Science of the Total Environment 408 (2010) 2985–2994

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Review

Current use pesticides in Arctic media; 2000–2007 ☆

Lisa Hoferkamp [a,*], Mark H. Hermanson [b,c], Derek C.G. Muir [d]

[a] Department of Natural Sciences, University of Alaska Southeast, Juneau, AK 99801 USA
[b] Department of Chemistry, University of Pennsylvania, Philadelphia, PA 19104 USA
[c] University Centre in Svalbard, N-9171 Longyearbyen, Norway
[d] Aquatic Ecosystem Protection Research Division, Environment Canada, Burlington, Canada ON L7R 4A6

ARTICLE INFO

Article history:
Received 20 June 2009
Received in revised form 18 November 2009
Accepted 18 November 2009

Keywords:
CUP
Current use
Pesticide
Arctic
Levels
Atmospheric transport

ABSTRACT

This review will summarize the levels of selected current use pesticides (CUPs) that have been identified and reported in Arctic media (i.e. air, water, sediment, and biota) since the year 2000. Almost all of the 10 CUPs (chlorothalonil, chlorpyrifos, dacthal, diazinon, dicofol, lindane, methoxychlor, pentachloronitrobenzene (PCNB), pentachlorophenol, and trifluralin) examined in the review currently are, or have been, high production volume chemicals i.e. >1 M lbs/y in USA or >1000 t/y globally. Characteristic travel distances for the 10 chemicals range from 55 km (methoxychlor) to 12,100 km (PCNB). Surveys and long-term monitoring studies have demonstrated the presence of 9 of the 10 CUPs included in this review in the Arctic environment. Only dicofol has not been reported. The presence of these chemicals has mainly been reported in high volume air samples and in snow from Arctic ice caps and lake catchments. There are many other CUPs registered for use which have not been determined in Arctic environments. The discovery of the CUPs currently measured in the Arctic has been mainly serendipitous, a result of analyzing some samples using the same suite of analytes as used for studies in mid-latitude locations. A more systematic approach is needed to assess whether other CUPs might be accumulating in the arctic and ultimately to assess whether their presence has any significance biologically or results in risks for human consumers.

Contents

1. Introduction

The Arctic is a region that is largely free from direct inputs of industrial and agricultural chemicals. Increasing urbanization in high latitude areas combined with atmospheric transport from lower latitude regions, however, provides for deposition of persistent organic pollutants (POPs) and other anthropogenic contaminants

☆ This paper is a contribution to the AMAP POPs assessment.

* Corresponding author. 11120 Glacier Hwy, Department of Natural Sciences, University of Alaska Southeast, Juneau, AK 99801 USA. Tel.: +1 907 796 6538; fax: +1 907 796 6447.
E-mail address: lahoferkamp@uas.alaska.edu (L. Hoferkamp).

0048-9697/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2009.11.038

into this unique part of the world. POPs are defined within the Stockholm Convention in terms of persistence, bioaccumulation, and adverse effects including human and ecotoxicity, as well as potential for long-range transport (LRT) (UNEP, 2001). Traditionally this definition has referred to legacy chlorinated organic contaminants, which are no longer in use or have much reduced global emissions as a result of past national and regional bans on use. Furthermore, Arctic monitoring data suggest that the environmental residues of legacy chlorinated organic chemicals are leveling off or declining (de Wit et al., 2004). In contrast, increasing levels of large-scale application agro-chemicals (current use pesticides; CUPs) have been found in regions isolated from their use and production, e.g. in the Arctic and in some alpine regions. By current use, we mean pesticides (acaricides, insecticides, herbicides, fungicides) which are currently registered for use by regulatory authorities worldwide although our emphasis is on products used in the USA, Canada, Russia, and western Europe. Not included here are banned organochlorine insecticides such as chlordane, DDT, mirex, and toxaphene. The CUPs most relevant to monitoring efforts are those exhibiting characteristics similar to those of POP substances currently listed in the Stockholm Convention. As of 2009 endosulfan has been proposed for inclusion in the Stockholm Convention while trifluralin, dicofol and pentachlorophenol (PCP) have been proposed to be added to the UNECE list of POPs (Rasenberg and van de Plassche, 2003; UNECE, 2007, 2008).

CUPs of concern in the Arctic have high production volumes and widespread current or past use, in temperate regions, relatively high air–water partitioning, and potential to bioaccumulate and biomagnify in fish and wildlife. When coupled with the fact that significant populations of arctic subsistence food users are reliant on high trophic level wildlife as part of their traditional diet, the relevance of these contaminants in Arctic monitoring activities becomes apparent. This review will summarize the levels of selected CUPs (Table 1) that have been identified and reported in Arctic media (i.e. air, water, sediment, and biota) since the year 2000. Endosulfan, an important CUP found in the Arctic, is reviewed in another article in this series (Weber et al., 2010-this issue).

1.1. CUP production and long-range transports

Production volume and use profile are important considerations in evaluating the arctic contaminant potential of CUPs (Muir and Howard, 2006). Almost all of the 11 CUPs listed in Table 1 currently are, or have been, high production volume chemicals i.e. >1 M lbs/y in USA or >1000 t/y globally. For two chemicals, methoxychlor and lindane, production and use have recently ceased in the USA, Canada, and the EU (US Environmental Protection Agency, 2004; UNEP, 2006). According to the UNEP lindane dossier, lindane production and use continues in Russia although other reports suggest that it has been banned along with other hexachlorocyclohexane (HCH) isomers (Li et al., 2005). PCP remains in use in the USA for wood treatment (US EPA, 2008) and is also permitted for wood treatment in Europe (UNECE, 2008) but has been phased out in Canada. Dicofol is a major acaricide in China and also has relatively wide use in the USA and Europe. Pentachloronitrobenzene (PCNB), chlorpyrifos, diazinon, and chlorothalonil remain high production CUPs and have undergone re-evaluation in the USA (US EPA, 1999, 2004, 2006b,a) although chlorpyrifos was phased out for residential and termite uses in the USA in 2000 (US EPA, 2006a). Dacthal underwent re-registration in the USA in 1998 (US EPA, 1998) and its use continues today (USGS, 2004). In general, quantities of PCNB, chlorphyrifos, chlorothalonil, diazinon, and dacthal used in other circumpolar countries and globally are not publically available to our knowledge.

The presence of CUPs in the Arctic environment suggests sufficient stability for transport to remote areas (Stocker et al., 2007). A degree of persistence is necessary for a pesticide's effectiveness. However, compared to the legacy organochlorine pesticides, most CUPs have

Table 1
Technical data for CUPs reviewed in this article.

Common name (acronym)	Class	IUPAC name	Production (t/y)[a]	Soil half-life (days)[b]	Water solubility (mg/L)[b,c]	Log Kow[c]	Log Kaw[c]	Log Koa[c]	AC-BAP[d]	CTD (km)
Chlorpyrifos	Organothiophosphate insecticide	Diethoxy-sulfanylidene-(3,5,6-trichloropyridin-2-yl)oxy-phosphorane	5000–7300[e]	30	0.4	4.96	−3.47	8.43	NA	430
Chlorothalonil	Organochlorine fungicide	2,4,5,6-Tetrachloro-1,3-benzene-dicarbonitrile	3600–5000[e]	30	0.6	4.81	−4.87	7.92	No	4420
Dacthal	Organochlorine herbicide	Dimethyl 2,3,5,6-tetrachlorobenzene-1,4-dicarboxylate	200[e]	100	0.5	4.28	−4.05	8.33	No	2690
Diazinon	Organothiophosphate insecticide	Diethoxy-(6-methyl-2-propan-2-yl-pyrimidin-4-yl) oxy-sulfanylidene-phosphorane	1820–3200[e]	40	60	3.81	−4.43	8.24	NA	130
Dicofol	Organochlorine insecticide	2,2,2-Trichloro-1,1-bis(4-chlorophenyl) ethanol	5500[f]	45	0.8	5.02	−5.01	10.03	Yes	640
Lindane	Organochlorine insecticide	(1*r*,2*R*,3*S*,4*r*,5*R*,6*S*)-1,2,3,4,5,6-hexachlorocyclohexane	3220[g]	400	7	4.14	−4.76	7.77	Yes	2500
Methoxychlor	Organochlorine insecticide	1-Methoxy-4-[2,2,2-trichloro-1-(4-methoxyphenyl)ethyl]benzene	193–2500[h]	120	0.1	5.08	−5.08	10.16	NA	55
Pentachlorophenol (PCP)	Chlorinated herbicide/fungicide	Pentachlorophenol	8500–50,000[i]	10–100	14	5.12	−6.0	11.1	NA	1320
Pentachloroanisole (PeCA)	PCP transformation product	1-Methoxy-2,3,4,5,6-pentachlorobenzene	–	55	0.35	5.45	−2.35	7.98	NA	2110
Pentachloronitrobenzene (PCNB)	Organochlorine fungicide	Pentachloronitrobenzene	350–450	189	0.44	4.64	−2.74	11.1		12,100
Trifluralin	Dinitroaniline herbicide	2,6-Dinitro-N,N-dipropyl-4-(trifluoromethyl)aniline	8700–10,500[j]	60	0.18	5.34	−2.38	7.72	NA	110

[a] Global for lindane and dicofol or for USA only as indicated below.
[b] ARS database.
[c] EPI Suite database (US Environmental Protection Agency, 2008).
[d] Arctic accumulation potential (Brown and Wania 2009). NA = not analyzed for AC-BAP.
[e] Kiely et al. (2004). USA production only — 2001.
[f] Global dicofol production (Rasenberg and van de Plassche, 2003) including China (Belfroid et al., 2005); USA estimated at 160 t/y for the period 1999–2004 (USGS, 2004).
[g] Global lindane usage, 1990–1995 (UNEP, 2006).
[h] Last produced in the USA in late 1990s. Production in 1975 estimated at 2500 t/y and in 1991 at 193 t/y (US Environmental Protection Agency, 2004).
[i] Range of global production estimates during the 1990s (UNECE, 2008).
[j] Combined USA and EU production based on Kiely et al. (2004) for 2001 in USA and UNECE (2007) for the EU.

been designed with reduced environmental persistence in mind. Since the 1970s manufacturers of these chemicals have been required to show that they have relatively short persistence in water, soil and sediment and low bioaccumulation potential (US EPA, 2007). Although several of the chemicals in Table 1 were in commerce prior to these regulations (e.g. methoxychlor, dicofol, and PCP) they remained in use after bans on chlorinated pesticides due to shorter persistence in soils and sediments, lower bioaccumulation potential and lower toxicity than most persistent organochlorines (Table 1). Long-range transport potential (LRTP) is not currently a consideration for registration of CUPs and thus data are not routinely generated by pesticide registrants. However, it can be estimated using LRT models such as the OECD LRT tool where suitable physical–chemical properties and the atmospheric oxidation half-lives of the chemicals are available (Scheringer et al., 2009).

Characteristic travel distances (CDTs) for the 10 chemicals plus pentachloroanisole (PeCA), a transformation product of both PCP and PCNB (US EPA, 2006b, 2008), calculated with the OECD LRT tool, range from 55 km (Methoxychlor) to 12,100 km (PCNB) (Table 1). While 5 of the chemicals have CDTs <1000 km, their presence in the arctic can be explained by transport in the gas phase or on dust during periods without rainout events (the OECD LRT tool and other similar models assume continuous low rates of precipitation), and lower rates of photodegradation than predicted from standard assumptions due to transport in the spring or fall (Muir et al., 2004). Use at rates of kg/ha in northern agricultural areas of Canada, Scandinavia, Russia and China may also explain the appearance of some compounds with low predicted CDT.

Wet deposition rates for these compounds have been shown to range from a few mg ha^{-1} y^{-1} to more than 1 g ha^{-1} y^{-1} in European regions removed from the site of application (VanDijk and Guicherit, 1999). In a study of contaminants in US western parks that spanned the years 2002 through 2007 it was shown that parks with the highest concentrations of semi-volatile organic compounds, CUPs dominated the distribution (Landers et al., 2008). In all parks sampled, including 3 parks in the Alaskan Arctic, total endosulfans, γ-HCH, chlorpyrifos and dacthal were commonly found and chlorpyrifos, dacthal and methoxychlor were identified above MDLs in more than 50% of fish samples. Due to the absence of significant local or regional pesticide sources, deposition in the Arctic parks is due to long-range transport from agricultural areas in lower latitudes.

1.2. Pre-2000 studies and reviews

Early surveys and monitoring of environmental levels of both legacy organochlorines and CUPs have been reviewed previously. The Canadian Arctic Contaminants Assessment Report (CACAR) of 2004 (Bidleman et al., 2003) and AMAP's POPs Assessment Reports (De March et al., 1998; de Wit et al., 2004) reviewed the previously published Arctic contaminants literature to about 2002. By the early 2000s there was considerable evidence for declining concentrations of legacy chlorinated pesticides in wildife and in arctic air. However, time trends of the numerous CUPs currently in large-scale use and production in the Arctic were limited or non-existent as of 2002. The major exception was lindane for which time trends in air, fish, seals, beluga and seabirds are available (de Wit et al., 2004). Time trend data are also available for endosulfan (Weber et al., 2010-this issue) and PeCA in air (Hung et al., 2002).

Beginning in the early 1990s studies showing the presence of other CUPs in the Arctic environment were published. Welch et al. (1991) reported methoxychlor, PeCA and trifluralin in snow from the Canadian arctic. The snow was associated with a “brown snow” event whose clay mineral composition, soot particles, and visible organic remains point to an Asian source, probably western China. Melted snow samples were found to contain lindane concentrations of 0.320 ng/L, methoxychlor at 0.066 ng/L and trifluralin at 0.660 ng/L. Particles filtered from the melted snow were found to contain significant levels of these analytes as well.

Earlier, several studies had reported the presence of lindane and methoxychlor (Franz et al., 1997) in Arctic snow. Methoxychlor and trifluralin were detected in air sampled at stations in the Canadian Arctic (Halsall et al., 1998; Hung et al., 2002) while trifluralin, chlorpyrifos and chlorothalonil were identified in surface water, ice and fog samples from the Bering and Chukchi Seas (Chernyak et al., 1996). Studies detailing the occurrence of these three latter CUPs and diazinon in Arctic fog sampled in the Bering and Chukchi Seas were published a short time later (Rice and Chernyak, 1997). Analysis of tissue from resident fish collected near Fairbanks, AK in the early eighties included dacthal, but levels were below the detection limits of the instrumentation used (Schmitt et al., 1985). Boyd-Boland et al. (1996) reported a wide range of CUPs in Russian ice caps using solid-phase micro-extraction followed by ion trap mass spectrometry. This latter study holds the record for the largest number of CUPs identified in arctic snow, with about 33 non-persistent organochlorine compounds.

Since then, proposals have been submitted to the UN Economic Commission for Europe (UNECE) for consideration of trifluralin, PCP and dicofol as candidate POPs for inclusion in the Long Range Transboundary Air Pollution (LRTAP) Convention (Rasenberg and van de Plassche, 2003; UNECE, 2007, 2008). Inclusion of these CUPs is based partly on their detection in remote regions.

2. Current use pesticides in the Arctic; post-2000 publications

Recent studies reporting concentrations of CUPs in the Arctic are limited in number but provide some data for comparison to the earlier findings discussed briefly above. Among the published results discussed here, a lack of consistency in sampling methodologies, sampling site selection, collection time and duration, selected analytes, analytical methods and detection limits leave direct comparisons and thus trend determination difficult. Another important issue is the lack of confirmation of some results, particularly those generated by Gas Chromatography-Electron capture detection (GC-ECD). While this detector is quite suitable for routine measurements of chlorinated pesticides and PCBs at the higher levels found in biota it is a major challenge to use it for low levels typical of abiotic samples because it is a non-specific detector. There is also the potential for false positives from GC–low resolution mass spectrometry detection of CUPs. This is discussed by Weber et al. (2010-this issue) in connection with analysis of endosulfan but could also apply to other CUPs.

The following review of arctic CUP levels minimizes reference to many of these variables and reports the most commonly found CUPs. Of the three HCH isomers, only the γ-isomer, also known as lindane, is still in current use (UNEP, 2006). Lindane is commonly measured in studies of organochlorine contaminants and reports of its occurrence in Arctic media outnumber those of all other CUPs discussed here. This review will therefore focus only on major temporal trend studies that have included lindane. Though smaller in number, reports of chlorpyrifos, chlorothalonil, dacthal, diazinon, methoxychlor and trifluralin in the Arctic are sufficiently numerous to necessitate review. We were unable to find any results for dicofol in arctic environmental media. However, dicofol was mentioned in the context of high levels of *o,p′*-DDT in river water samples collected from the Russian Arctic in 2003 and 2005 (Carroll et al., 2008).

2.1. Air

High volume air samples collected at several locations in the Arctic since 1993 consistently demonstrate measurable levels of lindane. These data have recently been assessed (Hung et al., 2005, 2010-this issue) and generally show declining concentrations in sampling sites

in Finland (Pallas), northern Ellesmere Island (Alert) and Svalbard (Zeppelin) throughout the 1990s and early 2000s.

Several studies using passive air samplers have reported lindane in Arctic air at locations distant from the high volume sampling stations. Shen et al. (2004) found lindane ranging from about 5 to 14 pg/m^3 at 7 Canadian arctic locations using passive XAD samplers. Pozo et al. (2006, 2009) reported lindane at 3 arctic locations (Alert, Barrow, and Ny Alesund) using passive PUF samplers at similar concentrations ranging from 5 to 9 pg/m^3. Concentrations at Snare Rapids (NT) a subarctic site about 1200 km north of a lindane use area in western Canada were 16 pg/m^3, or about double those found at high arctic sites.

As part of the Western Airborne Contaminants Assessment Program (WACAP) (Landers et al., 2008) passive air-sampling devices using XAD-2 as an absorbent were deployed from July 2005 to July 2006 at three Alaskan parks. Analysis indicated low levels of the CUPs investigated compared to similar measurements at parks of lower latitude. Total chlorpyrifos levels were at instrumental detection limits (1.6 pg/g XAD) while dacthal and trifluralin levels were below detection limits in the Arctic and interior Alaska parks. Lindane was found in all three parks at levels ranging from ~9 to 35 pg/g dry XAD.

Studies of Arctic air masses sampled at stations in the Canadian, Alaskan, Russian and Norwegian Arctic between 2000 and 2003 included direct measurements of the CUPs methoxychlor and trifluralin (Su et al., 2008). Positive results for air concentrations of these CUPs were noted but the levels were below analytical detection limits and thus values not specified. The presence of *o,p′*-DDT in some of the air samples was inferred by the authors as indicative of atmospheric transport of dicofol. This isomer of DDT has been identified as a common impurity in Chinese formulations of dicofol (Qiu et al., 2005). In an earlier report Hung et al. (2005) reported annual average levels of trifluralin in air approaching the practical quantitation limit (PQL); 0.13–0.18 pg/m^3 at Tagish, 0.1–0.05 pg/m^3 at Kinngait and 0.18–0.16 pg/m^3 at Dunai. In the same article Hung et al. also reported PeCA ranging from 2.6 to 4.0 pg/m^3 and methoxychlor ranging from 0.12 to 0.41 pg/m^3 at the same locations. A comparison of high volume sampling results from five arctic sites (Alert, Kinngait, and Little Fox Lake in Canada; Point Barrow in the USA; Valkarkai in Russia; and Zeppelin on Svalbard) typically showed seasonal variations in atmospheric levels of lindane with peaks in the spring and fall suggesting application and tilling at lower latitudes followed by LRT (Su et al., 2006). Mean annual concentrations were lowest at the Alaskan site (Point Barrow) at 2.7 pg/m^3 (range: 0.89–5.8) and highest at the Russian site (Valkarkai) at 7.4 pg/m^3 (range: 4.6–16).

A 2007 study of CUPs in the Arctic environment appears in the abstracts of the ArcticNet Fourth Annual Scientific Meeting published online at the organizational website. This Canadian-based, international collaborative project facilitated CUP sampling from a 2007 cruise of the Labrador Sea. Jantunen et al. (2007) analyzed air and water samples from the cruise, for chlorpyrifos, chlorthalonil, dacthal and trifluralin and presented their air-sampling results in a poster at this meeting. CUP levels were reported with chlorpyrifos at 0.36–30.4 pg/m^3, dacthal at 0.68–8 pg/m^3 and trifluralin at levels of 0.2–0.91 pg/m^3. Chlorthalonil was identified as present but levels had not been quantified at the time of the poster's publication.

2.2. Lake and river waters

At a 2000 AMAP workshop, Alexeeva et al. (2000) provided results from an extensive sampling dating from 1990 to 1996, of 14 Russian rivers flowing into the Arctic Ocean. Later publication of these results reported annual average lindane levels ranging from 114 ng/L in the Taz River to non-detect in the Anabar and Olenek Rivers (median value 3 ng/L) (Alexeeva et al., 2001). The median lindane level reported by Alexeeva is orders of magnitude larger than those reported by Carroll et al. (2008) who found lindane levels in freshwater from the Ob and Yenisei Rivers at pg/L concentrations (56 ± 5.7 pg/L and 58 ± 16 pg/L respectively). Wide variations in concentrations and fluxes with seasonal pesticide use and river flow could explain these differences. Kimstach and Dutchak (2004) found that γ-HCH was the predominant HCH isomer in the Pechora and Yenisey rivers in the Russian Persistent Toxic Substances study conducted in 2001–02. They reported fluxes of lindane of 1124 kg/y in the Yenisey and 1179 kg/y in the Pechora River. Alexeeva's 2001 publication reported similar fluxes of lindane in the Pechora (1460 kg/y) but 6-fold higher levels in the Yenisey (6420 kg/y).

By comparison Carroll's 2008 publication reported lindane fluxes of 33 kg/y in the Yenisey for 2003. These authors reported low pg/L concentrations of lindane in the Gulf of Ob (56.0–89.0 pg/L) and Yenesei Bay (47.0–71.0 pg/L) as well as the Kara Sea shelf (60.0–98 pg/L). Carroll et al. also reported trace levels of methoxychlor in the river water samples but results were below detection limits (<2.0 pg/L).

Water samples collected from six Canadian Arctic lakes between 1999 and 2001 had levels of 11 CUPs at or near analytical detection limits (Muir et al., 2004). Among those discussed in this review, chlorpyrifos was identified at the detection limit (<0.017 ng/L) in five of the lakes studied and at 1.6 ng/L in a sixth. The fungicide chlorothalonil was identified in three of the six lakes; two of the lakes had chlorthalonil concentrations at the MDL (<0.002 ng/L) while one lake carried concentrations of the fungicide (2.8 ng/L) that exceeded levels found for any of the 11 CUPs measured in the study. Dacthal was present at nominal concentrations (0.01–0.07 ng/L) in all of the six lakes tested. Levels of diazinon ranging from 2.0 to <0.003 ng/L (detection limit) were identified in two of the lakes. In a followup study, Muir et al. (2007) analyzed large volume water samples from Lake Hazen in northern Ellesmere Island and from Char Lake on Cornwallis Island, collected in 2005 and 2006. Chloropyrifos, dacthal, trifluralin, PeCA, and PCNB were among the CUPs detected in all samples at low pg/L concentrations.

2.3. Ocean waters

Bidleman et al. (2007) reported lindane concentrations across the Canadian arctic archipelago ranging from 0.19 ng/L in the east (Davis Strait) to 0.45 ng/L in the west (Viscount Melville Sound) based on the "Tundra-99" cruise. Strachan et al. (2000) published results from surface water samples collected from the Tundra '99 cruise and 3 other cruises in the Canadian arctic/Alaska/Greenland waters between the years 1993 and 1999. In general, concentrations were similar to those reported by Bidleman et al. (2007).

In a July 2000 study of air–sea exchange of the α- and γ-HCHs, Lakashus et al. sampled air and water on a cruise originating in Bremerhaven, Germany and terminating at Svalbard in the Norwegian Arctic (Lakaschus et al., 2002). Distinctively higher HCH levels were detected in the northern hemisphere compared to the southern hemisphere and the levels determined are noted as similar to those found in previous investigations of the same area. In comparing the seven water sampling sites and the five air-sampling sites that were within Arctic latitudes (74.1°–77.2°), a limited range of lindane concentrations was observed; 230 pg/L to 72 pg/L for aqueous samples and 7.1 pg/m^3 to 2.9 pg/m^3 for air samples.

Dacthal was reported in seawater and invertebrates from the Canadian Arctic by Morris et al. (2007) at the 2007 SETAC North America meeting but quantitative results were not provided in the published abstract.

2.4. Snow

Garbarino et al. (2002) analyzed CUPs in a study of contaminants in sea ice associated snow cores collected in 1996 from five Alaskan coastal locations on the Chukchi and Beaufort Seas. Lindane and PeCA

were not detected (PQL of 6 ng/L) while dacthal was detected at four of the five sites sampled but at levels near the PQL (4 ng/L). Chlorpyrifos was detected in snow from three of five sites at relatively high levels ranging from 70 to 80 ng/L.

Starting in 2003 and ending in 2005, the WACAP analyzed a suite of, CUPs including chlorpyrifos and its degradation product chlorpyrifos oxon (reported as total chlorpyrifos), dacthal, lindane and trifluralin were included. It is noted however, that lindane was considered an historic use pesticide in the WACAP report. The results of this project were published in a 2008 final report (Landers et al., 2008) and several portions of the report have appeared in the scientific literature as stand-alone articles (Usenko et al., 2005; Hageman et al., 2006; Usenko et al., 2007; Ackerman et al., 2008; Landers et al., 2008). Among the 8 primary parks only two, the Noatak National Preserve (NOAT) and Gates of the Arctic National Park and Preserve (GAAR), were within the Arctic (as defined by the 10 °C July air temperature isotherm). One other primary (Denali National Park and Preserve; DENA) and four secondary parks (from among 12 secondary parks sampled) were located within Alaska, USA. Only results from the three primary parks will be discussed in detail.

Both CUP concentration and fluxes ($ng/m^2/y$) in snow samples are provided in the WACAP report and fluxes are summarized in Table 2. Flux values indicate total input to the watershed from seasonal snowpack and provide information on the total amount of contaminant deposited via precipitation in a given area but these flux values are highly dependent on snowfall quantity. Because the three Alaskan parks do not typically receive as much snowfall as other mountainous regions of the conterminous United States, flux values are comparatively lower. Significant variation in flux of CUPs from snow deposition within and among the Alaskan parks is attributed to differences in annual snowfall. Hageman et al. (2006) have published results for measurements made during 2003 and these data generally reflect the trends described in the WACAP report for data collected from 2003 through 2005 (Landers et al., 2008). Concentrations of CUPs at three sites within DENA and one site within both NOAT and GAAR were consistently lower than those measured at lower latitude. The average concentration of total chlorpyrifos in snow from all five Alaskan sites showed little variation (0.023 ± 0.008 ng/L). Dacthal concentrations were lower and more varied across the five sites (average concentration $= 0.0097 \pm 0.0091$ ng/L) with the highest and lowest (non-detect) concentrations found at the DENA sites. Four sites had lindane concentrations below instrument detection limits (<0.012 ng/L) and the fifth, located in NOAT, showed lindane levels of 0.072 ng/L. Trifluralin and a large suite of other CUPs in use in the USA and Canada were not detected in snow from the Alaskan parks. Dacthal concentrations in snow declined significantly with latitude while concentrations of chlorpyrifos and lindane did not (Hageman et al., 2006).

Fluxes of the CUPs were calculated by multiplying contaminant concentrations (ng/L) by the accumulated amount of snow at the time of sampling, in L $(H_2O)/m^2/y$. Among total chlorpyrifos and dacthal, one DENA site had the highest flux values (32 and 27 ng/m^2 respectively) and the two other DENA sites had the lowest flux values for chlorpyrifos (0.48 and 1.0 ng/m^2). Flux of dacthal was also low at the latter two DENA sites and at the arctic GAAR site (0.49–0.67 ng/m^2). The highest flux of lindane was found at the NOAT site (43 ng/m^2). The lowest lindane flux was found at the two DENA sites that have already been identified as having low fluxes of chlorpyrifos and dacthal. These data accentuate the flux variations that can be expected with variable snowfall even within a relatively small region.

A 1998 ice core drilled from the Austfonna ice cap on Svalbard, Norway was analyzed for contaminants including some CUPs. Isaksson et al. (2003) noted that seasonal melting in surface layers could lessen the deposition timing accuracy of various components within the ice core ($\delta^{18}O$, conductivity, pesticides) but indicates that this issue was overcome by simultaneous use of reference horizons (volcanic eruptions, Cs-137 peak deposition) and glacial modeling for core dating purposes.

A large suite of CUPs extracted from the top 70 m of the Austfonna ice core and corresponding to the years 1906 to 1998 was quantified (Hermanson et al., 2005). The upper 48 m of the core, the oldest of which was deposited in 1936, contained various organochlorine and organophosphorus pesticides. Chlorthalonil and the oxidative degradation products (oxons) of chlorpyrifos and diazinon were among the analytes tested but reported as non-detectable. Lindane and dacthal were identified in isolated, non-continuous sections of the core. The highest concentration of lindane was 7.7 ng/L and corresponded to a core-section dating from 1979 to 1986. Peak levels of dacthal (0.3 ng/L) were associated with a section of the core from the same time period. Diazinon, methoxychlor and intact chlorpyrifos were found consistently and at various levels throughout the core suggesting a continuous source that the authors attribute to long-term atmospheric transport facilitated by lower concentrations of hydroxyl radical in the arctic atmosphere. Levels of diazinon peaked at 20.5 ng/L during the late 1980s and early 1990s and were the highest among the three consistently present CUPs discussed here. Declining diazinon levels in later core sections are attributed to the 2001 European ban on this pesticide. Methoxychlor was found in sections of the core dating to the early 1950s and concentrations were found to increase over subsequent years with a peak concentration of 4.7 ng/L associated with the early 1980s. The highest concentration of chlorpyrifos (16.2 ng/L) was found in sections of the core corresponding to the early to mid-1980s and levels were found to decline from that value into the 1990s. The absence of chlorpyrifos or its oxon in surface samples of the ice core, dating from 1990 to 1998, was noted.

Table 2
Fluxes ($ng/m^2/y$) for current use pesticides (CUPs) in Arctic snow and ice cores.

Location	Collection year	CUP	Recent flux ($ng/m^2/y$)	Notes	Reference
Devon ice cap (Canada)	2005	Chlorpyrifos	<0.1	Snow pit samples dated 2001–05	Muir unpublished 2007[a]
	2005	Dacthal	0.06–2.7		Muir unpublished 2007
	2005	Lindane	3–5		Muir unpublished 2007
	2005	Metolachlor	0.73–15		Muir and Zheng (2007)
	2005	Metribuzin	<0.1–1.1		Muir and Zheng (2007)
	2005	Methoxychlor	<0.1		Muir unpublished 2007
	2005	PeCA	0.4–0.6		Muir unpublished 2007
	2005	Phorate	10–42.0		Muir and Zheng (2007)
	2005	Trifluralin	0.16–0.22		Muir and Zheng (2007)
Noatak and Gates of the Arctic NP (USA)	2003	Chlorpyrifos	2–14	Samples from watersheds of Lakes Burial, Matcharak and Kangilipack	Landers et al. (2008); Hageman et al. (2006)
	2003	Dacthal	0.5–3.9		
	2003	Lindane	0.2–43		
Denali NP (USA)	2003	Chlorpyrifos	0.5–32	Samples from watersheds of Lakes Wonder, McLeod and Kahiltna base camp	Landers et al. (2008); Hageman et al. (2006)
	2003	Dacthal	0.5–27		
	2003	Lindane	0.08–6.0		

[a] Unpublished results, Environment Canada, Burlington ON.

Significantly higher lindane and methoxychlor levels were found in ice and snow samples collected in 1992 and 1993 from the Russian arctic by Boyd-Boland et al. (1996) than have been reported in more recent studies. For example, lindane concentrations were 2–3 orders of magnitude higher while methoxychlor was reported at levels 2–4 orders of magnitude higher than reported by Hermanson et al. (2005). Description of the Russian arctic sample preparation was limited with respect to temporal assignment and thus the levels reported may represent composite levels spanning several years or decades. A 1986 study of Canadian arctic snow samples was also cited and indicated levels of lindane comparable to Hermanson's results (2.98–4.55 ng/L) (Gregor and Gummer, 1989).

Muir and Zheng (2007) reported detection of 7 CUPs in the Devon Island ice cap in the Canadian arctic. Samples were obtained in a snow pit dug in 2005. The 7 CUPs detected in almost all recent horizons were dacthal, lindane, PeCA, methoxychlor, metolachlor, metribuzin, phorate, and trifluralin. Levels found for methoxychlor, metribuzin, and trifluralin were less than detection limits in surface and near surface layers but were found to increase slightly in deeper horizons. All CUPs detected are major-use chemicals in Canada and the USA, as well as in Europe. The higher amounts in deeper layers may reflect the phase out of methoxychlor and declining use of trifluralin in the USA over the period 1987–2001 (Kiely et al., 2004).

In Table 2, the fluxes of CUPs in the WACAP study sites in Alaska are compared with results from the Devon ice cap in Canada (Muir and Zheng, 2007). Fluxes of dacthal and lindane were comparable at NOAT and Devon ice cap. Chlorpyrifos fluxes were much greater at the Alaska locations while the opposite was the case for trifluralin. Unfortunately fluxes of CUPs for the Austfonna ice core are not available for comparison.

2.5. Sediments and soils

Evenset et al. (2004) report finding trifluralin and lindane residues in sediments, collected in 1996 from two lake sites on Bjornoya Island in the Barents Sea. Lake sediments showed levels of trifluralin that were at the PQL (0.04 ng/g dw) and low lindane levels (0.2 ng/g dw).

Stern et al. (2005) reported lindane in a sediment core from Lake DV on Devon Island in the Canadian arctic. Maximum concentrations were 0.17 ng/g (dry wt) and the maximum flux was estimated at 12.9 ng/m^2/y.

Sediments collected in 2004 from the Pasvik River in the Russian Arctic between latitudes 69.00° N and 69.40° N were analyzed for the CUP dacthal. From this study, a single value of 0.012 ng/g dacthal was reported at Dioxin 2005 in Toronto, Canada (Savinova et al., 2005).

The WACAP report (Landers et al., 2008) included lake bottom sediment cores from four lakes within the three primary WACAP parks in Alaska and provides information regarding temporal changes in contaminant load over the last ~150 y. Annual accumulations of contaminant are presented as ng/m^2/y and are corrected for differences in sediment accumulation within a given lake. One lake in each of the two arctic parks and two lakes in DENA were sampled. Total chlorpyrifos, lindane and trifluralin levels were reported as below detection limits. Dacthal was found in the NOAT-Burial Lake core at levels ranging from ~0.5 ng/m^2/y to ~0.2 ng/m^2/y and in the DENA-McLeod Lake core at levels ranging from 0.25 to 1.6 ng/m^2/y. The deposition timing associated with detection of dacthal in both these cores was somewhat inconsistent with peak production and usage of these chemicals. The three Alaskan parks had significantly lower CUP levels compared to cores from mid-latitudes lakes that were measured in the same study.

2.6. Freshwater fish

Evenset et al. (2004) report finding trifluralin and lindane in zooplankton and Arctic Char collected in 1996 from two lake sites on Bjornoya Island in the Barents Sea. However, trifluralin was below the limit of quantification and actual concentrations were not reported.

Fish of similar age and sex distributions were collected from the same four lake sites noted above for collection of sediment cores within the WACAP. Lake trout were collected from NOAT, GAAR and one site from DENA while burbot and whitefish were collected from a second DENA site. Comparisons of CUP levels and method specifics were detailed in a publication from Ackerman et al. (2008) that appeared prior to publication of the final WACAP report. CUP levels were not available in tabular form and levels reported here are approximated from graphical illustrations. Total chlorpyrifos levels ranged from 0.041 to 0.1 ng/g ww among the four lakes. Dacthal and lindane concentrations were very similar across fish from the four Alaskan lakes (0.011–0.015 ng/g ww and 0.11–0.13 ng/g ww respectively). The CUP methoxychlor was included in the suite of analytes measured but was not found at significant levels in any of the four Alaskan sites. Trifluralin levels were not discussed in publications describing analyses of fish tissue within the WACAP project.

Vorkamp et al. (2004) collected and analyzed an extensive array of tissue samples between 1998 and 2001 from terrestrial, freshwater and marine biota in Greenland but the only CUPs included in the suite of analytes were methoxychlor and PeCA. Only samples providing measurable results (i.e. non-detect results are neglected) and among those, only the median results from specific sampling locations are cited here. Muscle tissue in Arctic Char was found to contain methoxychlor at 0.079 ng/g lw. No PeCA was reported in freshwater fish.

2.7. Terrestrial plants and animals

A comparison of total pesticide concentrations (sum of endosulfan, chlorpyrifos, dacthal, HCB, γ- and α-HCH, dieldrin, DDTs and chlordane) in lichen samples collected under the WACAP indicated that the lowest levels (~5–10 ng/g lichen lipid) among all sites tested were found in the primary parks of Alaska (NOAT, GAAR, DENA). Levels increased (~100 ng/g lipid) moving south to the four secondary Alaskan parks, the most northern of which is located at the beginning of the Aleutian Peninsula (Katmai National Park and Preserve; KATM) and the most southern located at the southern end of Southeast Alaska (Stikine-LeConte Wilderness, Tongass National Forest; STLE). A portion of the geographical trend was attributed to species differences and winter snow burial of the most northerly samples but CUP levels in high latitude lichen were statistically less than those from the conterminous United States. Of the CUPs measured, total chlorpyrifos and trifluralin levels in lichen were reported at or below instrument detection limits. In less than 50% of the samples from the three Alaskan parks, lindane was detected. A mean level for DENA was given as 0.45 ng/g lipid and a value of ~0.13 ng/g lipid for NOAT and GAAR can be estimated from WACAP report figures. Mean levels of dacthal in lichen samples from all three parks were provided (mean = 0.60 ng/g). Two year old conifer needles from Sitka spruce were also analyzed but only from DENA because the two Arctic parks are largely treeless. Mean total chlorpyrifos levels were measured at 0.86 ng/g lipid, dacthal at 0.09 ng/g lipid and lindane at 0.80 ng/g lipid. Geographical trends in total pesticide levels in conifer needles showed the same pattern as lichen, increasing with decreasing latitude, but difference in highest and lowest levels was less pronounced.

An assessment (1998 to 2001) of terrestrial, freshwater and marine biota in Greenland provided methoxychlor and PeCA levels in several terrestrial mammals (Vorkamp et al., 2004). The muscle tissue of lamb (0.036 ng/g of lipid weight) and of muskox (0.57 ng/g lw) showed significant levels of methoxychlor while all tissue types in caribou were found to contain this pesticide; liver, 0.088 ng/g lw, muscle, 0.22 ng/g lw, kidney, 0.73 ng/g lw and blubber, 0.038 ng/g lw. Caribou muscle was found to have the highest concentration of PeCA

(0.20 ng/g lw) followed by median concentrations of PeCA in caribou blubber and kidney just above instrumental detection limits.

2.8. Marine biota

Arctic cod liver samples collected from the Vestertana Fjord on the Arctic coast of Norway for the period 1987–1998 were measured for a series of polychlorinated organic compounds including lindane (Sinkkonen and Paasivirta, 2000). The levels of lindane measured over the 12 year period were sufficiently different to suggest a decreasing temporal trend. In 1987 lindane was determined at 7.4 ng/g lipid weight (lw) and while concentrations from the following year were slightly greater (8.1 ng/g lw), succeeding years showed a distinct downward trend to 4 ng/g lw in 1998. Variable loadings of principal components indicated a statistically significant decrease of HCH over the study period.

Fisk et al. (2001) sampled the herbivorous copepod *C. hyperboreus* in the Northwater Polnya off the western coast of Greenland in 1998 and used the data to correlate POP zooplankton concentrations with POP water solubility. Only summed concentrations of HCH isomers were provided in that publication and those values ranged from 1.04 to 31.3 ng/g dry weight over the four month sampling period. A concurrent publication describing isomer distributions of HCH across a Northwater Polnya food web provides specific lindane concentrations for several types of biota also collected in 1998 (Moisey et al., 2001). Six species of zooplankton were sampled and showed lindane levels ranging from 1.9 ± 0.4 ng/g lipid (*M. occulata*) to 36.4 ± 12 ng/g lipid (*T. libellula*). Lindane levels in benthic invertebrates ranged from 31.7 ± 7.2 ng/g lipid (*A. nugax*) to 0.1 ± 0.1 ng/g lipid (clam). In seabirds the lowest lindane levels reported were 16.8 ± 2.9 ng/g lipid (dovekie) to 3.6 ± 0.5 ng/g lipid (glaucous gulls) while levels of 23.3 ± 4.6 ng/g lipid were reported in Arctic cod and 7.2 ± 1 ng/g lipid in ringed seal. Another assessment of seabirds from the Northwater Polnya, collected between May and June of 1998 reports lindane levels in liver and fat tissue (Buckman et al., 2004). Seven different seabird species were assessed and all carried relatively low levels of lindane. In samples of liver, concentrations ranged from 0.05 ± 0.01 to 0.19 ± 0.022 ng/g ww while in samples of subcutaneous fat, a 1–2 order of magnitude increase in concentration was observed (2.4 ± 0.2 to 12.2 ± 2.0 ng/g ww).

Amphipods collected from the front of northern sea ice, north of Svalbard in the Fram Strait during September of 1998 and 1999 were analyzed for organochlorine bioaccumulation by Borga et al. (2002). Three types of amphipods, each displaying a different feeding behavior, were collected from five different stations along the sea ice and total HCH concentrations were reported for each amphipod type. Average amounts of lindane in all three amphipod types at each of the five stations were reported. These lindane levels ranged from 5.7 ± 0.3 to 12.3 ± 2.0 ng/g lw with levels generally increasing as sample site moved southward. The small standard deviations on the average lindane levels indicate little variation among herbivorous versus omnivorous amphipods. Zooplankton samples collected from five locations in northern Alaska and the Canadian Arctic in July and September 1998, June 1999 and September 2000 were analyzed by Hoekstra et al. (2002) for a similar suite of organochlorines. Total HCH (sum of α, β, γ and δ-isomers) in zooplankton and associated water was reported as the dominant analyte group among those measured and with an α/γ ratio that was similar to other studies (Hargrave et al., 2000; Fisk et al., 2001). Mackay and Arnold (2005) reexamined HCH isomer results from the study of Hoekstra et al. (2002, 2003) and concluded that lindane did not biomagnify in the arctic marine food web.

Hobbs et al. (2003) assessed organochlorine pesticide levels in the blubber of Minke whales sampled in 1998 from seven locations throughout the North Atlantic and European Arctic. Lindane was among the analytes with the lowest and highest mean concentrations reported as 9.68 ng/g lipid and 55.9 ng/g lipid respectively. The highest lindane concentrations were found in North Sea whales. In contrast to other organochlorine pesticides, a trend of increasing $\sum$HCH levels from east to west was noted.

A study of heavy metal and organochlorine levels reported $\sum$HCH levels in three species of seabird eggs collected from the Canadian Arctic between 1975 and 2003 (Braune, 2007). In addition to total concentrations of all HCH isomers, data for α- and β-HCH were provided and thus lindane levels could be inferred. For two seabird species, black-legged kittiwakes and thick-billed murres lindane was present only in the 1993 samples at approximately 0.2 ng/g ww and 1.1 ng/g ww respectively. In the third species studied, northern fulmars, the presence of lindane was surmised in 1993 (0.3 ng/g ww) and in 1998 (0.3 ng/g ww).

Marine biota tissue samples collected by Vorkamp et al. (2004) between 1998 and 2001 in Greenland were analyzed for methoxychlor and PeCA. Snow crab muscle tissue and liver contained the highest levels of methoxychlor (29 ng/g lw and 12 ng/g lw respectively) among marine invertebrates and fish, followed by shorthorn sculpin liver (5.1 ng/g lw), Atlantic cod muscle and liver (5.0 ng/g lw and 1.9 ng/g lw respectively), shrimp muscle (4.0 ng/g lw) and wolfish muscle (3.3 ng/g lw). Positive results for other marine invertebrates and fish were less than 1 ng methoxychlor per gram of lipid weight. Equal median concentrations of 2.3 ng PeCA/g lw were found in Atlantic cod muscle, Atlantic cod liver and capelin muscle. Measurable concentrations of PeCA were found in snow crab muscle (0.66 ng/g lw) and snow crab liver (0.45 ng/g lw) but the median concentration was below the detection limit of 0.01 ng/g ww in Atlantic salmon muscle and redfish muscle. Of four common seabirds sampled, kittiwake and thick-billed murre tissues carried a methoxychlor burden; kittiwake muscle with 1.1 ng/g lw and murre liver with 2.6 ng/g lw. Only king eider and thick-billed murre livers were assessed for PeCA with median concentrations just above detection limits (0.36 and 0.22 ng/g lw respectively). Of five marine mammal species sampled, four indicated positive methoxychlor levels. Harp seal muscle and blubber showed 1.8 and 1.1 ng/g lw respectively, minke whale muscle and kidney were found to contain 1.0 and 0.21 ng/g lw respectively, and beluga liver had 1.8 ng/g lw. All types of narwhal tissue sampled had significant levels of methoxychlor. Levels in narwhal were in fact most consistent among tissue types and highest among all species; liver: 9.2 ng/g lw, muscle: 0.12 ng/g lw, kidney: 0.074 ng/g lw, skin: 6.2 ng/g lw and blubber: 9.3 ng/g lw. The highest concentrations of PeCA were found in Arctic mammal muscle with values ranging from 0.08 ng/g lw in harp seal muscle to 0.54 ng/g lw in narwhal muscle and 1.1 ng/g lw in beluga muscle. It was noted that in contrast to typical patterns of bioaccumulation in the food chain, PeCA concentrations in marine mammals did not exceed the concentrations in the marine fish.

In a study published at approximately the same time as the Greenland biota study, no detectable residues of methoxychlor was reported in liver and breast muscle samples from cormorants and sandpipers inhabiting the Aleutian Archipelago in Alaska, USA between 2000 and 2001(Rocque and Winker, 2004). Levels of other organochlorine legacy contaminants were reported but methoxychlor was the only CUP measured in the Aleutian bird tissues. Situated between 52° N and 54° N latitude, the Aleutian chain is at the southern limit of the Arctic but is considered part of this region when defining the southern Arctic boundary as the 10 °C July air temperature isotherm. The Aleutians have no local sources of agricultural pesticides and CUP levels found in biota further accentuate the long-range atmospheric transport potential of these contaminants.

Methoxychlor was detected in a 2001 sampling of the glaucous gull population of the Barentsburg area of the Svalbard Archipelago (Savinov et al., 2008). Among thirteen female and 7 male glaucous gulls sampled, average methoxychlor levels above MDL values were found in the liver tissue (1.20 ± 1.39 ng/g ww and 0.60 ± 0.65 ng/g ww respectively).

A 2005 publication described analysis of adipose tissue from adult and sub-adult polar bears, sampled between 1996 and 2002, from Arctic and Subarctic regions of Alaska, Canada, East Greenland, and Svalbard (Verreault et al., 2005). The ∑HCH concentrations were reported and ranged from 71.3 to 593 ng/g lipid. The highest concentrations were identified in Alaskan samples and the lowest in Svalbard. The higher concentrations at the western sampling sites were attributed to ongoing contributions of HCHs from China, southeastern Asia, and North America. No specifics regarding lindane concentrations were provided.

PCP was reported in polar bear plasma samples that were analyzed for phenolic compounds (Sandau et al., 2000). PCP was also reported in human plasma from Northern Quebec (Sandau et al., 2002). However in both cases PCP was assumed to be a degradation product of HCB.

Riget et al. (2008) assessed α, β and γ-HCH isomers in blubber taken from ringed seals harvested by subsistence hunters in West and East Greenland between 1986 and 2006. Lindane consistently showed the lowest concentrations of the three isomers and its concentration was lower in the more recent samples. West Greenland seals showed mean lindane levels of 1.91 ng/g lw in 2002 and 4.95 ng/g lw in 1995. East Greenland juvenile ringed seals had mean lindane levels of 2.57 ng/g lw in 2006 and 7.89 ng/g lw in 1986 while adult seals displayed an even larger temporal variation (2.7 ng/g lw in 2002 versus 9.3 ng/g lw in 1994). The higher concentrations in the seal populations of East Greenland are attributed to a prolonged period of higher lindane use in Europe compared to the Americas where the proximity of Europe allowed for greater atmospheric transport and delivery via ocean currents.

3. Conclusions

Surveys and long-term monitoring studies have demonstrated the presence of 9 of the 11 CUPs included in this review in the Arctic environment. Only dicofol has not been reported. The presence of these chemicals has mainly been reported in high volume air samples and in snow from Arctic ice caps and lake catchments in Alaska. Most of the CUPs that have been detected are or were at one time used at greater than 1000 t/y. Thus even when they have low predicted travel distances, the large amounts used, combined with relatively rapid transport in the atmosphere appear to result in detectable levels in abiotic samples. With the exception of lindane and α-endosulfan (Weber et al., 2006) there is insufficient data to assess whether air concentrations are resulting in net deposition to Arctic Ocean and lake waters or whether these waters are actually outgassing the chemicals. Other sources to the ocean could include long distance transport in ocean currents as has been postulated for β-HCH (Li et al., 2002). Glacier runoff could be an important source for some lakes and estuaries (Blais et al., 2001; Bizzotto et al., 2009).

In contrast to the growing dataset for abiotic matrices, there is only limited data indicating bioaccumulation of CUPs in arctic biota. Dacthal, trifluralin, methoxychlor, and PCP have been reported in biota. In most cases concentrations have been near or below limits of quantification. The exception is lindane which has been included in many studies and is routinely monitored for temporal trends in seals, beluga and other wildlife. Lindane has been shown not to biomagnify in arctic marine food webs.

However there are many other CUPs which have not been determined in Arctic environments. The discovery of the CUPs currently measured has been mainly serendipitous, a result of analyzing some samples using the same suite of analytes as used for studies in mid-latitude locations. A more systematic approach is needed to assess whether other CUPs might be accumulating in the arctic. A combination of modeling using the OECD LRT tool and more complex models which include emissions estimates such as GloboPOP (Wania and Mackay, 1995) would be useful to examine all CUPs for possible accumulation behavior.

Whether the presence of CUPs at low levels in arctic ecosystems has any significance biologically or results in risks for human consumers is another question that has not been adequately addressed. UNECE dossiers for pesticides trifluralin, PCP and dicofol have not addressed this issue for human or wildlife exposure in remote environments generally.

References

Ackerman LK, Schwindt AR, Simonich SLM, Koch DC, Blett TF, Schreck CB, et al. Atmospherically deposited PBDEs, pesticides, PCBs, and PAHs in Western U.S. National Park fish: concentrations and consumption guidelines. Environ Sci Technol 2008;42(7):2334–41.

Alexeeva LB, Strachan WMJ, Shlychkova VV, Nazarova AA, Nikanorov AM, Korotova LG, et al. Organochlorine pesticide monitoring of rivers of Russia flowing to the Arctic Ocean: 1990–1996. AMAP workshop on persistent organic pollutants (POPs) in the Arctic: human health and environmental concerns, Rovaniemi, University of Lapland Arctic Centre; 2000.

Alexeeva LB, Nazarova AA, Nikanorov AM, Korotova LG, Koreneva VI, Strachan WMJ, et al. Organochlorine pesticide and trace metal monitoring of Russian rivers flowing to the Arctic Ocean: 1990–1996. Mar Pollut Bull 2001;43(1–6):71–85.

Belfroid A, Blok H, Balk F. Addendum to the Risk Profile of Dicofol. Final report to the Ministry of VROM, Nijmegan, The Netherlands 2005 9R5744.01/R0007/ABE/ CKV/ Nijm.

Bidleman T, Macdonald R, Stow J. Sources, occurrence, trends and pathways in the physical environment. Canadian Arctic contaminants assessment report II. Ottawa: Ministry of Indican Affairs and Northern Development; 2003. 361 pp.

Bidleman TF, Kylin H, Jantunen LM, Helm PA, Macdonald RW. Hexachlorocyclohexanes (HCHs) in the Canadian Archipelago. 1. Spatial distribution and pathways of α-, β- and γ-HCHs in surface water. Environ Sci Technol 2007;41:2688–95.

Bizzotto EC, Villa S, Vaj C, Vighi M. Comparison of glacial and non-glacial-fed streams to evaluate the loading of persistent organic pollutants through seasonal snow/ice melt. Chemosphere 2009;74(7):924–30.

Blais JM, Schindler DW, Muir DCG, Sharp M, Donald D, Lafreniere M, et al. Melting glaciers: a major source of persistent organochlorines to subalpine Bow Lake in Banff National Park, Canada. Ambio 2001;30(7):410–5.

Borga K, Poltermann M, Polder A, Pavlova O, Gulliksen B, Gabrielsen GW, et al. Influence of diet and sea ice drift on organochlorine bioaccumulation in Arctic ice-associated amphipods. Environ Pollut 2002;117:47–60.

Boyd-Boland AA, Magdic S, Pawliszyn JB. Simultaneous determination of 60 pesticides in water using solid-phase microextraction and gas chromatography–mass spectrometry. Analyst 1996;121:929–38.

Braune BM. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975–2003. Environ Pollut 2007;148:599–613.

Buckman AH, Norstrom RJ, Hobson KA, Kamovsky NJ, Duffe J, Fisk AT. Organochlorine contaminants in seven species of Arctic seabirds from Northern Baffin Bay. Environ Pollut 2004;128:327–38.

Carroll J, Savinov V, Savinova T, Dahle S, McCrea R, Muir DCG. PCBs, PBDEs and pesticides released to the Arctic Ocean by the Russian rivers Ob and Yenisei. Environ Sci Technol 2008;42(1):69–74.

Chernyak SM, Rice CP, McConnell LL. Evidence of currently-used pesticides in air, ice, fog, seawater and surface microlayer in the Bering and Chukchi Seas. Mar Pollut Bull 1996;32:410–9.

De March BGE, De Wit C, Muir DCG, Braune BM, Gregor DJ, Norstrom RJ, et al. Persistent organic pollutants. AMAP assessment report. Arctic pollution issues, chapter 6. Oslo, Norway: Arctic Monitoring and Assessment Programme; 1998. p. 183–372.

de Wit CA, Fisk AT, Hobbs KE, Muir DCG, Gabrielsen GW, Kallenborn R, et al. AMAP assessment 2002: persistent organic pollutants in the Arctic. Oslo, Norway: Arctic Monitoring and Assessment Programme (AMAP); 2004. xvi + 310 pp.

Evenset A, Christenson GN, Skotvold T, Fjeld E, Schlabach M, Wartena E, et al. A comparison of organic contaminants in two high arctic lake ecosystems, Bjornoya (Bear Islan), Norway. Sci Total Environ 2004;318:125–41.

Fisk AT, Stern GA, Hobson KA, Strachan WJ, Loewen MD, Norstrom RJ. Persistent organic pollutants (POPs) in a small, herbivorous, Arctic marine zooplankton (*Calanus hyperboreus*): trends from April to July and the influence of lipids and trophic transfer. Mar Pollut Bull 2001;43(1–6):93-101.

Franz TP, Gregor DJ, Eisenreich SJ. Snow deposition of atmospheric semivolatile organic chemicals. In: Baker JE, editor. Atmospheric deposition of contaminants to the Great Lakes and coastal waters. Pensacola: SETAC Press; 1997. p. 73-107.

Garbarino JR, Snyder-Conn E, Leiker TJ, Hoffman GL. Contaminants in Arctic snow collected over northwest Alaskan sea ice. Water Air Soil Pollut 2002;139:183–214.

Gregor DJ, Gummer WD. Evidence of atmospheric transport and deposition of organochlorine pesticides and polychlorinated biphenyls in Canadian Arctic snow. Environ Sci Technol 1989;23(5):561–5.

Hageman KJ, Simonich SL, Campbell DH, Wilson GR, Landers DH. Atmospheric deposition of current-use pesticides in snow at national parks in the western United States. Environ Sci Technol 2006;40(10):3174–80.

Halsall CJ, Bailey R, Stern GA, Barrie LA, Fellin P, Muir DCG, et al. Multi-year observations of organohalogen pesticides in the Arctic atmosphere. Environ Pollut 1998;102: 51–62.

Hargrave BT, Phillips Georgina A, Peter Vass W, Philip Bruecker, Welch Harold E, Siferd Timothy D. Seasonality in bioaccumulation of organochlorines in lower trophic level Arctic marine biota. Environ Sci Technol 2000;34(6):980–7.
Hermanson MH, Isaksson E, Teixeira C, Muir DCG, Compher KM, Li Y-F, et al. Current-use and legacy pesticide history in the Austfonna ice cap, Svalbard, Norway. Environ Sci Technol 2005;39(21):8163–9.
Hobbs KE, Muir DCG, Born EW, Dietz R, Haug T, Metcalfe T, et al. Levels and patterns of persistent organochlorine in minke whale (*Balaenoptera acutorostrata*) stocks from the North Atlantic and European Arctic. Environ Pollut 2003;121:239–52.
Hoekstra PF, O'Hara TM, Texeira C, Backus S, Fisk AT, Muir DCG. Spatial trends and bioaccumulation of organochlorine pollutants in marine zooplankton from the Alaskan and Canadian Arctic. Environ Toxicol Chem 2002;21(3):575–83.
Hoekstra PF, O'Hara TM, Fisk AT, Borga K, Solomon KR, Muir DC. Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas. Environ Pollut 2003;124(3):509–22.
Hung H, Halsall CJ, Blanchard P, Li HH, Fellin P, Stern G, et al. Temporal trends of organochlorine pesticides in the Canadian Arctic atmosphere. Environ Sci Technol 2002;36(5):862–8.
Hung H, Blanchard P, Halsall CJ, Bidleman TF, Stern GA, Fellin P, et al. Temporal and spatial variabilities of atmospheric polychlorinated biphenyls (PCBs), organochlorine (OC) pesticides and polycyclic aromatic hydrocarbons (PAHs) in the Canadian Arctic: results from a decade of monitoring. Sci Total Environ 2005;342:119–44.
Hung H, Kallenborn R, Breivik K, Su Y, Brorström-Lundén E, Olafsdottir K, et al. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006. Sci Total Environ 2010;408:2854–73 (this issue).
Isaksson E, Hermanson M, Hicks S, Kgarashi M, Kamiyama K, Moore J, et al. Ice cores from Svalbard — useful archives of past climate and pollution history. Phys Chem Earth 2003;28:1217–28.
Jantunen, L.M., Wong, F., Bidleman, T.F. and Stern, G. (2007). Occurrence and levels of current-use and legacy pesticides in air: Leg 1 of ArcticNet 2007. Arctic Net. Collingwood, ONpp.
Kiely T, Donaldson D, Grube A. Pesticides industry sales and usage. 2000–2001 pesticide market estimates. Washington, DC: U.S. Environmental Protection Agency, Office of Pesticide Programs, Office of Prevention, Pesticides, and Toxic Substances; 2004. 48 pp.
Kimstach V, Dutchak S. Chapter 4. Persistent toxic substances (PTS) sources and pathways. Persistent toxic substances, food security and indigenous peoples of the Russian North. Final report. Oslo, Norway: Arctic Monitoring and Assessment Programme (AMAP); 2004. 33–80 pp.
Lakaschus S, Weber K, Wania F, Bruhn R, Schrems O. The air–sea equilibrium and time trend of hexachlorocyclohexanes in the Atlantic Ocean between the Arctic and Antarctica. Environ Sci Technol 2002;36(2):138–45.
Landers, D.H., Simonich, S.L., Jaffe, D.A., Geiser, L.H., Campbell, D.H., Schwindt, A.R., Schreck, C.B., Kent, M.L., Hafner, W.D., Taylor, H.E., Hageman, K.J., Usenko, S., Ackerman, L.K., Schrlau, J.E., Rose, N.L., Blett, T.F. and Erway, M.M. (2008). The fate, transport, and ecological impacts of airborne contaminants in western national parks (USA). Western Airborne Contaminants Assessment Project final report: volume 1. U. S. Environmental Protection Agency, Office of Research and Development, NHEERL, Western Ecology Division. Corvalis, ORpp.
Li Y-F, Macdonald RW, Jantunen LMM, Harner T, Bidleman TF, Strachan WMJ. The transport of β-hexachlorocyclohexane to the western Arctic Ocean: a contrast to α-HCH. Sci Total Environ 2002;291:229–46.
Li YF, Zhulidov AV, Robarts RD, Korotova LG. Hexachlorocyclohexane use in the former Soviet Union. Arch Environ Contam Toxicol 2005;48(1):10–5.
Mackay, N. and Arnold, D. (2005). Evaluation and interpretation of environmental data on endosulfan in Arctic regions. Report for Bayer CropScience, Report number 107. Cambridge Environmental Assessments (CEA). Cambridge, UK pp.
Moisey J, Fisk AT, Hobson KA, Norstrom RJ. Hexachlorocyclohexane (HCH) isomers and chiral signatures of αλπηα-HCH in the Arctic marine food web of the Northwater Polnya. Environ Sci Technol 2001;35(10):1920–7.
Morris AD, Sturman SK, Solomon KR, Muir DC, Teixeira C. Bioaccumulation and distribution of brominated flame retardants and current use pesticides in and Arctic marine food-web. SETAC North America 28th annual meeting, Milwaukee, WI; 2007.
Muir DCG, Howard PH. Are there other persistent organic pollutants? A challenge for environmental chemists. Environ Sci Technol 2006;40(23):7157.
Muir D, Zheng J. Environmental trends monitoring of new chemical contaminants in the Canadian High Arctic via ice and snow cores. In: Smith SL, Stow J, editors. Synopsis of research conducted under the 2006–2007, Northern Contaminants Program. Ottawa, ON: Indian and Northern Affairs Canada; 2007. p. 194–202.
Muir DCG, Teixeira C, Wania F. Empirical and modeling evidence of regional atmospheric transport of current-use pesticides. Environ Toxicol Chem 2004;23(10):2421–32.
Muir D, Alaee M, Teixeira C, Pacepavicius G, Spencer C, Scott B, et al. New contaminants in the Arctic and subarctic atmospheric and aquatic environments. CEPA resources report — FY 2006–2007. Burlington ON: Environment Canada, Aquatic Ecosystem Protection Research Division; 2007. 5 pp.
Pozo K, Harner T, Wania F, Muir DCG, Jones KC, Barrie LA. Toward a global network for persistent organic pollutants in air: results from the GAPS Study. Environ Sci Technol 2006;40(16):4867–73.
Pozo K, Harner T, Lee SC, Wania F, Muir DCG, Jones KC. Seasonally resolved concentrations of persistent organic pollutants in the global atmosphere from the first year of the GAPS Study. Environ Sci Technol 2009;43(3):796–803.
Qiu X, Zhu T, Yao B, Hu J, Hu S. Contribution of dicofol to the current DDT pollution in China. Environ Sci Technol 2005;39(12):4385–90.
Rasenberg MHC, van de Plassche EJ. Risk profile and summary report for dicofol. UNECE convention on long-range transboundary air pollution. Geneva, Switzerland; 2003. 42 pp.
Rice CP, Chernyak SM. Marine Arctic fog: an accumulator of currently used pesticides. Chemosphere 1997;35(4):867–78.
Riget F, Vorkamp K, Dietz R, Muir DCG. Levels and temporal trends of HCH isomers in ringed seals from West and East Greenland. J Environ Monitor 2008;10(8):935–40.
Rocque DA, Winker K. Biomonitoring of contaminants in birds from two trophic levels in the North Pacific. Environ Toxicol Chem 2004;23:759–66.
Sandau CD, Meerts IATM, Letcher RJ, Mcalees AJ, Chittim B, Brouwer A, et al. Identification of 4-hydroxyheptachlorostyrene in polar bear plasma and its binding affinity to transthyretin: a metabolite of octachlorostyrene. Environ Sci Technol 2000;34(18):3871–7.
Sandau CD, Ayotte P, Dewailly E, Duffe J, Norstrom RJ. Pentachlorophenol and hydroxylated polychlorinated biphenyl metabolites in umbilical cord plasma of neonates from coastal populations in Quebec. Environ Health Perspect 2002;110(4):411–7.
Savinov, V., Muir, D., Savinova, T., Gabrielsen, G.W., Alexeeva, L., Marasaev, S. and Zyryanov, S. (2008). Chlorinated hydrocarbons and polybrominated diphenyl ethers in glaucous gulls (*Larus hyperboreus*) from Barentsburg (West Spitsbergen). EMV-levels and trends of POPs in the Arctic pp.
Savinova T, Savinov V, Christensen G, Chernik G, Alexeeva L, Samsonov D, et al. Screening studies of POP levels in freshwater environment within the joint Russian–Norwegian border area. Organohalogens 2005;67:977–80.
Scheringer M, Jones KC, Matthies M, Simonich S, van de Meent D. Multimedia partitioning, overall persistence, and long-range transport potential in the context of POPs and PBT assessments. Integr Environ Assess Manag 2009;5(4):557–76.
Schmitt CJ, Zajicek JL, Ribick MA. National pesticide monitoring program: residues of organochlorine chemicals in freshwater fish, 1980–81. Arch Environ Contam Toxicol 1985;14:225–60.
Shen L, Wania F, Lei YD, Teixeira C, Muir DC, Bidleman TF. Hexachlorocyclohexanes in the North American atmosphere. Environ Sci Technol 2004;38(4):965–75.
Sinkkonen S, Paasivirta J. Polychlorinated organic compounds in the Arctic cod liver: trends & profiles. Chemosphere 2000;40:619–26.
Stern GA, Braekevelt E, Helm PA, Bidleman TF, Outridge PM, Lockhart WL, et al. Modem and historical fluxes of halogenated organic contaminants to a lake in the Canadian arctic, as determined from annually laminated sediment cores. Sci Total Environ 2005;342(1–3):223–43.
Stocker J, Scheringer M, Wegmann F, Hungerbuhler K. Modeling the effect of snow and ice on the global environmental fate and long-range transport potential of semivolatile organic compounds. Environ Sci Technol 2007;41(17):6192–8.
Strachan WMJ, Fisk A, Texeira CF, Burniston DA, Norstron R. PCBs and organochlorine pesticide concentrations in the waters of the Canadian Archipelago and other Arctic regions. AMAP workshop on persistent organic pollutants (POPs) in the Arctic: human health and environmental concerns, Rovaniemi, Arctic Monitoring Assessment Program/University of the Arctic; 2000.
Su Y, Hung H, Blanchard P, Patton GW, Kallenborn R, Konoplev A, et al. Spatial and seasonal variations of hexachlorocyclohexanes (HCHs) and hexachlorobenzene (HCB) in the Arctic atmosphere. Environ Sci Technol 2006;40(21):6601–7.
Su Y, Hung H, Blanchard P, Patton GW, Kallenborn R, Konoplev A, et al. A circumpolar perspective of atmospheric organochlorine pesticides (OCPs): results from six Arctic monitoring stations in 2000–2003. Atmos Environ 2008;42:4682–98.
UNECE. Trifluralin risk profile. UNECE convention on long-range transboundary air pollution. Geneva, Switzerland; 2007. 29 pp.
UNECE. Pentachlorophenol risk profile. UNECE convention on long-range transboundary air pollution. Geneva, Switzerland; 2008. 87 pp.
UNEP. Final act of the conference of plenipotentiaries on the Stockholm convention on persistent organic pollutants. Geneva, Switzerland; 2001. 44 pp.
UNEP. Lindane: risk profile. United Nations Environment Programme, Stockholm convention on persistent organic pollutants. Geneva, Switzerland; 2006. 25 pp.
US Environmental Protection Agency. Methoxychlor Reregistration Eligibility Decision (RED). EPA 738-R-04-010. Washington, DC: US Environmental Protection Agency, Office of Pesticide Programs; 2004. 9 pp.
US EPA. Reregistration Eligibility Decision (RED) DCPA (EPA 738-R-98-005). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 1998. 220 pp.
US EPA. Reregistration Eligibility Decision (RED) chlorothalonil (EPA 738-R-99-004). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 1999. 337 pp.
US EPA. Reregistration Eligibility Decision for diazinon (EPA 738-R-04-006). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 2004. 138 pp.
US EPA. Reregistration Eligibility Decision for chlorpyrifos (EPA 738-R-01-007). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 2006a. 260 pp.
US EPA. Reregistration Eligibility Decision for pentachloronitrobenzene (EPA OPP-2006-XXX). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 2006b. 127 pp.
US EPA. Data requirements for pesticide registration. Washington, DC: US Environmental Protection Agency; 2007. http://www.epa.gov/pesticides/regulating/data_requirements.htm#changes.
US EPA. Reregistration Eligibility Decision for pentachlorophenol (EPA 739-R-08-008). Washington, DC: Office of Prevention, Pesticides Protection and Toxic Substances, United States Environmental Agency; 2008. 103 pp.
Usenko S, Hageman KJ, Schmedding DW, Wilson GR, Simonich SL. Trace analysis of semivolatile organic compounds in large volume samples of snow, lake water, and groundwater. Environ Sci Technol 2005;39(16):6006–15.
Usenko S, Landers DH, Appleby PG, Simonich SL. Current and historical deposition of PBDEs, pesticides, PCBs, and PAHs to Rocky Mountain National Park. Environ Sci Technol 2007;41(21):7235–41.

USGS (2004). Pesticide National Synthesis Project, 2002 pesticide use maps (http://water.usgs.gov/nawqa/pnsp/usage/maps/). National Water-Quality Assessment (NAWQA) Program. US Geological Survey. Denver, COpp.

VanDijk HFG, Guicherit R. Atmospheric dispersion of current-use pesticides: a review of the evidence from monitoring studies. Water Air Soil Pollut 1999;115:21–70.

Verreault J, Muir DCG, Norstrom RJ, Stirling I, Fisk AT, Gabrielsen GW, et al. Chlorinated hydrocarbon contaminants and metabolites in polar bears (*Ursus maritimus*) from Alaska, Canada, East Greenland, and Svalbard: 1996–2002. Sci Total Environ 2005;351–352:369–90.

Vorkamp K, Riget F, Glasius M, Pecseli M, Lebeuf M, Muir D. Chlorobenzenes, chlorinated pesticides, coplanar chlorobiphenyls and other organochlorine compounds in Greenland biota. Sci Total Environ 2004;331:157–75.

Wania F, Mackay D. A global distribution model for persistent organic chemicals. Sci Total Environ 1995;160(161):211–32.

Weber J, Halsall CJ, Muir DCG, Teixeira C, Burniston DA, Strachan WMJ, et al. Endosulfan and gamma-HCH in the Arctic: an assessment of surface seawater concentrations and air–sea exchange. Environ Sci Technol 2006;40(24):7570–6.

Weber J, Halsall CJ, Muir D, Teixeira C, Small J, Solomon K, et al. Endosulfan, a global pesticide: a review of its fate in the environment and occurrence in the Arctic. Sci Total Environ 2010;408:2966–84 (this issue).

Welch HE, Muir DCG, Billeck BN, Lockhart WL, Brunskill GJ, Kling HJ, et al. Brown snow: a long-range transport event in the Canadian Arctic. Environ Sci Technol 1991;25(2):280–6.