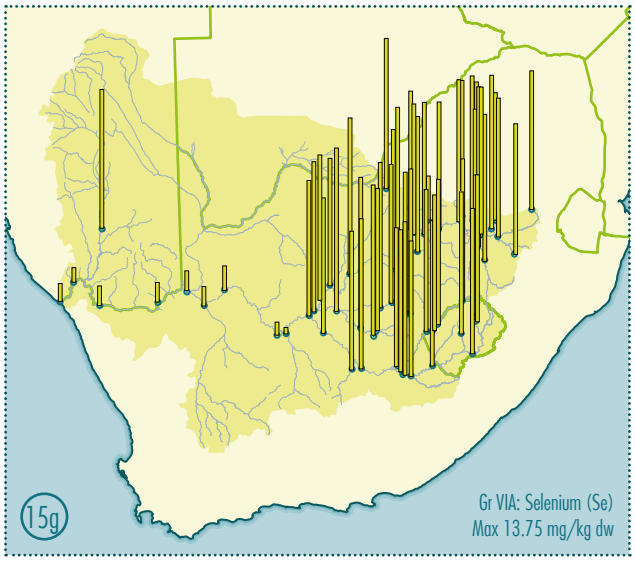
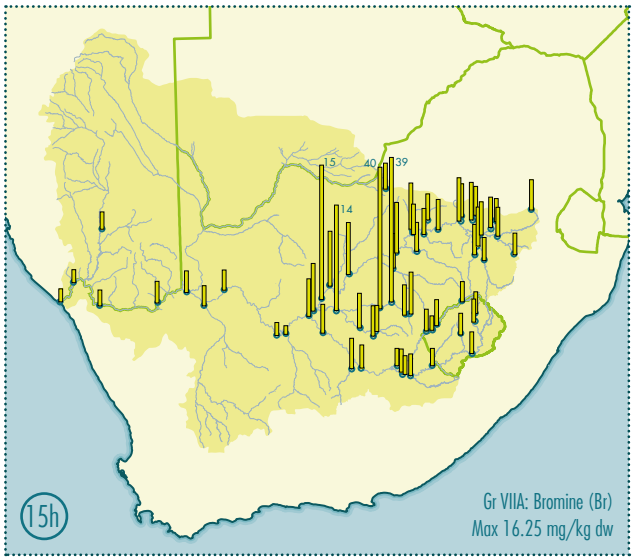
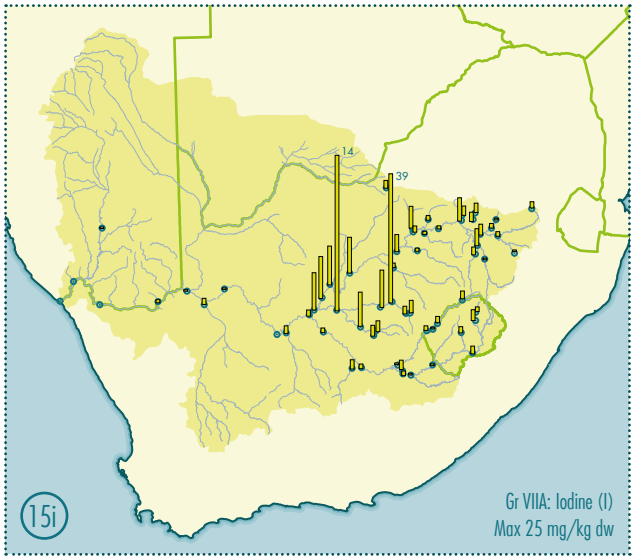
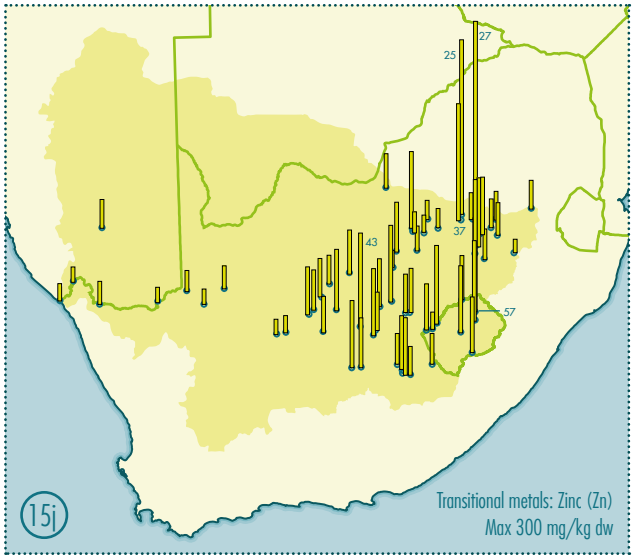
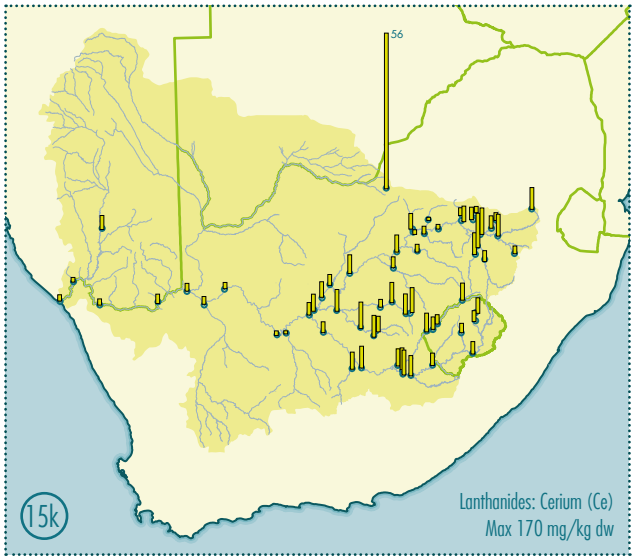
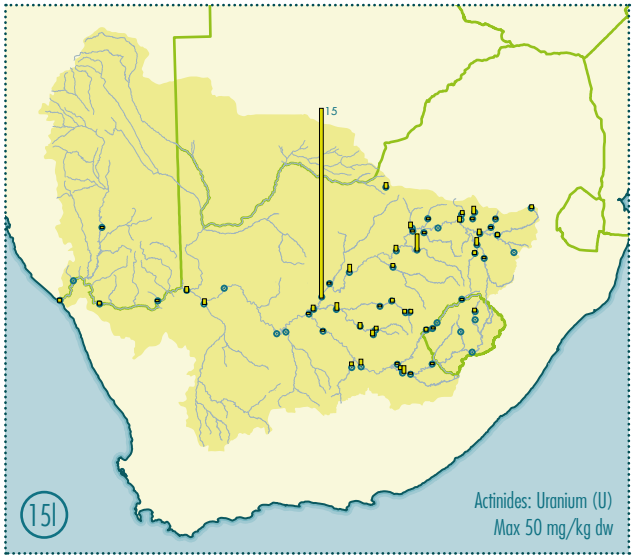

15g
Gr VIA: Selenium (Se)
Max 13.75 mg/kg dw


15
40
39
14
15h
Gr VIIA: Bromine (Br)
Max 16.25 mg/kg dw


14
39
15i
Gr VIIA: Iodine (I)
Max 25 mg/kg dw


27
25
37
43
57
15j
Transitional metals: Zinc (Zn)
Max 300 mg/kg dw


56
15k
Lanthanides: Cerium (Ce)
Max 170 mg/kg dw


15
15l
Actinides: Uranium (U)
Max 50 mg/kg dw

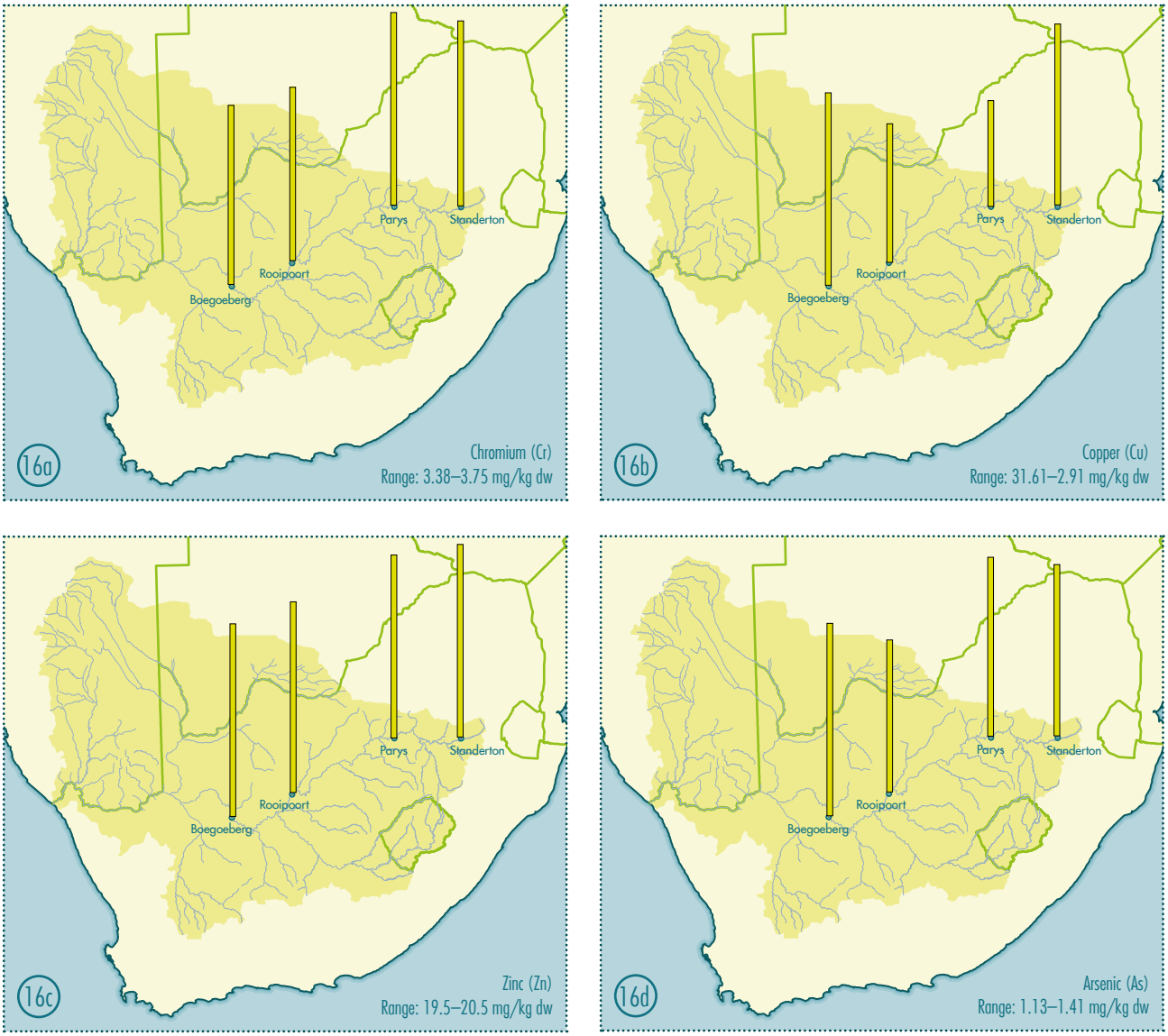
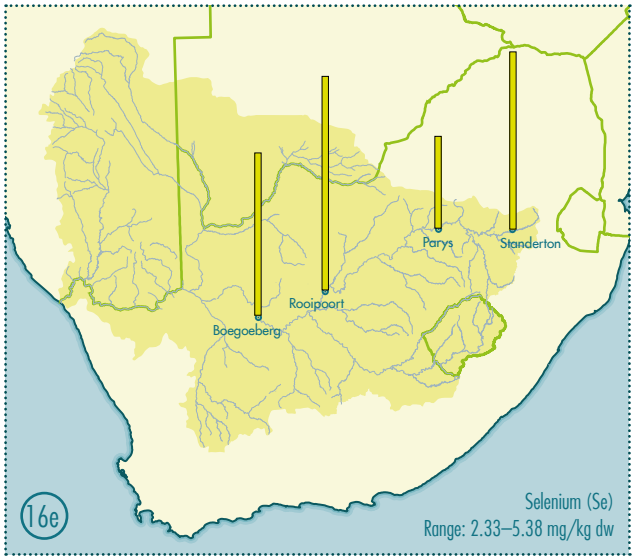


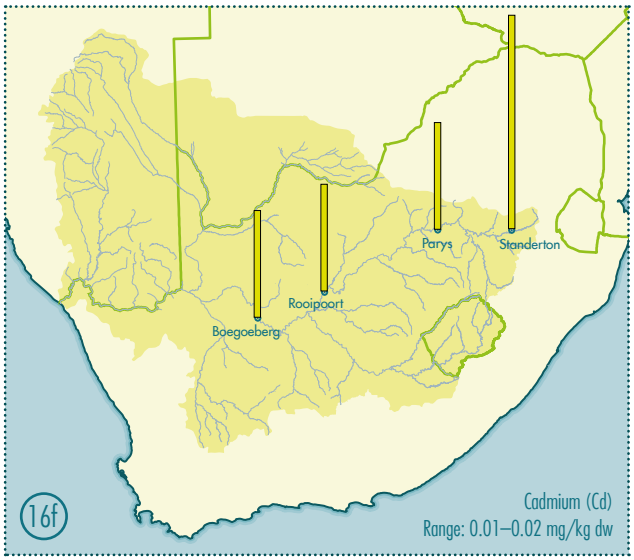
Figure 16: Levels of elements in fish from the sampling sites. Bar scale is relative for each element, but not between elements. Range = range of all samples.

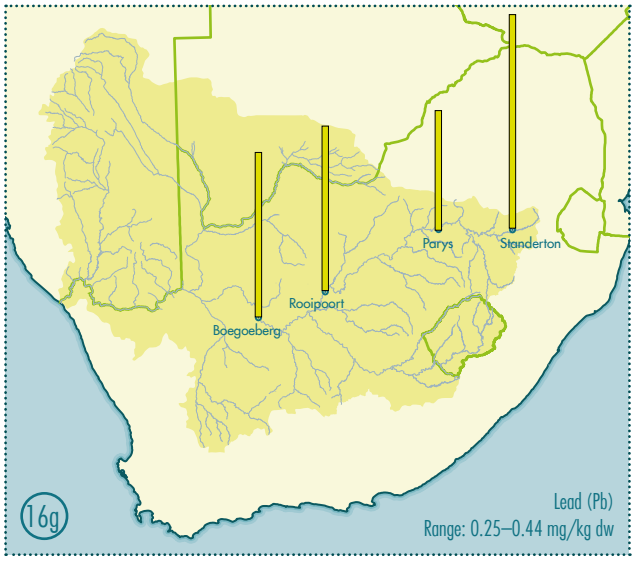
Fish

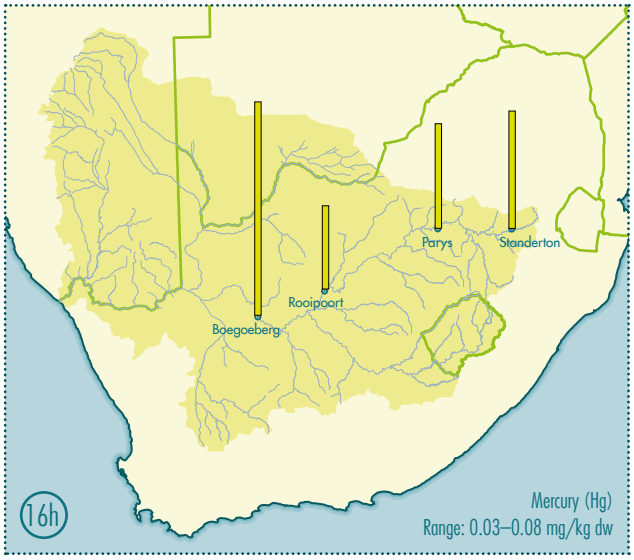
The investigation of elements in fish focused on those with known toxic effects and for which guideline levels in fish fillet (i.e. considered safe for human consumption) were found. Values for the heavy metals chromium (Cr), copper (Cu), zinc (Zn), arsenic (As), selenium (Se), cadmium (Cd) and lead (Pb) were sourced from Wagner and Boman (2003), who cited a 'range of international standards' compiled by the Food and Agricultural Organisation (FAO) from different countries for contaminants in fresh fish. Wagner and Boman (2003) also noted that values expressed as dry weight (dw) could be converted into wet weight (ww) by dividing them by factors ranging from four to six. This conversion factor is supported by the US-based Consortium for Risk Evaluation with Stakeholder Participation, which recommends multiplying wet weight values of contaminants in fish by five to give an approximate conversion to dry weight values (CRESP, 2006).

In recent years, food standards have been revised by individual countries and by the Codex Alimentarius Commission (CAC), an inter-governmental body to implement the Joint FAO/WHO Food Standards Programme. In the current (2010) version of









the Codex General Standard for Contaminants and Toxins in Food and Feed, the only heavy metals for which Standards in fish products are provided are lead (0.3 mg/kg ww) and methylmercury (0.5 mg/kg ww) (Codex Standard 193). The European Union has food safety limits for cadmium (0.05 mg/kg ww), lead (0.3 mg/kg ww) and mercury (Hg) (0.5 mg/kg ww) in freshwater fish (EC 2006).

According to Wagner and Boman (2003), the range of international standards for Cu in fish fillet is 10 to 100 mg/kg and for Cr it is 1 mg/kg wet weight (ww). This means that the Cu and Cr levels measured for pooled fish fillets in this study were well within guideline levels (Figures 16a and b).

The Zn levels found in fish fillet in this study were far below that allowed by international standards, which varied between 40 and 100 mg/kg ww (Wagner and Boman, 2003). The concentrations were similar between the four sites (Figure 16c).

The highest As levels were found in the fillet of fish caught in the only Orange River site (O2). According to Wagner and Boman (2003), As may vary between 0.1 and

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5 mg/kg ww depending on the fish species. The highest level for As in this study was 1.41 mg/kg dw, equivalent to approximately 0.28 mg/kg ww (Figure 16d).

Results for Se ranged between 2.3 and 5.4 mg/kg dw, with the highest measured in fish from site V3 (Figure 16e). The 'safe' range of 0.3 to 2 mg/kg ww (Wagner and Boman, 2003) would equate to 1.5–10 mg/kg dw, using a conversion factor of 5.

The international standards for Cd are 0.05 to 2 mg/kg ww (EC, 2006; Wagner and Boman, 2003), for Pb they are 0.5 to 10 mg/kg ww (Wagner and Boman, 2003), and for Hg they are 0.5 mg/kg ww (EC, 2006). The levels of Cd, Pb and Hg in fish analysed in this study (Figure 16f–h) were well within these thresholds. The highest Hg level was found in fish from the Orange River site (O2) and although the level was twice as high as fish from the other sites, it was still below international limits.

The elemental bioaccumulation factors for fish were determined from the data. A bioaccumulation factor is the ratio of the contaminant concentration in the organism to that of the potential ambient sources (Newman, 2010), in this case the sediment. The term biota-sediment accumulation factor (BSAF) is therefore used. The BSAF for each of the fish pools was calculated using the concentrations at one or two sediment sites closest to where the fish were sampled, and the mean BSAF determined. It was assumed that 100% of the measured concentration in the sediment was bioavailable to fish, which implies that BSAF would be much higher if bioavailability is in fact lower. Only elements for which the mean BSAF was found to be greater than one are reported, namely rubidium (Rb), rhodium (Rh), silver (Ag), tin (Sn), platinum (Pt) and gold (Au) (Table 11).

Table 11: The BSAF of elements that showed accumulation in fish tissue.

Fish pools	Sediment sites	Rb	Rh	Ag	Sn	Pt	Au
V1	35	0.3	2.9	1.2	0.9	5.6	2.3
V2	24	3.7	0.8	1.9	2.3	10.6	1.6
V3	12, 15	1.0	0.4	0.1	0.7	3.5	1.5
O2	2, 9	2.8	0.3	1.5	1.5	3.3	0.2
Mean		1.9	1.1	1.2	1.3	5.8	1.4

For each element that showed bioaccumulation, the highest BSAF was at fish pool V1 or V2 (Table 11). The mean for all fish pools showed that Pt had the strongest bio-accumulation, more than double that of the element with the next highest BASF, Rb.

Bird eggs

Only elements with the five highest concentrations at each site, irrespective of bird species, are reported here. The complete dataset is provided in the addendum. Black-headed herons were not included in this assessment because they are mainly terrestrial feeders. Tin (Sn) had the highest level in bird eggs at all of the sites, followed by iron (Fe) (Table 12). These two elements, together with boron (B) and zinc (Zn), were ranked among the top four at each of the egg collection sites.

Table 12: The five highest elemental concentrations measured in any of the wild bird eggs at each of the bird sites. Concentrations are in mg/kg dw and arranged in decreasing order.

Barbers Pan			Bloemhof Dam			Eldorado Park*			Potchefstroom		
African darter	Sn	235	Great white egret	Sn	280.6	Sacred ibis	Sn	240	Reed cormorant	Sn	255.4
Grey heron	Fe	230	Little egret	Fe	230.0	Sacred ibis	Fe	205	Reed cormorant	Fe	204.1
Grey heron	B	115	Great white egret	Zn	75.0	Sacred ibis	Zn	65	Cattle egret	Zn	75.0
African darter	Zn	70	Great white egret	B	65.0	Sacred ibis	B	40	Reed cormorant	B	34.1
Grey heron	Au	70	Great white egret	Au	19.5	Sacred ibis	Al	21	Cattle egret	Ba	17.0

*Only sacred ibis eggs collected at Eldorado Park were analysed.

The bioaccumulation of elements from fish to birds was evaluated. This could only be done for elements analysed in both fish and bird eggs, and it is assumed that 100% of the measured concentration in fish will be bioavailable to birds, although fish and egg samples were not taken at the same sites. The bioaccumulation factors were calculated for each bird species relative to each of the fish samples. This approach was followed because the birds would not necessarily feed on the fish species collected for elemental analysis, nor would the birds' uptake of elements be through food only. It was assumed that all the bird species in this study would consume sharptooth catfish. The mean bioaccumulation factor (MBF) for each element was calculated to provide a broad indicator of the range of bioaccumulation factors, irrespective of the species. A bird species-specific bioaccumulation factor (SSBF) was also calculated for each element to assess if the different species would accumulate the elements differently.

The MBF showed that all the elements bioaccumulated (at levels higher than in fish) in the bird eggs, except Cs (Table 13). Sn had the highest MBF of 313.6, followed by Au at 108.1. Six elements – Be, B, Mo, Pd, Tl and U – had MBFs between 10 and 20, and the rest had MBFs less than 10. Grey herons accumulated the greatest percentage of elements to the highest extent (42%), followed by cattle egret (26%). These results should be treated with caution, however, as the diet of cattle egrets is dominated by insects.

Table 13: The MBF of elements in bird eggs as well as the SSBF for the different bird species. MBF and SSBF were calculated assuming the birds consume sharptooth catfish. The highest SSBF for each element is shaded.

		SSBF							
	MBF	Grey heron	African darter	Great white egret	Reed cormorant	Sacred ibis	Little egret	Cattle egret	Glossy ibis
Be	10.7	22.3	9.8	10.9	13.0	12.1	3.7	3.0	4.5
B	10.9	25.5	10.9	18.8	8.5	8.8	5.1	2.1	6.5
Al	2.1	1.9	1.7	1.3	1.4	2.5	1.7	3.5	1.9
Sc	1.8	2.7	1.8	2.1	2.0	1.8	1.3	0.7	1.5
Ti	1.8	0.3	0.2	0.3	0.2	0.2	0.4	10.5	0.3
V	2.1	1.9	2.2	1.5	2.3	2.1	2.4	2.0	2.3
Cr	2.3	1.6	2.3	1.7	2.3	2.2	2.2	3.6	2.2
Mn	3.3	2.8	3.3	2.6	4.0	3.8	3.3	2.2	5.1
Fe	6.8	4.2	3.5	4.1	3.9	3.9	4.2	22.6	3.6
Co	2.8	4.7	1.7	2.3	1.8	3.5	1.6	1.9	4.8
Ni	3.5	4.7	3.5	3.7	3.6	3.9	3.7	1.7	3.2
Cu	3.2	3.5	4.0	3.4	3.2	3.8	2.7	2.2	2.3
Zn	4.5	3.1	3.2	3.7	3.3	3.3	3.7	10.7	3.5
Ga	1.4	1.3	1.4	1.2	1.3	1.6	1.5	1.1	1.8
Ge	1.9	1.8	2.1	1.9	2.1	2.2	2.1	1.3	1.9
As	1.6	1.7	1.7	1.5	1.6	1.6	1.6	1.5	1.7
Se	1.6	2.2	1.8	1.4	1.7	1.4	1.2	1.4	1.9
Rb	1.5	0.2	0.2	0.3	0.4	0.5	0.3	7.9	0.4
Sr	2.7	2.2	2.8	1.8	2.0	4.5	2.1	1.5	5.0
Mo	16.4	16.6	27.4	16.9	23.7	13.0	12.5	8.2	6.0
Pd	14.1	42.3	8.6	23.1	6.6	8.5	3.7	9.5	5.3
Cd	3.4	10.9	2.5	3.7	1.3	3.4	2.5	0.4	0.5
Sn	313.6	324.9	364.2	310.8	345.9	346.1	339.1	166.4	332.0
Cs	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.2	0.1
Ba	2.3	1.9	2.0	1.6	2.0	2.1	2.1	3.9	2.6
Ce	1.2	1.0	0.6	0.7	0.8	3.3	0.9	0.7	1.1
Pt	3.8	12.9	2.5	4.6	2.1	2.3	1.5	0.6	2.5
Au	108.1	432.7	54.3	206.6	34.4	46.3	18.5	9.6	26.0
Tl	11.3	49.8	4.4	20.1	2.7	4.1	1.4	0.7	1.9
Pb	1.7	3.3	1.3	2.0	1.4	1.9	1.2	1.0	1.5
U	10.9	48.3	3.8	16.6	4.7	3.2	2.1	0.8	2.0
% elements with highest SSBF		**41.9**	**9.7**	**0.0**	**0.0**	**6.5**	**3.2**	**25.8**	**12.9**

4. DISCUSSION

4.1 ORGANIC POLLUTANTS

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Dioxin-like TEQ was found in sediments in the eastern parts of the Orange–Senqu River basin, but was undetectable in the western parts. The presence of dioxin-like TEQ in fish from Rooipoort needs further investigation, and a species-specific analysis is needed for birds. Fish from the Gauteng region were not sampled in this study, and it would be useful to do so in the future to investigate the impact of industrial activities.

PCBs in sediment were also higher towards the eastern than the western parts of the catchment, but levels in fish from Rooipoort were higher than in any of the sediment samples. Appreciable levels were also found in some bird eggs, especially at Bloemhof Dam.

In sediments, OCPs were highest in the Gauteng region, and mostly undetectable in the rest of the basin. Although the highest levels of OCPs in fish were from Parys, in birds the highest levels were from Bloemhof Dam, which could possibly be acting as a biological retainer of some compounds coming from upstream. It would have been instructive to sample fish and sediments from Bloemhof Dam as well, but the bird colony at Bloemhof Dam was only located after the fish sampling was completed.

None of the sediment samples had detectable levels of PFOS, but fish from the two sampling sites in the central parts of the basin contained PFOS, while bird eggs from Barbers Pan and especially Bloemhof Dam had surprisingly high levels. This requires further exploration, and a species-specific assessment needs to be done.

The three sediment sites with the highest ∑PAH levels were downstream of urban and industrial areas in South Africa and Lesotho. The sources for PAHs were pyrogenic in nature, typically associated with coal combustion or smelter operations. The most common PAH was fluoranthene, followed by phenanthrene and benzo(b+k) fluoranthene.

In summary, sediment had higher levels of POPs towards the east, decreasing drastically downstream towards the west, but this distribution pattern was not reflected in biota. This study has shown that sediment analysis alone will not provide sufficient information to assess contaminant levels in biota and the risk to human health. However, using pollution distribution patterns on a basin scale assists in assessing the overall pollution picture and in identifying hotspots and areas of interest.

In order to clarify some aspects, it would be very informative to sample fish from a number of additional areas, including between Bloemhof Dam and Boegoeberg, Upington, Modder River, Klerksdorp, Potchefstroom, in or near Lesotho, and Gauteng, while bird eggs from the colony at Upington might yield helpful data, especially with regard to PFOS distribution.

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4.2 HEAVY METALS AND TRACE ELEMENTS

The MPI for sediment site 56, the Molopo Eye, was the highest, and when the I_{geo} values were determined the same site had the most elements with I_{geo} values regarded as polluted. This site, together with a cluster of sites in the Riet and Modder rivers, also belonged to the 25% of sites with the most elements with the highest concentrations. Shared geology between some of the sites could partially explain this, but in-depth investigation of the area is deemed necessary to determine the exact cause. High levels of elements at two sites in Lesotho are also likely to be due to geology rather than mining or any other anthropogenic activities.

Levels of the elements in the sediment warrant further investigation, especially in light of the I_{geo} values for Se, As and Hg. However, intensive investigation into the natural background levels for these elements is needed for clarification.

When the sediment levels are compared to sediment quality guidelines for the Netherlands (only for As, Ba, Cd, Cr, Co, Cu, Pb, Hg, Mo, Ni and Zn (Adriano, 2001), and U and Se (Hamilton, 2004; Sheppard et al., 2005), all of the sites had a value less than 0.5, which is considered to be a low probability of being toxic, apart from the site at the Molopo Eye. There, the value of 1.6 implies that the sediment levels of elements have a high probability of being toxic to the biota in the system.

The levels of Cu, Cr, Zn, As, Se, Cd, Pb and Ag in the fish fillets were compared to international food safety guidelines and standards, and were found to be well within acceptable limits, with the possible exception of Se. Those elements that seemed the most likely to bioaccumulate in the fish, if exposure to sediment is their only exposure pathway and 100% of the measured concentrations are bioavailable, were Pt > Rb > Au > Sn > Ag > Rh. None of the other elements had bioaccumulation factors greater than 1. Further assessment is needed on the bioavailable fractions of the elements at the sites identified.

Sn had the highest level at all of the bird egg pools, followed by Fe, and the same four elements – Sn, Fe, B and Zn – had the highest levels at each of the bird egg collection sites. All the elements had bioaccumulation factors greater than 1, except for Cs. However, this is only a broad indication of possible bioaccumulation in the catchment as these calculations were done under a number of assumptions. There is also a possibility that the various bird species bioaccumulate particular elements differently, but the grey heron seemed to bioaccumulate more elements than the other species. The eggs of the grey heron might be a good bioindicator and the species of choice in situations where only a single bird species may be sampled.

5. RISK ASSESSMENT

5.1 BACKGROUND AND METHODS

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A human health risk assessment was conducted to determine if the contaminants in the sediment, fish fillets and bird eggs tested would be likely to cause adverse health effects to humans consuming the fish or eggs from these areas or being in regular skin contact with the sediment (dermal exposure). The methodology with which this human health risk was assessed is described by the USEPA (1987, 1992, 1996) and the WHO (2002). Human health risk assessment consists of four steps:

- *Hazard identification*. Can exposure lead to toxic or carcinogenic health effects?
- *Dose-response assessment*. What is the relationship between the agent and incidence of adverse health effect?
- *Exposure assessment*. What is the sum total of expected exposure to the agent?
- *Risk characterisation*. What is the probability of an adverse effect due to exposure to the agent?

For agents that cause non-cancer toxic effects, a hazard quotient (HQ) can be calculated in order to compare the expected exposure to the agent to an exposure that is assumed not to be associated with toxic effects.

For oral or dermal exposures, the average daily dose (ADD) was compared to a reference dose (RfD):

$$HQ = ADD/RfD \qquad \text{Equation 1}$$

Any HQ less than 1 is considered safe for a lifetime exposure.

For chemicals that may cause cancer if ingested, risk is calculated as a function of oral slope factor and dose, and can be calculated using the following formula:

$$\text{Risk} = 1 - e^{-(\text{oral slope factor} \times \text{lifetime average daily dose})} \qquad \text{Equation 2}$$

In this report the human health risk assessment will be based on the HQ determined for toxic agents and risk calculated for the various carcinogens.

As a screening risk assessment, the maximum concentration detected in all sample sites was used as a worst-case scenario to determine what risks (if any) were involved. If a chemical was found to be responsible for risks considered by the USEPA and WHO to be unacceptably high, a detailed assessment for that contaminant was done, making use of the spread of the data and averages, and identifying which sampling site was responsible for the highest concentrations detected.

Cross-media transfer equations were used to determine the amount of contaminant that could transfer from the sediment into fish. The formulae used to generate the exposure concentrations based on sediment concentrations was that described by the USEPA (1990) for sediment to fish concentrations.

$$C(w) = \frac{C(sd)}{(K_{OC} \times OC \times DN)} \qquad \text{Equation 3}$$

$$BCF = (0.79 \times \log(K_{OW})) - 0.40 \qquad \text{Equation 4}$$

$$C(f) = BCF \times \left(\frac{fat}{3}\right) \times C(w) \qquad \text{Equation 5}$$

Abbreviations:

C(w)	concentration in water
C(sd)	concentration in sediment
C(f)	concentration in fish
DN	sediment density (relative to water density of 1.0 kg/ℓ) (1.9)
OC	organic carbon fraction of sediment (4.0%)
K_{OC}	octanol-carbon partition coefficient of the compound
K_{OW}	octanol-water partition coefficient of the compound
BCF	bioconcentration factor

These calculations, however, restrict the number of contaminants that can be investigated as there are limited contaminants for which K_{OW} and K_{OC} values are available. To determine the risk posed by contaminants, various exposure parameters are used to assess the average exposure of humans. The exposure parameters used in this risk assessment are listed in Table 13.

Table 13: Exposure parameters used to generate exposure estimates.

Exposure parameter	Amount
Events per year	350
kg per event (fish)	0.054
kg per event (eggs)	0.060
Body mass	70 kg
Exposure duration	30 years

The average daily dose was calculated taking into account the concentration of the chemicals in sediment, for a 70 kg adult, assuming an intake of 0.054 kg of fish and 0.060 kg of eggs from wild birds on a daily basis (equivalent to 378 g per week). A range of risks is presented making use of average and 95th percentile concentrations of chemicals detected in the sediment, calculated to represent concentrations expected in fish. The 95th percentile represents the 'reasonable maximum' risk.

5.2 RESULTS

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The results of the exposure calculations are presented as both average daily dose (ADD) and lifetime average daily dose (LADD) in mg/(kg × d) for fish, bird eggs and sediment (Tables 14 and 15). Based on the exposure assumptions described above, risks of developing cancer and toxic effects were calculated for those contaminants where sufficient data were available (Tables 14 and 15 and Figures 17 and 18). An HQ of less than 1 is considered to be safe for a lifetime exposure, with no negative health effects experienced (USEPA 1991). A cancer risk of greater than one in 100,000 is considered unacceptable (WHO 2002). Most of the chemicals were at concentrations below the 'unacceptable' risks level, as defined by both the WHO and USEPA.

Beryllium, and especially arsenic, presented high risks of developing cancer from all media investigated (Figure 17). The risk of developing cancer from the consumption of bird eggs is as high as one in 1,000. PAHs were only measured in sediment and the following pose between a two in 100,000 and five in 100,000 risk for the development of cancer: indeno(1,2,3-cd)pyrene, benz(a)anthracene, benzo(b)fluoranthene and benzo(k)fluoranthene. The other PAHs are still being investigated by the EPA and their slope factors are derived from other isomeric chemicals. Benzo(a)pyrene in sediment poses an 'unacceptable' cancer risk of one in 10,000. This sediment sample was collected from site 64 which also had significant pyrene, benz(a)anthracene, chrysene, fluoranthene, fluorene, indeno(1,2,3-cd)pyrene and pyrene concentrations. Of the POPs, PCBs in bird eggs pose a two in 10,000 cancer risk. This bird egg sample was taken from Bloemhof Dam, which also posed a significant arsenic cancer risk.

Arsenic in the bird eggs and fish samples presented a high toxic health risk too, with HQ values exceeding 3. The chromium and selenium HQ value in the bird eggs also exceeded 1, posing a possible health risk. These concentrations were measured in pool 15 and pool 1 respectively, and therefore pose individual risks. The only PAH which is an emerging health risk is indeno(1,2,3-cd)pyrene in sediment. The reference dose for this PAH is, however, still under investigation by the EPA, and is only derived from isomeric compounds.

Table 14: The ADDs, LADDs, toxic health risks and cancer risks associated with exposure to the maximum concentrations detected in bird eggs and fish.

Contaminant	Bird eggs				Fish			
POPs	ADD mg/kg/d	LADD mg/kg/d	HQ	Risk	ADD mg/kg/d	LADD mg/kg/d	HQ	Risk
Total PCBs	8.72E-05	3.74E-05		2.00E-04	4.89E-06	2.10E-06	0.048	1.61E-05
TetraBDE	1.06E-06	4.56E-07	0.011		1.30E-07	5.55E-08	0.0013	
PentaBDE	3.55E-06	1.52E-06	0.001		7.95E-08	3.41E-08	7.95E-04	
HexaBDE	5.33E-06	2.28E-06	0.027					
HeptaBDE	7.09E-06	3.04E-06						
HexaBB	3.30E-07	1.40E-07	0.0002					
PFOS	0.001997	0.000856						
β-HCH	4.8E-06	2.06E-06		3.70E-06				
HCB	1.00E-06	5.80E-07	0.002	9.00E-07				
Dieldrin	6.00E-06	2.00E-06	0.11	4.00E-05				
Heptachloroepoxide	8.57E-07	3.67E-07						
p,p'-DDE	2.47E-04	1.06E-04		4.00E-05				
o,p'-DDD	5.00E-06	2.00E-06		5.00E-07	6.94E-07	2.98E-07		1.24E-06
p,p'-DDT	7.00E-06	3.00E-06	0.02	1.00E-06				
Heavy metals								
Aluminium	1.43E-02	6.12E-03	0.002		1.79E-05	7.69E-06	2.56E-06	
Arsenic	1.58E-03	6.77E-04	5.26		1.10E-03	4.71E-04	3.66	3.14E-04
Boron	5.24E-02	2.24E-02	0.26		2.89E-03	1.24E-03	0.01	
Barium	1.46E-02	6.24E-03	0.07		5.40E-03	2.31E-03	0.027	
Beryllium	8.14E-05	3.49E-05	0.04	1.50E-04	5.79E-06	2.48E-06	0.003	5.77E-07
Cadmium	9.10E-05	3.90E-05	0.18		1.54E-05	6.61E-06	0.015	
Cobalt	3.00E-04	1.29E-04			6.36E-05	2.73E-05		
Chromium	5.86E-03	2.51E-03	1.17		2.89E-03	1.24E-03	0.96	
Copper	8.57E-03	3.67E-03			2.70E-03	1.16E-03		
Iron	1.97E-01	8.45E-02			5.98E-02	2.56E-02		
Mercury	8.14E-03	3.49E-03						
Manganese	4.29E-03	1.84E-03	0.031		9.45E-04	4.05E-04	0.007	
Nickel	1.76E-03	7.53E-04	0.088		1.27E-03	5.46E-04	0.06	
Lead	1.37E-03	5.88E-04			3.66E-04	1.57E-04		
Antimony	1.16E-05	4.96E-06	0.029		4.44E-03	1.90E-03	0.89	
Selenium	6.16E-03	2.64E-03	1.23					
Titanium	6.43E-03	2.76E-03			2.12E-02	9.09E-03		
Uranium	4.11E-05	1.76E-05			9.06E-05	3.88E-05		
Vanadium	2.53E-03	1.08E-03	0.28		9.64E-04	4.13E-04	0.11	
Zinc	4.62E-02	1.98E-02	0.15		1.83E-02	7.85E-03	0.06	

Table 15: The ADDs, LADDs, toxic health risks and cancer risks associated with exposure to the maximum concentrations detected in sediment.

Contaminant	Sediment			
POPs	ADD mg/kg/d	LADD mg/kg/d	HQ	Risk
Total PCB	9.57E-08	4.10E-08	9.57E-04	3.16E-07
TetraBDE	1.99E-08	8.52E-09	1.99E-04	
PentaBDE	5.92E-08	2.54E-08	5.92E-04	
HexaBDE	1.80E-07	7.72E-08	9.01E-04	
Lindane	7.00E-06	3.00E-06	2.27E-02	
p,p'-DDE	8.24E-08	3.53E-08		1.20E-08
o,p'-DDD	4.79E-08	2.05E-08		4.93E-09
p,p'-DDT	9.45E-08	4.05E-08	1.89E-04	1.38E-08
Heavy metals				
Arsenic	1.35E-04	5.79E-05	0.45	8.68E-05
Beryllium	7.88E-06	3.38E-06	3.94E-03	1.45E-05
Lead	7.24E-04	3.10E-04	3.62E-03	
PAHs				
Acenaphthene	2.18E-03	9.34E-04	3.63E-02	
Acenaphthylene	3.28E-06	1.41E-06		
Anthracene	1.10E-05	5.00E-06	3.60E-05	
Benz(a)anthracene	8.03E-05	3.44E-05		2.51E-05
Benz(a)pyrene	3.80E-05	1.60E-05		1.00E-04
Benzo(b)fluoranthene	1.54E-04	6.58E-05	1.75	4.81E-05
Benzo(ghi)perylene	1.02E-04	4.39E-05	3.41E-03	
Benzo(k)fluoranthene	1.54E-04	6.58E-05		4.81E-05
Chrysene	9.26E-05	3.97E-05		2.90E-07
Dibenz(ah+ac)anthracene	1.51E-05	6.46E-06	1.71E-02	
Fluoranthene	2.47E-04	1.06E-04	6.19E-03	
Fluorene	3.10E-05	1.30E-05	7.75E-04	
Indeno(1,2,3-cd)pyrene	7.65E-05	3.28E-05	0.869	2.39E-05
Naphthalene	2.64E-05	1.13E-05	1.32E-03	
Phenanthrene	8.81E-05	3.78E-05	2.94E-03	
Pyrene	8.60E-05	3.70E-05	2.87E-03	

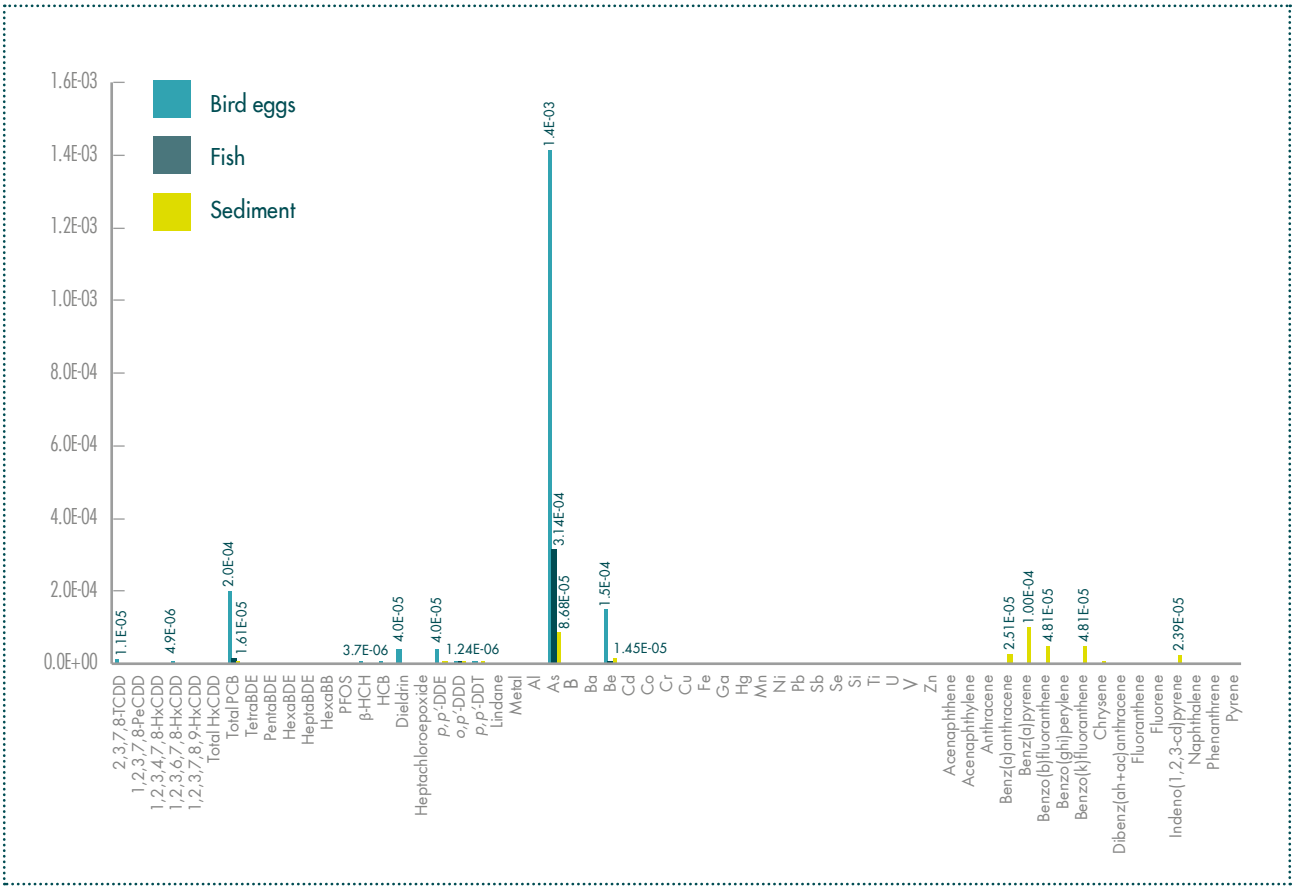

Figure 17: The risk of developing cancer determined from the maximum concentrations detected in wild bird eggs, fish and sediment.

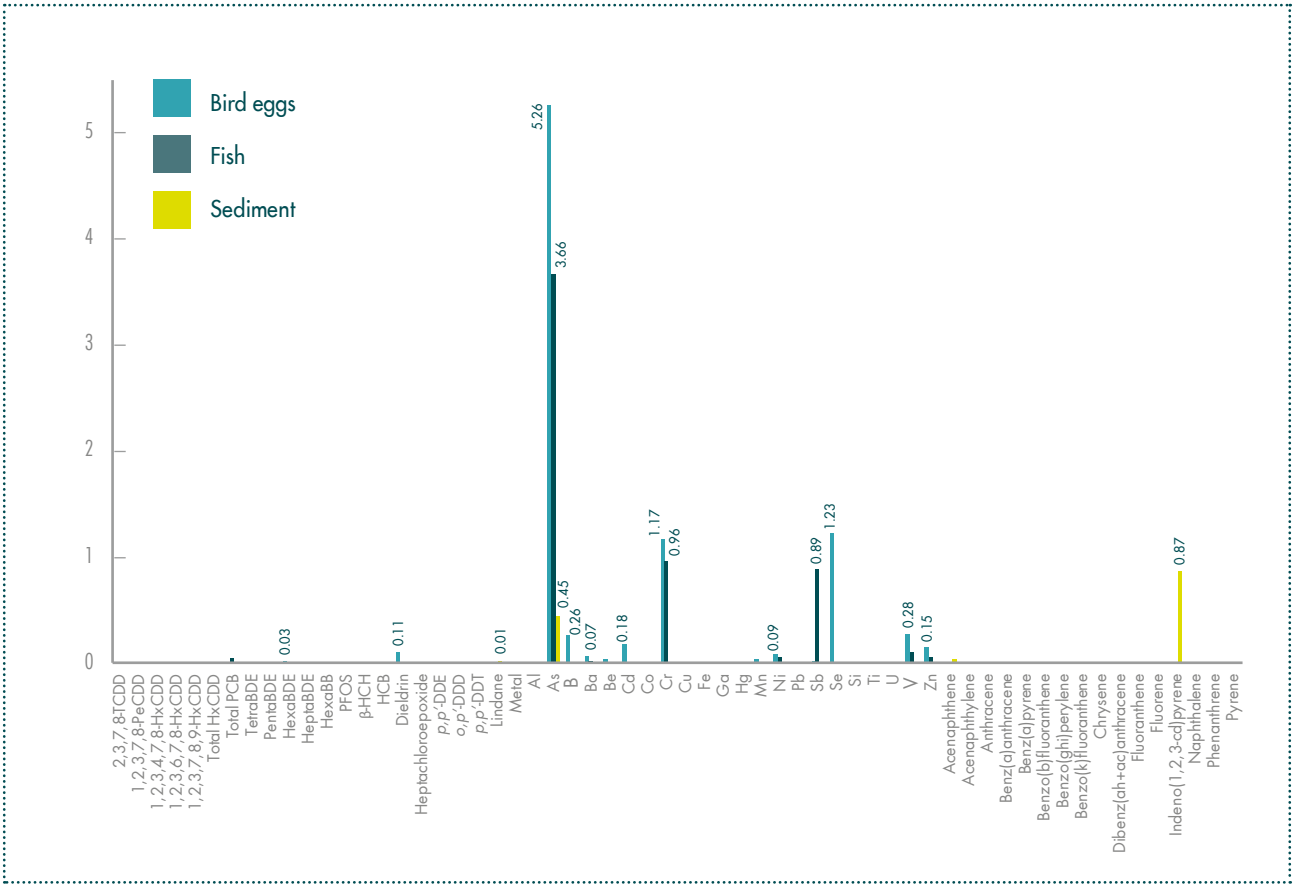

Figure 18: The hazard quotient determined from the maximum concentrations detected in wild bird eggs, fish and sediment.

5.3 DISCUSSION

The human health risk assessment examined whether possible human health effects might be anticipated based on chemical contaminants detected in sediment, bird eggs and fish samples. The methods involved modelling the contaminant concentrations expected in fish based on levels detected in sediments. Trans-media calculations (sediment to fish) were conducted based on individual chemical parameters described in the earlier sections. These used partition coefficients from the literature, although reported values vary (Bowman and Sans, 1983; Ahangar, 2011).

The risk assessment identified the chemicals that could potentially be responsible for adverse health effects if wild-harvested fish or bird eggs were to be eaten on a daily basis over a 30-year period. The contaminants exceeding the risk threshold were identified as arsenic, benzo(a)pyrene, PCBs, chromium and selenium. The type of adverse effect that might result was also identified as predominantly carcinogenic, associated with arsenic, benzo(a)pyrene and PCBs exposure, with other toxic effects being anticipated from heavy metal exposure to chromium, arsenic and selenium.

This risk assessment has highlighted that possible health risks can be anticipated resulting from regular ingestion of fish or eggs collected from the wild. However, there are uncertainties in any health risk assessment, and this study represents a screening of human health risk. It was intended to provide an indication of potential health risks that would require further investigation.

Arsenic is classified a human carcinogen by the EPA, and considered likely to cause skin, liver, bladder and lung cancer. Exposure to higher than average levels of arsenic occurs mostly in the workplace, near hazardous waste sites, or in areas with high natural arsenic levels in drinking or irrigation water. Ingesting very high levels of arsenic can result in death. Exposure to lower levels can cause nausea and vomiting, decreased production of red and white blood cells, abnormal heart rhythm, damage to blood vessels, and a sensation of 'pins and needles' in hands and feet. Studies in animals show that large doses of arsenic that cause illness in pregnant females can also cause low birth weight, foetal malformations, and even foetal death (ATSDR, 2007).

Benzo(a)pyrene is considered the most potent of the PAHs. For the public, the main route of exposure is from inhalation of smoke or ingestion of food. Chronic exposure in an occupational setting was followed by a reported decrease in lung function, as well as chest pain, respiratory irritation, cough, dermatitis and a depressed immune system, although in most cases it was not possible to evaluate the contribution of BaP to such effects. In animals, few adverse effects were observed in rats or hamsters exposed to BaP via inhalation, but myelotoxicity and hepatotoxicity were observed following ingestion. In mice, BaP has been shown to cross the placenta and cause adverse developmental and reproductive effects. Dietary administration during gestation reduced fertility and caused foetal abnormalities, whereas administration by gavage caused an increase in foetal death and decreased fertility (USEPA, 2007).

Polychlorinated biphenyls usually occur as mixtures of a possible 209 individual chlorinated compounds (known as congeners), primarily originating from anthropogenic sources. Health effects that have been associated with exposure to PCBs include neurobehavioural and immunological changes in children. Developmental delays occur at all ages and are greater in children smaller in size (ATSDR, 2000). PCBs are known to cause cancer in animals, and the EPA, the International Agency for Research on Cancer (IARC) and the WHO have classified PCBs as probable human carcinogens.

Chromium is known to be a human carcinogen. People are exposed to chromium by ingesting contaminated food or water, or breathing contaminated workplace air. Hexavalent chromium (VI) at high levels can damage the nose and cause cancer. Ingesting high levels of chromium (VI) may result in anaemia or damage to the stomach or intestines (ATSDR, 2008).

Selenium is needed at low doses to maintain good health, and inorganic selenium is not classified as a human carcinogen. However, exposure to high levels can cause adverse health effects. Short-term oral exposure to high concentrations of selenium may cause nausea, vomiting and diarrhoea, while chronic oral exposure to high concentrations of selenium compounds can produce a disease called selenosis. The major signs of selenosis are hair loss, nail brittleness, and neurological abnormalities such as numbness and other odd sensations in the extremities (ATSDR, 2003).

6. CONCERNS AND RECOMMENDATIONS

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6.1 AREAS OF CONCERN

Organic compounds

In general, the sediment levels of all compounds except PAHs were fairly low, but the distribution patterns indicated that industrial activities and combustion (pyrogenic) processes contribute to PAH pollution in the basin. The risk assessment suggested that, at the maximum level recorded, benzo(a)pyrene poses an unacceptably high cancer risk. The highest concentrations (and therefore the higher risks) were found at sites 16, 17, 22, 54, 60 and 64. The sources of the PAHs at all these sites were pyrogenic in nature, but the exact type of activity needs further investigation.

Dioxin-like TEQ and indicator PCBs were associated with industrial activities in Gauteng and possibly mining or residential combustion in North West Province. The relatively high levels in and close to Lesotho cannot be explained.

Organochlorine pesticides (OCPs) were generally at low levels at the sediment sites sampled, which is a positive finding. However, detectable OCP levels found at Blesbok Spruit, Suikerbosrand, Potchefstroom and Klerksdorp highlight the need for further monitoring.

The levels of organic compounds in biota did not reflect the levels in sediments. However, the results should be interpreted with caution, as the biota samples were not collected at sites that were sampled for sediments. The highest levels of dioxin-like TEQ, PCBs and PFOS were from isolated sites, far removed from industrial areas. The high levels of PFOS in fish at Boegoeberg and Rooipoort, and in bird eggs from Bloemhof Dam and Barbers Pan, suggest sources other than industry, possibly linked to unknown releases from agriculture (although PFOS should then be similar to OCP distribution), or unknown uses in mining in the drier, central parts of the country. The levels are quite high compared to European levels, so PFOS sources and environmental distribution demand closer scrutiny. Human consumption of PFOS via fish should be seen as a serious concern.

Elements

The elemental composition of sediments was difficult to associate with sediment source geology due to the very complex geology of the system, the huge drainage area covered, and the numerous tributaries and flood events.

The study has identified areas of concern where more in-depth assessments should be done to determine whether high levels of elements are due to natural factors or to disturbance, agricultural runoff, industry, urbanisation, mining, or a combination of these factors. Based on elemental analyses and risk assessment of sediments from 61 sites, the areas in the Orange–Senqu River basin that were deemed to be of concern are:

- Molopo Eye (site 56) due to Ga, Cr, Mn, Ni, Ag and Se.
- Vaal River at Schmidtsdrift (site 15) due to U.
- The areas associated with the Riet River and Koranna Spruit (sites 12, 14, 39, 41 and 44) due to a combination of higher than average levels of several elements.
- The Caledon and Malibamatso rivers draining into the Senqu and Orange rivers (sites 49, 50, 55, 57, 58 and 60) due to a combination of higher than average levels of several elements.
- Skoon Spruit (site 22) due to higher than average levels of Fe, Ni and other elements.
- Fish River, due to higher than average levels of As.

The risk assessment suggested that arsenic, chromium and selenium levels are potentially hazardous to human health in terms of toxic effects, while arsenic and beryllium may pose significant cancer risks to humans consuming bird eggs and fish. This aspect needs further investigation as the sites with high levels in eggs and fish were mostly far away from industry and may be related to local geology. The levels in bird eggs in particular need closer scrutiny, as very little data is available on the impacts of these elements on biota, or on rates of consumption by humans and other organisms. In fish, arsenic occurs predominantly in relatively non-toxic organic forms, such as arsenobetaine, rather than the highly toxic and carcinogenic inorganic arsenic (ATSDR, 2007).

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6.2 RECOMMENDATIONS

Organic compounds

Communities potentially exposed to hazardous levels of PAHs should be identified and the contributing pyrogenic sources investigated, so that interventions to reduce PAHs emissions can be proposed. Such interventions would also reduce releases of dioxins and PCBs.

There are likely more sites and communities potentially experiencing hazardous exposure to PAHs than revealed by this basin-wide survey. The project has identified both pyrogenic and petrogenic sources, and additional areas can now be identified by focusing on industrial hubs, mining sites and residential areas where these sources are known to occur.

The dynamics and sources of certain organic compounds (especially PFOS) in biota in isolated areas need further investigation to better understand the reasons for unexpectedly high concentrations, and to establish the associated risk to biota.

A monitoring programme should be instituted to track changes, and a selection of samples stored so that retrospective analyses can be conducted as new compounds are added to the Stockholm Convention.

Elements

Now that specific areas of concern have been identified, these should be investigated further to determine the sources and processes contributing to high levels of these elements in water and sediment, as well as their bioavailability.

Communities that may be exposed to higher than recommended levels of elements in water and water-associated food should be identified. Water used for irrigation, for instance, may contaminate produce.

Background levels of elements have not been collected on this scale previously, so the data collected should be curated to allow for future comparisons and trend monitoring.

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Site number/ name	Coordinates																				
Sediment mg/kg dw			Be	B	Al	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Rb	Sr
1	27.099	-26.683	0.35	67.5	3750	2.425	62.5	17.75	57.5	475	7500	9	25	12	32.5	0.825	0.5	2.75	3.75	1.7	5.5
2	21.242	-28.467	0.35	32.5	14000	2.75	800	40	22.75	237.5	15750	12.5	42.5	24.5	37.5	2.5	1.25	3.25	2.075	2.75	27.5
3	20.642	-28.858	0.4	25	5500	1.95	222.5	15.75	15.75	152.5	6750	3.5	14	9.75	25	1.4	0.55	2.75	1.675	6.25	10.5
4	20.154	-28.499	0.3	19.5	10500	2.325	700	32.5	18.5	177.5	12500	9.25	30	24.75	35	2.125	1.025	3	1.775	4.25	15.75
5	19.303	-28.737	0.235	16	6500	1.925	450	30	17	152.5	10750	6	20.25	13	25	1.5	0.9	2.475	1.775	4.5	10.5
6	17.608	-28.749	0.275	13.5	10000	2.175	575	27.5	20.25	170	12000	9	30	30	37.5	1.95	1	3.25	1.65	3.5	16.75
7	16.471	-28.602	0.215	11	8000	1.5	425	23.5	12.5	152.5	9500	6.25	22.75	25	27.5	1.65	0.775	3.25	1.525	2.025	16
8	16.890	-28.123	0.225	9	3500	1.075	242.5	22	10.5	105	7000	3.5	14.5	8.75	25	1.075	0.575	3.75	1.35	2.425	10.75
9	22.748	-29.659	0.178	6.25	6000	1.475	450	30	16.75	182.5	9000	7	25	15.75	25	1.275	0.775	2.5	1.225	1.775	10.25
10	23.016	-29.623	0.35	4.5	7750	2.5	172.5	19.75	25	192.5	8250	4.5	21.25	11.5	27.5	1.5	0.7	3.75	0.65	4.5	19
11	23.699	-29.162	0.575	20.25	23250	6	925	62.5	50	325	24000	19.5	70	42.5	57.5	6	2	4.25	11.5	6.75	27.5
12	23.841	-29.039	1.225	21.5	27500	8.25	425	67.5	95	1000	30000	25	112.5	42.5	75	8.25	2.5	8.5	12.75	16.75	30
13	24.111	-29.601	0.6	18.25	25000	5.75	1225	87.5	47.5	500	27500	21	80	45	65	7.5	2.25	5.25	11.5	5.5	47.5
14	24.503	-29.042	1.675	22	37500	11	725	75	105	875	35000	22	105	60	97.5	12	2.75	8.25	13.75	21.25	37.5
15	24.067	-28.722	1.125	17	20750	7.5	300	55	85	725	23250	22	97.5	42.5	60	7.5	2	6.75	12.25	13.5	40
16	24.303	-28.377	0.85	18	21500	6.25	250	52.5	82.5	775	21500	18	105	30	47.5	7	1.7	5.75	10.75	9.25	70
17	24.867	-28.095	1.375	14	24750	8.25	450	52.5	72.5	825	25000	25	87.5	47.5	70	11	2.125	6.5	13.25	18.25	25
18	26.127	-27.936	0.425	12.5	7500	2.15	170	20	30	120	7250	7.25	22.5	15.25	27.5	3.25	0.675	3.5	11.75	6.5	14.25
19	26.216	-27.516	1	15.5	24500	7.5	275	47.5	85	900	21000	24	100	40	80	7.5	1.775	5.5	12.25	17	16
20	26.813	-27.487	0.4	10.5	15750	2.5	275	55	77.5	650	16750	20.5	67.5	65	40	4.75	1.4	7.25	11.5	4.5	23.25
21	26.998	-27.040	0.475	11.5	10000	1.975	227.5	40	90	350	11500	21.5	67.5	22.5	27.5	3.5	1	3.5	11.25	6.25	9
22	26.664	-26.935	0.8	13.75	16250	5.5	227.5	65	152.5	925	40000	32.5	172.5	47.5	122.5	7.5	3.25	11.75	11.75	11	16.25
23	26.680	-27.014	0.275	11.5	5250	1.975	120	24.25	45	400	7250	15	55	23	35	2.5	0.65	3.5	11	4.25	7.75
24	27.397	-26.914	0.208	10	3500	1.275	125	18.25	35	300	5000	10.75	37.5	18	32.5	2.1	0.475	2.75	10.75	3.25	6.25
25	28.064	-26.549	0.575	10.75	11250	4.75	142.5	40	82.5	1375	20500	52.5	200	45	275	4	1.7	4	11.5	5.5	12.5
26	28.381	-26.647	0.975	9	20500	11	247.5	85	112.5	625	30000	32.5	147.5	65	42.5	7.75	2.5	4	12.25	9	14.5
27	28.426	-26.479	0.525	9.25	8000	3	50	25	57.5	1075	11250	40	160	32.5	300	2.75	0.95	4.75	11.25	3.5	6.75
28	29.025	-26.629	0.275	9	7000	2.25	185	55	57.5	1050	11500	32.5	87.5	30	47.5	2.5	1	3.25	10.75	3.25	8.75
29	30.026	-26.313	1.3	8.75	21500	9.75	575	152.5	67.5	325	32500	30	32.5	92.5	45	6.75	2.5	5	11.75	11	7.75
30	29.600	-27.470	0.5	8	3750	1.45	90	25	25	775	12000	12	16.75	17	22	3.5	1.05	5.25	11	3.25	5.5
31	28.475	-27.531	1.525	7	27500	11.75	400	75	95	1225	27500	25	80	52.5	57.5	12.5	2.4	4.25	12.5	17	25
32	28.750	-27.647	0.625	8	19250	3.5	1050	60	40	500	27500	21	60	45	50	7	2.325	9	12.5	3.5	30
33	28.534	-27.310	2.275	8	37500	14.25	450	77.5	92.5	1975	37500	30	85	57.5	110	19	3.25	8.25	13.5	25	40
34	28.598	-27.023	1.6	7.5	27500	12	275	67.5	105	900	27500	30	110	57.5	92.5	12.75	2.5	5.25	12.5	21.75	27.5
35	29.105	-27.037	1.075	7	21250	9	245	57.5	87.5	725	20250	23.25	75	42.5	52.5	8.25	1.8	4.25	11.75	15.25	16
36	28.914	-26.846	0.675	8.75	17000	7	230	50	65	750	16250	20.75	60	40	47.5	6.25	1.4	3.25	13	11	14.75
37	28.050	-26.682	1.05	8.5	17250	9.75	182.5	67.5	95	1175	30000	55	187.5	57.5	185	7	2.5	6.5	12	10	19.25
39	26.107	-28.807	1.575	16.75	27500	9.25	350	55	90	1700	27500	21.5	95	50	120	15.25	2.475	8.25	11.75	19	52.5
40	25.773	-28.945	0.575	16.25	25000	6.25	350	72.5	62.5	525	30000	21.25	167.5	77.5	75	7.25	2.5	6	12.25	8	155
41	26.636	-29.086	1.3	9.25	25000	6.75	600	102.5	90	1150	32500	25	87.5	42.5	70	11.5	2.75	16.5	12.25	14.25	42.5
42	26.525	-29.101	1	10.5	23750	6.25	750	62.5	72.5	825	24000	24.75	72.5	35	60	8.25	2.125	8	12	14.75	30
43	25.202	-29.474	2.3	17.25	45000	10.75	1150	115	85	800	52500	23	110	62.5	147.5	15.25	4.5	17	12.75	27.5	77.5
44	25.587	-29.681	1.35	15.75	37500	10	1400	82.5	87.5	600	32500	19.5	75	42.5	105	11.5	2.75	10.25	12.75	21.25	72.5
45	25.649	-29.563	0.8	12	21500	5.25	725	60	47.5	350	22250	14.5	50	45	60	6.75	2	7.25	12	12.75	55
46	24.963	-30.535	1.425	11.25	37500	14.25	1000	110	167.5	275	37500	21.25	95	55	105	7.5	3.25	5.25	11.75	15.5	55
47	25.242	-30.507	1.125	8.5	32500	10.25	1025	75	77.5	1175	32500	21	85	47.5	80	9.5	2.75	6	12.75	14	40
48	26.306	-30.428	0.625	6.5	19000	4.25	1175	67.5	35	800	25000	23.5	72.5	45	50	8.5	2.325	7.25	11.75	4.5	30
49	26.459	-30.575	1.325	10	50000	14.5	1150	92.5	97.5	875	42500	27.5	117.5	77.5	85	12	3.5	6.5	12	18	40
50	26.465	-30.650	1.6	9.25	27500	6.5	550	85	60	1425	40000	25	72.5	37.5	90	12.5	3.25	12	13	16	80
51	26.711	-30.684	0.55	13	21750	5	875	60	37.5	275	22500	14	57.5	37.5	47.5	5.25	1.975	4	11.5	8.25	27.5
52	27.337	-30.405	0.375	6.25	19750	4.75	1500	85	30	525	25000	19	65	45	50	5.25	2.275	4.25	12.25	3.75	27.5
53	27.137	-29.528	1.325	6.5	19750	5.75	350	97.5	117.5	850	35000	23	72.5	50	72.5	8	3	10.5	12	9.5	27.5
54	27.316	-29.490	0.275	5.75	8250	2.5	425	35	24.25	195	11500	8.75	30	22.75	27.5	3	1.05	3.25	11.5	3.5	8.75
55	28.151	-28.724	1.175	7	42500	12.75	1600	135	140	2400	52500	47.5	135	75	70	16.25	4.5	9.25	12	6	57.5
56	25.887	-25.887	3	8.75	13500	8.5	350	207.5	450	60000	45000	117.5	250	80	55	300	4	19.25	12.75	4	80
57	28.551	-29.020	1.05	14	35000	7.75	1250	87.5	62.5	2500	37500	23.75	100	67.5	207.5	11.75	3	6.75	11.75	10.5	82.5
58	28.563	-29.230	0.6	6.75	72500	21.25	4250	205	112.5	1325	72500	45	147.5	137.5	127.5	12	6	2.75	12.5	5.25	120
59	28.148	-29.553	0.425	5	57500	16	2500	170	97.5	1025	57500	40	150	117.5	105	8.25	4.75	2.75	12	3	75
60	27.454	-29.336	0.525	5.5	62500	13.25	4000	175	70	1125	62500	37.5	175	137.5	125	10	5.25	2.5	12.25	4	92.5
61	28.510	-30.062	0.7	7.75	50000	15.5	2000	117.5	92.5	950	47500	32.5	145	102.5	87.5	8	4	3.25	12.25	8.25	50
62	17.79	-26.8	0.525	7	8750	2.3	250	32.5	18.25	425	18500	9	32.5	42.5	47.5	4.75	1.625	13.75	11.75	5.25	24
Fish	mg/kg dw																				
V1	29.51	-26.86	0.00	3.75	15.00	0.09	25.63	1.20	3.63	1.14	62.50	0.06	0.99	2.76	20.50	0.07	0.02	1.26	4.50	4.25	0.70
V2	27.40	-26.92	0.01	3.25	4.88	0.09	22.00	1.21	3.75	0.94	50.00	0.08	0.26	1.61	19.50	0.07	0.02	1.33	2.33	12.00	0.58
V3	24.16	-28.56	0.01	3.50	6.00	0.09	24.13	1.16	3.38	0.93	53.75	0.05	0.30	2.10	20.38	0.07	0.02	1.13	5.38	15.00	0.79
O2	22.21	-29.21	0.01	3.38	10.38	0.08	23.50	1.11	3.50	0.96	52.50	0.03	0.33	2.91	20.50	0.07	0.02	1.41	4.13	6.25	0.88
Bird eggs	mg/kg dw																				
Barberspan	25.53	-26.59	0.153	115	16	0.23	7.5	2.95	9	4.25	230	0.35	2.05	8.5	70	0.095	0.039	2.25	10	2.4	2.95
Bloemhof Dam	25.67	-27.69	0.083	65	16.664	0.244	8.5	2.95	9	3.749	230	0.125	1.525	9.15	75	0.115	0.045	2.562	6.71	2.604	3.489
Eldorado Park	27.89	-26.3	0.095	40	21	0.195	5	2.2	7	3.75	205	0.205	1.25	10	65	0.11	0.039	1.8	5.5	5	2.9
Potchefstroom	27.19	-26.7	0.096	34.05	14	0.136	6	2.95	9.5	5	207.05	0.23	1.15	6.5	75	0.125	0.042	2.25	7	10.1	3.6

Mo	Rh	Pd	Ag	Cd	Sn	Cs	Ba	Ce	Pt	Au	Hg	Tl	Pb	U	I	MPI	Total PAHs	Dioxin TEQ	Indic PCBs	Sum OCPs	PFOS
																	mg/kg dw	ng/kg dw	ng/kg dw	ng/kg dw	µg/kg dw
1.65	0.245	1.325	4.25	0.153	0.975	0.173	23.5	4.75	0.148	2.3	3.5	1.825	5.25	0.65	0.85	8.681	0.290	0.17	0	8	0
0.725	0.75	0.85	4	0.093	0.6	0.325	90	7.5	0.173	0.675	1.525	0.5	5.75	0.475	0.4	10.598	0.046	0.16	0	0	0
0.425	0.218	0.475	2.5	0.055	0.675	0.55	40	8.75	0.015	0.35	1	0.3	4	1.5	1.1	6.841	0.035	0.16	0	0	0
0.35	0.4	0.475	2.375	0.07	0.65	0.425	62.5	9.75	0.018	0.213	0.9	0.275	4.25	1.5	0.3	8.230	0.021	0.16	0	0	0
0.233	0.12	0.3	2.3	0.048	0.675	0.325	40	11.5	0.105	0.18	0.65	0.21	3.5	0.8	0.525	6.790	0.050	0.16	0	0	0
0.235	2.15	0.325	1.7	0.053	0.375	0.4	55	7.5	0.168	0.108	0.65	0.195	4.25	1.275	0.1675	7.846	0.014	0.16	0	0	0
0.223	0.275	0.3	2	0.07	0.275	0.225	55	7	0.138	0.095	0.575	0.143	4.25	1.025	0.1725	6.313	0.009	0.16	0	0	0
0.22	0.148	0.218	2.45	0.038	0.75	0.3	37.5	6.5	0.04	0.128	0.4	0.108	4.75	0.45	0.14	4.936	0.017	0.16	0	0	0
0.138	0.13	0.17	1.5	0.035	0.245	0.228	42.5	5.5	0.010	0.063	0.3	0.09	3.5	0.3	0.1475	4.655	0.012	0.16	0	0	0
0.108	0.158	0.168	1	0.04	0.3	0.5	45	5	0.008	0.04	0.213	0.1	5	0.4	1.3	5.224	0.029	0.16	0	0	0
0.325	0.425	0.475	4.75	0.108	0.85	0.8	117.5	14.25	0.014	0.098	0.575	0.233	8.5	0.825	1.175	13.176	0.020	0.16	0	0	0
0.45	0.5	0.525	5	0.168	1.05	1.475	157.5	19.75	0.015	0.128	0.575	0.35	13.5	1.5	6	18.478	0.043	0.19	0	0	0
0.375	0.575	0.525	9	0.108	1.425	0.7	150	13	0.019	0.08	0.5	0.2	8.75	0.575	0.9	14.704	0.116	0.17	0	0	0
0.35	0.5	0.575	6.5	0.155	1.575	1.8	202.5	25	0.014	0.105	0.575	0.4	17.5	2.225	25	22.225	0.117	0.19	0	0	0
0.55	0.45	0.475	14.75	0.118	0.975	1.025	155	17.5	0.025	0.09	0.525	0.275	12.5	50	6.75	19.605	0.047	0.21	0	0	0
0.3	0.725	0.5	3.75	0.128	0.725	0.8	160	12.5	0.006	0.088	0.475	0.218	7.75	0.925	6.25	15.444	0.365	0.17	0	0	0
0.325	0.35	0.475	6	0.125	1.35	1.375	212.5	21.75	0.175	0.2	0.5	0.35	17	2	5.75	18.586	0.179	0.22	0	0	0
0.173	0.175	0.25	2.75	0.045	0.45	0.625	75	12.75	0.103	0.06	0.375	0.18	6.25	1.325	0.875	7.575	0.047	0.16	0	0	0
0.325	0.24	0.35	4.75	0.155	1.15	1.225	150	19.5	0.014	0.098	0.4	0.3	12.5	1.7	2.75	14.887	0.083	0.27	244	0	0
0.3	0.275	1.825	1.95	0.08	0.375	0.375	110	10	0.125	0.4	0.95	0.25	5.75	4.75	0.525	12.373	0.015	0.16	0	0	0
0.225	0.218	0.215	2.475	0.063	0.475	0.55	77.5	9.5	0.014	0.06	0.325	0.213	5.75	0.625	0.55	8.193	0.058	0.16	0	0	0
0.825	0.35	0.325	10.25	0.85	1.6	0.525	147.5	17.25	0.014	0.5	0.975	0.205	18.25	1.675	3.5	17.814	0.867	0.54	1702	1.7	0
0.375	0.13	0.245	3	0.063	0.525	0.35	57.5	6.5	0.017	0.105	0.375	0.128	4.75	0.975	1.1	7.067	0.052	0.22	2828	0	0
0.138	0.158	0.148	2.75	0.045	0.35	0.3	50	6.5	0.013	0.055	0.35	0.115	3.75	0.375	0.575	5.336	0.067	0.17	0	0	0
0.325	0.4	0.235	12.25	0.45	2.25	0.45	80	8.75	0.011	0.08	0.35	0.173	14.75	1.15	1.6	12.831	0.207	0.41	1807	0.7	0
0.25	0.25	0.325	4.25	0.05	0.725	0.475	152.5	15.25	0.023	0.05	0.375	0.2	11.75	0.775	1.7	12.361	0.059	0.2	0	0	0
0.195	0.218	0.24	5.25	0.4	0.625	0.875	60	8	0.16	0.103	0.325	0.115	10	2.3	1.575	10.012	0.345	0.32	1389	8.5	0
0.205	0.148	0.215	3.5	0.05	0.35	0.233	60	9.75	0.009	0.048	0.25	0.133	3.5	0.5	0.45	7.187	0.242	0.22	0	0	0
0.725	0.25	0.25	6.25	0.053	0.725	0.525	115	25	0.03	0.06	0.425	0.325	23.25	1.175	1.125	11.969	0.117	0.24	0	0	0
0.325	0.103	0.135	2.75	0.038	0.25	0.275	97.5	8.5	0.007	0.15	0.3	0.12	6	0.375	0.3	5.725	0.035	0.16	0	0	0
0.3	0.325	0.4	5.75	0.1	0.95	1.3	227.5	25	0.009	0.038	0.375	0.325	16.75	1.175	1.3	14.961	0.074	0.2	0	0	0
0.35	0.45	0.3	3.25	0.08	0.5	0.325	167.5	13	0.012	0.03	0.275	0.11	9.75	0.525	0.4	10.689	0.015	0.16	0	0	0
0.55	0.45	0.625	8.5	0.223	1.625	2.075	350	37.5	0.035	0.06	0.3	0.425	27.5	2.15	2.75	21.523	0.117	0.34	0	0	0
0.325	0.45	0.5	5.25	0.153	1.3	1.65	245	30	0.008	0.068	0.275	0.375	17.5	1.925	1.825	16.864	0.074	0.26	0	0	0
0.195	0.213	0.325	3.5	0.078	0.85	1.1	182.5	24.25	0.03	0.043	0.25	0.25	14.25	1.1	0.75	12.331	0.068	0.17	0	0	0
0.275	0.235	0.275	3.75	0.055	0.675	0.575	135	13.75	0.003	0.063	0.275	0.19	7.25	0.8	0.8	10.063	0.454	0.2	0	0	0
0.3	0.3	0.4	21.75	0.208	1.125	1.675	150	17	0.038	0.125	1.6	0.193	15.5	1.975	3.75	17.551	0.225	0.69	531	2.8	0
0.475	0.5	0.5	8.75	0.143	1.775	1.7	300	22.25	0.012	0.068	0.4	0.3	21.5	1.15	20.75	21.302	0.174	0.52	953	0.5	0
0.35	1.225	0.65	3	0.115	0.775	1.125	165	8.5	0.014	0.07	0.325	0.3	6.25	0.875	6	18.199	0.050	0.16	0	0	0
0.55	0.6	0.5	12.75	0.11	1.075	1.45	227.5	27.5	0.010	0.043	0.4	0.245	19	1.225	2.125	17.896	0.012	0.17	0	0	0
0.275	0.35	0.4	4.25	0.098	1.05	1.4	177.5	23.75	0.010	0.145	0.238	0.245	15.5	1.325	1.4	14.817	0.016	0.17	0	0	0
0.425	0.675	0.55	4.5	0.163	1.625	3.5	250	30	0.013	0.048	0.22	0.45	24.25	1.825	5.5	24.597	0.043	0.19	0	0	0
0.325	0.625	0.55	5.25	0.12	1.55	1.975	210	25	0.088	0.14	0.21	0.275	17.25	1.625	1.925	20.465	0.016	0.16	0	0	0
0.375	0.525	0.375	11.25	0.12	0.925	1.3	145	18.5	0.014	0.043	0.208	0.22	11	1.275	1.925	14.412	0.024	0.17	0	0	0
0.218	0.5	0.45	8	0.123	1.35	2.075	100	20.25	0.021	0.035	0.213	0.275	16.25	1.025	1.55	17.798	0.011	0.16	0	0	0
0.375	0.45	0.4	8.25	0.118	1.125	1.625	195	24	0.013	0.038	0.228	0.235	13.75	1.5	0.925	16.427	0.060	0.17	0	0	0
0.4	0.6	0.525	3.5	0.085	0.625	0.5	200	18.75	0.020	1	0.203	0.113	11	0.65	0.325	12.827	0.024	0.16	0	0.5	0
0.3	0.425	0.425	4.75	0.138	1.15	2.15	210	25	0.038	0.058	0.25	0.248	15	1.2	1.675	18.908	0.044	0.16	0	0	0
0.4	0.675	0.6	2.75	0.3	1.15	1.9	250	27.5	0.011	0.035	0.175	0.25	24.5	2.225	0.775	18.400	0.013	0.17	0	0	0
0.228	0.325	0.25	3.5	0.07	0.65	1.075	97.5	22.25	0.053	0.03	0.155	0.138	7.75	0.95	0.45	11.379	0.013	0.17	0	0	0
0.3	0.245	0.25	4	0.073	0.55	0.5	105	15	0.010	0.03	0.173	0.088	5.75	0.55	0.25	9.796	0.022	0.16	0	0	0
0.5	0.35	0.375	4.75	0.098	0.775	0.975	192.5	22	0.010	0.035	0.195	0.175	17.25	1.375	0.85	13.895	0.008	0.32	0	0	0
0.175	0.11	0.148	3.75	0.038	0.35	0.375	67.5	13.75	0.008	0.025	0.178	0.078	4.75	0.525	0.25	6.000	0.673	0.18	0	0	0
0.55	0.625	0.425	4.25	0.12	0.7	0.525	325	20.25	0.007	0.028	0.208	0.168	15.5	0.9	1.25	17.163	0.039	0.17	0	0	0
4	1.25	0.475	23	0.188	1	0.3	8500	170	0.175	0.03	0.185	3.25	80	1.575	1.475	33.944	0.179	0.19	2158	0	0
1.2	0.675	0.45	5.25	0.188	2.75	1.225	240	17.5	0.083	0.03	0.188	0.208	16.75	1.05	0.975	20.309	0.037	0.17	0	0	0
0.475	0.85	0.675	9	0.178	0.9	0.22	152.5	13.25	0.03	0.03	0.3	0.098	4.5	0.375	1.8	18.452	0.038	0.17	0	0	0
0.425	0.55	0.5	7	0.133	0.7	0.155	102.5	11	0.018	0.028	0.2	0.093	3.5	0.325	1.075	14.585	0.055	0.19	0	0.5	0
0.45	0.825	0.525	5.25	0.145	0.9	0.18	127.5	11.75	0.020	0.028	0.165	0.083	4.25	0.35	1.1	15.786	0.711	0.59	1204	0.7	0
0.3	0.5	0.4	5	0.143	0.875	0.825	110	14.25	0.015	0.028	0.178	0.145	7	0.375	1.475	15.705	0.033	0.18	0	0	0
1.15	0.3	0.3	1.875	0.115	0.6	0.75	125	15.5	0.003	0.023	0.135	0.11	10.25	0.775	0.35	9.228	0.006	0.17	0	0	0

																		ng/kg ww	ng/kg ww	ng/kg ww	µg/kg ww
0.10	0.63	0.12	4.11	0.02	0.75	0.02	5.75	0.03	0.17	0.10		0.08	0.44	0.04				0.16	0	1.6	0
0.11	0.13	0.08	5.25	0.01	0.79	0.03	6.38	0.01	0.14	0.09		0.12	0.25	0.01				0.16	454	18.9	0
0.10	0.21	0.11	1.28	0.01	0.66	0.21	6.50	0.02	0.07	0.16		0.10	0.34	0.10				0.23	5409	8.2	1.7
0.08	0.13	0.09	3.99	0.01	0.65	0.01	6.25	0.01	0.30	0.07		0.10	0.34	0.07				0.16	0	1.4	2.6

																		ng/kg ww	ng/kg ww	ng/kg ww	µg/kg ww
2.6	0.6	6.5		0.148	235	0.003	12.5	0.022	2.95	70	7	8.5	1.6	2.9				0.2	6077	90.8	846
2.604	0.125	2.2		0.072	280.6	0.005	13.019	0.015	0.6	19.5	9.5	1.95	0.65	0.55				1.61	87646	95	2330
1.55	0.125	1.1		0.038	240	0.005	13.5	0.09	0.36	6.5	2.05	0.6	0.6	0.21				0.44	32627	155.7	69
3.1	0.5	1.8		0.021	255.375	0.006	17	0.018	0.33	3.632	3.235	0.312	0.55	0.091				0.32	44941	153.6	201

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