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Mobilisation of antimony from microplastics added to coastal sediment

Elanor James & Andrew Turner*

School of Geography, Earth and Environmental Sciences
University of Plymouth
Drake Circus
Plymouth PL4 8AA
UK

*Corresponding author. Tel: +44 1752 584570; Fax: +44 1752 584710; e-mail: aturner@plymouth.ac.uk

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Abstract

Antimony (Sb) widely occurs in plastics as a pigment and reaction residue and through the use and recycling of electronic material enriched in Sb as a flame retardant synergist. In this study, clean estuarine sediment has been contaminated by different microplastics prepared from pre-characterised samples of different types of plastic (including a rubber) containing a range of Sb concentrations (256 to 47,600 μg g^{-1}). Sediment-plastic mixtures in a mass ratio of 100:1 were subject to 6-hour extractions in seawater and in seawater solutions of a protein (bovine serum albumin; BSA) and a surfactant (taurocholic acid; TA) that mimic the digestive conditions of coastal deposit-feeding invertebrates. Most time-courses for Sb mobilisation could be defined by a second-order diffusion equation, with rate constants ranging from 44.6 to 0.0216 $(\mu g\ g^{-1})^{-1}\ min^{-1}$. Bioaccessibilities, defined as maximum extractable concentrations throughout each time course relative to total Sb content, ranged from < 0.01 % for a polycarbonate impregnated with Sb as a synergist exposed to all solutions, to > 1 % for acrylonitrile butadiene styrene containing a Sb-based colour pigment exposed to solutions of BSA and TA and recycled industrial polyethylene exposed to BSA solution. The potential for Sb to bioaccumulate or elicit a toxic effect is unknown but it is predicted that communities of deposit-feeders could mobilise significant quantities of Sb in sediment contaminated by microplastics through bioturbation and digestion.

Keywords: antimony; plastics; sediment; mobilisation; bioaccessibility

Capsule: Antimony is mobilised from plastics added to sediment by seawater and solutions of a protein and a surfactant via a diffusion model

1. Introduction

Antimony and many of its compounds display both acute and chronic toxicity to a range of organisms (Tschan et al., 2010; Paoli et al., 2013; Yang, 2014; Yang et al., 2018). In humans, exposure to Sb has been related to respiratory, cardiovascular and gastrointestinal symptoms and antimony trioxide (Sb_2O_3) is classified as possibly carcinogenic (Sundar and Chakravarty, 2010; Tamás, 2016). Antimony is also a technology-critical element in that is has economic importance to emerging technologies but is subject to a high supply risk (Nuss and Blengini, 2018). Current applications of Sb include pigments for colour or colour protection, decolouring and refining agents in glass, catalysts for polycondensation, synergists for flame retardants, and alloys in batteries in ammunition, antifriction bearings, cable sheaths and roofing (Yellishetty et al., 2017).

Antimony is particularly pervasive in synthetic polymers (including textiles, plastics and rubbers) (Filella et al., 2020). Here it is used in various pigments, but its principal application that results in the highest concentrations is as a synergist for halogenated flame retardants (as Sb_2O_3, the pentoxide, Sb_2O_5, or sodium antimonite, Na_3SbO_4; Papazoglou, 2004). The trioxide is also used as a catalyst for the production of polyethylene terephthalate (PET), including fibrous polyester, and residues of the compound and other reaction products (including organic-complexes of Sb) remain in the material after manufacture (Welle and Franz, 2010). Antimony is more widely dispersed amongst consumer and commercial plastics through the recycling of end-of-life products. In this respect a particular problem arises when flame-retarded electronic plastics are recycled into consumer goods in which halogenated compounds and Sb are neither expected nor desired (Turner and Filella, 2017).

Concerns over the presence and possible migration of Sb in consumer products, and in particular plastics, has resulted in guidelines or regulations for the metalloid. In PET food-contact and storage items, where Sb concentrations are typically a few hundred $\mu g\ g^{-1}$ (Westerhoff et al., 2008), migration into liquids is effectively limited to 5 $\mu g\ L^{-1}$ according to EC Directive 2003/40 on natural mineral waters (European Commission, 2003). Migration into foods is limited to 40 μg per kg of food according to EU Regulation 10/2011 European Commission, 2011) and based on the tolerable

daily intake of 6 μg kg^{-1} body weight (World Health Organization, 2003). In plastic, rubber or painted toys, migration of Sb is limited by the amended EC Toy Safety Directive to between 11.3 and 560 μg g^{-1} depending on the characteristics of the material (based on pliability, size, thickness, brittleness) (European Parliament and Council of the EU, 2009). Despite concentrations of Sb in flame-retarded electronic plastics that can exceed several percent by weight (Papazoglou, 2004), the metalloid is currently not included in the list of substances defined by the original or recast Restriction on Hazardous Substances (RoHS) Directives (European Parliament and Council, 2003; 2011). However, the EU regards Sb as a "heavy metal" and defines waste as "ecotoxic" if heavy metal concentrations exceed certain thresholds (European Commission, 2018).

As a result of its widespread use in and contamination of polymers, Sb is predicted to be commonly encountered in marine litter. X-ray fluorescence (XRF) analysis of consumer, maritime and industrial plastics collected by hand from the strandlines of various beaches in south west England has revealed the presence of Sb in up to 50% of a wide variety of samples of relatively low density (but mainly polyethylene and polypropylene) (Shaw and Turner, 2019; Turner et al., 2019). However, given that microplastics buried below the sediment surface, including microfibers, appear to contain a higher proportion of denser materials like PET (Yu et al., 2018; Zheng et al., 2019), the overall abundance of Sb in marine litter may be significantly greater.

In coastal regions where the substrate is contaminated by synthetic polymers (hereafter referred to as plastics), benthic and infaunal animals may be exposed to elevated levels of Sb. Of particular significance in this respect are deposit-feeding invertebrates that non-selectively process large quantities of material through burrowing, ingestion and egestion. Exposure could increase should quantities of Sb be mobilised from microplastics through bioturbating activities, and in particular during digestion. Accordingly, the present study examines the potential for Sb to be released from plastics via coastal deposit-feeders by incubating preparations of clean sediment and a range of microplastics in fluids that simulate the invertebrate digestive environment. Specifically, we use seawater solutions of the protein, bovine serum albumin (BSA; molecular mass = 66,400 g mol^{-1}), to mimic the complexing capacity

of amino acids (Chen and Mayer, 1998), and the vertebrate bile acid surfactant, taurocholic acid (TA; $C_{26}H_{44}NO_7SNa$; molecular mass = 537.7 g mol^{-1}), as a surrogate for anionic surfactancy (Voparil and Mayer, 2004).

2. Materials and methods

2.1. Sourcing and characterisation of plastics

About 50 plastics items sourced from archived litter samples and consumer products were analysed for total Sb and polymer type according to protocols outlined elsewhere (Turner et al., 2019). Briefly, Sb was determined on three regions of each sample by portable XRF spectrometry using a battery-operated Niton XL3t 950 He GOLDD+ portable XRF. The instrument was operated for a period of 60 seconds in a "plastics" mode with thickness correction and performance was checked regularly by analysing a polyethylene reference disk reference (Niton PN 180-619) that contained Sb at a concentration of 96 $\pm$ 10 μg g^{-1}. Polymer identification on small (< 5 mg) sample offcuts was performed by attenuated total reflectance Fourier-transform-infrared (ATR-FTIR) spectrometry using a Bruker Vertex 70.

Based on the analyses above, seven plastics of varying Sb content and polymeric composition, including one sample of rubber, were selected for use in the extraction experiments (Table 1). The sample of PET likely contains Sb as catalytic residue arising from the manufacture of the material, while acrylonitrile butadiene styrene (ABS) contains Sb as a component of the pigment titanium yellow (CI Pigment Yellow 53; $NiO{\cdot}Sb_2O_3{\cdot}20TiO_2$, and confirmed by the presence of Ni and Ti in the XRF spectrum). Polystyrene (PS), polycarbonate (PC) and polyethylene (PE) contain Sb as a flame retardant synergist in the presence of brominated compounds, and polyvinyl chloride (PVC) and rubber (R) appear to contain Sb as a synergist in the presence of chlorinated compounds. Using a stainless steel grater, at least 1 g of each sample was formulated into microplastic particles (< 1 mm in at least two dimensions) that were stored in individual polyethylene specimen bags.

2.2. Sediment sampling and processing

Approximately 1.5 L of surficial, oxic sandy-silty sediment was collected using a plastic trowel from the lower, intertidal reaches of a protected estuary that is

relatively unpolluted with regard to both metals and plastics (Erme, south west England; 50.3111, -3.9457). On site, the sample was sieved though a 1 mm Nylon mesh into a plastic bucket. In the laboratory, subsamples of about 250 g were transferred to a series of clear zip-locked polyethylene bags and stored frozen until required. The chemical characteristics of the fractionated sediment sample, determined on freeze-dried aliquots, are given in Table 2. Here, metal concentrations were determined by inductively coupled plasma-optical emission spectrometry (ICP-OES) and Sb concentration by inductively coupled plasma-mass spectrometry (ICP-MS; see below) according to established protocols (Turner, 2019) and following digestion of triplicate 250 mg aliquots in aqua regia heated to 80 ^{o}C for 2 h. Loss on ignition (LOI) as a proxy for organic matter content was determined by mass loss on ignition at 500^{o}C for 8 h.

2.3. Experimental

One-litre solutions that mimic the chemical conditions (amino acid composition and surfactancy) encountered in the digestive environment of temperate, coastal deposit-feeding invertebrates were prepared in 0.45 μm-filtered English Channel seawater (*S* = 33; pH 7.8) in a series of borosilicate bottles. Specifically, these consisted of 4 g L^{-1} of the protein, BSA (> 96% fraction V; Sigma Aldrich), 4 g L^{-1} of TA (taurocholic acid sodium salt hydrate, 97%+; VWR International), and 4 g L^{-1} of BSA plus 4 g L^{-1} TA.

Experiments were undertaken according to protocols outlined in Jones and Turner (2010) and Martin and Turner (2019). Thus, bags of sediment were defrosted as required and aliquots of 25 g (or 18.26 g on a dry weight basis) were weighed into a series of eight screw-capped 200 mL polyethylene bottles. To seven of the bottles, 250 mg of the different microplastics were added, with the eighth bottle serving as a plastic-free control. The concentration of plastic in the sediment on a dry weight basis was, therefore, about 14 g kg^{-1}. This is higher than microplastic abundance reported in contaminated sediments (up to about 130 mg kg^{-1}; He et al., 2020) but was selected in order to allow the ready detection of mobilised Sb.

One hundred ml of seawater (SW) was pipetted into each bottle before the contents were agitated on a Stuart SSL1 benchtop orbital shaker set at 150 rpm at room temperature and in the dark. After 15 min, 30 min, 1 h, 2 h, 4 h, 5 h and 6 h, subsamples of 5 mL were pipetted into 15 mL polypropylene centrifuge tubes and centrifuged at 3000 rpm for 10 min before 2-mL supernatants were transferred to Sterilin tubes, diluted to 10 ml with 2% HNO_3 and stored at 4^{o}C and in the dark pending analysis. This approach was repeated using seawater solutions of BSA (SW + BSA), taurocholic acid (SW + TA) and BSA plus taurocholic acid (SW + BSA + TA).

2.4. Analysis of extracts

A ThermoScientific iCAP RQ ICP-MS with a Glass Expansion micromist nebuliser and cyclonic spray chamber was used to determine Sb concentrations in the diluted extracts. The instrument was calibrated with a blank and four standards (up to 20 μg L^{-1}) prepared by serial dilution of a LabKings 10000 mg L^{-1} Sb standard in 2% HNO_3. Radio frequency power was set at 1.5 KW with coolant, nebuliser and auxiliary flows of 1.4, 1.07 and 0.8 L Ar min^{-1} and a replicate (n = 3) read time of 10 ms. Detection limits arising from three standard deviations of blank measurements were between 0.05 and 0.10 μg L^{-1} and precision among replicate readings was better than 15% for mean concentrations above 0.5 μg L^{-1} and up to 25% for lower mean concentrations .

3. Results and Discussion

3.1. Antimony concentrations in the plastic-amended sediment samples

Table 3 presents the dry weight concentrations of total Sb in the estuarine sediment and in sediment amended by the different microplastics defined in Table 1. Note that in the sediment amendments concentrations are computed from the fractional contributions of sediment and plastic to total dry mass and the total concentrations of Sb in the different solids. Thus, mass contamination of coastal sediment by about 1.4% of these plastics results in total Sb concentrations that are orders of magnitude greater than the Sb content of sediment itself.

3.2. Antimony mobilisation kinetics

Figure 1 shows the dry weight normalised concentrations of Sb released from the sediment and sediment amendments, [Sb], for the different solutions as a function of time, *t*. In some cases, [Sb] increases throughout the incubation (e.g. sediment + R in SW + BSA) or there is a reduction in [Sb] following a period of rapid increase (e.g. sediment + PE in SW + BSA). In most cases, however, [Sb] increases over the experimental period and appears to approach equilibrium. Here, data were modelled using a second-order diffusion model (Ruby et al., 1992; Martin and Turner, 2019):

$$1/([Sb]_e - [Sb]) = 1/[Sb]_e + kt \quad (1)$$

where $[Sb]_e$ is the "equilibrium" concentration of the metalloid defined as the highest concentration reported among the last five data points (i.e. $t > 120$ min), and k is a second-order rate constant of units $(\mu g\ g^{-1})^{-1}\ min^{-1}$. Rate constants were derived from the slopes of $(1/([Sb]_e - [Sb]) - 1/[Sb]_e)$ versus t, provided that linear regressions were significant ($p < 0.05$), and are given in Table 4 along with values of $[Sb]_e$. Where timed data were more complex or equilibrium was not approached, maximum measured values of the metalloid, $[Sb]_{max}$, are given in the table.

For sediment alone, the greatest concentration of Sb mobilised (as $[Sb]_e$ or $[Sb]_{max}$) was in SW. Among the samples amended with plastics, however, there was no clear pattern of mobilisation among the different solutions, with each mobilising the greatest concentration of Sb from at least one sample. Overall, second-order rate constants ranged from 44.6 $(\mu g\ g^{-1})^{-1}\ min^{-1}$ for sediment amended with PS in the presence of SW + BSA + TA to 0.0216 $(\mu g\ g^{-1})^{-1}\ min^{-1}$ for sediment amended with PE in the presence of SW alone.

3.3. Antimony bioaccessibility

Bioaccessibility may be defined as the fraction of a contaminant that is available for dissolution in the digestive tract of an organism, and provides an upper-estimate of bioavailability, or the amount that is able to enter the systemic circulation (Weston and Mayer, 1998). Upper estimates of the percentage bioaccessibility of Sb in sediment and sediment amended with plastic to a range of deposit-feeding invertebrates (i.e. with varying proteins and degrees of surfactancy in their digestive

environments) were calculated from $[Sb]_e$ or $[Sb]_{max}$ relative to total Sb and are shown in Table 5. Overall, BA is greatest for sediment alone, where the metalloid is likely adsorbed to the particles surface and bound in different mineral phases of varying solubility, with estimates ranging from 0.67 for SW + TA to 2.67 % for SW itself. Where plastic is present, the highest values of bioaccessibility were observed for amendments with ABS, PE and R. For ABS, similar values of bioaccessibility were exhibited for each solution. However, sediment amended with PE exhibits a value that is greatest by two orders of magnitude when BSA is present without TA, and sediment amended with R exhibits values that are an order of magnitude greater for SW and SW + BSA than when TA is present. Among the remaining plastics, bioaccessibility is lowest overall for PC and greatest for PET.

3.4. Mechanisms of antimony mobilisation from plastics

The release of ions and molecules from polymers normally proceeds by diffusion (Nakashami et al., 2016; Town et al., 2018) but very little has been studied in respect of Sb release from marine plastics. Specifically, as part of a study into metal and metalloid bioaccessibility in various plastics subject to simulated, acidic avian digestion, Turner (2018) established that Sb mobilisation from two samples of micronized polypropylene under simulated avian digestive conditions proceeded via a pseudo-first-order diffusion process with rate constants of about 0.05 h^{-1}.

Because of human health concerns, a greater body of literature exists regarding the migration of Sb from polyethylene terephthalate (PET) food-contact plastics that contain the metalloid as catalytic residue (Haldimann et al., 2007; Rungchang et al., 2013; Chapa-Martínez et al., 2016). It appears that Sb diffusion is dependent on the physical and chemical nature of migrating species or compounds, the properties of the plastic, including crystallinity, polarity and molecular weight, plastic surface area to solution volume ratio, and the presence of any other additives in the plastic that may retard or facilitate the diffusion process through, for example, adsorption (Westerhoff et al., 2008; Haldimann et al., 2013).

In this study, Sb is likely present as Sb_2O_3 where the metalloid is employed directly (or introduced through recycling) as a flame retardant synergist (Papazoglou, 2004), and, additionally, as a series of glycolate complexes in PET as products of the

polymerisation process (El-Toufaili et al., 2006); in ABS, however, Sb is present as part of the complex pigment, titanium yellow. Accordingly, the comparatively rapid release of Sb from PET can be attributed to the relatively high diffusivity of rather small organic-complexes of Sb (Welle and Franz, 2010), while for ABS enhanced mobilisation could be due to the presence of Ti, which, despite its sorptive properties, has been observed to promote diffusion of Sb in plastic (Haldimann et al., 2013). Differences between the mobilisation of Sb from other materials can be related to polymer permeability; for example, published permeation coefficients (for oxygen) are around 10^3 cm^3 mm m^{-2} d^{-1} atm^{-1} for various rubbers but are an order of magnitude lower for PVC, PS and PC (Keller, 2017).

Despite these qualitative explanations, however, it is important to bear in mind that net mobilisation of Sb is also dependent on largely unknown interactions of migrating species with the sediment surface (e.g. through adsorption) and substances present in the aqueous medium (e.g. through complexation), and interactions among aqueous constituents and between these constituents and the sediment surface.

3.5. Bioaccessibility and mobilisation of Sb in the coastal zone

Antimony is commonly encountered in a wide range of plastics as a flame retardant synergist (often through the recycling of electronic waste), a pigment and a catalyst, and is one of the main inorganic residues in polyester fibres (Turner, 2019). Given the abundance of microplastics in many coastal sediments (up to several thousand kg^{-1} have been recorded; Lots et al., 2017; Lo et al., 2018; Wang et al., 2019) these particles could represent a significant anthropogenic source of Sb in the littoral zone. For example, 1 kg of the estuarine sediment used in the present study would double its Sb concentration (to 0.36 μg g^{-1}) if contaminated by just 3.6 mg of flame-retarded rubber particles.

Many invertebrates inhabiting the littoral zone incidentally and non-selectively ingest microplastics as part of their sedimentary diet (Van Cauwenberghe et al., 2015; Setala et al., 2016) and are thereby exposed to quantities of additives and residues that, in some cases, could be considerable. For example, colonies of the deposit-feeding lugworm, *Arenicola marina*, may actively process up to 80 cm^3 of sediment per m^2

per day (Kesy et al., 2016) while individual holothurians may ingest-defecate up to 82 kg per year (Renzi et al., 2018). Whether sufficient Sb is bioaccessible and accumulated to elicit some adverse effect is unknown because the toxicity of the metalloid in the marine environment is poorly documented. However, the processing and bioturbation of plastic-contaminated sediment is likely to be critical to the mobilisation and redistribution of more bioavailable forms of the element (and to polymer-bound contaminants more generally) in the local interstitial environment and overlying water column.

4. Conclusions

Antimony is a technology-critical element that is commonly present in plastics as an additive, residue or contaminant. This study has shown that Sb can be partially mobilised from micronized plastics added to sediment by seawater and solutions mimicking the digestive chemistry of deposit-feeding invertebrates. Mobilisation often proceeds via a second-order diffusion model and over a six-hour period can range from between about 0.01% and 3.5% of total Sb depending on the type of solution and nature of the plastic. Large communities of deposit-feeders could be instrumental to the dissolution and mobilisation of the element from sediment contaminated by microplastic.

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Table 1: Characteristics of the plastic samples employed in the experiments. Total Sb concentrations are reported as the mean and one standard deviation arising from three XRF measurements.

sample description	polymer	Sb, μg g^{-1}	Sb origin
clear water bottle	polyethylene terephthalate (PET)	365±66.6	catalytic residue
black Xmas bauble	polystyrene (PS)	890±29.6	flame-retardant synergist (recycled)
white beached litter fragment	polyvinyl chloride (PVC)	6260±14.7	flame-retardant synergist
white electrical casing	polycarbonate (PC)	27800±100	flame-retardant synergist
yellow-green Lego brick	acrylonitrile butadiene styrene (ABS)	256±23.9	colour pigment
black beached industrial biobead	polyethylene (PE)	2170±30.1	flame-retardant synergist (recycled)
black rubber	rubber (R)*	47600±321	flame-retardant synergist

*FTIR failed to determine the precise nature of the rubber because of the high absorbance of the material.

Table 2: Concentrations of geochemically important metals, total Sb and organic matter (as loss on ignition, LOI) in the sieved estuarine sediment sample. Concentrations of metals and Sb are totals recovered by aqua regia and are shown as the mean and one standard deviation arising from three independent determinations.

Al, mg g^{-1}	6.75±0.36
Ca, mg g^{-1}	82.2±8.9
Fe, mg g^{-1}	14.4±0.7
Mn, μg g^{-1}	290±11
Sb, μg g^{-1}	0.18±0.04
LOI, %	1.44

Table 3: Total concentrations of Sb in the sediment amended by the different micronized plastics (as defined in Table 1).

plastic-amended sediment	Sb, μg g^{-1}
sediment	0.18
sediment + PET	5.1
sediment + PS	12.2
sediment + PVC	84.8
sediment + PC	376
sediment + ABS	3.6
sediment + PE	36.8
sediment + R	643

Table 4: Equilibrium or maximum concentrations of Sb in the time-courses shown in Figure 1. Rate constants were derived from equilibrium concentrations by linear regression analysis according to equation 1, and values are not shown where regressions were non-significant ($p > 0.05$).

sample	constant	SW	SW+BSA	SW+TA	SW+BSA+TA
sediment	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	(0.0048)	0.0016	(0.0012)	(0.0044)
	k, (μg g^{-1})$^{-1}$ min^{-1}		8.88		
	r^2		0.655		
sediment + PET	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	0.0029	(0.0013)	(0.0013)	(0.0037)
	k, (μg g^{-1})$^{-1}$ min^{-1}	4.36			
	r^2	0.504			
sediment + PS	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	0.0043	0.0024	(0.0013)	0.0040
	k, (μg g^{-1})$^{-1}$ min^{-1}	6.58	2.78		44.6
	r^2	0.688	0.634		0.857
sediment + PVC	$[Sb]_e$, μg g^{-1}	0.013	0.0095	0.014	0.022
	k, (μg g^{-1})$^{-1}$ min^{-1}	1.18	7.99	10.8	9.25
	r^2	0.846	0.774	0.807	0.777
sediment + PC	$[Sb]_e$, μg g^{-1}	0.020	0.011	0.035	0.025
	k, (μg g^{-1})$^{-1}$ min^{-1}	0.240	2.43	0.908	0.917
	r^2	0.731	0.814	0.840	0.827
sediment + ABS	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	0.026	0.036	0.036	(0.055)
	k, (μg g^{-1})$^{-1}$ min^{-1}	1.54	0.74	3.19	
	r^2	0.379	0.692	0.887	
sediment + PE	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	0.0019	(1.30)	(0.015)	0.012
	k, (μg g^{-1})$^{-1}$ min^{-1}	2.87			4.25
	r^2	0.760			0.791
sediment + R	$[Sb]_e$ (or $[Sb]_{max}$), μg g^{-1}	1.65	(2.11)	0.13	(0.17)
	k, (μg g^{-1})$^{-1}$ min^{-1}	0.0216		0.132	
	r^2	0.933		0.882	

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560 Table 5: Percentage bioaccessibilities of Sb in sediment amended by different
561 microplastics derived from equilibrium or maximum concentrations reported in Table
562 4 relative to total concentrations given in Table 3.
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	SW	SW+BSA	SW+TA	SW+BSA+TA
sediment	2.67	0.89	0.67	2.44
sediment + PET	0.057	0.025	0.025	0.073
sediment + PS	0.035	0.020	0.011	0.033
sediment + PVC	0.015	0.011	0.017	0.026
sediment + PC	0.005	0.003	0.009	0.007
sediment + ABS	0.72	1.00	1.00	1.53
sediment + PE	0.005	3.53	0.041	0.033
sediment + R	0.26	0.33	0.020	0.026

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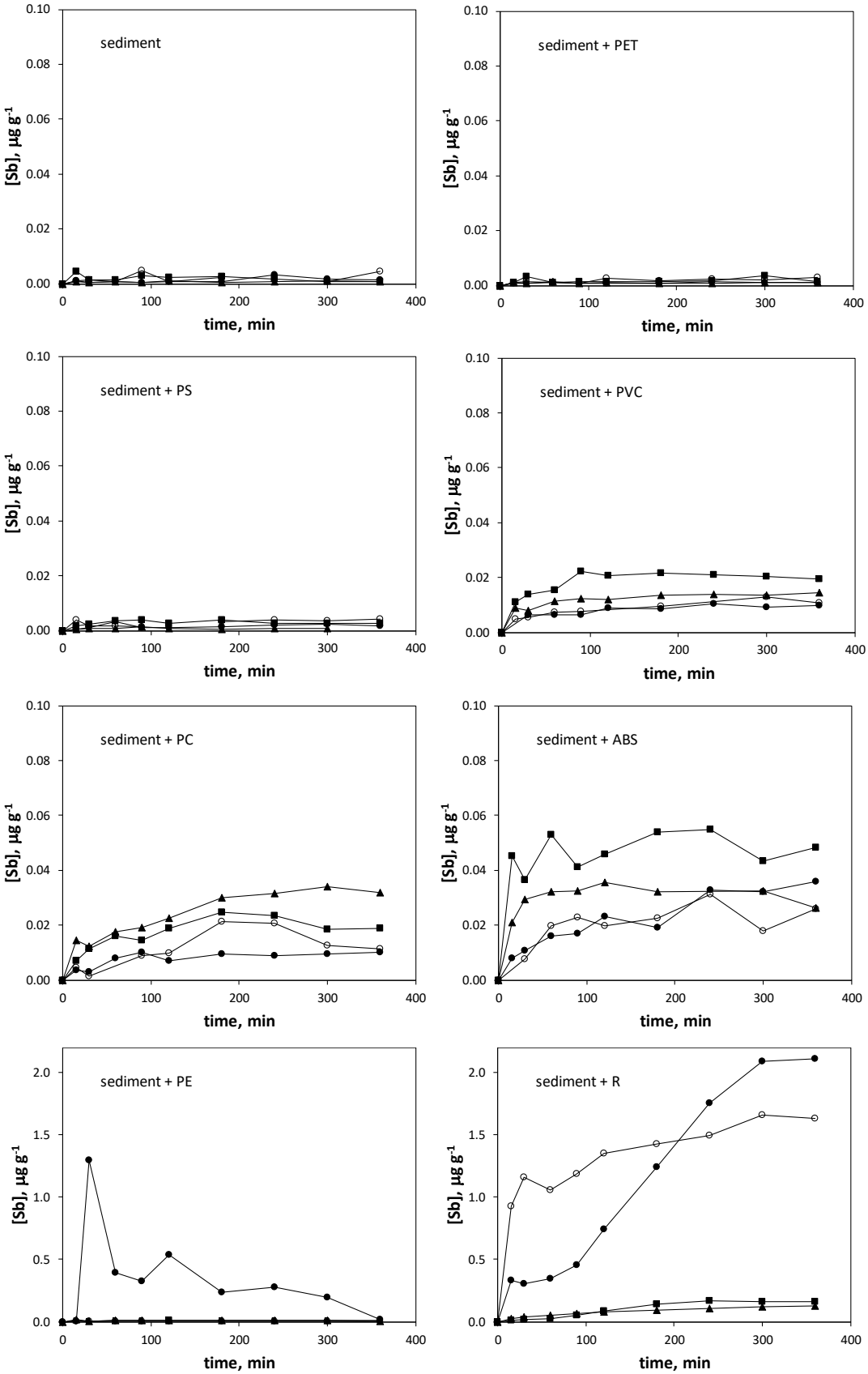
567 Figure 1: Concentrations of Sb released from 25 g of wet estuarine sediment (or 18.26 g on a dry
568 weight basis) and 25 g of wet sediment amended with 250 mg of different plastics by seawater (○) and
569 by seawater in the presence of BSA (●), taurocholic acid (▲) and BSA plus taurocholic acid (■) as a
570 function of time.


sediment
sediment + PET
sediment + PS
sediment + PVC
sediment + PC
sediment + ABS
sediment + PE
sediment + R
[Sb], µg g-1
time, min

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