

Analysis of the Fluoride Levels of Well Water Consumed by the Moroccan Population in Different Rural Areas

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Abstract—Fluoride plays an essential role in preventing cavities when consumed in small amounts. However, continuous excessive fluoride ingestion could cause significant adverse health effects in humans, ranging from mild dental fluorosis to debilitating skeletal fluorosis, depending on the level and period of exposure to fluorides. In rural areas, the source of fluoride in well water could be due to natural, industrial, or anthropogenic contamination. The purpose of our work was to assess the degree of contamination of groundwater by fluorides from different regions in Morocco located in agricultural campaigns, where the only source of consumable water is well water. A total of 40 well water samples were collected from 7 rural Moroccan areas. Fluoride level analyses were performed using a fluoride-specific ion electrode (HI-4110). The fluoride levels found in the various well waters of the different regions range from 0.2 mg/l to 6.58 mg/l. Five wells have levels that exceed the recommended adult standards, while 28 wells have levels above the recommended standards for children. These results help warn the public about the risk of developing dental fluorosis and encourage them to change the sources of water consumption from the well waters with high fluoride content.

Index Terms—Fluorides, water, wells, potentiometry, rural area, Morocco.

I. INTRODUCTION

Fluoride is an essential element for the growth and maintenance of bone tissue, and it is an important factor for the prevention of tooth decay. The recommended value of fluorides in drinking water according to the World Health Organization (WHO) is between 1.0 and 1.5 mg per day [1]. It also poses a significant health risk if the content exceeds 1.5 mg/day [2]. Excessive fluoride ingestion results in inhibition of ameloblasts, change in absorption characteristics, and surface properties of enamel crystals, with the formation of an unsightly porous enamel, which results in the appearance of dental fluorosis [3]-[5].

Fluoride is naturally found in the environment (through rock erosion and volcanic emissions, water, soil, plants, and other living organisms) and as a result of certain human activities such as mining and natural phosphate treatment,

aluminum manufacturing, or fluoridation of drinking water [6]-[8]. Many factors affect the concentration of fluorides in water, it has depended on the geological formations forming minerals like apatite, micas, amphiboles, and clay minerals traversed by water, temperature and pH. Its concentration also increases in summer and decreases in winter due to rain. [9], [10].

The exploitation of phosphate deposits in Morocco contributes to the manufacture of fertilizers for agriculture. Phosphoric acid, simple superphosphate, and concentrated superphosphate are simple fertilizers that can enter into the composition of complex fertilizers. These phosphates contain 3 to 4% fluoride in the form of apatite $3(\text{Ca}_3(\text{PO}_4)_2)_3(\text{Ca}_3(\text{PO}))$.

The main minerals containing fluoride sources are cryolite and aluminum fluoride (Na_3AlF_6). It is less dispersed, but often impure, and contains significant proportions of silica or silicates. It is used in aluminum manufacturing. These reprocessing industries are elsewhere causing chronic intoxication by fluoride ion: fluorosis, which can affect those who are exposed professionally, or populations: fluoridated pollution by effluents near plants, or by absorption of excess fluoridated water; or, animals put in pastures near plants releasing fluoride-rich fumes.

Agriculture in Morocco is characterized by the use of considerable quantities of plant protection products to protect crops from pests and increase yields. Their intensive and irrational use has consequences for the environment [11], [12].

In Morocco, various studies have revealed the exposure of the population to groundwater from the phosphate regions exposes, which show that groundwater pollution by fluorides is from phosphate origin, and this is the cause of dental fluorosis in these areas. In the region of Khouribga, the accumulation of fluorinated phosphate dust is the cause of groundwater and soil contamination and predisposes the population to fluorosis [13]. Few studies to are carried out to determine the levels of fluoride in well waters in non-phosphorus rural areas. However, few studies have been conducted at the level of food sources on fluoride-related toxicity in the Moroccan population.

The Moroccan population is characterized by the consumption of drinking water provided by the National Office for Drinking Water (ONEP) or other private distributors in urban areas. But most people in rural areas consider wells as their main source of water.

This study aims to highlight the fluoride levels contained in well water consumed by the population located in 7 rural Moroccans and to determine the risks of fluoride.

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II. MATERIALS AND METHODS

A. Geographic Meteorological Data of the Sampling Locations

Between September 2020 and February 2021, 40 samples were taken in 7 Moroccan regions: Had Soualem- Berrechid, Kenitra, Fez, Tit Mellil, Sidi Slimane - Sidi Kacem, Larache, Brachoua (Fig. 1). Well water samples were collected from the rural areas of Had Soualem located 10 km from the Atlantic Ocean and 30 km South of Casablanca at 33 degrees 25 degrees 04 degrees North, 7 degrees 50 degrees West and the rural area of Tit Mellil located at 33 degrees 33 degrees 29 degrees North, 7 degrees 29 degrees West.

The samples were taken in the rural areas of the Rabat-Salé-Kenitra region located between Sidi Slimane, in the North-West of Morocco at 34 degrees 15 degrees North, 5-55" West, the rural area of Sidi Kacem located at 34 degrees 13 degrees 00 degrees North, 5 degrees 42 degrees West, the rural area of Kenitra located at 34 degrees 15

degrees 00" North, 6 degrees 35 degrees 00" West and the rural area of Had El Brachoua located to the south of the region.

Samples were taken in rural areas of the Fez-Meknes region, on the outskirts of Fez, located at 34 degrees 03 degrees North, 4-58-59" West (Table I).

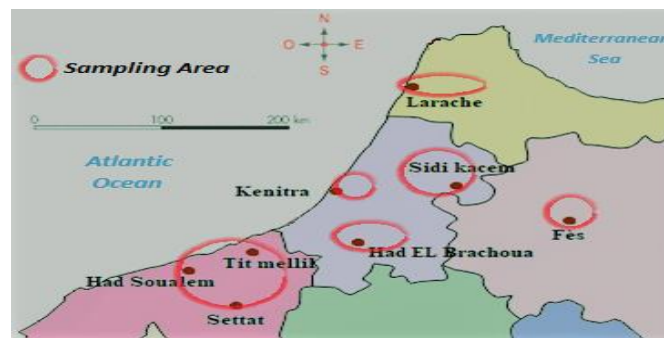


Fig. 1. Mapping of rural areas in Morocco indicating the different sampling point.

TABLE I: LOCATION AND CHARACTERISTICS OF WATER SOURCES AND THE WITHDRAWAL PERIOD

Sample	Region	Location	withdrawal period	Nature
P1	Had Soualem-Berrechid	Had Soualem	February 2020	Wastewater
P2	Had Soualem-Berrechid	Had Soualem	February 2020	Well
P3	Had Soualem-Berrechid	Harcha	February 2020	Well
P4	Had Soualem-Berrechid	Ouled Bouziri	February 2020	Well
P5	Kenitra	Douar Bourhma	February 2020	Well
P6	Kenitra	Douar Bourhma	February 2020	Well
P7	Kenitra	Douar Bourhma	February 2020	Well
P8	Kenitra	Kenitra	February 2020	Well
P9	Kenitra	Kenitra	February 2020	Water lagoon
P10	Fez	Douar Bsais	February 2020	Well
P11	Fez	Douar Bsais	February 2020	Well
P12	Fez	Douar Chejeaa	February 2020	Well
P13	Fez	Douar Chejeaa	February 2020	Well
P14	Fez	Taaounyat Birda , Btamat Boujlafa	February 2020	Well
P15	Fez	Taaounyat Elfarah	February 2020	Well
P16	Fez	Taaounyat Ghalya	February 2020	Well
P17	Fez	Moulay Yacoub	February 2020	Well
P18	Fez	Moulay Yacoub	February 2020	Well
P19	Fez	Moulay Yacoub	February 2020	Well
P20	Fez	Moulay Yacoub	February 2020	Well
P21	Fez	Moulay Yacoub	February 2020	Well
P22	Tit Mellil	Tit Mellil	May 2020	Well
P23	Tit Mellil	Tit Mellil	May 2020	Well
P24	Tit Mellil	Tit Mellil	May 2020	Well
P25	Sidi Kacem-Sidi Slimane	Sidi Slimane -Lkhawass	November 2021	Well
P26	Sidi Kacem-Sidi Slimane	Sidi Slimane -Lkhawass	November 2021	Well
P27	Sidi Kacem-Sidi Slimane	Zirara	November 2021	Well
P28	Sidi Kacem-Sidi Slimane	Zirara	November 2021	Well
P29	Sidi Kacem-Sidi Slimane	Sidi Kacem	November 2021	Well
P30	Larache	Larache Oulad Hamou Lghaba	December 2020	Well
P31	Larache	Larache Oulad Hamou Lghaba	December 2020	Well
P32	Larache	Larache Oulad Hamou Lghaba	December 2020	Well
P33	Larache	Larache Oulad Hamou Lghaba	December 2020	Well
P34	Larache	Larache Oulad Khssiss	December 2020	Well
P35	Brachoua	Enakhlat	January 2021	Well
P36	Brachoua	Enakhlat	January 2021	Well
P37	Brachoua	Enakhlat	January 2021	Well
P38	Brachoua	Enakhlat	January 2021	Well
P39	Brachoua	Brachoua	January 2021	Well
P40	Brachoua	Brachoua	January 2021	Well

B. Fluoride Analysis

Fluoride dosing measurements were performed using the potentiometric technique using the Fluorides specific ion electrode (HI-4110) at room temperature. HI-4110 is an ISE fluoride combination ideal for detecting free fluoride in drinking water, soft drinks, wine, plants, emulsion food products, electrodeposition acids.

The HI-4110 allows accurate measurement of the total concentration of fluoride. The Total Ion Strength Adjustment Buffer (TISAB) guarantees the pH and ionic strength of the sample solution between 5 and 8 and that the activity coefficient is constant.

The TISAB solution also preferentially becomes complex with various metal ions, such as aluminum and iron, which tend to form complexes with fluoride ions.

The TISAB solution thus ensures that these metal ions do not interfere, allowing an accurate measurement of total fluoride. Measuring all solutions with constant pH and ion force reduces the margin of error between measurements. The fluorides specific ion electrode HI-4110 can measure values from 1 M (0.02 mg/l) to F-saturated.

EHIs require calibration for accurate readings when performing measurement techniques such as direct measurement and incremental methods. The results are displayed directly on the ISE (HI-98191) ppm counter.

III. RESULTS

A. pH Variation and Fluoride Levels in Well Water in the Had Soualem-Berrechid Region

The pH values of the Region of Had Soualem - Berrechid range from 7.40 to 7.90 (Table II), and the fluoride levels found are 1.4 mg/l, 6.58 mg/l, 0.40 mg/l, and 2.55 mg/l in well waters P1, P2, P3, and P4, respectively, in the Berrechid - Had Soualem region (Table II). The P2 well located in the Had Soualem region has a higher content than the P4 and P1 wells. The P1 well has low fluoride levels. P2 and P4 well waters have above the WHO recommended standards. The P2 well is located on a farm near an aluminum manufacturing plant. The high fluoride level in the P2 well could be due to groundwater contamination by plant discharge. The P1 well has a low fluoride level that could be related to rain showers on the day of sampling. P3 and P4 wells are located on wheat, beet, and potato farms. The high fluoride content in the P4 well could be due to groundwater contamination through the use of fluoridated pesticides and fertilizers.

TABLE II: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN BERRECHID - HAD SOUALEM REGION

Sample	P1	P2	P3	P4
pH	7.90	7.40	7.87	7.69
Fluoride	1.40	6.58	0.40	2.55

B. pH Variation and Fluoride Levels in Well Water in the Kenitra Area

The pH values of well water in the Kenitra region range from 7.05 to 8.80 (Table III), and the fluoride levels found are 0.79 mg/l, 0.53 mg/l, 1.66 mg/l, 0.53 mg/l, and 0.29 mg/l,

in wells P5, P6, P7, P8, and P9, respectively, in the Kenitra region (Table III). The P7 well has a high fluoride level above the WHO recommended standards. The composition and depth of the P7 well are different from that of other wells. The water in the P7 well has a salty flavor.

TABLE III: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN KENITRA REGION

Sample	P5	P6	P7	P8	P9
pH	7.65	7.60	7.80	8.80	7.05
Fluoride	0.79	0.53	1.66	0.53	0.29

C. pH Variation and Fluoride Levels in Well Water in the Fez Region

The pH values of the Fez region range from 7.05 to 7.65 (Table IV), and the fluoride levels found are 0.73 mg/l, 0.68 mg/l, 0.45 mg/l, 1.64 mg/l, 0.77 mg/l, 0.98 mg/l, 0.85 mg/l, 0.087 mg/l, 0.09 mg/l, 0.14 mg/l, 0.18 mg/l in well waters P10, P11, P12, P13, P14, P15, P16, P17, P18, P19, P20, respectively, in the Fez region (Table IV).

The P13 well has a high value compared to other wells. This value is higher than the WHO reference standard. The P10, P14, P15, P16, P17, P18 wells have values above the recommended standards for children.

These wells are located on agricultural area (beets, potatoes farms), which could be the source of groundwater contamination with pesticides fluorinated and fluorinated fertilizers.

TABLE IV: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN FEZ REGION

Sample	P10	P11	P12	P13	P14	P15
pH	7.47	7.53	7.43	7.06	7.60	7.57
Fluor	0.73	0.68	0.45	1.64	0.77	0.97
Sample	P16	P17	P18	P19	P20	P21
pH	7.52	7.50	7.67	7.59	7.49	7.98
Fluor	0.98	0.85	0.87	0.09	0.14	0.18

D. pH Variation and Fluoride Levels in Well Water in the Tit Mellil Region

The pH values of well water in the Tit Mellil region range from 7.36 to 7.51 (Table V). Fluoride levels found are 1.29 mg/l, 1.47 mg/l, 0.5 mg/l in wells P22, P23, P24, respectively, in the Tit Mellil region (Table V). The fluoride content of the P23 well may be related to industrial contamination. The glass manufacturing and metal construction industry is highly developed in this region.

TABLE V: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN TIT MELLIL REGION

Sample	P22	P23	P24
pH	7.51	7.40	7.36
Fluoride	1.29	1.47	0.50

E. pH Variation and Fluoride Levels in Well Water in the Sidi Kacem-Sidi Slimane Region

The pH values in well waters in the Sidi Kacem-Sidi Slimane region range from 7.50 to 8.86 (Table VI), and the fluoride levels found in well water are 4.11 mg/l, 2.06 mg/l, 1.17 mg/l, 0.27mg/l, and 0.20mg/l in wells P25, P26, P27,

P28, and P29, respectively, in the Sidi Kacem-Sidi Slimane region (Table VI).

P26 well has a fluoride content above the WHO standard. P27 well has a fluoride content above the WHO child standards. P28 and P29 wells have normal fluoride levels. The high fluoride content in the P26 well could be due to contamination by the use of fluorinated pesticides and fertilizers.

TABLE VI: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN SIDI KACEM-SIDI SLIMANE REGION

Sample	P25	P26	P27	P28	P29
pH	8.86	7.91	7.90	7.70	7.50
Fluoride	4.11	2.06	1.17	0.27	0.20

F. pH Variation and Fluoride Levels in Well Water in Larache Region

The pH values of the Larache region range from 7.55 to 8.12 (Table VII), and the fluoride levels found are 0.75 mg/l, 0.013 mg/l, 0.004 mg/l, 0.09 mg/l and 0.20 mg/l, respectively, in the well waters of the Larache region (Table VII). P30 well has fluoride levels above standard children's standards. Wells in the Larache area have low fluoride levels. The samples were taken during the winter period with heavy rains that lead to the dilution of the water table.

TABLE VII: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN LARACHE REGION

Sample	P30	P31	P32	P33	P34
pH	7.72	8.12	7.60	7.55	7.58
Fluoride	0.75	0.013	0.004	0.09	0.20

G. pH Variation and Fluoride Levels in Well Water in Brachoua Region

The pH values of the Rabat Zaer region range from 7.13 to 7.75 (Table VIII), and the fluoride levels found in well water are 0.65 mg/l, 0.79 mg/l, 0.33 mg/l, 0.46 mg/l, 0.87 mg/l, and 0.27 mg/l, respectively, in the Brachoua regions (Table VIII). P38 and P40 wells have fluoride levels above recommended standards for children. The source of groundwater contamination may be related to the use of pesticides and fluorinated fertilizers.

TABLE VIII: pH VARIATION, FLUORIDE (MG/L) CONTENT OF WELL WATER SAMPLES IN BRACHOUA REGION

Sample	P35	P36	P37	P38	P39	P40
pH	7.51	7.36	7.20	7.13	7.75	7.67
Fluoride	0.65	0.79	0.33	0.46	0.87	0.27

H. Comparison of the Studied Regions

Berrechid-Had Soualem was the region with the highest fluoride concentrations, followed by Sidi Kacem-Sidi Slimane, Kenitra, Fez, Tit Mellil, Brachoua and Larache. However, Larache was the region with the lowest fluoride concentrations, followed by Fez then Sidi Kacem-Sidi Slimane, Brachoua, Kenitra, Berrechid-Had Soualem and Tit Mellil. The region with the highest average fluoride concentrations is Berrechid-Had Soualem, followed by Sidi Kacem-Sidi Slimane then, Tit Mellil, Kenitra, Fez, Brachoua

and Larache (Fig. 2).

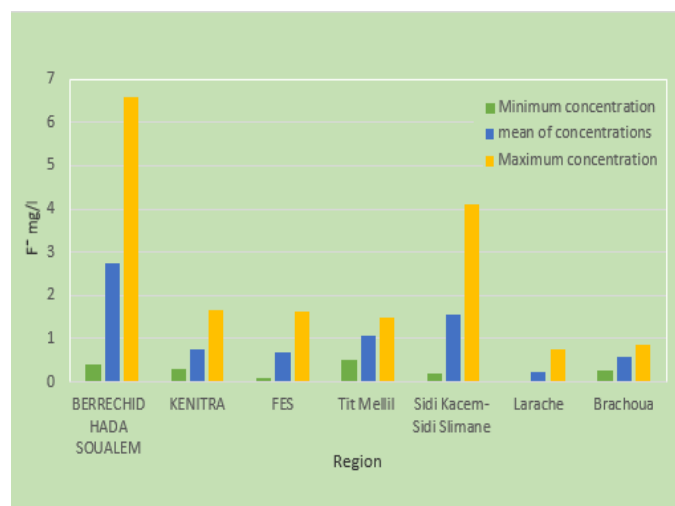


Fig. 2. Minimum - Average - Maximum fluoride (mg / l) concentration by region.

IV. DISCUSSION

Fluoride levels in well waters P2, P4, P7 and P26 in the Had Soualem, Berrechid, Kenitra, Sidi Kacem, Tit Mellil regions, respectively, exceed the WHO standard limit of less than 1.5 mg/day [1]. With a fluoride content of 6.58 mg/l, the P2 well water sampled from the rural region of Had Soualem had the highest fluoride value, making it the main cause of dental fluorosis in this area. We suspected that the high fluoride level in this well water was due to pollution from an aluminum plant. Barbier's research revealed that the naturally occurring presence of fluoride in the water table could be attributed to geological inputs, anthropogenic and industrial contaminations [14]. In sources from environments where calcium sulfate or magnesian fibrous clays are found, fluoride would be located in clays as a substitute for the OH group. These fossil geological deposits would be, for example, in France, those of the Terminal Triassic and those of the Terminal Eocene. The high groundwater fluoride values appear to be associated with the weathered basement complex containing biotite, which is a probable source of fluoride. The other suspected sources of fluoride in Nathenje groundwater could be due to the dissolution of hornblende, fluorite, and amphibole, all of which are reported to occur in rocks and soils in this area [9].

The P15, P16 and P17 wells in the Fez regions have fluoride levels above children's reference standards. Consumption of these waters could cause dental fluorosis in children [15] and animals [16]. The P13 well, which is located in the same region, exceeds the standards set by the WHO, with a value of 1.67 mg/l. This region is known as an agricultural area, and this well is located in agricultural land where the use of pesticides is excessive. This use includes treatments on cereals, oilseeds, vegetable crops, orchards. The study of A. Naamane revealed that in some Moroccan regions, 100% of farmers use pesticides, and 75% of agricultural producers are not aware of how these products are used [11]. Perhaps it is the use of fluorinated pesticides that seep through the soil and contaminate, increasing the level of fluoride in its water [17].

In the Sidi Kacem and Sidi Slimane regions, it was observed that a large number of the population suffers from dental fluorosis with different degrees; this is due to the presence of a well in the area, in which the fluoride reaches 4.11 mg/l, this value, which exceeds the standards set by WHO, confirms that water is the main cause of dental fluorosis in this region.

Wells in rural areas of Tit Mellil, Larache, Brachoua do not exceed the WHO standards, indicating that water is not the main source that causes dental fluorosis in these areas. It depends on the diet of the population [18]. Therefore, the daily consumption of well water in the different regions studied with fluoride concentrations ranging from 0.5 mg/l to 1 mg/l and its usage in the preparation of tea drinks, could cause the onset of dental fluorosis in children. The amount of fluoride contained in the water is in addition to that contained in some varieties of green and black teas that are widely consumed by the population in Morocco [19]. Children who eat cereals reconstituted with fluoridated water ingest a substantial amount of fluoride in this way [20]. Fruit juices and drinks are often prepared with water that is already fluoridated [21]. In their study, fluoride ingestion in children and babies results in a significant increase in the incidence of fluorosis, unless intervention measures are adopted [22].

Well water used for irrigation, which contain the fluoride, can indirectly affect the increase in the daily consumption balance of fluorine, this is because of the accumulation of F and biochemical constituents in crops and vegetables [23]. Eating these crops while using these wells for drinking, may increase the risk of infection with fluorosis.

There are many factors responsible for the high or low concentration of fluoride in well water, such as the depth of the well. According to Y. E. Ibrahim study, water in the depth well (45 m) was shown to have a low and very stable fluoride concentration of 0.25 ppm, while the depth well (93 m) had a fluoride concentration ten times higher and slightly variable (2.56 ppm) [24]. Another factor that affects the concentration of fluorine is rain precipitation, which lowers the level of fluorine in wells. Fluoride concentrations for the two seasons were significantly different from each other [9].

A group of samples was taken in the winter season, and on rainy days, so the fluorine value in these wells may be much greater if it was taken in the non-rainy seasons.

The study carried out by R. El Jaoudi, who conducted analyses on wells in rural areas, in which he proved that the concentration of fluoride in regions that contain phosphate is higher than that of regions that do not contain phosphate [25]. The rest of the works published are focused specifically on regions that contain phosphate. Such as A. Moufti, who found in his research, a concentration of fluoride ranging from 2.4 to 4.4mg/l in the region of Youssoufia [26].

On the other hand, this rate is high compared to that found in a study conducted in 2017 in the region of Khouribga by H. Lebrahimi, which revealed low values ranging from 0.126 and 0.924 mg/l [27]. The increasing risk of fluorosis in these regions is caused not only by excessive fluoride levels in the water meant for human consumption but also by the inhalation of phosphorus dust and the ingestion of food made with fluoridated water [28].

In Morocco, efforts are underway to provide villages with

safe drinking water, but despite this, many rural areas still do not have piped drinking water, forcing them to use well water for daily consumption, leaving them vulnerable to a variety of health problems.

V. CONCLUSION

The presence of fluorides in groundwater could be a constraint for the exploitation of these waters for agriculture and human consumption. At high concentrations, they pose risks to human health, such as dental and bone fluorosis. The presence of high levels of fluoride characterized in well water in different regions of Morocco could be linked to the contaminations of natural or human origins (industrial or agricultural by the use of fluorinated pesticides). Compliance with good agricultural practices minimizes the risk of pesticides outside cultivated plots. Improving water quality is, therefore, a major challenge. The likelihood of finding traces of pesticides in groundwater or surface water should be minimized. A mineralogy study of the aquifer is needed to clarify the depths of fluoride-high lithological facies. The need to be aware of the importance of water supply, sanitation, and personal hygiene is essential to promote the development of the localized population in rural areas.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

IE supervised the analysis and wrote the paper and YB contributed to analysis under the advice and follow-up of CO, who also contributed to data collection. CO, AM and AE supervised the work, proofread and corrected the revised version. EB supported this work to improve the scientific quality of the manuscript. All authors had approved the final version.

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