# Assessment of Heavy Metals and Pesticide Residues in Honey Samples Collected From Selected Villages in Five Local Government Areas of Adamawa State, Nigeria

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***Abstract:*** *This study was designed to assess the heavy metals and pesticide residues in honey samples collected from selected villages in Adamawa state, Nigeria. A total of five (5) different natural samples of wild harvested honey were bought from the honey harvesters in Dulwarchira, Gada, Gurum, Kwaja and Muva villages. Heavy metals in the honey samples were determined using Atomic Absorption Spectrophotometer, while pesticides residues were determined using High Performance Liquid Chromatography. The results show that honey sample from Gada contains high concentration of Cr and Mn. Fe and Cd were found to be highest in samples collected from Kwaja. The concentration of Pb was found to be highest in samples collected from Muva even higher than the permissible dose. The sample collected from Muva and Gurum are rich in essential metal Zn. Aldrin was detected in all the villages (Dulwarchira, Gada, Gurum, Kwaja and Muva) with the highest value recorded in Gurum (0.021 ± 0.03). Alpha-BHC were detected in Dulwarchira, Kwaja and Muva with the highest value recorded in Dulwarchira (0.032 ± 0.10). The levels of p-DDT were detected in Gada, Gurum and Muva. Deldrin levels in honey samples were recorded in Dulwarchira, Gada, Kwaja and Muva. Levels of Endosulfan I, Endosulphan II and Endosulfan Sulphate were detected in honey samples from Dulwarchira, Kwaja and Muva while not detected in samples from Gada and Gurum villages. Heptachlor (organochlorines) pesticide were also detected in honey samples from Dulwachira and Gada villages and not detected in samples from Gurum, Gada and Muva. Chlorpyriphos were detected in Gurum and Kwaja. Diazinon was found in honey samples from Dulwachira and Gurum. Dichlorovos were also detected in honey samples from Dulwachira, Gada, Gurum and Kwaja. Fenitrothion in honey samples from the different villages were recorded in Gada and Gurum. Fenithion residues were found in honey samples from Kwaja and Muva villages. Bifenthrin were detected in samples from Gada and Gurum. Cypermethrin were recorded in samples from Dulwachira, Gurum, Kwaja and muva villages. Permethrin residues were detected in honey samples from Dulwachira and Kwaja with the highest value recoded from Duwachira. From this study, it can be concluded that honey samples from these areas contain Fe, Zn, Cu and Mn at high concentrations which were virtually above the maximum permissible limits prescribed by FAO/WHO. While Pb was below the permissible limits prescribed by FAO/WHO.*

**Keywords: Honey, Heavy metals, Pesticides residue, Assessment, AAS.**

## Introduction

Recent clinical and epidemiological studies show that honey may act as an important mediator of human health (Khalil and Sulaiman, 2010; Eteraf-Oskouei, and Najafi, 2013; Samarghandian *et al*., 2017; Cianciosi *et al*., 2018; Nguyen *et al*., 2019). Honey is a natural substance which has been used as a sweetener for thousands of years, is produced by honey bees (*Apis mellifera* and *Apis dorsata fabricious*) from the nectars of plants. Its chemical consumption is influenced by its botanical origin, mode of processing, seasons, and environmental conditions (Cianciosi *et al*., 2018; Khan *et al.,* 2017). Honey is not only considered as food or a sweetener, its consumption has long been regarded as having beneficial effects on human health, as described in early Greek, Roman, Vedic, and Islamic texts (Hossen *et al.*, 2017).

In addition, more recent in vitro and in vivo studies have confirmed that honey possesses a range of antioxidant, antimicrobial, antiviral, anticancer, and antidiabetic properties, and it has been shown to demonstrate protective activities on the nervous, cardiovascular, gastrointestinal, and respiratory systems (Khalil and Sulaiman, 2010; Eteraf-Oskouei, and Najafi, 2013; Samarghandian *et al*., 2017; Cianciosi *et al*., 2018). It has been found that the effect of honey on the cardiovascular system depends on the bioavailability of various phytochemical compounds, and on their methods of absorption and metabolization (Cianciosi *et al*., 2018). Honey has some myriad health benefit as it could be used to treat coughs and sleep difficulty in children associated with childhood upper respiratory tract infection (Punch newspaper, 2020). Cenat *et al*. (2015), reported that honey has variation in taste, colour, and smell depending on the nectars of flowers, and the content of various plant pigments, especially carotenoids. (Khalil and Sulaiman, 2010) also described a correlation between colors. Honey is produced almost all over the world, with global production estimated to be approximately 1.2 million tons (Bogdanov *et al*., 2008).

The main source of heavy metal concentration in honey has been traced to the soil; being transported to honey plants via the root system, passing to the nectar and eventually to the honey produced by the bees (Stankovska *et al*., 2008). However, there is evidence that the presence of heavy metals in honey can emanate from anthropogenic factors (Bogdanov, 2006). Heavy metals originate from two primary sources; natural sources and anthropogenic sources including metalliferous mining and industries, agrochemicals and mineral fertilizers, vehicle exhaust, sewage sludge and industrial wastes (Jiao *et al.,* 2015). According to (Aljedani, 2017) although the metals poison the bodies of honey bees, the honey bees are still able to travel great distances in the search for food and may not die directly from the poisoning of these metals. The metals, however, accumulate in their bodies helping them to play the role of detecting heavy metals in the environment. The evaluation of heavy metals content in honey has a twofold significance: the former one lies in the toxicity of these metals, with the consequent necessity to develop adequate analytical procedures for their monitoring; the latter one is suggested by the possibility of using bees and their products as bio-indicator.

Chemical pesticides are conventionally synthetic materials that directly kill or inactivate the pest (Rama *et al*., 2011). Depending on their chemical properties they can enter the organism, bio accumulate in food chains and consequently influence also human health (PAN Europe, 2010). Pesticides use is usually accompanied with deleterious environmental and public health effects. Pesticides hold a unique position among environmental and public health effects. Pesticides hold a unique position among environmental contaminants due to their high biological activity and toxicity (acute and chronic). Although some pesticides are described to be selective in their modes of action, their selectivity is only limited to test animals. Thus pesticides can be best described as biocides i.e., capable of harming all forms of life other than the target pest (Zacharia, 2011).

## MATERIALS AND METHOD

### Equipments

Refrigerator, Analytical balance, Auto sampler G4513A, High Performance Liquid Chromatography (HPLC), Atomic absorption spectrophotometer (AAS).

**Chemicals**

- Concentrated $HNO_3$, $H_2O_2$ and $H_2SO_4$.
- Deionized water
- Hydroquinone 1, 10-phenanthroline hydrate.
- Sodium acetate trihydrate
- Ferrous ammonium sulfate hexahydrate

**Collection of Samples**

A total of five (5) different natural samples of wild harvested honey were bought from the honey harvesters in the different Metropolis. One hundred grams (100 g) each of the samples was stored in cleaned labeled glass jars, kept at room temperature and in the dark until analysis.

**Determination of Heavy Metals in Honey**

Five grams (5g) of the honey was weighed using an analytical balance, transferred into a beaker, digested using oxi-acidic mixture of $NO_3/H_2\ O_2$ and filtered into a 50ml volumetric flask. This mixture was heated up to 120$^{o}$C for 3hr and brought to a volume of 25ml with deionized water. The blank digestions were repeated using the same procedure (AOAC. 1985; Mbiri *et al*., 2011). Blanks were prepared to check for background contamination by the reagents used. For quality assurance, honey samples were digested thrice along with blanks to minimize error. All reagents were of analytical reagent grade. Double distilled deionized water (Milli-Q Millipore 18.2 MΩ-cm resistivity) was used for all dilutions. The instrument was calibrated with a series of standard solutions supplied by a reputable company. Also, all metal content determinations were performed with five replications. The digested honey samples were analyzed for the heavy metals (Cd, As, Pb, Cu, Ni, Mn, Cr and Zn) using atomic absorption spectrophotometer.

**Pesticides Analysis**

Five (5) samples of row honey was obtained from five selected villages from bee farmers of five different local government areas of Adamawa state Nigeria. The extraction and purification of the pesticides (organochlorine, organophosphate and pyrethroid) residues from the honey samples prior to instrumental analysis was carried out based on the procedure described by Lehotay *et al*., *(*2005). Five (5) grams of honey sample was taken and dissolved in 10 ml of de-ionized water. 10ml of acetonitrile acidified with acetic acid was added, 1.0 g sodium acetate and 4.0 g anhydrous magnesium sulphate was shacked for at least a minute. And further shaken vigorously for another one minute. Following the centrifugation at 4,000 rpm for 2 minutes, 6 ml of the extract was transferred into 15 ml glass tube containing 0.4 g primary and secondary amine (PSA) sorbent and 0.6 g anhydrous magnesium sulphate. The formed mixture was vigorously shaken for 1 minute and centrifuged at 4000 rpm for 2 minutes. The residue was dissolved in 2 ml of injection standard and passed through a 0.50 μm sized pore filter and quantified by HPLC.

**RESULTS AND DISCUSSION**

Table 1 shows the results of heavy metals results in honey samples collected from different villages in Adamawa State. Concentration of metals in honey sample from Gada contains high concentration of Cr and Mn. Highest concentration of Cu was recorded in samples from Dulwachira. Iron an essential element was highest in samples from Kwaja. Cd a non-essential element was highest in samples collected from Kwaja. The poisonous metal Lead (a non-essential element) was found to be highest in samples collected from Muva higher than the permissible dose. The sample collected from Muva and Gurum are rich in essential metal Zn. Concentration of Cd was slightly higher than the permissible range in this sampled.

Naturally, lead is found in the earth's crust and has been associated with illness in children and adult especially cardiovascular related diseases. Acute lead poisoning in humans causes severe damage in the kidneys, liver, brain, reproductive system and central nervous system, and even causes death (Dhahir and Hemed, 2015). Lead (Pb) residues in food are mostly linked to human activities such as farming, industrial and vehicular emissions and storage places. In this study, lead concentration in the honey samples was below the tolerable dose. The presence of lead may be due to the frequent farming activities and heavy vehicular emissions from the study area. Lead (Pb) in this study happens to be in little concentration than Cd, Cr, Zn, Cu, Mn, and Fe respectively. All the trace elements detected were below the permissible limits of <50mg/g. The number of different minerals and heavy metals in honey may be largely dependent on the soil composition, as well as various types of floral plants, because minerals are transported into plants through the roots and are passed to the nectar and finally into the honey produced from it (Chukwujindu *et al*. 2015). Also, the beekeeping practices, environmental pollution, and honey processing also contribute to the diversified mineral content found to be present in honey (Pohl, 2009). Another key trace mineral detected in both honey sample is Manganese, which acts as co-factor for up to 300 enzymes most of which are related to antioxidant reactions. Manganese deficiency contributes to aging and age-related disorders (Huskisson *et al*., 2007).

Manganese is required by the body for enzyme functioning, nutrient absorption, wound healing bone development. The element manganese deficiency results in poor bone growth, joints pains and related fertility problems. The results of Mn obtained in this study is contrary to the work of Chukwujindu *et al*. (2015) who reported highest concentrations of Mn (31.75 mg/kg) in honey samples from Calabar. Some other benefits of manganese include; preventing anemia, alopecia, alleviating premenstrual syndrome, epileptic seizures and antioxidant protection. Mn may also be contained in foods such as, whole grains, beans, nuts, okra and cocoa. Mn is one of the most abundant metals in the soil and normally occurs as oxides and hydroxides. Its effects in the body mostly occur in the respiratory tract and the brains. Some symptoms of manganese poisoning include; forgetfulness and hallucination. Manganese causes Parkinson disease and impotency in men when exposed for a longer time. The elevated levels of Mn in the honey samples from the study areas may be due its presence in the dust through the air, industrial activities, burning fossil fuels and the surface waters. The study also reported the pollution of Cu in the honey samples. Other heavy metals detected in the honey from the Dulwachira, Gada, Gurum, Kwaja and Muva includes Cd, Cr, Zn, Cu and Fe. The presence of these toxic metals in honey samples is an evidence of some micro polluting agents in the environment.

Most elements detected from honey samples are useful for good health at optimum concentrations, especially when they originate from plant or organic source, rather than that of inorganic and metallic source. They will have five times the specific gravity of water and become toxic. At this stage they are referred to as heavy metals. Which are known to be toxic or poisonous at low concentration because of their tendency to accumulate in living organism. Thereby leading to toxicity in humans. The toxicity occurs due to the inability of the heavy metal to be metabolized by the body, leading to accumulation in human or animal soft tissues without being fully inactivated or destroyed (Ajibola *et al*., 2012). Health problems caused by heavy metals include headaches, metabolic abnormalities, respiratory disorders, nausea, and vomiting. For instance, lead can cause damage to the brain, kidney, nervous system, and red blood cells (Chukwujindu *et al*. 2015).

**Table 1: Heavy Metals residues in the Honey Samples (µg/kg)**

| Parameters | Dulwarchira | Gada | Gurum | Kwaja | Muva |
|---|---|---|---|---|---|
| **Chromium** | $0.013 \pm 0.01^{a}$ | $0.019 \pm 0.01^{a}$ | $0.013 \pm 0.02^{a}$ | $0.015 \pm 0.01^{a}$ | $0.012 \pm 0.01^{a}$ |
| **Copper** | $0.55 \pm 0.30^{b}$ | $0.43 \pm 0.10^{a}$ | $0.12 \pm 0.10^{c}$ | $0.31 \pm 0.10^{d}$ | $0.43 \pm 0.03^{a}$ |
| **Iron** | $1.11 \pm 0.50^{c}$ | $1.21 \pm 0.15^{c}$ | $1.81 \pm 0.50^{a}$ | $1.92 \pm 0.13^{a}$ | $1.71 \pm 0.10^{a}$ |
| **Cadmium** | $0.022 \pm 0.01^{d}$ | $0.042 \pm 0.01^{a}$ | $0.042 \pm 0.03^{a}$ | $0.052 \pm 0.03^{b}$ | $0.050 \pm 0.01^{b}$ |
| **Lead** | $0.012 \pm 0.01^{a}$ | $0.013 \pm 0.01^{a}$ | $0.012 \pm 0.01^{a}$ | $0.012 \pm 0.01^{a}$ | $0.016 \pm 0.01^{a}$ |
| **Zinc** | $2.46 \pm 0.30^{a}$ | $2.12 \pm 0.10^{a}$ | $3.21 \pm 0.13^{b}$ | $3.06 \pm 0.15^{b}$ | $3.56 \pm 0.10^{c}$ |
| **Manganese** | $6.35 \pm 0.540^{a}$ | $13.15 \pm 0.53^{b}$ | $12.63 \pm 0.51^{b}$ | $8.06 \pm 0.14^{a}$ | $8.76 \pm 0.15^{a}$ |

Values are mean ± SD. Values with the same superscript are statistically not significant ($p<0.05$)

Table 2: shows the results of organochlorines pesticides residues in honey samples from different locations in Adamawa State.

Aldrin was detected in all the villages (Dulwarchira, Gada, Gurum, Kwaja and Muva) with the highest value recorded in Gurum (0.021 ± 0.03). Alpha-BHC were detected in Dulwarchira, Kwaja and Muva with the highest recorded in Dulwarchira (0.032 ± 0.10). The levels of p-DDT were detected in Gada, Gurum and Muva. Deldrin levels in honey samples were recorded in Dulwarchira, Gada, Kwaja and Muva. Levels of Endosulfan I, Endosulphan II and Endosulfan Sulphate were detected in honey samples from Dulwarchira, Kwaja and Muva while not detected in samples from Gada and Gurum villages. Heptachlor (organochlorines) pesticide were also detected in honey samples from Dulwachira and Gada villages and not detected in samples from Gurum, Gada and Muva.

Table 3 shows the organophosphates pesticides residues in honey samples from different villages of Adamawa State. Chlorpyriphos were detected in Gurum and Kwaja with Gurum having the highest value (0.0030 ± 0.011). Diazinon were found in honey samples from Dulwachira and Gurum the highest being recorded in sample from Gurum. Dichlorovos were also detected in honey samples from Dulwachira, Gada, Gurum and Kwaja with the highest value recorded at Gada (0.0034 ± 0.001). Fenitrothion in honey samples from the different villages were recorded in Gada and Gurum with the highest value recorded in samples from Gada (0.0034 ± 0.001). Fenithion residues were found in honey samples from Kwaja and Muva villages with the same concentration.

**Table 2: Organochlorines pesticides residues in the Honey Samples (µg/kg)**

| **Pesticides** | **Dulwachira** | **Gada** | **Gurum** | **Kwaja** | **Muva** |
|---|---|---|---|---|---|
| **Organochlorines** | | | | | |
| **Aldrin** | 0.025±0.10$^{a}$ | 0.013±0.01$^{b}$ | 0.021±0.03$^{a}$ | 0.015±0.01$^{b}$ | 0.011±0.01$^{b}$ |
| **Alpha-BHC** | 0.032±0.10$^{b}$ | ND | ND | 0.012±0.01$^{a}$ | 0.012±0.01$^{a}$ |
| **o, P-DDT** | ND | 0.012±0.01$^{a}$ | 0.012±0.03$^{a}$ | ND | 0.013±0.02$^{a}$ |
| **Deldrin** | 0.025±0.30$^{a}$ | 0.013±0.01$^{b}$ | ND | 0.011±0.03$^{b}$ | 0.013±0.01$^{b}$ |
| **Endosulfan I** | 0.011±0.01$^{a}$ | ND | ND | 0.015±0.05$^{a}$ | 0.012±0.02$^{a}$ |
| **Endosulfan II** | 0.015±0.03$^{b}$ | ND | ND | 0.012±0.01$^{b}$ | 0.013±0.03$^{b}$ |
| **Endosulfan Sulphate** | 0.014±0.01$^{a}$ | ND | ND | 0.013±0.01$^{a}$ | 0.013±0.01$^{a}$ |
| **Heptachlor** | 0.014±0.03$^{b}$ | 0.011±0.01$^{b}$ | ND | ND | ND |

Values are mean ± SD. Values with the same superscript are statistically not significant ($p<0.05$). ND = Not Detected

**Table 3: Organophosphates pesticides residues in the Honey Samples (µg/kg)**

| Pesticides | Dulwachira | Gada | Gurum | Kwaja | Muva |
|---|---|---|---|---|---|
| **Organophosphates** | | | | | |
| **Chlorpyriphos** | ND | ND | $0.0030{\pm}0.011^{a}$ | $0.0022{\pm}0.001^{b}$ | ND |
| **Diazinon** | $0.0011{\pm}0.001^{a}$ | ND | $0.0023{\pm}0.001^{b}$ | ND | ND |
| **Dichlorovos** | $0.0022{\pm}0.001^{a}$ | $0.0034{\pm}0.001^{b}$ | $0.0033{\pm}0.010^{b}$ | $0.0012{\pm}0.001^{c}$ | ND |
| **Fenitrothion** | ND | $0.0034{\pm}0.001^{b}$ | $0.0011{\pm}0.001^{a}$ | ND | ND |
| **Fenthion** | ND | ND | ND | $0.0012{\pm}0.001^{a}$ | $0.0012{\pm}0.001^{a}$ |

Values are mean ± SD. Values with the same superscript are statistically not significant ($p<0.05$). ND = Not Detected

Table 4 shows the pyrethroids pesticides residues in honey samples collected from different villages in Adamawa State. Bifenthrin were detected in samples from Gada and Gurum. Cypermethrin were recorded in samples from Dulwachira, Gurum, Kwaja and muva villages. Permethrin residues were detected in honey samples from Dulwachira and Kwaja with the highest value recoded from Duwachira.

The pesticides analysis was carried out for determination of different classes of compounds (organochlorines, organophosphates and pyrethroids). The result showed the presence of residual concentration in all honey samples. This is contrary to the results obtained by Bogdanov, (2006) who reported no measurable residues of insecticides in honey. The results were compared with the maximum residue limits (MRLs) for pesticides in honey (EC, 2005, Darko *et al.*, 2017). The MRLs for pesticides in honey were adopted from European MRL due to lack of available MRL for honey in Codex ~ 1409. The results from this study agrees with several studies detecting residual level of pesticides in honey (Bwatanglang *et al.*, 2019). Bwatanglang *et al.* (2019) reported residual levels of pesticides in honey samples collected alon Mbu-Yola road Adamawa State. Pesticide residues in honey samples collected from Hong and Mubi, Adamawa State was also reported by (Bwatanglang *et al.*, 2019) similar to this study. Panseri *et al.* (2014) reported the presence of organochlorines pesticides in honey samples. Several investigations conducted on different types of honey through various analytical methods (Buldini *et al.*, 2001; Zhu *et al.*, 2008; Zhang *et al.*, 2011; Blasco *et al.*, 2011) showed the presence of pyrethrins and pesticides in honey from India (Mukherjee, 2009) and Spain (Herrera *et al.*, 2005), respectively. In another study on honey from Spain and Portugal, residues of 42 different pesticides were examined (Blasco *et al.*, 2003; Blasco *et al.*, 2004) and most of the compounds found were organochlorines, like gamma-HCH, HCB and its isomers α-HCH and β-HCH, with concentrations ranging from 0.03 to 4.31 mg kg-1, but most of them were below 0.5 mg kg-1.

**Table 4: Pyrethroids pesticides residues in the Honey Samples (µg/kg)**

| Pesticides Pyrethroids | Dulwarchira | Gada | Gurum | Kwaja | Muva |
|---|---|---|---|---|---|
| **Bifenthrin** | ND | 0.0021±0.001[a] | 0.012±0.001[a] | ND | ND |
| **Cypermethrin** | 0.0012±0.003[a] | ND | 0.0100±0.001[a] | 0.0012±0.001[a] | 0.0012±0.001[a] |
| **Permethrin** | 0.0031±0.001[a] | ND | 0.0023±0.001[a] | 0.0026±0.003[a] | ND |

Values are mean ± SD. Values with the same superscript are statistically not significant ($p<0.05$)

## CONCLUSION

From this study, it can be shown that Iron (Fe), Zinc (Zn), and Copper (Cu) and Manganese (Mn) reported high concentrations which were virtually above the maximum permissible limits while Lead (Pb) was below the permissible limit as prescribed by FAO/WHO. Their high presence in the honey was probably due to the proximity of agricultural pesticides, emissions, use of fertilizers and the frequent cultivation of crops such as corn. Other heavy metals residues such as Cr and Cd were low and within the permissible limits proposed by WHO/FAO.

## CONSENT

It is not applicable.

## ETHICAL APPROVAL

It is not applicable.

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## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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