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Fluoride Contents in Groundwaters and the Main Consumed Foods (Dates and Tea) in Southern Algeria Region

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Abstract: In the South of Algeria, the dental fluorosis is a "silent" epidemic spreading within the populations. The present study was aimed to determine the fluoride content in the water, dates and the tea that are consumed. The results reveal that 35% of the water wells had excessive fluoride levels (> 1.5 mg L^{-1}). They are the waters of the Eastern areas that present the highest contents. These are, therefore, the areas where dental decay is the most widespread pathology. Although the dates and the tea are considerable sources of fluoride, with a daily contribution of 10 and 20%, respectively, it is the water which contributes up to 70%. For these three sources, the daily intake of fluoride ingested by an adult exceeds the proposed safe threshold for fluoride intake of 0.05-0.07 mg F.kg.day^{-1}. The defluoruration with hydrated lime shows that starting from one liter of water of 2.3 mg L^{-1} of fluoride; the content could be brought back to 1.2 mg L^{-1}. Maximum efficiency for one liter of treated water is 1.1 mg of fluoride by using 200 mg of lime.

Key words: Sahara, groundwater, date, tea, fluoride, defluoruration

INTRODUCTION

The fluoride is a beneficial oligo-element, essential to the growth and the upholding of bone tissues and teeth. As carioprotector (Arbab Chirani and Foray, 2005), it prevents the dental decay where its drinking water concentration is shown inversely correlated with the dental decay (Acharya, 2005): The fluoridation of water proved to be a potential solution (Gillespie and Baez, 2005) for the water deprived of fluoride. Many countries such as the United States (Miller-Ihli *et al*., 2003), Brazil (Aurélio Peres *et al*., 2004), go until fluoridate the water of consumption. Although it is beneficial and its chronic intoxication is relatively rare (Warren and Levy, 2003), an excessive fluoride intake, beyond acceptable levels (1.5 mg L^{-1}), leads to the dental and skeletal fluorosis (Onyango *et al*., 2004). Moreover, acute fluoride intoxication can have neurological complications (Long *et al*., 2002), urinary stone formation (Singh *et al*., 2001) and a hypocalcaemia (Pettifor *et al*., 1989) as consequences on the endemic patients. Even if it does not seem to have any link between fluoride and cancer (Harrison, 2005), the number of the cancer patients observed in the area of study, leads to think, in spite of the absence of the clinical data that a risk is possible. Research of several investigators in the last ten years has proved that life-long impact and accumulation of fluorides causes not only human skeletal and teeth damage, but also changes in the DNA-structure, paralysis of volition and cancer (Veressinina *et al*., 2001). Certain experiments carried out in Japan (Tohyama, 1996) the United States (Takahashi *et al*., 2001) and Taiwan (Yang *et al*., 2000) proved that various types of cancer were actually associated to fluoride. Classified as third worldwide, the dental and osseous fluorosis is the most widespread pathology in the world (Badet and Richard, 2004). It remains a problem of public health in many developing countries (Jones *et al*., 2005). The African continent counts among the most affected regions in the world where several homes of endemic osseous fluorosis were identified (Wondwossen *et al*., 2003). In the South of Algeria, we deal with a "silent" fluorosis among the populations, which has been reported through a great deal of work and epidemiologic investigations (Pinet *et al*., 1961; Poey *et al*., 1976). Because of the lack of information, as far as all the sources of fluoride are concerned, the present study which was mainly aimed to evaluate the daily fluoride intake and to determine its distribution in water and mostly consumed food, in order to locate the areas of risk and to seek a short-term solution, was conducted in a vast area (Biskra, Ouargla, El-Oued and Ghardaïa), where groundwaters (only source of drinking water), the dates and the tea (food practices) seem to be the primarily epidemic agents.

MATERIALS AND METHODS

Choice of the area of study: In 1980, a study of the National Institute of Algerian Public health raised the importance of the fluorosis in the Septentrional Sahara.

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Within the framework of the present study, the high number of the populations involved and their food practices, but especially fluoride quality of consumed water directed the choice of the studied zones. The fluoride intake was assessed through analyses of the fluoride contents present in water, tea and dates, at a community known to have the dental fluorosis. The four Wilayate (districts) of this arid band are Biskra, Ouargla, El-Oued and Ghardaïa. These areas are populated and show an important rate of affection compared with others of the Septentrional Sahara.

Choice of the material of study: For practical reasons, the material of the study consists of water, dates and green tea. The dates are in fact the main production of the area and hence their relatively wide consumption; similarly tea is a much appraised drink within the Saharian populations. After the meals, the ritual of the tea is a traditional practice. Each consumer must take 3 cups of approximately 30 mL each one. In fact, the consumption water is an inevitable element for the present study especially in arid environments. The above mentioned three food components may form a basis to establish a first balance of fluoride ingested by the population of the studied areas, in order to sort out the localities of high risk and to suggest a short-term solution. The studied water comes from 16 wells exploiting two main hydrogeologic reservoirs: The Continental Intercalary (of a depth >1200 m with hot water 55-60°C) and the Complex Terminal (a depth ranging between 100 and 500 m).

Analytical techniques: The total fluoride contained in water, dates and tea was dosed using a specific fluoride electrode (E lit 8221 F-55907). The samples were mixed with a Total Ionic Strength Adjustment Buffer (TISAB) to prevent interference with other ions (AL^{3+}, Fe^{3+}, Cu^{2+}, Ca^{2+}, Si^{2+}) during the fluoride measurement (Hao and Huang, 1986). The potentiometer readings were compared with a calibration curve ranging between 0.1 and 10 mg L^{-1}. The other physicochemical characteristics of water, such as calcium and magnesium, were dosed using the EDTA titrimetric method and bicarbonates by the volumetric method. The chlorides contents were determined according to the Mohr method. The pH and the electrical conductivity were measured respectively by a Hannas-212 pH-meter and a conductimeter WTW LF-315.

RESULTS AND DISCUSSION

Fluoride contents of groundwater: The fluoride of water varies with the source location. The contents vary from 0.4 to 2.3 mg L^{-1} (Table 1). The mineral content typically reflects the nature of geological formations through which the water comes in contact, probably resulting from the process of dissolution-precipitation of fluorinated minerals present in the aquifer. Concentration in fluoride shows a positive correlation to magnesium (R^2 = 0.448), to electrical conductivity (R^2 = 0.641) and to the alkalinity of water (R^2 = 0.803), thus suggesting a common geological origin. In addition, they are the waters of the Eastern regions (Biskra, Ouargla and El-Oued) which present the highest contents (>1 mg L^{-1}). These are consequently the areas where dental decay is the most widespread pathology. The analysis highlights that 24 % of exploited wells had contents lower than 1 mg L^{-1} whereas 41% contained between 1 and 1.4 mg L^{-1}. In terms of WHO directives, 38% of wells involving contents between 1.5 and 2.3 mg L^{-1} exceed the standard fixed quality for drinking water (1.5 mg L^{-1}). A rate of 57.5% with levels higher than 1.5 mg L^{-1} was already observed in the region (Djellouli *et al*., 2005). In comparison with the contents of 3 to 5 mg L^{-1} already obtained in the region of El-Oued (Poey *et al*., 1976) this difference probably seems to be related to the precision of the employed analytical technique. In addition, the concentrations of fluoride can change with the source of the water and the processing site. The groundwater lithology (Kim and Jeong, 2005) and the deep water tables exploited (Bhagavan and Raghu, 2005) are the series of factors which contribute to varying the fluoride contents in water.

Optimal dose of fluoride in water: In the studied zone, the major problem of the population is the presence of fluoride in water; approximately 70% of the entire fluoride intake is derived from consumed water. The determination of an optimal amount of reference is thus more than necessary. This later is calculated according to equation 1 (Galagan and Vermillon, 1957), which gives the optimal amount (D) as a function of the maximum average temperature of the air (T_m in °F):

$$D(mg.L^{-1}) = \frac{0.34}{-0.038 + 0.0062.T_m} \quad (1)$$

For an annual average temperature of 22.7 °C (72.86 °F), recorded in the last 20 years (1984-2004), the optimal fluoride amount, for drinking waters of these areas, is of 0.8 mg L^{-1} (Table 2).

Taking into accounts the hyper arid climatic conditions; transpiration and sweat involve high water consumption and consequently an accumulation of fluoride in the organism. In addition, the fluoride contribution by food (dates and tea in particular), the strong daily thermal amplitude and the high toxicity of fluoride for the diabetics (Banu Priya *et al*., 1997) make the application of this average amount inappropriate, even for the populations living in the areas where water is deprived of fluoride. Indeed, for most of the year, this amount exceeds that

Table 1: Chemicals composition of groundwaters study

Districts (Wilayate)	Municipal	EC mS.cm^{-1}	pH	TH °F	Contents (mg L^{-1})				
					Ca^{2+}	Mg^{2+}	HCO_3^-	Cl^-	F^-
Ouargla	Matmoura	4.25	7.40	300	782	251	171	1449	2.02
	Tazgrart	2.30	7.68	176	216	293	281	735	1.37
	Gharbouz	3.35	7.25	156	320	183	159	1033	1.07
	Touggourt	5.32	7.46	469	273	86	134	666	1.86
	Blidet Amor	4.35	7.50	396	269	106	171	533	2.30
Biskra	Sidi Okba	3.95	7.20	308	960	163	293	1390	1.67
	Biskra city	5.07	7.21	276	752	211	281	1985	1.68
	Oued-Biskra	2.50	7.46	240	272	413	256	93	1.20
	Mlili Hamra	2.43	7.04	288	928	135	61	417	1.42
	Ourlal	2.65	7.43	312	960	173	49	457	1.60
El-Oued	Al Chehada	3.82	7.24	154	592	144	159	1251	1.26
	Al Bayada	3.44	7.28	188	240	307	159	1033	1.18
	El Araire	2.47	7.64	218	592	168	61	219	1.32
Ghardaïa	El-Goléa	2.04	6.81	160	352	173	183	754	0.40
	Métlili	0.33	7.66	29	64	31	146	119	0.39
	Ghardaïa city	2.02	7.51	167	365	185	191	651	0.38

Table 2: Optimal dose (mg L^{-1}) of fluoride of the consumption waters

Month	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Aver.
T °C	11.1	13.7	17.2	21.1	26.2	32.0	34.9	34.9	30.0	23.7	16.1	12.0	22.7
T °F	51.85	56.66	62.96	69.98	79.16	89.6	94.82	94.82	86.0	74.66	60.98	53.59	72.86
Dose	1.2	1.1	1.0	0.9	0.8	0.7	0.6	0.6	0.7	0.8	1.0	1.2	0.8

recommended in hot areas, which is about 1 mg L^{-1} (WHO, 1993). The calculated optimal level needs to be corrected. On focusing on the high thermal amplitude during the day, ranging between 10 and15°C (50-59°F), a factor of correction of 0.7 is calculated. Thus, the average optimal fluoride amount for the area must be, after correction, between 0.5-0.6 mg L^{-1} instead of the calculated 0.8 mg L^{-1}.

Content fluoride in food: The fluoride content contained in dates: 2.9 mg.kg^{-1} is high and testifies the natural geochemical origin of fluoride in water and the grounds of the zone of study. Palm trees-date palms (*Phoenix dactylifera* L.) shows an accumulation of fluoride naturally present in these waters and grounds of the area. The tea leaves, favorite drink of the inhabitants of the Algerian South, are rich in fluoride which is easily released during infusion with a concentration of 1.81 mg L^{-1} in the first preparation and 1.45 mg L^{-1} in the second. Although the concentration decreases during the preparation of the tea, an adult consumption once a day leads to a total contribution of 3.3 mg L^{-1}.

Without taking account of the other contributions, a regular consumption of the tea can develop the fluorosis at the people who consume great quantity (Wong *et al*., 2003). In China, research (Cao *et al*., 2003) showed that the tea had much more harmful effects on human health compared with those of water.

Total consumed fluoride balance assessment: It is practically difficult to establish a balance of daily-ingested total fluoride because the sources are variable. It is almost present in all food at variable contents. The cereals (Haikel *et al*., 1986), the fish (Malde *et al*., 1997), salt (Martinez-Mier *et al*., 2005), the tea (Cao *et al*., 2006) and the drinks (Jimenez-Farfan *et al*., 2004) are the richest food-stuffs in fluoride. With the absence of a complete balance, the establishment of a partial balance, is quite necessity, as a guide and a basis reference. Considering the average daily consumption of water (1.9 L day^{-1}), tea (0.4 L day^{-1}) and dates (200 g day^{-1}), the total daily fluoride, in mg/day, ingested by an adult, is calculated according to Eq. 2 (Heikens *et al*., 2005):

$$Total\ daily\ consumption = \sum_i C_i I_i \qquad (2)$$

Where *i*, represents the source (water, tea and dates), *C*, the concentration in the source (mg L^{-1} or mg g^{-1}) and *I*, is the consumption of the source (g/day or L/day).

The results of Table 3 reveal that the daily fluoride intake coming from drinking water varies with the season, from 1.2 in winter to 7.6 mg day^{-1} in summer (4.4 mg day^{-1} as an average); of 1.2 mg day^{-1} for the tea and 0,6 mg day^{-1} for dates. A person living in these areas ingests between 3 mg day^{-1} and 9.41 mg day^{-1} of fluoride. Although this assessment is presented as an indication, it gives us an eloquent idea of the ingested fluoride mass. Approximately 70% of the entire fluoride intake derives from that of consumed water; the tea and the dates represent, respectively 20 and 10%

Table 3: Adult daily average fluoride consumption

Sources	Average sources consumption	$[F^-]$(mg day^{-1})
Dates (kg day^{-1})	0.2	0.58
Water (L day^{-1})	0.51 (winter) – 3.3 (summer)	1.17-7.59
Tea (L day^{-1})	0.38	1.24
Total (mg day^{-1})		2.99-9.41
Total (mg.kg.day^{-1})		0.05-0.16

of the daily total contribution. If one transposes these results as function of the average weight of an adult (60 kg for the average of the population of the studied areas), this one consumes in summer was twice more than the proposed safe threshold for fluoride intake of 0.05-0.07 mg F.kg.day^{-1} (Levy, 1994). Nevertheless, for the inhabitants of the areas of South-west (Ghardaïa, Métlili, El-Goléa), whose water is deprived of fluoride (=0.4 mg L^{-1}), the food contributions (dates and tea), seem to compensate the deficit and even be enough for an effective carioprophylaxis. The tea is in fact a localised fluoride delivery for the oral cavity (Simpson *et al.*, 2001) where 34% of fluorides are maintained after rinsing by the tea; whereas the dates can serve bone tissues. In addition, in Wilayate of Biskra, Ouargla and El-Oued, one note that the optimal amount recommended is exceeded, particularly in summer and a defluoruration of water becomes then necessary.

Waters défluoridation: For a population where 70% of the entire fluoride intake derives from water, defluoruration is the only possible alternative to prevent the fluorosis. Admittedly, many processes were developed, but their high cost limits their use in the developing countries. The lime treatment is, according to many authors (Sorg, 1978) the only economic, effective treatment for water with low fluoride content and much easier to implement. It presents the advantage of reducing simultaneously the carbonated waters hardness. By treating a water of 2.3 mg L^{-1} of fluoride, by addition of hydrated lime (purity 97% out of $Ca(OH)_2$), we were able to bring back the content to 1.2 mg L^{-1}. Maximum efficiency to defluoride one liter of water is 1.1 mg of fluoride by 200 mg of hydrated lime (Fig. 1), that is to say an output approximately 52% under our experimental conditions. This poor yield seems to be allotted to the limit of the effectiveness of the process as underlined by certain authors (Wang and Reardon, 2001).

Under our conditions, the co-precipitation with magnesium seems to dominate precipitation in the form of Calcium Fluoride (CaF_2). The good correlation between residual fluoride and precipitated magnesium highlights that only the co-precipitation is to be considered in this type of water (Fig. 2). It is shown (Savinelli and Black, 1958) that in waters containing magnesium, when treated by hydrated lime, the

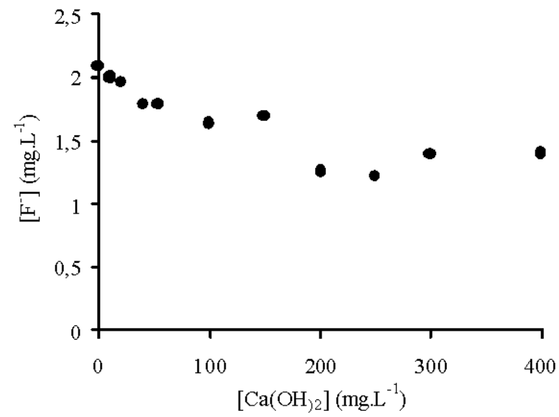

Fig. 1: Relationship between residual fluoride content and the hydrated lime added

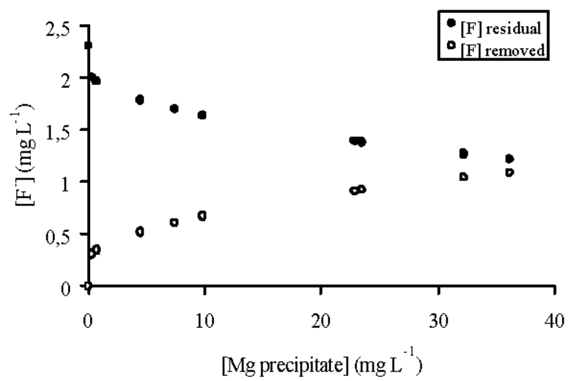

Fig. 2: Relationship between fluoride (removed and residual) and magnesium precipitate

fluorides are eliminated by adsorption on the hydroxide magnesium flocs.

CONCLUSION

The fluoride content of studied groundwaters lies between 0.4 and 2.3 mg L^{-1}. The results highlight that 62% of water drilling wells have fluoride contents lower than 1.5 mg L^{-1} and 38% of these exploited wells involve contents higher than the directives of WHO (1.5 mg L^{-1}). They are water of Eastern areas which present the highest contents and consequently the areas where the dental decay is spread, compared to the Southern West area. The optimal amount calculated for water exceeds, for the majority of the year that recommended in hot regions. Approximately 70% of the entire fluoride intake derives from consumed water, the tea and the date's account, respectively 10 and 20% of the daily total contribution. The defluoruration with hydrated lime shows that starting with one liter of water, 2.3 mg L^{-1} of fluoride could be brought back to 1.2 mg L^{-1} thanks to the use of 200 mg hydrated lime.

Maximum efficiency is 1.1 mg of fluoride for 200 mg of hydrated lime. The correlation between the residual fluoride and the precipitated magnesium shows that only the co-precipitation is to be considered in this type of water. From the point of view of public health, the oral health of the populations is an important challenge and should be a priority. Although the fluorosis is irreversible, it could be prevented in the future by reducing the consumption of tea and dates by establishing a program of dental medical education.

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