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Oceanic long-range transport of organic additives present in plastic products: an overview

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Abstract

Most plastics are made of persistent synthetic polymer matrices that contain chemical additives in significant amounts. Millions of tonnes of plastics are produced every year and a significant amount of this plastic enters the marine environment, either as macro- or microplastics. In this article, an overview is given of the presence of marine plastic debris globally and its potential to reach remote locations in combination with an analysis of the oceanic long-range transport potential of organic additives present in plastic debris. The information gathered shows that leaching of hydrophobic substances from plastic is slow in the ocean, whereas more polar substances leach faster but mostly from the surface layers of the particle. Their high content used in plastic of several percent by weight allows also these chemicals to be transported over long distances without being completely depleted along the way. It is therefore likely that various types of additives reach remote locations with plastic debris. As a consequence, birds or other wildlife that ingest plastic debris are exposed to these substances, as leaching is accelerated in warm-blooded organisms and in hydrophobic fluids such as stomach oil, compared to leaching in water. Our estimates show that approximately 8′100–18′900 t of various organic additives are transported with buoyant plastic matrices globally with a significant portion also transported to the Arctic. For many of these chemicals, long-range transport (LRT) by plastic as a carrier is their only means of travelling over long distances without degrading, resulting in plastic debris enabling the LRT of chemicals which otherwise would not reach polar environments with unknown consequences. The transport of organic additives via plastic debris is an additional long-range transport route that should also be considered under the Stockholm Convention.

Keywords: Plastic additives, Long-range transport, Leaching, Floating plastic, Plastic debris, Arctic, LRTP, Chemicals

Introduction

The potential for environmental long-range transport (LRTP) is a chemical-related property that determines a chemical's ability to travel long distances from its emission source. A chemical with high LRTP can reach remote regions, where it may be harmful to local wildlife and humans. To address this issue at the global level, the Stockholm Convention was established in 2001 and incorporates LRTP as one of its criteria for a Persistent Organic Pollutant (POP) [104]. For the POPs that have so far been included in Annex A, B or C of the Stockholm Convention, environmental long-range transport was assumed to occur either via air (in the gas phase or with particles in air) or via water (ocean currents, rivers) and was often demonstrated via measurements in air, water or biota at remote locations. However, LRTP may be difficult to prove for chemicals that are not routinely

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monitored in these matrices (danger of taking "*absence of evidence*" for *"evidence of absence*").

The long-range transport of chemicals via plastic debris is an additional transport route that has not been used to determine a chemical's LRTP. It is of particular importance for chemicals that are used as plastic additives, since they are present in plastic in concentrations of mostly 0.1 to 1 weight % (and of 20% and 50% for flame retardants and plasticisers, respectively) and are not chemically bound to the polymer. Most plastic products contain organic additives added as fillers, plasticisers, antioxidants, coupling agents, colourants, UV and heat stabilisers, polymeric impact modifiers, anti-static agents, flame retardants, blowing agents, lubricants, slip additives, antimicrobials and others [32, 40, 84, 128, 138]. Many of these chemicals, for example some brominated flame retardants or short- and medium-chain chlorinated paraffins, have been identified as PBT (persistent, bioaccumulative and toxic) chemicals and some are listed as POPs under the Stockholm Convention [13, 40]. These additives are of concern due to their persistence, bioaccumulation potential and toxicity together with their migration potential from the polymer matrix [40, 57]. Leaching of the chemicals from the polymer matrix has been shown to occur [132], but may take several years, during which the plastic debris may have been transported far from its emission source. Thus, the LRTP of plastic debris itself may become the basis for the LRTP of additives present in plastic matrices [54].

Plastic fragments have been consistently detected since the 1970s on remote beaches, in sea water, sea ice and glacier ice in remote regions as well as in wildlife living in less populated to remote areas [7–9, 17, 21, 43, 44, 58, 59, 71, 76, 92, 96, 100, 126, 130, 134]. Furthermore, commonly used additives such as phthalates, bisphenol A, and alkylphenols that can act as endocrine-disrupting chemicals (EDCs) but are not persistent have also been detected in high concentrations in plastic debris found in less populated areas and in the open ocean [45]. Thus, plastic debris can even transport chemicals to remote regions that would not have reached these areas by other means. Subsequently, these chemicals can then be taken up by organisms covering the whole food chain at these remote locations, which may lead to adverse effects with unknown consequences. Highly brominated PBDEs, for example, were detected in the abdominal adipose tissue and liver of seabirds (*Puffinus tenuirostris* and *Fulmar glacialis*) in the North Pacific Ocean and the Svalbard Archipelago, with strong indications that these additives originated from the plastic debris present in the birds' stomachs [82, 120]. However, other anthropogenic sources and transport pathways of some of these chemicals besides plastic particles exist, requiring a careful evaluation of fluxes and fate to identify the chemicals that are dominantly transported by plastic debris to remote regions.

The polymer composition of plastic debris observed in remote regions covers mostly four polymers: polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) [49, 118, 125]. Tire-wear particles (TWP) are an additional category and consist mainly of synthetic styrene–butadiene rubber (SBR) [42, 51, 56]. All of the mentioned five plastic/rubber types are known to contain a large number of additives, with TWP containing a different mix and amount of additives than the others.

This paper's aim is to review, reconcile and integrate information in order to evaluate the hypothesis that marine plastic debris is a relevant long-range transport carrier for plastic additives. Specifically, we discuss (i) the presence of marine plastic debris globally and its potential to reach remote locations; (ii) the organic additives present in plastics; and (iii) how these additives can leach into the marine environment. We finally estimate the mass of additives in plastic debris that can undergo oceanic long-range transport and summarise the available evidence for uptake of plastic additives by wildlife.

Plastic production and waste

Around 370 million tonnes (Mt) of plastics were manufactured in 2019 globally [93]. In 1950, this figure was 2 Mt globally, which means that plastic production has increased substantially since then. Half of the total amount was produced in just the past 13 years [37]. Today, almost half of all plastics are produced in Asia, while NAFTA (US, Canada and Mexico) and EU countries account each for about 20% [131]. Until 2017, the cumulative amount of plastics (resins and fibres) ever produced globally was around 8.3 gigatonnes (Gt), 1 Gt of it being fibres [37]. It has been suggested that 6.3 Gt of plastic waste has been generated thus far [37]. From this, around 79% has been accumulated in landfills or in the environment, 9% was recycled, and 12% was incinerated [37]. Although global recycling rates have increased in the last 10 years to over 30% [94], there is still a large share of mismanaged plastic waste that has already been released into the environment, with more to come [10].

The most-produced plastic polymers worldwide are PP (23%), low-density (LDPE) and linear low-density PE (17%), polyvinyl chloride (PVC) (16%), high-density PE (HDPE, 15%), PS and expandable PS (EPS, 6%), PET (7%), polyurethane (PU, 6%), and others (9%) [113]. The largest groups in non-fibre plastics are PE, PP, and PVC [37]. Polyester, most of which is PET, accounts for 70% of all

fibres [37]. Approximately 42% of all non-fibre plastics have been used for packaging and 19% in the building and construction sector [37].

In addition to plastic resins and fibres, around 13.6 Mt of natural rubber and 15.1 Mt of synthetic rubber were produced in 2019 [114, 115]). One of the largest uses for rubber is in tires where TWP are generated. TWP discharges to the environment can be expected to be highest in urban areas where traffic density is greatest. The quantity of TWPs ultimately entering marine environments strongly depends on the vicinity of urban areas to the coast and on the extent of collection and treatment of road runoff, which is highly variable [129].

Plastics in the ocean

Marine plastic waste has increased significantly over the last 60 years with highest numbers recorded in the Northern Hemisphere [89]. Borrelle et al. [10] estimated that at least 18.6 to 26 Mt, or 11% of macroplastic waste generated globally in 2019 entered the oceans (or lakes and major rivers that are connected to the oceans). They used recent spatial information on the generation of mismanaged plastic waste [63] to determine the emission rate of plastic waste as a function of distance from aquatic systems.

These estimates do not account for microplastics such as tire- and road-wear particles, microfibers emitted from washing of textiles, and microplastics added into personal care products as these do not originate from mismanaged plastic. A recent report estimated that 0.28 Mt to 0.87 Mt of microfibers are emitted every year from washing of synthetic textiles to the ocean and 0.23 Mt to 0.71 Mt are emitted as road-wear particles to the ocean, respectively [11]. With also microplastics in personal care products and marine coatings taken into account, it was estimated that 0.8 Mt to 2.5 Mt of microplastics are emitted in total to the ocean [11], additionally to the amount of mismanaged (macro) plastic (18.6–26 Mt, see above). Combining these numbers results in 5.2% to 7.7% of the global plastic production in 2019 that was emitted into the ocean in 2019 (19.4 Mt to 28.5 Mt), see Fig. 1.

Based on these figures and an estimated cumulative amount of 6.3 Gt of plastic waste generated over time, 330 to 485 Mt of plastic may have entered the oceans by now (global historical total). Around 6% of this amount has been reported to have entered the Atlantic Ocean

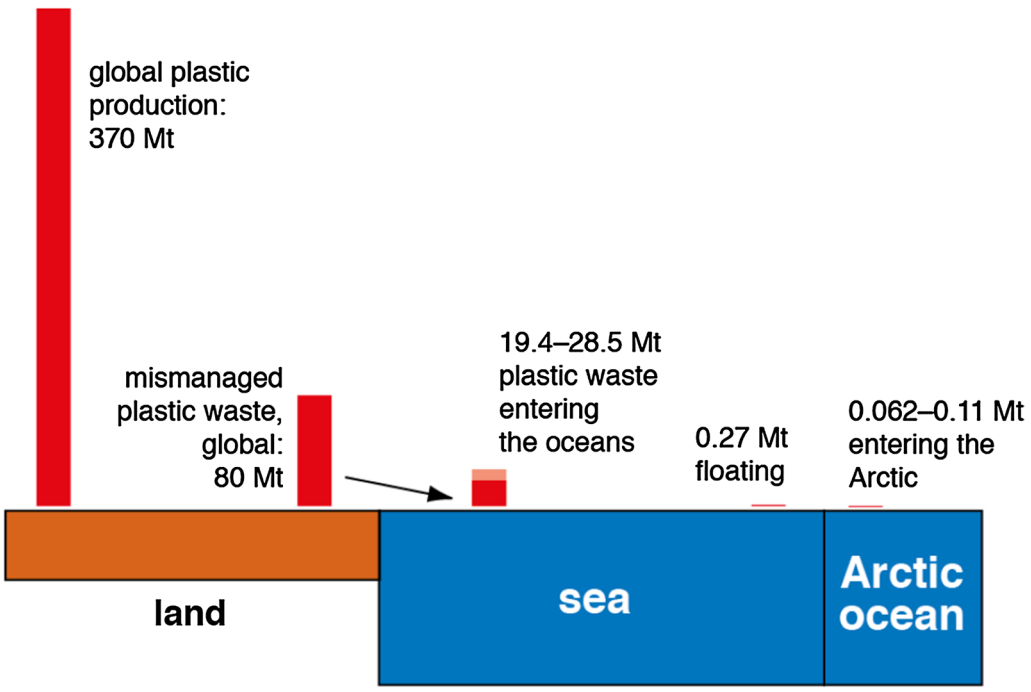


Fig. 1 Plastic waste entering the oceans in 2019. Breakdown of data similar to the scheme by Jambeck et al. (https://jambeck.engr.uga.edu/landplasticinput). Data for 2019 from Borrelle et al. [10]. Global plastic production for 2019 from Plastics Europe [93]. Amount of mismanaged plastic waste from Lebreton and Andrady [63], scenario B. Amount of plastic floating on the surface of the oceans from Eriksen et al. [27] and Van Sebille et al. [108]; amount of plastic entering the Arctic from Zarfl and Matthies [136], see below, section on Mass balance of chemicals additives that can reach remote regions. Mt: million tonnes

[90]. It was calculated that 11.6 to 21.1 Mt of PE, PP and PS are present as 32–651 µm size-class plastics in the top 200 m, below the surface of the Atlantic Ocean. This amount adds to the amount of larger plastic debris estimated to be dispersed in the oceanic surface waters (only approximately 0.1 Mt) and to the amount of plastic particles accumulated on the seafloor (5.6–13.5 Mt) of the Atlantic Ocean [90].

For the purpose of this study, we focus on marine floating plastic particles that are available to LRT, excluding plastic litter below the surface of the ocean that is settling to the seafloor (approximately 95% of the mismanaged plastic litter in the ocean) (see the Additional file 1: Sect. 1 for further information). This is a very conservative approach as also plastic particles below the surface of the ocean might be transported over longer distances. However, it is currently not possible to quantify the LRT of these plastic particles. The amount of floating plastic has been investigated in several studies, but all have calculated quantities of total floating plastic below 0.3 Mt. In 2014, it was estimated that 5 trillion plastic pieces were floating on the oceans' surface, which corresponds to around 0.27 Mt of plastic [27]. In a second study, the global estimate for floating MPs ranged from 15 to 51 trillion particles, weighing between 0.09 and 0.24 Mt [108]. A third study from 2010 reported lower numbers of floating plastics, 0.01–0.04 Mt [20]. Some of these floating pieces might be classified as microplastics (MPs, size < 5 mm), which can have primary or secondary sources and are most commonly PE and PP, followed by PS and PVC. Larger plastic fragments (above 5 mm), are considered meso- or macroplastics [18, 124]. MPs account for 13% of the global floating marine plastic debris mass and for 92% of global plastic pieces [1, 27].

However, the particle size of plastic particles in the environment continuously decreases due to physical stress and weathering, causing the particles to break into smaller fragments. This results in increasing particle numbers while the weight stays largely constant. Biofilm covering the surface of MPs can increase the density of otherwise floating MPs and cause them to sink. This seems to be particularly relevant for MPs smaller than 1 mm and indicates that MPs smaller than 1 mm may not be as easily available for oceanic long-range transport [20, 50, 68, 74]. This has been demonstrated in studies that analysed particle-size distributions of floating plastic in the ocean [20, 50], in a modelling study [68], and in a study that analysed a sediment core in the semi-closed Tokyo Bay. The latter study found that most MP in the sediment were in the 0.3–1 mm size class, with particles of 1–5 mm present in the sediment only in trace amounts [74]. A subsequent study that analysed the size distribution of floating plastic in the Tokyo Bay confirmed that MP larger than 1 mm were actually emitted to the Tokyo Bay [78]. As a caveat it needs to be mentioned that the methodology of sampling of MPs below 0.3 mm in seawater is generally very challenging, resulting in a lack of data for the smaller size classes.

Sunk plastic debris is believed to become trapped in low-circulation and high-sediment accumulation areas after drifting [5, 28] if it is not remobilised again after being ingested (followed by removal of the biofilm) by benthic feeders.

Floating plastic is transported by various processes (for more details see the next section) and accumulates in certain parts of the oceans. Among the oceanic accumulation zones, there are five major plastic gyres, the North and South Atlantic, North and South Pacific, and Indian Ocean gyres (Table 1, Fig. 2). These gyres are large systems of circular currents and can accumulate passive items such as floating marine debris [47]. The total amounts of floating plastics determined by Eriksen et al. [27] for the southern hemisphere oceans (South Atlantic, South Pacific and Indian Ocean) was within the same range as for the northern hemisphere oceans (North Atlantic, North Pacific, and the Mediterranean Sea) (Table 1). The highest plastic particle count and mass was calculated for the North Pacific Ocean Gyre (NPG) [27]. The estimates from Eriksen et al. [27] are thought to be conservative and other studies [137] have come up with even higher amounts of plastic in the NPG than Eriksen et al. [27]. Within the NPG there are three large accumulation zones for marine debris: the North Pacific Subtropical Convergence Zone, the "Eastern Garbage Patch", and the "Western Garbage Patch". The "Eastern Garbage Patch" is also called the "Great Pacific Garbage Patch" and is the largest accumulation zone in the NPG and also globally [47]. The "Eastern Garbage Patch" is increasing exponentially and escaped plastics are retained within or around the NPG [64]. Debris from all other gyres move here, but migration from southern gyres is much slower [106].

Table 1 Estimates of floating plastic debris in the five main oceanic gyres (North and South Atlantic, North and South Pacific, and Indian Oceans) and the Mediterranean Sea (data from [27])

Floating plastic debris	North Atlantic Ocean	South Atlantic Ocean	North Pacific Ocean	South Pacific Ocean	Indian Ocean	Mediterranean Sea	*Total*
Weight [kt]	56	13	96	21	59	23	*269*

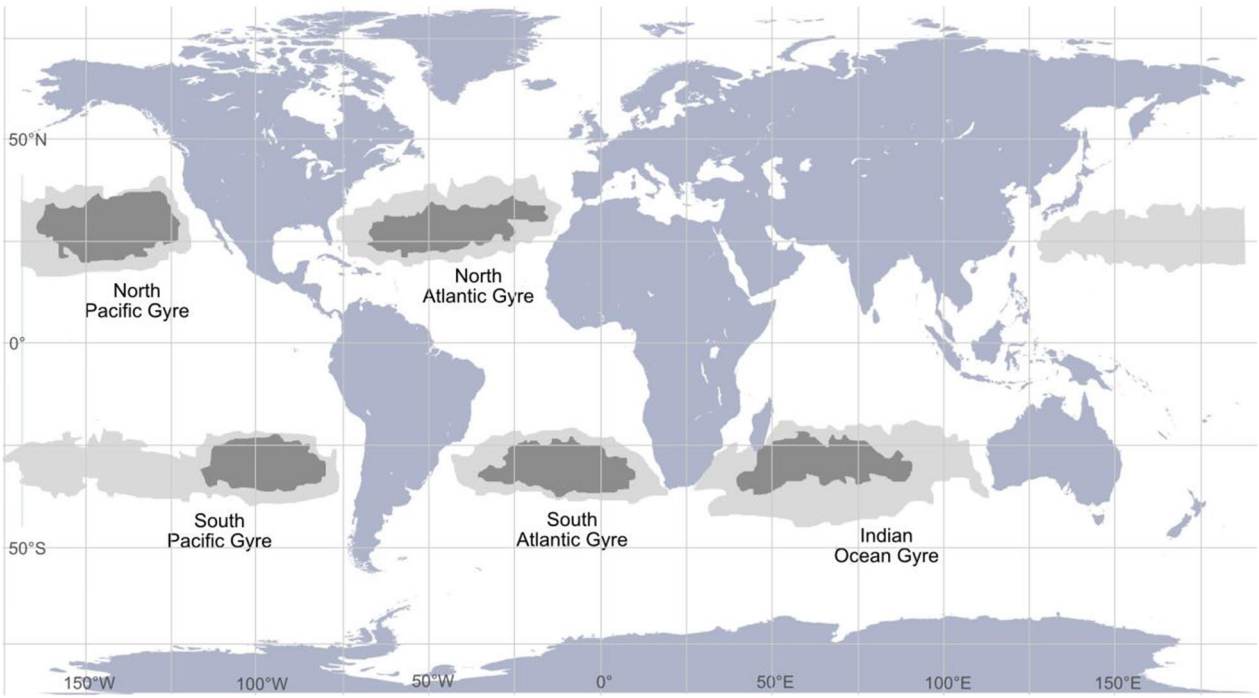


Fig. 2 Location of the five major plastic gyres. Dark and light grey represent inner and outer accumulation zones, respectively; white areas are non-accumulation zones. The locations of the accumulation zones taken from Cózar et al. [20], map created by the authors

Oceanic long-range transport of plastic debris

Plastic waste travels long distances in the oceans [5, 86], and has been detected in all regions of the world, such as in the whole latitude range of the Atlantic Ocean (68°S–78°N), at 1200 km off the Brazilian coast [112], by the Scottish coast [103], in the Eastern Pacific Ocean [62], on Amchitka Island in the Bering Sea [41] or on South Korean beaches, where 56% of the debris collected was ocean-derived [52]. Additionally, plastic resin pellets (3–4 mm) have been shown to occur at remote islands such as Cocos Island, St. Helena, Macquarie Island, Galapagos Island, Shetland Islands and Henderson Island [44, 134].

A significant amount of buoyant plastics will float and accumulate in regions of strongest convergence near the surface, such as in oceanic gyres. In general, PE, PP, EPS, and PU foam are less dense than seawater and, if the plastic fragments are larger than 1 mm, they may travel following wind and oceanic currents as they float [68, 105]. Polyester, PVC, polyamide, and acrylics, on the other hand, are denser than seawater and sink [30, 110]. However, as described before, the plastic's density is influenced by many factors, e.g. polymer, chemical additives, biofouling, and weathering [33, 55, 67]. Thus, for longer time-scales, it is difficult to determine the density of a piece of debris. The most common polymers found in the Northern gyres were PE and PP [25, 75]. The floating capability of synthetic fibres follows the same density relationship as particles from the same polymer, however, sinking velocities vary substantially, causing a broad range of residence times in subsurface waters as well reduced settling [33]. TWPs, on the other hand, are characterised by a high density, theoretically leading to fast settling. However, air enclosed in the rubber from the galvanisation process can cause rubber to stay in the water column and travel longer distances or be remobilised from the seafloor.

For each hemisphere, numerical models and tracers show that gyres are inter-connected and dynamic; Southern Hemisphere gyres show greater inter-connectivity than Northern ones, but there is relatively little exchange between Northern and Southern Hemispheres gyres. The travel time from the point of emission to gyres is years to decades, whereas the residence time of plastic pieces in the gyres themselves maybe even longer, depending on the specific characteristics of the gyre [106]. Plastic in the two Pacific Ocean Gyres is estimated to stay for hundreds of years [106]. Plastic in the North Atlantic Gyre is advected northeastward towards the Barents Sea, while plastics from the Barents Sea is advected southwestward back to the North Atlantic Gyre [106]. The Arctic is highly connected with adjacent seas, e.g. through the Fram Strait, the Bering Strait and the porous Alaska

Archipelago, and the propagation of plastic litter thus extends into the Arctic.

In the Southern Hemisphere, the South Pacific and Indian Ocean and, to a lesser extent, the Atlantic Ocean subtropical gyres are connected forming a Southern Hemispheric 'supergyre' [97]. The flow in the upper waters between the South Pacific and the Indian Ocean is dominated by the Indonesian Through Flow, but the Tasman Leakage (a connection south of Tasmania) constitutes a second pathway for Pacific waters to reach the Indian Ocean [97]. The Indonesian Through Flow is also significant for plastic debris migration, as it involves transport of surface waters in much larger volumes than the Tasman Leakage [107]. Recently, a robust and permanent super-convergent pathway was identified over 8,000 km, connecting the South Indian Ocean with the convergence zone of the South Pacific [72]. This shows that, although the convergence zones in the oceans accumulate plastic, these zones are rather leaky, with plastic being expelled from the gyres within a matter of years, although for most of the gyres this leakage is then re-accumulated by the same gyre [106]. The North Pacific Gyre is ultimately the largest 'attractor' of surface plastic debris of all, and it is suggested that a significant fraction of the marine debris reaching the open ocean outside of the North Atlantic will eventually end up in that gyre (Fig. 2).

Plastics in remote regions

Carried by large-scale oceanic currents, significant fractions of the plastics entering the oceans leave or miss the gyres, reach remote regions and accumulate, e.g. on shorelines. For example, Henderson Island in the South Pacific has the highest plastic-debris density on the beaches in the world [60]. This island has no significant local input, but is located near the super-convergent pathway [72]. Another remote region, the Cocos (Keelings) Islands in the Indian Ocean, also shows high concentrations of plastic debris on its beaches and no relevant nearby source [61]. At the isolated atoll of Saint Brandon's Rock in the Indian Ocean, plastic waste from Southeast Asia, the Indian sub-continent, and the Arabian Peninsula was detected on the shores [12]. It has also been shown that windward beaches of the Main Hawaiian Islands showed 1–2 orders of magnitude more plastic pollution than leeward beaches, despite smaller human populations on windward sides [14]. This shows that the majority of marine debris in Hawaii floats in from distant sources rather than originating from Hawaii's residents or tourists.

Recent studies have shown that also the Arctic may serve as an important accumulation zone for plastic debris and MPs, which can be transported there via the Thermohaline Circulation [8, 21]. MPs have been detected ubiquitously in the Arctic: in surface waters [6, 21, 38, 71], the water column [71, 100], benthic sediment [8], sea ice [87, 92], and in biota [58, 96]. The highest recorded value of MPs in benthic sediments were reported in the Arctic, in the range of 42–6595 MPs/kg, taken from 2340 to 5570 m depths in the Fram Strait [8]. The highest reported concentration of MPs in Arctic surface waters was 61.2 particles/m^3, found at Ny-Alesund, Svalbard, Norway [38]. In a recent study conducted in the Fram Strait, North Sea and Baltic Sea, average concentrations of MPs found in surface waters were 0.058 particles/m^3 or 0.009 particles/m^2 floating on the surface, with higher concentrations of MPs found closer to the Arctic [48]. 80% of the MPs were fragments and 16% were films, with a mean particle size of 2.66 mm. PE was the most abundant polymer material (50%), followed by PP (24%). In the Beaufort Sea, MPs were found throughout the water column, with an average concentration of 174 ± 21.2 particles/m^3 [100]. The MPs were mostly polyester fibres originating from Atlantic inputs. In addition, plastics and MP contamination has been documented in Arctic biota. Northern fulmars have been found to ingest plastics in the Canadian Arctic, with an incidence of 84%, and an average of 0.094 g of plastic mass per fulmar [96]. Non-fibrous MPs have been detected in the stomach of polar cods [58].

Plastic debris and MPs have also been detected in Antarctica, though in smaller amounts and not as ubiquitously as in the Arctic [126]. While plastic pollution of Antarctic waters primarily occurs due to local sources, it has been suggested that Antarctica may also receive plastic inputs from lower latitudes via the Polar Front [130]. Macro- and microplastics have been measured in Antarctic sea surface waters at concentrations of 1794 items/km^2 or 27.8 g/km^2 [59]. In another study, MPs measured in Antarctic surface waters had a mean concentration of 0.17 ± 0.34 particles/m^3. 72% of the MPs were fragments and 13% were fibres. PE and PP were the most abundant polymers in these MPs. Microplastic contamination of Antarctic biota has also been documented [9]. Analysis of Gentoo penguins' scat revealed an average of 0.23 MPs/scat, with a detection frequency of 20%. Data on microplastics in Antarctic ice cores are currently unavailable [126].

Chemical additives in marine plastic debris

Plastic waste is a concoction of chemicals originating from the actual plastic matrix, organic additives and their degradation products, as well as hydrophobic chemicals adsorbed to the plastic surface from the environment. Table 2 provides an overview of the most commonly used additives in plastic materials;

Table 2 Types of additives in plastic materials

Category/type of additive	Substances	Polymers in which applied	Typical range % w/w
Plasticisers	Chlorinated paraffins, diisoheptylphthalate (DIHP), diisodecylphthalate (DIDP), 1,2-benzenedicarboxylic acid, di-C_{7-11}-branched and linear alkyl esters (DHNUP), benzyl butyl phthalate (BBP), bis(2-ethylhexyl)phthalate (DEHP), bis(2-methoxyethyl) phthalate (DMEP), dibutyl phthalate (DBP), dipentyl phthalate (DPP), di-(2-ethylhexyl) adipate (DEHA), di-octyladipate (DOA), diethyl phthalate (DEP), diisobutylphthalate (DiBP), tris(2 chloroethyl)phosphate (TCEP), dicyclohexyl phthalate (DCHP), diheptyl adipate (DHA), heptyl adipate (HAD), and heptyl octyl adipate (HOA)	PVC, cellulose plastic	10–70
Flame retardants	Chlorinated paraffins, brominated flame retardants with antimony (Sb) as synergist (e.g. polybrominated diphenyl ethers (PBDEs), decabromodiphenylethane, tetrabromo-bisphenol A (TBBPA), tetrabromophthalic anhydride (TBPA), phosphorous flame retardants (TCEP or tris(2-chlorisopropyl)phosphate (TCPP)), hexabromocyclododecane (HBCDD)	Various	2–60
Blowing agents	Azodicarbonamide (ADC), p-toluenesulfonylhydrazide (TSH), 4,4′-oxibis (benzenesulfonylhydrazide) (OBSH), p-toluenesulfonyl semicarbazide (TSSC), 5-phenyltetrazole (5-PT), N,N′-dinitroso-pentamethylenetetramine (DNPT)	Various	0.05–20
Colourants	1,4-Diamino-2-methoxy-9,10-anthracenedione, 2-methyl-4-((2-methylphenyl)azo)-benzenamine	Various	0.25–5
Antifogging additives	Glycerol esters, polyglycerolester, sorbitan esters and their ethoxylates, nonylphenol ethoxylates, alcohol ethoxylates	PE, PP, EVA copolymers, PVC	1–3
Heat stabilisers	Nonylphenol (barium and calcium salts)	PVC	0.5–3
Anti-static additives	Fatty acid esters, ethoxylated alkylamines, diethanolamides, ethoxylated alcohol, alkylsulfonates, alkylphosphates, tetraalkylammonium salt, trialkylbenzylammonium salt	PE, PP, PVC, PS/HIPS, ABS/SAN	0.1–3
Lubricants	Fatty acid esters, hydrocarbon waxes, metal stearates, amide waxes, ester waxes	PVC, PS/ABS, PP, PE, engineered thermoplastics	0.1–3
Curing agents	4,4′-Diaminodiphenylmethane (MDA); 2,2′-dichloro-4,4′-methylenedianiline (MOCA), formaldehyde – reaction products with aniline; hydrazine, TGIC/β-TGIC		0.1–2
Primary Antioxidants	Irganox E 201, butylated hydroxytoluene (BHT), Irganox 1076, Irganox 1135, Irganox 2246, Irganox 259, Irganox 1098, Irganox 1222, Irganox 1425, Irganox 245	PP, PE, styrenics, engineered resins, PVC	0.01–1
Antimicrobials	2-*n*-octyl-4-isothiazolin-3-one in dioctylphthalate, Ag-Zn-zeolites, 2,3,5,6 tetrachloro-4 (methylsulfonyl) pyridine, N-(trichloro-methylthio) phthalamide, 10,10′-oxybisphenoxarsine (OBPA), triclosan	Plasticised PVC, PU, LDPE, polyester	0.001–1
Secondary Antioxidants	Irgafos TNPP, Irgafos 168, Irgafos 126, ADK Stab PEP 36, ADK Stab HP-10, Hostanox P-EPQ, Ethanox 398, Weston 618, Irgafos 12, Irgafos 38, Ultranox 641, Irganox PS 802, Irganox PS 800	PP, PE, styrenics, engineered resins, PVC	0.02–0.5
Nickel Quenchers	Cyasorb UV-1084, Irgastab 2002, UV Chek AM 101, Sanduvor NPU	PP	0.05–0.4
UV Absorbers	UV-P, UV-320, UV-326, UV-327, UV-328, UV-329, UV-350, UV-360, UV-571, Tinuvin 213, Tinuvin 234, Tinuvin 840, ADK Stab LA 51, Seesorb 1000, Cyasorb UV-9, Uvinul 400, Cyasorb UV-24, Cyasorb UV-531	Various	0.1–1
Hindered Amine Stabilisers	Tinuvin 770, Tinuvin 622 LD, Chimassorb 944, Chimassorb 119, Tinuvin 765, Tinuvin 144, Tinuvin 123, Chimassorb 2020		0.1–1.5
Slip additives	Erucamide, oleamide, stearamide	LDPE, PP	0.05–0.15
Polymer processing aids	Fluoropolymers (e.g. copolymer of vinylidene fluoride and hexafluoropropylene), polydimethyl siloxane oils	LLDPE	< 0.1
Polymeric impact modifiers	Methacrylate butadiene styrene compounds, chlorinated PE, acrylic polymers, ethylene vinyl acetate	PVC, PE, PP	

Information on the substances is from Zweifel et al. [138], Hahladakis et al. [40], OECD [84], OECD [85], Gauquie et al. [34], and van Oers et al. [88]. Information on the polymers is from Zweifel et al. [138] and van Oers et al. [88]. Information on the typical amount range is from Zweifel et al. [138], Hahladakis et al. [40], and Drobny [23]. Additives are listed according to their percentages used. The list of polymer types is not exhaustive and the additives listed may also be used in other polymers

Additional file 1: Table S2 provides CAS registry numbers and chemical structures of more than 100 additives. The EU report on the relative release potential of plastic additives lists 418 different additives that are used in the EU above 100 t/year [24]. Most of them are pigments, followed by plasticisers, flame retardants, and heat stabilisers.

The amounts in which additives are present in the final mixture vary widely among different polymer types and functions. Geyer et al. [37] stated that non-fibre plastics contain 7% additives by mass. Other sources consider a fraction of 15% chemical additives across plastic types [132]. For example, phthalates might represent up to 50% of the total weight of

PVC, which matches the fact that PVC uses the highest bulk of the world's production of additives by volume (73%), followed by PE and PP (10%), and styrenics (5%) [77, 98]. However, PVC has a density higher than water and will sink to deeper water layers. De Frond et al. [31] found in total 0.05% to 2.5% per mass of 20 different additives in LDPE, HDPE, PP, and PS plastics that were collected from beaches as litter. However, as most of the collected items were food-grade items, they might have had fewer additives than other types of plastics. Further, the list of chemicals was not inclusive of all additives used in these items. Suhrhoff and Scholz-Böttcher [116] measured additives in PVC, PET, PS and LDPE. Non-expanded PS contained 1.05% of additives and LDPE contained 0.23%. The general lack of data on additive identity and amounts used in plastic items hampers estimations of mass flows of additives. We here assume a range of organic additive concentrations across all floating plastic types of 3% to 7%, noting that there are differences across polymers types and uses and that this percentage does not cover brominated flame retardants, which are used at around 20% (e.g. BDE-209 in PE [138]), nor plasticisers, which are used in PVC at 33% on average [138].

Chemical additives leaching from plastic debris into the marine ecosystem

The aqueous leaching potential of additives depends on many factors, such as the plastic porosity, the additive's molecular size, concentration, and physical–chemical properties, the extent of weathering, the surface area-to-volume ratio of plastic particles (shape and size), pH, water temperature, and duration of exposure to water [70, 123, 133].

Higher temperature or surface-to-area ratio increase leaching. It has also been found that turbulence in the water and physical stress increases the leaching of additives [116]. Another factor influencing additive leaching is the properties of the polymer matrix. Polymers generally contain crystalline and amorphous parts and the fractions of crystalline vs. amorphous vary among polymers. Additives can only occupy amorphous regions of the polymer. A factor that affects the diffusivity of additives in a polymer is the polymer's glass transition temperature, T_g. Below T_g, the amorphous parts are in a glassy, more rigid state and above T_g, they are in a rubbery, more flexible state [111]. Diffusion of additives in the amorphous parts of a polymer is faster when the polymer is above its T_g [36, 117]. The T_g values of the most important plastic polymers follow the order $T_{g,PE} < T_{g,PP} < T_{g,PET} < T_{g,PVC} < T_{g,PS}$; approximate values of T_g are 150 K (PE), 260 K (PP), 345 K (PET), 360 K (PVC), 373 K (PS) [19, 117]. Accordingly, some polymers are below their T_g (i.e. glassy) at ambient temperatures (PVC, PS) and others (PE, PP) are above their T_g (i.e. rubbery) at ambient temperatures.

Furthermore, the leaching of additives depends on the physical–chemical properties of the additives themselves [26, 65, 79]. It has been shown for PE and PP that, as the hydrophobicity or the partition coefficient between plastic and water of the additives increases, the loss of the additives from plastic particles decreases [26, 65, 79]. Diffusion of substances with higher hydrophobicity (in PE and PP) is more likely controlled by the aqueous boundary layer diffusion (diffusion between particle and water) and not by the internal diffusion in the plastic particle [57, 65, 69]. For planar passive PE samplers, diffusion across the plastic–water boundary layer is usually the rate-limiting step for substances with a logarithmic octanol–water partition coefficient (log K_{OW}) of 4 or above. However, this might be different for other polymer types. Results for acrylonitrile butadiene styrene (ABS), a polymer that has a similar glass transition temperature as PS, indicate that diffusion for PBDEs with log K_{OW} values above 4 is also dominated by internal diffusion in the plastic particles [117]. This is in line with other studies that showed that the diffusivity of substances in polymers decreases as the glass transition temperature of the polymers increases [15].

Highest leaching rates are therefore expected for small PE fragments. According to the equations from Endo et al. [26], the desorption half-life from PE for substances with a log K_{OW} between 5.5 and 7 will be between 1 and 38 years, assuming medium-turbulent water (aqueous boundary layer 100 μm). Phthalates have been shown to leach from plastic materials; with 80–120 ng g^{-1} plastic over a period of 90 days, only a small share of their overall content (1–5 weight percent) is released in the first 3 months of exposure [91]. This shows that leaching is a relevant process but also that considerable fractions of the additives remain available for long-range transport with the plastic matrix.

Mass balance of chemicals additives that can reach remote regions

If we consider that at least 0.01–0.27 Mt of plastics are floating on the oceans and that 3% to 7% of the upper limit of 0.27 Mt corresponds to organic additives, approximately 8′100–18′900 t of organic additives are transported within buoyant plastic matrices globally [2, 86]. For comparison, De Frond et al. [31] estimated a much lower amount (190 t), only considering the litter of 7 distinct plastics items accounting for only 1% of the plastic that was estimated to have entered the oceans in 2019 [10]. In addition to the amount of additives in

floating plastics, 570′000 to 1′820′000 t of additives may be present as a large additional reservoir in plastics below the ocean's surface. A fraction of these additives has probably already been degraded, but low temperatures and lack of light may have also slowed down the degradation processes. Benthic organisms will be exposed to these deposits and may partly remobilise plastic particles and chemicals alike.

Zarfl and Matthies [136] estimated, based on the maximum volume of water transported to the Arctic, a flow of 0.062 Mt to 0.105 Mt plastic per year to the Arctic Ocean. They assume with this estimation that not only floating plastic, but also plastic debris in deeper water layers travels long distances. This is a less conservative approach than we have taken here, but not necessarily incorrect, given the work of Pabortsava and Lampitt [90], who reported high concentrations of plastic in deeper water layers (10–270 m). Based on the estimate of Zarfl and Matthies [136] and a percentage of 3% to 7% plastic additives, between 1′900 and 7′400 t organic additives may be transported to the Arctic Ocean with plastics every year. This number is much larger than the values that were calculated by Zarfl and Matthies [136] for chemicals adsorbed to the plastic such as PCBs. However, additives are present in concentrations higher by several orders of magnitude than concentrations of adsorbed contaminants such as PCBs. For example, Yeo et al. [135] reported two orders of magnitude higher concentrations of BDE-209 compared to PCBs in buoyant microplastics collected from surface waters. Therefore, the conclusion by Zarfl and Matthies [136] that PCBs from plastic-mediated transport are minor compared to PCBs from atmospheric transport is not valid for additives. In the same way, also the conclusion by Koelmans et al. [54] that plastics are unimportant as a transfer pathway for hydrophobic organic chemicals does not apply to additives.

The amount of organic additives that may be transported to the Arctic Ocean with plastics every year is also much larger than the amount of perfluorooctanoate that may reach the Arctic every year via ocean currents (8–23 t) [3]. For airborne transport, Zarfl and Matthies [136] estimated that 300 t of PCBs and 720 t of PBDEs might be transported every year to the Arctic. This flow is still considerably smaller than the 1′900 t to 7′400 t of plastic additives that may be transported every year into the Arctic via oceanic transport of plastic.

Environmental impacts of chemical additives released from oceanic plastic debris

More than 80% of the adverse impacts by debris in the marine environment are associated with plastic waste, as it can impact this environment physically and biochemically [46, 109]. Plastic waste has also been found in biota from different trophic levels, such as marine invertebrates [16, 22, 66], fish [73, 83, 101], marine mammals [29, 80, 81], seabirds [4, 82, 95, 96, 119], green turtles [127], and penguins [9]. See also Ryan [102] for a comprehensive overview.

Leaching processes are accelerated in warm-blooded organisms compared to leaching in water, due to elevated temperatures in organisms (body temperature of seabirds $\approx$ 40 °C), presence of hydrophobic fluids such as stomach oil and fish oil, and physical abrasion/degradation of plastic particles into smaller sizes within organisms upon plastic ingestion. In a study by Tanaka et al. [121], pieces of plastic compounded with deca-BDE were soaked in several leaching solutions. Trace amounts of deca-BDE leached into distilled water, seawater, and acidic pepsin solution. In contrast, over 20 times as much of deca-BDE leached into stomach oil, and over 50 times as much into fish oil (a major component of stomach oil) [121]. Moreover, Sun et al. [117] showed that the rate of leaching of BDE-209 increased with decreasing size of plastic particles, due to larger specific surface area and/or shorter distance for internal diffusion.

Direct evidence of plastic additives transfer into biota at higher trophic levels comes from feeding experiments with seabirds [122]. Tanaka et al. [122] prepared PE pellets compounded with five plastic additives (UV-327, UV-328, UV-326, BDE-209, and BP-12). The concentration of each additive in the pellets was 0.4% by weight. The pellets were fed to eleven 37-day-old streaked shearwater (*Calonectris leucomelas*) chicks in a natural colony on a cliff on Awashima Island, Japan, in 2017. Ten chicks were chosen for control. In the plastic-exposed chicks, all of the five additives were detected in the liver, abdominal adipose, and preen-gland oil, except BP-12 in preen-gland oil, on day 16 in significantly higher concentrations than in the control group [122].

There is also indirect evidence for the transfer of plastic additives into birds [120]. Tanaka et al. [120] analysed PBDEs in abdominal adipose tissue of short-tailed shearwaters (*Puffinus tenuirostris*) collected in the northern North Pacific Ocean. In 9 of the 12 birds, lower-brominated congeners (i.e. tetra- to hexa-brominated congeners such as BDE-47, BDE-99, and BDE-154) were dominant, similar to findings for pelagic fishes, the prey of shearwaters. Only one of the nine birds had measureable PBDE amounts in plastics in its stomach. However, in the other three birds, higher-brominated congeners (viz., BDE-209 and BDE-183), which are not present in the natural prey of the birds, were also detected. Instead, BDE-209 and BDE-183 were found in plastic in the stomachs of the three birds. Similar findings have been reported for northern fulmars (*Fulmarus glacialis*) in Norway [82].

Evidence for transfer of chemicals from plastic has also been shown for other species. For example, HBCDDs were detected in mussels growing on EPS marine debris [53]. Higher in the food chain, BPA, nonylphenol, and PBDEs were detected in fish from the Antarctic Ocean and there was a significant correlation between concentrations of BDEs-183 to -209 in myctophid fish and the concentration of plastic debris in pieces/km^2 [99]. Likewise, house crickets accumulated PBDEs after feeding on PU foam for 28 days [35].

Hundreds of studies have looked into the hazards of plastic additives and there are concerns over many of these additives. Examples are short-chain chlorinated paraffins, HBCDD, or commercial deca-BDE, all of which are now regulated under the Stockholm Convention. It is beyond the scope of this work to list the hazards of all additives, but we provide here some figures as an overview. A study that looked into chemicals associated with plastic packaging listed 906 chemicals likely associated with plastic packaging and another 3′377 substances that are possibly associated with plastic packaging as well [39]. Of the 906 additives, 63 had the highest possible score for human health hazard and 68 for environmental hazard based on information from the harmonised hazard classifications by the European Chemicals Agency (ECHA) under the EU's Classification, Labeling and Packaging (CLP) regulation. Further, of the 906 additives, seven are PBT/vPvB substances and 15 are EDCs. Moreover, according to UNEP 34 of the 906 chemicals are EDCs or potential EDCs [39]. However, there are most probably more hazardous substances associated with plastic packaging. Groh et al. [39] stated that there were significant data gaps in the hazard information for the 906 additives. For example, over 200 substances associated with plastic packaging without a harmonised CLP classification had advisory CLP classifications assigned by the Danish EPA based on quantitative structure–activity relationships. Thus, these chemicals might be hazardous as well, but are not yet officially classified as such, possibly due to the fact that confirming the predicted hazardous properties requires experimental toxicity testing data, which are often lacking [39].

Conclusion

The available information on plastic waste and its additives demonstrates that plastic pollution is ubiquitous and is often associated with chemicals of concern. Marine plastic waste does undergo long-range transport and reaches remote regions. The available data are not fully consistent (for example, the amount of floating plastics was taken from different studies dating back to different years). However, the overall picture is consistent and the order of magnitude of the data does match. Data gaps exist for the amount of additives present in the different polymers, which hampered a more accurate estimation of the mass flows of hazardous chemicals in mismanaged plastics. Chemical additives may partly be released from the plastic matrix during the transport in the ocean, but the available information shows that leaching of hydrophobic substances is slow in the ocean. It is therefore highly likely that substantial amounts of organic additives reach remote regions together with plastic debris. Birds or other wildlife that ingest plastic debris are then exposed to these substances, as leaching is accelerated in warm-blooded organisms and in hydrophobic fluids such as stomach oil, compared to leaching in ocean water. The oceanic transport of chemical additives via plastic debris is therefore an additional long-range transport route that can lead to adverse human health and environmental effects. A recommendation derived from these findings is that long-range transport via plastic debris be considered as an additional pathway for the evaluation of a chemical's potential for long-range transport under the Stockholm Convention.

Supplementary Information

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Additional file 1. Additional information.

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Authors' contributions
MS, HA and JG devised the study. HA, JG, DH and NA collected data on plastic transport, additives leaching, wildlife exposure and drafted text. SMN collected data on plastic transport and drafted text. MS drafted and revised text. All authors read and approved the final manuscript.

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Availability of data and materials
All data generated or analysed during this study are included in this published article.

Declarations

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