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# Assessing the Total Levels and Health Risk of Presence of Cadmium, Cobalt, Copper, Lead and Zinc in Most Smoked Tobacco Sold in Alassaba Municipality-Libya

Salem Irhema S. Irhema [1 *], Adel Almaprok S. Arhouma [2], Sarah Abdulhakim Al-Shaqlouf [3], Fatima Aboulqasim Haba [4]
[1] Environmental Sciences Department, Faculty of Science, University of Al-Zintan, Libya
[2,3,4] Chemistry Department, Faculty of Science- Alassaba, University of Gharyan, Libya

# تقدير التركيز الكلي والخطر الصحي الناتج عن وجود الكادميوم والكوبلت والنحاس والرصاص والزنك في أكثر أنواع السجائر مبيعاً بأسواق بلدية الأصابعة - ليبيا

سالم رحيمه سالم رحيمه [1*]، عادل المبروك سعد أرحومه [2]، سارة عبد الحكيم الشقلوف [3]، فاطمة بلقاسم حابه [4]
[1] قسم علم البيئة، كلية العلوم، جامعة الزنتان، ليبيا
[2،3،4] قسم الكيمياء، كلية العلوم -الأصابعة، جامعة غريان، ليبيا

[*]*Corresponding author:* s.irhema@uoz.edu.ly

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**Abstract:**
Tobacco is one of the sources that may introduce several heavy metals into the bodies of positive and negative smokers, leading to harmful effects on smokers' bodies. Because a major number of Libyan populations are smoking different brands of cigarettes alongside raw tobacco leaves which may affect our citizens' health, accordingly 10 cigarette brands comprising Bon International, Gold Mexico Original, Camel Yellow, D&G, Milano London, Milano, Oris Fliter. Karelia Ligts, Yes and Libyan brand named Ryadi, and another sample of raw tobacco leaves (LRT) planted locally were collected randomly from local shops distributed in Alassaba municipality with aims to determine the total concentration of Cadmium (Cd), Cobalt (Co), Copper (Cu), Lead (Pb) and Zinc (Zn) and calculate the health risk related to the presence of these metals in the most consumed tobacco. The obtained results showed that all collected tobacco samples contained various levels of Cd, Co, Cu Pb and Zn (mg/kg) ranged from 0.38 ± 0.07 to 0.84 ± 0.01, 0.33 ± 0.04 to 0.5 ± 0.07, 15.29 ± 0.88 to 18.75 ± 0.72, 0.42 ± 0.04 to 1.04 ± 0.11 and 14.33 ± 0.33 to 32.45 ± 2.09, respectively. The mean levels of Co, Cu and Zn were within the allowable levels set by the World Health Organization (WHO), whereas Pb and Cd levels were above the acceptable levels. The calculated harm quotient (HQ) of target metals were greater than 1 representing extreme health risk due to the presence of Cd, Cu Pb and Zn in studied tobacco, nevertheless, the HQ of Co were lower than 1 presenting minimal health risk to the smokers. In conclusion, there is a need to emphasize the regulation of the import and tread of tobacco and monitor the content of heavy metals in tobacco to reduce the health effects of these cigarette brands.

**Keywords:** Cigarette brands, Heavy metals, Health risk, Smoking, Tobacco.

**الملخص**
تعتبر السجائر من أحد المصادر الذي يسبب استهلاكها أو التعرض لأدخنتها تراكم محتواها من العناصر الثقيلة بأجسام المدخنين لها تدخين سلبي وإيجابي مما يسبب اخطار صحية لهؤلاء المستهلكين. لهذا تم تجميع 10 أنواع مختلفة من العلامات التجارية للتبغ الأكثر استخداما من قبل المدخنين، وعينة من أوراق تبغ مُزروع محليا وجميعها مُباع في أسواق بلدية الأصابعة. عُوملت العينات بالطرق الموصوفة والمعتمدة علميا بهدف تقدير تركيز كلا من الكادميوم والكوبلت والنحاس والرصاص والزنك فيها وحساب مؤشر الخطر الصحي (HQ) الناتج عن وجود واستنشاق هذه العناصر من قبل المستهلكين للتبغ والمتعرضين لأدخنته. بينت النتائج أن تراكيز الكادميوم والكوبلت والنحاس والرصاص والزنك بوحدة مليجرام/كيلوجرام تراوحت بين 0.38 ± 0.07 الى 0.84 ± 0.01 ومن 0.33 ± 0.04 الى 0.5 ± 0.07 ومن 15.29 ± 0.88 الى 18.75 ± 0.72 ومن 0.42 ± 0.04 الى 1.04 ± 0.11 ومن 14.33 ± 0.33 الى 32.45 ± 2.09 على التوالي. وكان متوسط تركيز كلا من الكوبلت والنحاس والزنك أقل من الحدود العليا المسموح بها من قبل منظمة الصحة العالمية في التبغ، بينما كان متوسط تراكيز الكادميوم والرصاص أعلى من الحدود المسموح بها. وكانت قيم مؤشر الخطر الصحي (HQ) المحسوبة نظريا أعلى من 1 لجميع العناصر المدروسة عدا الكوبلت منبها لخطر صحي عالي نتيجة

لوجود هذه العناصر في عينات السجائر المُدروسة. وخلصنا الى ان هناك حاجة ملحه لاستصدار لوائح تنظم عمليات توريد وتوزيع وبيع هذه الأنواع من التبغ ومراقبة محتواها من العناصر الثقيلة للتقليل من الاخطار الصحية الناتجة من استهلاكها.

**الكلمات المفتاحية**: التبغ، التدخين، الخطر الصحي، العناصر الثقيلة، ماركات السجائر.

## Introduction

Heavy metals may enter the environment, particularly soil from many sources such as using animal manure, inorganic fertilisers and sewage sludge in the agriculture sector as well as mining and smelting activities and daily use of some materials and products contain heavy metals, consequently, the plants have grown in these soils including tobacco may be susceptible to contaminated by these pollutants. Tobacco plants can absorb heavy metals from polluted soil or/and the atmosphere and accumulate them in aboveground parts and specifically in their leaves [1]. The tobacco leaves are the parts used by humans to smoke as a raw material or after manufacturing them to produce different brands of cigarettes. Cigarettes contain more than 700 hazard chemicals that are reported to cause cancer comprising heavy metals, thus, more than $8 \times 10^6$ people die annually due to smoking tobacco, also $1.38 \times 10^6$ person may lose their lives because of negative smoking [2]. Pinto et al. [3]; Afridi et al. [4] and Ashraf [5] mentioned that Pb and Cd are commonly found in tobacco tissues and can reach smoker's body. Similarly, Umar et al. [6] reported that heavy metals including Cu, Zn, Fe, Co, Ni, Cr and Mn are found in tobacco and its products. The cigarette contents of Cd approximately ranging from 1 – 3 mg $kg^{-1}$ thus smoking one piece of cigarette may lead to inter 2 – 4 µg of Cd into the lung tissues of smokers. Likewise, smoking 20 piece of cigarettes /per day cause to inhalant 1 – 5 µg of Pb into human lungs [1]. Several heavy metals are essential to a human body in low intake levels e.g., Cu, Co and Zn, however high intake levels of vital metals and any concentrations of non-essential elements such as Pb and Cd are poisonous. The toxicity route of heavy metals is similar comprising: oxidative stress, weakening of antioxidant defense, damaging the hematopoietic system, and inhibiting the activity of the enzymes that are involved in flora and fauna cell division. Therefore, this leads to severe diseases for people who are exposed to these metals from different sources, especially due to smoking cigarettes and raw tobacco including positive and negative smokers [7]. According to Gatzke-Kopp et al. [8] the concentrations of heavy metals are higher in some biological fluids of smokers than in the same biological fluids of non-smokers people. Smoking tobacco is a common hobby universally particularly in Libya as most families have one person or more used to smoke for prolonged one or more brands of cigarette ranging from a few pieces of cigarette/day to tens /day. In addition, Libyan markets are open to importing any cigarette brands from all countries without control over the quality and content of the tobacco products, therefore monitoring the content of imported cigarettes of heavy metals and other dangerous chemicals is highly required action to protect and attract the attention of Libyan citizen about the harm effects of smoking tobacco and may assistance to persuade them to quit of this bad hobby. Therefore, the aims of the current study are to 1) evaluate the total concentration of Cd, Co, Cu, Pb and Zn in commonly smoked cigarette brands by Libyan people and raw tobacco leaves cultivated locally and 2) ascribe the concentration of heavy metals to the possible health risks and to produce better awareness of tobacco-associated health issues.

## Material and methods

### *Sample collection*

Ten cigarette brands that are mostly smoked by Libyan ordinary people Including Bon International, Gold Mexico Original, Camel Yellow, D&G, Milano London, Milano, Oris Fliter. Karelia Ligts, Yes and a Libyan brand named Ryadi were collected. The collected samples were donated by the owners of two local markets distributed in Alassaba municipality, and another sample of local raw tobacco leaves (LRT) was planted locally few people still use to smoke it, also donated by a farmer from the studied area.

### *Sample preparation and analysis*

According to the procedure reported by Engida [9]. The collected samples were oven-dried for 24 hours at 70°C then grounded separately and stored in polyethylene plastic bags till the time of analysis. To evaluate the total levels of target metals 2.0g of dry tobacco (DW) was digested by adding 10 mL of concentrated nitric acid ($HNO_3$ heavy metal grade, BDH 65%) in 100 mL borosilicate digestion beaker, covered and left overnight to digest in the fume cupboard before being heated in water bath at 100°C till dryness and left to cool to room temperature followed by applying 4 mL of 30% $H_2O_2$ and heated again to dryness and left to cool to room temperature before 10 mL of 1% $HNO_3$ solution was added than the acid extracted solution were filtered through Whatman 42 filter paper. The borosilicate beaker and filter paper were washed three times with double-distilled water, and the acid-extracted solution was made up to 25 mL in a volumetric flask. All the extracting solutions were stored at 4°C before analysing the total concentration of Cd, Co, Cu, Pb and Zn by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) following the EPA3050B method at the Libyan Petroleum Research Centre.

### *Heath Risk assessment*

The health risk assessment can quantify by calculation and was calculated as an average daily dosage (ADD) (mg $kg^{-1}$ $d^{-1}$) and harm quotient (HQ) according to the following equations (1, 2) reported by [10]:

$$ADD = (C \times IR \times EF \times ED) / BW \times AT \quad (1)$$
$$HQ_{\text{for inhalation}} = ADD / R_fC \quad (2)$$

Where:
*C* = Total concentration of target metal in cigarette brand (mg/kg)
*IR* = The inhalation rate of cigarette smoke (20 $m^3$ /day, [11])
*EF* = The exposure frequency to cigarette smoke (365 days /year, [11])
*ED* = The exposure duration to cigarette smoke (30 years, [11])
*BW* = The average body weight of smokers (70 kg, [11])
AT = The average time of exposure to cigarette smoke ((ED) 30 year × 365 = 10950 days, [11])
$R_fC$ is the reference concentration of metal (mg $kg^{-1}$ $d^{-1}$) for Cd = 0.001, Pb = 0.0032, Cu = 0.04, Zn = 0.3 [12] and Co = 5.7 × $10^{-6}$ [13].

***Assurance of result quality***

The accuracy of the trial results was achieved as possible as we can using analytical grade chemicals and double-distilled water. 10% of $HNO_3$ solution was used to clean all equipment before and after usage. The metal extractions were run in triplicate and multi-metal standard solutions (5 solutions) were used to standardise the instrument. The blank was prepared following the same procedure of analysed samples without tobacco and used for zeroing the device.

***Statistical Analysis***

The obtained data were analysed statistically using SPSS software version 26 and the resulting statistics are presented in table (1) as a mean (M) of 3 replicates ± standard error (SE). The data of Cd, Cu and Zn separately in cigarette brands were normally distributed according to Shapiro-Wilk test, however, Co and Pb levels did not normally distribute also according to Shapiro-Wilk test. Therefore, the Kruskal Wallis test for non-parametric data was used to identify if there are differences between the mean levels of Co and Pb separately within different cigarette brands at $P< 0.05$. Additionally, one-way ANOVA tests were used to compare the difference between the mean levels of Cd, Cu and Zn separately within different cigarette brands at $P< 0.05$. In the cause of Zn data, the ANOVA P-value was significant, thus Tukey's HSD post hoc test was run to recognise the difference between the mean levels of Zn within different cigarette brands at $P< 0.05$ (table 1). The ADD and HQ were calculated using equations 1 and 2 by Excel software version 2409 and the resulting statistics are presented in table (2).

## Results and discussion

The obtained results of total target metal concentration are presented in Table (1) as a mean of 3 replicates ± SE. Table (2) illustrates the average daily dosage (ADD) in mg $kg^{-1}$ $d^{-1}$ and harm quotient (HQ) values accordingly these results were discussed as follows:

Table 1. The total concentration of target metals as a M ± SE in mg/kg DW in investigated cigarettes

| Cigarette brand | Studied heavy metals | | | |
|---|---|---|---|---|
| | Cd | Co | Cu | Pb |
| Bon INTERNATIONAL | 0.84 ± 0.01ᵃ | 0.50 ± 0.001 | 16.42 ± 0.65ᵃ | 0.71 ± 0.04 |
| Gold Maxico Original | 0.38 ± 0.01ᵃ | 0.46 ± 0.04 | 15.71 ± 0.61ᵃ | 0.88 ± 0.001 |
| CAMEL YELLOW | 0.54 ± 0.08ᵃ | 0.42 ± 0.08 | 15.37 ± 1.68ᵃ | 0.46 ± 0.08 |
| D&G | 0.42 ± 0.11ᵃ | 0.33 ± 0.04 | 15.92 ± 1.17ᵃ | 0.71 ± 0.17 |
| MILANO LONDON | 0.42 ± 0.09ᵃ | 0.42 ± 0.09 | 15.33 ± 0.74ᵃ | 0.71 ± 0.04 |
| MILANO | 0.41 ± 0.13ᵃ | 0.42 ± 0.12 | 16.21 ± 0.37ᵃ | 0.67 ± 0.15 |
| ORIS ELITE | 0.50 ± 0.07ᵃ | 0.50 ± 0.07 | 15.42 ± 1.01ᵃ | 0.71 ± 0.04 |
| KARELIA LIGTS | 0.46 ± 0.11ᵃ | 0.46 ± 0.11 | 15.29 ± 0.88ᵃ | 0.67 ± 0.11 |
| Yes | 0.42 ± 0.14ᵃ | 0.33 ± 0.04 | 17.88 ± 0.19ᵃ | 0.50 ± 0.19 |
| Ryadi (Libyan brand) | 0.42 ± 0.11ᵃ | 0.42 ± 0.03 | 15.54 ± 0.80ᵃ | 0.42 ± 0.04 |
| Local raw tobacco leaves (LRT) | 0.38 ± 0.07ᵃ | 0.39 ± 0.03 | 18.75 ± 0.72ᵃ | 1.04 ± 0.11 |
| WHO Limitations* | 0.05 | 1.2 | 100 | 0.05 |

***Cd:***

Cadmium is extremely poisonous and carcinogenic element. This metal can affect the skeletal, nervous, circulatory and respiratory systems in the human body due to its long biological half-life time ranging from 13.6 – 23.5 years. Therefore, it can accumulate biologically in the lung tissue of smokers with levels higher by 5-fold than non-smokers [7]. In this study, the levels of Cd were higher than its permissible values set by WHO (0.05 mg/kg) as Cd levels ranged from 0.38 ± 0.07 in LRT samples to 0.84 ± 0.01 in Bon In. samples and the differences between the mean concentration of Cd among all cigarette brands and LRT were not significant ($P< 0.05$). The levels of Cd obtained in this study are in the range of that found by Umar et al. [6] (0.14 ± 0.04 – 1.46 ± 0.24 mg/kg) and that reported by Ntarisa [2] (0.53 ± 0.09 mg/kg). Our results were higher than that documented by Dahlawi et al. [14] for Cd levels recording 0.09 ± 0.01 mg/kg as a mean value in imported cigarettes to Saudi Arabia. Conversely, the results of the current study for Cd levels were much lower than that (9.7 – 14.9 mg/kg) reported by Ndokiari et al. [15]. All the values of the average daily dosage (ADD) and harm quotient (HQ) values were in proportional trend with each other, as ADD increased the HQ also increased for one brand. The calculated values of ADD ranged from 0.109 mg $kg^{-1}$ $d^{-1}$ in LRT and Bon In. brand to 0.02 mg $kg^{-1}$ $d^{-1}$ in Gold M. O. brand. In addition, the HQ recorded values ranged from 109 for LRT and Bon In. brand to 240 for Gold M. O, which is much greater than 1, indicating significant health risk specially to the positive as well as negative smokers [10].

***Co:***

Co is a heavy metal categorised in IARC group 2b as a human carcinogenic metal. Consequently, the exposure to elevated levels of Co may cause several health complications for the nervous system such as Parkinson and Alzheimer diseases [19]. The concentration of Co in investigated cigarette brands recorded values ranged from 0.33 ± 0.04 mg/kg in Yes and D&G brands to 0.5 ± 0.07 mg/kg in Oris brand without any significant difference among the average of Co levels of all examined tobacco samples according to Kruskal Wallis test at $P < 0.05$, additionally, the levels of Co in the studied samples were less than the permissible levels of Co in tobacco set by WHO (1.2 mg/kg). The result of this study is in line with others reported by Umar et al. [6] (0.00 – 1.2 mg/kg) and Abu-Obaid et al. [20] (0.18 – 2.61 mg/kg). The ADD values ranged from 0.094 mg $kg^{-1}$ $d^{-1}$ in Yes and D&G brands to 0.143 mg $kg^{-1}$ $d^{-1}$ in Bon In. and Oris brands. In addition, the HQ recorded values ranged from $1.7 \times 10^{-4}$ in Yes and D&G brands to $2.5\times 10^{-4}$ in in Bon In. and Oris brands which is much less than 1 indicating minimal health risk due to the Cobalt content of studied products' cigarette brands and LRT.

***Cu*:**

Cu is vital for plants and fauna in protein synthesis, photosynthesis procedure and as a catalyst to encourage some redox enzymes. Cu levels ranged from 15.29 ± 0.88 mg/kg in Karelia Ligts brand to 18.75 ± 0.72 mg/kg in LRT without any significant difference between each other at $P < 0.05$ as the one-way ANOVA teas showed and lower by more than 5 times than the allowable levels set by WHO (100 mg/kg). Our results are higher than those obtained by Umar et al. [6] (0.1 – 0.62 mg/kg) and Dahlawi et al. [14] (2.61 ± 0.01 mg/kg) and Ntarisa, [2] (9.35 ± 3.0 mg/kg). However, in the range of Cu levels in some tobacco brands sold in Palestine that ranged from 11.86 to 20.53 mg/kg found by Abu-Obaid et al. [20]. The calculated DDA of Cu in the examined samples varied from 4.73 mg $kg^{-1}$ $d^{-1}$ for Karelia Ligts to 5.36 mg $kg^{-1}$ $d^{-1}$ in LRT leading to record values for HQ ranging from 109.2 for Karelia Ligts to 133.9 for LRT representing extreme health risk to the people smoking these brands and specifically LRT as the HQ of all studied tobacco greater than 1 by more than 100-fold.

***Zn*:**

This metal is required in the development and growth of the human body at trace levels, however, the elevated concentration of Zn may cause anemia and The concentration of Zn in all studied tobacco samples showed levels less by 3 to 6-fold than the permissible levels (100 mg/kg) suggested by WHO ranging from 14.33 ± 0.33 mg/kg in LRT to 32.45 ± 2.09 mg/kg in Camal brand. These results are lower than that obtained by Hussain et al. (2024) (60 – 70 mg/kg) and others found by Ndokiari et al. [15] (153 – 183 mg/kg), however our results are significantly higher than that recorded by Umar et al. [6] (0.52 – 6.2 mg/kg) and Ntarisa [2] (0.92 ± 0.43 mg/kg). The ADD values of Zn ranged from 4.09 mg $kg^{-1}$ $d^{-1}$ because of smoking LRT to 9.27 mg $kg^{-1}$ $d^{-1}$ due to using Camel Y. brand to smoke, consequently, the HQ for all examined brands ranged from 13.65 to 30.90 for LRT and Camel Y. brand posing high health risk to positively and negatively smokers.

Table 2. The average daily dosage (ADD) mg $kg^{-1}$ $day^{-1}$ and harm quotient (HQ).

| Cigarette brand | Studied heavy metals | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Cd | | Co | | Cu | | Pb | | Zn | |
| | AAD | HQ | AAD | HQ | AAD | HQ | AAD | HQ | AAD | HQ |
| Bon INTERNATIONAL | 0.240 | 240 | 0.143 | $2.5 \times 10^{-4}$ | 4.69 | 117.3 | 0.20 | 63.39 | 7.80 | 25.99 |
| Gold Maxico Original | 0.109 | 109 | 0.131 | $2.3 \times 10^{-4}$ | 4.49 | 112.2 | 0.25 | 78.57 | 4.92 | 16.39 |
| CAMEL YELLOW | 0.154 | 154 | 0.120 | $2.1 \times 10^{-4}$ | 4.39 | 109.8 | 0.13 | 41.07 | 9.27 | 30.90 |
| D&G | 0.120 | 120 | 0.094 | $1.7 \times 10^{-4}$ | 4.55 | 113.7 | 0.20 | 63.39 | 7.09 | 23.65 |
| MILANO LONDON | 0.120 | 120 | 0.120 | $2.1 \times 10^{-4}$ | 4.38 | 109.5 | 0.20 | 63.39 | 9.06 | 30.19 |
| MILANO | 0.117 | 117 | 0.120 | $2.1 \times 10^{-4}$ | 4.63 | 115.8 | 0.19 | 59.82 | 4.52 | 15.08 |
| ORIS ELITE | 0.143 | 143 | 0.143 | $2.5 \times 10^{-4}$ | 4.41 | 110.1 | 0.20 | 63.39 | 8.57 | 28.57 |
| KARELIA LIGTS | 0.131 | 131 | 0.131 | $2.3 \times 10^{-4}$ | 4.37 | 109.2 | 0.19 | 59.82 | 8.43 | 28.10 |
| Yes | 0.120 | 120 | 0.094 | $1.7 \times 10^{-4}$ | 5.11 | 127.7 | 0.14 | 44.64 | 7.41 | 24.69 |
| Ryadi (Libyan brand) | 0.120 | 120 | 0.120 | $2.1 \times 10^{-4}$ | 4.44 | 111.0 | 0.12 | 37.50 | 8.51 | 28.37 |
| Local raw tobacco leaves (LRT) | 0.109 | 109 | 0.111 | $2.0 \times 10^{-4}$ | 5.36 | 133.9 | 0.30 | 92.86 | 4.09 | 13.65 |

HQ > 1 Refers to significant health risk

Generally, the studied tobacco brands and LRT contain levels of Zn, Co and Cu lower than the maximum limits set by WHO, but the concentrations of Pb and Cd were higher than those set by WHO. The QH values of all target metals except Co were greater than 1. This indicated that smoking positively or negatively of these brands and LRT may cause severe health risks. Even though, the presence of heavy metals in tobacco at negligible levels and smoking for a long time can lead to the accumulation of these hazard elements biologically in the human body tissues causing severe health problems for smokers.
As mentioned earlier the pollution of tobacco may be caused by the natural occurrence of heavy metals or due to anthropogenic activity as well as the manufacturing of cigarettes process might introduce some heavy metals to tobacco products [2].
The variation between the levels of studied metals among different previous studies mentioned above including the results obtained from this study may be related to several factors that are: 1) The soil texture where the tobacco planted, 2) the soil content of heavy metals (naturally or anthropogenically), 3) some physiochemical characteristics of these soils e.g., pH and organic matter, which play roles in the availability of heavy metals to plants in soils, and 4) tobacco plant species [14].

## Conclusion

The obtained results deliver an indication of the total levels of Cd, Co, Cu, Pb and Zn in frequently smoked cigarette brands by Libyan people and raw tobacco leaves cultivated locally. The observation about the levels of target metals in studied brands of tobacco showed that all explored tobacco products contain target metals with different levels ranging from concentrations of Cd, Pb above the permissible levels of these metals suggested by WHO to levels lower than WHO reference in the cause of Zn, Co and Cu. Further, it was also calculated that the harm quotient factor indicates high to extreme health risk due to the presence of all examined metals except Co. Thus, the utilising of these types of cigarettes can cause health problems for the consumers. Consequently, the current study suggested monitoring the content of imported and local tobacco products heavy metals continually to mitigate the associated health risks due to smoking these processed or raw tobacco leaves and establishing regulatory actions to control the importing, distributing, cultivation and product processing of tobacco.


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## Author contributions

Salem Irhema S. Irhema: Conceptualization, Validation, Formal analysis, Resources, Data Curation and Writing Original Draft.
Adel Almaprok S. Arhouma: Resources, Investigation and Supervision of laboratory work.
Sarah Abdulhakim Al-Shaqlouf : Resources, Investigation.
Fatima Aboulqasim Haba: Resources, Investigation.

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