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The heron that laid the golden egg: metals and metalloids in ibis, darter, cormorant, heron, and egret eggs from the Vaal River catchment, South Africa

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Abstract Metal pollution issues are afforded the highest priority in developing countries. Only one previous study has addressed metals in African bird eggs. We determined the concentration of metals and metalloids in bird eggs from four sites in the Vaal River catchment (VRC) of South Africa to provide data on the current situation. We analysed 16 pools of 77 heron, ibis, darter, egret, and cormorant eggs for 18 metals and metalloids using ICP-MS. We found high concentrations of gold (Au), uranium (U), thallium (Tl), and platinum (Pt) in Grey Heron eggs from Baberspan. Great white egrets from Bloemhof Dam had high concentrations of mercury (Hg). Multivariate analyses revealed strong associations between Au and U, and between palladium (Pd) and Pt. The toxic reference value (TRV) for Hg was exceeded in seven pools. Selenium exceeded its TRV in one pool; in the same pool, copper (Cu) reached its TRV. Compared with other studies, VRC bird eggs had high concentrations of contaminants. Based on these high concentrations, human health might be at risk as Grey Herons and humans share similar food and are therefore exposed to the same contaminants.

Keywords Geology · Metals · Metalloids mining · Pollution · Waterbird eggs

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Introduction

Of 22 emerging chemical management issues in developing countries, 'heavy metal' pollution was considered the highest priority (STAP 2012). Metals and metalloids in the biosphere originate in the underlying geology (McCarthy 2011) and are mobilised through anthropogenic activities (Burger and Gochfeld 2004). Waterbirds are exposed to natural and anthropogenically mobilised metals that can accumulate through trophic transfer (Burger 2002) and direct uptake (Newman 2010). Waterbirds are excellent bio-indicators because they depend on both aquatic and terrestrial environments, they may occupy high trophic levels, have relatively long life-spans, are of a large size, often occur abundantly (Kim and Koo 2007; Medvedev 1995), and their eggs are relatively easy to collect. The concentrations of metals and metalloids in bird egg contents have often been used to assess environmental contamination (Braune 2007; Burger et al. 2004; Medvedev 1995). The eggs are relative accurate representations of the levels of contaminants present in the mother's body (Ackerman et al. 2016). Despite the large body of knowledge on contaminants in sediments and fish (e.g., Pheiffer et al. 2014), there is only one prior publication that lists heavy metal concentrations in bird eggs from Africa (Greichus et al. 1977).

The Orange-Senqu River Basin (OSRB), the largest river basin in South Africa, encompasses areas of social, economic, ecological, and biological importance (ORASECOM 2013), and supports more than 11 million people (Kistin and Ashton 2008). Since the discovery of diamonds in Kimberley and gold in Johannesburg in the 1800s, the OSRB supplied water to the mines in the region. The Vaal River catchment (VRC, a sub-catchment of the OSRB) also supplies 40 % of South Africa's residents with water (Conley and Van Niekerk 2000). Because of the prominence of this mining and industrial region in southern Africa, it is important to understand the potential for metal contamination in the catchment, including elements associated with mining such as gold (Au), platinum (Pt), and uranium (U). Agricultural activities and large scale industrial water usage may also add to pollution (Earle et al. 2005). Based on Greichus et al. (1977) and Pheiffer et al. (2014), we predict that trophic transfer to birds will result in metal and metalloid concentrations higher than in lower trophic levels. The aim of this study is to quantify the concentrations of toxic metals and metalloids in the eggs of aquatic birds breeding in the VRC—the largest tributary of the OSRB—and to identify issues that may need further investigation. For this reason, we assessed a number of metallic elements pertinent to mining activities and not just those that are often focussed on.

Materials and methods

This study formed part of a larger project that had the goal to determine management priorities of the OSRB. The collection of eggs was approved by the North-West University Ethics Committee (NWU-00055-07-S3), with the necessary permits obtained from provincial departments. We investigated and compared metals and metalloids in pooled bird egg samples and assessed the data against available data and toxicity reference values (TRVs) for bird eggs (Meyer et al. 2014). We used aerial surveys to locate breeding colonies and collected 77 bird eggs from the following: grey heron *Ardea cinerea*, black-headed heron *A. melancocephala*, great white egret *A. alba*, little egret *Egretta garzetta*, cattle egret *Bubulcus ibis*, sacred ibis *Threskiornis aethiopica*, glossy ibis *Plegadis falcinellus*, African darter *Anhinga rufa*, and reed cormorant *Phalacrocorax africanus*. The species' breeding seasons are not synchronised, and the sampling period extended between December 2010 and February 2011. The eggs were collected from four sites (Fig. 1), kept on ice, and frozen within 24 h. Upon thawing, the shells were cracked and the contents homogenised ultrasonically. Equal volume sub-samples of egg homogenates were pooled by species and site according to Table 1 and freeze dried.

The EPA 3050B method was used to determine metal and metalloid concentrations in the eggs. Two grams of egg contents was acid-digested by adding nitric acid and hydrogen peroxide. The solution was heated and diluted to a volume of 50 ml. The digested solutions were analysed using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500c ICP-MS). The laboratory regularly participates in inter-laboratory calibration exercises, and a standard reference material was used (SRM 1944—New York/New Jersey Waterway Sediment). Concentrations were within 25 % of certified values. Concentrations of 18 metals and metalloids (Table 1) were expressed as milligram per kilogram dry mass (dm). Pheiffer et al. (2014) used the same method and laboratory for analysing fish samples during approximately the same period of our study, but from different sites, making the results comparable.

Since the data are from pooled samples of different species and sites, only column statistics and multivariate analysis are appropriate. We investigated the relationships of concentrations among species, elements, guilds, and sites with non-metric multidimensional scaling (NMS, MjM Software PC-ORD version 6.07; www.pcord.com), with Sørensen as the distance measure. We relativised the data per sample to accommodate the large differences in concentrations and to provide relative proportional contributions as a 'fingerprint' for each sample. Six axes and 500 iterations were allowed with randomised data for Monte Carlo tests of significance for each dimension.

Results

Most of the pools had quantifiable concentrations of mercury (Hg), with great white egrets from Bloemhof Dam (BHD) having the highest at 9.5 mg/kg dm (Table 1). The pool with the greatest concentration for metals was that of the grey heron from Barberspan (BBP) with 70 mg/kg Au, 2.9 mg/kg U, 8.5 mg/kg thallium (Tl), and 3 mg/kg Pt dm (Table 1). The coefficient of variation (%CV) of the different elements across all 18 pooled samples (Table 1) gives some indication of which species and sites had more varied concentrations, indicating localised contamination in excess of natural background. Greater %CVs may indicate varied

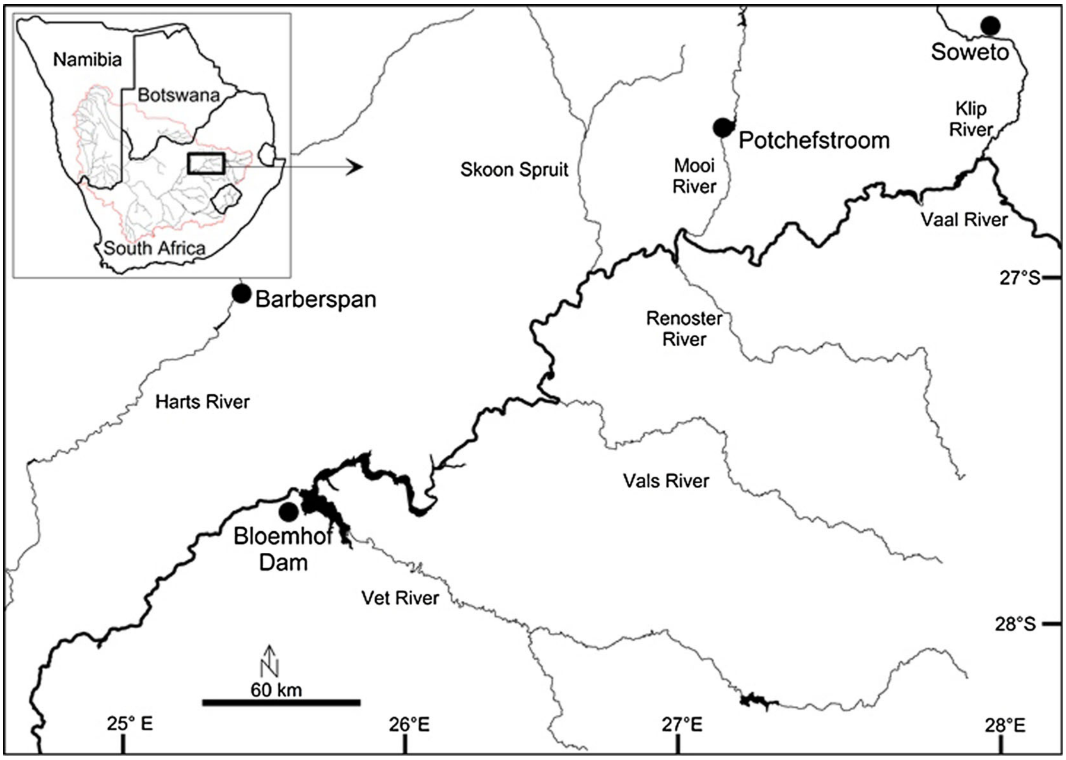

Fig. 1 Map of South Africa showing the locations of the collection sites

exposures of the birds from the different sites to the elements arguing for a pollutant source in the vicinity of one or more sites. Elements with a %CV greater than 50 % (arbitrarily chosen) were cadmium (Cd), Hg, Pt, Au, lead (Pb), palladium (Pd), and U. Elements with a %CV less than 10 % were zinc (Zn), tin (Sn), and iron (Fe), the three elements that occurred at the highest concentrations.

The NMS ordination (Fig. 2) had a final stress of 2.8 (very good) and a final instability of 0.000 reached after 54 iterations. Convex hulls (the outer extent of the sample ordinates) for the different sites are shown. Axis 1 explained 89.9 % of the variation, and axis 2, 7.2 %. U, Au, Pt, and Pd (and Cd to a lesser extent), were proportionally strongly present in the grey heron egg pool from BBP.

Discussion

The elements with a %CV greater than 50 % all had their highest concentrations in one pool, the grey heron from BBP (Table 1). The concentrations of Cd, Pt, Au, Pb, and U were at least an order of magnitude greater in the grey heron pool from BBP than the other pools. Other metals commonly considered as toxic, such as Cr, vanadium (V), manganese (Mn), Cu, arsenic (As), and Fe had remarkably low %CVs, suggesting they occur either as common contaminants throughout the system or are present as homogenous background in the environment. When the grey heron pool from BBP was removed, the %CVs dropped to below 50 % for Pt and Pb, but remained above 50 % for Hg, Au, Pd, and U (Table 1). This suggests that the latter metals were contaminants in the areas where some of the species from certain sites fed and that Pt and Pb were higher in the area of BBP than at the other sites.

All the collection sites, except BBP, are located in the Witwatersrand Supergroup geological formation. BBP is located in the Ventersdorp Supergroup. Due to the geological history of both regions, with rivers and streams depositing sediment into a shallow fossil sea, dense minerals accumulated in the area approximately 2800 million years ago (McCarthy and Rubidge 2005). Metals adsorbed to the sediment might become bioavailable if acid mine drainage enters the river system, lowering the pH, thus enhancing their water solubility (McCarthy 2011; Pheiffer et al. 2014).

Table 1 Concentrations (mg/kg) dry mass of metals in pooled eggs samples from different localities in 2010 and 2011

Species	Number of eggs pooled	Site	Guild	Cr	V	Mn	Ni	Cu	Zn	As	Se	Cd	Hg	Sn	Ti	Pt	Au	Pb	Pd	Fe	U
Grey heron	6	Barberspan	Aquatic	5.0	2.4	3.2	2.1	7	65	2.3	10	0.15	7.0	210	7.5	3.0	70	1.6	6.5	230	2.9
Grey heron	5	Bloemhof Dam	Aquatic	6.7	0	2.4	1.3	8.3	61	2.1	6.1	0.072	3.6	<LOQ	<LOQ	<LOQ	<LOQ	0.56	<LOQ	220	<LOQ
African sacred ibis	6	Bloemhof Dam	Aquatic	8.9	2.7	3.7	1.5	6.8	68	2.2	4.6	0.031	1.3	260	5.3	0.25	3.6	0.62	0.74	220	0.09
African sacred ibis	4	Soweto	Aquatic	7.0	2.2	3.8	1.3	10	65	1.8	5.5	0.038	2.1	240	5	0.36	6.5	0.6	1.1	210	0.21
African darter	4	Barberspan	Aquatic	7.5	2.8	3.4	0.95	8.5	70	1.7	6.5	0.026	2.4	250	5.2	0.24	2.2	0.37	0.51	180	0.09
African darter	3	Bloemhof Dam	Aquatic	8.5	<LOQ	3.2	1.5	9.2	61	2.6	6.7	0.026	2.5	<LOQ	<LOQ	<LOQ	<LOQ	0.51	<LOQ	200	<LOQ
Reed cormorant	5	Potchefstroom	Aquatic	8.5	2.2	4.9	1.1	5.4	62	2.0	6.2	<LOQ	3.2	240	5.5	0.31	5.5	0.41	0.85	200	0.12
Reed cormorant	5	Bloemhof Dam	Aquatic	7.8	2.9	2.9	1.5	8.9	73	2.1	6.3	0.022	2.2	280	5.7	0.33	4.8	0.52	0.79	220	0.13
Black-headed heron	5	Potchefstroom	Terrestrial	8.6	<LOQ	2.6	1.1	5.0	66	2.1	4.2	0.021	1.4	<LOQ	<LOQ	<LOQ	<LOQ	0.40	<LOQ	210	<LOQ
Black-headed heron	5	Potchefstroom	Terrestrial	8.5	2.6	2.9	1.2	5.0	70	2	5	<LOQ	0.9	250	5.1	0.2	1.8	0.39	0.39	210	0.06
Black-headed heron	6	Bloemhof Dam	Terrestrial	9.0	2.8	4.3	0.9	5.0	65	2.2	5.5	<LOQ	1.3	240	5.5	0.18	1.7	0.37	0.29	200	0.04
Cattle egret	5	Potchefstroom	Terrestrial	9.5	3.0	3.4	1.0	6.5	75	2.3	5	<LOQ	1.1	230	5	0.18	1.4	0.55	0.35	180	0.05
Cattle egret	6	Bloemhof Dam	Terrestrial	9.0	<LOQ	3.3	1.1	6.0	70	2.3	5.5	<LOQ	1.2	<LOQ	<LOQ	<LOQ	<LOQ	0.34	<LOQ	190	<LOQ
Great white egret	3	Bloemhof Dam	Aquatic	6.0	2.2	2.6	1.3	7.5	75	1.9	5	0.038	9.5	250	6.1	0.39	12	0.65	1.6	220	0.31
Little egret	4	Bloemhof Dam	Aquatic	8.0	2.7	3.3	1.3	6.0	75	2.1	4.4	0.026	1.5	230	5.2	0.29	2.9	0.38	0.52	230	0.22
Glossy ibis	5	Potchefstroom	Aquatic	8.0	<LOQ	5.0	1.2	5.0	70	2.2	7	<LOQ	1.6	<LOQ	<LOQ	<LOQ	<LOQ	0.48	<LOQ	200	<LOQ
Mean				7.9	2.4	3.4	1.3	6.9	68	2.1	5.8	0.045	2.7	240	5.6	0.52	10.0	0.55	1.2	210	0.4
%CV				15	34	22	23	24	7.1	10	24	88	88	7.4	13	160	200	55	140	7.6	220
%CV without grey heron BBP				12	35	23	16	25	7.2	10	16	47	89	6	6.5	27	76	22	56	7.3	67

(<LOQ) Concentration is lower than the limit of quantification

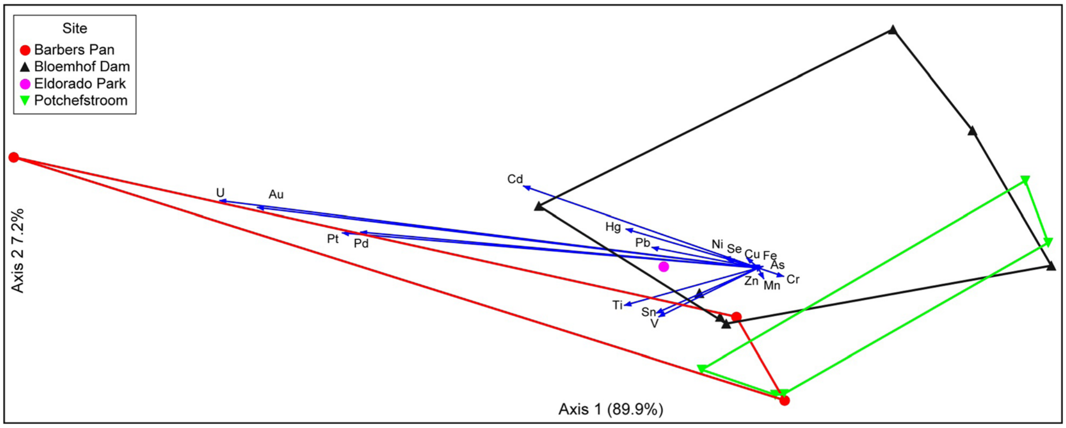

Fig. 2 Non-metric multidimensional scaling of the data. Convex hulls showing the different collection sites

Elements from the gold group (Au, Ag, and Cu) and platinum group (Pt, Pd, and Pb) are related and usually occur together (Klein and Dutrow 2007). The pairs of U and Au vectors, and the Pt and Pd vectors, are closely aligned (strongly co-variate) (Fig. 2 and Table 1), suggesting common sources for each pair, and possibly related sources for both. Mercury, mobilised from gold mining and illegal gold refining, enters the water environment at quite high concentrations in the North West Province (Lusilao-Makiese et al. 2014). Mercury is also volatile (Cukrowska et al. 2013) and can therefore travel long distances atmospherically. Its high %CV (Table 1) suggests this source as a common but unevenly distributed contaminant throughout the system we sampled. Since U and Au are mined together, and Pt and Pd belong to the platinum group of elements mined farther north, the pattern shown by the convex hulls in Fig. 2 (marginally overlapping) is suggestive of different combinations of mining-related sources of the metals at the different sites. An alternative source of Pt group elements might be attributed to automobile exhaust—Pt and Pd may be used as active components to remove harmful gasses such as CO, NO, and hydrocarbons from exhaust fumes (Rauch and Peucker-Ehrenbrink 2014). This does not, however, explain the correlation to Au group, and mining-related sources remains the more likely scenario.

The source of the high metal concentrations in the grey heron pool from BBP is not local as there are no mining activities in the BBP area. African darter from BBP had much lower concentrations (Table 1). BBP, however, is a Ramsar site and commonly used by many breeding birds. It is possible that the grey herons forage in the gold and platinum mining areas towards the east of BBP (approximately between Potchefstroom and Soweto in Fig. 1) outside the breeding season, and then breed at BBP which is a large wetland reserve. The grey herons from BHD are farther away from the mining areas and had lower overall metal concentrations. However, the breeding and foraging patterns of the grey heron in southern Africa are not well known.

We could not find any literature on Au in bird eggs, but compared to the other pools in this study, the concentration of gold in the eggs of the BBP grey heron population was very high. In general, metals and metalloids in the grey heron eggs we measured were higher than in eggs from comparable species from elsewhere (Table 2), except cattle egret eggs from Pakistan for Pb, chromium (Cr), nickel (Ni), and Cd (Bostan et al. 2007), and grey heron eggs from Turkey for Pb (Ayas 2007). We conclude that the birds we investigated had high concentrations of metals and metalloids in their eggs compared with similar birds elsewhere in the world. The concentrations of metals in bird eggs of this study were up to two orders of magnitude greater when compared to the bottom feeding catfish *Clarias gariepinus*, from a recent study by Pheiffer et al. (2014), confirming our prediction.

The developing embryo is probably one of the life stages most sensitive to xenobiotic disturbance. Only a few tissue-based TRVs for metals in bird egg contents are available (Meyer et al. 2014). The TRV for Hg in bird eggs is 2 mg/kg dm, which were exceeded by seven of the 16 pools (Table 1). The TRV for selenium (Se) is 8 mg/kg dm, and it was exceeded by the grey heron from BBP. Mercury and Se impair reproductive success in birds (Heinz et al. 2012). The TRV for Cu is between 10–20 mg/kg (Meyer et al. 2014). Copper

Table 2 Comparable data on concentrations of metals in bird eggs from elsewhere in the world

Species	Year	Location	Cu	Hg	Pb	Cr	Ni	Co	Cd	Zn	Se	As	Tl	Reference
Cattle egret	2004–2005	Pakistan			76	27	18	25	1.9					Bostan et al. (2007)
Cattle egret	2011	Potchefstroom	6.5	1.1	0.55	9.5	1.0	0.18	<LOQ	75	5.0	2.3	0.12	This study
Grey heron	2004	Turkey	6.8		6.8		0.41		0.93					Ayas (2007)
Grey heron	2010	Baberspan, South Africa	7.0	7.0	1.6	5.0	2.1	0.35	0.15	65	10	2.3	8.5	This study
Black-crowned night heron	2000–2002	Hong Kong	1.1	0.056	0.03	0.085		0.33	0.008	5.4	8.2		0.038	Lam et al. (2005)
Night heron	2004	Turkey	1.7		1.1		0.22		0.23					Ayas (2007)
Little egret	1999–2000	China		0.34	0.1	0.2			0.01	13	2.7	0.1		Zhang et al. (2006)
Little egret	2000–2002	Hong Kong	0.95	0.071	0.15	0.203		0.36	0.006		7.6		0.008	Lam et al. (2005)
Little egret	2010	Bloemhof Dam, South Africa	6.0	1.5	0.38	8.0	1.3	0.08	0.026	75	4.4	2.1	0.14	This study
Great white egret	2010	Bloemhof Dam, South Africa	7.5	9.5	0.65	6.0	1.3	0.11	0.038	75	5.0	1.9	2.0	This study
Darter	1974	Hartebeespoort Dam, South Africa	5.8	0.84	<.1				<.01	41		2.3		Greichus et al. (1977)
African darter	2010	Bloemhof Dam, South Africa	9.2	2.5	0.51	8.5	1.5	0.07	0.026	61	6.7	2.6	0.39	This study
Sacred ibis	2011	Soweto, South Africa	10	2.1	0.6	7.0	1.3	0.21	0.038	65	5.5	1.8	0.6	This study

concentrations in sacred ibis eggs from Eldorado Park, a wetland in Soweto, were 10 mg/kg dm. The TRV of Sr which varies between 66 and 73 mg/kg dm (Meyer et al. 2014) was not exceeded.

Based on these high concentrations in the pooled bird egg samples, we expect toxic effects and perhaps even impacts on reproduction, but more detailed analyses of individual eggs are required. There are no TRVs for elements such as Au, U, and Pd in bird eggs. However, these metals do have toxic properties and the effects of mixtures cannot be excluded (Nordberg et al. 2015). Further research is therefore needed on the elements not normally covered. More research is also needed on how elements such as gold and platinum in the environment are taken up by biota, an aspect of which little is known.

Conclusion

The birds in this study, especially the grey heron, consume prey such as fish, some of which may be utilised by humans as well (Pheiffer et al. 2014). Humans may even collect wild bird eggs as a food source (Burger and Elbin 2015). Some of the metals such as U and Hg are well known for their detrimental health impacts (Winde 2010; WHO 1980), especially on foetuses and young children (Ackerman et al. 2016). The potential negative effects of the accumulation of toxic metals and metalloids in humans and biota from the VRC (and possibly the OSRB catchment as a whole) cannot be ignored and need more investigation.

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