett and Fitzharris, 1998).

Warmer summer temperatures and drier continental interiors are likely to promote forest fires which will be accompanied by direct emissions of PAHs, PCDD/Fs and other POPs produced by combustion (see for example, Gribble, 1994; Yunker et al., 2002). Forest fires will also damage terrestrial soils leading to erosion and an increased release of organic carbon thence to impinge on aquatic systems.

4.2. Aquatic systems

4.2.1. Lakes, rivers and estuaries

Changes in snow and ice cover and in the hydrological cycle will alter the light and nutrient climate of freshwater systems. These changes together with loss of permafrost, which will enhance the supply of nutrients and particulates to lakes, will increase aquatic productivity and particle flux (Douglas et al., 1994; McDonald et al., 1996; Schindler, 1997). Spring bloom will probably occur earlier in spring with early loss of ice cover, but hydrological processes in a lake's drainage basin will probably also advance. Increased summer temperatures will disadvantage fish like trout and grayling whereas winter temperature increase may enhance microbial decomposition. Shifts in the seasonal light/temperature cycle may also advantage or disadvantage species lower in the food web including insects, phyto- and zooplankton. Change in water level will have obvious effects on important fish stocks especially species dependent on small refugia to over-winter (Hammar, 1989) or species dependant on freshwater coastal corridors for their life cycle (the arctic cisco (*Coregonus autumnalis*) provides a relevant Canadian example of the latter (see, Carmack and Macdonald, 2002; Gallaway et al., 1983)). Warming and loss of nearshore or estuarine ice may accordingly eliminate indigenous fish to be replaced by anadromous fish from the Pacific Ocean (see Babaluk et al., 2000). With warming, we can expect widely distributed shifts in zoogeographic distributions that have the potential to impact every step in the freshwater food chain, but prediction will probably founder on "counter-intuitive" surprises (Schindler, 1997).

4.2.2. The ocean

Those who harvest the sea have long recognized the effect of ice on arctic marine ecosystems (Bockstoce, 1986; McGhee, 1996; Scoresby, 1969; Vibe, 1967). Change in ice climate, therefore, has a large potential to modify marine ecosystems, either

through a *bottom-up* reorganization of the food web by altering the nutrient or light cycle, or a *top-down* reorganization by altering critical habitat for higher trophic levels (see for example Parsons, 1992). Any reorganization that changes the number of trophic levels in the food web or alters the flow of carbon between, for example, pelagic and benthic food webs would have particular significance for contaminants that biomagnify like mercury and the organochlorines: the complexity of the interaction between ice and aquatic ecosystems provides much scope for surprises in contaminant pathways (Fig. 23). Arctic and sub-arctic marine ecosystems are also altered by ocean climate changes such as regime shifts involving the displacement of water masses and associated populations or temperature change or other reasons (Dippner and Ottersen, 2001; Feder et al., 2003; Hare and Mantua, 2000; Helland-Hansen and Nansen, 1909; Hunt et al., 1999; Loeng, 2001; Saar, 2000; Sakshaug et al., 1991, 1994).

4.2.2.1. Bottom up biological effects. There are far too many examples of how ice climate variation can affect ecosystem structure to list them all (see for example Sakshaug and Slagstad, 1992; Tynan and DeMaster, 1997) and it is not likely that we are aware of all changes that have occurred in arctic systems simply because we have not had a broad, long-term observational network. The thickness and distribution of ice can influence the amount of organic carbon produced, the types of algae that produce it, and connections between the algal production and communities in the water column or sediments (Niebauer and Alexander, 1985). Ice controls wind mixing and light penetration especially when covered with snow, and it may support

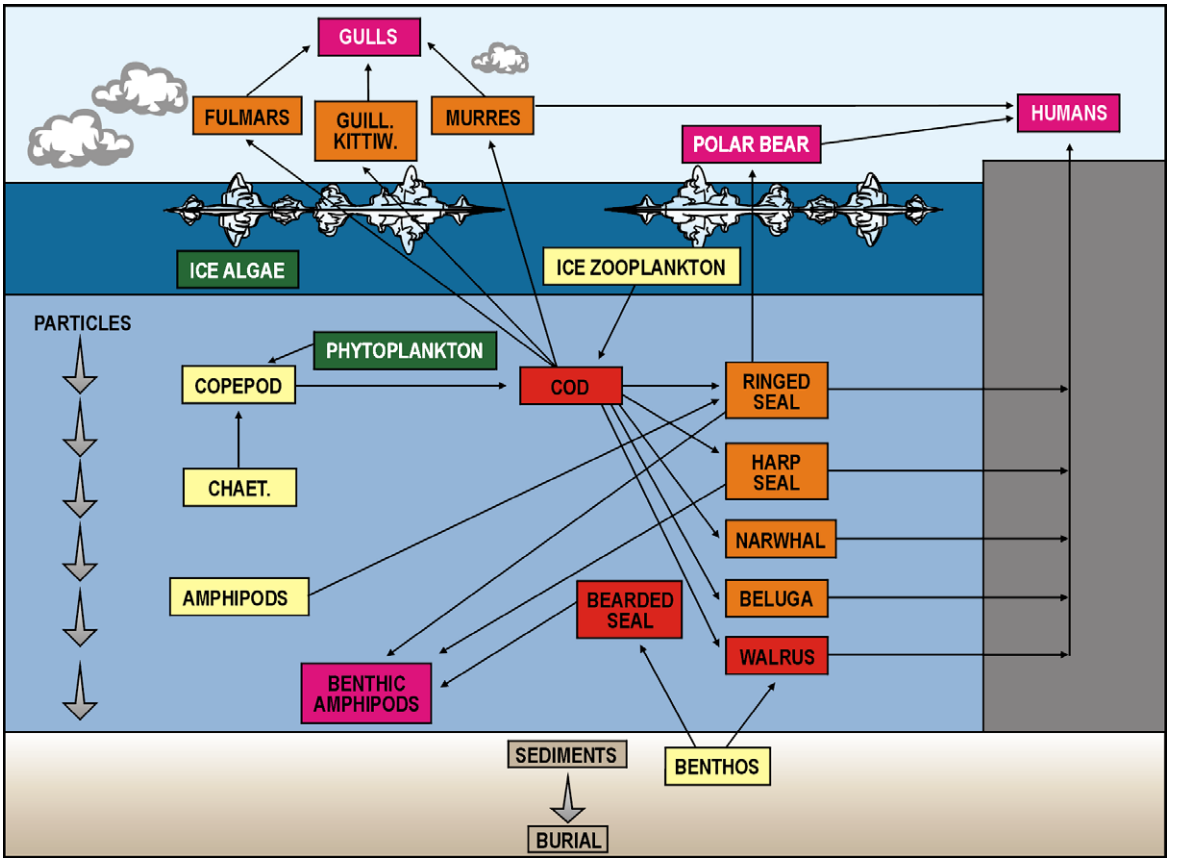

Fig. 23. A simplified schematic diagram showing the marine food web (based on Welch et al., 1992).

upwelling at the ice edge but suppress it beneath the ice. Through its annual cycle, ice formation decreases stratification in winter but increases it when the ice melts in spring. These physical factors impact the nutrient supply to surface water, the light climate, and the water column stability which together control primary production. Furthermore, mats of algae that grow on the bottom of the ice support an epontic food web that feeds polar cod (*Boreogadus saida*), ringed seals (*Phoca hispida*) and polar bears (*Ursus maritimus*) or, alternatively, by being shed from melting ice in spring, support a benthic food web that feeds molluscs, walrus (*Odobenus rosmarus*), bearded seals (*Erignathus barbatus*) and king eiders (*Somateria spectabilis*). Similarly, primary production in the water column may be partially grazed to support a pelagic food web, or descend ungrazed and, together with fecal pellets and zooplankton carcasses, feed the benthos (Grebmeier and Dunton, 2000). The bifurcation between pelagic and benthic food webs is very much influenced by the distribution of ice and its impact on nutrient and light climate.

Shifts in benthic species distribution due to temperature, carbon flux or other climate-related change have the potential to alter completely the coupling between sediments and bottom water. In one well-documented example from a temperate region, the invasion of Echiura (*Listroiolobus pelodes*) into coastal benthic communities off California, for as yet unknown reasons, resulted in aerated and biomixed sediments that reduced the evidence of wastewater impacts regionally (Stull et al., 1986).

The projected loss of ice for the Arctic Ocean, particularly over the shelves, intuitively should increase primary production in the marginal seas through enhanced mixing, penetration by light and upwelling at the shelf edge. In other words, arctic shelves would begin to look more 'temperate.' Greater new production implies greater particle flux and greater secondary production, but the complexity of marine ecosystems should alert us to possible surprises. Massive blooms of jellyfish were observed in the Bering Sea during in the 1990s (Brodeur et al., 1999; Hunt et al., 1999) and their emergence was ascribed to sea-surface temperature increase and loss of ice cover—the same two key changes poised over the Arctic Ocean.

Parsons (1979) has drawn attention to the fundamental ecological differences between western seaboards, where coastal water exhibits divergence and upwelling, and eastern seaboards which are convergent. The former have been of greater commercial interest to humans but they are also characterized by jellyfish (Parsons, 1979). The AO does not cause reversal of large-scale wind circulation but it does produce more divergent Arctic Ocean margins under AO^-/anti-cyclonic conditions and less divergent margins under AO^+/cyclonic conditions. The inherently noisy events of coastal upwelling and downwelling could then act together with the AO in a form of 'stochastic resonance' (Rahmsdorf and Alley, 2002) to enhance upwelling during AO^- conditions which might then have the capacity to produce large-scale modal shifts in shelf ecosystems and their commercial significance. Alternatively, the withdrawal of the ice boundary into the interior ocean, as seen recently (Fig. 13), may strengthen the coupling between winds and ocean surface thereby enhancing shelf-edge upwelling and producing more productive arctic shelves (Carmack and Chapman, 2003). Changes in ocean climate like those associated with the AO/NAO have long been known to impact fisheries in sub-polar seas either directly through water property changes (T, S) or indirectly through changes in the community structure (Hare and Mantua, 2000; Klyashtorin, 1998; Marteinsdottir and Thorarinsson, 1998).

A dramatic example of large-scale, bottom up biological change was witnessed during the SHEBA drift across the Beaufort and Chukchi Seas in 1997–1998 (Melnikov et al., 2002). Compared to Soviet observations from drifting stations that passed over the same region 20 years earlier, there was a marked decrease in large diatoms in the water column and microfauna within the ice. The freshening and strong stratification of the surface water, due to river discharge diverted into the basin under the high AO conditions of the early 1990s (described above), reduced the supply of nutrients from below, and produced species more typical of fresh water. Consequentially, there was a high proportion of recycled production and less new production implying a reduction in vertical flux of organic carbon. The loss of relatively large diatoms could reduce the size of herbivores, potentially inserting an extra 'small-carnivore' step at the bottom of the food web which

would increase the number of trophic levels. Biomagnification of organochlorines is often exponential (Fisk et al., 2001a) such that slightly higher concentrations at low trophic levels (e.g., copepods) can have a large impact on apex feeders. Stratification, which is altered at the basin scale under AO/NAO shifts, affects plankton composition and vertical flux dramatically as evident from studies in the Barents Sea (Wassmann, 2001). For example, Wassmann et al. (1990) showed that algal blooms by *Phaeocycstis* sp. along the Greenland coast and in the Barents Sea tend not to get grazed resulting in a large transfer of primary-produced organic carbon to the benthos. Climate change in the form either of loss of ice cover or increase in stratification has the potential to alter the quantity of available food and redistribute its flow between epontic, pelagic and benthic habitats.

The Bering Sea provides another outstanding example of recent change from the bottom up permeating an entire ecosystem. It is particularly regrettable that these observations of ecosystem change since the 1970s have not been matched by contaminant pathway studies considering the region's vulnerability to airborne contaminants from Asia (Bailey et al., 2000; Li et al., 2002). The changes in the Bering Sea may relate to a larger picture of change throughout the North Pacific, which had significant and widespread impacts on biota (Board, 2003; Hare and Mantua, 2000) and, in particular, to the switch in the Pacific Decadal Oscillation from cold to warm phase in the mid-1970s. The change in regime rapidly permeated the entire ecosystem of the Bering Sea altering fish community structure and seabird and mammal populations (Springer, 1998). More recently, blooms of small phytoplankton (*Emiliana huxlei*) were observed in 1997 and 1998 (Saar, 2000). These phytoplankton are smaller than the diatoms that typically bloom in the Bering Sea and they were, therefore, grazed by smaller copepods instead of larger euphausids. This, in turn, probably led to die-offs of the short-tailed shearwaters (*Puffinus tenuirostris*) that feed on the latter (Stockwell et al., 1999). Similarly, the alteration of primary production both in quantity and distribution probably decreased food availability for fish, whale, seal and walrus populations forcing die-offs, migration or redistribution throughout the foodweb (Botsford et al., 1997; Grebmeier and Cooper, 1995; Grebmeier and Dunton, 2000; Hare and Mantua, 2000; Rugh et al., 1999; Stabeno and Overland, 2001). Large as these ecosystem changes appear to have been, they may pale in comparison to the natural fluctuations that have occurred during the past two millennia (Finney et al., 2002). Clearly, the dramatic changes in the Bering Sea system could spill over into the Chukchi Sea, and the decline of Bering inflow by ~15% since the 1940s suggests a matched decline in new and advected production in the Chukchi Sea simply due to reduced nutrient and organic carbon supply.

In the Barents and Nordic Seas it has long been recognized that fish populations respond to climate variability (Helland-Hansen and Nansen, 1909). Indeed, the distribution of capelin (*Mallotus villosus*), the single most important food species for Arcto-Norwegian cod, is known to vary from year to year dependent on the inflow of Atlantic water (Sakshaug et al., 1994). Fluctuations in large- and regional-scale atmospheric pressure conditions affect winds and upper ocean currents, modify water temperature, alter drift patterns of fish larvae, and change availability of prey items. Mixing during summer alters the nutrient cycle and the coupling between primary production and benthos (Peinert et al., 2001; Wassmann, 2001).

4.2.2.2. Top-down biological effects. Ice-covered seas have a unique capacity for top-down trophic change. To understand and predict how the partial or complete loss of ice will impact the trophic structure requires a detailed understanding of how top predators take advantage of ice (Carmack and Macdonald, 2002; Lowry, 2000; Vibe, 1967). In an incisive review, Tynan and deMaster (1997) describe how whales, walrus, seals, bears and cod, are likely to be impacted by change in ice climate showing that their response to change depends on how 'plastic' their dependence on ice might be.

Change in the landfast ice may give the advantage to seals or to bears with the result that arctic cod (*Boreogadus saida*) would be subject to more, or less, predation, respectively. Walrus not only use *drifting ice* to haulout in winter because it provides better access to benthos but they also use terrestrial haulouts in ice-free periods, perhaps with detrimental energy costs (Lowry, 2000; Tynan and DeMaster, 1997). In contrast, eiders and other benthic-feeding birds prefer *open water* with a relatively shallow bottom (<50 m)

(Dickson and Gilchrist, 2002; Grebmeier et al., 1988; Suydam et al., 2000). Loss of ice (landfast or drifting) in critical regions or at critical times of the year, or movement of the ice edge to deeper water where benthos can no longer be accessed, therefore, can mean a substantial rearrangement of the top of the food web advantaging some animals, disadvantaging others and possibly causing wholesale migration (Dyke et al., 1996b, 1999, Dyke and Savelle, 2001; Fay, 1982; Lowry, 2000; Moore and Clarke, 1986; Tynan and DeMaster, 1997; Woollett et al., 2000). With benthos not readily available, walrus might turn to predation on seals thereby raising their trophic position–and their contaminant intake–considerably (Muir et al., 1999), or with absence of ice, killer-whale predation on bowhead whales might decimate their population leaving their prey (zooplankton) to feed something else. Even within a single species such as beluga (*Delphinapterus* leucas), differences between males and female foraging behaviour (Barber et al., 2001) could produce differing exposures to contaminants, and change in ice conditions could accordingly operate differently between sexes on that exposure. Early breakup in the Bering and Beaufort Seas during 1995–1998 probably led to the observed abandonment of seal pups in 1998 and the decline or starvation of walrus.

The Hudson Bay polar bear population provides perhaps one of the clearest warnings to us of the consequence of change. Polar bears rely on ringed seals for food and ringed seals prefer *landfast* or stable first-year ice for pupping (Finley et al., 1983; Stirling, 2002; Wiig et al., 1999). The loss of landfast ice in spring, the loss of food supply for seals, unusual rain events, or the inability of bears to access seals during the few critical weeks in spring when pupping occurs, means life or death and can produce large population shifts (Harwood et al., 2000; Smith and Harwood, 2001; Stirling et al., 1999; Stirling and Smith, in press). In Hudson Bay, bears probably accumulate most of their annual energy requirements during the few months of late spring prior to breakup when they can access older pre-weaning ringed-seal (*Phoca hispida*) pups or naïve post-weaning pups—exactly the period of time that has seen recent dramatic change. Furthermore, permafrost is a critical habitat for bears because they dig maternity dens in frozen peat, and this habitat is threatened by warming or increased incidence of fire initiated by more frequent lightning strikes. In Hudson Bay, at the southern limit of their population, polar bears presently appear in a very precarious position (Stirling and Derocher, 1993; Stirling et al., 1999). Near starvation for these bears, on one hand, will lead to re-mobilization of contaminants in stored fat, whereas shifting to the terrestrial foodweb (e.g., see Brook and Richardson, 2002) might reduce biomagnifying contaminant intake but possibly at a crippling energy cost.

Arctic cod is the most important forage fish in Arctic Ocean's food web (Fig. 23, Bradstreet et al., 1986; Tynan and DeMaster, 1997; Welch, 1995). The loss of ice, either in the marginal seas or, as projected by models, for the entire ocean (Fig. 5, Flato and Boer, 2001), would have a massive impact on the distribution and life history of arctic cod and, therefore, on seals, beluga and birds who depend heavily on them. One thing is clear: the ice edge is an especially critical habitat for cod and marine mammals and it is this region that is most vulnerable to change. Again we can turn to work in Hudson Bay to illustrate the potential effect on biological systems of recent change in the ice climate. Through an analysis of the diets of thick-billed murres (*Uria lomvia*) over the period 1981–2002, Gaston et al. (2003) have shown a regional shift in diet from arctic cod (*Boreogadus saida*), sculpins (*Triglops* and others) and benthic Zoarcidae, toward capelin (*Mallotus villosus*) and sandlance (*Ammodytes* spp.). This shift has implications not only for the marine pathways of lipophilic contaminants but also for the energetics involved in species like murres and seals whose dietary preference would normally be the cod which provide a better energy return. Increase in the energy burned relative to the energy stored has the likely consequence of increasing biomagnification (e.g., see Macdonald et al., 2002a) although we presently lack studies to inform us of how big a change might be involved. Whether the trophic changes observed in Hudson Bay can be described as top down or bottom up is not yet clear, but they signal the distinct possibility of large-scale change in one keystone species-arctic cod. Going further back in history, commercial whaling all but eliminated bowhead whales from Hudson Bay with as yet poorly understood top-down consequences for the foodweb. The

recent resurgence of bowhead whale populations may, therefore, once again alter the foodweb to the disadvantage of other species.

Finally, climate change can alter the routes and destinations of migratory species. For example, under the AO^+ conditions of the early 1990s, Pacific salmon began to enter arctic rivers (Babaluk et al., 2000). Similarly, bowhead whales and belugas range widely in search of food, and because the ice edge and lead systems are critical parts of their ranges, their migration varies in time and space commensurate with change in ice climate (Dyke et al., 1996b; Dyke and Savelle, 2001; McGhee, 1996). It is interesting to note that benthic feeding Gray whales (*Eschrichtius robustus*) of the North Pacific, who have likely suffered from starvation during the late 1990s due to reduction of prey (Moore et al., 2003), appear to have responded by expanding their foraging range into the Chukchi Sea. A consequence for these animals is that they are now accessing an arctic contaminant pathway via Chukchi benthos and 'withdrawing' these contaminants back into the Pacific Ocean. These sorts of migratory options, forced by climate variation, make interpretation of Gray whale contaminant burdens and trends difficult especially in view of the very few data we have (Tilbury et al., 2002). Nor are long migrations limited to whales. Harp seals of the Northwest Atlantic undergo 8000 km round trips to feed on arctic cod in Baffin Bay (Finley et al., 1990) and bird species migrate inordinately long distances often depending on critical areas along their migration pathways where they may ingest contaminants (see, for example, Braune et al., 1999; Savinova et al., 1995; Springer, 1998).

5. The effect of climate change on human activities

There are at least six very different ways in which climate change may lead to an alteration of contaminant pathways through a modification of human activities. Firstly, people on the margins of the Arctic Ocean will make dietary choices, as they have always done, based on the availability of country foods including terrestrial and marine animals (Krupnik, 2000; McGhee, 1996; Vibe, 1967). Secondly, a marginal sea that clears of ice for large portions of the year will encourage shipping, tourism, oil exploration and other industrial activities each of which brings with it associated contaminants. Furthermore, enhanced shipping increases the risk of introducing exotic species or diseases which then affect indigenous species. Thirdly, the encroachment of commercial fisheries (fish, shellfish and marine mammals) into the arctic could alter the food web structure in arctic seas (Bockstoce, 1986; Myers and Worm, 2003; Parsons, 1992; Pauly et al., 1998, 2001) and lakes (de Graff and Mychasiw, 1993; de Wit et al., AMAP, 2005). Fourthly, climate change may promote the spread of insect pests globally and some countries may choose to re-introduce or increase the use of banned or restricted pesticides especially those that are proven to be cheap and effective. Fifthly, climate change toward conditions suitable for domestic crops may encourage further expansion of agriculture or silviculture within the Arctic's drainage basin. Sixthly, the various changes listed above will likely contribute to demographic shifts and population increases in northern regions which will then lead to increased contaminant release locally (for example from burning, power and fuel consumption, use of industrial or agricultural products).

It is well known that the dietary composition (e.g., marine vs. terrestrial, fat vs. protein, old fish vs. young fish) of the human 'food basket' controls the amounts and kinds of contaminants ingested (AMAP, 1998; Kinloch et al., 1992; Mos et al., in press; Van Oostdam et al., 1999). Dietary change–and therefore change in contaminant exposure–can be forced by fluctuations in the populations of target species as discussed above (e.g., beluga, bowhead whales, walrus, seals, bears, birds, fish, caribou, muskox) or by changes in access to the species (early melt, permafrost degradation, open water, loss of multiyear ice, late freeze-up (see, for example, Fast and Berkes, 1998; Riedlinger, 2001; Vibe, 1967). Because the aquatic and terrestrial food webs are so differently impacted by contaminants, the balance between food items from each of these sources in the food basket will be a pivotal point of change in exposure to biomagnifying contaminants.

With marginal seas clear of ice for long periods of the year, it is inevitable that the Arctic will become a favoured shipping route between Europe, Asia and North America either via the Northern Sea Route (Russia) or the Canadian Archipelago. Shipping

brings with it specific, well-known contaminants like hydrocarbons and marine antifoulants (e.g., tributyl tin) and, potentially, exotic species in ballast water and these latter may have greater opportunity to expand their ranges simply due to climate change (Elliot, 2003). The major future concern, however, is likely to come from accelerated oil exploration and development on the arctic continental shelves of North America and Eurasia (Bakke et al., 1998).

Outside the Arctic, global warming and alteration of hydrological cycles will probably cause insects and other pests to flourish in some locations. Many of the Arctic's problematic pesticides (toxaphene, DDT, HCH) continue to be used in central America, Africa and Asia, particularly by developing countries, and it is these countries that will be forced to rely more heavily on pesticides in coming decades (Harner, 1997).

Within arctic drainage basins, warming may expand the area suitable for agriculture. Much of the southern portion of the Mackenzie Basin in North America is presently cultivated; under a global warming scenario this region is projected to contribute an additional 10 million hectares of land suitable for small grain crops (Cohen, 1997b), an area that might be further expanded with the development of new climate-tolerant crops. In Russia, most of the major arctic river basins contain agricultural land particularly within the river valleys and as far north 65°N (including the Sv. Dvina, Ob, Yenesey and Lena River basins). Agriculture brings with it industrial pesticides and other chemicals and, if pests also thrive in a warmer climate, farmers may resort to increased reliance on pesticides to protect crops.

6. The effects of climate change on contaminant pathways

We now consider how the changes described above will impact specific contaminant categories—heavy metals, radionuclides, organochlorines, and hydrocarbons. For each category we will generally discuss the significance of the recent shift to high AO index and then consider the more general, long-term changes facing us. To avoid repetition, we will make direct, unsupported statements for which arguments and citations have already been provided in preceding sections. We will emphasize connections in the contaminant pathways (Figs. 1 and 2) such as: (1) mobilization from global primary or secondary contaminant sources and/or a change in delivery pathways to arctic ecosystems; (2) entry into the base of the food web from water, snow pack, ice, soil, and runoff; (3) shifts in the relative importance of source of primary productivity in aquatic systems (ice vs. aquatic or coastal vs. deep ocean); (4) change in food web structure affecting the degree of biomagnification (bottom up effects); (5) change in the feeding ecology of key higher order consumers (top down effects) and (6) change in the age structure of higher trophic order populations where contaminant concentrations in tissue are age-dependent.

6.1. Heavy metals

6.1.1. Lead, cadmium, zinc

These three elements are commonly released to the atmosphere through high-temperature processes or, in even greater quantities, to water through runoff, municipal discharges and dumping (Pacyna et al., 1995). Of these metals, contaminant lead has had a unique, predominant source in leaded gasoline combustion. The strong atmospheric connection in winter between Eurasia and the high Arctic (Fig. 8a) has long been recognized in phenomena like arctic haze (Hileman, 1983) and brown snow deposition (Welch et al., 1991). Accordingly, much attention has been focused on the air as the means of transporting contaminant metals to the Arctic (Akeredolu et al., 1994; AMAP, 1998; Boutron et al., 1995; Gong and Barrie, 2003; Pacyna, 1995; Rosman et al., 1993; Sirois and Barrie, 1999; Sturges and Barrie, 1989). Back trajectories, models and stable lead isotope composition have established the sources of atmospheric metals to be primarily Eurasia (Norilsk), then western and eastern Europe and, finally, North America—each of which has the opportunity at particular times during the year to supply air masses to the Arctic (Fig. 8a,b). The shift between AO^- and AO^+ conditions alters mean wind fields thereby effecting change on the balance and timing of air movement from the three sources, but the connections remain intact. Air transport from eastern North America and Western Europe–both of which have abandoned lead in gasoline–strengthen under high AO index, especially in winter, due to the

intensification and extension northward of the Icelandic Low. Air mass trajectory changes, while probably shifting the pathway and rate of transport between temperate sources and arctic sinks, will not likely change the net delivery of airborne contaminants substantially. This hypothesis could be tested by running transport models (e.g., see Akeredolu et al., 1994) under AO^+ and AO^- conditions.

The greatest leverage for change with aerosol metals resides in the wet and dry removal processes within the Arctic for which our knowledge is not very complete. The Arctic is a poor trap for atmospheric contaminants, sequestering <10% of the emissions that pass through it (Akeredolu et al., 1994; Pacyna, 1995). Therefore, there is a great deal of scope to enhance the deposition of airborne contaminants to surfaces by altering location and intensity of precipitation as occurs under AO shifts (Fig. 9a,b) and/or by changing the balance between snow and rain fall (see, for example, Sherrell et al., 2000). Clearly, these changes can affect both terrestrial and marine systems. Under AO^+ conditions, the atmospheric corridor from eastern North America and Western Europe will become a much more efficient trap for particulates, raining them out in the Nordic Seas and in the southern portion of the Eurasian Basin. Particle scavenging will generally increase wherever higher precipitation prevails, such as over northern Europe and the Eurasian Basin in general. Contaminants deposited on the eastern side of the Nordic Seas will then enter the Barents Sea and the Eurasian Basin via ocean currents. Contaminants deposited to the west will be delivered back into the North Atlantic via the East Greenland Current. Given the above set of circumstances, it seems likely that under AO^+ conditions, metal-contaminated aerosols entering the Arctic near the prime meridian will be subject to enhanced scavenging en route. Larger areas of open water in this same region (Fig. 13) mean that scavenging will tend to place a greater proportion of these airborne contaminants directly into the surface ocean rather than on the sea ice. The decline of aerosol metal concentrations at Alert after about 1991, ascribed by Sirois and Barrie (1999) to the collapse of industry following the fragmenting of the former Soviet Union, could alternatively be explained by changes in wind and precipitation patterns at the end of the 1980s which could divert the transport upwind and/or remove particles to surface before they arrived at Alert (Figs. 8 and 9). Enhanced loadings to sea-ice surfaces under AO^+ conditions are most likely to occur within the southern Eurasian Basin and this ice would then be exported back into the Greenland Sea. Melting at the southern boundary of the ice would inject metal contaminants into the water column, either to sink to the bottom on particles (e.g., cryoconites), or travel south in the water, or recirculate back into the Arctic (Untersteiner, 1988). The connections at the entrance to the Arctic Ocean at Fram Strait are extremely complex, poorly understood even for the transport of water itself, and subject to climate variation.

The focus on the atmosphere as a contaminant metal pathway to the Arctic has to some degree diverted attention from the ocean. Sediment cores collected along the margins of the Eurasian and Canadian Basins (Fig. 24) suggest that a major route for contaminant lead to the Arctic Ocean has been the same ocean current that transports radionuclides northward from the European reprocessing plants (Gobeil et al., 2001a). The residence time of lead in surface water, which is relatively short (<5 years), is still long enough to permit transfer of contaminant lead from the North Atlantic and Nordic Seas into the Arctic (Gobeil et al., 2001a). We may expect under AO^+ conditions an even more vigorous transfer of metals from western Europe to the Arctic either via rainout in the air transport corridor to the northwest of Europe (Figs. 8 and 9) or via coastal discharges to the North or Baltic Seas. Lead comprises four stable isotopes, ^{204}Pb (1.48%), ^{206}Pb (23.6%), ^{207}Pb (22.6%) and ^{208}Pb (52.3%) with the composition varying among the world's geological reservoirs (Sangster et al., 2000). That variation has provided an incisive way to determine the sources of contaminant lead in global environmental media including arctic aerosols and ice (Rosman et al., 1993; Sturges and Barrie, 1989; Sturges et al., 1993). Accordingly, Gobeil et al. (2001a) were able to relate the contaminant lead accumulating in sediments along the Barents Sea margin to a western European source ($^{206}Pb/^{207}Pb \sim 1.14$) with ocean currents acting as the major transporting mechanism (Figs. 20 and 24). In contrast, the contaminant lead in sediments near the North Pole had a distinctly eastern Europe or Russian composition ($^{206}Pb/^{207}Pb \sim 1.18$) (Fig. 24). Based on

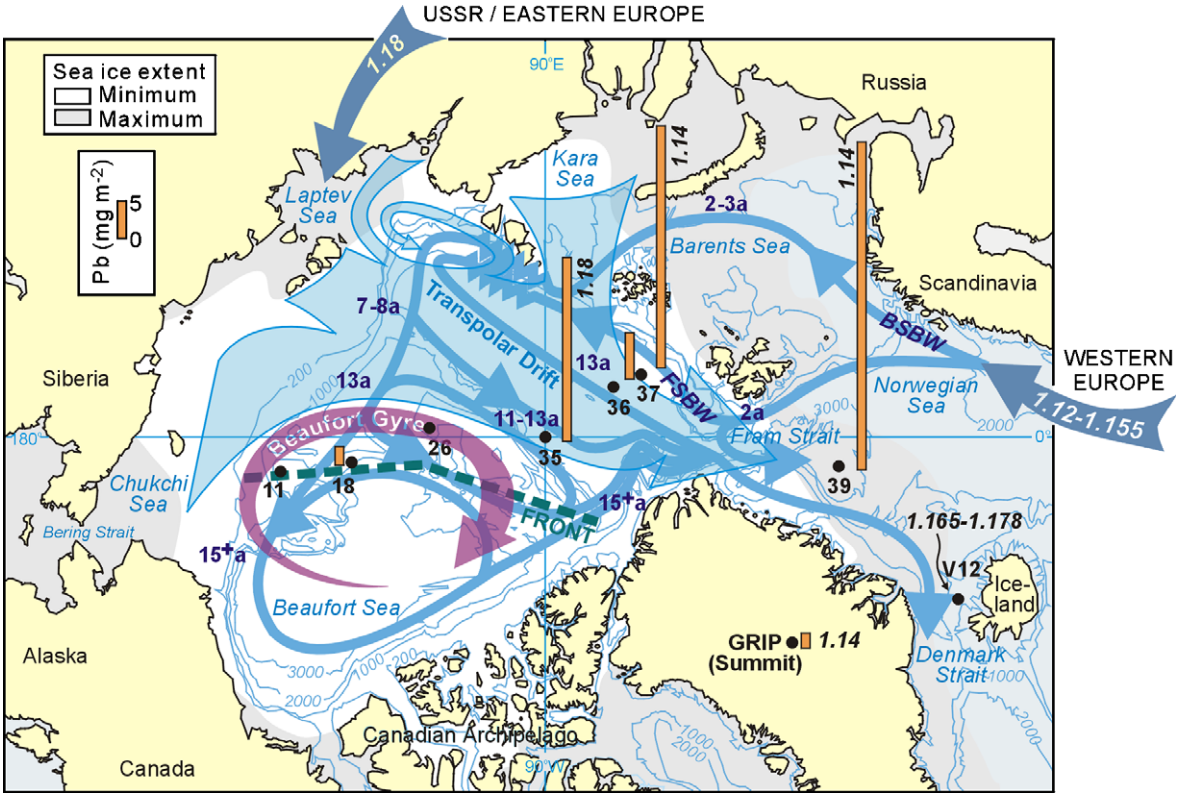

Fig. 24. The transport of lead into the Arctic Ocean following boundary currents shown by the contaminant lead inventory in sediment cores. Source of the lead (western Europe or Eurasia) is shown by the stable lead isotope composition (figure adapted from Gobeil et al., 2001a).

the composition of contaminant lead in North Pole sediments, these authors proposed a second transport route wherein contaminant lead enters the Arctic Ocean via the Laptev Sea, either in ice or, perhaps more likely, in water of the TPD (Gobeil et al., 2001a). The observation that contaminant lead was conspicuous in the Eurasian Basin margins but not in the Canadian Basin, led these authors to conclude that ocean and ice transport pathways during peak lead emission years (~last 60 years) must have been predominantly those associated with the AO^- conditions. Under AO^+ conditions, pathways for ice, ocean currents and runoff change dramatically (Figs. 8, 14 and 19) such that contaminant metals entering the Russian Shelves (atmospherically or by runoff) would be diverted to the east into the Canada Basin and toward the Archipelago (Gobeil et al., 2001a; Mysak, 2001). The pathways illuminated by lead contamination probably have relevance for other contaminants that attach to particles—for example the highly chlorinated PCBs, PBDEs or high molecular weight PAH.

Atmospheric aerosols of cadmium and zinc will to some degree behave like lead excepting the predominant source for the latter in leaded gasoline. In the Arctic, time series of aerosol composition at Alert (Sirois and Barrie, 1999) and records from ice cores (Boutron et al., 1991, 1995) and glacial snow (Sherrell et al., 2000) reveal contamination due to industrial activity in Asia, Europe and North America. Like lead, these elements are poorly captured within the Arctic (<15%, Akeredolu et al., 1994; Pacyna, 1995) and changes in precipitation patterns are likely to have the greatest potential to change metal delivery to surfaces.

Of these three heavy metals, cadmium provides the greatest risk through bioaccumulation and biomagnification especially into liver and kidney of marine and terrestrial mammals (Braune et al., 1999; Muir et al., 1999). Observed high concentrations of cadmium in

arctic biota, however, appear to be natural and not obviously related to human activities except, possibly, at locations close to sources like Norilsk (<100 km). In consequence, significant changes in cadmium exposure are likely to be delivered by changes in the natural biogeochemical cycle of cadmium rather than changes in contaminant cadmium pathways. An exception to this rule may occur locally when cadmium contamination is accompanied by or followed by system changes that alter cadmium biogeochemistry. Croteau et al. (2002) provide a clear example where reductions in cadmium loadings to a contaminated lake were accompanied by increases in pH with the consequence that organisms actually exhibited increasing cadmium uptake.

In the Arctic Ocean, natural cycles completely dominate cadmium fluxes and budgets (Macdonald et al., 2000a). Cadmium follows soft body parts in the marine biogeochemical cycle and exhibits a strong correlation with phosphate (Boyle, 1988; de Baar et al., 1994). The interaction between the biogeochemical cycle and circulation of the world ocean results in sub-surface water of the North Pacific containing naturally higher cadmium concentrations than those of the North Atlantic (by a factor of about 5—see Bruland and Franks, 1983). In turn, this makes the Pacific inflow through Bering Strait a dominant source of cadmium to surface waters of the Arctic Ocean (Macdonald et al., 2000a). Reduced Bering inflow since 1940 could entail a similar, 15%, reduction of the supply of cadmium to the Arctic Ocean from that source, depending on whether or not the reduced inflow pertains to nutrient- and Cd-rich Anadyr water or whether it pertains to Alaska coastal current water which contains fewer nutrients. The encroachment of Atlantic water into the surface of the Makarov Basin, seen under recent AO^{+} conditions, will further reduce the domain of cadmium-rich water within the Arctic (Figs. 18 and 20). The accompanying increased stratification and recycled production in the smaller Pacific domain of the Canada Basin will, however, tend to maintain cadmium entering in runoff or by atmospheric deposition at the surface. It is noteworthy that the Archipelago is the downstream recipient of water from the Pacific Ocean and, therefore, the recipient of cadmium and nutrients from that source. Again, change in the volume inflow and composition of Pacific water or change in the distribution of water masses within the Arctic Ocean both have the potential to alter the Cd content of water flowing through the Archipelago. With a relatively short resident time of ~6 months for water in the Archipelago, it is likely that only changes in ocean structure (currents and upwelling) could supply sufficient Cd to support a change in water properties of the Archipelago for any length of time.

Recent work on metal-impacted lakes near a copper-smelting center in Quebec, Canada, shows that metal loadings (Cd, Cu, Zn) can alter the ecosystem structure, causing the demise of medium to large benthic invertebrates and producing fish populations shifted to smaller sizes (Sherwood et al., 2000, 2001). This suggests a strong possibility that contamination by these heavy metals could interact with the accumulation and biomagnification of other contaminants like mercury and the organochlorines, actually reducing concentration of the latter in apex feeders.

Even without contaminant metal loadings, climate change can alter the natural cycle of metals like Zn and Cd such that biological burdens, exposure or sensitivity to them is altered. There has been very little research on this topic but recent findings by Köck and Triendl (1996) and Köck et al. (2001) show that concentrations of Cd and Zn in livers from Arctic char (*Salvelinus alpinus*) are sensitive to temperature, both seasonally and interannually. Other limnological factors likely play a role, but higher water temperatures associated with projected climate change, imply enhanced metal bioaccumulation and concomitant detoxification responses. Surprisingly, Arctic char are extremely susceptible to rising temperatures which lead to greater metabolic rates, increased pumping of water over gill surfaces and increased uptake of metals from the water. Do other freshwater or marine species have similar metal–temperature interactions? We do not know.

6.1.2. Mercury

Mercury must be considered separately from other heavy metals due to its volatility and tendency to undergo biogeochemical transformation (see, for example, Fitzgerald et al., 1998; Mason and Fitzgerald, 1996; Mason et al., 1994). We must pay particular attention to aquatic environments because it is there that mercury poses its greatest threat through meth-

ylation and biomagnification (Atwell et al., 1998; Driscoll et al., 1998; Evans and Lockhart, 1998; Gill et al., 1999; Muir et al., 1999). To a degree, processes leading to enhanced mercury concentration in the environment can be considered as either "solvent switching" or "solvent reduction" (see Section 6.3 and Macdonald et al., 2002a). In the former, mercury moves between phases such as air, water and particles based simply on partition coefficients, whereas in the latter mercury may achieve high fugacity through the loss of surfaces or through chemical reactions mediated by photons or microbes (see for example processes described in Lindberg et al., 2002; Malcolm and Keeler, 2002).

The natural mercury cycle has been enhanced by human activities such that two to three times as much mercury is presently cycling through the atmosphere and upper ocean than before the rise of industry (Lamborg et al., 2002; Mason and Fitzgerald, 1996; Mason et al., 1994; Pacyna and Keeler, 1995). For example, Pacyna and Pacyna (2001) estimated that human activities in 1995 released approximately 2235 tonnes of mercury with fossil fuel consumption contributing over half of that. This may be compared with 2500 tonnes estimated for natural emissions (Nriagu, 1989). The largest emitter of mercury from fossil fuel consumption is China (495 tonnes in 1995) which is directly upwind from the Bering Sea, Alaska and the western Arctic.

Atmospheric mercury is almost entirely gaseous Hg^0. It would be tempting to assume that the polar regions are global sinks simply due to low temperature and, accordingly, make simple projections of the effects of climate change based only on temperature. In fact, thermal forcing for Hg^0 to arctic aquatic systems either by rain or by air–water exchange is weak simply due to a relatively high Henry's Law Constant (Macdonald et al., 2000a; Mason et al., 1994). Nevertheless, the Arctic is uniquely vulnerable to global atmospheric mercury transport because of an extraordinary set of circumstances at polar sunrise that deposit reactive (and bioavailable) mercury to the surface (Fig. 25, Lindberg et al., 2002; Lu et al., 2001; Schroeder et al., 1998; Scott, 2001). A relatively long residence time for Hg^0 of 1–2 years in the atmosphere (Lamborg et al., 2002) ensures that winds can transport mercury to the Arctic. With polar sunrise, Hg^0 is converted to reactive gaseous mercury (RGM) through reaction sequences in which bromine, chlorine and compounds like BrO and ClO play prominent roles (Fig. 25 and see Lindberg et al., 2002; Lu et al., 2001). RGM is then very effectively removed from the atmosphere by particles/snow, with this process estimated to account for the deposition of ~50 tonnes (>90% of the annual total) on the Arctic Ocean and Hudson Bay during spring (Lu et al., 2001) and perhaps as much as 150–300 tonnes/year (Lindberg et al., 2002). The distribution of BrO from satellite measurements implies that reactive or particulate mercury is probably deposited on most of the land in northern Canada and in the Archipelago, and on much of the Arctic's marginal seas (Richter et al., 1998).

During spring and summer warming, some of the mercury deposited in snow is re-volatilized with estimates ranging from about two-thirds (AMAP, 2005) to only 10% (Dommergue et al., 2003). The remaining mercury likely enters aquatic environments through melt water. For example, in the Amituk Lake basin (26 km^2) on Cornwallis Island, Nunavut, it has been estimated that about 15 g of Hg were deposited during the year through rain and snow (Diamond et al., 2003). Of this, about 4.5 g Hg passed into the lake with, finally, 2.8 g Hg exiting the lake in the outflow. Before projecting the impact of global change on the mercury cycle in the Arctic, one more fact needs to be grasped. The invasion of mercury to global aquatic reservoirs via wet and dry deposition of reactive forms, is balanced by gaseous evasion of reduced forms of mercury such as Hg^0 or methylmercury (Mason et al., 1994). Therefore, one of the ways in which the Arctic is vulnerable to global mercury emissions lies in the mismatch between mercury invasion and evasion processes. Melt water and runoff, which contain deposited reactive mercury, can drain into surface water below the ice cover (in lakes or the ocean) whereas the evasion of gaseous mercury is partly or completely blocked by ice cover (Fig. 25). Indeed, this set of circumstances may partly explain trends of increasing mercury or elevated mercury observed in arctic aquatic biota (Evans and Lockhart, 1998; Macdonald et al., 2000a; Muir et al., 1999; Wagemann et al., 1995, 1996) despite the lack of any increase in total gaseous mercury (TGM) concentrations in air, at least during the past 5 years (Steffen et al., 2005). Unfortunately, coherent mon-

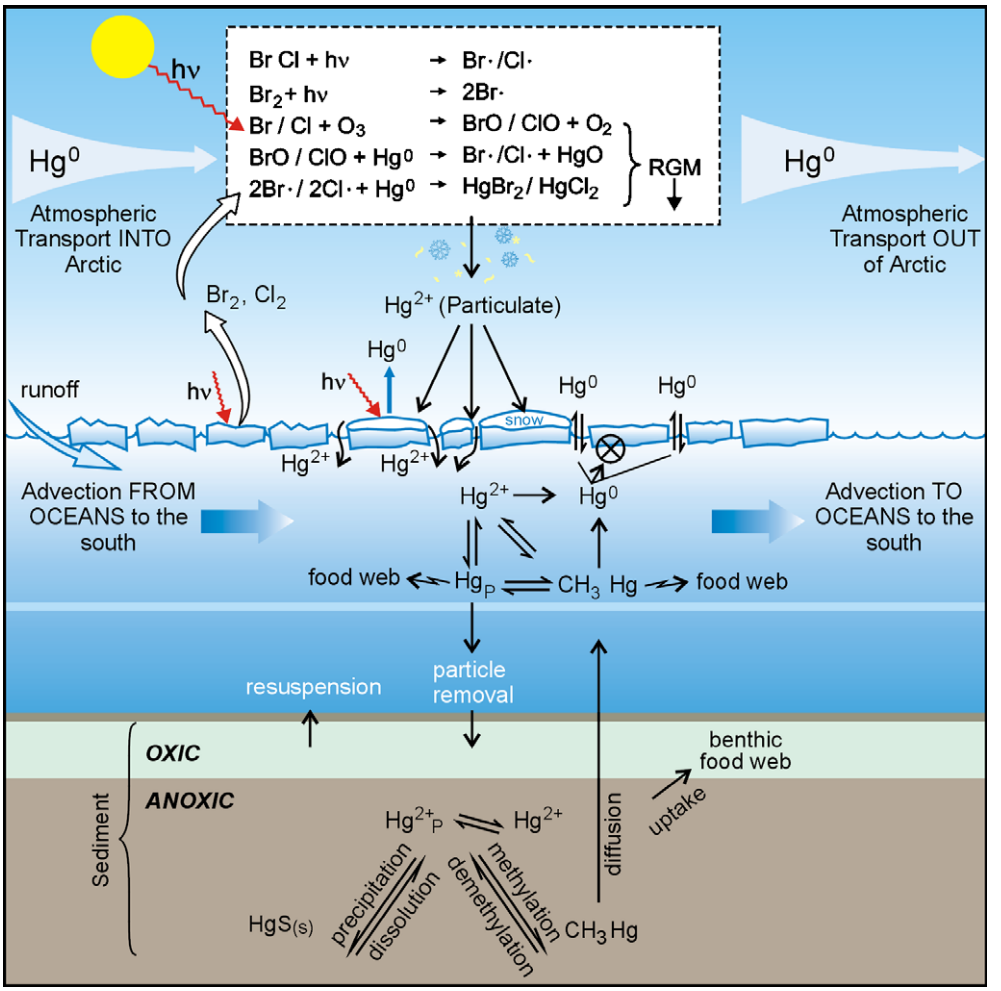

Fig. 25. A schematic diagram illustrating the mercury cycle in the Arctic. The chemical processes initiated by light radiation after polar sunrise are listed in the inset box at top right (for mercury reactions schemes in atmosphere and ocean see Lamborg et al., 2002; Lindberg et al., 2002). The mercury cycle in aquatic environments is illustrated in detail for lakes—the same processes apply to oceans but have not been shown due to space limitations.

itoring of different phases (air, water, biota) and appropriate geochemical studies investigating the complete pathways of mercury in arctic aquatic systems (Fig. 25) have not been done and we are forced at this time to the above speculations.

Climate change can impact both the invasion and evasion routes for mercury. Springtime depletion of atmospheric mercury depends on the availability of sea salt or salt flowers, calm weather, a temperature inversion, the presence of sunlight and sub-zero temperatures (Lindberg et al., 2002; Lu et al., 2001); these components of the system provide a tremendous scope for change in boundary-layer chemistry (Shepson et al., 2003) but, unfortunately, detailed studies of chemistry involved in ocean–atmosphere–sea ice–snowpack interactions at polar sunrise have not yet been conducted. Initially with climate change, it is likely that increased amounts of first-year ice around the polar margins will contribute to generally saltier ice and snow in spring which will enhance the production of BrO/ClO. Depending on how the rate of supply of gaseous mercury to the Arctic is controlled, increased BrO/ClO will either enhance scavenging or maintain it at present levels, possibly

extending the area of springtime mercury depletion beyond that implied by recent satellite measurements of the distribution of BrO/ClO (see Lu et al., 2001). Considering that global emissions of mercury have actually been decreasing of late, Lindberg et al. (2002) proposed that the observed recent increases in mercury levels in arctic biota are, in fact, evidence that MDEs may be a recent phenomenon due to change in sea-ice climate over the past decade or two. Mercury depletion events leading to the production of bio-available mercury have been confirmed at Barrow, Alaska (Lindberg et al., 2002). It seems likely that the Bering/Chukchi Sea region may be especially vulnerable to further increased mercury loadings given that this region receives airborne contaminants from Asia (Li et al., 2002; Wilkening et al., 2000), that China is increasing its reliance on coal for energy, and that coal is a leading source of anthropogenic mercury (Nriagu and Pacyna, 1988).

Before MDEs can be implicated in increased mercury concentrations in apex feeders in aquatic systems, all components of the mercury pathway must be considered. Larger areas of open water in spring, either for ocean or lakes, will enhance exchange allowing gaseous forms of mercury to escape back to the atmosphere (Fig. 25). With further warming, parts of the Arctic will become more temperate in character implying that atmospheric mercury depletion would decrease and evasion from the water increase leading eventually to lower total mercury concentrations in water.

Aquatic food webs have strong leverage for change in mercury exposure in the Arctic because methylmercury biomagnifies, exhibiting a concentration increase from particulate organic matter to apex predators of about 1000–3000 (Atwell et al., 1998; Kidd et al., 1995a; Muir et al., 1999). Mercury concentration also increases with age and size in predatory fish such that large, old fish often exceed thresholds considered safe for unrestricted human consumption (Lockhart and Evans, 1999), containing anywhere from two to five times the mercury concentration in smaller fish. Therefore, adding an extra step in the food web could enhance mercury concentrations in higher trophic levels by a factor of 5. Likewise, altering the population age distribution of fish in a lake could produce a change rivaling or exceeding that caused by alteration of physical pathways.

With warming (Fig. 3) will come the loss of permafrost (Fig. 21) which then leads to altered hydrology, potentially more wetland, and enhanced fluxes of soil and organic carbon to rivers, lakes and estuaries. The coupling between mercury deposition to arctic surfaces and entry into lakes will likely be enhanced by projected changes in the hydrological and organic carbon cycles (Diamond et al., 2003; Stanley et al., 2002). Warming of drainage basins in the Arctic, therefore, would appear to provide a widespread mechanism to increase mercury fluxes to northern aquatic environments and to the atmosphere. A recent study of the pathway of mercury from snow-covered land to streams in Vermont (Stanley et al., 2002) showed that mercury export from soils correlated with particulate organic carbon, and that mercury concentrations in runoff increased with flow—unlike most solutes (see also Bishop et al., 1995). These two factors together suggest that episodic, large releases of organically bound mercury (contaminant and natural) may accompany permafrost degradation. Clearly, arctic lakes would be most vulnerable to this process, but enhanced input of terrestrial carbon is projected to occur to arctic seas as well (Benner et al., 2004; Kabat et al., 2001) suggesting that mercury loadings there may, similarly, be increased. In the ocean, Hudson Bay would seem especially vulnerable partly due to its large drainage basin, already impacted by reservoir flooding (Bodaly and Johnston, 1992), and partly due to the likelihood of permafrost melting within that drainage basin (Gough and Wolfe, 2001). It seems noteworthy that enhanced snow concentrations for mercury are observed generally in that region (Lu et al., 2001) and that a recent increase in mercury flux to Hudson Bay sediments has, likewise, been observed (Lockhart et al., 1995).

Historical records from dated sediment cores have been used to infer mercury fluxes increasing by factors of 3–7 in some arctic lakes during the past two centuries (Bindler et al., 2001; Landers et al., 1995; Lockhart et al., 1998). What is not so clear is whether such increases are due to increased atmospheric deposition or to alteration of processes that transfer mercury from wetlands to atmosphere or from the drainage basin into lakes or transfer mercury from the water to the sediments. In regard to the latter, Gajewski et al. (1997) have shown major increases in

diatom fluxes to varved sediments from a lake on Devon Island, which they attribute to climate change (i.e., longer ice-free summers). Not only could such a mechanism explain enhanced mercury fluxes to arctic lake sediments but it could also have the non-intuitive result of *reducing* the exposure of higher trophic levels to mercury through bloom dilution at the algal stage (Pickhardt et al., 2002).

Finally, methylmercury, which presents the greatest health hazard of the various mercury species shown in Fig. 25, may be produced in greater quantity by climate changes that lead to a more vigorous carbon cycle and higher temperatures. Wetlands and wetland sediments are net producers of methylmercury (Driscoll et al., 1998; Suchanek et al., 2000) and methylmercury observed in fish from small lakes appears to correlate with the amount of watershed occupied by wetlands (Greenfield et al., 2001). Flooding of terrestrial landscape has the well-known consequence of releasing mercury from submerged soils (Bodaly et al., 1984). Therefore, alteration of wetland distribution or area as a result of permafrost melting in the Arctic is yet another way to release mercury or convert it to its environmentally most toxic form—all the more serious if arctic soils contain an inventory of contaminant mercury accumulated during the past century or two.

In summary, the above discussion and Fig. 25 reveal mercury to have a particularly complex set of pathways in the Arctic which can be altered at many stages including deposition from the atmosphere (MDEs), ice- and snow-mediated coupling into the hydrological cycle, entry into the organic carbon cycle, release from permafrost melting, biomagnification or bioaccumulation in foodwebs, and transformation to methylmercury due to change in wetlands or organic carbon cycling.

6.1.3. Arsenic

The sediment geochemistry of arsenic can be altered by changing the carbon cycle. Mining and smelting have been major sources to the environment of arsenic (Nriagu, 1989) where it may accumulate in aquatic sediments (Martin and Pedersen, 2002). In the Arctic, it has also been suggested that underwater nuclear weapons testing may have provided a significant contaminant arsenic source to the Pechora Sea (Loring et al., 1995). Diagenesis often produces large natural surface sediment arsenic enrichments by remobilizing solid-phase As(V) at depth in sediments through reduction to As(III) which then diffuses upward to re-precipitate through reactions with nitrate, oxides of manganese or oxygen (Fig. 26). This natural redistribution of arsenic in sediments makes it exceptionally precarious to infer contamination from sediment surface arsenic enrichments (Loring et al., 1995, 1998; Siegel et al., 2001). However, strong sediment-surface enrichments serve as a warning that alteration of the organic geochemistry of aquatic sediments by enhanced organic

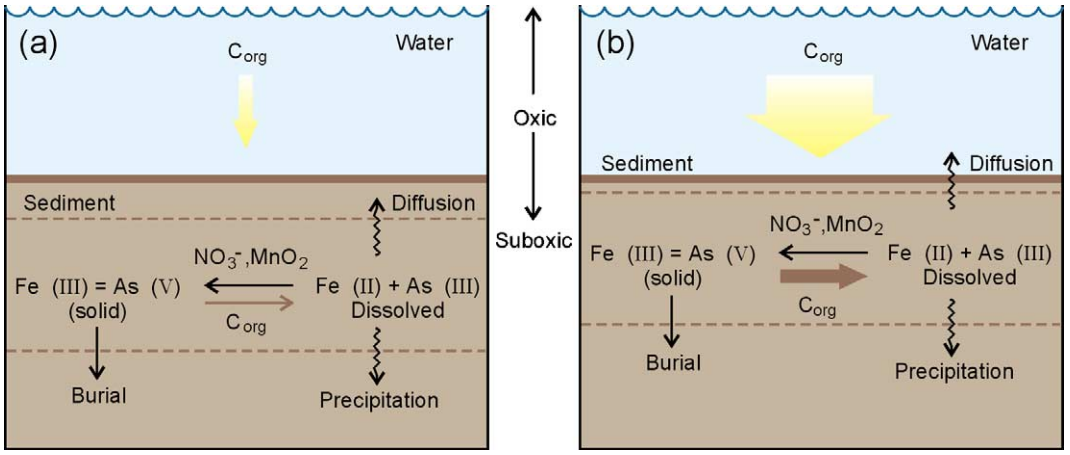

Fig. 26. A schematic diagram showing how arsenic cycles in sediments (modified from Sullivan and Aller, 1996). The solid-phase form of arsenic (As(V)) is released to pore water through reduction to As(III) which then may diffuse back out of sediments. Enhanced fluxes of organic carbon to sediments may therefore lead to a release of dissolved arsenic to bottom waters.

carbon fluxes may have unanticipated consequences for arsenic mobility.

A geochemical study of a temperate, seasonally ice-covered lake impacted by copper, zinc, nickel and arsenic is especially instructive (Martin and Pedersen, 2002). When action was taken to reduce metal loadings, the lake's response was enhanced phytoplankton production which then invigorated carbon fluxes to sediments. The enhanced carbon fluxes then reduced oxygen through metabolism in sediments, thus mediating the conversion of solid-phase As(V) to dissolved-phase As(III), which then diffused back into the lake's bottom water. The unanticipated result of decreasing metal loadings to the lake was to produce higher arsenic concentrations. If one of the responses to change in arctic aquatic environments is enhanced aquatic productivity or enhanced organic carbon loadings, release of solid-phase arsenic, especially from sediments with high natural or contaminant burdens (e.g., see Loring et al., 1995, 1998; Siegel et al., 2001) is a possible response.

6.2. Radionuclides

Previous assessments have outlined the atmospheric and oceanic pathways that transport artificial radionuclides to the Arctic (AMAP, 1998; Macdonald et al., 2000a). Atmospheric sources of artificial radionuclides derive mainly from atmospheric weapons testing predating the mid-1960s, and Chernobyl in 1986. Accordingly, predominant food web contamination from the atmosphere has occurred to terrestrial ecosystems through fallout (AMAP, 1998) and there appears little opportunity for present or future climate changes to have much impact on historical atmospheric sources which are decaying away with half lives of about 30 years (^{137}Cs and ^{90}Sr, predominantly). The distribution of fallout from Chernobyl was very much controlled by wind and rainfall patterns (see Figs. 8.4 and 8.5 in AMAP, 1998), and these pathways are clearly subject to climate change. An important lesson to be learned from the Chernobyl accident is that climate patterns can predispose the Arctic in how it will receive contaminants from such accidents and, for example, wetter conditions in the Nordic Seas and Northern Europe during AO$^+$ conditions (Fig. 9) would favour deposition of ^{137}Cs fallout in that region.

In the ocean, the predominant artificial radionuclides (e.g., ^{137}Cs, ^{90}Sr, $^{239+240}$Pu) do not biomagnify sufficiently (excepting perhaps Pu (and ^{99}Tc) in brown macroalgae and see Berrow et al., 1998; Brown et al., 1999; Fisher et al., 1999) to contribute significantly to the radiation dose for humans (Layton et al., 1997; Macdonald and Bewers, 1996). The above facts suggest we must look to the ice and surface water motion of the Arctic Ocean, both of which undergo alteration with the AO/NAO (Figs. 14 and 19), to find pathway changes that might be cause for concern.

Clearly, the enhanced northward transport of water in the Nordic Seas under the AO$^+$ conditions of the 1990s strengthened the delivery to the Arctic of radionuclides discharged by the European nuclear reprocessing plants (Fig. 20) where they continued to spread into the surface waters of the Makarov Basin (Figs. 18 and 20, Smith et al., 1998).

A shift in the climate regime toward increased NAO and stronger wind fields will probably lead to radionuclides undergoing a faster transport closer to the Norwegian coast, and a larger proportion of them will enter the Barents Sea. It is expected that other European contaminants entering the North Sea and southern Norwegian Sea will encounter similar change in their oceanic transport route as projected for the radionuclides.

The enhanced coupling between release points for European reprocessing plant nuclear wastes and the Arctic Ocean will be more than offset by reduction in releases that have occurred since the 1970s (Macdonald et al., 2000a), and input of these tracers to the Arctic Ocean should continue to decline. The extensive data collections under the Arctic Nuclear Waste Assessment Program (ANWAP, Layton et al., 1997) and from icebreakers (Smith et al., 1998) have led to a reasonable understanding of the distribution of artificial radionuclides in Arctic surface waters and confidence that they pose little risk to human or ecosystem health. The conclusion of the ANWAP assessment was that the largest radiation doses to individuals living on the Alaskan coast and consuming subsistence seafoods were, in order of importance, ^{210}Po (a natural radionuclide), followed by ^{137}Cs and ^{90}Sr from atmospheric fallout. It seems that the nuclear reprocessing radionuclides have made elegant tracers of water motion and, although they will reflect

recent changes in that motion, they will continue to provide almost no risk to arctic marine biota (Macdonald and Bewers, 1996).

The ice and surface water pathway changes forced by the AO (Figs. 15 and 19) strongly suggest that radionuclides discharged to the Russian shelves may, under AO^+ conditions, enter the Canadian Basin and subsequently find their way into the Canadian Archipelago. The estimated total release of radionuclides to the Kara Sea via river water (Ob, Yenisey) is about 1.1×10^{15} Bq (^{90}Sr and ^{137}Cs) (Paluszkiewicz et al., 2001), or about an order of magnitude less than the fallout and reprocessing plant sources to the Arctic Ocean (Aarkrog, 1994). It seems likely, therefore, that diversion of Russian river runoff from the Eurasian Basin to the Canadian Basin under AO^+ conditions will be matched by a diversion of associated radionuclides (see Cooper et al., 1999) which, nevertheless, will provide little threat to ecosystems there.

Ice drift, the remaining transport pathway to consider, provides a distinct, but difficult to quantify risk. Sediments from the Russian shelves, known to have been contaminated by weapons testing and accidental and deliberate radio-waste discharges, have been found to be heavily contaminated in several locations (Josefsson, 1998; Matishov et al., 1999; Smith et al., 2000). Suspension freezing provides an efficient mechanism to entrain fine sediments into newly formed ice in the Russian Seas (Eicken et al., 2000) and ice has been shown to carry radioactive sediments (Dethleff et al., 2000a; Landa et al., 1998; Meese et al., 1997). High radioactivity has been found in ice-entrained sediments in the Canada Basin (>70 Bq/kg, Cooper et al., 1998) and in the Archipelago (Darby, personal communication) but the origin of the sediment in the ice, based on mineralogy, has not been assigned to Russian shelves. Given the very few samples together with their uncertain provenance, it is impossible to quantify risks to biota in the Canada Basin and Canadian Archipelago from contaminated ice except to say that the AO^+ conditions of the early 1990s appear to produce ice transport pathways more conducive to carrying sediment and surface water from the Russian Shelves into the Archipelago.

Perhaps the most significant increase in radioactivity exposure to northerners will come from the natural ^{226}Ra decay series that supports ^{222}Rn, ^{210}Pb and ^{210}Po. ^{210}Pb in aquatic systems derives partly from in-situ production supported by ^{226}Ra and partly from ^{222}Rn which has diffused out of soils and, with a short 3.8 day half-life, decays to ^{210}Pb which is scavenged by particles. This latter component, called excess ^{210}Pb, often exceeds the 'supported' ^{210}Pb in aquatic sediments. Presently, excess ^{210}Pb tends to be very low in the Arctic because ^{222}Rn remains trapped in the soil by permafrost and snow/ice cover. With warming, ^{222}Rn evasion will increase as will excess ^{210}Pb activity matched by the activity of ^{210}Po, its granddaughter. Since ^{210}Po and ^{222}Rn together account for about 75% of the radiation dose to native northern residents (Layton et al., 1997; Macdonald and Bewers, 1996) any increase in ^{222}Rn evasion due to warming/permafrost melting would have a widespread and parallel increase (doubling or tripling) of the radioactive dose.

6.3. Organochlorines

Of all the contaminants, the organochlorines (OCs) provide the greatest challenge to predict consequences of change because they have been so widely released, comprise so many compounds and exhibit such a wide range of chemical properties. Furthermore, the important chemical properties, volatility, phase partitioning, and degradation kinetics, are all sensitive to temperature and hydrological change. Efforts to determine where in the environment these compounds end up has improved our understanding of global pathways enormously, but surprises alert us that intuition often fails because we do not have a complete grasp of environmental processes (Macdonald et al., 2000b; Oreskes et al., 1994; Schindler, 1997). Recent OC budgets underscore the importance of the atmosphere–ocean partnership in the transfer of OCs to the Arctic from their temperate and tropical release points (Li et al., 2002, 2004; Macdonald et al., 2000a,b; Shen et al., 2004). These same budgets show that the relative importance of atmosphere versus ocean in transporting contaminants varies widely between OC compounds and over time. Therefore, change forced by the AO or by general global change will have similarly varied impact depending on the particular OC and the time period in question. All of the OCs of concern (DDT, toxaphene, chlordane, PCBs, HCHs) have transient emissions: that is, they were first released in the 1930s to 1940s, peak sometime in

the 1970s to 1990s and emissions will continue to decline or cease, or possibly increase, depending on bans, further controls, or invigorated assaults by pests encouraged by climate change. As a general rule, particularly with transient releases of chemicals that partition strongly into water, the atmosphere is initially the dominant transporting medium but as aquatic reservoirs (lakes, rivers, upper ocean) become loaded, these then contain the important if not dominant budget and flux terms (Li et al., 2002, 2004; Macdonald et al., 2000a,b). The dominant budget terms posses the greatest leverage for change.

Before discussing how climate change is likely to impact organochlorines, it is important to understand the manner in which organochlorines become concentrated in the environment. Building on earlier perspectives developed by Wania (1999), Macdonald et al. (2002a) suggest there to be two fundamental concentrating processes which they termed *solvent switching* and *solvent depletion*. The distinction between these two processes is particularly important in the context of change. Solvent switching is a natural process wherein a given contaminant distributes itself between various phases (solid, liquid, gas) according to well-described thermodynamic rules. This process can lead to elevated concentrations—for example, the partitioning of HCH into cold water will produce concentrations in the water that far exceed those in the air, and indeed this process alone can cause unexpected divergences of chemicals over large scales (Li et al., 2002). But this process cannot cause concentrations to exceed thermodynamic equilibrium; rather, during partitioning chemicals redistribute themselves in the direction of thermodynamic equilibrium (that is, solvent switching does not increase fugacity). The transfer of a POP from water to lipid at the bottom of the food web is another very important solvent-switching process; again, this process enhances POPs concentrations in phytoplankton or small zooplankton strictly according to thermodynamic forcing.

Solvent depletion, however, differs from solvent switching in that it can lead to concentrations in selected media that exceed thermodynamic equilibrium (i.e., fugacity is increased), but to do this requires a source of energy. Perhaps the clearest example of solvent depletion occurs in the food web where lipid transfers on going from one trophic level to a higher one are inefficient. Metabolism effectively burns much of the lipid, leaving the contaminant to accumulate in a decreasing volume of stored fat. This process can lead to organochlorine concentrations in aquatic apex feeders that are well above thermodynamic equilibrium with the water, and the situation can be exacerbated by starvation cycles during which individuals even further reduce their fat content. Arctic and other cold environments offer a number of solvent depletion processes, all of which can be manipulated by climate change, by which contaminants can be ramped up well above thermodynamic equilibrium (Macdonald et al., 2002a). For example, a POP can be scavenged by absorption to snow and thence transported to ground. The initial large snow surface ($0.1\ m^2\ g^{-1}$) can be reduced by a factor of over 100 as the snow sinters and, indeed, during melting, the snow surface can disappear entirely, which effectively removes the solvent (the snow's surface) from under the contaminant. The fugacity can be increased enormously during snow sintering/melt with the consequence that the organochlorine is forced to go somewhere else—diffuse back to the atmosphere, enter the meltwater or adsorb to other particles. Similarly, fogwater can provide a temporary large surface area which is thermodynamically attractive (that is, solvent switching favours distribution of the chemical onto the surface). Upon coalescence, most of the surface is destroyed and contaminant fugacity increases in the remaining large water drops. Finally, cryo-concentration may occur in shallow lakes or seas that form a thick ice cover. The withdrawal of water into the ice, leaving behind most of the POPs, can easily increase concentrations in the water beneath the ice to values that exceed thermodynamic equilibrium with the atmosphere by factors of 2 to 5. Diffusion of the chemical back into the air is not an option until spring breakup due to ice cover.

Under change scenarios, the solvent switching processes can be modeled by taking into account changes in partition coefficients and vapour pressures with temperature. However, changes in the solvent depletion processes are much harder to project especially since the elevated fugacities imply transports that will be sensitive to time. For example, the effect of snow on contaminants entering an arctic lake will depend on when the snow accumulates, how long it sinters, how porous and how deep the snow is, how

and when snow melt occurs, and how quickly snow melt enters the lake. None of these transfer processes have been investigated in sufficient detail to provide guidance to modeling.

6.3.1. The influence of the Arctic Oscillation on organochlorines

There appear to be several consequential ways that physical pathways could change in response to the AO. In the Nordic Seas, the atmospheric coupling of eastern North America and western Europe with the Arctic becomes more intense, especially during winter/spring (Fig. 8a). Spraying of pesticides in these regions will, if anything, intensify 'events' like those seen at Alert for α- and γ-HCH (see, for example, Fig. 22 in Macdonald et al., 2000a). Furthermore, re-emission of old OC residues in soils and aquatic reservoirs of Europe or eastern North America will enter these same air pathways to be transported north. In this regard it is worth noting that the highest cumulative use of PCBs was in western Europe and Eastern United States (see Fig. 2.3.1. in AMAP, 2005)—both are source regions for air masses entering the Arctic between Greenland and Europe (Fig. 8a). Similarly, the United States has been historically the largest user of toxaphene (490 kt up to 1982 when it was banned) such that soils in the south-east regions continue to provide fluxes to the atmosphere through re-emission (Harner et al., 2001). The higher precipitation in the Nordic Seas and southern Eurasian Basin (Fig. 9) will provide more effective scavenging of particle-associated OCs (high molecular weight PCBs, for example) and for OCs with low Henry's Law Constants (HLCs) that partition strongly into water (HCHs, toxaphene).

In the Bering Sea, gas exchange with the ocean or washout by rain can provide a mechanism to remove β-HCH selectively from the air as it heads northward simply due to its exceptionally low HLC (Li et al., 2002). This process does not prevent the entry of the contaminant to the Arctic Ocean; rather, it switches the mode of delivery from winds to ocean currents which consequently slows the rate of transport from m s^{-1} to cm s^{-1}. At the large scale, the HLC provides a hemispheric 'chromatographic' separation that impacts the buildup and release of the HCHs over time periods measured in decades (Shen et al., 2004) which then permits decadal climate changes to modulate the process. Under AO^{+} conditions, therefore, we predict a more rapid atmospheric transport of OCs into the Arctic from western Europe, with the delivery shifting toward the ocean pathway for OCs that partition strongly onto particles or have low HLC. On the Pacific side, OCs will continue to enter the Arctic via the atmosphere and ocean currents (see for example Bailey et al., 2000) but the 15% reduction in Bering Sea inflow over the past several decades would effect a proportional reduction in this pathway. Variation in precipitation over the North Pacific and Bering Sea will alter the balance between atmosphere and ocean as transport pathways to the Arctic. The relatively good spatial and temporal database for α-HCH (atmosphere, surface ocean) has allowed a plausible reconstruction of the history of its hemispheric transport and ocean loading. Beginning in the 1940s and continuing until bans in China in the early 1980s and India in the late 1980s, α-HCH was gradually loaded into the Arctic Ocean under the permanent pack. The final stage of unloading the Arctic Ocean surface reservoir is now occurring as α-HCH-enriched water exits through the Canadian Archipelago and/or Fram Strait and transports southward to the eastern seaboard of Canada where through warming the α-HCH evades back into the atmosphere (Shen et al., 2004). During this 50-year period of loading and unloading, climate change has had many opportunities to affect the process of α-HCH distribution, ocean loading, and degradation through alteration in (1) ice cover; (2) surface water temperatures; (3) precipitation distribution; (4) surface-water residence times in the Arctic; (5) diversion of river inflow pathways; (6) coupling between the Eurasian and Canadian Basins; and (7) the exit pathways (Fram Strait/Canadian Archipelago).

Larger areas of open water observed under AO^{+} conditions or from general climate change (Figs. 11–13) will accelerate equilibrium between air and sea by an amount that scales with the expanded open areas. Furthermore, increased numbers of polynyas in winter will enhance the production of fog over sea ice, acting to scavenge and deposit contaminants to surface (Chernyak et al., 1996) at locations known to be important for biota (Stirling, 1997). Due to the drastic reduction in atmospheric concentration through emission controls, α-HCH became oversaturated in ice-covered areas of the Arctic Ocean (Jantunen and

Bidleman, 1995; Macdonald et al., 2000b). The opening of the pack and the clearance of shelves seasonally will, in this case, result in evasion and drawdown of α-HCH from the upper ocean. In contrast, PCBs and toxaphene appear still to be loading into the Arctic Ocean via the atmosphere (Macdonald et al., 2000b) and, therefore, the same loss of ice cover will lead to increased loading of the Arctic's surface water with these two chemical groups. To emphasize this point, the impact of altered ice cover depends on what point the chemical is in its transient loading history—something that may even differ between congeners within a single OC group (e.g., PCBs). A ΣPCB budget estimated a net gas exchange into the Arctic Ocean of about 20 tonnes/year (Macdonald et al., 2000b). Reduced ice cover illustrated in Fig. 13 might lead to as much as a doubling of the area of open water which would similarly double net air–sea exchange of PCB.

The diversion of Russian river inflow toward the east under AO^{+} conditions (Fig. 19) will have a significant effect on OC pathways within the Arctic Ocean. Inherent with this diversion is a shift of all the OC loadings from these Russian rivers out of the Eurasian Basin and into the Canada Basin (Table 1 and see, Macdonald et al., 2000a; Sericano et al., 2001). As the Canada Basin has a longer residence time (10 years compared to 2 years in the TPD), there would be an added consequence of increased contaminant inventories for the Arctic Ocean in general and the Canada Basin in particular. Instead of tracking across the Eurasian Basin to exit into the East Greenland current, OCs discharged by the Russian rivers might now exit via the Archipelago (see Fig. 19). The changes here are consequential to budgets (Table 1) and to distribution within the water column keeping in mind that the same river water that delivers contaminants also stratifies the ocean and potentially reduces new production and vertical particle flux which together will act to maintain river-borne contaminants near the surface where it can partition into algae. Although there are few data from which to evaluate the relative importance of organochlorine pathways in the ocean, the findings of Andersen et al. (2001) provide a strong warning that there are sources of PCB in the region around the Kara Sea and Franz Jozef Land, possibly a consequence of river inputs.

Table 1
Russian river loadings for selected organochlorine compounds

Organochlorine	Loading (tonnes/year)	% of Arctic ocean input budget	Reference
α-HCH	25	13	(Alexeeva et al., 2001; Macdonald et al., 2000a)
γ-HCH	44	51	(Alexeeva et al., 2001; Macdonald et al., 2000a)
ΣPCB	15	23	(Macdonald et al., 2000b)
ΣDDT	18		(Alexeeva et al., 2001)

Water in the Canadian Archipelago channels, which is supplied from surface water in the Arctic Ocean (~0–200 m), has the potential to undergo change in its organochlorine content due to alterations in the distribution of water masses in the Canada or Eurasian Basins. As shown in several studies (Carmack et al., 1997; Li et al., 2002, 2004; Macdonald et al., 1999b, 2000a), HCHs are not distributed uniformly within the Arctic Ocean and it is likely that other organochlorine compounds are, likewise, not uniformly distributed. For example, α-HCH is highest near the surface, decreasing to very low values in water deeper than several hundred meters, and the Canada Basin under the permanent pack exhibits much higher HCH concentrations than are observed in the Chukchi Sea or the Eurasian Basin surface waters. The redistribution of Pacific and Atlantic water masses in surface water of the Arctic Ocean (Figs. 18 and 20) may therefore have been accompanied by change in the composition of water flowing into the Archipelago. Such change could occur in two ways, either by horizontal displacement of water mass domains or by vertical displacement of water properties. We have few data from which to evaluate how the water composition in the Archipelago channels might respond to the Arctic Oscillation. An extraordinary set of data collected by Hargrave et al. (1997) illustrate clearly that change in upstream water composition can have important consequences for contaminant concentrations in water flowing through the Archipelago and, by inference, in locations to the south of the Arctic (Shen et al., 2004). Seasonal measurements of organochlorine compounds made at Resolute in 1993 (Fig. 27) reveal a coherence between the surface and 50 m water depth in contaminant trends. This coherence, together with the fact that the patterns

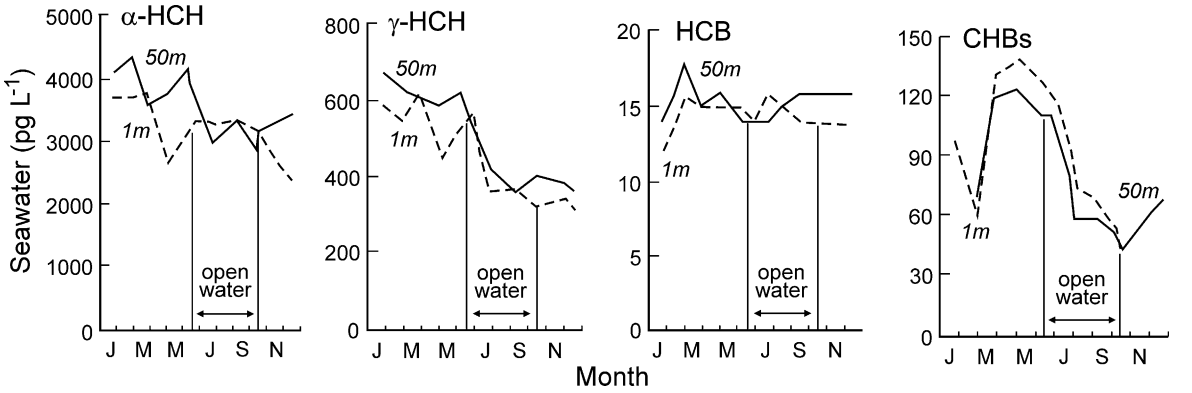

Fig. 27. Organochlorine concentrations measured at Resolute (Canadian Archipelago) during 1993. Shown are time series at the surface and 50 m water depth for α- and γ-HCH, hexachlorobenzene (HCB) and toxaphene (CHB). The coherence in trend between the surface and 50 m water depth for all four contaminants strongly suggests the mechanism to be change in upstream (Canada Basin) water properties (source: Hargrave et al., 1997).

differ among organochlorine compounds, argues strongly that the observed time trends at Resolute are produced by variation in the composition of upstream water drawn into the Archipelago from the Canada Basin. This single season's data alone contain concentration variations exceeding a factor of 2.

6.3.2. The effect of glacial melt back

Glacier ice-mass loss and snow melt back due warming (cyclical or trend) can release archived contaminants accumulated during years of higher fluxes (Blais et al., 1998). Based on the total amount of glacial melt back (Fig. 22) and the range of concentration of OC contaminants measured in ice and snow during the 1960s to 1990s for the Agassiz Ice Cap (Table 2, Franz et al., 1997; Gregor et al., 1995) we would expect the maximum total input to be about 3 kg for PCBs to perhaps as much as 400 kg for DDT. In the year exhibiting the most substantial melt back on record (1993), we estimate a maximum release of about 0.5 kg PCBs and 74 kg DDT. The amounts for HCH and PCB are miniscule compared to Arctic Ocean budgets (Macdonald et al., 2000b) and are also relatively small compared to the flux of these contaminants through the Archipelago (Table 2, right hand column) with the exception of DDT for which glacial melt appears potentially to provide an important, climate-modulated source. For arctic glaciers, most of the melt occurs in zones where old ice of pre-industrial age is emerging. This ice would contain little or no contaminant and, indeed, would act to dilute any released contaminants. For smaller ice caps, more recent layers of snow and ice might be involved. We conclude, therefore, that glacial melt back could only be of significance for DDT and even there it is likely to have only local and short-lived significance.

6.3.3. The effect of warming on organochlorine cycling in lakes

Arctic lakes presently tend to retain only a small fraction of the contaminants delivered to them,

Table 2
Potential input of selected organochlorines from glacial melt

Compound	Concentration (pg/L)	Total glacial input (kg)	Glacial input for 1993 (kg/year)	Flux through the Archipelago[a] (kg/year)
α–HCH[b]	256	205	39	195,000
γ-HCH[b]	115	92	18	27,900
ΣDDT[b]	480	384	74	161
CHLOR[b]	35	28	5	96
HCB[b]	65	52	10	810
PCB[c]	3.5	2.8	0.5	2700

[a] Flux through Archipelago was estimated assuming a mean flow of 54,000 km^3/year (Macdonald et al., 2000a) and using concentration data collected in the Archipelago during 1993 (HCH, HCB, CHLOR, Hargrave et al., 1997) or for the Canada Basin in 1997–1998 (PCB, DDT, Macdonald et al., 2001).

[b] Concentration data from Franz et al. (1997). Samples collected for 1987, 1990 and 1992 in snow layers after first year loss.

[c] Concentration data from Gregor et al. (1995); average concentration over 30-year period from 1964/1965 to 1992/1993 (n=34).

something that should alert us to the potential for change. Studies and models (Helm et al., 2002; Macdonald et al., 2000a) show that the snowmelt and runoff cycle connects with an arctic lake's hydrological cycle such that most of the contaminants deposited in the drainage basin throughout winter transport across the lake surface in a low density layer under the ice to exit in out-flowing water. The lack of a strong particle flux due to oligotrophic conditions further decouples deep lake water from contaminants at the surface. Reduced ice cover and loss of permafrost, leading to greater mixing and stronger primary production, will enhance the ability of arctic lakes to retain OCs. How this added retention will play out in the food web is less certain. Enhanced primary production and settling of ungrazed phytoplankton in early spring might draw down contaminant burdens in lake surface water and thereby reduce entry of contaminants into the food web or act so as to dilute the organochlorine concentration in algae as has been shown for mercury (Pickhardt et al., 2002). However, if a given lake has a very low sedimentation rate and most of the organic carbon depositing to sediments becomes metabolized, it is likely that OCs associated with the particle flux will be released to the bottom water and re-mixed within the lake as has been observed in Lake Superior (Jeremiason et al., 1994). This process of drawdown and remineralization could slowly ramp up water column concentrations below the lake's thermocline, especially if ice cover is sufficient to hinder exchange during lake turnover. It appears that climate change has the potential to cause substantive physical and biological changes in northern lakes that would alter OC pathways, but we are presently woefully ill prepared to predict what those changes might be.

6.3.4. The effect of warming on chemical partitioning and degradation

Physical–chemical properties sensitive to temperature include vapour pressure (p^0), Henry's Law Constant (HLC, alternatively expressed as the air–water partition coefficient, K_{AW}), octanol–air partition coefficient (K_{OA}) and the octanol–water partition coefficient (K_{OW}) (Table 3). The particle–gas partition coefficient (K_p), which depends both on particle composition and chemical composition, also varies with temperature. These properties by themselves exert a lot of control on whether or not a contaminant can arrive in the Arctic (Wania, 2003).

The extent to which chemicals are associated with aerosols is key to their atmospheric transport. Association with particles may on one hand slow or reduce transport to the Arctic through temporary or permanent deposition to surfaces. On the other hand, association with particles may protect a chemical from oxidation during transit to the Arctic. The potential for temperature to alter partitioning between gas and aerosol phases appears greatest for chemicals that exhibit log K_{OA} values in the range of 11 to 14 (Fig. 28). For example, with a temperature rise from winter (−30 °C) to summer (0 °C), DDT goes from being over 70% on particles to being almost entirely in the gas phase. Similar changes in partitioning are evident for many of the other organic compounds illustrated in the figure. Chemicals with K_{OA} values at −30 °C of about 11–12, therefore, will be most sensitive to change in atmospheric transport during temperature rise.

At 0 °C most of the chemicals shown in Fig. 28 are in the gas phase implying they are easily advected by air but will be prone to photolytic degradation. Timing is important: the transport processes that produce

Table 3
Physical parameters sensitive to temperature change

Parameter	Abbreviation	Unit	Description/application
Henry's Law Constant	HLC	Pa m^3 mol^{-1}	Partitioning between air and water
Air–water partitioning coefficient	K_{AW}	Unitless	K_{AW}=HLC/RT
Octanol–air partition coefficient	K_{OA}	Unitless	Used as a proxy to model partitioning between air and organic phases such as vegetation, soil, sediment organic carbon, and particles in air and water.
Vapour pressure	p^0	Pa	Describes tendency of a chemical to volatilize
Particle–gas partition coefficient	K_p	m^3 μg^{-1}	The ratio of chemical concentration on atmospheric particles (ng μg^{-1}) to concentration in the gas phase (ng m^{-3})

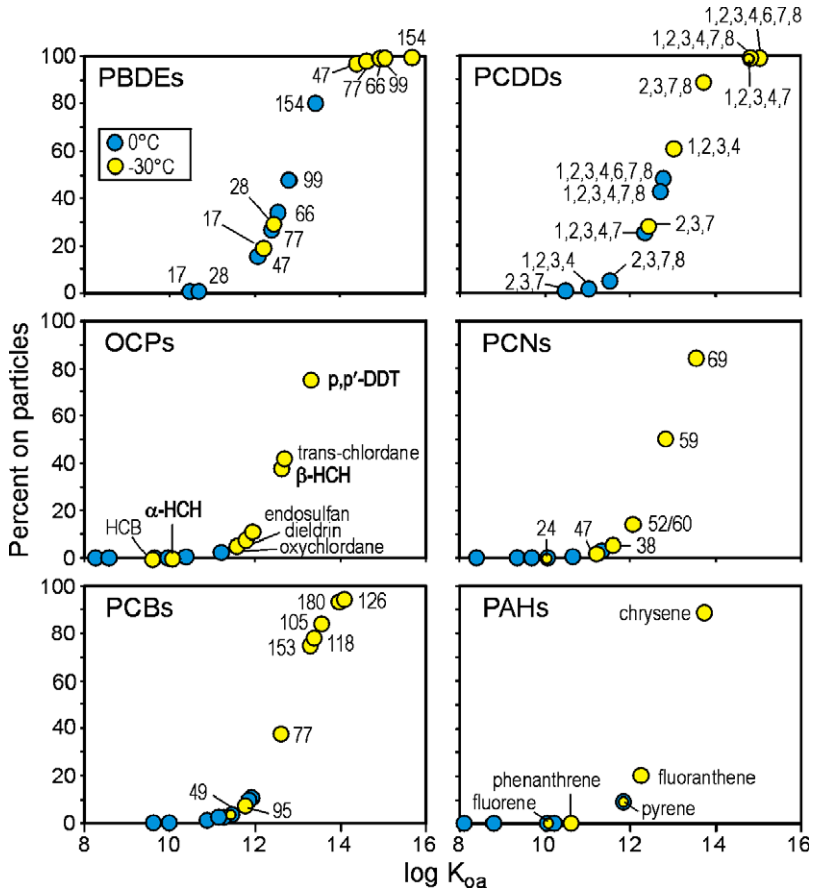

Fig. 28. The partitioning of selected POPs as a function of K_{OA}. Shown are the changes in partitioning that occur between winter (−30 °C; solid circle) and summer (0 °C; open circles). Particulate fractions were calculated based on equations developed by Finizio et al. (1997) from field measurements for organochlorines. Calculations are based on measurements of particulate organic carbon in Alert air during winter and summer 1998–1999 (Sharma et al., 2002). Temperature-adjusted K_{OA} values were taken from the literature (Harner and Bidleman, 1996, 1998; Harner et al., 2000a; Harner and Shoeib, 2002; Shoeib and Harner, 2002).

arctic haze in spring may alter substantially if warming comes earlier in the season with consequent change to the particle–gas partitioning. Several higher molecular weight polybrominated diphenyl ethers (PBDEs) and polychlorinated dibenzo-*p*-dioxins (PCDDs) are appreciably associated with particles even at 0 °C. Depending on their susceptibility to photolysis, particle-association may actually protect these compounds during transport which might explain why the increase in PBDEs in the Arctic closely follows the production and usage to the south (Ikonomou et al., 2002)—something that was not true for the PCBs and other organochlorine compounds which are mostly in the gas-phase during summer. Despite cold winter temperatures many PCB, OCP and PCN compounds remain in the gas phase. Winter warming projected by climate models may therefore facilitate their transport during the period of year when reduced sunlight makes them less vulnerable to degradation.

Chemical partitioning between environmental media (air, water, soil, biota) can be described using only three partition coefficients—K_{OA}, K_{AW}, and K_{OW} Gouin et al., 2000). K_{OW}, which is a ratio of two solubilities that both increase with temperature, tends to vary weakly with temperature as shown by the modest 20% increase for HCB in response to a 5 °C increase (Fig. 29, Bahadur et al., 1997). A 5 °C

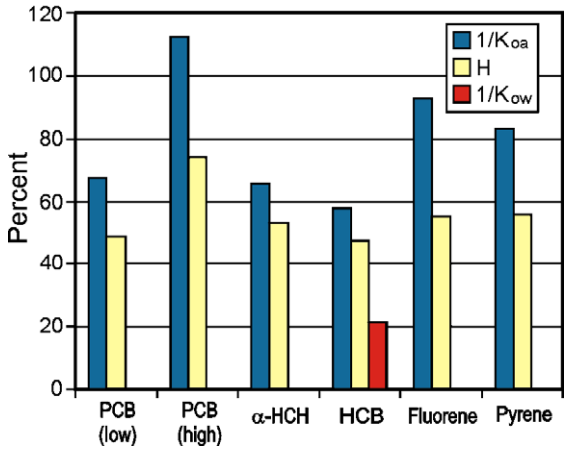

Fig. 29. Predicted percent changes in H and $1/K_{OA}$ associated with a 5 °C increase in temperature for selected chemicals. "High" and "low" represent the upper and lower values reported for PCBs. Values are based on ΔH_H (enthalpy of phase change associated with transfer from water to air, kJ mol^{-1}), ΔH_{OA} (enthalpy of phase change from octanol to air), and ΔH_{OW} (enthalpy of phase change from octanol to water: see Table A.1.1. in Harner et al., 2003).

temperature rise produces a more substantive 60–100% increase in $1/K_{OA}$, which will be manifest as an increase in volatility and greater potential for atmospheric transport. Although this is not as dramatic as the winter/summer differences for partitioning to aerosols (Fig. 27), it would result in more chemical partitioning out of surface soils and aerosols to enter the gas phase. If global warming occurs, cycling of chemicals through the atmosphere will increase. On the other hand, models suggest that temperature contrast between equator and pole will decrease (Zwiers, 2002). Accordingly, kinetic processes will increase with temperature rise but the overall thermodynamic forcing toward polar regions will decrease with reduced global thermal contrast. K_{AW} increases significantly (40–70%) which would favour evasion from surface waters. This would be particularly important for OCs like the hexachlorocyclohexanes (HCHs) which are at, or over, saturation in arctic waters. The predicted increase in K_{AW} associated with a 5 °C increase in water temperature corresponds to tonnes of HCHs that could be forced back into the atmosphere each year. We note that temperature increases of 5 °C will not likely apply to partially ice-covered regions where temperatures will be buffered by melting ice. Nevertheless, under recent changes in the AO index (Fig. 6a,b) and with change projected by models (Figs. 4 and 5), a large number of lakes and major areas of arctic shelves could be so impacted.

Loss of chemicals occurs during transport in the atmosphere through reactions with hydroxyl (OH) radical, nitrate radical (NO_3) or ozone (O_3), through photolytic oxidation and through sorptive partitioning to other phases (e.g. aerosols, precipitation, vegetation) with subsequent deposition.

Although photolytic reactions do not have a strong dependence on temperature they will be affected by cloud cover which is predicted to increase with global warming (IPCC, 1995). Increased cloud cover will also result in lower OH radical concentrations and less chemical removed by this and other photolytic pathways.

The dominant removal processes in soil and water include hydrolysis, photolysis, redox reactions, microbial degradation and removal through soil–surface–air partitioning. Of these, only photolysis is not strongly dependent on temperature. The influence of temperature on the rate constant, k, is usually described using the Arrhenius expression,

$$k = A\mathrm{e}^{-E_a/RT} \quad (1)$$

where A is a constant and E_a is the activation energy. Based on hydrolysis activation energies of 78–85 kJ mol^{-1} (Ngabe et al., 1993) for α-HCH and γ-HCH, respectively, a 5 C° increase would increase removal rates by ~85–95%. The increase would be even greater if we consider that the dissociation constant for water (K_w) (e.g. pH=8) increases with temperature resulting in more OH^- ions. Activation energies associated with redox reactions are not reported for OCs but are usually assumed to be about 50 kJ mol^{-1} (Tratnyek and Macalady, 2000) which imply about a 50% increase in reaction rate with a 5 °C rise.

Microbial degradation also follows the Arrhenius equation, but few studies report E_a. As a general rule, the biological activity in the mesophilic range (5–35 °C) doubles for every 10–15 °C temperature rise which implies an E_a of 30–45 kJ mol^{-1} (Viessman and Hammer, 1985). Arctic microbial populations exhibit a large diversity and abundance (Ravenschlag et al., 2001; Sahm and Berninger, 1998) and are

typically cold-adapted, able to maintain efficient rates of organic degradation and mineralization down to the freezing point of seawater (−2 °C) (Arnosti, 1998; Sagemann et al., 1998). Indeed, even the extreme environments of brine pockets in ice, where liquid phases are maintained at temperatures well below zero, appear to harbor microbial activity (Krembs et al., 2002). These facts suggest that warming may be accompanied by adaptation or population change but not necessarily that microbial degradation rates will increase. For example, reduced degradation of methyl dichlorprop was observed in experiments where field plots were warmed 5 °C above normal for several years (Peterjohn, 1994).

Harner et al. (1999, 2000b) found in situ microbial removal rates for α- and γ-HCH in cold Arctic Ocean waters to be surprisingly fast ($t_{1/2}$ for (+)α-HCH (5.9 years); (−)α-HCH (22.8 years); γ-HCH (18.8 years)). Assuming an E_a of 50 kJ mol^{-1}, a 2–5 °C temperature rise in the upper Arctic Ocean would imply a reduction of these half lives by 20–50%. Microbial degradation was estimated to account for over 30% of the removal of HCH (Harner et al., 2000b) suggesting that a small temperature rise could push this proportion to over 50%, reducing exported of HCH to the Archipelago or the Atlantic Ocean by tens of tons per year.

Alterations in environmental characteristics other than temperature (e.g. soil moisture, soil and water pH, nutrient levels, vegetation cover and type) will be tied to global warming and impact the composition and density of microbial populations. For instance Lewis et al. (1999) found differences in microbial preference for microbes inhabiting forested versus pasture soils. They also showed that the enantioselectivity (preference of the microbial population for a + or − enantiomer of a chiral compound (compound exhibiting mirror-image forms)) shifted with organic nutrient enrichments. Other studies have observed high rates of microbial degradation of HCHs in arctic lakes (Law et al., 2001) and watersheds (Helm et al., 2000). Law et al. (2001) found that enantioselective degradation of α-HCH was greater in small, high arctic lakes and streams compared to temperate lakes and wetlands. They concluded that low nutrient levels in the arctic systems resulted in an adapted microbial population that was more capable of degrading organic contaminants. Thus biodegradation of chemical residues in soil and water will be altered as microbial populations adapt to the changing climate. The complexity and uncertainty associated with these changes however, does not allow us to predict whether global warming will enhance or diminish chemical removal by this pathway.

Putting many of the concepts outlined above into a numerical model, McKone et al. (1996) asked specifically what would be the effect of a 5 °C temperature rise on the health risk from hexachlorobenzene (HCB) in a temperate location. They found surprisingly little consequence from the projected 5 °C temperature increase. Indeed, their results showed that warming would actually reduce exposure to this compound for aquatic organisms because it would enhance degradation and tend to force HCB out of water and into air. The critical step, controlled by the sensitivity of air–water partitioning to temperature, acts at the bottom of aquatic food webs which through biomagnification provides most of the health risk.

6.3.5. The effect of altering food web structure on organochlorines

Biomagnification can concentrate fat-soluble compounds, like the organochlorines, by factors as high as 10^5 to 10^9 from water to apex predator (Fisk et al., 2001a,b; Kidd et al., 1995b; Muir et al., 1999; Muir and Norstrom, 1994). The distribution of contaminants in air, water, and for the first step in the food web (phytoplankton, particulate organic carbon), can be predicted simply by applying appropriate partition coefficients like K_{AW} and K_{OW} (e.g., see Wania, 2003). Chemical partitioning, which is based solely on thermodynamics, provides a crucial platform upon which biomagnification can then operate (Fig. 30). A chemical at equilibrium will have identical fugacity in the media in question (e.g., air, water, oil), making it relatively simple to predict how temperature will alter the distribution as discussed above.

Biomagnification, however, cannot be explained solely by thermodynamics and because it involves energetic processes to produce the elevated concentrations in the top predators. These processes can be considered in part as a reduction of the solvent containing organochlorines (fat) through metabolism. The processes are complex and can lead to variability simply due to bio-energetics for which the offloading of contaminants by nursing mammals is a

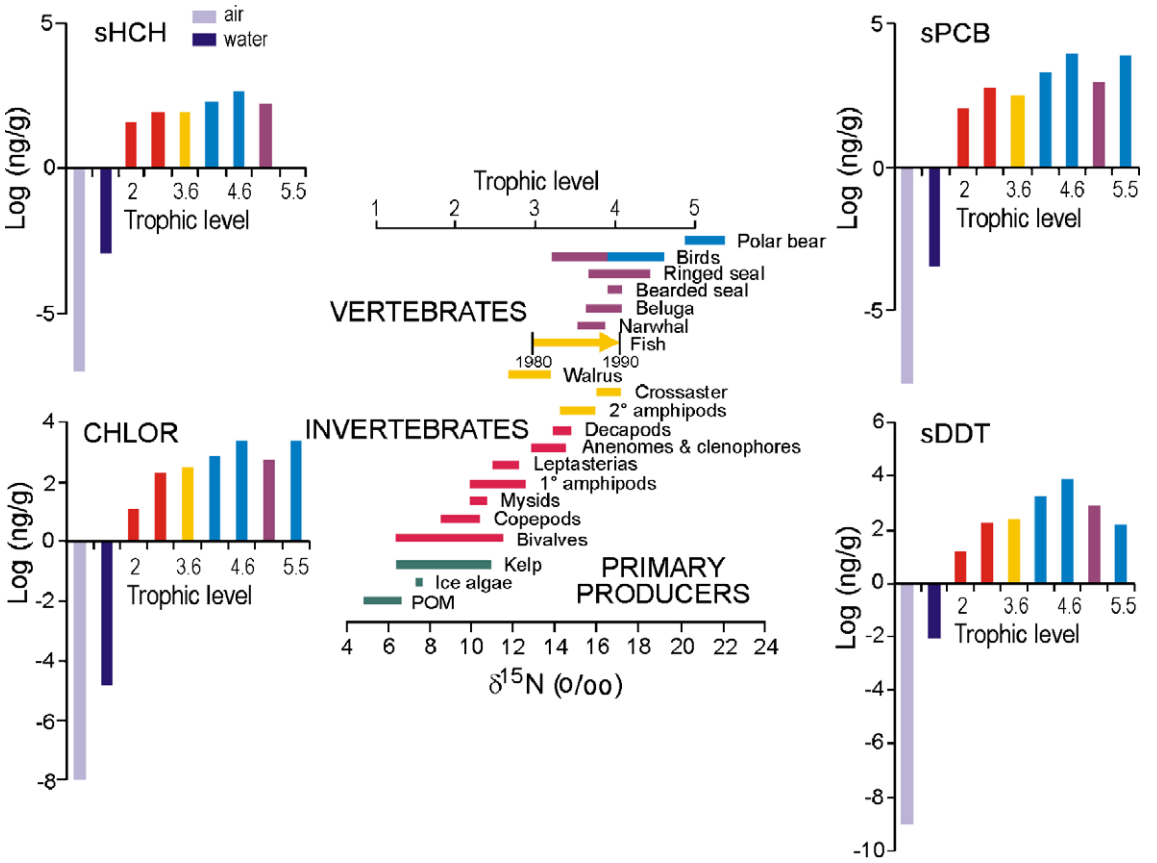

Fig. 30. The marine food web structure observed during the Northwater Project. Shown in the panels are average concentrations of selected organochlorine compounds in air, water, and various trophic levels as represented by *Calanus hyperboreus* (2), *Themisto libellula* (2.6), Arctic Cod (3.6), Black guillemot (4.3), glaucous gull (4.6), Ringed Seal (4.5), Polar Bear (male, 5.5) (see Fisk et al., 2001a,b; Hobson and Welch, 1992).

classic example (Addison and Smith, 1998). It is not our intent to conduct a thorough review of bioaccumulation and biomagnification except to admit that the complexity of the process offers the opportunity for climate change to act in subtle ways. The data in the panels of Fig. 30 show an almost linear relationship between the log (contaminant burden) and trophic level. The slope of this relationship indicates the multiplication factor involved for each step in trophic level so that, for example, one step would multiply DDT concentration by perhaps as much as 6 (neglecting polar bears) whereas the other contaminants experience factors of 2 (HCH), 3 (PCB) and 4 (chlordane). The removal or addition of trophic levels in the food web mediated by climate change, therefore, will not have the same effect for all contaminants; for example, Fig. 30 suggests that DDT will be the most sensitive to this kind of change.

Bifurcation is another way in which food web structure can effect change. For instance, altering the coupling between pelagic and ice primary production and the benthos can change the relative proportions of organic carbon (and contaminant) that enter pelagic or benthic food webs. However, these changes in pathways do not alter the relationship between contaminant concentration and trophic level. In this context it can be seen that walrus feeding on a benthos enriched by strong coupling with primary production will expose them to lower organochlorine concentrations by factors of 10 or more than if they switch to predation on seals.

6.3.6. The epontic food web and changes in ice climate

The entry of contaminants to a stratifying surface layer from ice melt in spring offers a mechanism vulnerable to climate change. However, studies seem not to show higher concentrations in epontic fauna than in zooplankton (Borgå et al., 2002). Organochlorine concentrations in epontic amphipods (*Apherusa glacialis*, *Gammarus wilkitzkii*, *Onisimus* spp.) and zooplankton (*Calanus hyperboreus*, *Thysanoessa inermis*, *Parathemisto libellula*, *Chaetognatha*) from the marginal ice zone near Svalbard could be explained mostly by diet, with habitat (sea-ice underside versus the pelagic zone) accounting for a smaller part of the variance. Epontic amphipods had higher concentrations of HCB, γ- and α-HCH while DDTs, PCBs and chlordanes did not differ between epontic and pelagic habitats. This pattern of uptake can be explained by the vertical distribution of OCs in the water column. Higher concentrations of HCHs and HCB are found near the sea surface (Harner et al., 1999; Jantunen and Bidleman, 1998; Macdonald et al., 2000a; Tanabe and Tatsukawa, 1983) whereas particle reactive compounds like DDTs and highly chlorinated PCBs adsorb to sinking particles to produce a more homogenous vertical distribution (Tanabe and Tatsukawa, 1983). Furthermore, most particles transported by sea ice are not available to epontic biota because they are released in the marginal ice zone when the ice melts (Ramseier et al., 1999) and descend rapidly, carrying adsorbed contaminants with them. For OCs that exhibit strong gradients in the upper ocean, the loss of ice and hence of epontic fauna can alter the dietary exposure of higher trophic levels like seabirds and seals.

6.3.7. Food deprivation or shifts in diet

Many of the Arctic's top predators undergo periods of fasting forced by lack of food, seasonality of food, inability to access food or periods when foraging does not occur due to, for example, spawning migrations. Perhaps the best documented example of fasting due to climate variation is the stress to the Hudson Bay polar bear population deprived of their ability to hunt seals during spring due to change in spring ice climate (Stirling, 2002; Stirling and Lunn, 1997; Stirling et al., 1999). The consequent burning of stored fat through metabolism results in release of archived fat-soluble contaminants and, potentially, an increase of contaminant burden in the remaining fat reservoir. Longer periods of starvation due to change in ice or change in prey populations lead to higher doses of OCs released from fat. In this way, starvation and contamination work together to stress the animal. Although the concern with nourishment-deprived polar bears has received much attention, similar circumstances probably apply to other species, examples including common eider (*Somateria mollissima*, Olafsdottir et al., 1998), arctic charr (AMAP, 2005) and migratory salmon (Ewald et al., 1998; Krümmel et al., 2003).

Species that have dietary flexibility may respond to ecosystem change by switching to alternate prey, again with consequences to their organochlorine intake. For example, the large variation in organochlorine concentration in the livers of glaucous gulls from the western Barents Sea reflects wide range in the gull's prey trophic level as reflected by parallel variations in $\delta^{15}N$ as a proxy for trophism (AMAP, 2005). Recent decreases in organochlorine concentrations for Svalbard Minke whales (*Balaenoptera acutorostrata*) might superficially be ascribed to banning of PCB manufacture during the 1970s. However, it seems just as likely that this decline reflects a dietary switch from capelin, whose stocks collapsed in 1992–1993, to krill which are further down the foodweb (AMAP, 2005). Polar bears also display a range in prey that can explain regional variation in organochlorine burdens. For example, Chukchi and Bering Seas bears feed more heavily on Pacific walrus which are less contaminated than ringed seals because they are at a lower trophic level, whereas bears from Svalbard feed on more heavily contaminated harp seals. These observations indicate that climate variables expressed through prey availability and biological condition have much influence on apex feeders exposure to organochlorines.

6.3.8. Altered migration pathways, invading species and animal behaviour

Migratory species (whales, fish, birds) can obtain contaminant loadings in one location and release them in another, or migrating animals can be subject to varied exposure as they feed along their migration paths. The degree of PCB contamination in Alaskan lakes can be predicted from the strength of the

anadromous fish returning expressed on a per hectare basis; for those lakes receiving the largest sockeye (*Oncorhynchus nerka*) returns, PCB fluxes and concentrations in sediments are as much as eight times higher than in lakes receiving only atmospheric input (Krümmel et al., 2003). Furthermore, the contaminants returned to these lakes by spawning fish then enter the lake's aquatic foodweb (Ewald et al., 1998), where they may provide a risk to salmon recruitment. Recent change involving new–or more intensive–migration of Pacific salmon into arctic rivers (Babaluk et al., 2000) could, likewise, have an impact on contaminant budgets for rivers or lakes they enter. Certainly, these fish provide a new 'vector' for delivering contaminants to species that feed on them or depend on a food web supplied by their carcasses. Nor need contaminant biotransport from oceans to lakes be restricted to fish. On Bear Island, Evenset et al. (2004) have made a convincing case that birds can provide a significant pathway through their guano. Here, two lakes were compared; the one favoured by seabirds had higher trophic levels (δ^{15}N) and contaminant burdens due to the delivery of trophically higher guano—a product of ocean feeding by the birds.

If the spatial distribution of contaminants is controlled by processes subject to climate change, then exposure during population migrations can also alter through climate change. An intriguing example of such a process has recently been described for the Bering/Beaufort bowhead whale migration. The whales reflect in their body burdens the change in α- and β-HCH composition between the Bering and Beaufort Seas (Hoekstra et al., 2002). In turn, the ocean composition for the HCHs is likely controlled by large-scale physical processes (e.g., rainfall and air–sea partitioning—see Li et al., 2002) as are migratory routes (Dyke et al., 1996b; Moore et al., 1995) each which is sensitive to climate change. Because bowhead whales tend to follow the ice edge in their migrations, changes already witnessed in the location of the ice (Fig. 13) likely have contributed to changes in contaminant uptake by these animals. Likewise, the recent response of gray whales (*Eschrichtius robustus*) to insufficient benthic feedstock in the Bering Sea, probably a consequence of climate variation, has led them to forage benthos in the Chukchi Sea (Moore et al., 2003) and, therefore, become an 'arctic' animal for part of their diet, withdrawing such contaminants back into the Pacific Ocean.

Invasions of new species or extensions of ranges fostered by climate change or by introduced exotic species both have the potential to re-structure food webs. For example, the recent introduction of rainbow smelt (*Osmerus mordax*) into Hudson Bay's coastal river systems (Franzin et al., 1994; Remnant et al., 1997) and the subsequent expansion of their range provides a mechanism to alter foodwebs either directly or indirectly through damage to coastal fisheries which then changes predation by humans.

An elegant example of how dramatic such change can be, both in trophic organization and contaminant pathways, was provided by the invasion of the zebra mussel into the Great Lakes (Morrison et al., 1998, 2000; Whittle et al., 2000). Although the widespread invasion of the zebra mussel into arctic freshwaters may appear unlikely, this one example well illustrates how the extension of the range of a single species could have unexpected impact on contaminant cycling in lakes (or oceans). Anadromous and freshwater fish distributions are sensitive to climate variables with temperature acting as a first-order control (Wrona et al., 2005). To discuss change, distributions of fish have been roughly characterized as an *Arctic Guild*, a *Coldwater Guild* and a *Coolwater Guild*, each of which contain characteristic species with varying ranges of temperature tolerance. Climate change–especially change in temperature regime–therefore, offers the potential to alter boundaries of tolerance and so shift species distribution. Although we can predict such changes are likely to occur on the heels of temperature shifts, the understanding of biological systems is not presently up to the task of accurate prediction of that change or its timing.

Sublethal concentrations of contaminants may affect animal behaviour in important, unappreciated, ways which then have climate ramifications. For example, Atlantic salmon underyearlings exposed to low doses of DDT shifted their preference for water temperature downward whereas fish exposed to high doses shifted upward (Ogilvie and Anderson, 1965). It is effects such as these that hold the greatest potential for surprises when, for example, climate change produces altered thermal regimes in aquatic systems lightly impacted by endocrine-disrupting chemicals.

6.3.9. Organochlorines, disease and epidemics

During the past decade or so, there has emerged much evidence that mass mortality in marine mammals may occur due to a combination of factors including disease vectors, population stress and contaminants, each of which may be affected by climate change (cf. Lavigne and Schmitz, 1990; Ross, 2002). The complexity of this interaction provides fertile ground for surprises. Some disease outbreaks have been observed following migrations associated with large-scale ecological change and some have derived from the introduction of viruses from domestic animals. But it is the addition of immunotoxic chemicals, like many of the POPS, that may provide the trigger for disease to emerge (Ross et al., 2000; Vos and Luster, 1989). The widespread distribution of canine distemper virus or a closely related morbillivirus in seals from Greenland, led Dietz et al. (1989) to speculate on the possibility that large-scale migration of harp seals from the Barents Sea to north Europe in 1986–1987 might have provided a disease vector. The co-factors of a naïve marine mammal (seal) population in coastal Europe manifesting suppressed immune systems through high contaminant PCB burdens would then have provided the foundation for an epidemic (Heide-Jørgensen et al., 1992).

Within the Arctic, top predators would be at greatest risk due to their high exposure to contaminants, and marine mammals likely face the added stress of change in ice climate. Accordingly, indications of immunosuppression have been found in polar bears, northern fur seals and glaucous gulls (AMAP, 2005). In particular, the polar bears of the Kara Sea, Franz Jozef Land, East Greenland and Svalbard would seem especially vulnerable. Firstly, they exhibit inordinately high contaminant burdens (Andersen et al., 2001; Norstrom et al., 1998) and these high burdens may very well derive partly from the enhanced connectivity between this region and Europe/North America under the high AO/NAO indices of the 1990s (Fig. 8). Secondly, as discussed in previous sections, change in ice climate and in marine ecosystems may have provided the added stress of malnourishment. Finally, it seems these bears already have sufficient contaminant burdens to exhibit health effects (Bernhoft et al., 2000; Skaare et al., 2001).

6.4. Hydrocarbons

Contaminant hydrocarbons in the Arctic derive from combustion and petrogenic sources (Yunker et al., 1995). The pathways of these two sources of hydrocarbon differ substantially as will their sensitivity to climate change. Contaminant hydrocarbons pose two kinds of problems; polynuclear aromatic hydrocarbons (PAHs) and their oxidation products are toxic (Zedeck, 1980) and spilled oil has direct, well known impacts on biota, especially those like seals, birds and benthos which inhabit surfaces (Patin, 1999; Wolfe et al., 1994).

6.4.1. Combustion PAHs

Combustion PAHs are well-known products of natural fires and combustion or high-temperature processes related to human activities (automobiles, liquid and solid fuel burning, waste incineration, metallurgy). In the atmosphere, PAHs partition between the vapor phase and particulates (see for example Figs. 14 and 15 in Macdonald et al., 2000a) and are transported long distances to be detected at remote arctic locations (Halsall et al., 1997; Macdonald et al., 2000a; Patton et al., 1991; Yunker and Macdonald, 1995). There is a strong seasonality in PAH concentration in air at Alert with colder months (October–April) displaying concentrations about 10 times higher than warmer months (May–September). This suggests that the arctic winter haze phenomenon, which transports heavy metals across the pole from Eurasia, similarly transports industrial combustion products. Therefore, much of what has been said about aerosol metals and climate change relates directly to PAH; altered wind patterns (Fig. 8) and enhanced precipitation (Fig. 9) have the potential to change pathways and to deposit PAH aerosols over parts of the Arctic Ocean especially toward the southern Eurasian Basin. Furthermore, temperature increases may shift the equilibrium from particulate to vapor phase for PAHs like pyrene, fluoranthene, phenathrene and anthracene which at arctic temperatures are partially distributed between air and solid phase (see Fig. 28 and similar Figs. 14 and 15 in Macdonald et al., 2000a).

In addition to the strong seasonal signal of industrial PAH observed in the Arctic, outliers (samples with abnormally high PAH concentrations) are also

observed during summer months, particularly at Tagish, and these have been assigned to forest fires (Macdonald et al., 2000a). Forest fires are projected to increase through climate change as a result of warmer continental temperatures (Fig. 3a) and less precipitation in continental interiors. We may expect, therefore, a general increase in atmospheric PAH in the Arctic deriving from such biomass burning, and this increase will likely have a greater impact on small rivers in northern Canada which already receive almost all of their PAH from combustion sources (Yunker et al., 2002). Loss of permafrost and enhanced erosion of peat may further contribute enhanced amounts of relict PAH to lakes and rivers (Yunker et al., 1993).

6.4.2. Petrogenic hydrocarbons and oil

The risk of oil spills from offshore exploration and production is a leading concern for the Arctic (Bakke et al., 1998; Patin, 1999). Climate change that produces an ocean margin substantially clear of ice will undoubtedly encourage further exploration, perhaps in more remote locations. Producing oil from shelves, especially remote shelves, has the associated problem of transporting produced oil south either by ship or pipeline.

It is clear that changes in the ice drift associated with changes in the AO index will have a dramatic influence on where spilled oil will go if it gets into the ice pack (Fig. 31). In the Canadian and Alaskan sector of the Arctic Ocean during AO^- conditions, spilled oil will follow ice into the East Siberian Sea to traverse the Russian shelves and thence exit to Greenland Sea. During AO^+ conditions, oil spilled from the same location would tend to remain within the Beaufort Gyre potentially to return within a few years from whence it was spilled.

Oil spilled over the Russian shelves, or entering their coastal seas from spills into rivers, would tend

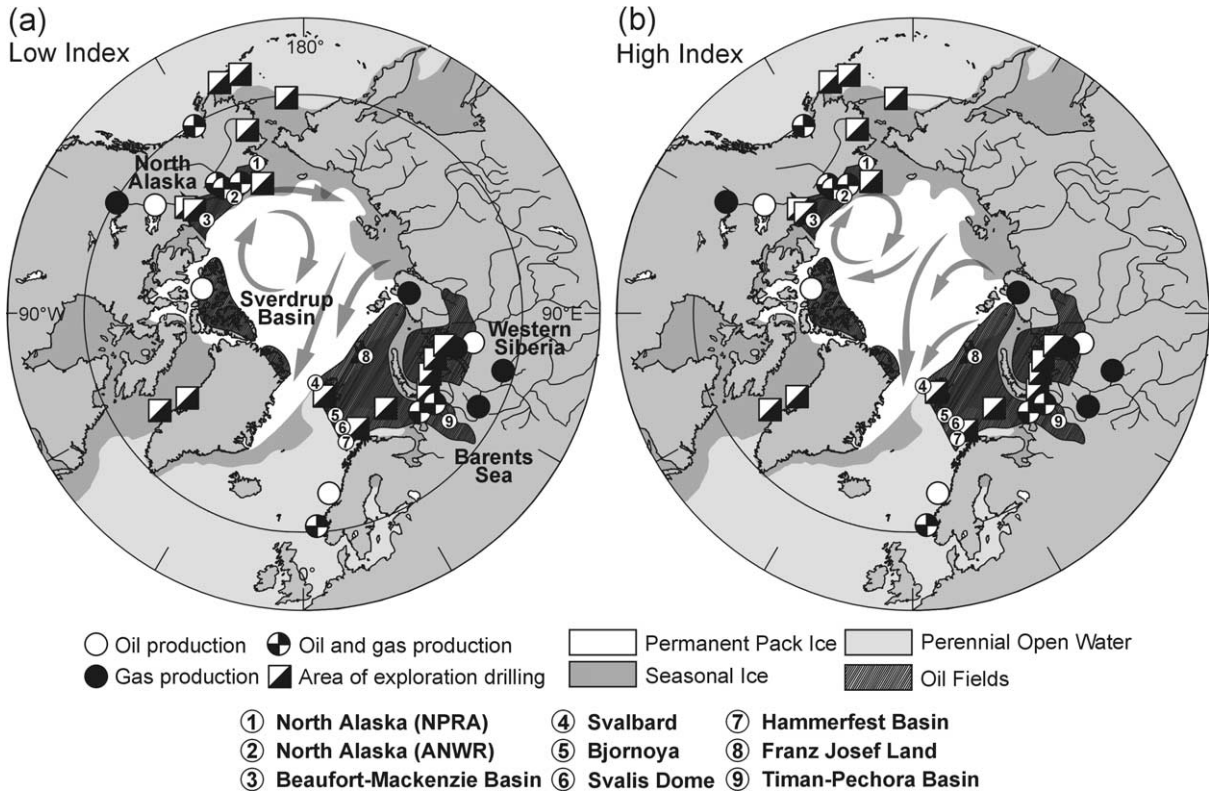

Fig. 31. A map of the Arctic showing oil-bearing regions and locations of oil production and pipelines together with the ice-drift field under (a) Low Arctic Oscillation index and (b) High Arctic Oscillation index (ice motion based on Rigor et al., 2002 and oil data are from Bakke et al., 1998).

to track directly across the Arctic under AO^- conditions (Fig. 31a). However, oil spilled under AO^+ conditions could move more to the east, with a slight chance of getting into the Canada Basin and thence arriving in the Canadian Archipelago through which it would then have to pass. It is uncertain how viable this latter route might be for oil spilled in the Kara or Laptev Seas, but evidence from tree dendrology suggests that there have been periods since the Holocene when communication from Siberia to the Archipelago has occurred, mediated by ice drift (Dyke and Savelle, 2000; Dyke et al., 1997).

A final connection between spilled oil and climate change derives from the projected increase in incident UV radiation (Weatherhead and Morseth, 1998) which could lead to an increase in photo-enhanced toxicity of spilled oil (see, for example Barron and Ka'aihue, 2001; Pelletier et al., 1997). Toxicological assessments of oil made in the presence of UV light reveal a toxicity up to 1000 times greater than that measured under the traditional fluorescent light. Furthermore, photoenhanced toxicity of oil can occur at the intensities and wavelengths measured for UV in aquatic water columns suggesting that increased incident UV radiation projected for polar regions would, in addition to many other effects on ecosystems (Weatherhead and Morseth, 1998), enhance damage done by spilled oil.

7. Time series

The value of time series is undisputed both for climate-related variables (Hare and Mantua, 2000; McGowan, 1990) and for contaminants (AMAP, 1998). However, recognition of the potential of climate variables to produce variance in contaminant time-series has all but been neglected (Macdonald et al., 2002a). We have discussed numerous examples of how global change can alter delivery of contaminants to and within the Arctic, alteration in wind fields and precipitation forced by the Arctic Oscillation being but one simple example. The fact that the leading 'global distillate,' water, provides one of the clearest detectors of global temperature change in its isotopic composition (Fischer et al., 1998) should generate considerable anxiety about time series of volatile and semi-volatile compounds even if the pathway between emission and point of measurement is reasonably direct (e.g., atmospheric concentrations, ice cores, sediment cores). Examples closer to contaminant time series can be found. For instance, the mercury record in ancient Antarctic ice has been suggested as a proxy for ocean productivity (Vandal et al., 1993) and cadmium has been applied to paleo ocean-productivity histories (Saager and deBaar, 1993; Shen et al., 1987).

The environment can be monitored at many points (Fig. 32) each of which will tell a separate story; together, these stories might tell us much not only about contaminant cycling but about the impact of climate change on that cycling. For example, we might measure PCB concentration: in air at Alert every 2 weeks; in a dated sediment core or ice core; in biota or surface sediments collected every 5 years; or in an ocean profile collected annually. What these hypothetical PCB time-series data will tell us depends on how many environmental processes have an opportunity to operate on the original signal (the emission), and how the recorder (the medium being monitored) itself actually works. Much research has been conducted to understand and account for the latter (e.g., organochlorine cycles in lactating female mammals; increase in mercury with age of fish, diagenesis in sediment cores) but the difficulty of climate variability creeping into the record remains virtually ignored. Climate variability by itself may cause aliasing, which arises when sampling intervals are chosen close to natural variation frequencies. For example, the Arctic Oscillation's time scale of 5–7 years (Proshutinsky and Johnson, 1997) would make it very difficult to assess the role of such sub-decadal variation in time series whose resolution is of the order of five years. Furthermore, a trend from a time series collected from say 1980 to 1995 will potentially carry a large bias, either positive or negative, produced by a switch in the middle of the record from generally AO^- to strong AO^+ conditions (Fig. 6, top panels). The importance of such bias would be impossible to evaluate unless time series were extended to include several AO phases. Climate change often may provide an alternate hypothesis for time-series contaminant data. We describe briefly below a few examples.

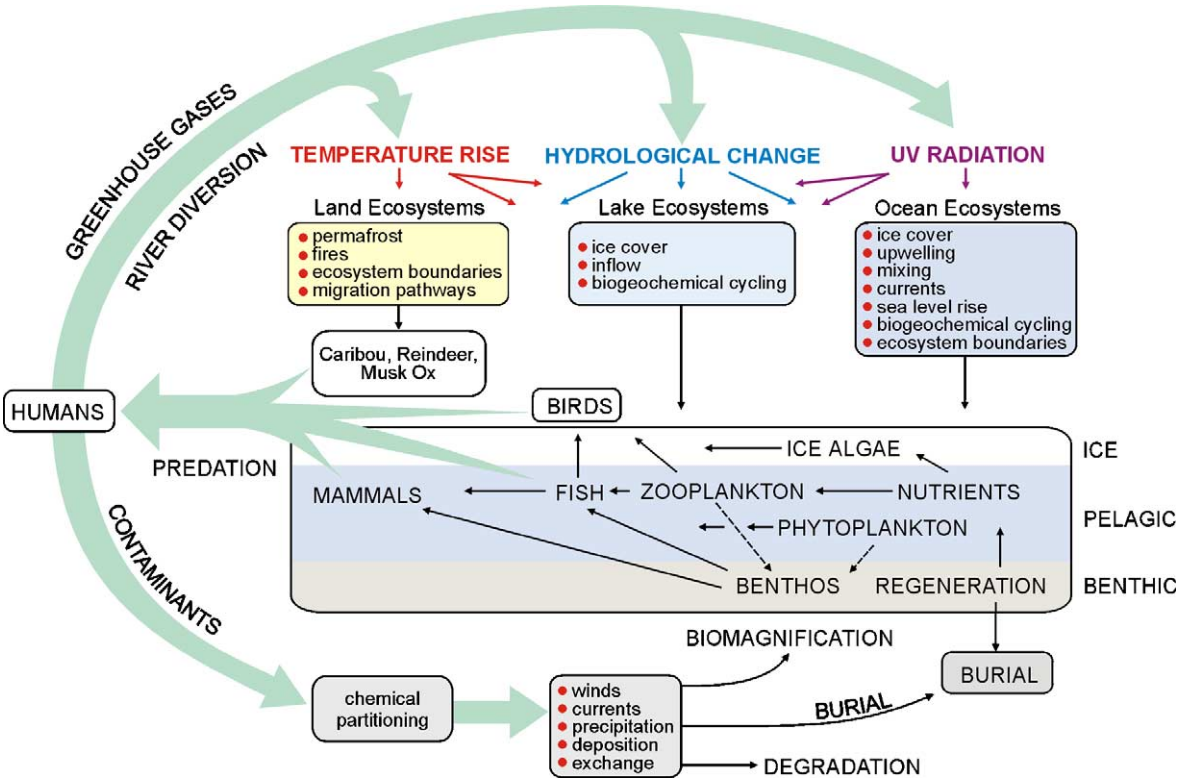

Fig. 32. A schematic diagram showing components of the pathways contaminants must traverse between emission and points at which environmental time series are collected for the Arctic.

7.1. Sediment-core records and surface sediments

Well-dated sediment cores from lakes and oceans provide a traditional way to estimate contaminant fluxes and trends (Lockhart et al., 1995, 1998; Muir et al., 1996). Fluxes to sediments estimated from a sediment core are sometimes corrected for focusing to produce an estimate of flux to the surface of the water in order to compare it with fluxes at other sites or with emissions. Increases or decreases with depth in a sediment core are then often used to infer historical changes in emissions. However, between the emission and the sedimentation at the bottom of the lake or ocean lie atmospheric transport, deposition to the water's surface or to the drainage basin, attachment to particles, and settling to accumulate at the bottom (Fig. 32). These physical pathways may in some cases be augmented by biotransport (Evenset et al., 2004; Ewald et al., 1998; Krümmel et al., 2003). Climate change may operate within this system to (1) change wind fields (important if there are atmospheric gradients); (2) change the efficiency of the air to ground transport (e.g., by altering precipitation or temperature); (3) change the efficiency of capture to sediments (by processes outlined in Section 6.3.3); (4) change the strength of fish or bird migration; or (5) change the diet of migratory species.

The case of biotransport by bird guano, described by Evenset et al. (2004) for a small lake on Bear Island near Spitsbergen, provides an instructive example of how climate-related variables could interact with contaminant records in sediments. What would a dated sediment core record look like from a lake receiving contaminants directly from the atmosphere and indirectly from bird guano? It is clear that the record would contain two components of flux, one relating to source regions (emissions) in the context of downwind transport and the other relating to the biological vector. The atmospheric flux component would respond partly to emission

strength and partly to the transport path which includes wind fields and precipitation. Given the changes that can occur with the AO/NAO, for Bear Island we might see the relative importance of American and European sources vary (Fig. 8) and the rate of capture through precipitation vary (Fig. 9). For the biotransport component, we have the added complication of the population dynamics of birds. This will depend on the actual number of birds, their nesting locations, the source and trophic level of food which derives from the Barents and Nordic Seas (AMAP, 2005) and on the contaminant burdens in the upper ocean from the foraging domain around Bear Island; all of these are subject to climate variation. This example, which is particularly compelling in the way local and perhaps regional PCB patterns have been impacted by biological sources (guano—AMAP, 2005; Enge et al., 1998; Evenset et al., 2004) is likely not alone. It is just that we have studied so few places with sufficient intensity. One is reminded of the importance of fish as transporting agents for organochlorines and mercury (Ewald et al., 1998; Krümmel et al., 2003; Zhang et al., 2001), which together with high and variable anadromous fish escapements (Finney et al., 2000, 2002), very likely imprints as yet uninvestigated contaminant records in coastal lakes of Alaska.

Finally, natural variability in organic carbon flux to sediments can enhance sediment foraging (including small infauna and large animals such as diving birds, walrus, seals and belugas). This, together with the potential for episodic colonization of sediments by new species under changing ocean climate (see for example Stull et al., 1986), provides a strong caution on using surface sediment contaminant distributions to infer spatial or temporal trends in contaminants even where normalizing factors such as aluminum or organic carbon have been applied.

7.2. Atmospheric time series

Data collections from ground stations in the Arctic are of relatively short duration, extending back a couple of decades for metals (Gong and Barrie, 2003; Sirois and Barrie, 1999), and only a few years for OCs (see for example Bailey et al., 2000; Halsall et al., 1998; Hung et al., 2001; Macdonald et al., 2000a; Stern et al., 1997). There are fewer processes between the emission sources and the recorder for these measurements (Fig. 32) but as discussed above, changes inherent in the Arctic Oscillation (winds, ice cover, precipitation) are sufficient to imprint themselves on the emission before it reaches a monitoring station. In this context, a very recent paper (Ma et al., 2004) has found correlation between large-scale atmospheric pressure fields (NAO, El Niño–Southern Oscillation (ENSO), Pacific North American (PNA) patterns) and air concentrations of POPs measured in the Great Lakes and Arctic regions. Although such correlations strongly imply that there is a large-scale variation in atmospheric forcing for contaminant transport, further work is required to reveal the underlying mechanism(s). For example, change in wind fields arising from sea-level pressure changes might provide the simplest explanation. However, temperature, precipitation distribution and form of precipitation (snow/rain) also vary with the state of these indices (e.g., see Fig. 9) and it is therefore possible that correlation is due in some part to change in upstream scavenging (precipitation) or in revolatilization from contaminated soils (temperature). Furthermore, it is crucial to distinguish between ongoing emissions and re-emissions from environmental reservoirs that have been loaded with POPs during the past several decades. These two sources of contaminants to the atmosphere respond differently to climate variables and will have different implications regarding compliance with international regulations.

7.3. Biological tissue time series

Marine and terrestrial biota have been collected, usually at very sparse intervals (typically 3–6 periods over a couple of decades), to monitor bioaccumulating substances, especially the organochlorines and mercury (Addison and Smith, 1998; AMAP, 1998; Braune et al., 1999; Wagemann et al., 1995). α-HCH concentration in seals collected at Holman (bars in Fig. 33) clearly do not follow atmospheric emissions (line in Fig. 33) nor do they follow arctic air concentrations which mimic emissions quite closely (see Fig. 9 in Macdonald et al., 2000a). Instead, they approximately follow Canada Basin surface water concentrations, which makes sense given that seals obtain their HCH burden from a marine diet. The burden of α-HCH is large in surface

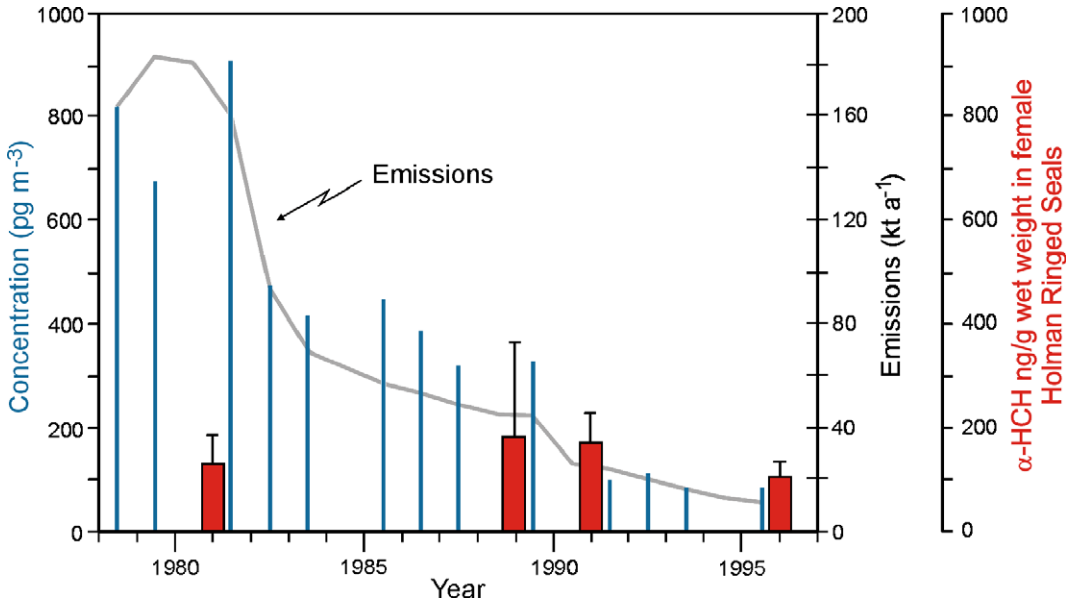

Fig. 33. A comparison of various HCH time series. Shown are (1) global emissions of α-HCH (the curve), (2) mean concentrations of α-HCH in Arctic air (the narrow blue bars) from 1979 to 1996 and (3) α-HCH concentration in Holman Island seals (thick red bars). Air concentration data of α-HCH in the Arctic have been measured at different stations by several research groups (for data sources see Addison and Smith, 1998; Li et al., 2002). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

waters of the Canada Basin (estimated at 1750 tonnes in the early 1990s, Macdonald et al., 2000a), and not quickly changed. However, the burden can be altered by diversion of Russian rivers or by removal of ice cover, both of which have occurred in the 1990s. Furthermore, change in the seal burdens can be caused by a regional change in the food web—something that has likely occurred as a consequence of changing ice cover and stratification (Melnikov et al., 2002). The food web factor might be 'controlled for' by monitoring trophic level through isotopic measurements (δ^{15}N) or by monitoring other components of the food web (e.g., arctic cod) over the same time interval. The advantage of monitoring species at high trophic levels–that they are very sensitive to OCs and that they are important to human diets–is somewhat offset by their sensitivity to variance from many factors other than emission strength (Fig. 32). Given the 5–7 year time scale inherent in change forced by the Arctic Oscillation, time series data for biological tissue will be vulnerable to an unaccounted aliasing. More insightful interpretations of biological time series would be possible if such records were matched by ocean contaminant concentrations.

8. Conclusions

Remarkable physical changes have occurred in the Arctic's climate during the 1990s. These changes have been manifest in wind and weather patterns, ice cover, ice thickness, ice drift patterns, permafrost, hydrology, ocean currents, precipitation and temperature patterns. Such changes were not recognized during previous assessments and their potential to alter pathways has not been adequately considered. Coherent studies of how these changes may have affected contaminant pathways have not been conducted but the evidence reviewed here convinces us that such changes have been occurring. Furthermore, change will continue to occur globally, especially within the Arctic, and inevitable surprises will inform us that our understanding of complex environmental systems remains woefully incomplete. Previous assessments have discussed physical and chemical transport processes separately from biological processes. In the equation of change, physical and biological systems interact with one another and both may have dramatic influence on the outcome for a contaminant headed for the Arctic. A contaminant's journey from its point

of emission to its accumulation in an arctic ecosystem is a complex sequence of many steps each of which can be altered by global change (Fig. 32). The risk of contaminants to arctic ecosystems cannot be fully understood by studying isolated components and no step can be considered more or less important than another. Rapid transport of a contaminant to the Arctic in air may or may not be followed by entry into foodwebs there.

The response time of natural systems is a key factor in establishing the role for a particular process in the equation of change (Ruddiman, 2000). Almost all contaminants exhibit a 'transient' emission profile that begins with a rapid rise followed by a dramatic decrease when bans or controls are instituted. Depending on environmental response times and reservoir sizes (e.g., atmosphere, ocean, soils, plants), the unloading stage for a given contaminant, following bans on its emission, is not a simple reverse of its loading stage. The atmosphere often predominates initially as a transporting medium, but with controls, other slower-moving or stationary media (water, soil, vegetation) may take over, especially if they comprise the largest reservoirs of contaminant in the environment. Models are crucial to understand contaminant transport, but many processes remain insufficiently understood to be included realistically. Furthermore, scenarios reflecting the true complexity of biogeochemical systems which include, for example, biotransport, remain some distance in the future.

We lack spatial and temporal time series. Many of the important climate variations occur at time scales from a few years to centuries with a dramatic shift associated with the Arctic Oscillation having occurred during the late 1980s. As a consequence, the short time-series we presently possess for most contaminants are subject to unresolvable influences from natural cycles (i.e., aliasing). For the Arctic, changes associated with ice and precipitation (the zero-degree isotherm), and with ecosystems (trophic structure, pathway bifurcation) have the greatest potential to alter pathways in ways that are particularly difficult to anticipate. Natural climate variation, such as the Arctic Oscillation, provides an opportunity to study change in the Arctic and to improve our ability to project the consequences of change. The marginal seas (Beaufort Sea, Archipelago, Hudson Bay, Chukchi and Bering Seas) appear to be the locations where change will occur first and most dramatically and it is there that change will most impact ecosystems valued by humans.

Alteration of the coastal margin's ice climate promises an acceleration of new risks due to more economic transport corridors, tourism and increased mineral exploration/exploitation. Perhaps the greatest concern will come from the growing attractiveness of arctic marginal seas for oil production and transport with reduced ice cover. Even though ice may clear from substantial portions of the marginal seas annually, the perspective must be maintained that these seas maintain all of the risks from seasonal ice cover—an ice cover that may, indeed, become more mobile.

Local sites (sewage lagoons, dumps, tailings ponds) and pipelines whose strategy of contaminant containment strongly depends on permafrost stability are at risk. Of the metals, mercury continues to provide the greatest concern. Despite declining anthropogenic emissions, the arctic ecosystem appears to be increasingly exposed to mercury for reasons that are not at all clear. Mercury depletion events show the Arctic to possess a unique, climate-sensitive process that may explain much of the Arctic's susceptibility to mercury contamination. However, the pathway for mercury after deposition until it eventually accumulates in apex aquatic feeders is very poorly known. Increase in the distribution of wetlands may provide a mechanism similar to the "reservoir effect" to release methyl mercury from mercury inventories accumulated in organic carbon phases of frozen ground.

Artificial radionuclides in the Arctic have proven not to be a radiological hazard but, rather, excellent tracers of ocean pathways. One pathway for artificial radionuclides continues to prove difficult to assess—sea ice. Evidence for concentrated amounts of radionuclides in ice far from known sources suggests that the ice-transport vector may cause exposure to humans and animals that use sea-ice surfaces. This pathway appears especially vulnerable to change as evidenced by the influence of the AO on ice drift patterns.

Organochlorine compounds continue to pervade global ecosystems despite success in curtailing many of the emissions. The risk organochlorines provide to arctic ecosystems is predicated on a scheme involving

physical and biological pathways that produces relatively high concentrations in apex feeders far from known sources. Given the length and complexity of the organochlorine pathways into top predators of aquatic systems in the Arctic, exposure to these chemicals is particularly sensitive to global change. Subtle effects of the organochlorines (immune function, reproduction and endocrine disruption) can be exacerbated by, and work together with, nutritional stress brought about by ecosystem and ice climate change. The significance of contaminants as added stressors to predators already suffering from habitat and ecosystem change, while widely suspected, is not at all confidently understood. Time series for organochlorine compounds in air and in top predators from aquatic systems are helpful in determining trends and evaluating recent contaminant controls. Because there are only sporadic, opportunistic time series for organochlorine compounds in oceans, rivers and lakes, the connection between the atmosphere, which responds rapidly to emissions, and the aquatic biota, which respond to the water, is missing.

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