Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 74 (2015) 294 – 300

Energy Procedia

International Conference on Technologies and Materials for Renewable Energy, Environment and Sustainability, TMREES15

Distribution and removal of fluoride ions in the drinking waters in the Algerian South (Ouargla as a showcase)

Kais Baouia[a], Amar Messaitfa[b]

[a] *Assistant Professor, University of Ouargla.. University of Kasdi Merbah Ouargla. Laboratory of water Engineering and Environement within the Saharian milieu. Faculty of Nature and Life Sciences. Ouargla 30000, Algeria. E-mail: kais_baouia@yahoo.fr*

[b] *Professor, Research Laboratory of water Engineering and Environement within the Saharian milieu. Faculty of Nature and Life Sciences. Ouargla 30000, Algeria.*

Abstract

In certain countries, the Algerian South in particular, where the scarcity of drinking water resources of good quality has constrained the local populations to consume the underground waters that are rich in fluoride. Fluoride constitutes an essential component for the human body in moderate rates, between 0.5 to 1.5 mg/l of the consumed water. Nonetheless the population of the Southern Algeria is characterized by a very particular diet marked by tea and dates consumption that contain fluoride in an unidentified and accurate amount within the absence of a full knowledge of all fluoride sources .

The present study aims at evaluating the daily amount of fluoride taken and to determine its distribution in water and the main consumed food, as well as localizing the zones of risk and seeks at the end a short term solution to this matter.

This work focuses on the evaluation of the rate of fluoride contained in the diet. The dosage of fluoride of the two main watertables feeding the region of the study shows that the rates exceed by far the standards of the WHO. They are comprised in rates oscillating between 1.23 to 2.01 mg/l, as for the main food stuff, tea and dates are respectively (0.86 – 2.10 mg/l) and (10.48 – 18.24) mg/kg. Actually four methods of fluoride removal from water have been tested, as for the method using Aluminum sulfate and Lime, they provide good findings compared to the other methods.

Key words: Fluoride, Ground water, fluoride removal, Lime, aluminium sulfate;

Peer-review under responsibility of the Euro-Mediterranean Institute for Sustainable Development (EUMISD)

* Corresponding author. Tel.: +213-0778-068072; fax: +213-0778-068072.

E-mail address: kais_baouia@yahoo.fr

Peer-review under responsibility of the Euro-Mediterranean Institute for Sustainable Development (EUMISD)
doi:10.1016/j.egypro.2015.07.610

1. Introduction

In south of Algeria, dental fluorosis constitutes a silent epidemy that the local population suffers from. the physicochemical analysis of waters of four regions (Ouargla, Touggourt, El Oued and Biskra) of the Northern Sahara, reveal that the dosage of fluoride exceeds frequently the standards of the WHO (Messaitfa, 2007). The risk of fluorosis rise when it took into consideration the amount of fluoride provided by the specific day to day diet of the region of study (tea and dates cosumption). On the physiological basis, fluoride is a necessary nutritive and beneficial for human organism and growth as well as the maintenance of the bonal tissues and teeth, It prevents teeth decay where its concentration in water is inversely correlated with dental decay (Acharya, 2005) : fluoride concentration in waters has been a potential solution to the waters lacking fluoride. Many countries such as the USA (Aurélio Peres and Al., 2004), go beyond up to adding fluoride to the drinking waters. Although beneficial, passing the allowed limits (1.5 mg/l) (Chavaissieux P; Meuinier PJ, 1995), fluoride could lead to dental and bonal fluorosis (Onyango and Al., 2004). In the other hand, a cute intoxication of fluoride could lead to neurological complications (Long and al., 2002), urine stone formation (Singh et al., 2001) and a hypocalcaemia (Pettifor and Al., 1989) of the endemic patients. Though it seems to have no link between fluoride and cancer (Harrison, 2005), the number of the observed cancerous people in the region of study shows that even within the absence of clinical data that a risk is possible. Some experiences made in Japan (Tohyama, 1996), in USA (Takahashi and Al., 2001) and in Taiwan (Yang et al., 2000) have proven that many types of Cancer have been frankly associated with fluoride. In Southern Algeria, dental fluorosis is a "silent" epidemy that local population suffers from. the physicochemical analysis of the waters of four regions (Ouargla, Touggourt, El Oued et Biskra) of Northern Sahara, shows that the concentration of fluoride frequently exceed the standards of the WHO (Messaitfa, 2007). The fluorosis risk rises when it took into consideration the amount of fluoride contained in the very specific diet of the region of study (tea and dates consumption). The aim of this study is to determine the fluoride concentration in the drinking waters for the watertables feeding the networks of drinking water supply, and that of the different food stuffs locally produced. In order to efficiently and economically remove fluoride within the waters, four methods have been proposed (lime, Aluminum sulfate, the ferrous and ferric sulfate).

2. Materials And Methods

2.1. Localizing the study zone

The region of Ouargla is situated in Southern-East Algeria, it stretches over a surface of 211980 Km², according to 2010 statistics, the population is estimated over 633967 inhabitants, distributed over 21 municipality. It is limited by Djelfa in the North and El Oued, in the South by the city of the city of Illizi and Tamanrasset, in the Ouest by Ghardaia and in the East by Tunsia (Figure 1). The basin of Ouargla is caracterized by an arid climate that was very hot (exceeding 42 °C in July) and cold in winter (3 .02 °C in January) with a very high thermic amplitude, a very weak and irregular flow (12.5 mm in January), a very high evaporation (340 mm in July). The region of the study presents quite important potentials represented by three types of watertables, the first is free, corresponding to the watertable, the remaining two are captive, corresponding to the terminal complex watertables terminal (Mio-pliocene , Senonien) of a depth of almost 60 to 250 m respectively with an exploited flow between 10 and 45 l/s , and the Albien water table that is caught from a depth oscillating between 1300 to 1400 m and exploited with a flow of 100 to 120 l/s.

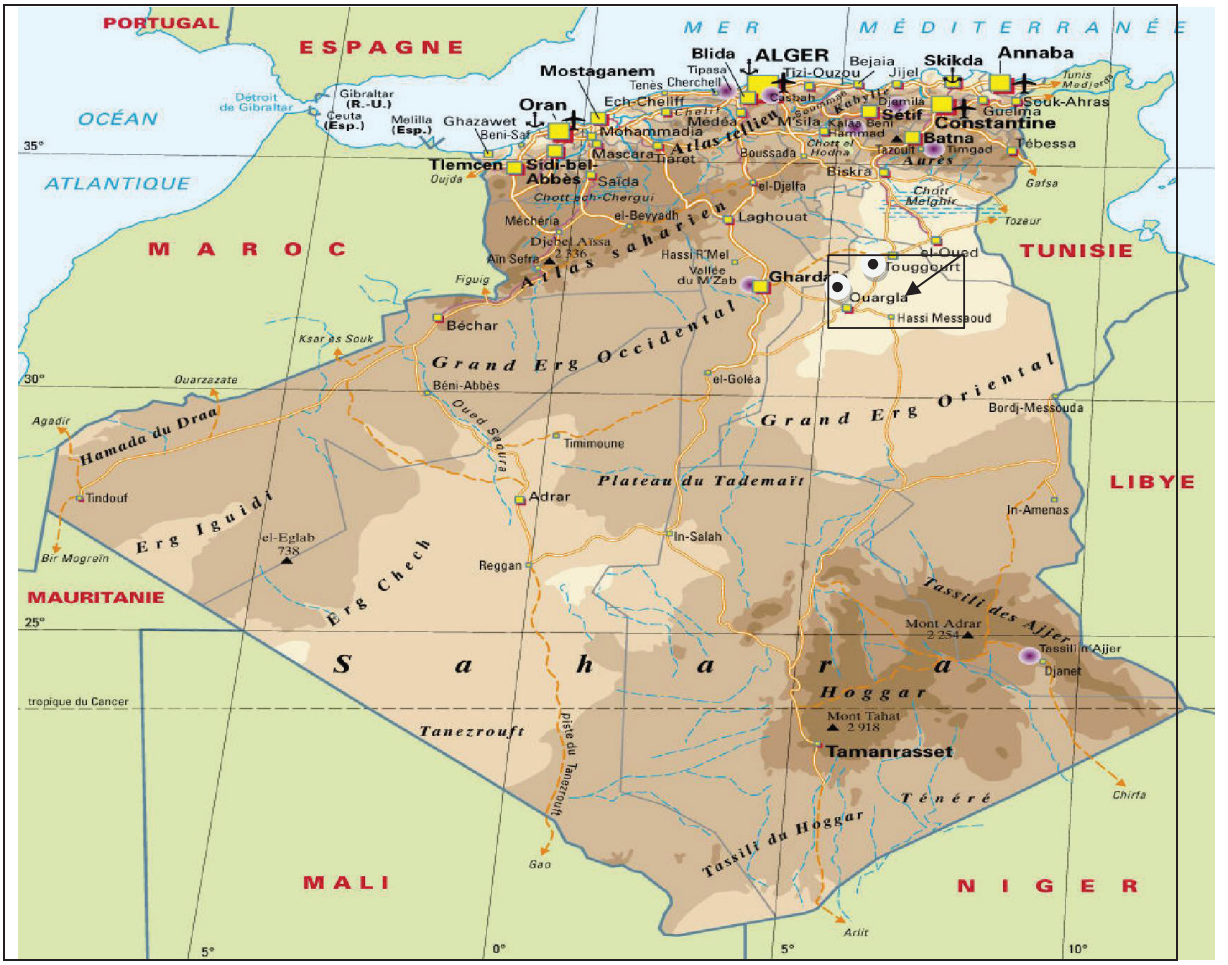

Fig 1. A Map of the situation of the region of Ouargla**.**

2.2. Methods

The analysis of fluoride dosage de fluorure were made on water samples of 25 drills of different watertables (Mio-pliocene, Senonien, Albien), the consumed food stuffs (dates, lentil, potato, semolina, carrots, pumpkin) and tea. The chosen method of analysis so as to determine the content of the chemical elements (F^-, Ca^{2+}, Mg^{2+}, Fe^{2+}, Al^{3+}) is the spectrophotometer method DR 2800. Fluoride tenor of consumed food is obtained by decomposing the organic substance by at a temperature of 500 °C during two hours time (Bock 1979, Messaitfa 2007), once cooled, the ashes are smoothly crushed until reduced into a homogenic powder. The mineralization consists in putting an amount of 5 g of powder with a relatively concentrated chlorhyde acid volume, after 24 hours time reaction, the mineralization process is adjusted on 100 ml with distilled water, afterwards filtered and the obtained solution is ready for analysis.

Our experiment consists of utilizing the removal method, with lime, Aluminium sulfate, ferous and ferric by adding growing doses of the reaction to the taken water sample, from the drill of "Beldet Amor" (Touggrout), of 200 ml to be treated by the four above mentioned methods. The purpose of reducing the studied water fluoride dosage, the process in which lime is used aims at minimizing the water hardness and reducing the fluoride dose in the same time. The process of softening relying softening lime only consists in reducing calcic carbonate that is to say the precipitation of calcium related to carbonate (HCO_3). The optimal dose in lime necessary to the softening, in equivalence with $CaCO_3$, is calculated according to the equation (Raymond, 1990): $[Ca(OH)_2]= [HCO_3^-]$.

3. Results And Discussion

3.1. Fluoride of water consumption of the region of Ouargla

Considering the results of twenty-five (25) samples, fluoride tenor for the Mio-pliocene watertable varies from 1.62 to 2.01 mg/l, as for the Senonien, it presents tenors less increased than 1.15 to 1.62 mg/l, finally the Albien watertable, they oscillate from 0.79 - 0.96 mg/l. In order to better illustrate the variability in the space a map was realized (tableau 1).

Table1. Fluoride Tenor of the drills of the region of Ouargla.

watertable	Drill	PH	TH (F°)	Ca^{2+} (mg\l)	Mg^{2+} (mg\l)	HCO_3^- (mg\l)	Cl^- (mg\l)	SO_4^{2-} (mg\l)	F^- (mg\l)
Mio -pliocene	Blidet Amour	7.5	187	529.05	134.5	518.5	1014.31	1475	2.01
	Gharbouz	7.47	94	220.44	94.74	133.03	298.23	625	1.86
	Elgoog	7.32	167	388.7	170.37	170.12	856.96	1075	1.83
	Sokra	7.37	105	280.56	85.07	120.06	375.78	725	1 .74
	Frane	7.41	112	272.54	106.93	229.41	489.6	950	1.98
	Bour Elhaicha	7.64	80	196.39	75.34	250.25	422.33	525	1 .62
Senonien	Beni Thour	7.22	145	456.91	75.33	130.71	908.88	1375	1.58
	Sellise	7.13	99	260.52	82.63	129.83	483.92	825	1.25
	Mekhadma 2	7.36	115	280.56	109.37	553.35	661.66	1050	1.44
	Mekhadma 3	7.10	110	244.48	119.09	518.5	665.3	1350	1.62
	Université	7.15	115	284.56	106.94	147.32	709.4	875	1.47
	Ain Baida	7.55	86	204.41	85.06	199.63	392.7	525	1.58
	F 22 HMS	7.45	115	244.84	131.24	128	199.95	675	1.35
	El bour 1	7.25	105	248.49	104.51	125.35	433.31	850	1.46
	Ifri	7.42	98	264.52	77.77	129.83	430.15	775	1.58
	Sidi Khouiled 1	7.36	128	300.6	128.81	136.47	616.91	1025	1.23
	Sidi Khouiled 2	7.39	100	260.52	85.06	128.2	533.21	1050	1.37
	Said Otba	7.27	95	224.44	94.79	158	313.44	800	1.6
	Hassi Ben Abde	7.73	152	396.79	128.81	135.07	516.24	1050	1.25
	Hassi Miloud	7.41	88	248.49	63.19	112.61	467.18	650	1.58
Albien	Elhadeb 1	7.26	91	192.38	104.51	589.11	396.1	650	0.93
	Elhadeb 2	7.36	88	184.36	102.08	597.91	392.29	625	0.96
	Blidet Amour	7.34	102	232.46	106.94	566.42	345.37	675	0.89
	Sidi Mehdi 2	7.33	96	244.48	85.06	541.53	514.17	850	0.85
	Sidi Slimane	7.36	105	240.48	109.37	165.65	480.58	625	0.79

According to the results of fluoride dosage of drinking water of the exploited underground watertable in the region of Ouargla, all the drills of the Mio-pliocene contain high condensations compared to the standards of the WHO (1.5 mg/l), nonetheless the waters of the Albien watertable are characterized by an acceptable dosage of fluoride but that of the Senonien watertable exceeds the standards of the WHO more than 38% of fluoride..

3.2. Fluoride removal from drinking water in the region of Ouargla

In many countries, different processes of fluoride removal from water have been chosen and their optimization has been developed whether in the laboratory, pilot stations or even in the industrial scale. The effectiveness of these processes varies according to the nature of the process and the characteristics of the treated water. The most adopted techniques are the chemical process by precipitation, the classical physical and chemical process (coagulation-floculation, absorption, ions exchange) and membranes (electrodialysis, inverse osmosis and nanofiltration). The results of the four tested methods (lime, aluminum sulfate, ferrous and ferric) are given in the tables 2,3,4,5.

Table 2. Results of treatment by lime ($Ca(OH)_2$)

Parameter / sample N°	1	2	3	4	5	6	7	8	9
Dose de $Ca(OH)_2$ (mg/l)	00	50	100	150	200	250	300	350	400
F^- (mg/l)	2.01	1.79	1.59	1.53	1.27	1.26	1.13	1.09	0.88
Ca^{2+}(mg/l)	529.05	465	440	420	350	360	360	330	235
Mg^{2+}(mg/l)	134.5	130	120	120	115	120	125	125	120
Fe^{2+}(mg/l)	0.1	0.11	0.1	0.08	0.1	0.11	0.12	0.11	0.11
Al^{3+}(mg/l)	0.13	0.09	0.098	0.119	0.11	0.09	0.1	0.099	0.094
TH (F°)	187.29	170.41	160	155	135.41	140	142.08	134 .58	108.7
PH	7.5	8.33	9.6	10.75	10.77	10.8	10.74	10.83	10.88
EC (ms/cm)	4.61	4.5	4.33	4.2	4.24	4.23	4.33	4.23	4.23

Table 3. Results of treatment by aluminum sulfate ($Al_2(SO_4)_3$)

Parameter /sample N°	1	2	3	4	5	6	7	8	9	10
Dose de $Al_2(SO_4)_3$ (mg/l)	00	10	20	30	40	50	100	150	200	300
F^- (mg/l)	2.01	1.88	1.73	1.66	1.45	1.55	1.42	1.19	0.98	0.84
Ca^{2+}(mg/l)	529.05	460	515	515	540	505	465	395	310	305
Mg^{2+}(mg/l)	134.5	110	115	115	115	125	125	110	125	120
Fe^{2+}(mg/l)	0.1	0.1	0.11	0.09	0.09	0.1	0.11	0.11	0.11	0.11
Al^{3+}(mg/l)	0.12	0.12	0.12	0.07	0.1	0.1	0.11	0.14	0.105	0.1
TH (F°)	187.41	160.33	176.66	176.66	175	178.33	168.33	144	129.5	126
PH	7.5	6.6	6.6	6.6	6.6	6.7	6.6	6.6	6.1	5.2
EC (ms/cm)	4.61	4.15	4.03	4.04	3.97	3.85	3.83	3.65	3.58	3.17

Table 4. Results of treatment by ferrous sulfate ($FeSO_4$)

Parameter / sample N°	1	2	3	4	5	6	7	8
Dose de $Fe(OH)_2$ (mg/l)	00	50	100	150	200	250	300	350
F^- (mg/l)	2.01	1.89	1.77	1.7	1.56	1.54	1.32	1.25
Ca^{2+}(mg/l)	529.05	510	460	405	365	345	320	280
Mg^{2+}(mg/l)	134.5	130	125	120	130	135	135	135
Fe^{2+}(mg/l)	0.1	16.4	21.7	70.25	99	102	120	135.5
Al^{3+}(mg/l)	0.12	0.079	0.096	0.1	0.087	0.091	0.086	0.076
TH (F°)	187.29	181.7	167.1	151.3	145.4	142.5	136.3	126.3
PH	7.5	5.9	5.8	5.7	5.6	5.5	5.4	5.3

Table 5.Results of treatment by ferric sulfate ($Fe_2(SO_4)_3$)

Parameter / sample N°	1	2	3	4	5	6	7	8	9	10
Dose of $Fe(OH)_3$ (mg/l)	00	10	20	30	40	50	100	150	200	300
F^- (mg/l)	2.01	1.83	1.79	1.75	1.76	1.76	1.74	1.7	1.64	1.45
Ca^{2+}(mg/l)	529.05	455	500	500	505	505	515	510	405	320
Mg^{2+}(mg/l)	134.5	125	125	120	120	125	130	125	110	125
Fe^{2+}(mg/l)	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.11	0.11	0.12
Al^{3+}(mg/l)	0.12	0.06	0.095	0.079	0.12	0.1	0.11	0.11	0.12	0.115
TH (F°)	187.29	166	177	175	176	178	183	180	147	132
PH	7.5	6	6.1	6.2	6.2	6.2	6.3	6.3	4	3.3
EC (ms/cm)	4.61	4.11	4.12	4.06	4.12	4.13	4.18	4.11	4.14	4.24

Fluoride removal by lime provided an outcome of 56.22 % with a decrease of fluoride amount equivalent to a rate between 2.01 mg/l to 0.88 mg/l after adding 400 mg/l of lime, it was noted that the tenor of fluoride ions decreased exponentially with the increase of the lime dosage, this result are explained by the precipitation of CaF_2 and $CaCO_3$ (Raymond, 1990). In addition to that the rate of outcome of fluoride removal by aluminum sulfate are 58.21%, with a decrease of fluoride of 2.01 mg/l à 0.84 mg/l after adding an amount of 350 mg /l. Finally, the outcome of ferrous sulfate and ferric which are respectively 37.81 and 27.86 %. this phenomenon could have been explained by the decrease of the initial tenor fluoride during the treatment by coagulation and flocculation (aluminum sulfate, ferrous and ferric sulfate) would be a phenomenon of absorption (fixation), according to Rabosky (1975), the frost of hydroxide formed ($Al(OH)_3$, $Fe(OH)_3$) could serve as fixation surface or absorption of iron ions. This occurred during the floculation, the fluoride ion are thus absorbed in the ionic layer (double layer) of the floculant than removed by the latter during decantation.

Conclusion

The population of the region of Ouargla is characterized with a diet that is very marked with tea, dates, lentil, carrots potato and pumpkin consumption, where fluoride ions dosage was very significantly. Moreover, the arid climatic conditions (T °C > 40 °C in summer) lead to a heavy water consumption and a strong sweating of the body, and thus a condensation of fluoride in the organism. In fact, it seems that standard of how drinkable the waters are vis-à-vis of the fluoride ions prescribed by the WHO are from being applied in these condition. These findings incite us to fluoride removal from water that which the tenor of fluoride ions exceed 0.85 mg/l according the WHO recommendation. These findings allowed us to conclude that the water table of Mio-pliocene is the most charged with fluoride as regards the standards of the WHO nonetheless the Albien one contains a dosage of fluoride lesser that that of the WHO standards. In order to decrease fluoride in water, the four methods have provided outcomes and remarkable values, especially in the case where the treatment was made by lime (56.22 %) and the aluminum sulfate (58.21%). As regards the food stuffs where the dosage of fluoride is quite high, they vary between 0.03 and 18.24 mg\kg. Finally, the establishment of a feeding equation (water + food) is advocated, in such a way to obtain an all in all fluoride dosage estimated of 4.4 mg/day /inhabitant.

References

[1] Aurélio Peres M, Simara Fernandes L, Glazer Peres K (2004) Inequality of water .uoridation in southern Brazil-the inverse equity hypothesis revisited. Social Sci Med 58:1181-1189
[2] Acharya S (2005) Dental caries, its surface susceptibility and dental fluorosis in South Int Dent J 55:359-364
[3] Badet C, Raghard B (2004) Etude Clinique de la carie. EMC-Dent 1 : 40-48
[4] Bock R (1979) Decomposition methods in analytical chemistry, translated by I.L. Marr, International Text Book Co,Glasgow,138p.

[5] Chavaissieux P, Meuinier PJ, (1995) bénéfique et risque des apports fluorés. Arch Pédiatre 2, 568-572.
[6] Gillespie GM, Baez R (2005) Development of salt fluoridation in the Americas. Schweiz Monatsschr Zahnmed 115: 663-669
[7] Long Y-G, Wang Y-N, Chen J, Jiang S-F, Nordberg A, Guan Z-Z (2002) Chronic fluoride toxicity decreases the number of nicotinic acetylcholine receptors in rat brain. Neurotoxicol Teratol 24: 751-757
[8] Messaitfa Amar (2007) Fluoride contents in ground waters and the main consumed foods (dates and tea) in southern Algeria region 1 :7
[9] M.G. Sujana , G. Soma, N. Vasumathi, S. Anand : Studies on fluoride adsorption capacities of amorphous Fe/Al mixed hydroxides aqueous solutions , 15 June 2009,pp753.
[10] Mille-Ihli NJ, Pehrsson PR,Cutrifelli RL, Holden JM (2003) Fluoride content of municipal water in the United states: what percentage is fluoridated .J food Compost Anal 16:621-628
[11] Onyango MS, Kojima Y, Aoyi O, Berrnardo EC, Matsuda H (2004) Adsorption equilibrium modelling and solution chemistry dependence of fluoride removal from water by trivalent-cation-exchanged zeolite F-9.Colloid Interface Sci 279: 341-350
[12] Pettifor JM,Schnizler CM,Ross FP, Mdoodley GP(1989) Endemic skeletal fluorosis in children: hypocalcaemia and the presence of renal resistance to parathyroid hormone. Bone Miner 7:275-288
[13] Pinet F, Pinet A, Barrierre J, Bouche B (1961) Endemic fluorosis of aqueous origin in Souf. Darmous and fluorosis osteopetroses. A Report on 51 cases of condensing osteoses.Alger Med 65:737-749
[14] Poey J, ElsairJ, Morgan P, Reggabi M, Hataab F (1976) Evaluation of biologic balance as a function of radiologic status in a population living in a endemic fluoride zone of southern a Algeria. Eur J Toxicol Environ Hyg 9:179-186
[15] Raymond Desjardins (1990) le traitement des eaux, 2émé édition, école polytechnique de Montréal 141 -180
[16] Singh PP, Barjatiya MK, Dhing S, Bhatnagar R, Kothari S, Dhar V (2001) Evdence suggesting that high intake of fluoride provokes nephrolithiasis in tribal populations. Urol Res 29:238-244
[17] Tohyama E (1996) Relationship between fluoride concentration in drinking water and mortality rate from uterine cancer in Okinawa prefecture, Japon. J Epidemiol 6:184-191
[18] Takahashi K, Akiniwa K, Narita K (2001) Regression analysis of cancer incidence rates and water fluoride in the U.S.A based on IACR/IARC(WHO) data (1978-1992).International agency for research on cancer. J Epidemiol 11:170-179
[19] Veressinina Y, Trapido M, Ahelik V, Munter R (2001) Fluoride in drinking water: the problem and its possible solutions. Proc Estonian Acad Sci Chem 50:81-88
[20] Wondwossen F, Asttrom AN, Bardsen A, Bjorvatn K (2003) Perception of dental fluorosis amongst Ethiopian children and their mothers. Acta Odontol Scand 61:81-86