



# Trophodynamics of inorganic pollutants in a wide-range feeder: The relevance of dietary inputs and biomagnification in the Yellow-legged gull (*Larus michahellis*)

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## ABSTRACT

The suitability of sentinel species to monitor environmental pollution is often hampered by an insufficient knowledge on pollutant trophodynamics. We simultaneously evaluated the influence of individuals' trophic position (as revealed by  $\delta^{15}\text{N}$  values) and dietary exploitation of particular systems (using  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  as proxies) on inorganic pollutant concentrations measured on fledglings' feathers of a wide-range feeder, the Yellow-legged Gull (*Larus michahellis*), sampled at four locations throughout the Western Mediterranean. Concentrations of total Hg and Se in fledgling feathers ( $2.43 \pm 1.30$  and  $1.16 \pm 0.43$   $\mu\text{g/g}$ , respectively) were under the threshold points for deleterious effects on seabirds. On the contrary, alarming Pb concentrations were found in one colony (mean:  $1.57 \pm 2.46$   $\mu\text{g/g}$ , range: 0.16–12.13). With the exception of Pb, pollutant concentrations were positively influenced by consumption of marine resources (as suggested by the positive relationship with  $\delta^{34}\text{S}$  values), whereas trophic position played a minor role in determining pollutant body burdens.

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## 1. Introduction

Anthropogenic activities during the last few decades have resulted in contamination of a wide range of environmental media (Halpern et al., 2008; Mason and Sheu, 2002; Peterson et al., 2003; Votier et al., 2005). Aquatic organisms, and those exploiting aquatic resources, are particularly exposed to increasing levels of pollutants as aquatic systems are usually the ultimate pollutant sink, either due to diffuse sources, or direct discharges from the terrestrial and atmospheric compartments. In addition, the increasing fish consumption by most human populations (FAO, 2005) raises the need of understanding the trophodynamics of these contaminants, particularly in those marine and freshwater foodwebs (Dewailly and Knap, 2006; Risher et al., 2002; Simmonds et al., 2002; Tchounwou et al., 2003). In this sense, top predators are often suggested as ideal biomonitoring candidates as they integrate pollutant levels along food chains (Becker, 2003; Burger and Gochfeld, 2004; Cabana and Rasmussen, 1994; McIntyre and Beauchamp, 2007). Since contaminant emissions are not spatially uniform, top predators also become helpful integrators of the

spatial distribution of pollutants, as many species have vast breeding ranges, are able to move freely at large scales and some of them even exploit resources ascribed to different ecosystems (Braune et al., 2005; Christensen et al., 2005; Hobson et al., 2004; Ricca et al., 2008; Roscales et al., 2010). However, there is a marked variability in these factors affecting the dynamics of contaminants throughout foodweb (i.e. pollutant trophodynamics), which may potentially lead these apical species to a different pollutant exposure; and this knowledge becomes critical for effective environmental health monitoring and pollutant body burden assessments (Becker et al., 2002; McIntyre and Beauchamp, 2007; Verreault et al., 2009).

Since Cabana and Rasmussen (1994) questions related to bio-magnification of contaminants to deleterious levels in biota, have largely been addressed by considering species' trophic positions (Becker et al., 2002; González-Solís et al., 2002; McIntyre and Beauchamp, 2007; Morel et al., 1998; Stewart et al., 2004). However, direct trophic transfer of pollutants derived from local feeding opportunities and individual feeding behaviours is also considered an important pathway for contaminant acquisition (Bearhop et al., 2000; Bergeron et al., 2007; Loseto et al., 2008; Ramos et al., 2009a), adding complexity to the study of pollutant trophodynamics. Thus, although it is generally accepted that both

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individual trophic level and differential exploitation of particular trophic chains condition the pollutant exposure of species, most of previous contaminant studies partially neglected some of these effects due to the difficulty in distinguishing and accurately quantifying them (e.g. Bearhop et al., 2000; Bond and Diamond, 2009; González-Solís et al., 2002; Ricca et al., 2008). Additionally, precise and individual dietary information is often cornerstones which has also hampered our appropriate understanding of the influence of several trophic aspects on pollutant body burdens in first term, but also has ultimately hampered our general ability to assess and comprehend pollutant dynamics in natural systems.

Naturally occurring stable isotopes of carbon ( $^{13}\text{C}/^{12}\text{C}$ ,  $\delta^{13}\text{C}$ ), nitrogen ( $^{15}\text{N}/^{14}\text{N}$ ,  $\delta^{15}\text{N}$ ) and sulphur ( $^{34}\text{S}/^{32}\text{S}$ ,  $\delta^{34}\text{S}$ ) have become powerful tools in ecology to tease apart the effect of exploitation of particular food resources from the effects of the trophic position on animals' pollutant body burden. On one hand,  $\delta^{15}\text{N}$  values can be used as a reliable proxy of trophic position, since consumers' tissues are typically and predictably enriched in  $^{15}\text{N}$  relative to their diet (Caut et al., 2009; Post, 2002). On the other hand, those signatures of  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  are commonly used to discriminate among dietary exploitation of disparate ecosystems (such as marine vs. continental environments (Bearhop et al., 2003; Connolly et al., 2004)). Therefore, through the isotopic composition of appropriate tissues, one might be able to disentangle between individuals' trophic positions and feeding habits for an accurate understanding of pollutant pathways throughout foodwebs.

Here, we propose the use of  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  signatures as a tool to evaluate the effect of individual dietary preferences on the exposure of organisms to several inorganic pollutants. In particular, we evaluated the influence of individuals' trophic position (using  $\delta^{15}\text{N}$ ) and foraging habitat exploitation (using of  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$ ) on the concentrations of total mercury (THg), selenium (Se) and lead (Pb) in fledgling feathers of Yellow-legged gull (*Larus michahellis*). The Yellow-legged gull was selected as a suitable model species as (1) it is widely considered a top predator, (2) it is abundant and easily accessible, and (3) its diet is highly diverse, feeding on disparate trophic chains and environments (Ramos et al., 2011). Due to the expected differences in pollutant availabilities among foraging habitats of this gull (particularly between marine and terrestrial environments; Clark, 1992; Dewailly and Knap, 2006; Fitzgerald et al., 2007), and the biomagnification processes affecting pollutant dynamics throughout foodwebs (Cabana and Rasmussen, 1994; McIntyre and Beauchamp, 2007; Stewart et al., 2004), we expected those individuals preferentially exploiting marine food resources and occupying higher trophic positions to show the highest pollutant concentrations. Finally, this study not only brought clue insights into the trophodynamics of inorganic pollutants, but might also be relevant for studies of human food risk assessment as most of the trophic resources of these gulls are also present in human's diet (e.g. commercial/marketable fish, meat wastes).

## 2. Material and methods

### 2.1. Study area and sampling strategy

During the late chick-rearing period of the 2004 breeding season, Yellow-legged gull fledglings were sampled in four colonies along the Eastern Iberian coast (Fig. 1): the Medes Islands ( $n = 20$ ), the Ebro Delta ( $n = 23$ ), the Columbretes Islands ( $n = 40$ ), and Mazarrón Island ( $n = 14$ ). To avoid pseudoreplications coming from siblings being fed with the same prey, a single fledgling from each brood was captured, marked and sampled. We collected 6–8 growing scapular feathers from each bird as well as some food samples spontaneously regurgitated which were individually placed in sealed plastic bags and stored at  $-20\text{ }^{\circ}\text{C}$  until laboratory analyses. To our interest, chick feathers grow slowly and constantly throughout the chick-rearing period (personal observation) and they are formed from the dietary inputs received at the colony site (Becker et al., 1994). Although feather elements are routed from blood and liver before being excreted into growing feathers, with the sampling

of chick feathers we avoided bioaccumulation biases and severe biogeochemical routing from other internal organs (such as muscle or fat) which might cause great impact in biogeochemical profiles due to potential seasonal movements and dietary shifts of the adult individuals (Bearhop et al., 2000; Sanpera et al., 2007). Chicks and fledglings diet of these colonies of Yellow-legged gull is described in detail in Ramos et al. (2009b) through conventional analysis of regurgitates.

### 2.2. Sample preparation and laboratory analyses

At the laboratory, single food items from fledglings' regurgitates were identified and then assigned to three categories according its origin: a) marine prey, b) continental invertebrates, and c) refuse dumps. Marine prey was mainly composed by bogues (*Boops boops*) and several *Clupeiformes* fish, continental items included several families of invertebrates (larvae of *Muscidae* and *Syrphidae* as well as adults of *Lumbricidae*, *Forficula auricularia* and *Tenebrio molitor*) and refuse samples were mainly composed by meat wastes coming from refuse dumps. The best preserved food samples of the main prey items consumed in each locality were selected and prepared for stable isotope and pollutant determinations. Food samples were freeze dried, ground to powder and half of each sample was lipid extracted with several rinses of chloroform-methanol (2:1) solution (Bligh and Dyer, 1959). Lipid extracted dietary samples were analysed for stable isotopes, while non-extracted samples were analysed for pollutant concentrations. Feathers were washed in a 0.25 M sodium hydroxide solution, rinsed thoroughly in distilled water to remove any surface contamination (Valladares et al., 2010), dried in an oven at  $60\text{ }^{\circ}\text{C}$  to constant mass, and ground to a fine powder in a freezer mill (Spex Certiprep 6750; Spex Inc., Metuchen, New Jersey, USA) operating at liquid nitrogen temperature.

Isotope analyses were carried out at the Serveis Científic-Tècnics of the University of Barcelona (SCT-UB; Spain) by means of elemental analysis-isotope ratio mass spectrometry using a Thermo Finnigan Flash 1112 (CE Elantech, Lakewood, NJ, USA) elemental analyzer coupled to a Delta-C isotope ratio mass spectrometer via a CONFLU interface (Thermo Finnigan MAT, Bremen, Germany). Stable isotope ratios were expressed in the standard  $\delta$ -notation (‰) relative to Vienna Pee Dee Belemnite (VPDB;  $\delta^{13}\text{C}$ ), atmospheric  $\text{N}_2$  (VAIR;  $\delta^{15}\text{N}$ ) and Vienna Cañon Diablo Troilite (VCDT;  $\delta^{34}\text{S}$ ). Isotopic ratio mass spectrometry facility at the SCT-UB applies international inorganic standards (IAEA CH7, IAEA CH6 and USGS 24 for C, IAEA N1, IAEA N2 and IAEA NO3 for N and IAEA-S1, IAEA-S2, IAEA-S3, NBS-127 and YCEM for S) as well as internal organic standards (Human Hair CRM 397, Community Bureau of Reference, Commission of the European Community;  $\delta^{13}\text{C}$ :  $-19.46 \pm 0.15$ ,  $\delta^{15}\text{N}$ :  $10.01 \pm 0.20$ ,  $\delta^{34}\text{S}$ :  $4.48 \pm 0.17$ ) inserted every 12 samples to calibrate the system and compensate for any drift over time. Replicate assays of standard materials indicated measurement errors of  $\pm 0.1$ ,  $\pm 0.2$  and  $\pm 0.2$ , for carbon, nitrogen and sulphur respectively but these are likely underestimates of true measurement error for complex organics like feathers.

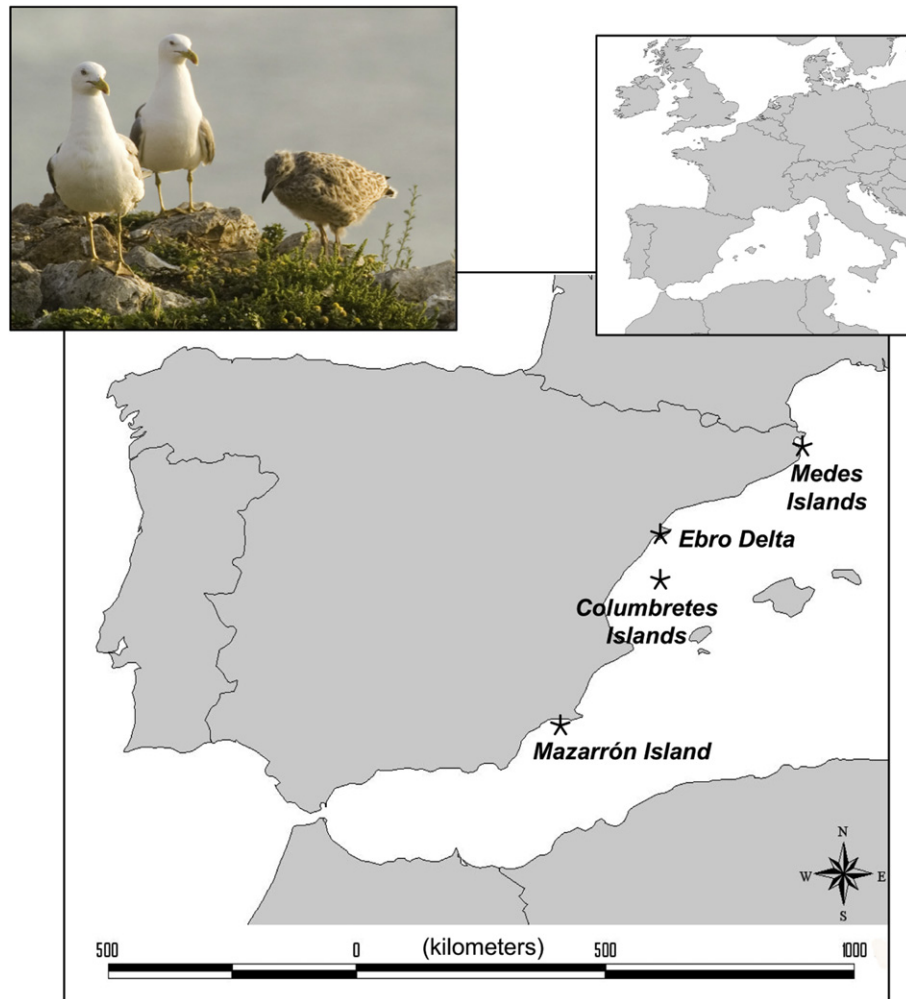
Trace element concentrations were also determined at SCT-UB by means of Induction Coupled Plasma–Mass Spectrometer (ICP–MS, Perkin Elmer Optima 6000, Connecticut, USA). The accuracy of the analysis was also checked by measuring certified Human Hair standard (CRM 397). Mean recoveries on reference tissues account for 102% for THg, 101% for Se and 98.5% for Pb, and no corrections were applied to the original results. ICP–MS LOD values were 0.2 ng/g for THg, 1 ng/g for Se and 0.1 ng/g for Pb which correspond approximately to 0.17  $\mu\text{g/g}$  dw (THg), 0.83  $\mu\text{g/g}$  dw (Se) and 0.08  $\mu\text{g/g}$  dw (Pb) in the tissue samples analysed.

### 2.3. Statistical analysis

Normality checks for distributions of THg, Se and Pb concentrations (partitioned by colony), were made using Q–Q plots. THg, Se and Pb concentration values were log transformed to symmetrise their distributions. Linear mixed models (LMMs) were used to fit log-transformed concentrations of pollutants in fledgling feathers. A set of competing models was built by considering the three stable isotopic signatures observed in fledglings' feathers ( $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  as proxies of foraging habitat exploitation, and  $\delta^{15}\text{N}$  as a proxy of individual trophic position) and all potential double interactions between the three signatures. To account for the potential spatial heterogeneity in pollutant availabilities the colony site was also included in the LMMs as a random term. Model selection was done using the Akaike information criteria ( $\text{AIC}_c$ ) and the corresponding  $\text{AIC}_c$  weights (Johnson and Omland, 2004). LMMs were conducted in R version 2.8.1 (R Development Core Team, 2010) with additional functions provided by the R packages lmer4 (lmer; Bates et al., 2008) and MuMIn (dredge; Barton, 2009).

## 3. Results

First of all, we did not find significant differences in isotopic signatures among localities for none of the food categories (one-way ANOVA with Welch's correction, all  $P > 0.1$ ), although in some cases this testing was hampered either by the absence of the resource or its low abundance in a given locality (see sample sizes in Table 1). We then represented the overall mean and the 95%



**Fig. 1.** Map of the Iberian Peninsula (South West Europe) indicating the Western Mediterranean breeding colonies of the Yellow-legged gull (*Larus michahellis*) included in the study. Photo shows a couple of Yellow-legged gulls with one of their chicks begging for food (photo credit R. Ramos).

confidence intervals for each food source without considering the locality effect in diet signatures (see Fig. 2). Values of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  for refuses were significantly lower than those for both marine and continental prey ( $F_{\text{WELCH } 2,27} = 24.26$ ,  $P < 0.001$ , and  $F_{\text{WELCH } 2,20} = 30.31$ ,  $P < 0.001$ , respectively; post hoc Tamhane's multiple comparison test,  $P < 0.01$ ), whereas  $\delta^{34}\text{S}$  signatures of refuses and continental invertebrates were much lower than those of marine prey invertebrates ( $F_{\text{WELCH } 2,23} = 218.51$ ,  $P < 0.001$ ; post hoc Tamhane's multiple comparison test,  $P < 0.001$ ; Fig. 2).

Values of  $\delta^{13}\text{C}$  in feathers showed significant differences between localities ( $F_{\text{WELCH } 3,23} = 69.06$ ,  $P < 0.001$ ; Fig. 2a). Post hoc comparisons showed that Columbretes chicks had the highest  $\delta^{13}\text{C}$  values ( $-17.5 \pm 0.2$ ). Ebro Delta and Medes Islands chicks showed intermediate values ( $-18.2 \pm 0.3$  and  $-18.4 \pm 0.5$ , respectively), which are not significantly different, whereas Mazarrón chicks ( $-18.8 \pm 0.6$ ) showed the lowest values, which were different from those of the Ebro Delta but not different from those of the Medes. Feathers also exhibited significant differences between localities in  $\delta^{15}\text{N}$  ( $F_{\text{WELCH } 3,23} = 44.08$ ,  $P < 0.001$ ; Fig. 2). Post hoc comparisons showed that values from Columbretes ( $+11.1 \pm 0.3$ ), Ebro Delta ( $+11.4 \pm 0.5$ ) and Mazarrón ( $+11.6 \pm 1.1$ ) were similar, whereas values from the Medes site ( $+9.9 \pm 0.5$ ) were significantly lower. The  $\delta^{34}\text{S}$  values of feathers differed among the four localities ( $F_{\text{WELCH } 3,23} = 222.46$ ,  $P < 0.001$ ; Fig. 2b, all post hoc Tamhane's multiple comparison test with  $P < 0.01$ ) in the same direction

apparent in  $\delta^{13}\text{C}$  values, although with a higher discrimination power. Feathers from Columbretes ( $+18.9 \pm 0.5$ ) exhibited higher  $\delta^{34}\text{S}$  values than those of Mazarrón ( $+11.6 \pm 1.5$ ). The Ebro Delta and Medes sites also had intermediate values ( $+16.1 \pm 1.5$ ; and  $+13.7 \pm 1.8$ , respectively).

Using model information criteria, we evaluated those competent models to assess pollutant dynamics (Table 2; Fig. 3). Regarding concentrations of THg and Se in fledgling feathers, and after controlling for locality, the best-supported models included, in both cases,  $\delta^{34}\text{S}$  signatures as the main explanatory variable (explaining these models up to 24.5 and 15.1% of the original variance, respectively). Concentrations of THg and Se were both positively related to  $\delta^{34}\text{S}$  values (Fig. 4). The selected model to fit Pb values showed this variable to be related to none of the isotopic values (Table 2). However, the random colony component accounted for a considerable amount of the total variability in Pb concentrations (Table 3). In fact, the impact of baseline levels on fledgling burdens (i.e. spatial heterogeneity), was only remarkable in Pb concentrations, which was probably due to the high values of this heavy metal in fledglings from the polluted Mazarrón area (Fig. 3c). In addition, residual variance, i.e. variability not explained by the effects included in fitted models (assumed to mainly derived from intrinsic factors), was particularly high when modelling Pb concentrations, but considerably low in both THg and Se modelling (Table 3).

**Table 1**  
Descriptive statistics (mean  $\pm$  SD (min, max)) of stable isotopes signatures ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  in ‰) and pollutant concentrations (THg, Se and Pb in  $\mu\text{g/g dw}$ ) in feathers from fledglings of Yellow-legged gull and in its main prey types. Values are split by colony when available. ND: non-detectable values.

	n	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	$\delta^{34}\text{S}$ (‰)	n	THg ( $\mu\text{g/g dw}$ )	Se ( $\mu\text{g/g dw}$ )	Pb ( $\mu\text{g/g dw}$ )
<b>Fledgling feathers</b>								
Columbrete Is.	40	-17.5 $\pm$ 0.2 (-18.0, -17.1)	+11.1 $\pm$ 0.3 (+10.6, +11.6)	+18.9 $\pm$ 0.4 (+17.9, +19.7)	40	3.6 $\pm$ 1.1 (1.8;6.3)	1.3 $\pm$ 0.3 (ND;1.7)	0.8 $\pm$ 1.5 (0.2;7.7)
Ebro Delta	23	-18.2 $\pm$ 0.3 (-18.9, -17.7)	+11.4 $\pm$ 0.5 (+10.3, +12.1)	+16.1 $\pm$ 1.5 (+12.9, +18.3)	23	1.9 $\pm$ 0.5 (1.0;2.8)	1.4 $\pm$ 0.3 (0.9;2.2)	0.5 $\pm$ 0.3 (0.2;1.4)
Medes Is.	20	-18.4 $\pm$ 0.5 (-19.3, -17.5)	+9.9 $\pm$ 0.5 (+9.1, +11.1)	+13.7 $\pm$ 1.8 (+10.6, +17.1)	20	1.8 $\pm$ 0.9 (0.6;3.7)	1.1 $\pm$ 0.4 (ND;1.4)	0.6 $\pm$ 0.3 (0.3;1.6)
Mazarrón Is.	14	-18.8 $\pm$ 0.6 (-20.2, -17.9)	+11.6 $\pm$ 1.1 (+9.3, +13.6)	+11.6 $\pm$ 1.5 (+8.1, +13.9)	14	1.3 $\pm$ 0.7 (0.5;3.5)	0.7 $\pm$ 0.5 (ND;1.2)	5.4 $\pm$ 3.1 (1.4;12.1)
<b>Marine diet</b>								
Columbrete Is.	11	-18.8 $\pm$ 0.3 (-19.1, -18.3)	+9.4 $\pm$ 0.5 (+8.4, +10.15)	+17.6 $\pm$ 0.4 (+16.9, +18.2)	3	0.2 $\pm$ 0.1 (ND;0.2)	3.2 $\pm$ 0.0 (3.2;3.3)	3.8 $\pm$ 3.0 (1.8;7.3)
Ebro Delta	9	-16.4 $\pm$ 2.7 (-19.1, -14.2)	+9.9 $\pm$ 1.6 (+8.0, +12.4)	+16.4 $\pm$ 1.5 (+13.9, +18.1)				
Medes Is.	9	-18.4 $\pm$ 0.4 (-18.8, -17.4)	+9.4 $\pm$ 0.9 (+8.6, +11.5)	+17.2 $\pm$ 0.5 (+16.7, +18.3)	6	0.3 $\pm$ 0.1 (ND;0.4)	2.7 $\pm$ 0.5 (1.9;3.2)	1.7 $\pm$ 1.1 (0.4;3.3)
Mazarrón Is.	2	-18.8 $\pm$ 0.4 (-19.0, -18.5)	+9.6 $\pm$ 0.3 (+9.3, +9.8)	+14.6 $\pm$ 4.9 (+11.1, +18.1)	2	0.2 $\pm$ 0.2 (0.1;0.4)	1.9 $\pm$ 0.3 (1.7;2.1)	9.1 $\pm$ 3.9 (6.3;11.8)
<b>Continental invertebrates</b>								
Ebro Delta	6	-17.9 $\pm$ 1.8 (-21.3, -16.2)	+11.0 $\pm$ 3.2 (+7.6, +16.1)	+6.7 $\pm$ 0.7 (+5.7, +7.7)				
Medes Is.	3	-18.4 $\pm$ 2.8 (-21.1, -15.6)	+10.8 $\pm$ 5.3 (+5.2, +15.6)	+6.8 $\pm$ 0.6 (+6.1, +7.3)				
Mazarrón Is.	7	-20.5 $\pm$ 3.0 (-25.3, -18.4)	+14.1 $\pm$ 6.9 (+7.5, +27.4)	+8.6 $\pm$ 2.6 (+3.5, +11.0)	5	0.1 $\pm$ 0.0 (ND;0.1)	1.0 $\pm$ 0.3 (ND;1.4)	168.4 $\pm$ 124.2 (62.7;359.2)
<b>Refuse diet</b>								
Ebro Delta	2	-19.9 $\pm$ 0.1 (-19.9, -19.9)	+6.0 $\pm$ 1.6 (+4.9, +7.2)	+7.0 $\pm$ 1.0 (+6.3, +7.7)				
Medes Is.	5	-22.0 $\pm$ 1.6 (-23.3, -20.0)	+4.8 $\pm$ 1.3 (+3.4, +6.4)	+5.4 $\pm$ 1.8 (+3.8, +8.3)	4	0.1 $\pm$ 0.0 (ND;0.1)	0.7 $\pm$ 0.2 (ND;1.0)	6.3 $\pm$ 5.3 (1.2;11.3)
Mazarrón Is.	5	-22.0 $\pm$ 1.1 (-23.7, -20.9)	+6.0 $\pm$ 2.3 (+4.3, +9.6)	+7.9 $\pm$ 1.9 (+5.3, +10.5)	5	0.2 $\pm$ 0.1 (ND;0.3)	0.9 $\pm$ 0.4 (ND;1.3)	360.1 $\pm$ 298.2 (64.7;746.1)

## 4. Discussion

### 4.1. Isotopic landscape of Yellow-legged gull diet in the Western Mediterranean

As previous studies reported (Knoff et al., 2002),  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  levels were higher in marine prey, and consequently, also in those gull populations which largely rely on marine consumption (i.e. Columbretes Is.; Ramos et al., 2009b). However, only sulphur signatures differed enough among prey types and gull localities to be considered a useful tracer to assess the origin of the food consumed (Fig. 2b). In fact, carbon signatures of continental invertebrates and marine prey were rather similar, and therefore its values in fledgling feathers were rather homogeneous (Fig. 2a). Thus,  $\delta^{34}\text{S}$  had the greatest discrimination power that allowed us to distinguish between continental-terrestrial and marine-based diets even better than with  $\delta^{13}\text{C}$  signatures (see also Moreno et al., 2010).

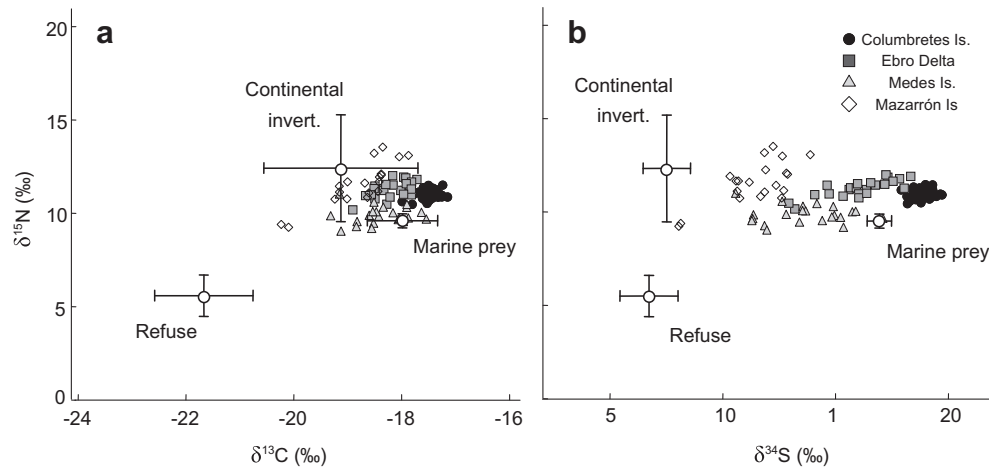
We also found that samples from refuse dumps provided the lowest values for  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$ , and consequently, those populations which consume refuses abundantly (i.e. Medes and Mazarrón Is.; Ramos et al., 2009b) showed the lowest values for these three isotopes (Table 1, Fig. 2). In particular, we expected those  $\delta^{15}\text{N}$  values from refuses to be the least enriched owing to a short food chain (Hebert et al., 1999). Accordingly,  $\delta^{15}\text{N}$  values of the Medes colony were significantly lower than those from other localities (Fig. 2), since almost half of its dietary biomass came from refuse dumps (Ramos et al., 2009b). Although Mazarrón chicks also received half of their dietary biomass from refuses, the relatively high  $\delta^{15}\text{N}$  values in that colony were explained by a moderate consumption of continental invertebrates (28.1% of chicks diet; Ramos et al., 2009b), which substantially increases  $\delta^{15}\text{N}$  values of consumers (Vanderklift and Ponsard, 2003). The fact that these invertebrates were mostly involved in foodwebs based on dead organic matter contributed to explaining their relatively high  $\delta^{15}\text{N}$  signatures (Ponsard and Ardit, 2000).

Thus, based on that isotopic landscape, we selected  $\delta^{34}\text{S}$  and  $\delta^{15}\text{N}$  signatures to evaluate, respectively, the potential impacts of feeding habitat and individual's trophic position in pollutant dynamics.

### 4.2. Monitoring environmental pollution

Oceans have been exposed to great anthropogenic pressure, and the Mediterranean basin in particular, has been largely considered one of the most polluted marine water masses around the world (Danovaro, 2003; Halpern et al., 2008). Although marine predators are expected to be exposed to increasing levels of such pollutants, there is a considerable lack of information on contaminant concentrations of these marine organisms which hampers our general ability to understand and interpret the dynamics of contaminants and the pollutant status of this ecosystem. Arcos et al. (2002) previously reported that THg concentrations in mantle feather of adult Yellow-legged gulls from the Ebro Delta ranged between 0.75 and 4.36  $\mu\text{g/g dw}$  (median = 1.49, mean = 2.10,  $n = 14$ ), which are very close to the results we found for this area (Table 1). However, concentrations of THg and Se that we found were generally lower to those previously reported for other strictly ichthyophagous seabirds (i.e. Audouin's gull *Larus audouinii* or Cory's shearwater *Calonectris diomedea*) throughout the Western Mediterranean (Arcos et al., 2002; Renzoni et al., 1986; Sanpera et al., 2007). Accordingly to our a priori expectations, this result could be easily due to the greater proportion of terrestrial items in the diet of Yellow-legged gull, which is known to generate, in general, a lower exposure to such contaminants (Lasorsa and Allen-Gil, 1995).





**Fig. 2.** Bivariate plots of  $\delta^{13}\text{C}$ – $\delta^{15}\text{N}$  (a) and  $\delta^{34}\text{S}$ – $\delta^{15}\text{N}$  (b) showing isotopic signatures of the three potential food prey (white circles; mean  $\pm$  95% CI and sampled size in brackets) and feathers of fledglings in relation to the breeding colony (Columbrete Is. in black circles, Ebro Delta in grey triangles, Medes Is. in greyish squares, and Mazarrón Is. in white diamonds).

Overall, concentrations of THg, Se and Pb that we found in fledgling feathers were under the threshold points for deleterious effects defined for several top predators and aquatic birds (5.0  $\mu\text{g/g}$  dw for Hg (Eisler, 1987; Furness et al., 1989); 3.8  $\mu\text{g/g}$  dw for Se (Eisler, 1985; Heinz, 1996); and 4.0  $\mu\text{g/g}$  dw for Pb (Burger and Gochfeld, 2000); Fig. 3). However, alarming Pb concentrations both in fledglings' feathers and in their diet were found in the Mazarrón area, which went largely above the adverse limits (Table 1; Fig. 3c). The presence of an abandoned mining site in the area (closed in 1991), where lead, zinc, copper, iron and silver had been intensively extracted for more than 2500 years (Gómez-Ramírez et al., 2011), seemed clue in explaining these high values. About 60 millions tones of polluted sediments and soils, containing high concentrations of heavy metals, are accumulated with scarce monitoring around this area of the South Eastern Iberia as a result of that ancient miner activity (Cesar et al., 2004; Fernández et al., 2010). The high Pb concentrations found in Mazarrón area, not only in gull feathers, but also in several trophic chains from that locality (Table 1), and in particular in those resources related with human intake (such as Clupeiform fish and refuses derived from poultry and beef scraps) highlighted up to the human health concern the necessity for an appropriate monitoring of the area by the competent authorities. Finally, these results support the suitability of feathers from resident top predators to biomonitor environmental pollution at local and regional scales. Even though gulls exploited very different trophic chains, the reported contaminant concentrations on Yellow-legged gull chicks reflected well the pollutant availability in each sampled locality, which ultimately derives from inherent soil/water composition as well as from previous contamination episodes.

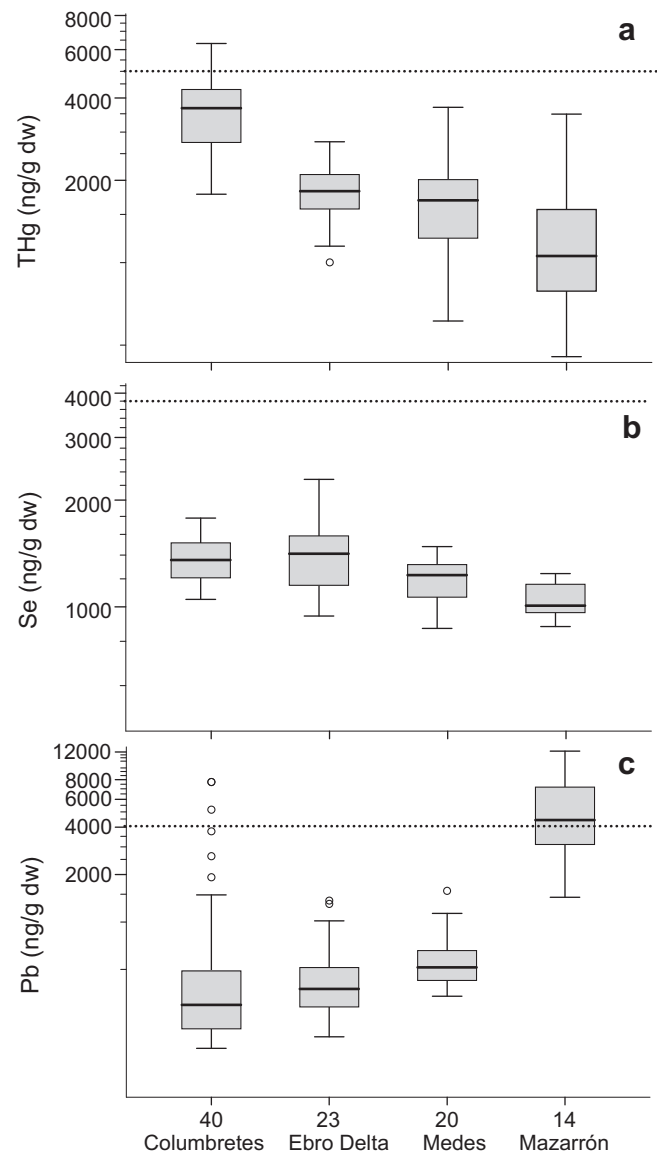
#### 4.3. Unravelling the trophodynamics of pollutants

In addition to the reported geographic variability in pollutant availability, there are many other important sources of variability that condition pollutant exposure both at inter-specific and intra-specific levels. In previous papers, several authors have speculated on the causes underlying the high intra-specific variation in contaminant concentrations in many marine top predators (McIntyre and Beauchamp, 2007; Muir et al., 2003; Takahashi et al., 2000; Verreault et al., 2009; Wan et al., 2007). In this regard and in addition to intrinsic factors, such as detoxification capability, age, sex, body size, genetics, reproductive status and nutritional condition, those factors related with diet and trophic aspects are proved to be the most relevant in affecting pollutant burdens of the individuals (Becker et al., 2002; McIntyre and Beauchamp, 2007; Verreault et al., 2009). In particular, several studies have previously reported on the influence of particular dietary contributions on THg and Se concentrations (Bergeron et al., 2007; González-Solís et al., 2002; Loseto et al., 2008; Ramos et al., 2009a). In addition, many other studies have already reported on biomagnification of these elements throughout foodwebs, considering  $\delta^{15}\text{N}$  as a measure of trophic positions (Atwell et al., 1998; Becker et al., 2002; Cabana and Rasmussen, 1994; González-Solís et al., 2002; Jarman et al., 1996; McIntyre and Beauchamp, 2007; Morel et al., 1998). Some of these studies even found this positive pollutant– $\delta^{15}\text{N}$  relationship at intra-specific level (Ramos et al., 2009a). However, to our knowledge, none of them were able to evaluate or distinguish between exploited foraging habitats and individual trophic positions, ultimately hampering their ability to understand pollutant dynamics throughout foodwebs. In this sense, our study provided quantitative evidence of the greater influence of marine

**Table 2**

Candidate models evaluated to fit the data corresponding to log-transformed THg, Se and Pb concentrations measured in chick feathers from four colonies and their associated measures of information (AIC<sub>c</sub> – corrected AIC;  $\Delta\text{AIC}_c$  – AIC<sub>c</sub> increments and AIC<sub>c</sub> Wgt – AIC<sub>c</sub> weights). The most complete model included  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  signatures, as well as their potential interaction. All evaluated models included the colony as a random effect. Preferred models are highlighted in bold.

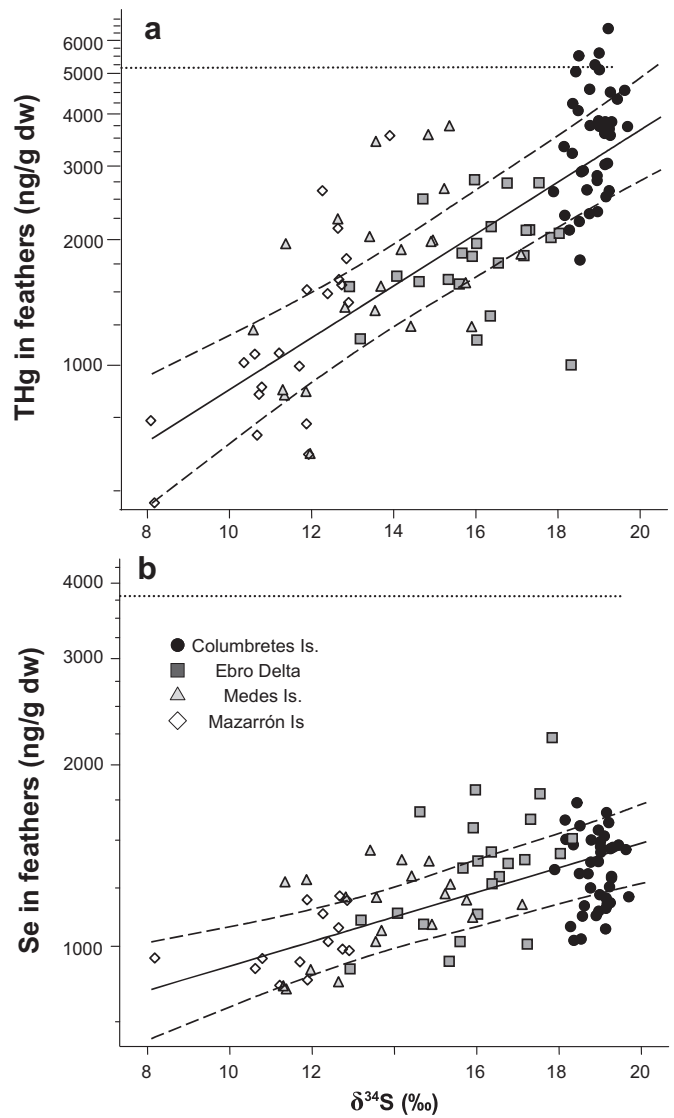
Model	k	log THg			log Se			log Pb		
		AIC <sub>c</sub>	$\Delta\text{AIC}_c$	AIC <sub>c</sub> Wgt	AIC <sub>c</sub>	$\Delta\text{AIC}_c$	AIC <sub>c</sub> Wgt	AIC <sub>c</sub>	$\Delta\text{AIC}_c$	AIC <sub>c</sub> Wgt
Constant	3	–57.1	25.35	0.000	–233.9	7.15	0.025	<b>80.9</b>	<b>0.00</b>	<b>0.923</b>
$\delta^{15}\text{N}$	4	–64.1	18.37	0.000	–235.1	5.92	0.046	87.0	6.05	0.045
$\delta^{34}\text{S}$	4	<b>–82.4</b>	<b>0.00</b>	<b>0.941</b>	<b>–241.0</b>	<b>0.00</b>	<b>0.885</b>	87.8	6.85	0.030
$\delta^{15}\text{N} + \delta^{34}\text{S}$	5	–76.9	5.55	0.059	–235.0	6.02	0.044	93.1	12.19	0.002
$\delta^{15}\text{N} + \delta^{34}\text{S} + \delta^{15}\text{N}:\delta^{34}\text{S}$	6	–66.8	15.66	0.000	–224.8	16.26	0.000	100.0	19.12	0.000



**Fig. 3.** Log-scale box-plots of (a) THg, (b) Se and (c) Pb concentrations in mantle feathers of Yellow-legged gull fledglings (in ng/g dw) are shown in relation to the breeding colony (sample size is 40, 23, 20 and 14 for the Columbretes, Ebro Delta, Medes and Mazarrón colonies, respectively). Horizontal dotted lines represent the approximate level for potential adverse effects of each pollutant estimated by Burger and Gochfeld (2000), Eisler (1987) and Heinz (1996) in seabird feathers.

prey contribution, as revealed by  $\delta^{34}\text{S}$  values (Fig. 2b), in explaining feather THg and Se concentrations compared to the low variability explained by the trophic status of the organism (as indicated by  $\delta^{15}\text{N}$  values) is particularly novel in the context of marine pollution sciences.

Although our study was based in a single predator (i.e. at intra-specific level), the high range in the  $\delta^{15}\text{N}$  levels (ranging from 9.09 to 13.56; Table 1) suggested that Hg bioaccumulation might have also occurred within this species. However, once the variability in diet origin was accounted for, we found no effect of the proxy of trophic position on any pollutant burden. This may suggest that those previous biomagnification relationships found at intra-specific level could be, at least in part, a subconsequence of individuals foraging on habitats with different pollutant availabilities. Additionally, those biomagnifications found along foodwebs could be also occurring at lower rates than previously thought, as they



**Fig. 4.** Relationships of THg (a) and Se (b) concentrations (in log-scale) in fledgling feathers with their  $\delta^{34}\text{S}$  values (mean and 95% confidence intervals in solid and dashed lines, respectively). Data from each colony is represented differently: Columbretes Is. in black circles, Ebro Delta in grey triangles, Medes Is. in greyish squares, and Mazarrón Is. in white diamonds. Horizontal dotted lines represent the approximate level for potential adverse effects of THg and Se estimated by Burger and Gochfeld (2000), Eisler (1987) and Heinz (1996) in seabird feathers.

could have been overestimated when feeding preferences of the individuals were not considered. Although this dietary influence could be exaggerated in those species with diverse feeding spectrum, particularly for those feeding in both terrestrial and marine

**Table 3**  
Parameter estimates from linear mixed models fitted to log-transformed THg, Se and Pb concentrations in Yellow-legged gull fledgling's feathers from four Western Mediterranean colonies.

	log THg	log Se	log Pb
<i>Fixed effects (estimate ± SE)</i>			
Intercept	2.326 ± 0.119	2.771 ± 0.063	2.912 ± 0.254
$\delta^{34}\text{S}$	0.063 ± 0.008	0.020 ± 0.004	
<i>Random effect (variance ± SE)</i>			
Locality	0.002 ± 0.048	0.001 ± 0.025	0.254 ± 0.504
Residual	0.021 ± 0.146	0.005 ± 0.067	0.101 ± 0.317

biomes (Lasorsa and Allen-Gil, 1995; Mason and Sheu, 2002), considering individual foraging preferences when assessing pollutant burdens of the species seems advisable from now on. This last point highlighted the necessity to consider and disentangle among diverse trophic aspects, by using isotopic approaches for instance, to ascertain the role and dynamics of heavy metals and trace elements within the biosphere.

## 5. Conclusions

Understanding pollutant dynamics throughout coastal and estuarine foodwebs has become critical to wildlife management and environmental sciences, as a consequence of the increasing human activities on these environments. Within this framework, monitoring animal exposure to pollutants derived from local foodwebs is crucial to evaluate the actual impact of such pollution on wildlife communities and on the ecosystem as a whole. In this regard and as many previous studies had found, THg, Se and Pb levels in fledgling feathers varied geographically confirming that baseline levels of these elements at different colony sites easily influence tissue burdens, especially at high trophic levels (Bearhop et al., 2000; Gochfeld, 1997; Ramos et al., 2009a; Ricca et al., 2008; Thompson et al., 1992). However, more importantly, this study concluded that exploited feeding habitats may affect pollutant burden of a given individual, in a greater manner than its own trophic position does. In addition, quantifying the diet of individuals based on isotopic approaches seemed advisable since differing individual feeding strategies may appear within populations resulting in a differential exposure to pollutants. Finally, we also suggested that those isotopic studies investigating diets through diverse trophic chains should seriously consider to evaluate  $\delta^{34}\text{S}$  in addition to the omnipresent  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  signatures, as greater discrimination among resources might be provided. Overall, this work constitutes an instructive case study aimed at understanding pollutant acquisition in wildlife, and its conclusions might be crucial for effective biomonitoring of the environmental health.

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## References

- Arcos, J.M., Ruiz, X., Bearhop, S., Furness, R.W., 2002. Mercury levels in seabirds and their fish prey at the Ebro Delta (NW Mediterranean): the role of trawler discards as a source of contamination. *Marine Ecology Progress Series* 232, 281–290.
- Atwell, L., Hobson, K.A., Welch, H.E., 1998. Biomagnification and bioaccumulation of mercury in an arctic marine food web: insights from stable nitrogen isotope analysis. *Canadian Journal of Fisheries and Aquatic Sciences* 55, 1114–1121.
- Bartoń, K., 2009. MuMIn: R Functions for Model Selection and Model Averaging. R package version 2.12.2. <http://r-forge.r-project.org/projects/mumin/>.
- Bates, D., Maechler, M., Dai, B., 2008. lme4: Linear Mixed-effects Models Using Eigen and Variance Components. R package version 2.12.1. <http://lme4.r-forge.r-project.org/>.
- Bearhop, S., Furness, R.W., Hilton, G.M., Votier, S.C., 2003. A forensic approach to understanding diet and habitat use from stable isotope analysis of (avian) claw material. *Functional Ecology* 17, 270–275.
- Bearhop, S., Phillips, R.A., Thompson, D.R., Waldron, S., Furness, R.W., 2000. Variability in mercury concentrations of great skuas *Catharacta skua*: the influence of colony, diet and trophic status inferred from stable isotope signatures. *Marine Ecology Progress Series* 195, 261–268.
- Becker, P.H., 2003. Biomonitoring with birds. In: Markert, B.A., Breure, A.M., Zechmeister, H.G. (Eds.), *Bioindicators and Biomonitoring: Principles, Concepts and Applications*. Elsevier Ltd., Amsterdam, pp. 677–736.
- Becker, P., González-Solís, J., Behrends, B., Croxall, J.P., 2002. Feather mercury levels in seabirds at South Georgia: influence of trophic position, sex and age. *Marine Ecology Progress Series* 243, 261–269.
- Becker, P.H., Henning, D., Furness, R.W., 1994. Differences in mercury contamination and elimination during feather development in gull and tern broods. *Archives of Environmental Contamination and Toxicology* 27, 162–167.
- Bergeron, C.M., Husak, J.E., Unrine, J.M., Romanek, C.S., Hopkins, W.A., 2007. Influence of feeding ecology on blood mercury concentrations in four species of turtles. *Environmental Toxicology and Chemistry* 26, 1733–1741.
- Bligh, E.G., Dyer, W.J., 1959. A rapid method of total lipid extraction and purification. *Canadian Journal of Biochemistry and Physiology* 37, 911–917.
- Bond, A.L., Diamond, A.W., 2009. Mercury concentrations in seabird tissues from Machias Seal Island, New Brunswick, Canada. *Science of the Total Environment* 407, 4340–4347.
- Braune, B.M., Outridge, P.M., Fisk, A.T., Muir, D.C.G., Helm, P.A., Hobbs, K., Hoekstra, P.F., Kuzyk, Z.A., Kwan, M., Letcher, R.J., Lockhart, W.L., Norstrom, R.J., Stern, G.A., Stirling, I., 2005. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: an overview of spatial and temporal trends. *Science of the Total Environment* 351–352, 4–56.
- Burger, J., Gochfeld, M., 2000. Metal levels in feathers of 12 species of seabirds from midway atoll in the northern Pacific Ocean. *Science of the Total Environment* 257, 37–52.
- Burger, J., Gochfeld, M., 2004. Marine birds as sentinels of environmental pollution. *EcoHealth* 1, 263–274.
- Cabana, G., Rasmussen, J.B., 1994. Modelling food chain structure and contaminant bioaccumulation using stable nitrogen isotopes. *Nature* 372, 255–257.
- Caut, S., Angulo, E., Courchamp, F., 2009. Variation in discrimination factors ( $\Delta^{15}\text{N}$  and  $\Delta^{13}\text{C}$ ): the effect of diet isotopic values and applications for diet reconstruction. *Journal of Applied Ecology* 46, 443–453.
- Cesar, A., Marín, A., Marín-Guirao, L., Vita, R., 2004. Amphipod and sea urchin tests to assess the toxicity of Mediterranean sediments: the case of Portmán Bay. *Scientia Marina* 68, 205–213.
- Christensen, J.R., Macduffee, M., MacDonald, R.W., Whittar, M., Ross, P.S., 2005. Persistent organic pollutants in British Columbia grizzly bears: consequence of divergent diets. *Environmental Science & Technology* 39, 6952–6960.
- Clark, R.B., 1992. *Marine Pollution*. Clarendon Press, Oxford, UK.
- Connolly, R.M., Guest, M.A., Melville, A.J., Oakes, J.M., 2004. Sulfur stable isotopes separate producers in marine food-web analysis. *Oecologia* 138, 161–167.
- Danovaro, R., 2003. Pollution threats in the Mediterranean Sea: an overview. *Chemistry and Ecology* 19, 15–32.
- Dewailly, É., Knap, A., 2006. Food from the oceans and human health: balancing risks and benefits. *Oceanography* 19, 84–93.
- Eisler, R., 1985. *Selenium Hazards to Fish, Wildlife, and Invertebrates: a Synoptic Review*. US Fish and Wildlife Service, Biological.
- Eisler, R., 1987. *Mercury Hazards to Fish, Wildlife, and Invertebrates: a Synoptic Review*. US Fish and Wildlife Service, Biological.
- FAO, 2005. *Fishery statistical collections*. In: *FIGIS Data Collection Data and Statistics Unit*. Fishery Information. Food and Agricultural Organization, Rome, Italy.
- Fernández, B., Campillo, J.A., Martínez-Gómez, C., Benedicto, J., 2010. Antioxidant responses in gills of mussel (*Mytilus galloprovincialis*) as biomarkers of environmental stress along the Spanish Mediterranean coast. *Aquatic Toxicology* 99, 186–197.
- Fitzgerald, W.F., Lamborg, C.H., Hammerschmidt, C.R., 2007. Marine biogeochemical cycling of mercury. *Chemical Reviews* 107, 641–662.
- Furness, R.W., Johnston, J.L., Love, J.A., Thompson, D.R., 1989. Pollution burdens and reproductive success of golden eagles *Aquila chrysaetos* exploiting marine and terrestrial food webs in Scotland. In: Meyburg, B.U., Chancellor, R.D. (Eds.), *Raptors in the Modern World*. World Working Group on Birds of Prey, Berlin, Germany, pp. 495–500.
- Gochfeld, M., 1997. Spatial patterns in a bioindicator: heavy metal and selenium concentration in eggs of Herring gulls (*Larus argentatus*) in the New York bight. *Archives of Environmental Contamination and Toxicology* 33, 63–70.
- González-Solís, J., Sanpera, C., Ruiz, X., 2002. Metals and selenium as bioindicators of geographic and trophic segregation in giant petrels *Macronectes* spp. *Marine Ecology Progress Series* 244, 257–264.
- Gómez-Ramírez, P., Martínez-López, E., María-Mojica, P., León-Ortega, M., García-Fernández, A.J., 2011. Blood lead levels and d-ALAD inhibition in nestlings of Eurasian Eagle owl (*Bubo bubo*) to assess lead exposure associated to an abandoned mining area. *Ecotoxicology* 20, 131–138.
- Halpern, B.S., Walbridge, S., Selkoe, K.A., Kappel, C.V., Micheli, F., D'Agrosa, C., Bruno, J.F., Casey, K.S., Ebert, C., Fox, H.E., Fujita, R., Heinemann, D., Lenihan, H.S., Madin, E.M.P., Perry, M.T., Selig, E.R., Spalding, M., Steneck, R., Watson, R., 2008. A global map of human impact on marine ecosystems. *Science* 319, 948–952.
- Hebert, C.E., Shutt, J.L., Hobson, K.A., Weseloh, D.V.C., 1999. Spatial and temporal differences in the diet of Great Lakes herring gulls (*Larus argentatus*): evidence from stable isotope analysis. *Canadian Journal of Fisheries and Aquatic Sciences* 56, 323–338.

- Heinz, G.H., 1996. Selenium in birds. In: Beyer, W.N., Heinz, G.H., Redmond-Norwood, A.W. (Eds.), *Environmental Contaminants in Wildlife: Interpreting Tissues Concentrations*. Lewis Publishers, Boca Raton, Florida, pp. 447–458.
- Hobson, K.A., Riget, F.F., Outridge, P.M., Dietz, R., Born, E., 2004. Baleen as a bio-monitor of mercury content and dietary history of North Atlantic minke whales (*Balaenoptera acutorostrata*): combining elemental and stable isotope approaches. *Science of the Total Environment* 331, 69–82.
- Jarman, W.M., Hobson, K.A., Sydeman, W.J., Bacon, C.E., McLaren, E.B., 1996. Influence of trophic position and feeding location on contaminant levels in the Gulf of the Farallones food web revealed by stable isotope analysis. *Environmental Science & Technology* 30, 654–660.
- Johnson, J.B., Omland, K.S., 2004. Model selection in ecology and evolution. *Trends in Ecology & Evolution* 19, 101–108.
- Knoff, A.J., Macko, S.A., Erwin, R.M., Brown, K.M., 2002. Stable isotope analysis of temporal variation in the diets of pre-fledged laughing gulls. *Waterbirds* 25, 142–148.
- Lasorsa, B., Allen-Gil, S., 1995. The methylmercury to total mercury ratio in selected marine, freshwater, and terrestrial organisms. *Water, Air, and Soil Pollution* 80, 905–913.
- Loseto, L.L., Stern, G.A., Ferguson, S.H., 2008. Size and biomagnification: how habitat selection explains beluga mercury levels. *Environmental Science & Technology* 42, 3982–3988.
- Mason, R.P., Sheu, G.-R., 2002. Role of the ocean in the global mercury cycle. *Global Biogeochemical Cycles* 16, 40.
- McIntyre, J.K., Beauchamp, D.A., 2007. Age and trophic position dominate bio-accumulation of mercury and organochlorines in the food web of Lake Washington. *Science of the Total Environment* 372, 571–584.
- Morel, F.M.M., Kraepiel, A.M.L., Amyot, M., 1998. The chemical cycle and bio-accumulation of mercury. *Annual Review of Ecology and Systematics* 29, 543–566.
- Moreno, R., Jover, L., Munilla, I., Velando, A., Sanpera, C., 2010. A three-isotope approach to disentangling the diet of a generalist consumer: the yellow-legged gull in northwest Spain. *Marine Biology* 157, 545–553.
- Muir, D., Savinova, T., Savinov, V., Alexeeva, L., Potelov, V., Svetochev, V., 2003. Bio-accumulation of PCBs and chlorinated pesticides in seals, fishes and invertebrates from the White Sea, Russia. *Science of the Total Environment* 306, 111–131.
- Peterson, C.H., Rice, S.D., Short, J.W., Esler, D., Bodkin, J.L., Ballachey, B.E., Irons, D.B., 2003. Long-term ecosystem response to the Exxon Valdez oil spill. *Science* 302, 2082–2086.
- Ponsard, S., Ardit, R., 2000. What can stable isotopes ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) tell about the food web of soil macro-invertebrates? *Ecology* 81, 852–864.
- Post, D.M., 2002. Using stable isotopes to estimate trophic position: models, methods, and assumptions. *Ecology* 83, 703.
- R Development Core Team, 2010. R: a Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. <http://www.rproject.org>.
- Ramos, R., González-Solís, J., Forero, M.G., Moreno, R., Gómez-Díaz, E., Ruiz, X., Hobson, K.A., 2009a. The influence of breeding colony and sex on mercury, selenium and lead levels and carbon and nitrogen stable isotope signatures in summer and winter feathers of *Calonectris* shearwaters. *Oecologia* 159, 345–354.
- Ramos, R., Ramírez, F., Sanpera, C., Jover, L., Ruiz, X., 2009b. Diet of Yellow-legged gull (*Larus michahellis*) chicks along the Spanish Western Mediterranean coast: the relevance of refuse dumps. *Journal of Ornithology* 150, 265–272.
- Ramos, R., Ramírez, F., Carrasco, J.L., Jover, L., 2011. Insights into the spatiotemporal component of feeding ecology: an isotopic approach for conservation management sciences. *Diversity and Distributions* 17, 338–349.
- Renzi, A., Focardi, S., Fossi, C., Leonzio, C., Mayol, J., 1986. Comparison between concentrations of mercury and other contaminants in eggs and tissues of Cory's shearwater *Calonectris diomedea* collected on Atlantic and Mediterranean islands. *Environmental Pollution Series A* 40, 17–35.
- Ricca, M.A., Miles, K.A., Anthony, R.G., 2008. Sources of organochlorine contaminants and mercury in seabirds from the Aleutian archipelago of Alaska: inferences from spatial and trophic variation. *Science of the Total Environment* 406, 308–323.
- Risher, J.F., Murray, H.E., Prince, G.R., 2002. Organic mercury compounds: human exposure and its relevance to public health. *Toxicology and Industrial Health* 18, 109–160.
- Roscales, J.L., Muñoz-Arnan, J., González-Solís, J., Jiménez, B., 2010. Geographical PCB and DDT patterns in shearwaters (*Calonectris* sp.) breeding across the NE Atlantic and the Mediterranean archipelagos. *Environmental Science & Technology* 44, 2328–2334.
- Sanpera, C., Moreno, R., Ruiz, X., Jover, L., 2007. Audouin's gull chicks as bio-indicators of mercury pollution at different breeding locations in the western Mediterranean. *Marine Pollution Bulletin* 54, 691–696.
- Simmonds, M.P., Haraguchi, K., Endo, T., Cipriano, F., Palumbi, S.R., Troisi, G.M., 2002. Human health significance of organochlorine and mercury contaminants in Japanese whale meat. *Journal of Toxicology and Environmental Health, Part A* 65, 1211–1235.
- Stewart, A.R., Luoma, S.N., Schlegel, C.E., Doblin, M.A., Hieb, K.A., 2004. Food web pathway determines how selenium affects aquatic ecosystems: a San Francisco Bay case study. *Environmental Science & Technology* 38, 4519–4526.
- Takahashi, A., Tanabe, S., Kawaguchi, K., 2000. Organochlorine and butyltin residues in mesopelagic myctophid fishes from the Western North Pacific. *Environmental Science & Technology* 34, 5129–5136.
- Tchounwou, P.B., Ayensu, W.K., Ninasvili, N., Sutton, D., 2003. Environmental exposure to mercury and its toxicopathologic implications for public health. *Environmental Toxicology* 18, 149–175.
- Thompson, D.R., Furness, R.W., Barrett, R.T., 1992. Mercury concentrations in seabirds from colonies in the northeast Atlantic. *Archives of Environmental Contamination and Toxicology* 23, 383–389.
- Valladares, S., Moreno, R., Jover, L., Sanpera, C., 2010. Evaluating cleansing effects on trace elements and stable isotope values in feathers of oiled birds. *Ecotoxicology* 19, 223–227.
- Vanderklift, M.A., Ponsard, S., 2003. Sources of variation in a consumer-diet  $\delta^{15}\text{N}$  enrichment: a meta-analysis. *Oecologia* 136, 169–182.
- Verreault, J., Letcher, R.J., Sonne, C., Dietz, R., 2009. Dietary, age and trans-generational effects on the fate of organohalogen contaminants in captive sledge dogs in Greenland. *Environment International* 35, 56–62.
- Votier, S.C., Hatchwell, B.J., Beckerman, A., McCleery, R.H., Hunter, F.M., Pellatt, J., Trinder, M., Birkhead, T.R., 2005. Oil pollution and climate have wide-scale impacts on seabird demographics. *Ecology Letters* 8, 1157–1164.
- Wan, Y., Jin, X., Hu, J., Jin, F., 2007. Trophic dilution of polycyclic aromatic hydrocarbons (PAHs) in a marine food web from Bohai bay, North China. *Environmental Science & Technology* 41, 3109–3114.