**Evidence of Increased Anthropogenic Emissions of Platinum:
Time-Series Analysis of Mussels (1991-2011) of an Urban Beach**

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Abstract

The anthropogenic emissions of Pt to the environment have increased significantly over the past decades, especially after the introduction of the catalytic converters in motor vehicles. In order to check whether this is affecting the levels of this trace metal on living organisms, time-series analysis of wild mussels (*Mytilus galloprovincialis*) covering the period from 1991 to 2001 and collected at an urban beach in the city of Vigo (NW Iberian Peninsula) were conducted. Concentrations ranged from 0.30 to 0.68 ng g⁻¹ with an average concentration of 0.47 ± 0.10 ng g⁻¹ (n=21); these concentrations were higher than those obtained for samples collected at a control location away from anthropogenic pressure (0.31 ± 0.10 ng g⁻¹; n=5). Platinum concentrations followed a statistically significant temporal trend (at the 0.020 level), and the excess of Pt in mussels over the 1991-2011 period compared to the control location were correlated with the European Pt autocatalyst demand (p=0.0006) and, especially, the car sales in Spain (p=0.0001). A bioaccumulation factor of ~ 5 10³ was derived, which is greater than those previously calculated for Pt from exposure experiments, but 1-2 orders of magnitude lower than other trace elements (e.g. Zn, Cu, Pb, Cd).

Keywords:
Platinum; Bioaccumulation; *Mytilus galloprovincialis*; Urban contamination

1. Introduction

The natural environmental concentrations of platinum in the biosphere are extremely low, with average crustal abundance of 0.5 ng g^{-1} (Rudnick and Gao 2003). However, Pt concentrations are currently increasing in the environment due to its use in a variety of anthropogenic activities, the most important including the use of this element in automobile catalytic converters and the manufacture of jewellery, representing 43% and 31%, respectively, of the total Pt demanded during the last decade (Johnson Matthey 2013). The anthropogenic disturbance of the cycle of Pt at the Earth's surface is such that it has been estimated that more than 80% of the Pt flux is derived from anthropogenic activities (Sen and Peucker-Ehrenbrinck, 2012).

As a result of the Pt emissions from catalytic converters in motor vehicles, this element currently represents a new category of traffic related trace metal contaminant in the environment (Haus et al., 2007). Accordingly, elevated Pt concentrations have been reported in areas and environmental compartments subject to vehicular traffic pressure such as road dust, roadside soils, sewage sludge and sediments of urban rivers and harbour basins (Schäfer et al., 1999; Ely et al., 2001; Fritsche and Meisel, 2004; Lesniewska et al., 2004; Zimmermann and Sures, 2004); also, evidence for a long range transport and global platinum environmental perturbation has been given (Soyol-Erdene et al., 2011).

The bioavailability and uptake of Pt emitted from catalytic converters was demonstrated in exposure studies using different aquatic organisms, including freshwater isopods (Moldovan et al., 2001), European eels (Zimmermann et al., 2004), zebra mussels (Sures and Zimmermann, 2007), or common periwinkles (Mulholland and Turner, 2011). However, to date only few studies have been carried out reporting ambient concentrations of Pt in biological matrices. Among these, (i) Jensen et al. (2002) analyzed the contamination of feathers from raptor species showing an increase temporal trend of Pt concentrations reflecting the introduction of automobile catalytic converters; also, (ii) Haus et al. (2007) demonstrated the bioavailability of traffic-borne Pt in field samples of freshwater crustaceans from the Ruhr district (Germany) showing bioaccumulation factors in the range of other traffic related metals.

In this study we report Pt concentrations in time-series samples (1991-2011) of wild mussels (*Mytilus galloprovincialis*) collected in an urban beach of the Vigo Ria (NW Iberian Peninsula; Figure 1). Mussels are ideal organisms for use as bio-indicators because as filtering organisms tend to accumulate dissolved substances in the environment (e.g. Goldberg, 1986; Soto et al., 1997; Bellas et al., 2014) and have been widely used in the study area for biomonitoring of metal contamination (Besada et al., 2002, 2011). The main aim of this study is therefore to determine the temporal variation of Pt biological uptake in an urban beach during the past two decades and results will be discussed in terms of the potential link between the temporal variation of Pt accumulation in mussels and the anthropogenic use of this element from the early 1990's.

2. Material and methods

2.1. Sampling area

Wild mussels (*Mytilus galloprovincialis*) were collected manually and during low tides, in the period of September-November, which corresponds to the second prespawning period in this area (Caceres-Martinez and Figueras, 1998) in order to minimize variations caused by differences in the mussel physiology and therefore minimize seasonal environmental variations. Sampling strategies during this two decade long program are described elsewhere (Besada et al., 2014).

Mussels were collected at Samil beach, an urban beach located in the city of Vigo (Figure 1) which holds a population of aprox. 300,000 inhabitants; an urban stream (Lagares, mean annual flow of $\sim 4 \text{ m}^3\text{s}^{-2}$; Perez-Arlucea et al. 2005) discharges at the western part of

the beach. For comparison purposes, mussel samples from a control point (Oia, Figure 1) located around 40 km southward and far from any significant contamination influence were analyzed for selected years (1991, 1995, 2000, 2005, 2010).

2.2. Pretreatment

Each sample of mussels was prepared from 50 or more individuals representing the available size range (35-60 mm) existing in the sampling location. Soft tissues were separated from the shells, triturated with Ultraturrax and freeze-dried. An aliquot of the liophilized sample was withdrawn to calculate its water percentage (drying at 105 °C for 24 hours, until constant weight). After a freeze-drying process, samples were homogenised again with a mixer mill and stored in acid-washed glass vials at room temperature until analysis.

2.3. Pt analysis

Platinum analysis were performed by means of catalytic adsorptive cathodic stripping voltammetry after appropriate digestion (Cobelo-García et al., 2014); briefly, around 100 mg of sample was ashed up to 800°C in quartz crucibles in order to eliminate refractory organic material that may interfere during the voltammetric determination. Once cooled, a mixture of 3 mL of HCl and 1 mL of HNO₃ were added to the crucible and allowed to rest for at least 1h. Then, the acid mixture was transferred to uncapped 30 mL PFA vials (Savillex) and placed on a Teflon-coated hot plate at 195°C and allowed to evaporate until near dryness. After cooling down, 1 mL of H₂SO₄ was added to the vials and placed again on the hot plate until no fumes were observed (only sulphuric acid was present). After cooling, samples were diluted with 0.1M HCl and transferred to 25 mL polypropylene volumetric flasks pending analysis. Typically, 3 independent digestions were performed for each sample and the relative standard deviation (RSD%) was generally below 15%.

In order to avoid contamination of samples, sample pretreatment and manipulation was performed in a laminar flow bench (ISO-5) housed inside and ISO-7 lab. Voltammetric determinations were carried out using a µAutolab Type III potentiostat (Metrohm Autolab B.V.) connected to a polarographic stand (Metrohm model 663VA). Details of the voltammetric procedure for the Pt determination are given in Cobelo-García et al. (2014). The detection limit was determined as three times the standard deviation of the blanks ($n=15$) and was found to be 0.04 ng g⁻¹. Concentrations are expressed as in ng g⁻¹ dry weight. Since at present there are no certified reference materials (CRM) for platinum in biological matrices, the accuracy of the analytical procedure was checked using BCR-723 (road dust; Institute for Reference Materials and Measurements, Belgium) and JSd-2 (river sediment; Geological Survey of Japan), obtaining recoveries greater than 95% (BCR-723) and 80% (JSd-2).

3. Results and Discussion

3.1 Temporal trend of Pt accumulation in mussels

The results obtained for the time series (1991-2011) of mussels collected in the Samil urban beach ranged from 0.30 to 0.68 ng g⁻¹ (Figure 2), with an average concentration of 0.47 ± 0.10 ng g⁻¹ ($n=21$). Lower concentrations were obtained for the samples collected at the control location (Oia, Figure 1): 0.31 ± 0.10 ng g⁻¹ ($n=5$), indicating the higher anthropogenic pressure at the urban beach. These concentrations are in the range of those given for other aquatic organisms previously reported in the literature (with the exception of the elevated concentrations given for the freshwater crustacean *Asellus aquaticus* in two Swedish rivers subject to traffic pressure; Table 1) which indicate that Pt is typically present at the nanogram or sub-nanogram per gram level in aquatic biota. These values are also similar to the crustal abundance of Pt (0.5 ng g⁻¹; Rudnick and Gao 2003) or its concentrations in uncontaminated coastal sediments (0.3-0.6 ng g⁻¹; Cobelo-García et al. 2011).

In order to check whether the Pt concentrations followed a statistically significant temporal trend, the Kendall's Tau-b correlation coefficients were calculated as described earlier (Besada et al. 2002, 2014). A Kendall Tau-b coefficient of 0.324 was obtained, which represents a significance level of 0.020. This significant increase in Pt concentrations is accompanied by a decrease in Pb (Figure 2; Kendall Tau-b -0.663, significance level 0.000), a metal historically linked to traffic-borne pollution due the use of leaded gasoline in the past. The phasing-out of leaded gasoline during the mid to late 1990's was simultaneous with the introduction of catalytic converters (containing precious metals – Pt, Pd, and Rh) and thus explains their opposite temporal trend observed (Figure 2)

3.2 Link between Pt accumulation and traffic-borne contamination

Since the most important anthropogenic input of Pt to the environment is currently its use in vehicle catalytic converters (e.g. Cobelo-García et al. 2011), the excess (or anthropogenic) fraction of Pt in mussels from the urban beach were compared to the autocatalyst platinum demand in Europe and the car sales in Spain – owing that the Pt release rate from new cars is significantly greater than for old cars; Palacios et al. 2000) – for the studied period (Figure 3). The excess Pt fraction was calculated subtracting the background concentrations obtained from the control point (see Material and Methods). Inspection of Figure 3 shows that the increase of the excess fraction of Pt starts in the late 1990's, well before the sharp increase in the Pt autocatalyst demand during the 2000's but coincident with the rise of car sales for those years. It is also evident a decrease in the excess Pt in the last years in agreement with the drop in Pt demand and car sales provoked by the recent economic crisis. A significant correlation was obtained between the excess Pt and the autocatalyst demand ($p=0.0006$) and, especially, the car sales in Spain ($p=0.0001$), which provides a strong evidence of the traffic-borne contamination as the source of the increased Pt concentrations in the mussel samples in the past two decades at this urban beach.

3.3 Extent of bioaccumulation of Pt in *Mytilus galloprovincialis*

The bioaccumulation factor (BF) of platinum, defined as:

$$BF = [Pt_m]/[Pt_w]$$

where $[Pt_m]$ is the Pt concentration in the mussels (expressed as nanograms of Pt per kilogram of mussel in a dry weight basis) and $[Pt_w]$ is the dissolved Pt concentration in the water column (expressed as nanograms of Pt per kilogram of water), was calculated using the average concentrations in the mussels (470 ng kg⁻¹; Table 1) and a typical dissolved Pt concentration in the Vigo Ria of 0.5 pM, i.e. 0.1 ng kg⁻¹ (Cobelo-García et al. 2013), resulting in a BF of $\sim 5 \cdot 10^3$. This indicates that Pt is effectively bioaccumulated by aquatic organisms, as already observed in previous exposure studies (e.g. Zimmermann et al. 2005; Sures and Zimmermann 2007); however, bioaccumulation factors derived by the latter yielded systematically lower values. For example, Sures and Zimmermann (2007) reported BF's ranging from 40 to 100 for the freshwater zebra mussel (*Dreysena polymorpha*) exposed to dissolved Pt added in the form of PtCl₄; similarly, Veltz et al. (1996) obtained a BF of 96 for the worm *Lumbriculus variegatus* exposed to PtCl₆²⁻. For seawater organisms, Mulholland and Turner (2011) derived a BF of 300 for the macroalgae *Ulva lactuca* and 20-30 for the marine gastropod *Littorina littorea* exposed to Pt in the form of PtCl₆²⁻/PtCl₅.

The higher accumulation factor obtained in this study compared to exposure experiments may be explained by (i) the low incubation periods – generally from few days to one month – used in exposure studies which may underestimate the bioaccumulation due to non-equilibrium conditions (Veltz et al. 1996), and (ii) the uptake of suspended particles containing elevated Pt concentrations in real ambient conditions; accordingly, in coastal areas subject to urban pressure, whereas dissolved Pt may be similar or only slightly higher than uncontaminated waters, Pt concentrations in suspended matter may greatly

exceed typical background values due to the input of strongly-bound Pt-rich particles from catalytic converters (Cobelo-García et al. 2013).

Compared to other trace metals, the BF obtained in this study for Pt ($\sim 5 \cdot 10^3$) is significantly lower. For example, using the metal concentrations in the same mussels samples from the Vigo Ria reported earlier (Besada et al. 2014) and the typical dissolved metal concentrations in this basin (Santos-Echeandía et al. 2009), BFs around 1-2 orders of magnitude higher are obtained for Zn (BF $\sim 5 \cdot 10^5$), Cu (BF $\sim 2 \cdot 10^4$), Pb (BF $\sim 9 \cdot 10^4$) or Cd (BF $\sim 1 \cdot 10^5$).

3. Conclusions

The present study reports, for the first time, the temporal variation of Pt concentrations in aquatic organisms and shows evidence of the link of the observed increased levels in the recent years with the use of this metal in anthropogenic activities (i.e. catalytic converters in motor vehicles). This is especially relevant since the potential health risk from exposures to Pt is greater than once thought (Wiseman and Zereini, 2009) and, therefore, increasing Pt concentrations may lead in the future to toxic effects on living organisms. These results point the need for more field studies on the accumulation of Pt in organisms subject to traffic-borne contamination, as well as including other elements also used in automobile catalytic converters, i.e., Rh and, especially, Pd since it has shown a higher degree of mobilization from particles emitted by catalytic converters than Pt or Rh (Dahlheimer et al. 2007).

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Table 1. Summary of Pt concentrations reported in the literature for aquatic organisms

Species	Pt (ng/g)	Location	Technique	Reference
<i>Freshwater species</i>				
Fish liver	0.23-0.33	German river	?	Hees et al, 1998
Freshwater crustacean (<i>Asellus aquaticus</i>)	330-1100	Swedish urban river (Kvillebäcken)	ACSV	Rauch and Morrison, 1999
	5-119 (38±38)	Swedish river close to car traffic (Mölndal)	ICP-MS	Moldovan et al., 2001
	<0.36-0.54	Aquatic habitats close to car traffic in the Ruhr district (Germany)	ICP-MS	Haus et al., 2007
Freshwater crustaceans (<i>Gammarus pulex</i> and <i>Dikerogammarus villosus</i>)	<0.36-1.3	Aquatic habitats close to car traffic in the Ruhr district (Germany)	ICP-MS	Haus et al., 2007
Kidney of European eel (<i>Anguilla anguilla</i>)	<0.06	River Weser at Petershagen (Germany)	SF-ICP-MS	Sures et al. 2003
<i>Seawater species</i>				
Seaweed (various species)	0.25-1.75 (0.74±0.40)	California coast (USA)	AAS	Yang, 1989
	0.08-032	California coast (USA)	AAS	Hodge et al. 1986
Oysters (<i>Crassostea gigas</i>)	0.16-0.53 (0.35±0.08)	Gironde Estuary (France)	ACSV	Abdou 2014
Mussels (<i>Mytilus galloprovincialis</i>)	0.30-0.68 (0.47±0.10)	Vigo Ria (Spain)	ACSV	This work

Figure 1. Sampling sites for mussels at Samil Beach and Oia (control point, insert)

Figure 2. Pt (ng g⁻¹) and Pb (µg g⁻¹; Besada et al. 2014) concentrations in mussels at Samil beach (see Figure 1).

Figure 3. Excess of Pt in mussels from Samil beach respect to the background levels and compared to car sales in Spain (data from www.anfac.com) and European autocatalyst Pt demand (data from www.platinum.matthey.com).