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Fluoride in drinking water and its removal

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Abstract

Excessive fluoride concentrations have been reported in groundwaters of more than 20 developed and developing countries including India where 19 states are facing acute fluorosis problems. Various technologies are being used to remove fluoride from water but still the problem has not been rooted out. In this paper, a broad overview of the available technologies for fluoride removal and advantages and limitations of each one have been presented based on literature survey and the experiments conducted in the laboratory with several processes. It has been concluded that the selection of treatment process should be site specific as per local needs and prevailing conditions as each technology has some limitations and no one process can serve the purpose in diverse conditions.

Keywords: Fluoride; Fluorosis; Ground water; Soil water; Drinking water; Treatment

1. Introduction

Water is an essential natural resource for sustaining life and environment that we have always thought to be available in abundance and free gift of nature. However, chemical composition of surface or subsurface is one of the prime factors on which the suitability of water for domestic, industrial or agricultural purpose depends. Freshwater occurs as surface water and groundwater. Though groundwater contributes only 0.6% of the total water resources on earth, it is the major and the preferred source of drinking water in rural as well as urban areas, particularly in the developing countries like India because treatment of the same, including disinfection is often not required. It caters to 80% of the total drinking water requirement and 50% of the agricultural requirement in rural India. But in the era of economical growth, groundwater is getting polluted due to urbanization and industrialization.

Over the past few decades, the ever-growing population, urbanization, industrialization and unskilled utilization of water resources have led to degradation of water quality and reduction in per capita availability in various developing countries. Due to various ecological factors either natural or anthropogenic, the groundwater is getting polluted because of deep percolation from intensively cultivated fields, disposal of hazardous wastes, liquid and solid wastes from industries, sewage disposal, surface impoundments etc. [1–4]. During its complex flow history, groundwater passes through various geological formations leading to consequent contamination in shallow aquifers.

Presence of various hazardous contaminants like fluoride, arsenic, nitrate, sulfate, pesticides, other heavy metals etc. in underground water has been reported from different parts of India [5–9]. In many cases, the water sources have been rendered unsafe not only for human consumption but also for other activities such as irrigation and industrial needs. Therefore, now there is a need to focus greater attention on the future impact of water resources planning and development taking into consideration all the related issues. In India, fluoride is the major inorganic pollutant of natural origin found in groundwater. In this paper detailed review on sources, ill effects and techniques available for fluoride removal is done.

2. Occurrence and sources

Fluoride in minute quantity is an essential component for normal mineralization of bones and formation of dental enamel [10]. However, its excessive intake may result in slow, progressive crippling scourge known as fluorosis. There are more than 20 developed and developing nations that are endemic for fluorosis. These are Argentina, U.S.A., Morocco, Algeria, Libya,

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Table 1
Districts known to be endemic for fluoride in various states of India [18]

States	Districts	Range of fluoride concentration (mg/L)
Assam	Karbianglong, Nagaon	0.2–18.1
Andhra Pradesh	All districts except Adilabad, Nizamabad, West Godhavari, Visakhapattnam, Vijzianagaram, Srikakulam	0.11–20.0
Bihar	Palamu, Daltonganj, Gridh, Gaya, Rohtas, Gopalganj, Paschim, Champaran	0.6–8.0
Delhi	Kanjhwala, Najafgarh, Alipur	0.4–10.0
Gujarat	All districts except Dang	1.58–31.0
Haryana	Rewari, Faridabad, Karnal, Sonipat, Jind, Gurgaon, Mohindergarh, Rohtak, Kurukshetra, Kaithal, Bhiwani, Sirsa, Hisar	0.17–24.7
Jammu and Kashmir	Doda	0.05–4.21
Karnataka	Dharwad, Gadag, Bellary, Belgam, Raichur, Bijapur, Gulbarga, Chitradurga, Tumkur, Chikmagalur, Manya, Banglore, Mysore	0.2–18.0
Kerala	Palghat, Allepy, Vamanapuram, Alappuzha	0.2–2.5
Maharashtra	Chandrapur, Bhandara, Nagpur, Jalgaon, Bulduna, Amravati, Akola, Yavatmal, Nanded, Sholapur	0.11–10.2
Madhya Pradesh	Shivpuri, Jabua, Mandla, Dindori, Chhindwara, Dhar, Vidhisha, Seoni, Sehore, Raisen and Bhopal	0.08–4.2
Orrissa	Phulbani, Koraput, Dhenkanal	0.6–5.7
Punjab	Mansa, Faridcot, Bhatinda, Muktsar, Moga, Sangrur, Ferozpur, Ludhiana, Amritsar, Patila, Ropar, Jallandhar, Fatehgarh sahib	0.44–6.0
Rajasthan	All the 32 districts	0.2–37.0
Tamilnadu	Salem, Periyar, Dharampuri, Coimbatore, Tiruchirapalli, Vellore, Madurai, Virudunagar	1.5–5.0
Uttar Pradesh	Unnao, Agra, Meerut, Mathura, Aligarh, Raibareli, Allahabad	0.12–8.9
West Bengal	Birbhum, Bhardaman, Bankura	1.5–13.0

Egypt, Jordan, Turkey, Iran, Iraq, Kenya, Tanzania, S. Africa, China, Australia, New Zealand, Japan, Thailand, Canada, Saudi Arabia, Persian Gulf, Sri Lanka, Syria, India, etc. [11]. In India, it was first detected in Nellore district of Andhra Pradesh in 1937 [12]. Since then considerable work has been done in different parts of India to explore the fluoride laden water sources and their impacts on human as well on animals [13–17]. At present, it has been estimated that fluorosis is prevalent in 17 states of India (Table 1).

The safe limit of fluoride in drinking water is 1.0 mg/L [19]. The endemic fluorosis in India is largely of hydrogeochemical origin. It has been observed that low calcium and high bicarbonate alkalinity favor high fluoride content in groundwater [20,21]. Water with high fluoride content is generally soft, has high pH and contains large amount of silica. In groundwater, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, temperature, the action of other chemicals and the depth of wells. Due to large number of variables, the fluoride concentrations in groundwater range from well under 1.0 mg/L to more than 35.0 mg/L [22].

As the amount of water consumed and consequently the amount of fluoride ingested is influenced primarily by air temperature, USPHS [23] has set a range of concentrations for maximum allowable fluoride in drinking water for communities based on the climatic conditions as shown in Table 2.

Fluorine is highly reactive and is found naturally as CaF_2. It is an essential constituent in minerals like topaz, fluorite, fluorapatite, cryolite, phosphorite, theorapatite, etc. [24]. The fluoride is found in the atmosphere, soil and water. It enters the soil through weathering of rocks, precipitation or waste run off. Surface waters generally do not contain more than 0.3 mg/L of fluoride unless they are polluted from external sources. Though drinking water is the major contributor (75–90% of daily intake), other sources of fluoride poisoning are food, industrial exposure, drugs, cosmetics, etc. [25]. The fluoride content of some major food products is given in Table 3.

3. Health impacts of fluoride

Fluorine being a highly electronegative element has extraordinary tendency to get attracted by positively charged ions like calcium. Hence the effect of fluoride on mineralized tissues like

Table 2
USPHS recommendations for maximum allowable fluoride in drinking water

Annual average of maximum daily air temperature (°C)	Recommended fluoride concentration (mg/L)			Maximum allowable fluoride concentration (mg/L)
	Lower	Optimum	Upper	
10–12	0.9	1.2	1.7	2.4
12.1–14.6	0.8	1.1	1.5	2.2
14.7–17.7	0.8	1.0	1.3	2.0
17.8–21.4	0.7	0.9	1.2	1.8
21.5–26.2	0.7	0.8	1.0	1.6
26.3–32.5	0.6	0.7	0.8	1.4

Table 3
Fluoride concentration in agricultural crops and other edible items [26]

Food item	Fluoride concentration (mg/kg)
Cereals	
Wheat	4.6
Rice	5.9
Maize	
Pulses and legumes	5.6
Green gram dal	2.5
Red gram dal	3.7
Soyabean	4.0
Vegetables	
Cabbage	3.3
Tomato	3.4
Cucumber	4.1
Lady finger	4.0
Spinach	2.0
Lettuce	5.7
Mint	4.8
Potato	2.8
Carrot	4.1
Fruits	
Mango	3.7
Apple	5.7
Guava	5.1
Nuts and oil seeds	
Almond	4.0
Coconut	4.4
Mustard seeds	5.7
Groundnut	5.1
Beverages	
Tea	60–112
Aerated drinks	0.77–1.44
Spices and condiments	
Corriander	2.3
Garlic	5.0
Turmeric	3.3
Food from animal sources	
Mutton	3.0–3.5
Beef	4.0–5.0
Pork	3.0–4.5
Fishes	1.0–6.5
Others	
Rock salts	200.0–250.0
Areca but (supari)	3.8–12.0
Beetle leaf (pan)	7.8–12.0
Tobacco	3.2–38

bone and teeth leading to developmental alternations is of clinical significance as they have highest amount of calcium and thus attract the maximum amount of fluoride that gets deposited as calcium–fluorapatite crystals. Tooth enamel is composed principally of crystalline hydroxylapatite. Under normal conditions, when fluoride is present in water supply, most of the ingested fluoride ions get incorporated into the apatite crystal lattice of calciferous tissue enamel during its formation. The hydroxyl ion gets substituted by fluoride ion since fluorapatite is more stable than hydroxylapatite. Thus, a large amount of fluoride gets bound in these tissues and only a small amount is excreted through sweat, urine and stool. The intensity of fluorosis is not merely dependent on the fluoride content in water, but also on the fluoride from other sources, physical activity and dietary habits.

The various forms of fluorosis arising due to excessive intake of fluoride are briefly discussed below (Table 4) [27,28].

Table 4
Effects of fluoride in water on human health

Fluoride concentration (mg/L)	Effects
<1.0	Safe limit
1.0–3.0	Dental fluorosis (discoloration, mottling and pitting of teeth)
3.0–4.0	Stiffened and brittle bones and joints
4.0–6.0 and above	Deformities in knee and hip bones and finally paralysis making the person unable to walk or stand in straight posture, crippling fluorosis

3.1. Dental fluorosis

Due to excessive fluoride intake, enamel loses its lustre. In its mild form, dental fluorosis is characterized by white, opaque areas on the tooth surface and in severe form, it is manifestated as yellowish brown to black stains and severe pitting of the teeth. This discoloration may be in the form of spots or horizontal streaks [29]. Normally, the degree of dental fluorosis depends on the amount of fluoride exposure up to the age of 8–10, as fluoride stains only the developing teeth while they are being formed in the jawbones and are still under the gums. The effect of dental fluorosis may not be apparent if the teeth are already fully grown prior to the fluoride over exposure. Therefore, the fact that an adult shows no signs of dental fluorosis does not necessarily mean that his or her fluoride intake is within the safety limit.

3.2. Skeletal fluorosis

Skeletal fluorosis affects children as well as adults. It does not easily manifest until the disease attains an advanced stage. Fluoride mainly gets deposited in the joints of neck, knee, pelvic and shoulder bones and makes it difficult to move or walk. The symptoms of skeletal fluorosis are similar to spondylitis or arthritis. Early symptoms include sporadic pain, back stiffness, burning like sensation, pricking and tingling in the limbs, muscle weakness, chronic fatigue, abnormal calcium deposits in bones and ligaments. The advanced stage is osteoporosis in long bones and bony outgrowths may occur. Vertebrae may fuse together and eventually the victim may be crippled. It may even lead to a rare bone cancer, osteosarcoma and finally spine, major joints, muscles and nervous system get damaged.

3.3. Other problems

This aspect of fluorosis is often overlooked because of the notion prevailing that fluoride only affects bones and teeth [30]. Besides skeletal and dental fluorosis, excessive consumption of fluoride may lead to muscle fibre degeneration, low haemoglobin levels, deformities in RBCs, excessive thirst, headache, skin rashes, nervousness, neurological manifestations (it affects brain tissue similar to the pathological changes found in humans with Alzheimer's disease), depression, gastrointestinal problems, urinary tract malfunctioning, nausea, abdominal pain, tingling sensation in fingers and toes, reduced immunity, repeated abortions or still births, male sterility, etc. It is also responsible for alterations in the functional mechanisms of liver, kidney, digestive system, respiratory system, excretory system, central nervous system and reproductive system, destruction of about 60 enzymes. The effects of fluoride in drinking water on animals are analogous to those on human beings. The continuous use of water having high fluoride concentration also adversely affects the crop growth.

3.4. Solutions to the problem

A community with excessive fluoride in its water supply may meet the local MCL in one or more of several ways. Fluoride poisoning can be prevented or minimized by:

1. Using alternate water sources.
2. By improving the nutritional status of population at risk.
3. By removing excess fluoride (defluoridation).

3.4.1. Alternate water sources

Alternate water sources include surface water, rainwater and low-fluoride groundwater. Since surface water is often heavily contaminated with biological and chemical pollutants, it cannot be used for drinking purposes without treatment and disinfection making it too expensive and complex for application in poor communities. Rainwater is usually a much cleaner water source and may provide a low-cost simple solution. The problem however is its uneven distribution limited storage capacity in communities or households. The fact that fluoride is unevenly distributed in groundwater and its concentration keeps on changing with time both vertically and horizontally, implies that every well has to be tested individually and regular monitoring has to be done, which is not always possible in rural areas. Thus the option of using alternate water sources has its own limitations.

3.4.2. Better nutrition

Clinical data indicate that adequate calcium intake is directly associated with a reduced risk of dental fluorosis [31]. Vitamin C also safeguards against the risk [32]. Though, measures to improve the nutritional status of an affected population might be an effective supplement to the technical solutions of the problem, practically it sounds non-feasible.

3.4.3. Defluoridation of water

Defluoridation of drinking water is the only practicable option to overcome the problem of excessive fluoride in drinking water, where alternate source is not available. During the years following the discovery of fluoride as the cause of fluorosis, extensive research has been done on various methods for removal of fluoride from water and wastewater. These methods are based on the principle of adsorption [33], ion-exchange [34], precipitation–coagulation [35,36], membrane separation process [37,38], electrolytic defluoridation [39], electrodialysis [40–42], etc.

3.4.3.1. Adsorption. Several adsorbent materials have been tried in the past to find out an efficient and economical defluoridating agent. Activated alumina, activated carbon, activated alumina coated silica gel, calcite, activated saw dust, activated coconut shell carbon and activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tricalcium phosphate, bone charcoal, activated soil sorbent, carbion, defluoron-1, defluoron-2, etc., are different adsorbent materials reported in the literature [43–51]. The most commonly used adsorbents are activated alumina and activated carbon. The fluoride removing efficiency of activated alumina gets affected by hardness and surface loading (the ratio of total fluoride concentration to activated alumina dosage). Chloride does not affect the defluoridation capacity of activated alumina. The process is pH specific, so pH of the solution should be between 5.0 and 6.0 because at $pH > 7$, silicate and hydroxide become stronger competitor of the fluoride ions for exchange sites on activated alumina and at pH less than 5, activated alumina gets dissolved in acidic environment leading to loss of adsorbing media [52]. The process is highly selective but it has low adsorption capacity, poor physical integrity, requires acidification and pretreatment and its effectiveness for fluoride removal reduces after each regeneration.

Mckee and Johnston 1934, investigated the use of powdered activated carbon for fluoride removal and achieved good results [53]. The process is pH dependent with good results only at pH 3.0 or less. Therefore, the use of this material is expensive due to need of pH adjustment.

Activated alumina technique for defluoridation is being propagated in several villages by the voluntary organizations funded by UNICEF or other agencies to provide safe drinking water. Sarita Sansthan, Udaypur, Rajasthan is disseminating the technique with the practical assistance of UNICEF by providing a bucket (approximately 20 L capacity) fitted with a microfilter at the bottom containing 5 kg of activated alumina.

3.4.3.1.1. Advantages.

- The process can remove fluoride up to 90%.
- Treatment is cost-effective.

3.4.3.1.2. Limitations.

- The process is highly dependent on pH and works best only in a narrow pH range (5–6).
- High concentration of total dissolved salts (TDS) can result in fouling of the alumina bed.
- Presence of sulfate, phosphate or carbonate results in ionic competition.
- The process has low adsorption capacity, poor integrity and needs pretreatment.
- The regeneration is required after every 4–5 months and effectiveness of adsorbent for fluoride removal reduces after each regeneration.

- Disposal of fluoride laden sludge and concentrated regenerant is also a problem.

3.4.3.2. Ion-exchange. Fluoride can be removed from water supplies with a strongly basic anion-exchange resin containing quarternary ammonium functional groups. The removal takes place according to the following reaction:

$$\text{Matrix-NR}_3{}^{+}\text{Cl}^{-} + \text{F}^{-} \rightarrow \text{Matrix-NR}_3{}^{+}\text{F}^{-} + \text{Cl}^{-}$$

The fluoride ions replace the chloride ions of the resin. This process continues until all the sites on the resin are occupied. The resin is then backwashed with water that is supersaturated with dissolved sodium chloride salt. New chloride ions then replace the fluoride ions leading to recharge of the resin and starting the process again. The driving force for the replacement of chloride ions from the resin is the stronger electronegativity of the fluoride ions.

3.4.3.2.1. Advantages.

- Removes fluoride up to 90–95%.
- Retains the taste and colour of water intact.

3.4.3.2.2. Limitations.

- Efficiency is reduced in presence of other ions like sulfate, carbonate, phosphate and alkalinity.
- Regeneration of resin is a problem because it leads to fluoride rich waste, which has to be treated separately before final disposal.
- The technique is expensive because of the cost of resin, pre-treatment required to maintain the pH, regeneration and waste disposal.
- Treated water has a very low pH and high levels of chloride.

3.4.3.3. Coagulation–precipitation. Lime and alum are the most commonly used coagulants. Addition of lime leads to precipitation of fluoride as insoluble calcium fluoride and raises the pH value of water upto 11–12.

$$\text{Ca(OH)}_2 + 2\text{F}^{-} \rightarrow \text{CaF}_2 + 2\text{OH}^{-}$$

As lime leaves a residue of 8.0 mg F^-/L, it is used only in conjunction with alum treatment to ensure the proper fluoride removal [54–56].

As a first step, precipitation occurs by lime dosing which is followed by a second step in which alum is added to cause coagulation. When alum is added to water, essentially two reactions occur. In the first reaction, alum reacts with some of the alkalinity to produce insoluble aluminium hydroxide [$Al(OH)_3$]. In the second reaction, alum reacts with fluoride ions present in the water. The best fluoride removal is accomplished at pH range of 5.5–7.5 [57].

3.4.3.3.1. Advantages.

- The Nalgonda technique of defluoridation is based on combined use of alum and lime in a two-step process and has been claimed as the most effective technique for fluoride removal [58,59].
- Under Rajiv Gandhi Drinking Water Mission, several fill and draw (F&D) type and handpump attached (HPA) plants based on Nalgonda technique have come up in rural areas for which design and technology has been developed by NEERI, Nagpur.

3.4.3.3.2. Limitations. After having 10 years experience with these plants, the following serious drawbacks have been experienced:

- The process removes only a smaller portion of fluoride (18–33%) in the form of precipitates and converts a greater portion of ionic fluoride (67–82%) into soluble aluminium fluoride complex ion, and therefore this technology is erroneous. Also, as the soluble aluminium fluoride complex is itself toxic, adoption of Nalgonda technique for defluoridation of water is not desirable [60].
- Due to use of aluminium sulfate as coagulant, the sulfate ion concentration increases tremendously and in few cases, it crosses the maximum permissible limit of 400 mg/L, which causes cathartic effect in human beings.
- The residual aluminium in excess of 0.2 mg/L in treated water causes dangerous dementia disease as well as pathophysiological, neurobehavioural, structural and biochemical changes. It also affects musculoskeletal, respiratory, cardiovascular, endocrine and reproductive systems [61].
- Due to organoleptic reasons, users do not like the taste of treated water.
- Regular analysis of feed and treated water is required to calculate the correct dose of chemicals to be added, because water matrix keeps on changing with time and season as evident from our earlier studies conducted in laboratory.
- Maintenance cost of plant is very high. On an average as experienced in the recent years, a plant of 10,000 L per day capacity requires Rs. 3000 every month on maintenance.
- The process is not automatic. It requires a regular attendant for addition of chemicals and looking after treatment process.
- Large space is required for drying of sludge.
- Silicates have adverse effect on defluoridation by Nalgonda technique. Temperature also affects the defluoridation capacity.

3.4.3.4. Membrane process. Although various conventional techniques of water purification described earlier are being used at present to solve the problem of groundwater pollution, none of them is user-friendly and cost-effective technique due to some or the other limitation and has either no or very long pay back period. In the recent years, RO membrane process has emerged as a preferred alternative to provide safe drinking water without posing the problems associated with other conventional methods. RO is a physical process in which the contaminants are removed by applying pressure on the feed water to direct it through a semipermeable membrane. The process is the reverse of natural osmosis as a result of the applied pressure to the concentrated side of the membrane, which overcomes the natural osmotic pressure. RO membrane rejects ions based on size and electrical charge. The factors influencing the

Table 5
Comparative analysis of various techniques for fluoride removal

Sample no.	Initial fluoride concentration (mg/L)	Fluoride concentration after treatment (mg/L)			
		Activated alumina	Activated saw dust	Nalgonda	Reverse osmosis
1	4.2	1.13 (73.10)	1.42 (66.19)	1.32 (68.57)	0.32 (92.38)
2	7.8	1.96 (74.87)	2.32 (70.26)	2.24 (71.29)	0.63 (91.93)
3	8.6	2.23 (74.07)	2.56 (70.23)	2.47 (71.30)	0.78 (90.93)
4	9.3	2.11 (77.31)	2.42 (73.98)	2.31 (71.16)	0.88 (90.54)
5	8.2	2.17 (73.54)	2.43 (70.37)	2.34 (71.46)	0.77 (90.61)
6	6.8	1.81 (73.38)	2.16 (68.24)	1.95 (71.32)	0.56 (91.76)

Values in parentheses show the percent fluoride removal.

membrane selection are cost, recovery, rejection, raw water characteristics and pretreatment. Efficiency of the process is governed by different factors such as raw water characteristics, pressure, temperature and regular monitoring and maintenance, etc.

There are two types of membranes that can remove fluoride from water: NF and RO. NF is a relatively low pressure process that removes primarily the larger dissolved solids as compared to RO. Conversely, RO operates at higher pressures with greater rejection of all dissolved solids. Fluoride removal efficiencies upto 98% by membrane processes have been documented by many researchers.

In the past, the use of membrane technology for water treatment, particularly for drinking water production had been considered uneconomical in comparison with conventional means, but in the recent years the increased demand and contamination of water, rise in water quality standards and the problems associated with other methods have led to reconsideration of membrane technology for water purification. The progressive technical improvements in design and materials of the membranes have made the water treatment process economically competitive and highly reliable. Also, the capital and operational costs of RO plant go on decreasing with increasing plant capacity [62]. Thus with improved management, this new technology for drinking water production might be the best option. Furthermore, membrane processes present several advantages as compared with other treatment methods [63].

3.4.3.4.1. Advantages.

- The process is highly effective for fluoride removal. Membranes also provide an effective barrier to suspended solids, all inorganic pollutants, organic micropollutants, pesticides and microorganisms, etc.
- The process permits the treatment and disinfection of water in one step.
- It ensures constant water quality.
- No chemicals are required and very little maintenance is needed.
- Life of membrane is sufficiently long, so problem of regeneration or replacement is encountered less frequently.
- It works under wide pH range.
- No interference by other ions is observed.
- The process works in a simple, reliable automated operating regime with minimal manpower using compact modular model.

3.4.3.4.2. Limitations.

- It removes all the ions present in water, though some minerals are essential for proper growth, remineralization is required after treatment.
- The process is expensive in comparison to other options.
- The water becomes acidic and needs pH correction.
- Lot of water gets wasted as brine.
- Disposal of brine is a problem.
- The performance of all the above processes has been tested in the laboratory. A comparative analysis of the fluoride removal by various processes is presented in Table 5.

4. Conclusion

The literature survey and the laboratory experiments have indicated that each of the discussed techniques can remove fluoride under specified conditions. The fluoride removal efficiency varies according to many site-specific chemical, geographical and economic conditions, so actual applications may vary from the generalizations made. Any particular process, which is suitable at a particular region may not meet the requirements at some other place. Therefore, any technology should be tested using the actual water to be treated before implementation in the field.

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