Efficacy of *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder to reduce nitrate: N and total hardness in groundwater in karstified limestone aquifer

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Abstract

Groundwater is a vital resource in the northern region of Sri Lanka, as it is the only water resource used for domestic, agricultural and industrial activities in that area. However, due to excessive usage of synthetic fertilizer, the groundwater in this part of the country is highly contaminated with nitrates. In addition, due to the effect of underlain limestone aquifer, water hardness is also high. The present study aimed to study the effect of filtration through *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder on reducing nitrate concentration and hardness. The results indicated that 21.4% reduction of nitrate concentration can be achieved by filtration through *Terminalia arjuna* mature leaf powder and 9.3% reduction of total hardness by filtration through *Phyllanthus emblica* bark powder. Therefore, further research is recommended on testing the potential and side effects of using *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder as a combined home remedial treatment technique for treating nitrate contaminated hard water.

Keywords Home remedial techniques · Groundwater · Drinking water pollution · Water purification

Introduction

The Jaffna peninsula, Sri Lanka, is underlain by 100–150-m-thick Miocene limestone formation which is uniquely bedded and well established and highly karstified. The unique geological features of this part of the country include lagoonal and estuarine deposits, unconsolidated brownish grey coastal sands, highly karstified Miocene sedimentary limestone formations, red beds and dune sands (Kumara et al. 2013). The whole population in Jaffna Peninsula is primarily dependent on groundwater resources to meet the domestic, agricultural and industrial water requirements (Wijeyaratne and Suvendran 2017).

The groundwater aquifers in Jaffna peninsula are shallow karstic-type aquifers and are recharged only by infiltration

of rainfall. This shallow groundwater aquifers form mounds or complexes of freshwater lenses of up to 25 m thickness that floats over the saline water and reach their peak during the monsoon rains in November and December (Panabokke and Perera 2005; Vijakanth et al. 2017). Jaffna peninsula receives an average approximate annual rainfall of 1200 mm per 1000 km² area. However, after all the natural losses of precipitation, approximately 5–50% of rainwater is lost to sea as subsurface flow thereby leaving approximately 450 million m³ water annually to fulfil the requirements of the residing population in the peninsula. According to the Falkenmark indicator, this amount of annual water availability indicates that this region is facing water scarcity (Vijakanth et al. 2017). For more than five decades, the groundwater resources in Jaffna peninsula were sustainably extracted due to less population resided in the area during that period. However, the recent new developmental activities in the peninsula, the resettlement of population and rapid industrial and agricultural expansion have resulted in extensive use of groundwater resources leading to unsustainable groundwater extraction.

Increased concentration of nitrates and nitrites in some aquifer systems in Jaffna peninsula is identified as a significant issue over the decades. The high nitrate and nitrite

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concentration in drinking water can result in serious health issues in the population specifically affecting the newborns, pregnant mothers and elderly people (Sivarajah 2003; WHO 2011; Sutharsiny et al. 2014). Elevated levels of nitrates and nitrites in the soil and groundwater of the peninsula are primarily due to application of large quantities of nitrogenous fertilizers such as urea in an improper manner. Further, this issue is intensified due to poor planning and construction of soakage pits and latrines. According to previous studies, water samples in the intense agricultural areas in Jaffna Peninsula have recorded nitrate–nitrogen levels ranging from 20 to 50 ppm which is 2–5 times higher than the standard level (Mageswaran and Mahalingam 1983; Navaratnarajah 1994). Also, a recent study conducted in Chunnakam and Vadamaradchi areas of Jaffna peninsula indicated that the water is unsuitable for drinking purposes in terms of nitrate concentrations (Emmanuel et al. 2013; Wijeyaratne and Suvendran 2017; Mahagamage et al. 2019).

Increased hardness of water is also a major water quality problem in this area, and researches have recorded that this is primarily due to underlain Miocene limestone formation (Navaratnarajah 1994). Increased hardness of drinking water can also result in severe health effects including higher risk for urinary and salivary stone formation, urolithiasis and chronic kidney disease (CKDu) (Kozisek 2005; Kodikara et al. 2015; Wasana et al. 2016). In addition to health effects, increased hardness in water makes it unsuitable to be used for irrigational, industrial and domestic purposes (Emmanuel et al. 2013; Padmapriya et al. 2015; Wasana et al. 2016; Mahagamage et al. 2019).

Remediation of drinking water in terms of treatment for excess nitrate and hardness involves various techniques including chemical and biological methods. However, the advanced technologies such as electrochemical processes, enzyme catalysation, nanofiltration, electrodialysis, ultrasound methodologies, ultra-filtration methodologies and ion-exchange membranes are costly and not environmentally friendly (Saeed and Hamzah 2013; Kannan and Mani 2014; Megersa et al. 2014).

Therefore, providing safe drinking water in the developing countries is a challenging task due to economic and environmental issues (Megersa et al. 2014). Household water treatment and safe storage (HWTS) inventions have become a key solution and lead to enormous improvements in drinking water quality and reductions in waterborne diseases. This could make a difference to the lives of those who rely on water from polluted rivers, lakes and unsafe wells or piped water supplies (Buhar 2017). In some parts of the world, traditional home water treatment techniques such as utilization of roots, barks and leaves of various native plant species to remove impurities of water are practised. These methods are cost-effective and environmental friendly and have minimal side effects.

A study by Megersa et al (2014) identified about 40 plant species that can be used as coagulants and disinfectants in water treatment (Megersa et al. 2014). *Moringa oleifera* seeds were recorded to be an effective purifying agent that can remove 90% copper, 80% lead, 60% cadmium and 50% zinc and chromium in the polluted water (Nand et al. 2012). Also, bark powder of *Acacia catechu* (Thakur and Choubey 2014), *Moringa oleifera* seeds (Sotheeswaran et al. 2011) and seed coats of *Elettaria cardamom* (Khurana and Sen 2008) are recorded to have effective coagulant properties in treating high turbid water.

Phyllanthus emblica is widely used in ayurvedic, unani and siddha medicine in Sri Lanka (Sathish et al. 2012; Dasaroju and Gottumukkala 2014) and *Terminalia arjuna* is a medicinal plant native to India and Sri Lanka and often planted near wells, rivers and other water bodies as its root is believed to purify and cool the water (Schmelzer and Ameenah 2008). These two plants have medicinal properties, and immersing wood, roots and leaves of these two species in water pots are practised as a traditional water purification method in Sri Lanka. Studies have proved that *Phyllanthus emblica* wood can reduce the TDS, electrical conductivity, calcium (71.7%), magnesium (69.6%), total alkalinity (89.7%) and microbial loads in the polluted water (Kannan and Mani 2014; Padmapriya et al. 2015). Also, the roots and seed powder of *Terminalia arjuna* are recorded to have effective hardness and turbidity treatment properties (Illeperuma et al. 2012; Ravindara et al. 2014).

However, use of plant matter in drinking water treatment is not very popular among the population in the northern region of Sri Lanka. *Phyllanthus emblica* and *Terminalia arjuna* are two commonly grown plant species in the Jaffna peninsula. However, the effectiveness of parts of these two plant species in addressing the common water quality problems prevailing in this area such as increased hardness and nitrate concentration is not addressed. Therefore, the present study was carried out with the intention of testing the effectiveness of *Phyllanthus emblica* bark powder and *Terminalia arjuna* mature leaf powder in reducing the total hardness and nitrate concentration of drinking water collected from the domestic wells in the Kondavil region Jaffna peninsula, Sri Lanka.

Materials and methods

Study area and collection of samples

Jaffna municipal area is supplied with water pumped from wells located in Kondavil and Thirunelvely. This study focussed on selected domestic wells in the Kondavil area which is fed by karstic aquifer. Kondavil belongs to Nallur

Divisional Secretariat and is situated 5 kms from the Jaffna town (Panabokke and Perera 2005).

Fifteen domestic wells were randomly selected from the study area, and five replicate water samples were collected from each well. The location of the sampled wells is given in a map in Fig. 1. Among the sampled wells, W3 ($9^{\circ}42'8.70''\text{N}$, $80^{\circ}2'18.72''\text{E}$) was a service well that supplies water to the Jaffna municipal area.

The temperature, salinity, pH, electrical conductivity, dissolved oxygen concentration, total dissolved solids (TDS) of each water sample were measured in situ using pre-calibrated multi-parameter water quality checker (HACH model: H940). The water samples were preserved in accordance

with APHA 1992, and were transported to the laboratory of the Department of Zoology and Environmental Management, University of Kelaniya, Sri Lanka. In the laboratory, chemical oxygen demand (COD), nitrate concentration, total phosphorus concentration and total hardness were measured following the methodologies described in APHA 1992. Faecal coliforms were determined using the membrane filtration technique (WHO 2011).

Water treatment assays using plant powder

The sampled water from the domestic wells (Fig. 1) was subjected to filtering through *Terminalia arjuna* mature

Fig. 1 Map of the study area showing the sampled wells

leaf powder and *Phyllanthus emblica* bark powder using a column filter and boiling at 100 °C for 10 min, and the total hardness and nitrate–N concentration were measured in the filtered water samples, following the methodologies described in APHA 1992.

The *Terminalia arjuna* mature leaf powder was prepared as described by Saduzaman et al, 2013. The *Terminalia arjuna* mature leaves were obtained from the *Terminalia arjuna* tree and washed well with distilled water and were shade-dried. Then, these leaves were ground using grinder (Bright elegant-240V6A), and then, powder was sieved through a 600 µm sieve and stored in a desiccator at a cool dry place (Saduzaman et al. 2013).

Phyllanthus emblica bark powder was prepared as described by Padmapriya et al 2015. The coat of the bark of *Phyllanthus emblica* was removed and washed well with distilled water and was shade-dried. Shade-dried bark parts were ground using grinder (Bright elegant- 240V6A), and the powder was sieved through a 600 µm sieve and stored in a desiccator at a cool dry place (Padmapriya et al 2015).

A column experiment was set up using separate vertical columns containing *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder. Each column contained 5 g of respective plant powder, and 5L of water was filtered through the column maintaining 0.25L/min flow rate. The filtered water samples were again filtered separately through Whatman No. 1 filter paper. Each test was replicated five times using fresh plant powder at each replication.

The TDS and nitrate concentration of the filtered water sampled were measured following the methodologies described in APHA 1992.

Statistical analysis

After confirming the normality using Anderson–Darling test, the data were analysed using ANOVA followed by Tukey's pairwise comparison test to study the spatial variation of water quality parameters. Paired t test was used to analyse the significance of differences in water quality parameters before and after the treatments. MINITAB 14 statistical software package was used in the statistical analysis.

Comparison with drinking water quality standards and classification of the groundwater

The measured physico-chemical parameters in domestic wells were compared with the drinking water quality standards established by Sri Lanka Standards Institution (SLSI 1983, SLSI 2013).

Results

Water quality parameters

The mean $\pm$ standard deviation (SD) and the minimum and maximum values of water quality parameters sampled from the domestic wells in Kondavil and the drinking water standards established by the Sri Lanka Standards Institution (SLSI) and the percentage of sampled wells exceeding each SLSI drinking water standard are given in Table 1.

The pH, total phosphorous concentration (TP) and chemical oxygen demand (COD) of the sampled wells were within the drinking water standards established by Sri Lanka

Table 1 Mean $\pm$ SD of water quality parameters of the domestic wells in Kondavil region during the study period, drinking water standards established by the Sri Lanka Standards Institution (SLSI 1983, SLSI

2013) and the percentage of sampled wells exceeding SLSI drinking water standards

Parameter	Study area	SLSI drinking water standards	Percentage of wells exceeding the drinking water standards (%)
Temperature (°C)	29.87 $\pm$ 0.18 (28.86–31.31)	Not mentioned	–
pH	7.00 $\pm$ 0.05 (6.65–7.25)	6.3–8.5	0
Total dissolved solids (mg/L)	601.0 $\pm$ 44.7 (315.8–901.1)	500	66.7
Electrical conductivity (µs/cm)	1201.0 $\pm$ 81.1 (645.6–1742.9)	Not mentioned	–
Salinity (‰)	0.61 $\pm$ 0.05 (0.31–0.91)	Not mentioned	–
Dissolved oxygen concentration (mg/L)	4.89 $\pm$ 0.24 (2.88–6.09)	Not mentioned	–
Nitrate–N (mg/L)	12.07 $\pm$ 1.59 (0.82–20.99)	11.2	60
Total phosphorous (mg/L)	0.127 $\pm$ 0.03 (0.067–0.447)	2.0	0
Total hardness (mg/L, CaCO ₃)	644.15 $\pm$ 29.5 (454.8–784.4)	250	100
COD (mg/L)	5.33 $\pm$ 0.25 (3.17–6.73)	10	0
Faecal coliform	Not detected		
Oil and grease	Not detected		

The range of water quality parameters of the domestic wells is given within parentheses

Standards Institution (SLSI 1983, SLSI 2013). However, all the sampled wells exceeded the total hardness concentration and 66.7% of the sampled wells exceeded the total dissolved solids (TDS) concentrations and 60% of the wells exceeded total nitrate concentrations established by Sri Lanka Standards Institution (SLSI 1983, SLSI 2013) for safe drinking water (Table 1).

The spatial variation of mean total hardness, TDS and nitrate N concentration of the sampled wells are given in Fig. 2. A total of nine wells representing 60% of the sampled wells exceeded standards for the nitrate N concentration established by SLSI for safe drinking water (Table 1, Fig. 2). The mean nitrate N concentration of well 3 (W3),

which serves as a service well to supply water to water-scarce areas, was 12.16 mg/L and was also higher than the SLSI standards (Fig. 2a). The nitrate concentration of the wells W1, W2, W4, W5, W7 and W10 was below the SLSI standards for safe drinking water (Fig. 2a).

The total hardness concentration of wells 2, 4, 5 and 7 was significantly lower than that of the other sampled wells. However, all the wells exceeded total hardness concentration established by SLSI (SLSI 1983, SLSI 2013) for safe drinking water (Table 1, Fig. 2b).

The interpolated spatial variation pattern of total hardness in the study area by inverse distance weighted method using ArcGIS 10.2.2 software is given in Fig. 3. It shows a spatial

Fig. 2 The spatial variation of mean total hardness, TDS and nitrate N concentration of the sampled wells (**a**: nitrate N concentration; **b**: total hardness; **c**: total dissolved solids)

Fig. 3 The spatial interpolation of total hardness concentration in the study area

pattern of 3 zones of hardness (Fig. 3). The northern and western parts of Kondavil region can be characterized by high water hardness compared to other regions of the study area (Fig. 3). Increased hardness in water indicates the richness of calcium and magnesium. However, in this region of the country, the increased groundwater hardness is mainly due to high calcium concentrations leached by the underlain limestone aquifer (Mikunthan and Silva 2008).

The mean total dissolved solids concentration (TDS) of the well 2 was significantly lower, and it was significantly higher in well 9 compared to other sampled wells (Fig. 2). Many physical and chemical properties of water are dependent on the TDS content of water. The TDS of the sampled wells varied in a large range, and 66.7% of the wells had

higher TDS content than the standard limit for safe drinking water.

Water treatment assays

The mean nitrate, TH and TDS of the water samples before and after filtration through *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder and boiling at 100 °C are given in Table 2.

Filtration through *Terminalia arjuna* mature leaf powder or *Phyllanthus emblica* bark powder did not change the total dissolved solids of the water samples significantly. Filtration through *Terminalia arjuna* mature leaf powder significantly reduced the nitrate concentration of the water samples. The

Table 2 The mean nitrate, TH and TDS of the water samples before and after filtration through the *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder and boiling at 100 °C for 10 min

Parameter	Raw water	<i>Terminalia arjuna</i> mature leaf powder	<i>Phyllanthus emblica</i> bark powder	Boiling at 100 °C for 10 min
TDS (mg/L)	601.0 ± 4.7 ^a	606.9 ± 4.7 ^a	601.2 ± 4.8 ^a	608.52 ± 2.6 ^a
Nitrate-N (mg/L)	12.07 ± 1.59 ^a	9.49 ± 1.5 ^b	12.06 ± 1.54 ^a	16.63 ± 1.8 ^c
TH (mg/L, CaCO ₃)	644.15 ± 9.5 ^a	635.8 ± 7.6 ^a	584.0 ± 5.8 ^b	580.25 ± 2.8 ^b

The mean values indicated by different superscript letters at each row are significantly different from each other (paired *t* test, *p* < 0.05)

percentage reduction of nitrate concentration after filtration through *Terminalia arjuna* mature leaf powder was 21.4%, and the mean nitrate N concentration of the filtered water was less than the SLSI standard for safe drinking water (Table 2). However, filtration through *Phyllanthus emblica* bark powder did not have a significant effect on nitrate N concentration (Table 2).

The total hardness concentration was significantly reduced by filtration through *Phyllanthus emblica* bark powder, and the percentage reduction was 9.3%. The *Terminalia arjuna* mature leaf powder had no significant effect on the total hardness concentration of the well water (Table 2).

Boiling at 100 °C for 10 min significantly reduced total hardness concentration, and the percentage reduction was 9.9%. However, the nitrate N concentration was significantly increased after boiling (Table 2).

Discussion

Groundwater is the major natural water resource in the Jaffna Peninsula and is used for domestic, agricultural and industrial purposes. However, the quality of groundwater is very important in serving for the intended purposes. The results of the present study indicated that in terms of nitrate–nitrogen concentration and total hardness, the well water in the study area is not suitable to be used for drinking. Increased hardness and nitrate in the groundwater of this region of the country have been a prevailing problem for a long period of time. Mikunthan and De Silva identified high levels of nitrates and increased hardness as major problems in the service wells of Kondavil region (Mikunthan and De Silva 2008). Further, a previous study carried out in the study area has resulted in average nitrate concentration that ranges from 8.2 to 29.8 mg/L with a mean value of 19.3 mg/L (Jeevaratnam et al. 2018). Another study recorded nitrate concentrations exceeding 100 mg/L in some agricultural and urban wells (Rajasooryar et al. 2002) in the area. A recent study conducted by Mahagamage et al. 2019 recorded nitrate concentrations in groundwater ranging between 0.01 and 38.51 mgL⁻¹. The nitrate concentrations recorded in the present study are comparatively lower than the previous studies. However, the values of the present study exceed the

SLSI nitrate concentration standards for safe drinking water. The high concentration of nitrates in the area can be due to application of artificial fertilizers and the disposal wastes in this area.

Mahagamage et al. 2019 recorded water hardness between 45.33 and 611.33 mgL⁻¹ and recorded hard or very hard water in most sampling locations (Mahagamage et al. 2019). The results of the present study record higher hardness values compared to the values recorded by Mahagamage et al. 2019. Further, the USGS (USGS 2016) categorizes water into 4 categories based on CaCO₃ hardness as soft (0 to 60 mgL⁻¹; moderately soft/moderately hard (61 to 120 mgL⁻¹); hard (121 to 180 L⁻¹); and very hard (> 180 mgL⁻¹). According to this classification, all the sampling locations of the present were categorized as very hard water. The increased hardness can be a result of CaCO₃ in the limestone aquifer (Mikunthan and De Silva 2008, Jeevaratnam et al. 2018).

Therefore, application of suitable water treatment methods to reduce high nitrate and hardness levels is very important to improve the drinkability of water.

According to the World Health Organization guidelines for safe drinking water, water containing more than 1000 mg/L of TDS is not recommended as suitable for drinking (WHO 2011). However, the TDS of the well water in the present study did not exceed this limit, and therefore, even the SLSI standards are exceeded, the water cannot be considered totally unfit for drinking purposes in terms of TDS.

In Sri Lanka, the most common water treatment method is boiling at 100 °C for 10 min. Although this is practised as a common disinfecting method throughout the country, this method cannot be recommended for areas which contain high nitrate concentration in the drinking water. The present study confirms this, and a previous study conducted in Chunakkam and Vadamarachchi areas in Jaffna Peninsula also confirmed that boiling increases nitrate concentrations (Wijeyaratne and Suvendran 2017).

Natural plant extracts have been used for water purification for many centuries and Egyptians inscription afforded the earliest recorded knowledge of plant materials used for water treatment, dating back to 2000 BC (Fahey 2005; Megersa et al. 2014). Plant materials are of great interest

for low-cost water treatment (Ghebremichael et al. 2005) which are also relatively more environmentally friendly than chemicals that are used for water treatment (Megersa et al. 2014) and could be utilized as a household water treatment method. Household water treatment interventions may play an important role in protecting public health especially in developing countries.

The present study indicated that the *Terminalia arjuna* mature leaf powder has a significant effect of reduction of nitrate N concentration of groundwater but no significant effect on total hardness. The bark powder of *Phyllanthus emblica* showed contrasting results indicating significant reduction of total hardness but no significant reduction in nitrate N concentration.

Primary active components of *Terminalia arjuna* leaves are flavonoids (phenolic compound) and glycosides. They have antioxidant properties which are very important when they are used as medicines in treatment for cancer patients (Amalraj and Gopi 2017). In addition, *Terminalia arjuna* leaves contain proteins, carbohydrate, vitamin and other ions which are responsible for the plant's ability of coagulation. The proteins and other active components that produce positive charges act like magnets and attract the predominantly negatively charged particles. (Ghebremichael et al. 2005; Saduzaman et al. 2013). This property is also important in using *Terminalia arjuna* leaves in water treatment as they can bind the negatively charged nitrates from the contaminated water (Saduzaman et al. 2013).

Terminalia arjuna is often planted near wells, rivers and other water bodies as its root is believed to purify and cool the water (Schmelzer and Ameenah 2008). Furthermore, studies of *Terminalia arjuna* seedlings (Illeperuma et al. 2012) and seeds (Ravindara et al. 2014) have showed significant reduction of hardness. However, the results of the present study indicate that the *Terminalia arjuna* leaves are not effective in treating hard water.

Use of *Phyllanthus emblica* wood is commonly practiced for drinking water treatment in rural areas in India and in many African countries (Padmapriya et al. 2015). This reduction of hardness may be due to chelation property of *Phyllanthus emblica* wood (Padmapriya et al. 2015). Further, *Phyllanthus emblica* wood has been identified as an effective treatment agent for water hardness due to its coagulation property to remove hardness causing Ca^{2+} and Mg^{2+} ions (Sathish et al. 2012). In the present study, a significant reduction (9.3% reduction and significant at 95% level of significance, Table 2) of total hardness was observed after filtering through *Phyllanthus emblica* bark powder. However, this treatment did not reduce the total hardness level to meet the SLSI standards for safe drinking water.

Terminalia arjuna and *Phyllanthus emblica* are two species that have a high medical importance. Different parts of these plants are used as important medicines for treatment

of cardiovascular diseases and diabetes. Some studies have proven that *Terminalia arjuna* is effective in treating water contaminated with cadmium and lead (Rao et al., 2016; Buhar 2017). However, the effects of the plant parts *Phyllanthus emblica* in water treatment assays are very rarely studied at the laboratory scale. Therefore, the results of the present study provide very important information on the hardness reduction ability of *Phyllanthus emblica* bark powder.

Conclusion

The water collected from the domestic wells in the Kondavil region, Jaffna peninsula, Sri Lanka, is not suitable for drinking in terms of the nitrate concentration and the total hardness concentration. However, filtration of this water through mature leaf powder of *Terminalia arjuna* can significantly reduce nitrate N concentration (21.4% reduction) to meet the standards stipulated by SLSI for safe drinking water. Further, filtration through *Phyllanthus emblica* bark powder can approximately account for 9.3% reduction of the total hardness of the raw water. Therefore, *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder can be used as a combined home remedial treatment technique for treating nitrate contaminated hard water. Further, boiling water before or after plant-based treatment may also help further reduce nitrate and hardness as well. It is also recommended that further research needs to be carried out to test the synergistic effects of using *Terminalia arjuna* mature leaf powder and *Phyllanthus emblica* bark powder in combined water treatment techniques.

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Data availability The raw data of the study can be made available upon request.

Compliance with ethical standards

Conflict of interest The author(s) declare(s) that there is no conflict of interest regarding the publication of this article.

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