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Effect of Addition of a Sudanese Herb (*Cymbopogon proximus*) on Drinking Water Fluoride, Nitrate and Total Dissolved Salts Concentration Levels

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ABSTRACT

The *Cymbopogon proximus* (CP) (*Cymbopogon schoenanthus*) is commonly used in Sudan to purify and give a favorable aromatic flavor to drinking water. In this study, we examined experimentally the chemical effect resulting from the addition of CP on drinking water F⁻, NO₃⁻ and TDS levels. Screening and confirmatory laboratory tests were conducted to investigate the changes of the concentration levels of F⁻, NO₃⁻ and TDS. For screening tests, we used two types of water, (1) drinking groundwater that naturally contaminated with nitrate, (2) Deionized water that artificially (experimentally) contaminated by nitrate. For confirmatory tests, we used only artificially nitrated deionized water. The results obtained indicated that the addition of 3.3 g CP L⁻¹ water entirely removed NO₃⁻ contamination, as a promising result and at the same time increased both F⁻ and TDS levels by different magnitudes depending on CP weight added and immersion time after the first 24 h passed. Increase of fluoride content in drinking water increases the risk of dental fluorosis prevalence and the other related diseases.

Key words: Ground water, nitrate, fluoride, total dissolved salts, Sudanese herb

INTRODUCTION

The natural water analyses for physical and chemical properties are very important for public health studies (Jafari *et al.*, 2008). Groundwater contamination can be resulted from lack of proper waste management (Nkolika and Onianwa, 2011). Domestic wastewater, surface water discharge and industrial waste have been reported among main factors that cause groundwater contamination (Baghvand *et al.*, 2006). Treatment of drinking water with herbal plants as traditional medicine is worldwide. In particular, the CP is widely used in Sudan in a purpose of folk medicine. The CP is a herbal plant springs from the family Poaceae (Gramineae) with a sub-species proximus (Gramineae). Common name is camel's hay and is known locally as "maharab". It is a perennial herb, erect, tufted 9 cm long, culms slender, glabrous and 3-4 noded. Leaf simple, alternate, linear 5-7 cm long, 1 cm wide, sheathed apex spiny entire, inflorescence spikelets highly branched 5 cm log (Eltahir and EReish, 2010). The plant is widely distributed in Africa (northwest tropical, northeast tropical and east tropical), temperate Asia (western Asia and Arabia) and tropical Asia (Indin and Indo-China) (Clayton *et al.*, 2005). In addition, CP is found in Northern and Central Sudan (Andrews, 1963).

The CP is highly reputed in folk medicine as an antispasmodic and urolithiasis (renal stone removal), for gout, diuretic agent (Boulos, 1983), flatulent, biotonic, analgesic for gastrointestinal track (GIT) disorders and menstrual pains as well; the plant is used externally as anti-rheumatic and cosmetic herb (Jomaha, 1988). The methanolic extraction of CP showed relaxant properties when investigated on isolated rabbit jejunum without showing toxic manifestations on the tissue, confirming the safety for the plant extract (Hamad-Alneel, 2006). The plant is used in prostate inflammation, kidney disease, inhibits kidney shrinkages, anthelmintic and for stomach pains (Eltahir and EReish, 2010). The CP exerted antihyperglycemic effects and consequently alleviates liver and renal damage caused by alloxan-induced diabetes (Mansour *et al.*, 2002).

The CP contains essential oils e.g., citronellal, careen-2, piperitone and sterol which are widely used as pesticides. The plant contains triterpenes and coumarins when fractionated by different solvents with the absence of saponins, tannins, alkaloids, anthraquinone glycosides and flavonoids. Studies on the toxicological effect of the CP showed no toxicity effects when used in traditional medicine (Hamad-Alneel, 2006).

Gas Chromatography (GC) and Mass Spectrophotometry (MS) analyses performed on the oil prepared from samples obtained from Burkina Faso and Sudan revealed significant differences in the composition of the herb. Piperitone was identified as the major component in samples obtained from Burkina Faso while those of Sudan were free from this component (Menut *et al.*, 2000).

Eight compounds were isolated from CP by preparative thin layer chromatography (TLC). The isolated compounds were found are sesquiterpenes. Five of them (compounds 1, 2, 3, 5 and 7) are derivatives of eudesmol. While, compound 4 is a ketonic monocyclic sesquiterpene and compound 8 is an alcoholic monocyclic sesquiterpene, compound 6 was the only straight chain aliphatic unsaturated aldehyde (Al-Assiuty *et al.*, 2006). The eudesm compounds are: 5, 11 dihydroxy-1, 3-diene, eudesm 4 hydroxymethyl-11-hydroxy 1, 4 diene, eudesm 7, 11 dihydroxy 3 ene, atlantone, eudesm 4 methylene-5-methoxy-11-ol, 2 methyl undec-2-ene-10-al, eudesm-11-ol and elemol. The following compounds also were isolated from the unsaponifiable fraction of the petroleum ether extract of CP: proximadiol, 5 α - hydroperoxy- β -eudesmol, 1 β -hydroxy- β -eudesmol, 1 β -hydroxy- α -eudesmol, 5 α -hydroperoxy- β -eudesmol and 7 α , 11dihydroxy cadin-10(14)-ene (El-Askary *et al.*, 2003).

As being observed, the treatment of drinking water with CP is widely spread in the Sudan and it is commonly used in folk or traditional medicine or for aesthetic reasons in order to add a favorable aromatic flavor. Therefore, this study, intends to examine experimentally the effect of CP on groundwater F⁻, NO₃⁻ and TDS levels.

MATERIALS AND METHODS

Collection of samples and materials: Adequate amounts of CP were purchased from Rufaa' market, east of Gezira locality of Sudan. Large plastic container (4 liter volume) were used for collecting groundwater naturally containing 1.1 mg F⁻L⁻¹, 4.5 mg NO₃⁻ L⁻¹ and 360 mg TDS L⁻¹ from Umm-Dawanban, Sharkelneel Locality to be used for the screening test. Deionized water was used in both the screening and the confirmatory tests.

Chemical analyses: The determinations were carried out according to the standard methods for the examination of water and wastewater (APHA, 1998) and Rhoades (1982). The electric conductivity (EC) (dS m⁻¹ at 25°C) in groundwater samples was measured by Beckman Solu Bridge type equipment in 100 mL of the water sample. To convert the EC readings to TDS, the results

for the EC dS m^{-1} were multiplied by 640 according to Rhoades (1982). Fluoride was determined by Alizarin Visual method according to APHA (American Public Health Association): 1998. Nitrate is reduced almost quantitatively to nitrite (NO_2^-) in the presence of cadmium. The NO_2^- thus obtained is determined colorimetrically using instrument model SPECORD 40-analytik Jena's spectrophotometer (Germany).

Screening tests

Natural well drinking water screening test: Screening tests were conducted to investigate the effect of CP on F^- , NO_3^- and TDS concentration levels in a well drinking water sample. The F^- , NO_3^- and TDS levels in the groundwater sample were analyzed before and after the addition of CP. A random weight of CP (5 g) was added to 4 liters of well water test sample containing naturally $1.1 \text{ mg F}^- \text{ L}^{-1}$, $4.5 \text{ mg NO}_3^- \text{ L}^{-1}$ and $360 \text{ mg TDS L}^{-1}$ and allowed to stand for about 3 days (72 h) at room temperature (30-32°C). The treated sample was filtered and the filtrate was kept in a plastic bottle in the refrigerator (4°C) pending analysis.

Artificially nitrated deionized water screening test: A laboratory-screening test was also conducted to investigate the effect of CP on F^- , NO_3^- and TDS levels in an artificially nitrated deionized water samples. A total of 0.36099 gram of potassium nitrate (KNO_3) was accurately weighed, transferred into a one-liter volume long-neck flask, thoroughly dissolved and completed to the mark with deionized water and a solution containing $221 \text{ mg NO}_3^- \text{ L}^{-1}$ was thus obtained. This solution was divided into four equal portions, 250 mL each and then each portion was transferred into 600 mL volume plastic bottle, completed to volume with deionized water to achieve a solution containing $92.25 \text{ mg NO}_3^- \text{ L}^{-1}$. This solution was devoid of any F^- and its TDS was found to be 600 mg L^{-1} which was substantially induced by the addition of KNO_3 to the deionized water. Four different weights of the CP (2.5, 5.0, 10, 20 g) were carefully and gently washed with deionized water, in order to remove any adhering soil residues and were then immersed into the 600 mL plastic bottles containing the $92.25 \text{ mg NO}_3^- \text{ L}^{-1}$ and allowed to stand for about 3 days (72 h). The treated sample of each bottle was transferred into 600 mL plastic bottle and kept in the refrigerator at 4°C pending analysis.

Confirmatory laboratory test: A 0.902475 g weight of KNO_3 was accurately and quantitatively transferred into one-liter volume long neck flask and completed to the mark with deionized water. Thus, a solution containing $553.57 \text{ mg NO}_3^- \text{ L}^{-1}$ was obtained. About 150 mL of this solution were transferred into each of six 1.5-liter plastic bottles and the volume was completed to 1.5 liter with deionized water to obtain a final concentration of $55.36 \text{ mg NO}_3^- \text{ L}^{-1}$ and was then allowed to stand under laboratory conditions for 24 h. Five different weights of CP (5, 10, 15, 20 and 25 g) were then weighed and immersed (after being washed with deionized water) into each of five 1.5-liter volume bottles and the sixth bottle was left untreated i.e., containing $55.36 \text{ mg NO}_3^- \text{ L}^{-1}$ solution for control. The six bottles (five treated samples and one control) were allowed to stand under laboratory conditions (temperature 30-32°C). After one day each of the 6 bottles was sampled. Sampling was repeated after two and three days and all samples were kept in the refrigerator at 4°C pending analysis.

RESULTS AND DISCUSSION

Screening tests: Table 1 summarizes the results obtained for the screening tests of the addition of CP on both natural groundwater (NGW) and artificially nitrated deionized water (ANDW),

Table 1: Effect of addition of CP on the levels of NO_3^- , F^- and TDS in NGW and ANDW (i.e., screening test)

Water constituent studied	NGW		ANDW		2.5		5		10		20	
	Level in control sample (SA) (mg L^{-1})	Level after addition of 5 g CP (mg L^{-1})	Total decrease/increase	Level in control sample at 0 time (mg L^{-1})	Level after 24 h	Total decrease/increase	Level after 24 h	Total decrease/increase	Level after 24 h	Total decrease/increase	Level after 24 h	Total decrease/increase
NO_3^-	4.5	0.4	4.1 (1025%)*	92.25	91*	1.25 (1.4%)*	66*	26.3 (29%)*	54.5*	37.75 (41%)*	1.7*	90.55 (98%)*
F^-	1.1	2.5	1.4 (127%)*	0.0	1.5#	1.5 (NA)#	2.7#	2.7 (NA)#	3.5#	3.5 (NA)#	4.9#	4.9 (NA)#
TDS	360	850	490 (136%)*	600	810#	210 (35%)*	985#	385 (64%)*	1150#	550 (92%)*	1560#	960 (160%)*

NGW: Natural groundwater (Naturally nitrated, fluoridated and salted), ANDW: Artificially nitrated deionized water (free from F^- and the 600 mg L^{-1} TDS level resulted from the addition of KNO_3). SA: Study analysis, CP: *Cymbopogon proximus* (*Cymbopogon schoenanthus*), *: Percentage decrease in NO_3^- levels, #: Percentage increase in both F^- and TDS levels, NA: Not available

devoid of fluoride. After the addition of 5 g of CP, nitrate level in NGW was decreased from 4.5 to 0.4 mg L⁻¹ (1025%). On the contrary, both F⁻ and TDS were increased from 1.1 to 2.5 mg L⁻¹ (127%) and from 360 to 850 mg L⁻¹ (136%), respectively. In ANDW, NO₃⁻ level was decreased from 92.25 to 91 mg L⁻¹ (1.4%), 66 mg L⁻¹ (28%), 54.5 mg L⁻¹ (17%) and 1.7 mg L⁻¹ (97%) with an increasing weight of CP of 2.5, 5 g, 10 and 20 g, respectively. Conversely, a gradual increase was observed in the concentration levels of both F⁻ and TDS with an increasing weight of CP. According to Table 1, F⁻ level was increased from 0.0 to 1.5 mg L⁻¹, 2.7, 3.5 and 4.9 mg L⁻¹ while TDS level was increased from 600 to 810 mg L⁻¹ (35%), 985 mg L⁻¹ (64%), 1150 mg L⁻¹ (92%) and 1560 mg L⁻¹ (160%) with an increasing weight of CP of 2.5, 5, 10 and 20 g, respectively.

Confirmatory test

Control sample: Table 2 shows the levels of F⁻, NO₃⁻ and TDS in ANDW control sample at zero time and after 24 h. No changes were observed in both F⁻ and TDS levels while NO₃⁻ level was apparently affected. It has been confirmed that NO₃⁻ level in groundwater usually decreases as time progresses due to auto-dissolution of NO₃⁻ (if the source of NO₃⁻ is eliminated) (Alvarez and Illman, 2006); this is demonstrated and confirmed in this study by observing the trend of control sample in Fig. 1 which showed the reduction of NO₃⁻ with time. The initial calculated NO₃⁻ level in the control sample of the artificially nitrated deionized water is 55.36 mg NO₃⁻ L⁻¹. After 24, 48 and 72 h of incubation in the laboratory NO₃⁻ levels were reduced to 38.4 (31% reduction), 36.6 (34% reduction) and 31.5 mg NO₃⁻ L⁻¹ (43% reduction), respectively. The degradation of NO₃⁻ contaminant with time in control sample may be attributed to the redox reactions involving electron transfer as follows:

Table 2: Fluoride, nitrate and TDS changes in control samples in confirmatory test of artificially nitrated deionized water (ANDW)

Time (h)	Constituent/property		
	NO ₃ ⁻ level (mg L ⁻¹)	F ⁻ level (mg L ⁻¹)	TDS level (mg L ⁻¹)
0	55.36	0	410
24	38.4 (31%)	0 (0%)	410 (0%)
48	36.6 (34%)	0 (0%)	410 (0%)
72	31.5 (43%)	0 (0%)	410 (0%)

ANDW is free from F⁻, The 55.36 mg L⁻¹ NO₃⁻ and the 410 mg L⁻¹ TDS levels resulted from the addition of KNO₃

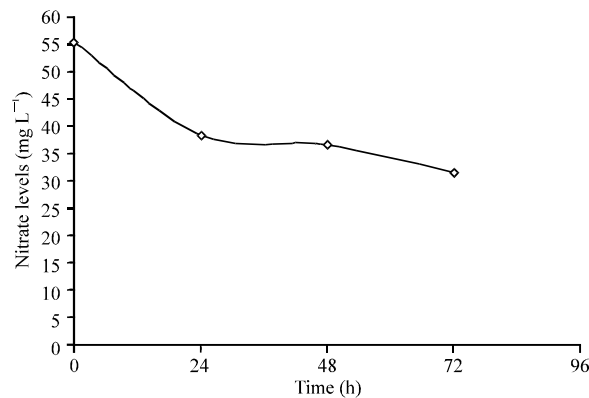
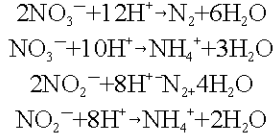


Fig. 1: Nitrate auto-dissolution in control sample



The contaminant is reduced if it gains electrons and the electron donor becomes oxidized. Redox reactions are often mediated by microorganisms (Alvarez and Illman, 2006).

Effect of CP on water NO_3^- level: A high level of NO_3^- is seen as a threat to both public health and natural waters, of these threats the latter is the more immediate but the health issue has attracted more public concern, as the presence of nitrate in drinking water has been linked to a number of medical conditions such as blue baby syndrome (methaemoglobinaemia) in infant (Curley *et al.*, 2009). Nitrate is effective plant nutrient and moderately toxic (Naeem *et al.*, 2007). Nitrate can be removed from drinking water by carbon. The denitrification depends on the carbon source and reactor type (Magram, 2010).

It may be inferred from Fig. 2 that the addition of 5 g CP to the artificially nitrated deionized water reduced NO_3^- level from 55.36 mg $\text{NO}_3^- \text{L}^{-1}$ to 31.1 mg $\text{NO}_3^- \text{L}^{-1}$ (44% reduction) after the first period (24 h). The NO_3^- level was reduced to zero (i.e., the solution was free from NO_3^-) after each of the second and the third periods (48 and 72 h), respectively, i.e., reduction percentage of 100%. The addition of 10 g CP to the nitrated solution reduced NO_3^- level from 55.36 mg $\text{NO}_3^- \text{L}^{-1}$ to 21.8 mg $\text{NO}_3^- \text{L}^{-1}$ (60% reduction) after the first period, reduction was 100% complete during the second and third periods (48 and 72 h) of incubation. The addition of 15 g CP to nitrated solution decreased the NO_3^- concentration level from 55.36 to 16.7 mg L^{-1} (69.83% reduction) after the first period (24 h) while during the second and the third period (48 and 72 h) the nitrated solution was devoid of any NO_3^- i.e. (100% reduction). The addition of higher weights i.e., 20 and 25 g of CP reduced NO_3^- concentration level from 55.36 mg L^{-1} to zero level after the first period (24 h) of incubation i.e., 100% reduction. Noteworthy that, this reduction includes both nitrate auto-dissolution (in control) and the effect of addition of CP). Commercially, nitrate can be removed from drinking water by either standard anion resins or nitrate-selective resins. The functional

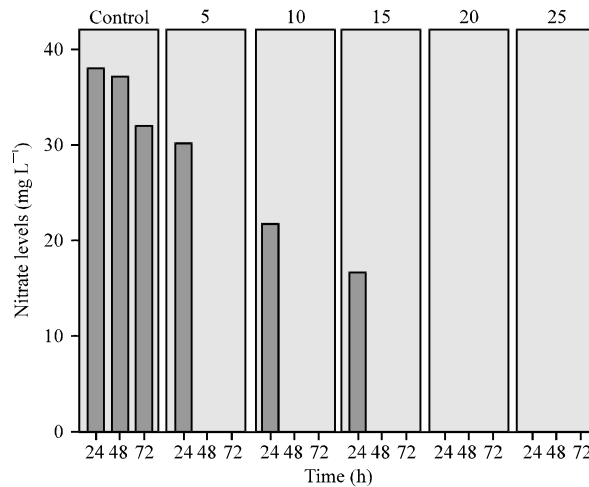


Fig. 2: Effect of addition of CP on NO_3^- level of deionized water

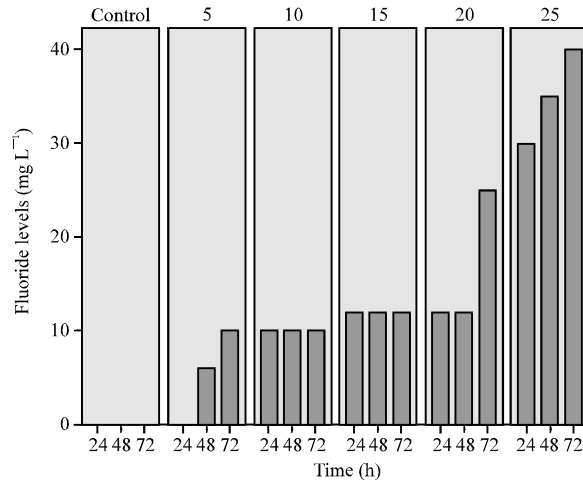


Fig. 3: Effect of addition of CP on fluoride level of deionized water

groups of the synthesized standard anion resins commonly used for nitrate removal are trimethylamine (NC_3H_9) and dimethyl ethanolamine ($\text{NC}_4\text{H}_{10}\text{OH}$). In nitrate-selective resins, the functional groups are tributylamine ($\text{NC}_{16}\text{H}_{33}$) and triethylamine (NC_6H_{15}). The natural functional group in the chemical compounds of CP could be the $-\text{C}_3\text{H}_6\text{OH}$ group that characterizes eudesmol which is the main component of CP herb (Al-Assiuty *et al.*, 2006; El-Askary *et al.*, 2003) and the nitrogen component may be offered by plant tissues. Thus, the CP herb may acts as a natural anion exchange and/or nitrate-selective resin which, may be obtained after immersing CP into water. In addition, redox reactions that mediated by microorganisms (Alvarez and Illman, 2006; Chitra and Lakshmanaperumalsamy, 2006) induced by addition of CP to the water sample may be also contribute to NO_3^- reduction.

Effect of CP on water F^- level: Figure 3 shows the ascending level trends of F^- levels with the increasing CP weights. Obvious increase of F^- level can be noticed with the progress of immersion time. Addition of 5 g CP to 1.5 liter F^- free deionized water conferred $0.6 \text{ mg F}^- \text{ L}^{-1}$ after duration of 24 h and this level was sustained during 48 h. Fluoride level increased from 0.6 to 1.0 mg L^{-1} during the 72 h incubation period and remained constant at this level even after the addition of 10 g CP. At the CP rate of 15 g F^- level was increased to 1.2 mg L^{-1} and remained so at the 20 g CP rate during the incubation periods of 24 and 48 h, however, at the duration of 72 h, the F^- level increased to about 2.5. At the rate of 25 g CP high levels of fluoride were observed amounting to 3.0, 3.5 and $4.0 \text{ mg F}^-/\text{l}$ at the incubation periods of 24, 48 and 72 h, respectively.

It has previously been reported that plants can uptake and accumulate F^- into their tissues (Elhadi, 2004). Therefore, CP plants (herbs) that grow in soils with high F^- content or irrigated with water of high F^- content may accumulate F^- into their tissues. This infers that when CP was immersed into or added to water, may add extra amounts of F^- to it, thus elevating its total F^- content.

Effect of CP on water TDS level: The palatability of water with a TDS level of less than 600 mg L^{-1} is generally considered to be good (Matloob, 2011). It has been reported that continuous pumping of groundwater increases TDS level (Al-Salamah and Nassar, 2009). The TDS level in

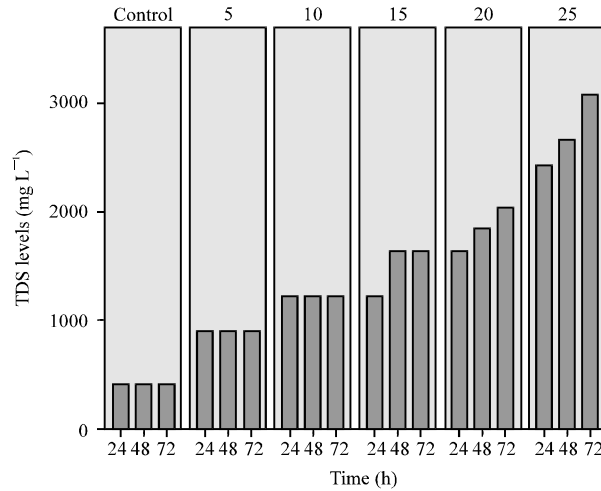


Fig. 4: Effect of addition of CP on TDS level of deionized water

control sample (deionized water) is 410 mg L^{-1} which resulted from addition of potassium nitrate. According to Fig. 4, the TDS level was positively affected by both immersion time and weight of CP. In the control, as in F^{-} , the TDS levels remained constant at 410 mg L^{-1} with the progress of time. The TDS level increased to 899 mg L^{-1} after adding 5 g CP and remained constant at 899 mg L^{-1} during all of the incubation duration of 24, 48 and 72 h. At the rate of 10 g CP the TDS level increased to 1229 mg L^{-1} and remained constant during all of the incubation period of 24, 48 and 72 h. At the rate of 15 g CP, the TDS level remained at 1229 mg L^{-1} after 24 h, then the level jumped to 1638 mg L^{-1} after 48 and 72 h of incubation and remained at this level even after 24 h incubation at the rate of 20 g CP. Afterwards, at the rate of 20 g CP, the TDS level was increased to 1843 and 2038 mg L^{-1} after 48 and 72 h, respectively. The TDS increased progressively to 2417, 2662 and 3072 mg L^{-1} after 24, 48 and 72 h of incubation at 25 g of CP, respectively. The increase in the magnitude of the TDS in water, due to the addition of CP, may be explained as follows: During immersion time of the CP herb in water, chemical constituents in the CP herb may have been released into the solution thus elevating the TDS level (Elhadi, 2004).

From the previous results pertaining to the effect of CP addition on the level of F^{-} , NO_3^{-} and TDS to deionized water, it can be concluded that the addition of CP to artificially nitrated deionized water entirely eliminates NO_3^{-} contamination and at the same time increases both F^{-} and TDS levels. Addition of 5 g of CP to 1.5 liter of water (3.3 g L^{-1}) entirely reduces NO_3^{-} contamination and at the same time increases both F^{-} and TDS levels by different magnitudes depending on CP weight used and immersion time. It is advisable to add CP to drinking water of high level of NO_3^{-} . Addition of CP to drinking water of high level of F^{-} should be avoided. On the other hand, the CP may be added, cautiously, to drinking water deficient in F^{-} in order to increase its F^{-} content to the optimum level that removes the concern related to dental caries and at the same time to impart pleasant flavor to drinking water that preferable in the study area.

CONCLUSIONS AND RECOMMENDATIONS

This study revealed that the addition of CP "maharaib" at the rate of 5 g CP/1.5 liter water (3.3 g L^{-1}) to groundwater removes NO_3^{-} in 48 h and at the same time increases TDS and F^{-} levels and consequently increases the risk of dental fluorosis incidence and other fluoride adverse effects.

The recommendation are as follows:

- Addition of "maharaib herb", as practiced in folk medicine, to groundwater of high levels of fluoride should be avoided because as has been demonstrated in this study, the herb increases water fluoride content. On the other hand, the "maharaib herb" may be added, cautiously, to drinking groundwater deficient in fluoride ion in order to increase its fluoride content to the optimum level that removes the risk of dental caries
- It is recommended to add "maharaib herb" to drinking groundwater of high levels of nitrate in order to reduce its nitrate level in communities where nitrate ion contamination is excessively beyond the maximum permissible level set by the SSMO (2002)

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