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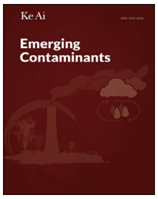

Levels and trends of current-use pesticides (CUPs) in the arctic: An updated review, 2010–2018

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ABSTRACT

Global regulations and many regional and national controls restrict the use of substances that exhibit the potential for environmental persistence and long-range transport. Nevertheless, many current-use pesticides (CUPs) continue to be newly discovered in remote regions, including the Arctic. The present review serves as an update, summarizing newly available information for CUPs in the Arctic environment and biota published from 2010 to 2018. Since 2010, at least seven new CUPs have been measured in Arctic media: 2-methyl-4- chlorophenoxyacetic acid (MCPA), metribuzin, pendimethalin, phosalone, quizalofop-ethyl, tefluthrin and triallate. Considering the large number of pesticides in current use, the number measured in the Arctic is very limited, however, modelling studies have identified additional CUPs as potential Arctic contaminants that have yet to be investigated in the Arctic. Owing to their recent detection, reports of CUPs in the Arctic are limited, but growing. CUPs have been reported in a wide range of abiotic Arctic matrices, including air, snow, ice, freshwater and seawater, indicating their capacity for long-range atmospheric transport, however, concentrations are generally low in comparison to legacy pesticides and other persistent organic pollutants (POPs). Recent food-web studies indicate CUPs can enter Arctic terrestrial and marine food chains, however, in contrast to POPs, the highest concentrations of many CUPs were found in lower trophic-level organisms, and the lowest concentrations detected in animals at the highest trophic levels (i.e, ringed seals, polar bear, caribou, and wolves) indicating significant trophic dilution. The detection of CUPs in the remote Arctic ecosystem reinforces the need for continued monitoring of both known and potential Arctic pollutants to prevent impacts on human and environmental health as the global arsenal of pesticides used in agriculture continuously changes.

1. Introduction

Since legacy pesticides such as DDT began to be regulated in the 1970s and 80s, there has been a massive increase in the use of replacement chemicals that serve similar purposes (i.e., reducing exposure to insect-borne diseases, protecting crops, and maintaining agricultural yields). In theory, these current-use pesticides (CUPs) can be licensed for use if no persistence in the environment is observed long after the period of use and if the chemicals have low bioaccumulation potentials, thus CUPs should have less environmental impact than the legacy pesticides they are replacing [1,2]. Nevertheless, many CUPs continue to be discovered in remote regions, including the Arctic, indicating that they are sufficiently

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persistent to undergo long-range transport and may represent an environmental concern [3]. Moreover, recent studies have indicated that several CUPs can bioaccumulate in Arctic biota [4,5].

Because CUPs were the subject of several recent reviews examining emerging Arctic contaminants [3,6,7], the present review, prepared as part of the recent Arctic Monitoring and Assessment Programme (AMAP) report on ‘Contaminants of Emerging Arctic Concern [8], serves primarily as an update (2010–2018), summarizing recently published and unpublished information for CUPs previously identified in the Arctic and several pesticides newly identified in the Arctic environment. For comparison purposes, Table 1 provides a summary of all CUP data reported in this updated review and previous ones for Arctic media (i.e. air, water, sediment, and biota).

1.1. Physical-chemical properties

The CUPs currently reported in the Arctic span diverse structural classes (Fig. 1 and Table 2), making it difficult to describe them as a group based on similar physical-chemical properties. However, owing to their occurrence in remote, high-latitude regions, they do share several general characteristics [3]. Many exhibit a moderate to low solubility in water and relatively low air-water partitioning (log K_{AW} values ranging from −3.5 to −6), allowing them to reach the Arctic, primarily through long-range atmospheric transport, with some contribution possible via ocean currents [9–12]. In addition, intermediate lipid solubility (log K_{OW} values ranging from 3 to ≈5) and high octanol-air partitioning (log K_{OA} values ranging from 7 to 11) impart the potential for many CUPs to bioaccumulate in food webs, especially marine and terrestrial food chains that contain multiple air-breathing consumers [4,5,13,14].

1.2. Sources, production, use and trends

According to the most recent report from the Food and Agriculture Organization (FAO), 4.1 million tonnes (Mt) of pesticides were used globally in 2016, with herbicides accounting for the largest proportion of pesticide use (Fig. 2) [15]. In the northern hemisphere, pesticide usage has increased 27% since 1996, reaching 3.2 Mt in 2016. Reports regarding the use of individual chemicals within countries are scarce, making it difficult to identify source areas and global trends of their production and use. However, most CUPs of Arctic concern have been recognized as high production volume (HPV) chemicals being produced or imported in amounts greater than 1000 tonnes per year by at least one of the 34 OECD member nations (Table 3) [16]. The CUPs detected in Arctic media most likely originate from agricultural applications as none of the chemicals listed in Table 2 were identified in a recent global survey of pesticides used to control insect-borne diseases, such as malaria or dengue fever [17].

The CUPs presently detected in the Arctic fall under varying levels of regulation [18–20] (Table 3). Most are still approved for use in the United States, Canada and Europe; however, some (e.g., chlorpyrifos; pentachloronitrobenzene (PCNB)) have restrictions on their use and others (e.g., endosulfan; dicofol) are beginning to be subject to domestic and international regulations. In 2011, endosulfan was added to the UN Stockholm Convention on Persistent Organic Pollutants (POPs) (Annex A) and as of 2016, all uses have expired in the United States, Canada and Europe [21]. In 2017, the POP Review Committee (POPRC) recommended the listing of dicofol to the Conference of the Parties to the Stockholm Convention [22,23] (www.pops.int/poprc13). In 2010, endosulfan and trifluralin, were under review for inclusion in the UNECE Long-Range Transboundary Air Pollution (LRTAP) Convention [24], however, as of 2018 no further action had been taken.

2. Environmental fate

2.1. Transformation processes

In agricultural and urban settings, pesticides are directly and extensively applied to large land areas. Therefore, degradation and transformation represents an important pathway for the removal of CUPs from the environment. As a group, pesticides span a wide range of structural conformations and physical-chemical properties, therefore the type(s) of transformation processes a given pesticide will undergo depends on its molecular configuration as well as the environmental conditions to which it is exposed.

Although degradation of pesticides can involve both abiotic and biotic transformation processes, biodegradation by microorganisms is generally recognized as the most important route of removal from the environment [25]. Organophosphate pesticides (i.e., chlorpyrifos; diazinon), phenoxy-herbicides (i.e., 2-methyl-4-chloro-phenoxyacetic acid, MCPA); pyrethroids (i.e., tefluthrin) and carbamates (i.e., triallate) are primarily degraded via microbial transformation. Several CUPs including chlorpyrifos [26], dacthal [27], endosulfan [28], and PCNB (also known as quintozene) [29] are biotransformed in animals, and rapidly depurated from the

Table 1
Summary of Arctic media for which CUP data have been previously (+) and newly (×) reported.

Common name	Air		Terrestrial		Freshwater			Marine		
	Air	Snow	Soil	Biota	Water	Sediment	Biota	Water	Sediment	Biota
Chlorothalonil	+,×	+,×		×	+			+,×		
Chlorpyrifos	+,×	+,×		×	+		+	+,×		+
Dacthal	+,×	+,×		+,×	+	+	+	+,×		+,×
Diazinon		+			+					
Dicofol	+,×							+,×		
Endosulfan	+,×	+,×		+,×	+		+	+,×	×	+,×
MCPA	×									
Methoxychlor	+	+		+	+		+	×		+
Metribuzin	×									
Pendimethalin	×									
Pentachloronitrobenzene (PCNB)	×	+,×		×	+			×		×
Phosalone	×									
Quizalofop ethyl	×									
Tefluthrin	×									
Triallate	×									
Trifluralin	+,×	+,×				+		+,×	×	

Historical data summarized from Hoferkamp et al. [3]; Weber et al. [6] and Vorkamp and Rigét [7].

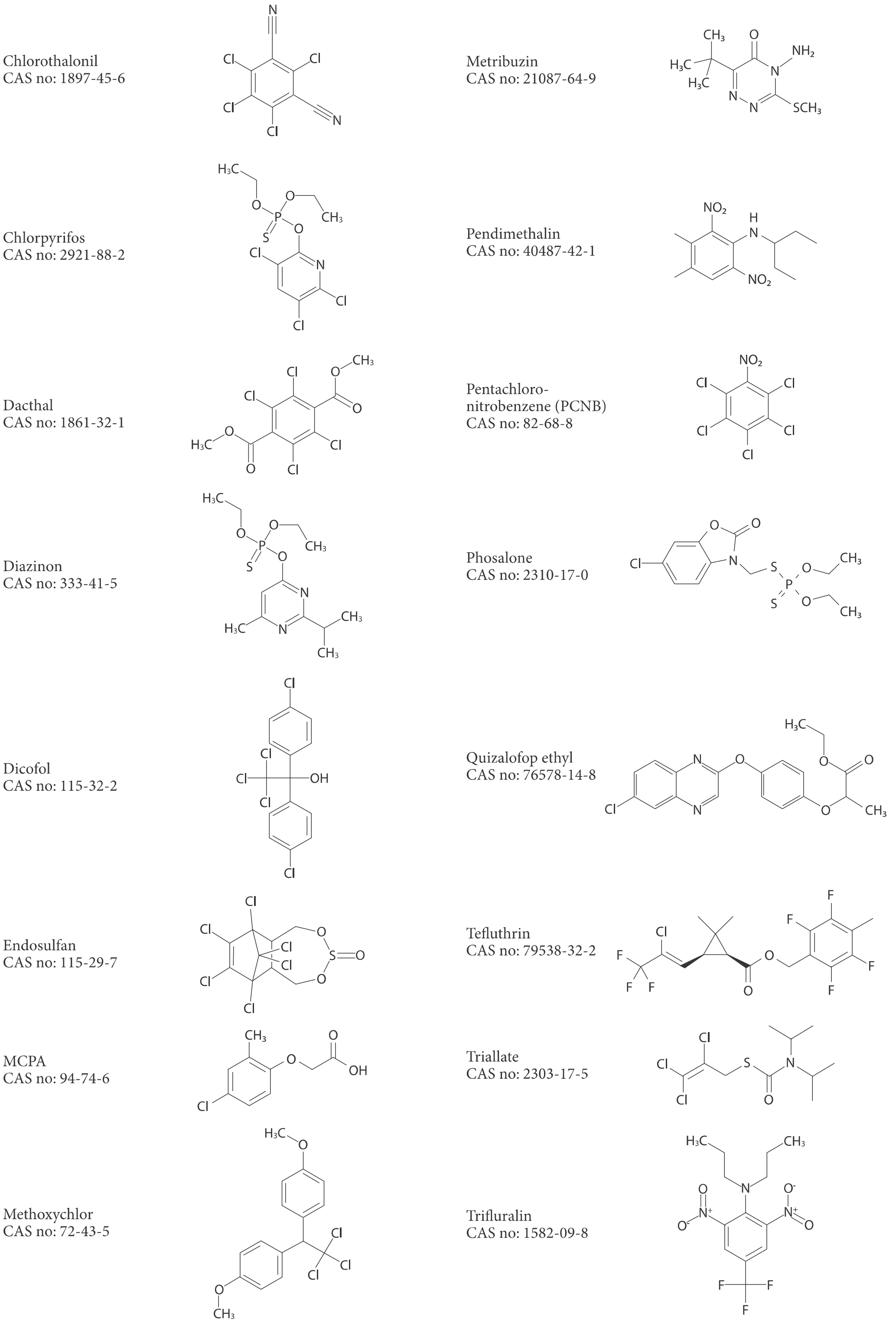

Fig. 1. Structure of current-use pesticides detected in Arctic media. Source: AMAP [8].

tissues of most mammals, particularly in comparison to recalcitrant organochlorine pesticides (OCPs). Chlorothalonil also seems to be eliminated rapidly from mammals, but this is more likely due to poor absorption across the gut epithelium than to biotransformation [30]. However, differences in biotransformation capacities do exist between mammalian species. Through evolution, some marine mammals (cetaceans and select pinnipeds), but not terrestrial mammals or polar bears, have lost a functional version of the paraoxonase 1 enzyme which hydrolyzes the -oxon metabolites of organophosphate compounds, such as those of chlorpyrifos and diazinon (chlorpyrifos oxon and diazoxon) [31]. Functional loss of this enzyme impairs the ability of some marine mammals to detoxify and excrete chemicals relative to terrestrial mammals and polar bears, and thus may contribute to observed differences in the

Table 2
Current-use pesticides detected in Arctic media.

Common name	CAS #	High production volume (HPV) chemical[a]	Class
Chlorothalonil	1897-45-6	×	Organochlorine fungicide
Chlorpyrifos	2921-88-2	×	Organothiophosphate insecticide
Dimethyl tetrachloro-terephthalate, Dacthal (DCPA)	1861-32-1		Organochlorine herbicide
Diazinon	333-41-5	×	Organothiophosphate insecticide
Dicofol	115-32-2 (*p,'p*-dicofol); 10606-46-9 (*o,p'*-dicofol)	×	Organochlorine insecticide
Endosulfan	115-29-7	×	Organochlorine insecticide
2-methyl-4-chloro-phenoxyacetic acid (MCPA)	94-74-6	×	Phenoxy herbicide
Methoxychlor	72-43-5		Organochlorine insecticide
Metribuzin	21087-64-9		Triazinone herbicide
Pendimethalin	40487-42-1	×	Dinitroaniline herbicide
Pentachloronitrobenzene (PCNB; Quintozene)	82-68-8		Organochlorine fungicide
Phosalone	2310-17-0		Organothiophosphate insecticide
Quizalofop ethyl	76578-14-8		Aryloxyphenoxypropionic herbicide
Tefluthrin	79538-32-2		Pyrethroid insecticide
Triallate	2303-17-5	×	Thiocarbamate herbicide
Trifluralin	1582-09-8	×	Dinitroaniline herbicide

[a] Produced or imported in greater than 1000 tonnes per year [16].

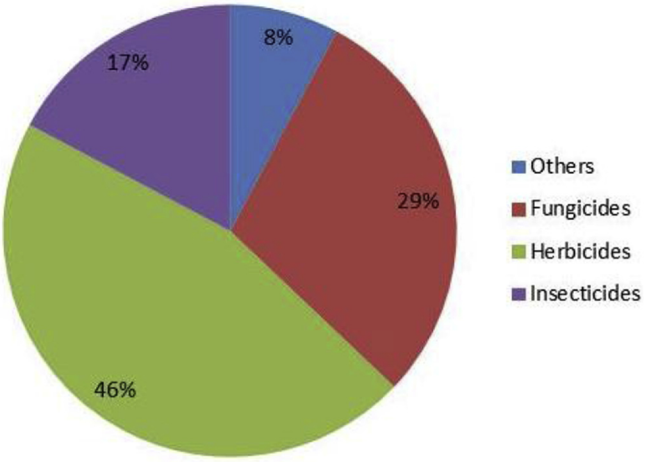

Fig. 2. Global pesticide market by use reported for 2016 [15]. 'Others'category includes rodenticides, botanicals, inorganics (sulfur, sulfuric acid) and mineral oil.

bioaccumulation and biological effects of CUPs and other pesticides among Arctic biota.

Although CUPs are effectively metabolized by many species, in some cases, the biotransformation products themselves can be bioaccumulative, as in the case of endosulfan sulfate, a biotransformation product of the isomers of endosulfan [4,32] and in the case of chlorpyrifos oxon, a more potent toxicant than the parent compound [33].

For some CUPs, abiotic processes such as chemical and photochemical reactions can also be important degradation pathways. Pyrethroids, carbamates, and dinitroaniline derivatives (i.e., trifluralin) are particularly susceptible to phototransformation, especially when discharged into surface waters. The primary degradation routes of the major pesticide classes were reviewed by Fenner et al. [25].

2.2. Modeling studies

Modelling efforts have been used to determine the global inventory, distribution, environmental fate, and Arctic contamination potential (ACP) of dicofol [34]. Data compiled from literature and field surveys suggest that 28,200 tonnes of dicofol were released into the environment between 2000 and 2012, with East and Southeast Asia, the Mediterranean coast, and Northern and Central America being the major sources. Applying two fugacity-based multimedia environmental models, the Berkeley-Trent (BETR)-Global simulation [35] and the Globo-POPs model [36,37], showed that dicofol is transported northward via atmospheric and oceanic transport and exhibits an ACP greater than several POPs such as heptachlor, aldrin, and hexabromocyclododecane (HBCDD).

Computational screening methods have been used to identify other CUPs with ACP. These predictive approaches entail the screening of large chemical databases for substances with the structural or functional properties to make them sufficiently persistent, bioaccumulative, and capable of long-range transport to reach the Arctic [12,38,39]. Screening of European and North American databases identified approximately 1200 current-use substances with the potential to reach the Arctic [40], including CUPs such as nitrapyrin [38], picloram [38,39], nitrofen [12,38], 2,4,6-trichlorophenol [38,39] and dinoseb [38,39]. Despite the predicted ACP for these pesticides, to our knowledge, they have yet to be investigated in the Arctic environment.

Models have also been used to assess the influence of a warming climate on the delivery of CUPs from the atmosphere via sea ice to the Arctic Ocean [41]. Legacy OCPs and CUPs were measured in air and surface seawater in the Beaufort Sea in 2008 and used to predict levels in sea-ice melt pond water. Predicted melt water concentrations were higher for CUPs (ranging from 27 ± 28 pg/L for dacthal to 258 ± 443 pg/L for chlorothalonil) than for legacy OCPs (ranging from 0.7 ± 0.4 pg/L for *trans*-chlordane to 41 ± 18 pg/L for dieldrin). The total annual release via melt water was estimated at 6 kg for dacthal, 16 kg for chlorpyrifos, 6 kg for α-endosulfan, and 54 kg for chlorothalonil, which equates to 2% of dacthal, 4% of chlorpyrifos, 10% of α-endosulfan, and 4% of chlorothalonil found in the standing stock of CUPs present in the upper mixed layer of the Beaufort Sea. Overall, the data suggest that sea ice meltwater is an important route for CUPs to the Arctic Ocean that will influence contaminant pathways as the Arctic's ice-scape changes under a warming climate.

Table 3
Regulatory status of current-use pesticides of relevance to the Arctic as of 2018 [18–20].

Chemicals	United States	Canada	European Union	Stockholm Convention	LRTAP[a] Convention
Chlorothalonil	Approved	Approved	Approved		
Chlorpyrifos	Approved for restricted use	Approved for restricted use	Approved for restricted use		
Dacthal	Approved	Approved	Approved		
Diazinon	Approved	Approved	Not approved		
Dicofol	Voluntarily phased out 2011	Not approved	Not approved	Under review	Under review (2010)
Endosulfan	Phased out, all uses expired July 2016	Phased out, all uses expired December 2016	Not approved	Listed, Annex A (2011)	Under review (2010)
MCPA	Approved	Approved	Approved		
Methoxychlor	Not approved	Not approved	Not approved		
Metribuzin	Approved	Approved	Approved		
Pendimethalin	Approved	Approved	Approved		
PCNB	Approved for restricted use	Approved for restricted use	Not approved		
Phosalone	Not approved	Not approved	Not approved		
Quizalofop ethyl	Approved	Approved	Not approved		
Tefluthrin	Approved	Approved	Approved		
Triallate	Approved	Approved	Approved		
Trifluralin	Approved	Approved	Not approved		Under review

[a] LRTAP: Long-Range Transboundary Air Pollution.

Table 4
Air concentrations of CUPs measured at the Canadian High Arctic station of Alert (Nunavut) between August 2006 and October 2009 under the Northern Contaminants Program [43,44]. Samples ($n = 68$) obtained using a high volume air sampler. Data shown are field and laboratory blank-corrected.

Chemical	Mean ± SD	Range	% detection
Chlorpyrifos	0.27 ± 1.0	<MDL – 6.8	19
Chlorothalonil	0.076 ± 0.15	<MDL – 0.65	31
Dacthal[a]	0.041 ± 0.057	<MDL – 0.23	66
α-Endosulfan[a]	3.4 ± 3.5	0.0060–14	100
β-Endosulfan	0.16 ± 0.54	<MDL – 3.2	37
Endosulfan sulfate[a]	1.9 ± 4.1	<MDL – 24	56
Metribuzin	0.029 ± 0.089	<MDL – 0.49	32
Pendimethalin	0.087 ± 0.19	<MDL – 1.1	25
PCNB	0.19 ± 0.80	<MDL – 5.5	26
Phosalone	0.12 ± 0.41	<MDL – 2.6	13
Quizalofop ethyl	0.042 ± 0.23	<MDL – 1.9	28
Trifluralin[a]	0.043 ± 0.056	<MDL – 5.5	76
Tefluthrin[a]	0.029 ± 0.041	<MDL – 0.15	59

MDL: Method detection limit. [a] Detectable in more than 40% of all samples.

3. Environmental concentrations

3.1. Air and precipitation

In 2005 and 2007, passive air samplers were installed at nine Canadian locations, including three sites within the Canadian Arctic [42]. Samplers were deployed at Coral Harbour, Cape Dorset and Arviat (Nunavut) for 90 days and screened for seven currently used herbicides, including 2,4-dichlorphenoxyacetic acid (2,4-D), bromoxynil, ethalfluralin, MCPA, mecoprop, triallate and trifluralin. Although most of the target analytes, including trifluralin, were undetectable; both triallate and MCPA were detected in air samples from Arviat, providing the first evidence of these CUPs in the Arctic region.

Starting in August 2006, air samples were collected at Alert (Nunavut, Canada) using a high volume air sampler (separate from the routine monitoring) equipped with a glass fiber filter and a PUF-XAD sandwich to collect CUPs (in both the particulate and the gas phases) [43,44]. Thirteen CUPs were sought and confirmed in these samples (gas and particulate phases were analyzed together). Table 4 summarizes results in air samples from Alert from August 2006 to October 2009. Air concentrations of all CUPs were low (<1 pg/m^3), and only dacthal, α-endosulfan, endosulfan sulfate, trifluralin and tefluthrin were detectable in more than 40% of samples.

During a GAPS (Global Atmospheric Passive Sampling) network special pilot study both PUF disks and sorbent impregnated polyurethane foam (SIP) disks collected CUPs at five locations: Alert (Nunavut, Canada), Little Fox Lake (Northwest Territories, Canada), Barrow (Alaska, USA), Ny-Ålesund (Svalbard, Norway), and Stórhöfði (Iceland) [45]. Dacthal, trifluralin, chlorpyrifos, pendimethalin and chlorothalonil were sought, but not always detected in air (Table 5). The highest concentrations of chlorothalonil, dacthal and pendimethalin were found in Iceland [45]. CUPs were also determined at several GAPS network sites as well as at other Arctic sites by Shunthirasingham et al. [46] over the period 2004–2008 (Table 6). These data were determined based mainly on 1-year deployments using XAD samplers and were reported by the authors as ng/sample. Results in Table 6 were expressed as pg/m^3 as described by Wania et al. [47] for comparison with the results of Koblizkova et al. [45] in Table 5. Dacthal and chlorothalonil, along with endosulfan were the most widely detected compounds.

Endosulfan has been routinely detected at Arctic sites using PUF disks deployed quarterly since 2005 [48]; the results of which have been reviewed previously [6]. Most recently, endosulfans were determined from XAD passive air samplers deployed for three one-year periods at Korean polar research stations at Ny-Ålesund between 2005 and 2009 [49]. Concentrations of α-endosulfan (range 17.7–130 pg/m^3; mean 41.3 pg/m^3) were in agreement with previous measurements of α-endosulfan at Ny-Ålesund (mean 37 pg/m^3) [50], while endosulfan sulfate was detected at concentrations below 1 pg/m^3 and β-endosulfan was not detected. α-Endosulfan was also investigated in air (sum of gaseous and particulate phases) at Villum Research Station, Station Nord in northern Greenland over the period 2008–2013 [51]. α-Endosulfan was detected in all 61 samples at a mean concentration of 3.3 pg/m^3 (range 0.09–14.1 pg/m^3). Bossi et al. [52] reported that α-endosulfan measured between 2008 and 2010 did not show any correlation with temperature indicating direct transport from primary sources as the origin.

Air concentrations of CUPs were measured onboard the *Oden* Icebreaker in July 2005 in the North Atlantic and the Canadian

Table 5
Arctic air concentrations (pg/m^3) of current-use pesticides measured via passive sampling at five Global Atmospheric Passive Sampling (GAPS) network sites [45]. Range of values based on PUF disk and SIP disk passive air samples deployed for ~3 months during May–June 2009. Malathion, chlorpyrifos and trifluralin were also determined by Koblizkova et al. [45] but were not detected (<LOQ) so is not included in the table.

		Chlorothalonil	Dacthal	Pendimethalin	Metribuzin
Alert, Canada	May–June 2009	<LOQ	<LOQ	<LOQ	<LOQ
Barrow, Alaska, USA	May–June 2009	<LOQ – 110	4.7–9.4	<LOQ	<LOQ
Little Fox Lake, Canada	May–June 2009	<LOQ	<LOQ – 37	<LOQ	<LOQ – 190
Stórhöfði, Iceland	May–June 2009	47–870	8.8–28	<LOQ – 270	<LOQ
Ny-Alesund, Norway	May–June 2009	<LOQ	19–37	<LOQ	<LOQ

Limit of quantification (LOQ) = ~1–5 pg/m^3.

Table 6
Arctic air concentrations (pg/m^3) of current-use pesticides measured via passive sampling at eight worldwide locations by Shunthirasingham et al. [46]. Data was reported by the authors as ng/sample, but are expressed here as pg/m^3 for comparison with the results of Koblizkova et al. [45] (Table 5). Conversion: ng/sampler converted to pg/m^3 using days of deployment and sampling rate of 0.5 m^3 per day (Wania et al. [47]) for Arctic locations.

Locations	Years	Trifluralin	Chlorothalonil	Dacthal	Pendimethalin	α-Endosulfan	β-Endosulfan	Endosulfan sulfate
Alert, Canada	2006–2007	<MDL	<MDL	<MDL	<MDL	<MDL – 8.8	<MDL	<MDL
Barrow, USA	2004–2007	<MDL – 0.3	<MDL – 1.1	<MDL – 0.3	<MDL	<MDL – 13	<MDL	<MDL
Dyea, USA	2005–2008	<MDL – 0.3	<MDL – 6.3	<MDL – 0.3	<MDL	<MDL – 2.8	<MDL	<MDL
St. Lawrence Island, USA	2004–2007	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Little Fox Lake, Canada	2006–2008	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Ny-Ålesund, Norway	2004–2007	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Pallas, Finland	2005–2008	<MDL	<MDL – 99	<MDL – 1.1	<MDL	<MDL – 15.6	<MDL – 0.3	<MDL-0.6
Stórhöfði, Iceland	2004–2008	<MDL	<MDL – 64	<MDL – 2.7	<MDL	<MDL – 17.2	<MDL – 0.5	<MDL-0.5

Method detection limit (MDL) = ~0.5 pg/m^3.

Table 7
Average air concentrations (pg/m^3) of current-use pesticides measured during oceanographic surveys in the western Arctic Ocean between 1999 and 2007–2013 [53].

Chemical	1999 Mean ± SD	2007–2013 Mean ± SD
Chlorpyrifos	NA	1.1 ± 1.3
Chlorothalonil	0.10 ± 0.20	0.90 ± 1.8
Dacthal	1.9 ± 1.6	1.1 ± 1.6
α-Endosulfan	4.8 ± 3.6	2.5 ± 1.5
β-Endosulfan	0.20 ± 0.46	0.12 ± 0.19
Endosulfan sulfate	NA	0.26 ± 0.55
Trifluralin	NA	0.39 ± 0.69

NA (not analyzed).

Table 8
Concentration range of current-use pesticides in Arctic air and seawater measured during an oceanographic cruise from the North Pacific to the Arctic Ocean in 2010 [55].

Chemical	Air (gas phase; pg/m^3)	Seawater (dissolved; pg/L)
Chlorpyrifos	0.50–2.0	0.08–0.85
Chlorothalonil	0.1–2.1	< MDL – 0.17
Dacthal	0.03–0.1	< MDL – 4
Dicofol	0.9–2.5	< MDL – 2.3
α-Endosulfan	0.40–1.6	0.04–0.4
β-Endosulfan	< MDL	< MDL
Trifluralin	0.9–2.2	< MDL – 0.009

MDL: Method detection limit.

Arctic Archipelago [43]. CUPs analyzed in these samples include dacthal, trifluralin, chlorothalonil, metribuzin, pendimethalin and endosulfans (α-, β-isomers and endosulfan sulfate). Back-trajectory analysis showed that during the entire sampling period there was no strong atmospheric input from potential source regions at lower latitudes. Although the samples were collected in summer when CUP use is generally high, the intrusion of more southerly air masses into the Arctic was minimal. Thus, these samples likely represented background Arctic concentrations. Accordingly, only a few CUPs were inconsistently detected at levels above method detection limits (MDLs). Dacthal concentrations exceeded the MDL of 0.6 pg/m^3 in two out of 20 samples and were higher than those observed at Alert (Table 4). Back trajectory analysis for one of the air samples with an elevated dacthal concentration indicated potential transport from the United Kingdom, an area known to have ongoing use of this pesticide. α-Endosulfan concentrations only exceeded the MDL of ~9 pg/m^3 once, and the trifluralin concentration was also high in this sample. β-Endosulfan concentrations were very low and endosulfan sulfate, metribuzin, and pendimethalin were not detectable. Air samples collected from oceanographic cruises in the northern Canada Basin and Canadian Arctic Archipelago have also been monitored for CUPs (Table 7) [53,54] and are discussed in section 4.1.

Air and seawater samples were collected during a research cruise extending from the East China Sea to the High Arctic between June and September 2010 and analyzed for six CUPs: chlorpyrifos, chlorothalonil, dacthal, dicofol, endosulfan and trifluralin (Table 8) [55]. Levels of α-endosulfan, chlorpyrifos and dicofol were one to two orders of magnitude higher in East Asia and the North Pacific compared to the Arctic, indicating Asian countries as potential sources. Gaseous concentrations of chlorothalonil, dacthal and dicofol also increased with latitude from the North Pacific toward the Arctic, which Zhong et al. [55] attributed to direct input from Russia and North America and/or transport via ocean currents.

In a separate study, chlorpyrifos, trifluralin, dacthal, chlorothalonil were investigated in Arctic air samples collected from Zeppelin (Svalbard, Norway) in 2017. With the exception of dacthal, which was measured in all five air samples at a mean concentration of 0.06 pg/m^3 (range 0.02–0.12 pg/m^3), these pesticides were found at levels below detection limits [56].

From 2009 through 2010, bi-monthly bulk atmospheric deposition samples (precipitation + dry particle) were also taken at an Arctic site near Abisko National Park in Stordalen mire, Sweden [57]. Two CUPs, trifluralin and chlorothalonil, were detected. Trifluralin was measured in 75% of samples, with an overall mean monthly deposition flux of 0.57 ng/m^2. Chlorothalonil was detected in 33% of samples, but at levels just above the instrument detection limit, therefore an average monthly flux could not be calculated.

In ice and snow, Ruggirello et al. [58] reported the historical profiles of CUPs in the Holtedahlfonna ice cap, one of the major ice fields on Svalbard, Norway. The ice core and snow samples were collected in 2005 and analyzed for 47 CUPs as well as legacy OCPs. Nine CUPs were observed in at least one of six core segments dating to 1953 and chlorpyrifos, dacthal, trifluralin and endosulfan-related compounds were the most prominent (Fig. 3). Chlorpyrifos had the highest fluxes of the CUPs, peaking at 808 pg/cm^2/y compared to 12.4 pg/cm^2/y for Σendosulfan (α-endosulfan, β-endosulfan and endosulfan sulfate). Ruggirello et al. [58] also compared fluxes from the Holtedahlfonna ice cap with earlier results in the Austfonna ice cap (220 km east-northeast of Holtedahlfonna), for the same time horizon [59]. They found that α-endosulfan and chlorpyrifos burdens at Austfonna were much higher by factors of about 60 and 13, respectively. The β-endosulfan burden had the opposite trend, being roughly 12 times greater at Holtedahlfonna. The dacthal burden at Holtedahlfonna was almost 3-fold higher than at Austfonna, although the peak inputs occurred over the same periods. The divergence of burdens or peak input periods of these compounds between these sites suggested that the general sources of these pesticides were different at least part of the time, and that Austfonna generally received the greater input. Air mass back trajectories over a 10-year period of comparison (1986–1995) showed air mass flow from Eurasia 74% of the time to Austfonna and 45% of the time to Holtedahlfonna, which may account for some of the differences.

Zhang et al. [60] also reported annual deposition profiles of CUPs in the Devon ice cap (Devon Island, Nunavut, Canada) based on a 6-m snow pit sampled in 2008. Dacthal and endosulfan sulfate were the most frequently detected CUPs with peak deposition fluxes of 1.0 and 0.4 pg/cm^2/y (Fig. 4). Endosulfan (sum of α and β isomers) predominated in 2003 and 2006, which together with air mass back trajectories suggested ongoing use in Eurasia as a possible origin. Comparison of results for deposition of CUPs in Arctic ice caps showed generally higher fluxes in Holtedahlfonna and Austfonna than in the Canadian Arctic [58,60]. This may be due to the proximity of the Svalbard glaciers to populated and agricultural regions in northern Eurasia compared to the remote Canadian High Arctic.

Chlorpyrifos, α- and β-endosulfan, endosulfan sulfate, dicofol and methoxychlor were measured in snow collected from northeast Greenland as part of a larger study (discussed in greater detail in section 3.4) examining the redistribution of organochlorine pesticides between snow, sea ice, and seawater during the spring melt period [61]. Samples were collected from the vicinity of the Villum Research Station, Station Nord in northeast Greenland over a five-week period during the early stages of the spring melt period in May–June 2015. Only chlorpyrifos and α-endosulfan were detected at concentrations above detection limits (Table 9). A similar study measured chlorpyrifos, chlorothalonil, dacthal, α-endosulfan, PCNB, and trifluralin in air and snow collected from Resolute Passage of the Canadian Arctic in 2012, along with sea-ice melt-pond water and seawater (discussed in greater detail in section 3.4) to determine atmosphere-ice-seawater exchange during the winter-summer transition [62]. With the exception of chlorothalonil and trifluralin, all pesticides were detected in air samples, and all six pesticides were detected in snow (Table 10).

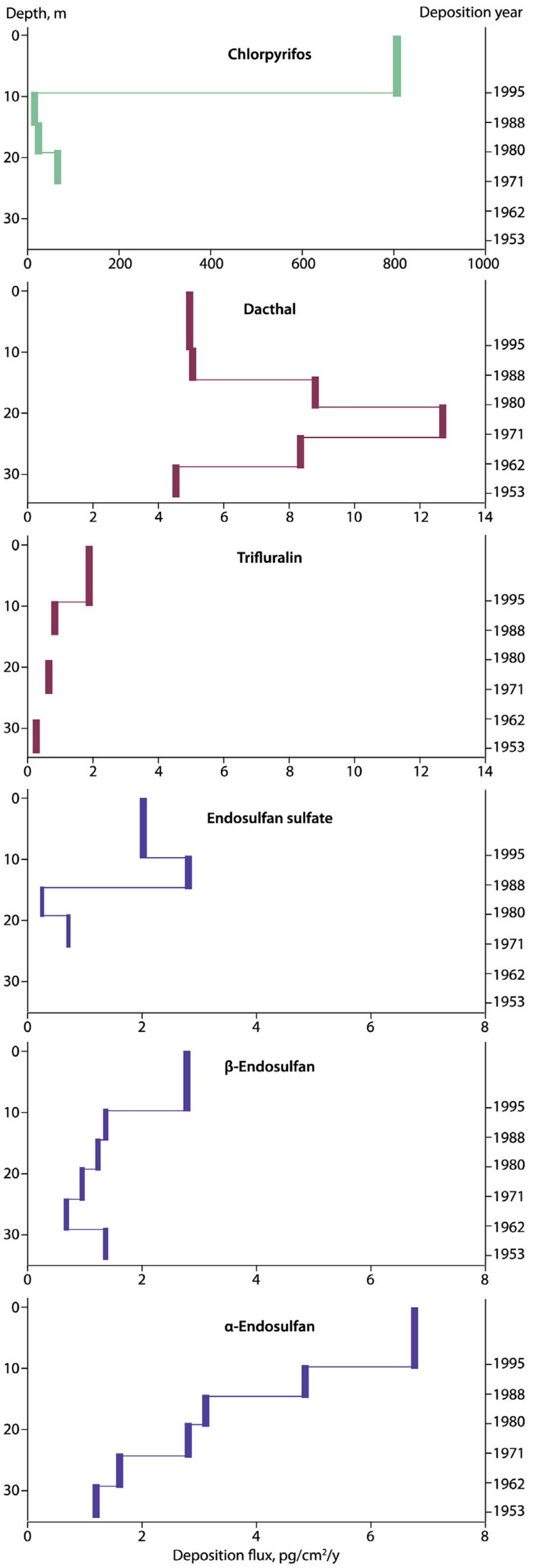

Fig. 3. Trends in the deposition of current use pesticides in the Holtedahlfonna ice cap (Svalbard, Norway) [58]. Source: AMAP [8].

Dacthal, chlorpyrifos and endosulfans have been determined in snowpack from Noatak National Preserve and Gates of the Arctic National Park and Preserve in Alaska, USA [63] and reviewed previously [3]. Despite the remote location of these parks and lack of agriculture in the region, CUP concentrations in snowpack were within range of those detected in parks at lower latitudes. New analyses of this data [64] have yielded air-to-snowpack flux estimates for CUPs at these Arctic locations. Pesticide flux (ng/m^2) was

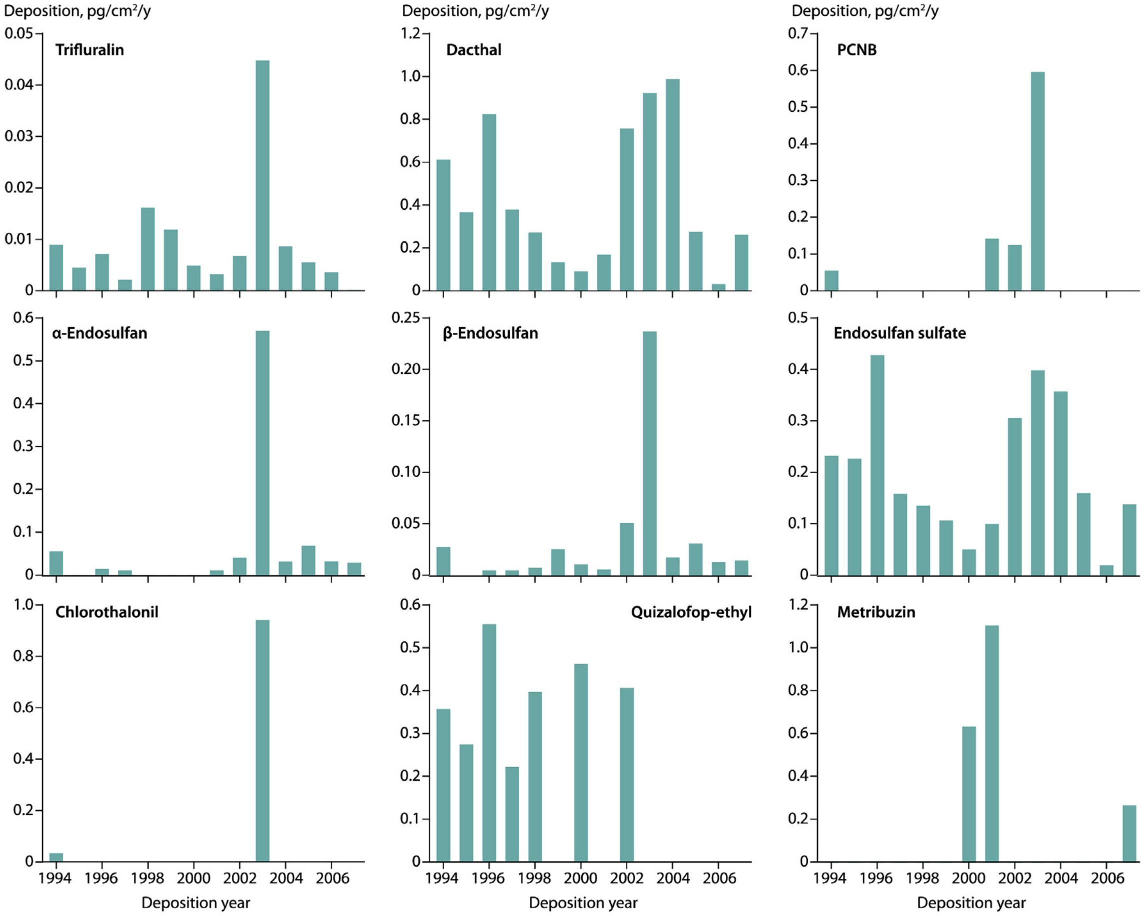

Fig. 4. Annual net deposition fluxes of CUPs in snow segments from a snow pit dug on the Devon Island Ice Cap in 2008 [60]. Source: AMAP [8].

Table 9

Concentration range of current-use pesticides in dissolved and particle phases of Arctic snow, sea ice, and seawater samples (pg/L) measured during the spring melt period in northeast Greenland in 2015 [61].

Chemical	Snow		Sea-ice		Seawater	
	Meltwater	Particles	Meltwater	Particles	Dissolved	Particles
Chlorpyrifos	6.2–11.5	<0.66	5.2–12.0	<0.86–15.5	0.74–1.0	<0.099–0.29
α-Endosulfan	2.6–58	<0.33–1.11	<2.2–6.2	<0.58	<0.50–1.1	<0.049
β-Endosulfan	<7.7	<0.97	<6.0	<1.5	0.69	<0.15
Endosulfan sulfate	<10.5	<1.3	<3.6	<1.9	3.2–4.2	<.0.19
Dicofol	<60.8	<137	<21.1	<179	<7.3	<21
Methoxychlor	<24	<6.4	<30.9	<3.8	<3.6	<0.38

Notes: < values are the higher of A) the method detection limit or B) the instrument limits of detection established for each individual sample.

Table 10

Concentrations (mean ± standard deviation) of current-use pesticides in Arctic air (pg/m^3), snow, sea ice melt pond water, and seawater (pg/L) in Resolute Passage of the Canadian Arctic during the winter-summer transition in 2012 [62].

Chemical	Air (pg/m^3)	Snow (pg/L)	Meltponds (pg/L)	Seawater (0 m) (pg/L)	Seawater (5 m) (pg/L)
Chlorpyrifos	0.10 ± 0.04	4.8 ± 1.3	14.4 ± 2.5	14.1 ± 6.1	10.5 ± 1/7
Chlorothalonil	<MDL	537.0 ± 563.7	<MDL	322 ± 180.0	<MDL
Dacthal	1.47 ± 1.18	30.4 ± 31.1	95.2 ± 71.0	9.3 ± 2.9	5.9 ± 1.4
α-Endosulfan	1.63 ± 0.13	2.4 ± 0.2	<MDL	3.7 ± 4.4	3.0 ± 2.8
PCNB	0.18 ± 0.07	2.6 ± 0.7	2.9 ± 0.9	2.9 ± 1.4	<MDL
Trifluralin	<MDL	1.2 ± 0.5	2.6 ± 0.6	2.1 ± 0.9	1.4 ± 0.2

determined as the concentration of pesticide measured in the melted snowpack sample (ng/m^3) multiplied by the snow depth in water equivalents (m) (SWE) for a specific winter season and provides an estimate of the total quantity of pesticide delivered to the ecosystem during the snow-accumulation period. Flux estimates calculated for snowpack samples collected between 2003

Table 11
Lipid equivalent (L_{eq})-normalized concentrations (ng/g lw) of current-use pesticides detected in vegetation of the Bathurst caribou herd region on the border of the Northwest Territories and Nunavut (geometric mean and 95% confidence interval). Values that share capital superscript letters are not significantly different (one-way ANOVA or Kruskal-Wallis ANOVA on ranks, $p > 0.05$). Selected results from Morris et al. [4].

	Lichen		Moss	Willow[a]	Grass and Sedge[a]	Mushroom
	(*Cladonia* sp.)	(*Flavocetraria* sp.)				
Samples (n)	6	6	6	6	8	5
L_{eq} (%)	1.7 ± 0.12	2.0 ± 0.18	0.61 ± 0.039	1.9 ± 0.40	1.2 ± 0.096	1.4 ± 0.67
PCNB	0.73^{AB}	0.89^{A}	0.10^{BC}	0.14^{ABC}	0.48^{ABC}	0.072^{C}
	(0.19–2.9)	(0.13–6.1)	(0.065–0.17)	(0.052–0.40)	(0.23–0.98)	(0.023–0.23)
Chlorothalonil	0.20^{A}	0.38^{A}	0.69^{A}	0.12^{A}	0.21^{A}	0.12^{A}
	(0.039–0.98)	(0.18–0.79)	(0.29–1.7)	(0.045–0.34)	(0.074–0.57)	(0.043–0.32)
Chlorpyrifos	0.20^{A}	0.32^{A}	0.49^{A}	0.18^{A}	0.17^{A}	0.85^{A}
	(0.053–0.75)	(0.051–2.0)	(0.22–1.1)	(0.075–0.43)	(0.073–0.42)	(0.11–6.4)
Dacthal	0.74^{AB}	0.99^{B}	0.19^{ABC}	0.14^{AC}	0.085^{C}	0.043^{C}
	(0.26–2.1)	(0.56–1.7)	(0.074–0.48)	(0.043–0.45)	(0.036–0.20)	(0.012–0.16)
ΣEndosulfan	7.7^{A}	7.4^{A}	2.1^{B}	2.0^{B}	0.92^{B}	ND
	(4.5–13)	(4.1–13)	(0.94–4.5)	(1.4–3.0)	(0.56–1.5)	(0.13–0.36)

[a] Only leaves of willow and leaves and stems of grasses were included in the extractions.

Table 12
Lipid-normalized tissue and body burden concentrations (ng/g lw) of current-use pesticides detected in caribou and wolves sampled from the Bathurst caribou herd region on the border of the Northwest Territories and Nunavut, Canada (geometric mean and 95% confidence interval). Mammalian total body burden concentrations were compared using Student's *t*-tests ($\alpha = 0.05$); where significant differences were detected in pair-wise concentrations, the significantly greater concentration is marked by an asterisk. Differences in concentration between tissues were tested using one-way ANOVA or Kruskal-Wallis ANOVA on ranks, with appropriate post-hoc tests; values that have capital superscript letters in common or have no superscripts are not significantly different. Selected results from Morris et al. [4].

	Caribou		Wolf		Caribou	Wolf
	Muscle	Liver	Muscle	Liver	Total body burden	Total body burden
Samples (n)	6	6	7	7	6	7
Lipid (%)	2.1 ± 0.85	4.4 ± 1.2	3.1 ± 1.5	4.2 ± 1.2	–	–
PCNB	0.044^{A}	0.21^{B}	0.11^{AB}	0.13^{B}	0.032	0.069
	(0.019–0.10)	(0.11–0.39)	(0.054–0.21)	(0.075–0.23)	(0.015–0.069)	(0.037–0.13)
Chlorothalonil	0.38^{A}	0.038^{B}	0.44^{A}	0.20^{AB}	0.25	0.27
	(0.25–0.56)	(0.015–0.098)	(0.12–1.5)	(0.13–0.3)	(0.16–0.37)	(0.076–0.92)
Chlorpyrifos	0.40^{A}	<MDL	<MDL	0.060^{B}	0.26*	0.023
	(0.20–0.79)	(0.035–0.060)	(0.045–0.11)	(0.030–0.12)	(0.14–0.52)	(0.015–0.035)
Dacthal	0.069	0.024	0.018	0.023	0.046*	0.0079
	(0.037–0.13)	(0.0092–0.060)	(0.009–0.036)	(0.012–0.042)	(0.024–0.085)	(0.0049–0.013)
ΣEndosulfan	0.57^{A}	2.0^{B}	0.17^{C}	0.14^{C}	0.40*	0.10
	(0.36–0.84)	(1.1–3.6)	(0.11–0.27)	(0.071–0.26)	(0.27–0.6)	(0.058–0.17)

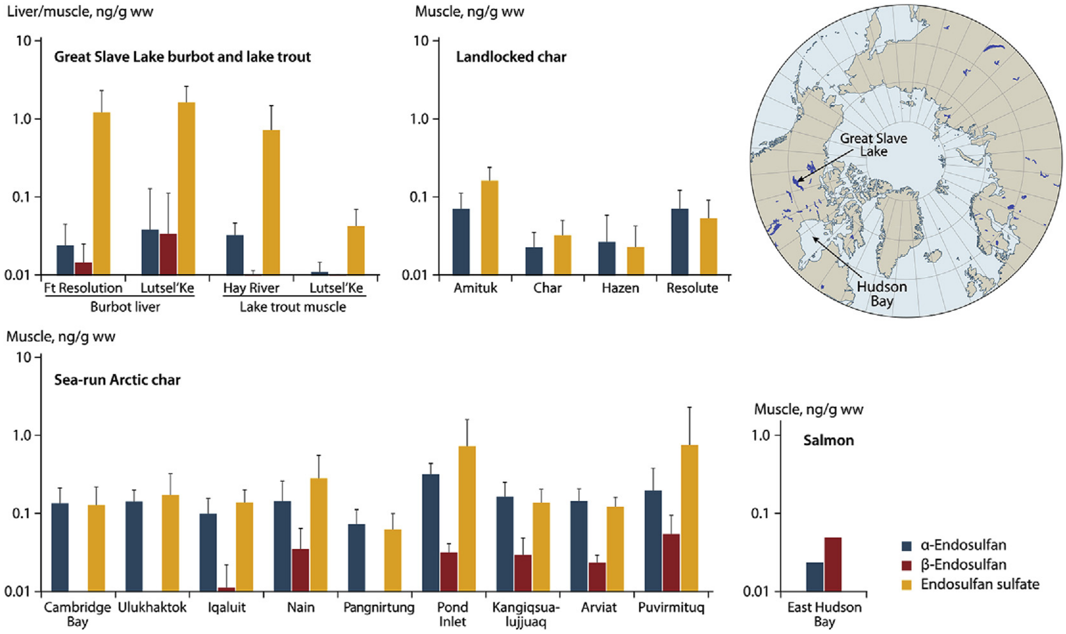

Fig. 5. Mean concentrations (±standard deviation) of α- and β-endosulfan and endosulfan sulfate in Great Slave Lake burbot liver and lake trout muscle and sea-run char muscle, landlocked Arctic char muscle, and salmon muscle from east Hudson Bay [70]. Source: AMAP [8].

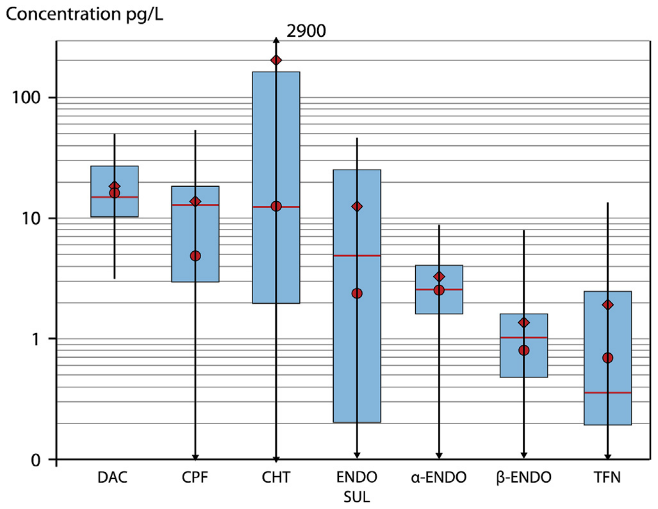

Fig. 6. Organochlorine pesticides and current-use pesticides in surface seawater collected during oceanographic cruises in the western Arctic Ocean between 1993 and 2013. Red bars are median concentrations, while red diamonds and dots are arithmetic means and geometric means, respectively. Vertical bounds of boxes indicate 25th and 75th percentiles, whiskers indicate the range, with lower arrows for samples below the instrument detection limit. Dacthal (DAC), chlorpyrifos (CPF), chlorothalonil (CHT), endosulfan sulfate (ENDO SUL), α-endosulfan (α-ENDO), β-endosulfan (β-ENDO), trifluralin (TFN) (Sources: Jantunen et al. [53],/AMAP [8].

and 2005 from Noatak and Gates of the Arctic parks ranged from 0.49 to 3.9 ng/m^2 for dacthal, from 6.7 to 31 ng/m^2 for chlorpyrifos and from 6.5 to 440 ng/m^2 for Σendosulfans. Although these parks are not subject to regional CUP usage, the authors hypothesize that the elevated pesticide levels observed there may be the result of enhanced air-to-snowpack partitioning of pesticides at cold temperatures.

3.2. *Terrestrial environment*

Current-use pesticides were investigated in a terrestrial food chain on the border of the Northwest Territories and Nunavut, Canada [4]. Between 2008 and 2010, samples were collected from regional flora and fauna, including lichens (*Cladonia* sp. and *Flavocetraria* sp.), crumpled leaf moss (*Rhytidium rugosum*), Arctic willow (*Salix pulchra*), cotton grass (*Eriophorum vaginatum*), aquatic sedge (*Carex aquatilis*), brown mushrooms (an unknown species), caribou (*Rangifer tarandus*), and wolves (*Canis lupus*). Samples were analyzed for seven CUPs, including chlorothalonil, chlorpyrifos, dacthal, PCNB, and endosulfans (α-endosulfan, β-endosulfan, and endosulfan sulfate). Low ng/g, lipid-equivalent corrected concentrations of the CUPs were detected in all vegetation species analyzed (Table 11). CUPs tended to bioconcentrate most effectively (on a volumetric basis) in lichens and moss, which have the greatest surface area to volume ratio of the vegetation and are relatively long-lived compared to the other vegetation species investigated. There was a positive correlation between the log K_{OA} and the bioconcentration factors for the CUPs, although dacthal did not fit this pattern and showed higher bioconcentration than would be predicted from its lipophilicity [4].

Concentrations of CUPs were lower in mammalian tissues than in vegetation (Table 12). The highest concentrations detected were for Σendosulfan in caribou liver, chlorothalonil in wolf muscle, as well as chlorpyrifos and Σendosulfan in caribou muscle [4]. A long-term study of reindeer and moose for the period 1987–2006 in Sweden also reported endosulfan concentrations below the detection limit in reindeer muscle [65]. Although measurable concentrations and bioconcentration factors demonstrated that these CUPs can effectively enter the terrestrial Arctic food chain, a subsequent calculation of trophic magnification factors by Morris et al. [4] indicated significant trophic dilution for all CUPs investigated, probably through biotransformation and excretion of these compounds, particularly in wolves. There was no intertrophic-level biomagnification from vegetation to caribou or caribou to wolves on a total body burden basis, and only limited tissue-specific biomagnification of some of the CUPs.

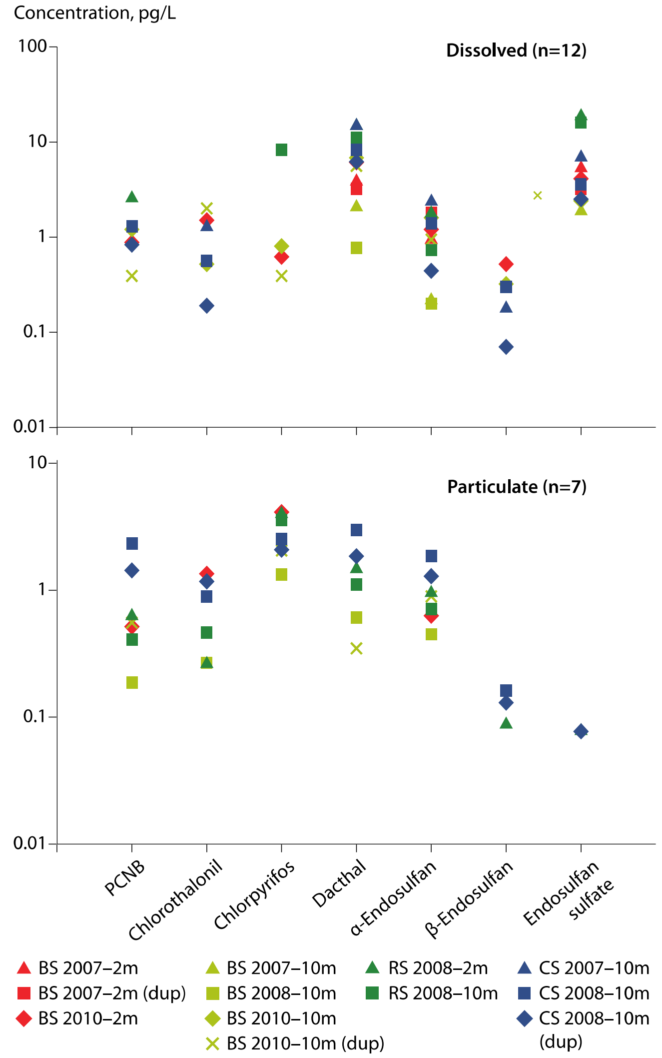

Fig. 7. Concentrations of current-use pesticides in the unbound and dissolved phase and the particulate phase of seawater collected from three Arctic locations: Barrow Strait (BS), Cumberland Sound (CS) and Rae Strait (RS), between 2007 and 2010 [5]. Only concentrations above the method detection limit (MDL) are shown. Source: AMAP [8].

In other terrestrial studies from Canada, dacthal, endosulfans (α-endosulfan, β-endosulfan and endosulfan sulfate), methoxychlor, and PCNB were measured in moose (*Alces alces*) liver from the Dehcho and South Slave regions of southwestern Northwest Territories, Canada in 2006 and 2010. With the exception of the endosulfans (Σendosulfan range <0.002–0.10 ng/g ww), all other CUPs measured were below detection limits (<0.02 ng/g ww) [66]. ΣEndosulfans in liver tissue of mountain goats (*Oreamnos americanus*) from the same regions were below detection limits, however, the detection limit reported was relatively high (29 ng/g lw) [67].

3.3. Freshwater environment

Törneman et al. [68] investigated the presence of chlorothalonil in surface water, sediment and soil samples collected in 2008 at Lake Abiskojaure (a background site) in northern Sweden but found no detectable concentrations of the pesticide. In another study, Törneman et al. [69] sought EU Water Framework Directive (WFD) priority substances mostly in water but also in some sediment samples from Lake Abiskojaure. Chlorpyrifos was not detected in water and α-, β-endosulfan and trifluralin were not detected in water or sediment.

CUPs have been determined in lake trout (*Salvelinus namaycush*), burbot (*Lota lota*), landlocked char and sea-run char (*Salvelinus alpinus*) from the Canadian Arctic and results to 2011 were summarized by Muir et al. [70]. Endosulfan was detected in burbot liver and lake trout muscle from Great Slave Lake, NWT, Canada (Fig. 5). Burbot liver had higher concentrations of endosulfan-related compounds than lake trout. Endosulfan sulfate, the stable degradation product of endosulfan was the main component, accounting for 95% of Σendosulfan in burbot liver and 89% of Σendosulfan in lake trout muscle. In landlocked char, α-endosulfan and endosulfan sulfate were present at similar sub-ng/g concentrations while β-endosulfan was below detection limits unlike in lake trout and burbot liver. Mean concentrations of α-endosulfan and endosulfan sulfate were higher in sea-run char (Fig. 5) than in landlocked char. Sea-run char α- and β-endosulfan concentrations were similar across the coastal arctic. Finally, dicofol was investigated in perch (*Perca fluviatilis*) from several lakes in Arctic Finland sampled in 2014 [71]. Four pooled samples were analyzed, each comprising 10 to 20 individuals. Dicofol was below the limit of quantification in each one.

3.4. Marine environment

3.4.1. Seawater

Surface seawater samples were collected during oceanographic cruises in the western Arctic Ocean and monitored for selected OCPs and CUPs between 1993 and 2013 (Fig. 6). Across all years, the detection rate for CUPs was highest for dacthal (98%), followed by α-endosulfan (97%), chlorpyrifos (95%), chlorothalonil (78%), β-endosulfan (76%), endosulfan sulfate (68%) and trifluralin (60%). Chlorothalonil, which exhibited the highest maximum concentration (2.9 ng/L), was also highly variable. In 2011, a year with high variation, the highest concentrations were associated with samples taken in regions with no ice cover and low salinity, indicating a potential relationship with recent melt episodes [53].

Surface seawater samples were collected from a research cruise along transects extending northward from the Sea of Japan into the Chukchi Sea and western Arctic Ocean in 2008 and were analyzed for α- and β-endosulfan, endosulfan sulfate, and methoxychlor [72]. Concentrations of α-endosulfan (<MDL–0.0980 ng/L), β-endosulfan (<MDL–0.0265 ng/L), endosulfan sulfate (<MDL–0.0875 ng/L) and methoxychlor (<MDL–0.0353 ng/L) in the Chukchi sea were comparable to those found in water samples from the Arctic Ocean collected during the same trip: α-endosulfan (0.0192–0.0528 ng/L), β-endosulfan (<MDL–0.0398 ng/L), endosulfan sulfate (<MDL–0.1225 ng/L) and methoxychlor (<MDL–0.0523 ng/L). Air and seawater samples were collected on a research cruise extending from the East China Sea to the High Arctic between June and September 2010 and analyzed for chlorpyrifos, chlorothalonil, dacthal, dicofol, endosulfan and trifluralin (Table 8) [55]. Spatial trends for CUPs in seawater were generally consistent with those observed in air.

The distribution of CUPs in seawater was also investigated at three Canadian Arctic locations: Barrow Strait, Cumberland Sound, and Rae Strait (Fig. 7) [5]. Overall, endosulfan sulfate (<MDL–19 pg/L) and dacthal (0.76–15 pg/L) were found in the highest concentrations and were the most consistently detected CUPs in the dissolved phase, followed by chlorpyrifos (<MDL–8.1 pg/L), PCNB (<MDL– 2.6 pg/L) and α-endosulfan (0.20–2.3 pg/L). Moreover, chlorpyrifos was detected in all particulate phase samples and had the highest concentrations in that phase (1.3–4.1 pg/L) (Fig. 7). In contrast to the dissolved phase, endosulfan sulfate was detected in only two particulate phase samples, at the lowest concentrations (<MDL–0.079 pg/L). Generally, the CUPs with the highest detection frequencies and concentrations in the particulate phase were those with higher K_{OW} values. However, environmental transformation of the endosulfans likely affected the consistency of this trend (e.g., β-endosulfan was detected in only three samples at low concentrations despite a relatively large K_{OW} value) [5].

As detailed in section 3.1, chlorpyrifos, α- and β-endosulfan, endosulfan sulfate, dicofol and methoxychlor were measured along with other organochlorine pesticides in snow, sea-ice, and seawater collected from northeast Greenland as part of a study examining the redistribution of organochlorine pesticides during the spring melt period [61]. Samples were collected from the vicinity of the Villum Research Station, Station Nord in northeast Greenland over a five-week period during the early stages of the spring melt in May–June 2015. Similar to findings in snow, chlorpyrifos and α-endosulfan were the only CUPs measured in sea-ice and seawater at levels above detection limits (Table 9). Although not found above detection limits in snow or sea ice, endosulfan sulfate was measurable in seawater. The authors note that chlorpyrifos and α-endosulfan exhibited different snow/sea ice/seawater distribution patterns compared to most other organochlorine pesticides (i.e, dieldrin) measured in the study. While most pesticides showed little change in concentration in snow or sea-ice throughout the duration of the study, concentrations of chlorpyrifos and α-endosulfan both appeared to increase in sea-ice during snowfall, suggesting that these chemicals may migrate downward from freshly fallen snow into the top layer of sea-ice.

In another study, sea-ice melt-pond water and seawater, collected from Resolute Passage of the Canadian Arctic in 2012, were analyzed along with air and snow (discussed in section 3.1) for a suite of CUPs to determine their atmosphere-sea ice-ocean dynamics during the winter to summer transition [62]. All six current-use pesticides investigated, including chlorpyrifos, chlorothalonil, dacthal, α-endosulfan, PCNB and trifluralin, were detectable in at least two of the matrices measured (Table 10). Chorothalonil and α-endosulfan were detected in snow, but not melt-pond water, suggesting that they either out-gassed back into the atmosphere with the commencement of the snow melt or were flushed out of snow into the ocean through widening brine channels before melt onset. However, four pesticides (chlorpyrifos, dacthal, PCNB and trifluralin) were present in both snow and melt-pond water, with melt-pond water having higher concentrations that would be reached by gas deposition only, suggesting there was atmospheric loading from falling and melting snow. Dacthal reached the highest concentration of all current use pesticides in melt-ponds, and exceeded concentrations in seawater by up to 16-fold, suggesting that drainage of dacthal-enriched pond water to the ocean during ice break-up provides an important ice-mediated annual delivery route to seawater.

3.4.2. Sediments

Some emerging pesticides were analyzed in surface sediments from a glacial fjord in Svalbard, Norway [73]. α-Endosulfan was detected in all samples (range 0.1–9.5 pg/g dw), while β-endosulfan was detected less frequently and at lower concentrations (range < MDL–3.0 pg/g dw). Both α- and β-endosulfan increased in

Table 13
Concentrations (geometric mean and 95% confidence interval) of current-use pesticides (ng/g lw) in Canadian Arctic marine biota from three locations [5]. Lipid-equivalent (% L_{eq}) normalization was applied for algae and plankton, all other animals were normalized to lipid content (% lipid). Values that have capital superscript letters in common or have no superscripts are not significantly different (One way ANOVA or Kruskal-Wallis ANOVA on Ranks with appropriate post hoc tests, $p < 0.05$).

Barrow Strait	Ice-algae	Plankton	Amphipods	Arctic cod	Seal blubber	Polar bear fat
n	10	18	8	23	18	8
ϕ_{Leq} (%)^{a}	1.5 ± 0.82	2.4 ± 0.73	2.7 ± 1.6	7.0 ± 3.9	84 ± 13	84 ± 5.9
Trophic Level	1.5 ± 0.29	2.1 ± 0.29	2.5 ± 0.31	3.6 ± 0.18	4.2 ± 0.18	5.4 ± 0.11
PCNB	0.24^{A}	0.25^{A}	0.15^{A}	0.35^{A}	0.011^{B}	<MDL
	(0.11–0.53)	(0.14–0.45)	(0.068–0.34)	(0.21–0.58)	(0.0039–0.029)	–
Chlorothalonil	1.1^{A}	<MDL	<MDL	0.10^{B}	<MDL	<MDL
	(0.65–2.0)	–	–	(0.072–0.15)	–	–
Chlorpyrifos	<MDL	0.41^{A}	<MDL	<MDL	<MDL	0.022^{B}
	–	(0.33–0.51)	–	–	–	(0.013–0.035)
Dacthal	<MDL	0.15^{A}	0.16^{A}	0.13^{A}	0.017^{B}	<MDL
	–	(0.075–0.29)	(0.089–0.29)	(0.076–0.22)	(0.011–0.027)	–
ΣEndosulfan	0.74^{A}	1.3^{AB}	2.9^{B}	1.8^{B}	0.070^{C}	0.13^{C}
	(0.45–1.2)	(0.87–1.8)	(1.9–4.4)	(1.4–2.4)	(0.054–0.092)	(0.048–0.36)

Rae Strait	Plankton	Polar cod	Seal blubber	Polar bear fat
n	4	6	6	7
ϕ_{Leq} (%)^{a}	3.82 ± 0.018	6.62 ± 2.61	90.1 ± 2.94	68.7 ± 8.24
Trophic Level	2.3 ± 0.25	3.6 ± 0.48	4.6 ± 0.32	5.6 ± 0.19
PCNB	0.50^{A}	0.071^{B}	0.059^{B}	0.011^{C}
	(0.13–1.9)	(0.026–0.20)	(0.044–0.081)	(0.0056–0.021)
Chlorothalonil	0.28	0.40	<MDL	0.16
	(0.00095–85)	(0.27–0.60)	–	(0.096–0.28)
Chlorpyrifos	0.33^{A}	<MDL	<MDL	0.032^{B}
	(0.11–0.95)	–	–	(0.013–0.076)
Dacthal	1.7^{A}	0.075^{B}	<MDL	0.015^{C}
ΣEndosulfan	11^{A}	0.79^{B}	0.082^{C}	0.27^{D}
	(5.3–24)	(0.58–1.1)	(0.043–0.16)	(0.11–0.65)

Cumberland Sound	Plankton	Char	Capelin	Ringed seal blubber	Polar bear fat
n	3	5	5	8	8
ϕ_{Leq} (%)^{a}	3.9 ± 0.11	8.9 ± 4.2	1.5 ± 0.36	90 ± 11	82 ± 13
Trophic Level	2.1 ± 0.075	3.1 ± 0.039	2.8 ± 0.065	3.3 ± 0.13	4.4 ± 0.25
PCNB	0.28^{ABC}	0.058^{B}	0.49^{C}	NM	0.0093^{D}
	(0.046–1.7)	(0.033–0.10)	(0.063–3.9)	–	(0.0037–0.023)
Chlorothalonil	0.47^{AB}	0.11^{AB}	1.4^{A}	NM	0.082^{B}
	(0.12–1.8)	(0.048–0.24)	(0.81–2.3)	–	(0.039–0.17)
Chlorpyrifos	1.1^{A}	0.11^{AB}	0.31^{A}	NM	0.016^{B}
	(0.010–131)	(0.013–0.93)	(0.017–5.5)	–	(0.0078–0.033)
Dacthal	0.074^{AB}	0.49^{AB}	2.1^{A}	NM	0.010^{B}
	(0.00033–17)	(0.15–1.6)	(1.3–3.5)	–	(0.0047–0.020)
ΣEndosulfan	8.7^{A}	2.2^{A}	5.1^{A}	0.22^{B}	0.22^{B}
	(3.1–25)	(1.0–5.0)	(2.2–12)	(0.14–0.27)	(0.099–0.49)

NM: not measured.

concentration from the outer fjord to the inner fjord, suggesting that runoff of glacial melt water may influence concentrations within the fjord. Trifluralin was also detected in fjord sediments but at lower concentrations; with the exception of an elevated concentration of 1.97 pg/g dw at a single station, trifluralin concentrations were all <0.15 pg/g dw.

3.4.3. Biota

With the exception of endosulfan, very few studies have reported CUP concentrations in Arctic marine animals. In 2012, eggs from common eider (*Somateria mollisima*), European shag (*Phalacrocorax aristoelis*), European herring gull (*Larus argentatus*) and American herring gull (*Larus smithsonianus*) were collected from the remote island Røst on the Norwegian coast and analyzed for three CUPs: pendimethalin, trifluralin, and methoxychlor; however all were found below the limits of detection [74]. Blood/plasma samples from polar cod (*Boreogadus saida*), capelin (*Mallotus villosus*), kittiwake (*Rissa tridactyla*) and glaucous gull (*L. hyperboreus*) and eggs from eider duck (*Somateria* sp.) and common guillemot (*Uria aalge*) collected from Arctic Norway in 2010–2011, contained no detectable levels of chlorpyrifos, dacthal, methoxychlor, or trifluralin. Chlorpyrifos, trifluralin, dacthal, chlorothalonil were also investigated, but not detected in eggs of common eiders, European shags, glaucous gulls, and black-legged kittiwakes, livers of American mink (*Neovision vison*) and blood of polar bears collected from Svalbard and Tromsø, Norway [56]. However, chlorpyrifos was detected in a single ringed seal (*Pusa hispida*) blubber sample from Arctic Norway at a concentration of 1.4 ng/g [75].

In a study of ringed seals from eastern Greenland between 1986 and 2012, α-endosulfan was above the detection limit (0.10 ng/g ww) in only 7 of 54 blubber samples, and none had concentrations of β-endosulfan above detection limits (0.10–0.25 ng/g ww) [32]. In contrast, 98% of the blubber samples had detectable levels of endosulfan sulfate, which was found at a mean concentration of 0.24 ng/g ww (0.27 ng/g lw). Dacthal, methoxychlor, α-, β-endosulfan and endosulfan sulfate were generally detected in <10% of ringed seals collected across the Canadian Arctic (2000s-2016) at concentrations around the limits of detection [76].

Moreover, total endosulfans (sum α- and β-endosulfan) were measured in blubber of killer whales (*Orcinus orca*) stranded or subsistence-harvested in southeast Greenland and the Faroe Islands between 2008 and 2014 [77]. Blubber concentrations of total endosulfans ranged from 197 to 13,000 ng/g lw (mean 2800 ng/g lw), however, the authors note that presence of killer whales in Arctic and sub-Arctic marine waters has historically been limited, thus the individuals sampled may represent Northern Atlantic populations that have expanded their ranges northward with recent declines in sea ice extent.

Bioaccumulation of PCNB, chlorothalonil, chlorpyrifos, dacthal, α-endosulfan, β-endosulfan and endosulfan sulfate was measured in three marine food chains in the Canadian Arctic Archipelago over the period 2007–2010 [5]. Samples comprised ice algae, plankton, amphipods, Arctic/polar cod, char, capelin, ringed seal blubber and polar bear (*Ursus maritimus*) fat (Table 13). Generally, low to sub-ng/g (lipid-equivalent normalized) concentrations of CUPs were observed, with lower trophic level biota such as ice algae, plankton and fish having the highest concentrations within the marine food web (Table 13). The bioaccumulation factors (BAFs) calculated for the CUPs (in water-respiring organisms only) indicate that CUPs are bioaccumulative with a broad range of log BAFs ranging from 3.8 ± 2.7 to 7.4 ± 7.1 (arithmetic means ± SEs, L/kg lipid weight). The lowest detectable concentrations were found in polar bear fat; PCNB, chlorpyrifos and dacthal were detected at very low concentrations (0.0061–0.032 ng/g lw). Endosulfan sulfate was not detected in polar bear fat, but both α- and β-endosulfan were detected at low concentrations (Σendosulfan <0.13–0.27 ng/g lw).

Combining the results for all three food chains into a food web spanning across the three locations showed that these CUPs all underwent trophic dilution (i.e. concentrations were highest in invertebrates and lowest in marine mammals, decreasing with increasing trophic level; Fig. 8). The relative consistency of marine mammal concentrations, lack of detection of endosulfan sulfate in polar bears, and the poor absorption and rapid depuration of many of the CUPs previously observed in laboratory studies [26–30] and trophic dilution of the CUPs through the food web, together suggest metabolic modification of many CUPs in mammals. Endosulfan sulfate is a biotransformation product of the isomers of endosulfan, and is bioaccumulative in a range of terrestrial and aquatic biota (seen in all lower trophic level biota reported by Morris et al. [5], however, it can also be further metabolized in some mammals to polar products that are more easily excreted (endosulfan ether, diol and lactone for example) [6,28]. Given that polar bears must be exposed to endosulfan sulfate from their diet of seal blubber, as well as forming it through the metabolism of endosulfan isomers, lack of detection of this compound in polar bear fat suggests further metabolism of endosulfan sulfate. This seems particularly likely given their high capacity to metabolize other organohalogen compounds [78], and that in more recent samples (post-banning of endosulfan under the Stockholm Convention) α-endosulfan, β-endosulfan and endosulfan sulfate as well as dicofol were sought but not detected in fat samples from western and southern Hudson Bay polar bears in Canada collected in 2012–2014 [79].

Although the CUPs investigated underwent food web dilution, it is important to note that significant biomagnification occurred in specific consumer-prey interactions [5]. ΣEndosulfan concentrations have been observed to biomagnify to a low degree between algae-plankton, plankton-amphipod and ringed seal-polar bear, with biomagnification factors (BMFs) ranging from 1.5 to 3.8. The highest BMFs observed were for the trophic transfer of β-endosulfan from blubber of seal to fat of polar bears (seal-bear) at Barrow Strait (BMF = 16 ± 4.9), dacthal in plankton-capelin from Cumberland Sound (BMF = 13 ± 5.0) and α-endosulfan in seal-bear from Rae Strait (BMF = 9.3 ± 2.8). Chlorothalonil also exhibited a small amount of biomagnification in bears from Rae Strait (BMF = 3.7 ± 0.63) and in capelin from Cumberland Sound (BMF = 2.7 ± 0.70) [5]. Although these compounds did biomagnify in some predator-prey interactions, concentrations remained very low relative to recalcitrant POPs in top predators [79].

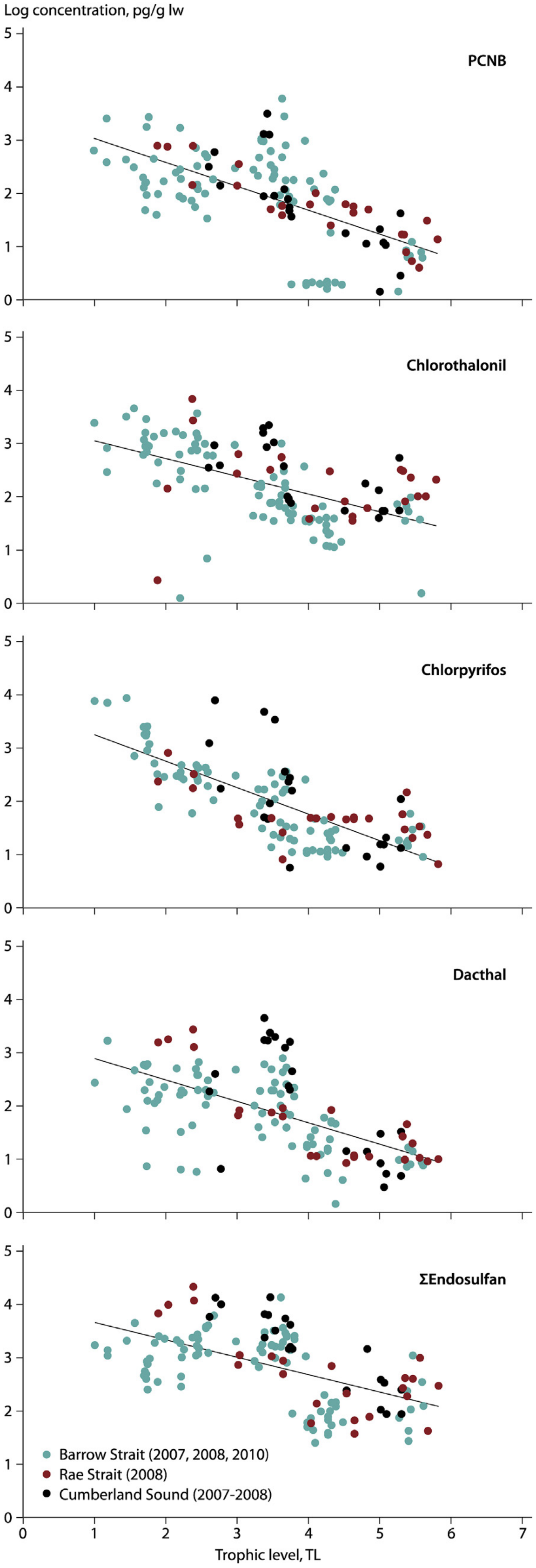

Fig. 8. Trophic dilution of current-use pesticides (CUPs) in the polar bear-ringed seal food web, based on data from three food chains sampled across the Canadian Arctic, illustrated by log plots of CUP concentration versus trophic level [5]. Source: AMAP [8].

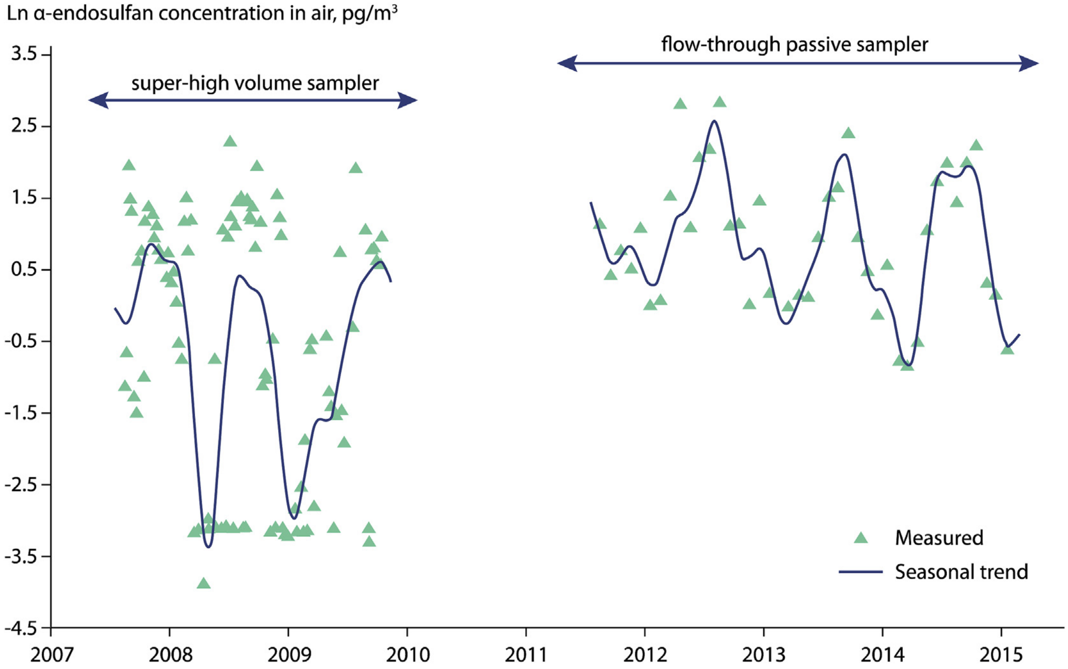

Fig. 9. Trends in concentration of α-endosulfan at Little Fox Lake (Yukon) from 2007 to 2014 using a super-high volume sampler and a flow-through passive sampler [80]. Source: AMAP [8].

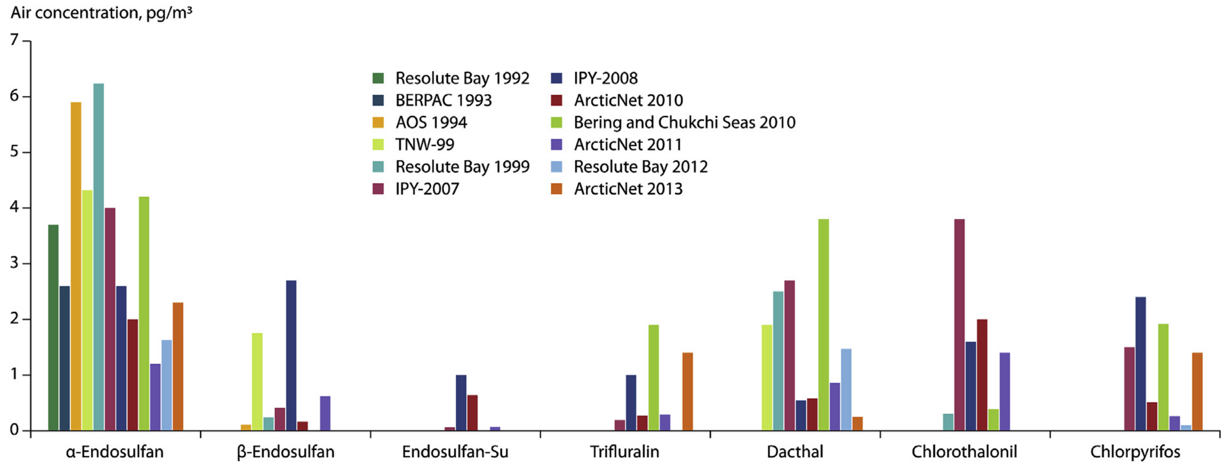

Fig. 10. Temporal trends of current use pesticides measured in air in the western Arctic Ocean collected during oceanographic surveys between 1999 and 2013 [53,54]. Data show arithmetic mean concentrations. Expeditions, listed by year, traversed the Bering-Chukchi Seas (BERPAC-1993), central Canada Basin to the North Pole (AOS-1994) and the Canadian Archipelago (TNW-1999, IPY-2007, IPY-2008, ArcticNet-2010, ArcticNet-2011, ArcticNet-2013). Endosulfan-Su = Endosulfan sulfate. Source: AMAP [8].

4. Temporal trends

4.1. Air and seawater

Owing to their recent discovery in the Arctic, data on long-term regional temporal trends for most of the CUPs highlighted in this review are scarce.

Atmospheric concentrations of α-endosulfan at Alert (Nunavut, Canada) were reported in the recent AMAP Trends overview [80]. At Alert, α- and β-endosulfan were sought in the routine air monitoring samples obtained with a super-high volume air sampler between 1993 and 2011. β-endosulfan was mostly found below detection limits. The time series of α-endosulfan showed little change or a slightly declining trend with a half-life of 37 years [80], indicating the restrictions on endosulfan put in place in Europe and North America prior to its addition to the Stockholm Convention in 2011 were not evident in its temporal trend in Arctic air.

Yu et al. [81] determined α-endosulfan in air at Little Fox Lake (Yukon, Canada) using a flow-through sampler with collection on polyurethane foam [82] as well as a high-volume sampler similar to the one used at Alert. Concentrations of α-endosulfan showed a decrease during the period in which the flow-through sampler was used (2011–2014). In the earlier period, in which a high-volume sampler was used (2007–2009), average concentrations determined using a digital filter technique were relatively constant (Fig. 9).

Several CUPs have been monitored in air and surface seawater samples collected from oceanographic cruises in the Bering and Chuckchi seas, northern Canada Basin, and Canadian Arctic

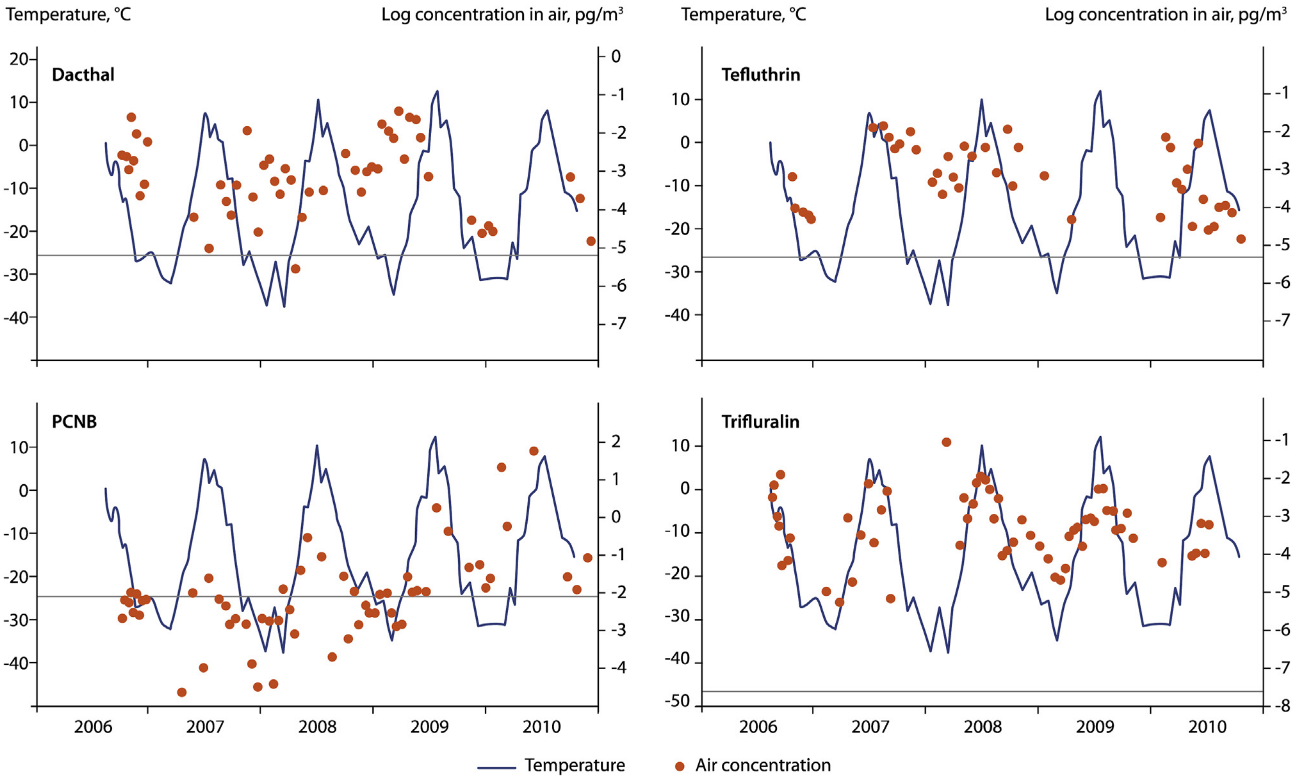

Fig. 11. Trends in concentration of dacthal, PCNB, tefluthrin, and trifluralin in air at Alert, Nunavut (2006–2010) [43]. Horizontal grey lines indicate mean blank concentrations. Source: AMAP [8].

Archipelago between 1993 and 2013 (Table 7, Figs. 6 and 10) [53,54]. Concentrations of most CUPs in air were stable or had lower concentrations in 2010–2013 compared to earlier sampling cruises. Within a year of a temporary ban of PCNB by Canada and the U.S. in December 2010, mean atmospheric levels of pentachlorothioanisole (PCTA), a degradation product of PCNB, dropped by a factor of 4 (from 0.21 ± 0.11 pg/m^3 (2007–2010) to 0.051 ± 0.020 pg/m^3 (2011/2013) [53]. At the tail end of a period during which global use of dicofol was thought to have decreased (from 3400 tonnes in 2000 to 730 tonnes in 2012) [34],

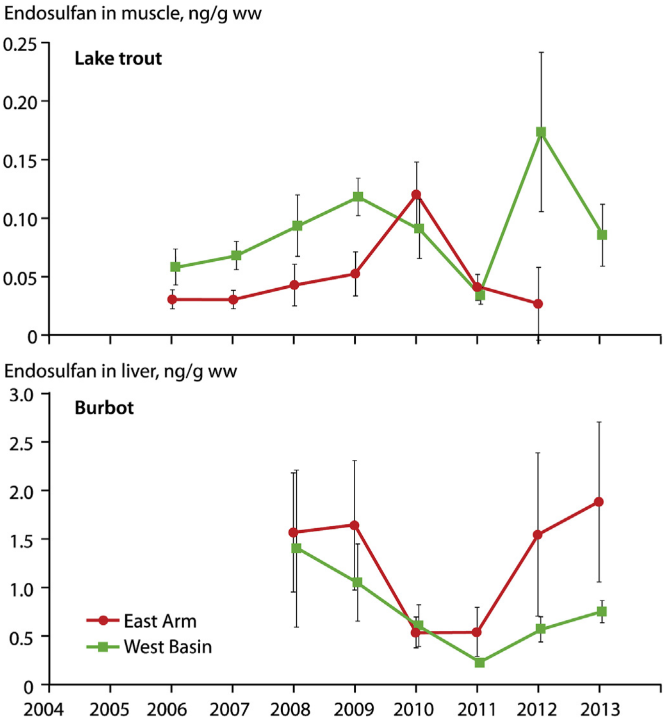

Fig. 12. Temporal trends in the concentration of Σendosulfan (sum α, β and endosulfan sulfate) in lake trout muscle and burbot liver (±95% confidence intervals) from two sites in the Great Slave Lake in the Canadian Arctic (Muir et al. [70], Evans and Muir, unpubl. data). Source: AMAP [8].

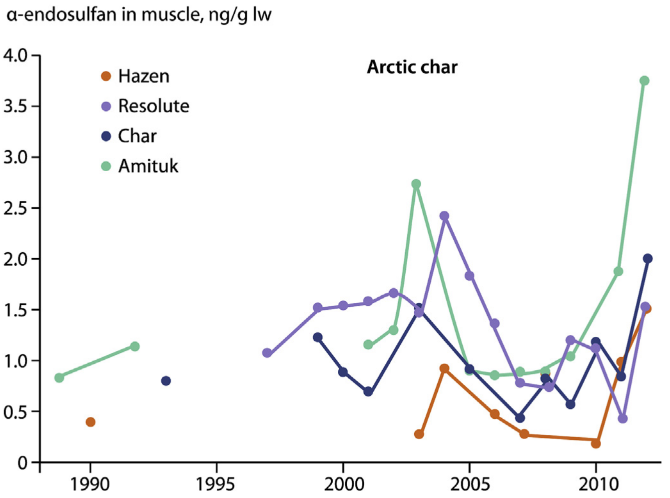

Fig. 13. Temporal trends in concentration of α-endosulfan (geometric means, ng/g lw) in landlocked Arctic char from Canadian lakes from Muir et al. [70] and Muir, unpubl. data. Error bars are omitted for clarity. Source: AMAP [8].

concentrations in air sampled onboard the *Amundsen* Icebreaker in the Canadian Arctic Archipelago during 2011–2013 averaged 1.9 pg/m^3 [54]. In surface seawater collected during the same oceanographic cruises, concentrations of some CUPs increased (e.g. α-endosulfan, chlorothalonil, trifluralin), while concentrations of others (e.g. chlorpyrifos, dacthal) showed no significant change over time [53].

Short term seasonal trends for some CUP concentrations in Arctic air have also been identified. Dacthal, PCNB, tefluthrin, and trifluralin have been monitored in air at Alert (Nunavut, Canada) since mid-2006 [43]. Concentrations of dacthal and trifluralin were found to be high in summer, whereas PCNB concentrations were high in winter (Fig. 11). No apparent seasonality was seen for air concentrations of chlorpyrifos, endosulfan sulfate, and tefluthrin at this site. Yu et al. [81] also observed seasonal trends in α-endosulfan at Little Fox Lake (Yukon, Canada) (Fig. 10). Short-term temporal trends could be the result of seasonal changes in the prevailing air trajectories; using source-contribution function calculations, Yu et al. [81] determined the endosulfan in air at Little Fox Lake (Yukon, Canada) likely originates from the Pacific, East Asia, and northern Canada during the warm seasons while in cold seasons, the Pacific Rim region is the predominant source. Alternatively, seasonal trends could be attributed to differences in the deposition and volatilization of CUPs from air to snow (winter) or soil and vegetation (in summer) [83].

4.2. Freshwater fish

Long-term monitoring of burbot, lake trout, and landlocked char has yielded temporal trends of endosulfan for several Canadian lakes [70; Evans and Muir, unpubl. data]. In lake trout from the Great Slave Lake, Σendosulfan concentrations generally increased between 2006 and 2010 and then declined, although more recent measurements indicate a shift to higher concentrations in west basin lake trout (Fig. 12). In burbot, Σendosulfan declined between 2008 and 2011 but showed an increase from 2011 to 2013.

Landlocked Arctic char from Resolute, Char and Amituk Lakes on Cornwallis Island and from Lake Hazen in Quttinirpaaq National Park on Ellesmere Island have been consistently monitored for endosulfan isomers since the early 1990s; measurements prior to 2009 did not include endosulfan sulfate [70; Muir, unpubl. data]. Although no temporal trend for α-endosulfan was apparent in char from Lake Hazen, fish from other lakes, notably Amituk and Resolute, showed increases until 2003–2004 and then declined. More recent data show striking increases in lipid-normalized concentrations of α-endosulfan in Amituk, Char and Resolute Lakes (Fig. 13).

4.3. Marine mammals

Endosulfan was investigated in a temporal study of ringed seals from eastern Greenland between 1986 and 2012 (Fig. 14) [32]. Blubber concentrations of endosulfan sulfate decreased over this time period by −5.6% annually, however temporal trends of α- and β-endosulfan could not be established because concentrations were below detection limits in the majority of the samples.

Trends in Σendosulfan were also determined in ringed seal blubber in the Canadian Arctic starting in 2007 (Fig. 15). No significant trends were apparent in samples from East Baffin Island, Lancaster Sound, western Hudson Bay or the Beaufort Sea [76].

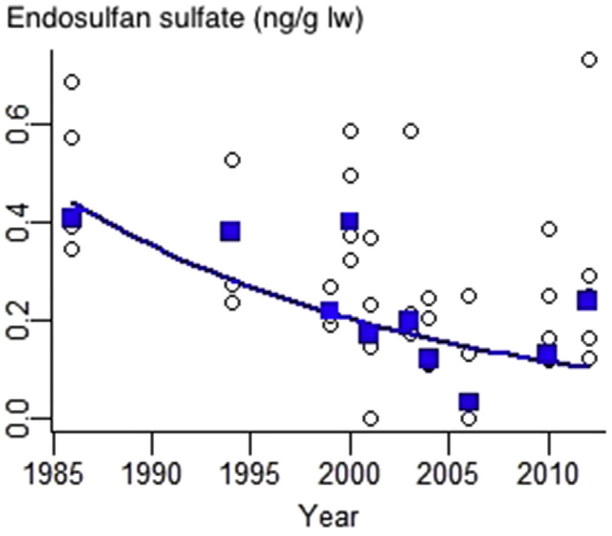

Fig. 14. Concentrations of endosulfan sulfate in individuals (open circles) and annual median concentrations (filled squares) in ringed seal blubber from Ittoqqortoormiit, East Greenland. The blue line is the log-linear regression line. Reproduced from Vorkamp et al. [32].

5. Conclusions and recommendations

Global regulations and many other regional and national controls restrict the use of substances that exhibit environmental persistence and long-range transport potential. Accordingly, CUPs should have minimal, if any, presence in the Arctic environment and biota. Yet, since 2010, at least seven new pesticides have been measured in Arctic media: MCPA, metribuzin, pendimethalin, phosalone, quizalofop-ethyl, tefluthrin and triallate. Considering the large number of pesticides in current use, the number measured in the Arctic is still very limited.

Owing to their recent detection in the Arctic, reports of CUPs in the circumpolar region are limited, but growing. CUPs have been measured in a wide range of abiotic Arctic matrices, including air, snow, ice, freshwater and seawater, indicating long-range atmospheric transport. Identifying spatial and temporal trends in CUPs within the Arctic region is difficult due to the general lack of data, however, results for ice caps in the Norwegian and Canadian Arctic provide some insights into spatial differences. In general, a higher deposition of CUPs was observed in Arctic Norway (Holtedahlfonna

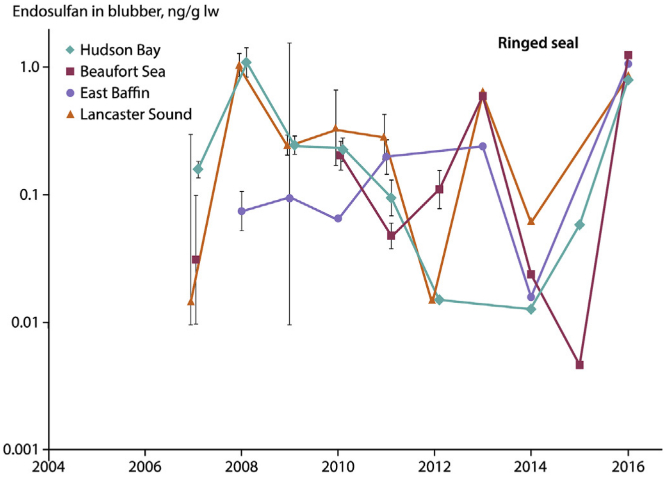

Fig. 15. Temporal trends in concentration (geometric means ± 95% CI) of Σendosulfan (sum of α-endosulfan, β-endosulfan and endosulfan sulfate) in blubber of ringed seals (females and juveniles) from four regional locations in the Canadian Arctic [76]. East Baffin samples were collected from Pangnirtung, Nunavut (2008–2012) and Nain, Labrador (2013–2016). Figure adapted from AMAP [8].

ice cap) than in the Canadian Arctic (Devon ice cap). Regional differences likely stem from differences in prevailing air trajectories and the closer proximity of the Norwegian Arctic to the populated and agricultural regions of northern Eurasia.

With the exception of endosulfan, long-term temporal trends of CUPs in the Arctic are scarce. Endosulfan has been found to be declining very slowly in air at Alert (Nunavut, Canada), however, temporal trends of endosulfan in biota are inconsistent. Endosulfan was reported to be declining in eastern Greenland ringed seals, but no trends were observed in ringed seals from the Canadian Arctic. Freshwater fish from the Canadian Arctic showed recent increases in endosulfan concentrations, but whether this is a consistent trend reflecting increased deposition or is related to food web or climatic factors is yet to be investigated. Given that endosulfan was added to the Stockholm Convention in 2011, and all uses in the U.S. and Canada expired in 2016, future monitoring will be important for assessing the impacts of these regulations on levels within the Arctic.

Recent food-web studies indicate that CUPs reaching the Arctic via long range transport can enter Arctic terrestrial and marine food chains and bioaccumulate, however, in comparison to legacy pesticides and other POPs, CUP concentrations reported for Arctic biota are generally low. Also in contrast to legacy POPs (e.g. DDT, chlordane, toxaphene and PCBs), which typically undergo trophic magnification, many CUPs have been found at the highest concentrations in lower trophic-level organisms (vegetation and invertebrates) and at the lowest concentrations in animals at the highest trophic levels (i.e, ringed seals, polar bear, caribou, and wolves), suggesting they have undergone significant trophic dilution, a likely result of the ability of certain mammals to biotransform and excrete these compounds. However, food web bioaccumulation studies have only been conducted for a limited number of CUPs (PCNB, chlorothalonil, chlorpyrifos, dacthal, and endosulfan), and some compounds have been reported at elevated levels in top predators (i.e, killer whales), therefore additional studies are warranted.

The arsenal of pesticides used to meet agriculture and public health needs is continuously changing. As new regulations force older pesticides out of the market, new chemicals are taking their place. Changes in climate [84], the increasing appearance of pesticide-resistant insects and weeds [85], and the widespread use of genetically-modified crop varieties [86] will also affect the type and volume of pesticides used in future agricultural practices. Although modelling studies have identified additional CUPs as potential Arctic contaminants, to our knowledge, they have yet to be investigated in the Arctic environment. Thus, continued monitoring of both known and potential Arctic pollutants will be important for ensuring that CUPs and their replacement chemicals have minimal impacts on human and environmental health.

Conflict of interest statement

The authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

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