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Are marine plastic particles transport vectors for organic pollutants to the Arctic?

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ABSTRACT

Plastic litter accounts for 50–80% of waste items stranded on beaches, floating on the ocean surface and lodged in the seabed. Organic pollutants can be absorbed onto plastic particles from sea water, attached to their surfaces or included in the plastic matrix as additives. Such chemicals may be transported to remote regions by buoyant plastics and ocean currents. We have estimated mass fluxes of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and perfluorooctanoic acid (PFOA) to the Arctic via the main ocean currents and compared them to those in the dissolved state and in air. Substance fluxes with atmospheric or sea water currents account for several tons per year, whereas those mediated by plastics are four to six orders of magnitude smaller. However, the significance of various pollutant transport routes does not depend only on absolute mass fluxes but also on bioaccumulation in marine food chains.

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1. Introduction

Since the initial widespread development of plastics, their worldwide production has expanded from 1.5 million tons in 1950 to 245 million tons in 2008 (PlasticsEurope, 2009). The five main high production volume plastics, which account for approximately 90% of the total demand, are polyethylene (including lowdensity LDPE, and high-density, HDPE, polyethylene), polypropylene (PP), polyvinylchloride (PVC), polystyrene (PS) and polyethylene terephthalate (PET) (Andrady and Neal, 2009). The useful properties of plastics that have led to numerous technological advances, energy savings, improved consumer health, and reduced transportation costs also mean that plastics have become one of the most drastic and observable impact factors on the environment. It is estimated that plastic litter accounts for 10% of municipal waste mass worldwide and comprises 50-80% of the waste items stranded on beaches, floating on the ocean surface and lodged in the seabed (Barnes et al., 2009), even though a complete ban on dumping of all forms of plastics into the sea was enacted in 1989 with the MARPOL regulation Annex V.1 In addition, so-called micro-plastics have attracted the attention of environmental researchers. These particles, which are smaller than 5 mm (Betts, 2008), account for over 80% of stranded plastics in the Tamar Estuary of the UK (Browne et al., 2007), whereas in the Los Angeles watershed they contribute 90% by count and 13% by weight to the total plastic debris (Moore, 2008).

In addition to the mechanical impacts of plastics on marine organisms, such as suffocation or starvation due to entanglement or ingestion of plastic pieces or bags, a relationship between chemical burden and plastic ingestion has been reported. In a population of Great Shearwaters, a large migrating seabird, the mass of polychlorinated biphenyls (PCBs) was positively correlated with the mass of ingested plastic (Ryan et al., 1988). Other studies have indicated that micro-plastics absorb PCBs from the surrounding sea water (Carpenter et al., 1972; Mato et al., 2001). Respective sorption coefficients on PP pellets turned out to be two orders of magnitude higher than for suspended particles (Mato et al., 2001). Besides sorption from surrounding sea water, several additives are contained in the plastic matrix. Colorants, UV-stabilizers and matting agents, brominated flame retardants, phthalate plasticizers, bisphenol A and anti-microbial agents are additives of particular concern (Thompson et al., 2009). Bound to the plastic matrix, these chemical compounds escape rapid degradation and may thus become "persistent" and subject to long-range transport. When ingested by organisms, plastic particles could be a "biomagnification" route of organic chemicals sorbed to or contained in the plastics, as shown by investigations of polybrominated diphenyl ethers (PBDE), polycyclic aromatic hydrocarbons (PAH), triclosan and nonylphenol (NP) (Teuten et al., 2009).

Persistence, long-range transport potential (LRTP) and bioaccumulation potential are fundamental criteria for assessing organic chemicals according to the Stockholm convention.² Up to now,

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¹ International Maritime Organization, 1982. International Convention for the Prevention of Pollution from Ships, 1973, as modified by the Protocol of 1978 relating thereto (MARPOL). http://www.imo.org/.

² Text of the "Stockholm Convention on Persistent Organic Pollutants" (online): www.pops.int.

evaluation of a compound's persistence has been based on environmental half-lives, bioaccumulation potential has been derived from a bioconcentration factor (BCF), and LRTP has been based on monitoring data in remote regions or an atmospheric half-life greater than two days. If plastics are important pollutant vectors to remote regions, assessment of organic pollutants might have to be supplemented by an additional aspect. However, the significance of plastic particles as transport vectors of organic pollutants to marine organisms and to remote regions such as the Arctic (long-range transport) remains uncertain and has not been quantified. Thus, the objectives of our study were to combine the actual knowledge of the quantities of plastics in oceans, pollutant sorption into plastic particles and environmental substance concentrations in order to roughly quantify the pollutant mass transported to the Arctic Ocean by plastics. Further, we will discuss the subsequent availability of plastic-associated pollutants for uptake by organisms relative to other abiotic modes of transport (via air and ocean currents). Substances were chosen as examples to emphasize that the relevance of specific abiotic transport routes depends on chemical properties and partitioning behavior. Although the uptake of contaminated plastic particles into marine organisms, transfer into food chains and subsequent transport by migrating animals may be another important (biotic) transport route, it is not considered here and will be the subject of another study.

2. Methods

2.1. Estimation of plastics and pollutant fluxes into the Arctic

The transport of pollutants to the Arctic by plastic particles depends on (i) the quantity of plastic debris in the oceans, (ii) the specific surface per mass of plastic particles, (iii) the sorption of pollutants into plastics, and (iv) the velocity of ocean currents.

First, the flux of plastics (G_P in mass per time) was estimated using reports on pieces of plastic (n_P) per km² of ocean surface and knowledge of water volumes transported to the Arctic via the main ocean currents. The sampling depth (d_s) with manta trawls, a net construction for sampling the surface of the ocean, is approximately 1 m (Day and Shaw, 1987; Ryan et al., 2009). This depth allows a rough estimate of plastic concentrations in the upper surface layer of the ocean. Because pollutant concentrations on plastic particles refer to the plastic weight, mass fluxes of plastics are required. Assuming thus an average mass of plastic per piece (w_P), the gross flux G_P of plastics (in mass of plastic per time) into the Arctic can be calculated based on an estimation of sea water volume flowing annually into the Arctic Ocean (G_W in water volume per time):

$$G_P = \frac{n_P}{d_s} \cdot w_P \cdot G_W \tag{1}$$

Subsequently, the flux of pollutants attached to plastics ($G_{\mathrm{poll},P}$ in mass of pollutant per time) is estimated. Two approaches can then be compared. On the one hand, pollutant flux $G_{\mathrm{poll},P}$ is based on pollutant concentrations (C_{plastic}) measured on plastic particles, which results in:

$$G_{\text{poll},P} = G_P \cdot C_{\text{plastic}} \tag{2}$$

On the other hand, the same flux $G_{\text{poll},P}$ can be estimated using measurements of the pollutant concentration in the ocean (C_{ocean}) and respective partition coefficients (K_{e} in L kg⁻¹) between sea water and plastics (if available):

$$G_{\text{poll},P} = K_e \cdot C_{\text{ocean}} \cdot G_P \tag{3}$$

The comparison of pollutant fluxes calculated using Eqs. (2) and (3) can be used to check the consistency of the underlying data. Because reported values are sparse or vary over several orders of magnitude, each single data set is taken to estimate flux rather than applying statistical averaging.

Pollutant transport with plastics $(G_{\text{poll},P})$ is compared with fluxes by atmospheric $(G_{\text{poll},A})$ in mass of pollutant per time) and ocean $(G_{\text{poll},W})$ in mass of pollutant per time) currents. For this purpose, average transport rates for air (G_A) in air volume per time) and an estimated flux of sea water (G_W) in water volume per time) via the main ocean currents to the Arctic Ocean are combined with typical pollutant concentrations in remote air (C_A) and sub-polar waters (C_{ocean}) , respectively:

$$G_{\text{poll},A} = G_A \cdot C_A \tag{4}$$

$$G_{\text{poll},W} = G_W \cdot C_{\text{ocean}} \tag{5}$$

Detailed information on the calculations of the single mass fluxes is given in Appendix A in the Supplementary material of this article.

3. Results and discussion

3.1. Selected substances

Substances were selected on the basis of three considerations: (i) availability of concentration data on marine plastic debris. (ii) available knowledge of sorption behavior of the pollutant into plastics from the surrounding ocean, and (iii) contrasting environmental partitioning. Reports on concentrations of various pollutants in plastics are summarized in Table 1. The most numerous accounts and respective sorption data can be found for PCBs. These were detected in PS spherules in southern New England as early as at the beginning of the 1970s (Carpenter et al., 1972), in resin pellets on Japanese coasts (Mato et al., 2001; Endo et al., 2005; Teuten et al., 2009), in plastic samples on beaches of Hawaii and California (Rios et al., 2007), in the North Pacific gyre, and on several other coasts all over the world (Ogata et al., 2009; Teuten et al., 2009, therein Fig. 7). An important category of plastic additives includes brominated flame retardants, such as PBDEs, which mainly partition into air and are virtually water insoluble. Surfactants such as perfluorooctanoic acid (PFOA) are acids that partition into water and that are designed to adsorb to textiles.

3.2. Annual water volume flowing into the Arctic

The Arctic Circle separates the Arctic from the Northern Temperate zone at approximately 66°34' northern latitude. In order to study the transport of marine plastics across the Arctic Circle, knowledge of the main ocean currents into the Arctic is essential. Three main transport routes into the North can be differentiated. (1) One cold ocean current follows the Bering Strait, which connects the Pacific Ocean (Bering Sea) to the Arctic Ocean (Chukchi Sea) and is located between Alaska (USA) and Russia. (2) The Fram Strait is a route between the Atlantic Ocean and the Arctic (Norwegian Sea), which stretches between Greenland and Spitsbergen. Here, the West Spitsbergen Current carries warm and saline ocean water from the North Atlantic Current to the inner Arctic. (3) The North Atlantic Current splits into a second current, which finds its way into the Arctic between Spitsbergen and the Siberian coastline. Simonsen and Haugan (1996) collected published data on the inflow of water volumes into the Arctic via these transport routes. The dominant ocean current is the West Spitsbergen Current, which carries 2–7.2 Sverdrup (Sv = 10^6 m³ s⁻¹) into the Arctic, followed by the Bering Strait and the ocean current along the Siberian coastline, which carry 1.2-1.5 Sv and 0.7-1.0 Sv, respectively. Recently, Fahrbach et al. (2001) published measurements of northward flowing volume transport through the Fram Strait, which ranges from 5 Sv in August to 20 Sv in February. They estimated an annual average of 9.5 ± 1.4 Sv. The maximal annual averages of the water volumes transported by the different ocean currents (9.5 Sv by the West Spitsbergen Current, 1.5 Sv via the Bering

Table 1Selected references reporting pollutant concentrations (ng g⁻¹) on plastic particles.

Reference	Sampling location	PCB	DDE	PAH	PBDE	NP ^a
Carpenter et al. (1972)	Southern New England	5000	n.a.	n.a.	n.a.	n.a.
Mato et al. (2001)	Japanese coast	3.97-117	0.16-3.1	n.a.	n.a.	130-16000
Endo et al. (2005)	Tokyo	<28 ^b -1260 ^c	n.a.	n.a.	n.a.	n.a.
Rios et al. (2007)	Hawaii	55-980	22 ^d	500	n.a.	n.a.
	California	27-730	100-1100 ^d	1200-6200	n.a.	n.a.
	Los Angeles industrial area	n.d.	42-7100 ^d	39-12000	n.a.	n.a.
Ogata et al. (2009)	Worldwide	5-605	0.14-128	n.a.	n.a.	n.a.
Teuten et al. (2009)	Japanese coast	12-254	0.2-276	<60-9370	0.9-2.1	n.a.
	North Pacific Central Gyre	1-23	0.1-4.7	<100-959	0.4-57	24.9-2660

n.a. - not analyzed.

n.d. - not detected at detection limit.

- a Nonviphenol.
- ^b Limit of detection.
- Two extreme values of 18,600 and 18,700 ng g^{-1} are also reported.
- ^d Including DDT and DDD (dichlorodiphenyldichloroethane, a degradation product of DDT).

Strait, and 1.0 Sv along the Siberian coastline) add up to a maximum volume of $37.8\times10^4\,\text{km}^3$ of sea water expected to be transported annually into the Arctic.

3.3. Plastic flux to the Arctic

The estimated plastic flux to the Arctic Ocean ranges from 62,000 to 105,000 tons per year, assuming the maximum volume transport of ocean water. This calculation is based on average quantities of plastic floating on the world's ocean surface, reported by the United Nations Environment Program ("exceeding 13,000 pieces per km²", UNEP, 2005; approximately 18,000 pieces per km², Ferris, 2009). Some investigations into plastics consider the quantity and weight of the total plastic collected (Day and Shaw, 1987; Moore et al., 2001). Because all samples contained several thousand to hundreds of thousand plastic pieces, the average weight of one plastic piece is fairly constant over the various samples taken (12.6, 13.6 and 15.4 mg, respectively, in the previous studies). Considering quantities of plastic collected in different parts of the world's oceans (Table 2), however, the estimated plastic mass transported to the Arctic Ocean ranges from 16,200 to 1.9 million tons per year. This variation is due to a number of aspects, including:

(1) Spatial heterogeneity: Plastic is spread heterogeneously across ocean surfaces depending on the location (e.g. close to populated and industrial areas), on ocean currents transporting the buoyant debris and on the level of the sea surface, which is influenced by wind and the relief of the seabed. Thus, the North Pacific gyre, one of the world's major gyres, has become an infamous example of marine pollution, concentrating waste material by oceanic rotational currents. Investigations that report the largest quantities of plastic were conducted within this area (Table 2). In other regions, plastics may be washed up on the shore in greater numbers.

- (2) Temporal variability: Over the years, greater plastic production and consumption has led to an increase in the release of plastics into the environment. This time trend has already been observed at different monitoring sites (e.g. Thompson et al., 2004).
- (3) Different sampling methods: The measurements themselves are based on varying methods, and consider different types or sizes of plastics within a single investigation (Table 2), leading to different counts.

In addition, the volume of sea water flowing to the Arctic Ocean also includes deeper layers of the oceanic water body. However, the abundance of plastics at 10–30 m depth is expected to be approximately two orders of magnitude lower than on the ocean surface (Ryan et al., 2009). Thus, the calculation of volume-based plastic fluxes from plastic counts per surface area is probably an upper boundary for the plastic flux to the Arctic Ocean. Nevertheless, a preliminary estimate assuming actual UNEP data (62,000–105,000 tons of plastic per year) seems to be most appropriate. A refined plastic mass balance including estimations of emissions and mass flow analysis is necessary to improve the assessment of possible risks connected to plastics in the marine environment.

3.4. Plastic-mediated pollutant flux to the Arctic

Reported PCB concentrations on plastic fragments and pellets range from 4 to 1260 ng g⁻¹ (Table 1). This results in an estimated annual PCB flux of 250 g to 130 kg per year to the Arctic, with an arithmetic mean of 12 kg a⁻¹. For reasons of consistency, these calculations are compared to an estimated PCB flux based on partition coefficients K_e between ocean water and plastics. Again, reported log K_e values include several orders of magnitude. Mato et al. (2001) determined log K_e values for the absorption of PCBs in PP ranging from 5 to 6, while Pascall et al. (2005) reported sorption into PE, PVC and PS with log K_e values of 3–6, 3–4 and 2–3, respec-

Table 2 Reported quantities (n_P) of plastics collected per km² of the ocean surface, sampling location and sampling year, as well as type of plastic and considered range of plastic particle size.

Reference	Sampling location	Sampling year	Plastic type	Plastic particle size	n_P
Carpenter et al. (1972)	Southern New England	1972	PS spherules	0.1-2 mm	138,000
Day and Shaw (1987)	Subarctic North Pacific	1984	Not specified	>0.5 mm	3400
	Subtropical North Pacific				96,000
Moore et al. (2001)	North Pacific gyre	1999	Not specified	>0.355 mm	332,000

tively. Recent investigations into the sorption of PCBs into polypropylene resulted in $\log K_e$ values of 4.5–7.5, increasing linearly with the octanol-water partition coefficient of the PCB congeners (Beckingham and Ghosh, 2009). Because the amount of PS on plastic particles collected in the ocean seems to be negligible (Moore et al., 2001), we focus on a range of 10^3 – 10^6 L kg⁻¹ for PCB partition coefficients, i.e. $log K_e$ of 3-6. An average PCB concentration of $15 \times 10^{-9} \text{ mg L}^{-1}$ in sub-polar waters, as proposed by Wania (1998), is within the range of more recently determined PCB concentrations of $3.75-21.8 \times 10^{-9} \text{ mg L}^{-1}$ (Smith and McLachlan, 2006). Thus, using this data in combination with Eq. (3) gives a plastic-mediated PCB flux to the Arctic of approximately 1 g to 1.6 kg per year. The difference between these calculations and previous estimations based on reported PCB concentrations in plastic fragments can be attributed to several factors. First, sorbed PCB concentrations in plastic particles used for the first kind of estimation were observed at contaminated sites and are thus high. whereas ocean concentrations used for the K_e -based calculation come from a remote region (the Arctic) and are thus low. Unfortunately, measurements of PCB concentrations in plastics from the Arctic region are not available. In addition, we do not know if sorption of PCBs between sea water and plastic is in equilibrium. Nevertheless, because the equilibrium concentration (C_{ocean}^*) of PCB in sea water would be smaller than actual measurements (C_{ocean}), which might be far from equilibrium, the PCB concentration (C_P) in plastics, estimated via K_e according to Eq. (3), represents an upper boundary for the equilibrium concentration in plastics (C_p^*) :

$$C_P = K_e \cdot C_w \ge K_e \cdot C_w^* = C_P^* \tag{6}$$

The estimated mass of the plastic additive PBDE transported to the Arctic Ocean ranges from 25 g to 5.9 kg per year, using measured PBDE concentrations in plastics of the North Pacific gyre and the Japanese coast, which amount to 0.4 and 57 ng g $^{-1}$ (Teuten et al., 2009). Partition coefficients between ocean water and different plastic materials are not yet available for PBDE. Thus, a consistency check as shown for PCBs is not possible for PBDEs.

Even less data is available for PFOA: ocean concentrations range from 52×10^{-9} to 439×10^{-9} mg L⁻¹ (Yamashita et al., 2008) and organic carbon normalized partition coefficients ($K_{\rm OC}$) are 10^{1} – 10^{3} L kg⁻¹ (Armitage et al., 2009). Because PFOA is an acid with a dissociation constant p $K_{\rm a}$ below 3.8 (Armitage et al., 2009) this compound is mainly available in its ionic state (>99.9%) at oceanic pH (pH 8.1, Orr et al., 2005). Equilibrium sorption of ionizable chemicals to organic carbon ($K_{\rm OC}$) was described by Franco et al. (2009) as the fraction weighted sum of species specific sorption coefficients:

$$K_{\rm OC} = \alpha_n \cdot K_{\rm OC,n} + \alpha_{\rm ion} \cdot K_{\rm OC,ion} \tag{7}$$

 $K_{\text{OC},n}$ and $K_{\text{OC},\text{ion}}$ are the species specific sorption coefficients for the neutral and the ionic molecules, respectively. α_n is the fraction of the neutral species calculated as follows:

$$\alpha_n = \left(1 + 10^{a \cdot (pH - pK_a)}\right)^{-1} \tag{8}$$

where a is 1 for acids and -1 for bases. $\alpha_{\rm ion}$ in Eq. (7) is the fraction of the ionic species (1 $-\alpha_n$ for monovalent acids and bases). Jenke (1993) expressed the overall partition coefficient of acids to plastic as a function of the partition coefficient of the nonionized form ($K_{e,n}$) and the neutral fraction in dependence of solution pH and substance p K_a :

$$K_e = \alpha_n \cdot K_{e,n} \tag{9}$$

and showed that sorption of the anionic fraction of butylbenzoic acid is negligible.

Assuming that sorption of neutral hydrophobic compounds to plastics is two orders of magnitude higher than to organic carbon ($K_e = 10^2 K_{OC}$), as observed for PCBs (Mato et al., 2001), an upper boundary for PFOA sorption to plastics can be derived from Eqs. (7) and (9):

$$K_{e}(PFOA) = \alpha_{n} \cdot K_{e,n} = \alpha_{n} \cdot 10^{2} \cdot K_{OC,n}$$

$$= 10^{2} \cdot (K_{OC} - \alpha_{ion} \cdot K_{OC ion}) < 10^{2} \cdot K_{OC}$$
(10)

Thus, a maximal annual PFOA flux of up to 4.6 kg is expected. Nevertheless, in contrast to PCBs and PBDEs, the sorption of surfactants depends to a great extent on the accessible surface area, and cannot necessarily be described by volume-based partition coefficients. Thus, further research into the surface-dependent sorption behavior of surfactants to plastic particles is required.

3.5. Significance of various pollutant transport routes

Although these rough estimations include considerable uncertainty as far as quantities of plastic and scattered concentration data are concerned, the resulting orders of magnitude for each of the calculated pollution fluxes enable various transport routes to be compared. Assuming an average PCB concentration of 15×10^{-9} mg L⁻¹ in the ocean, the transport of PCBs with ocean currents would result in an annual PCB flux of 5.7 tons. This mass exceeds the flux with plastic particles by two to four orders of magnitude. Reported PCB and PBDE concentrations in Arctic air of $20 \times 10^{-12} \, g \, m^{-3}$ (Wania, 1998) and of $7.8 \times 10^{-13} \, g \, m^{-3}$ to 4.8×10^{-11} g m⁻³ (Su et al., 2007), respectively, allow the respective mass flux with atmospheric currents to be estimated according to Eq. (5). Assuming an average transport rate for air (G_A) of $1.50 \times 10^{19} \,\mathrm{m}^3 \,\mathrm{a}^{-1}$ (Wania, 1998), we obtain a pollutant flux of 300 tons of PCBs and 720 tons of PBDEs per year. Annual mass fluxes of PCBs and PBDEs with atmospheric currents are thus the largest fluxes, exceeding plastic-mediated transport by five orders of magnitude. For PFOA, the mass flux with plastics to the Arctic is five orders of magnitude smaller than from ocean currents, assuming the above-mentioned PFOA concentration of 439×10^{-9} mg L⁻¹ (Yamashita et al., 2008). Further knowledge of surface-dependent sorption coefficients to plastic particles will improve estimates for surfactants.

Because plastic fluxes are assessed by an upper boundary, contaminant fluxes with plastic may even be smaller than estimated, meaning that the contribution of plastic vectors to the mass transport of pollutants to the Arctic would appear to be negligible. However, the number of micro-plastics (<5 mm) probably exceeds actual numbers of collected plastics, because their stake in the total plastic sample exceeds 90% per count (Moore et al., 2001; Moore, 2008) and has been increasing since the 1960s (Thompson et al., 2004). This may also increase the actual pollutant mass transported to the Arctic by plastics, because mass fluxes are not sufficient to assess the relevance of transport routes and the resulting environmental risk as far as availability for uptake by organisms is concerned. It is still unknown how many pollutants that are prone to LRT via atmospheric currents finally enter the aquatic food chain. Plastic particles, in contrast, have been shown to be swallowed by accident by various marine organisms (Carpenter et al., 1972; van Franeker, 1985; Derraik, 2002; Thompson et al., 2004: Colabuono et al., 2009) and to accumulate from prev to predator (Eriksson and Burton, 2003). Filter feeders such as krill (which are the basic source of food for many marine organisms), forage fish (herrings, sardines and others), jellyfish, sharks, whales and sea birds are particularly affected by ingested plastics due to their modes of food intake. Thus, plastic particles are pollutant vectors that can be transported directly into the food chain. In conclusion, the quantification of transported mass fluxes is no sufficient to assess the LRT potential of chemicals. It is even more important to identify transport routes that may directly serve as pollutant vectors into organisms and the food chain, such as plastic particles.

Plastic-mediated transport may be of greater importance for substances that are not transported in air or by ocean currents, e.g. due to rapid degradation or sorption to sediment particles. Because it has been shown that apparent sorption coefficients for PCBs and DDE (dichlorodiphenyldichloroethylene, a degradation product of DDT) in PP pellets are two orders of magnitude higher than for suspended particles (Mato et al., 2001), substances with a high affinity for organic matter may be attached even more strongly to buoyant plastic particles and thus be transported to remote regions by ocean currents. This may be the case for polycyclic aromatic hydrocarbons (PAHs), which show strong sorption behavior to organic carbon (log K_{OC} 3–6, Xu et al., 2010; EPISuite³ v4.0). In addition, because PAH are natural constituents of oil, they are continuously emitted with oil production (and extensively by accidents) directly into the ocean. Nevertheless, substance screening for preferred modes of long-range transport is necessary to assess the relevance of plastics as transport vectors for organic pollutants in general. Future research should therefore focus on refining the mass balance for plastics, with special consideration for micro-plastics, and on concentrations in and sorption into plastic material of substances that are not transported by atmospheric or ocean currents.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.marpolbul.2010.05.026.

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