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Assessment of Metals Levels and their Potential Risks in Local Brands of Honey from Bayelsa State, Nigeria

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ABSTRACT: This study provides information on the levels and potential risks of six metals (Cd, Pb, Ni, Mn, Zn and Fe) in some local brands of honey in Bayelsa State. The honey samples were digested using hydrochloric acid, nitric acid and hydrogen peroxide mixture and their metal levels were quantified by atomic absorption spectrometry (AAS). The concentration of Cd, Pb, Ni, Cu, Mn, Zn and Fe ranged from 0.226 to 0.294 mg kg^{-1}, 0.14 to 1.601 mg kg^{-1}, not detected to 0.299 mg kg^{-1}, not detected to 0.248 mg kg^{-1}, 0.055 to 0.420 mg kg^{-1}, 0.094 to 0.218 mg kg^{-1} and 1.721 to 6.072 mg kg^{-1} respectively. The estimated daily intakes of the metals in the honeys were lower than their respective tolerable daily intakes. The THQ values for the individual metals and ∑THQ were < 1 which indicates that there is no safety concern for consumers of these honey samples.

Keywords: Target hazard quotients, Honey, Bayelsa State, Metals

Introduction

Honey is an unfermented, natural sweet substance produced by *Apis melifera* bees from the nectar of plants or from the secretions of living parts of plants or excretions of plant-sucking insects on the living parts of plants, which they collect, transform and combine with given matters of their own, deposit, dehydrate, store and leave in honeycombs to ripen and mature (European Commission (EC) Directive 2001/110; Codex Alimentarius Commission, 1983/84). Honey can be multi-floral or mono-floral depending on its sources (Ahmida *et al*., 2013). Honey comprised 65 % fructose and glucose, 18 % water and minerals which constituted 0.04 % for pale honey and 0.2 % for darker honey (Bogdanov *et al*., 2007). The mineral composition of honey is influenced by beekeeping practices, environmental conditions, climate, geographical origin and plant species (Nkansah *et al*., 2018). Honey has various nutritional, medicinal and preventive properties contributed by its chemical constituents. Despite its several health benefits, the occurrence of contaminants such as heavy metals in honey may pose a health risk to consumers because certain heavy metals are known to cause cancer, neurological and developmental and reproductive disorders, renal, kidney and cardiovascular problems (Iwegbue *et al*., 2015).
Honey is contaminated by heavy metals in two ways: the environment where bees collect honey and the beekeeping method. The foraging area of bees is approximately 7 km^2 and includes various environments, plants and foods (Sereviciene *et al*., 2022). When going from flower to flower and foraging for nectar, honeydew, and pollen, bees also come in contact with air, water, soil, leaves and branches of plants (Sereviciene *et al*., 2022; Solayman *et al*., 2021). Thus, honey is the result of a bio-accumulation process that is useful for collecting information about the environment and may be considered a bio-indicator of environmental pollution (Dobrinas *et al*., 2008; Hammel *et al*., 2008; Fredes and Montenegro, 2006; Bratu and Georgescu, 2005). Heavy metals determination in honey using several methods has been the focus of many studies worldwide. In Nigeria, some studies have also been carried out to determine the levels of heavy metals in honey (Lekduhur *et al*., 2021; Iwegbue *et al*., 2015; Ajao *et al*., 2013; Agbagwa *et al*., 2011; Achudume and Nwafor, 2010). However, there is a need for continuous monitoring as food safety is of great importance in health policy and providing the best

possible quality of food will protect public health and preserve consumer confidence. Thus, the goals of this study are to quantify the levels and risks of heavy metals in honey samples from Bayelsa State.

Materials and methods

Sample collection: Eight samples of bottled honey were collected from different superstores across Bayelsa State, Nigeria. Samples were collected in glass bottles and stored at 4 °C in the dark prior to analysis.
Reagents: The reagents used were analar grades, hydrochloric (HCl) and nitric (HNO_3) acids and hydrogen peroxides (H_2O_2). Working standard solutions of the heavy metals were prepared from their stock solutions.
Sample digestion: The method of Iwegbue et al. (2015) was adopted. 1 g of honey was weighed into a crucible and 20 mL of HCl, HNO_3 and H_2O_2 mixture (3:1:1 v/v) was added. The crucible was covered and digested. At the end of digestion, the solution was cooled and filtered and made to 25 mL mark with 0.25 mol L^{-1} nitric acid. The levels of heavy metals in the digested samples were determined with AAS (GBC XplorAA).
Quality control and statistical analysis: The quality control methods used include blank determination, spike recoveries method and use of high purity reagents. The levels of metals in blank samples were below the detection limit. The % recovery of the heavy metals varied from 95.7 to 99.2 %. Analysis of variance was utilized to check if the levels of heavy metals in the honeys varied significantly at $p = 0.05$ level of significance.
Assessment of estimated dietary intake (EDI): The EDI of metals from the honey ingestion was assessed with equation 1 (Tesi *et al*., 2020):

$$EDI\ (\mu g/kg\ bw/day) = \frac{Ingestion\ Rate \times Concentration\ of\ metals\ in\ honey}{Body\ Weight} \quad (1)$$

The ingestion rate of 1.4 g/day was used based on the per capita consumption of 0.5 kg per annum per person (Iwegbue *et al*., 2015). The body weights used was 60 kg (adults) and 15 kg (children).
Evaluation of target hazard quotients (THQ): The THQ was employed to assess the degree of concern ensuing from the ingestion of metals in honeys. The THQ was evaluated with equation 2 (Tesi *et al*., 2020).

$$THQ = \frac{EF \times ED \times EDI}{RFD \times AT} \times 10^{-3} \quad (2)$$

where: EF is exposure frequency,
ED is exposure duration,
AT is averaging time for non-carcinogens and
RFD is the oral reference dose.

The values of these variables were adopted from Tesi *et al*. (2020) and USDOE (2011). A THQ value less than unity suggests no health concern whereas greater than unity suggests health concern (Iwegbue *et al*., 2015). Because of the synergistic effects of metals, the THQ values of the specific metals were summed up to obtain the total THQ (∑THQ) which also has the same interpretation as THQ.

Results

The results of the heavy metals levels in the honey samples are displayed in Figure 1 while the heavy metals levels in honey from this study in comparison with others in literature are shown in Table 1. The EDI and THQs of the heavy metals from honey ingestion are given in Tables 2 and 3 respectively.

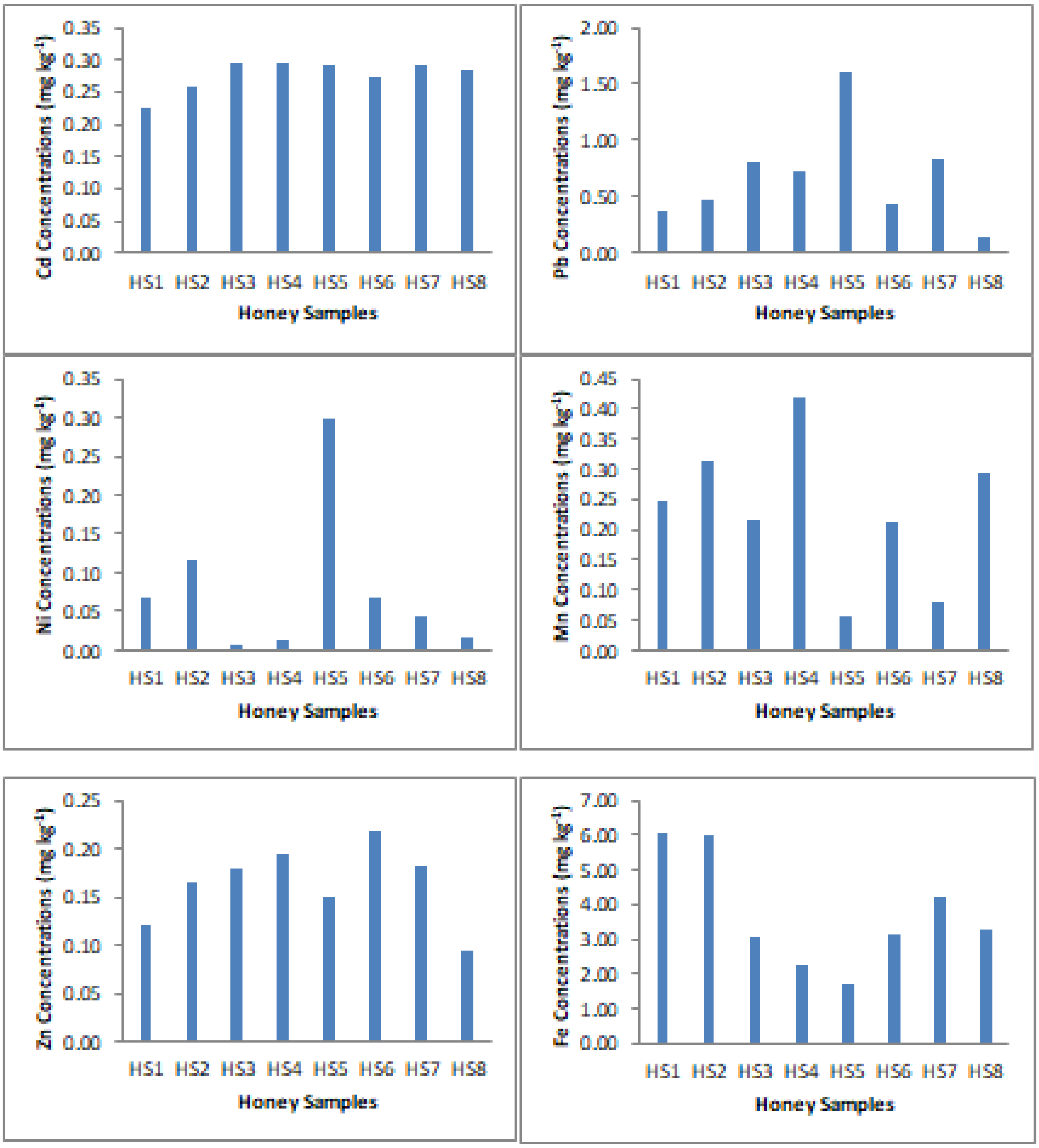

Figure 1: Heavy metals levels (mg kg^{-1}) in honeys

From Figure 1, the levels of Cd, Pb, Ni, Cu, Mn, Zn and Fe ranged from 0.226 to 0.294 mg kg^{-1}, 0.14 to 1.601 mg kg^{-1}, not detected to 0.299 mg kg^{-1}, not detected to 0.248 mg kg^{-1}, 0.055 to 0.420 mg kg^{-1}, 0.094 to 0.218 mg kg^{-1} and 1.721 to 6.072 mg kg^{-1} respectively.

Table 1: Computed EDI (μg kg^{-1} bw day^{-1}) of metals in the honeys

Category	Honeys	Cd	Pb	Ni	Mn	Zn	Fe
Child	HS1	0.02	0.03	0.006	0.02	0.01	0.57
	HS2	0.02	0.04	0.011	0.03	0.02	0.56
	HS3	0.03	0.08	0.001	0.02	0.02	0.29
	HS4	0.03	0.07	0.001	0.04	0.02	0.21
	HS5	0.03	0.15	0.028	0.01	0.01	0.16
	HS6	0.03	0.04	0.006	0.02	0.02	0.29
	HS7	0.03	0.08	0.004	0.01	0.02	0.39
	HS8	0.03	0.01	0.002	0.03	0.01	0.31
Adult	HS1	0.005	0.008	0.002	0.006	0.003	0.14
	HS2	0.006	0.011	0.003	0.007	0.004	0.14
	HS3	0.007	0.019	0.0002	0.005	0.004	0.07
	HS4	0.007	0.017	0.0003	0.010	0.005	0.05
	HS5	0.007	0.037	0.007	0.001	0.004	0.04
	HS6	0.006	0.010	0.002	0.005	0.005	0.07
	HS7	0.007	0.019	0.001	0.002	0.004	0.10
	HS8	0.007	0.003	0.0004	0.007	0.002	0.08

From the Table 1, the EDI in μg kg^{-1} bw day^{-1} of Cd, Pb, Ni, Mn, Mn, Zn and Fe ranged from 0.02 to 0.03, 0.01 to 0.15, 0.001 to 0.028, 0.01 to 0.04, 0.01 to 0.02, and 0.16 to 0.57 for children ingestion. For the honey ingestion by adults, the EDI in μg kg^{-1} bw day^{-1} ranged from 0.005 to 0.007, 0.003 to 0.037, 0.0002 to 0.002, 0.001 to 0.01, 0.002 to 0.005 and 0.04 to 0.14 for Cd, Pb, Ni, Mn, Zn and Fe respectively.

Table 2: Target hazard quotients (THQs) of metals in the honeys

		Target Hazard Quotients (THQs) of individual metals						
		Cd	Pb	Ni	Mn	Zn	Fe	∑THQs
Child	HS1	2.11×10^{-2}	9.65×10^{-3}	3.13×10^{-4}	1.66×10^{-4}	3.80×10^{-5}	8.10×10^{-4}	3.21×10^{-2}
	HS2	2.41×10^{-2}	1.28×10^{-2}	5.41×10^{-4}	2.09×10^{-4}	5.13×10^{-5}	7.95×10^{-4}	3.85×10^{-2}
	HS3	2.74×10^{-2}	2.17×10^{-2}	3.27×10^{-5}	1.43×10^{-4}	5.60×10^{-5}	4.11×10^{-4}	5.02×10^{-2}
	HS4	2.74×10^{-2}	1.94×10^{-2}	6.07×10^{-5}	2.80×10^{-4}	6.00×10^{-5}	3.03×10^{-4}	4.75×10^{-2}
	HS5	2.71×10^{-2}	4.27×10^{-2}	1.40×10^{-3}	3.67×10^{-5}	4.70×10^{-5}	2.29×10^{-4}	7.20×10^{-2}
	HS6	2.54×10^{-2}	1.17×10^{-2}	3.13×10^{-4}	1.41×10^{-4}	6.78×10^{-5}	4.21×10^{-4}	3.77×10^{-2}
	HS7	2.71×10^{-2}	2.21×10^{-2}	2.10×10^{-4}	5.27×10^{-5}	5.63×10^{-5}	5.64×10^{-4}	5.01×10^{-2}
	HS8	2.67×10^{-2}	3.73×10^{-3}	7.93×10^{-5}	1.97×10^{-4}	2.92×10^{-5}	4.36×10^{-4}	3.12×10^{-2}
Adult	HS1	5.27×10^{-3}	2.41×10^{-3}	7.82×10^{-5}	4.15×10^{-5}	9.49×10^{-6}	2.02×10^{-4}	8.02×10^{-3}
	HS2	6.02×10^{-3}	3.19×10^{-3}	1.35×10^{-4}	5.23×10^{-5}	1.28×10^{-5}	1.99×10^{-4}	9.61×10^{-3}
	HS3	6.86×10^{-3}	5.43×10^{-3}	8.17×10^{-6}	3.58×10^{-5}	1.40×10^{-5}	1.03×10^{-4}	1.26×10^{-2}
	HS4	6.86×10^{-3}	4.85×10^{-3}	1.52×10^{-5}	7.00×10^{-5}	1.50×10^{-5}	7.58×10^{-5}	1.19×10^{-2}
	HS5	6.77×10^{-3}	1.07×10^{-2}	3.49×10^{-4}	9.17×10^{-6}	1.17×10^{-5}	5.74×10^{-5}	1.80×10^{-2}
	HS6	6.35×10^{-3}	2.93×10^{-3}	7.82×10^{-5}	3.53×10^{-5}	1.70×10^{-5}	1.05×10^{-4}	9.43×10^{-3}
	HS7	6.77×10^{-3}	5.53×10^{-3}	5.25×10^{-5}	1.32×10^{-5}	1.41×10^{-5}	1.41×10^{-4}	1.25×10^{-2}
	HS8	6.67×10^{-3}	9.33×10^{-4}	1.98×10^{-5}	4.92×10^{-5}	7.31×10^{-6}	1.09×10^{-4}	7.79×10^{-3}

In Table 2, the THQs of the metals for children's ingestion ranged from 2.11×10^{-2} to 2.74×10^{-2} for Cd, 3.73×10^{-3} to 4.27×10^{-2} for Pb, 3.27×10^{-5} to 1.40×10^{-3} for Ni, 3.67×10^{-5} to 2.80×10^{-4} for Mn, 2.92×10^{-5} to 6.78×10^{-5} for Zn, 2.29×10^{-4} to 8.10×10^{-4} for Fe. For adults' ingestion, the THQs values were 5.27×10^{-3} to 6.86×10^{-3} for Cd, 9.33×10^{-4} to 1.07×10^{-2} for Pb, 8.17×10^{-6} to 3.49×10^{-4} for Ni, 9.17×10^{-6} to 7.00×10^{-5} for Mn, 7.31×10^{-6} to 1.70×10^{-5} for Zn, 5.74×10^{-5} to 2.02×10^{-4} for Fe. The ∑THQ values of the metals in the honeys ranged from 3.12×10^{-2} to 7.20×10^{-2} and 7.79×10^{-3} to 1.80×10^{-2} for children and adults ingestions respectively.

Table 3: Heavy metals levels (mg kg^{-1}) in honey from this study in comparison with others in literature

Location	**No of samples**	**Method**	**Cd**	**Pb**	**Ni**	**Mn**	**Zn**	**Fe**	**References**
South-south Nigeria	8	AAS	0.226-0.294	0.14-1.601	ND-0.299	0.055-0.42	0.094-0.218	1.721-6.072	This study
Tlemcen Province, north-western Algeria	18	ICP-MS	ND –0.0081	ND –0.1327	–	1.36 – 13.9	0.223 –13.9	11.7 –59.6	Bereksi-Reguig *et al.* (2020)
Tamale Metropolis, Ghana	20	AAS	0.400 – 7.300	44.000 – 111.100	1.200 – 44.100	0.900 – 15.100	4.200 – 8.100	–	Magna *et al.* (2018)
Nigeria			<0.001	<0.50-39.8	<0.25-6.98	11-31.75	1.0-31.0	5.0-163	Iwegbue *et al.* (2015)
Baringo and Keiyo Counties, Kenya	14	AAS	0.044–0.224	0.063–0.491	–	–	0.012– 0.259	0.073–1.295	Maiyo *et al.* (2014)
Podkarpackie region, Poland	10	ICP-OES	ND – 0.07	ND –0.77	ND – 2.43	0.15–16.72	–	–	Tomczyk *et al.* (2020)
Iranian markets	10	ICP-AES	0.08–0.39	0.05–0.11	–	0.16–0.42	2.53 –2.93	2.29–5.31	Akbari *et al.* (2012)
Lithuania	12	GFAAS and FAAS	0.002–0.013	0.008–1.649	0.012–0.087	–	–	–	Šereviciene *et al.* (2022)
Northeast region of Romania	52	ICP-MS	1.14–3.81	26.0–78.0	122.1–325.4	868.7–2528.8	2421.6–3870.7	19156–28285	Oroian *et al.* (2016)
Kenya	9	FAAS	0.02–0.03	0.08–0.28	–	–	0.016–0.43	0.20 – 1.12	Mbiri *et al.* (2011)
Central Anatolia	34	ICP-OES	0.09–0.24	0.02–1.50	0.03–1.44	0.24–1.56	0.50–5.39	0.57–8.74	Leblebici and Aksoy (2008)
Plateau State, Nigeria	5	GC-MS	ND– 0.0013	0.0007 - 0.0025	–	15.67 - 35.19	–	0.001–0.005	Lekduhur *et al.* (2021)
Turkey	65	ICP-OES	ND– 0.297	ND–3.035	–	0.096–29.496	1.734–245.205	3.506–1278	Altunatmaz *et al.* (2019)
Pakistan	52	HPLC	–	–	–	0.42– 0.59	0.37 –3.20	2.50–14.88	Yaqub *et al.* (2020)
Annaba, Northeast Algeria	4	FAAS	0.009–0.020		0.392–0.753			8.96 – 7.947	Chafik *et al.*, 2022
Ardabil, North West of Iran	25	ICP-OES	1.36– 125.88	117.46–1627.82	65.04–1094.49	–	122.86–6638.55	–	Aghamirlou *et al.* (2015)
North-Western Regions of Iran	72	AAS	–	0.06–0.12	–	–	2.03–6.84	–	Mahmoudi *et al.* (2015)

Discussion

Statistical analysis using ANOVA suggests significant variation ($p < 0.05$) in the levels of metals in the honeys. This significant variation could be from beehive locations, bee-keeping methods, environmental pollution, honey production stages (Iwegbue *et al*., 2015; Ioannidou *et al*., 2005). Cadmium was detected in all the honeys. The maximum Cd concentration was obtained in sample HS3 and HS4 while the minimum Cd concentration was obtained in sample HS1. The level of Cd in these honeys was comparable to others reported in literature (Table 3). However, higher levels of Cd were reported by Magna *et al.* (2018), Oroian *et al.* (2016) and Aghamirlou *et al.* (2015). Like Cd, Pb was also detected in all the honeys. The highest Pb concentration was found in sample HS5 while the lowest Pb concentration was obtained in sample HS8. The level of Pb in these honeys were lower than the allowable 1 mg kg^{-1} Pb in food specified by the European Commission (2006). The level of Pb in these honeys was similar to others reported in literature (Table 3). However, higher Pb levels were reported in honeys from Ghana (Magna *et al.*, 2018) and Iran (Aghamirlou *et al.*, 2015).

The highest Ni concentration was observed in sample HS5 while Ni was below the detection limit in sample HS6. The level of Ni in the honeys was lower than those reported by Magna *et al.* (2018), Oroian *et al.* (2016) and Aghamirlou *et al.* (2015). Copper was detected only in samples HS3 and HS5. The highest Mn concentration was detected in sample HS4 while the lowest was found in HS5. A wide range of Mn level in honeys has been reported in literature (Table 3). The maximum and minimum Zn levels were detected in samples HS6 and HS8 respectively. The highest Fe concentration was found in sample HS1 while the lowest concentration was found in sample HS5. Wide ranges of Mn levels in honey have been reported in literature (Table 3). Higher levels of Fe have been reported in Algeria (Bereksi-Reguig *et al.*, 2020; Chafik *et al.*, 2022), Romania (Oroian *et al.*, 2016), Central Anatolia (Leblebici & Aksoy, 2008), Turkey (Altunatmaz *et al.*, 2019), and Pakistan (Yaqub *et al.*, 2020).

The EDI of the heavy metals in these honeys were below their tolerable daily intake (TDI) (Tesi *et al*., 2020; Iwegbue *et al*., 2015). The THQ values for the individual metals and ∑THQ were <1 in the honeys for both children and adults ingestions (Table 2). This indicates that there is no safety concern for consumers of these honey samples. Among the metals, Cd has the greatest contribution to the ∑THQ values. The THQ of the individual metals was in the order of Cd > Pb > Fe > Ni > Mn > Cu > Zn.

Conclusion

This study has shown that the honeys from Bayelsa State studied were contaminated with metals (Cd, Pb, Ni, Mn, Zn and Fe). However, the EDI values of the metals were below their respective TDI values. The THQ of the individual metals was in the order of Cd > Pb > Fe > Ni > Mn > Cu > Zn. The THQ and ∑THQ of the metals were < 1 indicating no safety concern for consumers of these honeys.

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