Defluoridation of potable water by Nalgonda technique

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Fluoride, although beneficial when present in concentrations of 0.8–1.0 mg/l, has been associated with mottled enamel of teeth when present in potable waters in excess of 1.5 mg/l and skeletal fluorosis beyond 3 mg/l. A simple and effective process, the Nalgonda technique of difluoridation, has been developed by the National Environmental Engineering Research Institute. The process has been adapted for defluoridation of potable water at domestic as well as community level. The process details along with design data and cost of defluoridation are discussed here.

Fluoride and human health

Fluorine, a fairly common element of the earth's crust, is present in the form of fluorides in a number of minerals and in many rocks[1]. Industrial activities involving the use of fluorine-containing substances result in fluoride contamination of the environment. Thus, plants, food stuffs and water all contain traces of fluoride[2].

Fluorine has been fairly conclusively demonstrated to be an essential element for some animal species, in particular, fertility and growth rate are improved as a result of relatively small doses of fluorine[3]. The routes of human exposure to fluorides are essentially through drinking water and food and the exposure through air is insignificant in comparison with ingested fluorine[4]. Fluoride ingested with water is almost completely absorbed while that in the diet is not as fully absorbed[1]. Absorbed fluoride is distributed rapidly throughout the body and is retained mainly in the skeleton with a small portion in the teeth[3]. Once incorporated in the teeth, it reduces the solubility of the enamel under acidic conditions and thereby provides protection against dental caries.

Fluoride, although beneficial when present in concentrations of 0.8–1.0 mg/l, has been associated with mottled enamel of the teeth when present in potable waters in concentrations in excess of 1.5 mg/l. Skeletal fluorosis has been observed at concentrations beyond 3 mg/l. According to *WHO Guidelines for Drinking Water Quality* (1984), the excessive limit for fluoride is fixed as 1.5 mg/l, after which the water needs treatment for its removal.

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Excessive fluorides in drinking water

While traces of fluorides occur in many waters, higher concentrations are often associated with groundwaters. Excess fluoride in groundwaters is encountered in many parts of the world such as Afghanistan, Algeria, China, Egypt, India, Iran, Iraq, Jordan, Kenya, Libya, Morocco, Sudan, Tanzania, Thailand and Turkey. In India, it occurs in the states of Andhra Pradesh, Bihar, Gujarat, Haryana, Karnataka, Madhya Pradesh, Maharashtra, Orissa, Punjab, Rajasthan, Tamil Nadu and Uttar Pradesh[5]. There are 8700 villages in India which have the problem of excessive fluoride in water affecting 25 million people. Summarized information on the occurrence of excess fluoride in groundwater in various parts of India is presented in Table 1.

Methods of defluoridation

Since it became known that excessive fluoride in drinking water caused mottling of teeth in children, many methods have been suggested from time to time for removal of fluorides from water. These may be divided into two basic groups – those based on an exchange process or adsorption and those based on addition of chemicals to water being treated.

Reviews of these studies have been reported by Maier[6–8] and Savinelli and Black[9]. Boruff[10] was the first to investigate a variety of materials for fluoride removal, viz. aluminium sulphate, sodium silicate, ferric chloride, sodium aluminate, zeolites, bauxite, silica gel and lime. With the possible exception of lime and aluminium sulphate, none of these materials was found to be very practical. In addition to the studies on alum coagulation by Boruff and co-workers[10, 11], this technique has also been investigated by Kempf[12], Scott[13] and Culp and Stoltenberg[14]. Because the alum

Table 1. Occurrence of excessive fluoride in groundwaters in India

State	District	Total no. of samples analysed	No. of samples with fluoride > 1.5 mg/l	No. of samples with fluoride > 1.5 mg/l but TDS and nitrate within limits
Andhra Pradesh	Kurnool	143	82	55
Bihar	Palamu	191	35	28
Bihar	Rohatas	209	9	4
Gujarat	Mehsana	200	116	64
Haryana	Faridabad	200	46	28
Haryana	Gurgaon	429	116	68
Karnataka	Gulbarga	477	163	81
Madhya Pradesh	Jhabua	55	12	10
Maharashtra	Nagpur	114	8	7
Maharashtra	Latur	221	8	5
Maharashtra	Satara	1001	13	7
Maharashtra	Chandrapur	46	17	12
Orissa	Phulbani	225	5	5
Orissa	Koraput	324	8	4
Rajasthan	Barmer	351	180	16
Tamil Nadu	Ramnathpuram	66	3	0
	Total	4252	821	394

doses required for fluoride removal are much higher than those commonly used for turbidity and colour removal, the method of alum coagulation was not considered by the investigators, who concentrated on other methods using contact beds as a practical solution. McKee and Johnston[15] investigated powdered activated carbon for fluoride removal and showed that good results are obtainable only at pH 3 or less. Boruff[10] showed that lime softening removes fluoride and Scott[13] showed that the removal was a function of the amount of magnesium removed during the softening process. Others[10,16,17] also studied the use of lime and pointed out that water must be treated to a caustic alkalinity of 30 mg/l, at pH of 10.5 or above and as such recarbonation is necessary.

Granular tricalcium phosphate has been used[18–20] in several small-scale defluoridation systems in California but large-scale application has not shown it to be a good fluoride removal medium[21].

Activated alumina for fluoride removal was first studied by Boruff[10] and shortly thereafter by Fink and Lindsay[22]. Sorg[23] reviewing the methods of defluoridation, summarized that alum coagulation and lime softening treatments have very limited application and are not practical for defluoridation. Sorg opined that activated alumina is the most accepted and proven for fluoride removal from drinking water.

Strong base anion exchange resins remove fluorides either on hydroxyl cycle or chloride cycle along with other anions[24–26]. Since the proportional quantity for fluoride compared to other anions is very small, the effective capacity of such resins for fluoride works out quite low. There are no known commercial anion exchange resins which are selective for fluoride only. Some inorganic ion-exchangers, e.g. complex metal chloride silicate, formed from barium or ferric chloride with silicic acid, also exchange fluoride for chloride. Cation exchange resins impregnable with alum solution have been found to act as defluoridating agents[27–29].

Work done in India

Hariharan[30] reported the research carried out at the Indian Institute of Science on paddy husk carbon which, when processed chemically by cooking with caustic soda under pressure and washing free of alkali followed by impregnation with alum by soaking in this solution, has the capacity to remove fluoride from water when percolated through a bed of material.

Srinivasan[31] prepared a carbon from paddy husk by digestion in 1% KOH and soaking overnight in 2% alum solution. The material removed about 320 mg of F per kg (150 mg of F per litre) and showed a maximum removal efficiency at pH 7. The carbon was regenerated by using 2% alum solution for 12–14 h. A pilot plant with this material was installed at Guntakal, Andhra Pradesh to treat water containing 2.8 mg F/l. Activated carbons prepared by other workers[32,33] from cotton waste, coffee waste and coconut waste were tried for defluoridation but all these materials proved to be of academic interest only. Viswanadhan *et al.*[34] listed about 20 readily available materials for defluoridation. Magnesium oxide, calcium phosphate and aluminium oxide were reported effective media among the substances reported to have been examined, viz. calcium carbonate, lime ($CaCO_3$ heated to 950°C), granular activated carbon, 80-mesh activated carbon, chromatographic grade aluminium oxide, animal charcoal, bauxite (54% Al_2O_3, 7.2% Fe_2O_3), lateritic bauxite (31.5% Al_2O_3, 40.4% Fe_2O_3), laterite (15% Al_2O_3, 3.5% Fe_2O_3), silica gel, ordinary charcoal, saw-dust char, rice husk char, coconut shell char, magnesium carbonate, magnesium oxide, aluminium hydroxide heated to 140°C, iron hydroxide heated to 140°C, dibasic calcium phosphate, quartz and gypsum.

As a result of the studies on the comparative defluoridating capacities of different materials, Viswanadhan *et al.*[35] concluded that magnesium oxide had the highest capacity of removing fluorides from water at both acidic and alkaline pH. Venkateswarlu and Narayan Rao[36,37] studied the magnesia and calcined magnesite for fluoride removal from water and reported better fluoride removal capacities at higher temperatures.

Contribution by NEERI

Extensive studies were undertaken at NEERI since 1961 on materials like clays, minerals, ion exchange resins, activated carbons, sulphonated coals, activated alumina, activated magnesia, tamarind gel and serpentine minerals for removal of excess fluoride from water. Bulusu *et al.*[38] analysed and discussed the results of laboratory and field studies. The capacity of various defluoridating media, and problems in operation and limitations in their application are brought out. In order to overcome these, a method referred to as Nalgonda technique has been evolved by NEERI. The detailed cost analysis of all the methods has shown that the Nalgonda technique is by far the most economical and simple method available in India for defluoridation of water apart from its relative ease of operation and maintenance.

Nalgonda technique of defluoridation

The technique involves addition of aluminum salt, lime and bleaching powder followed by rapid mixing, flocculation, sedimentation and filtration. Aluminium salt may be added as aluminium sulphate or aluminium chloride or combination of these two. Aluminium salt only brings about the removal of fluoride from water. The dose of aluminium salt increases with increase in the fluoride and alkalinity levels of the raw water. The choice of the aluminium salt also depends on the sulphate and chloride contents of raw water so that their permissible limits are not exceeded. The dose of lime is empirically 1/20th that of the dose of aluminium salt. Lime facilitates forming denser floc or rapid settling. Bleaching powder is added to the raw water at the rate of 3 mg/l for disinfection. Based on extensive studies with fluoride-bearing waters, the approximate doses of alum required to obtain permissive limit (1 mg F/l) in water at various fluoride and alkalinity levels are given in Table 2.

Mechanism of defluoridation by Nalgonda technique

The chemical reaction involving fluorides and aluminium species is complex. It is a combination of polyhydroxy aluminium species complexation with fluorides and their adsorption on polymeric alumino hydroxides (floc). Besides fluorides, turbidity, colour, odour, pesticides and organics are also removed. The bacterial load is also reduced significantly. All these are by adsorption on the floc.

Alkalinity natural or supplemented by the addition of sodium carbonate or sodium bicarbonate, ensures effective hydrolysis of aluminium salts leaving no residual aluminium in the treated water.

The reactions which depend upon the nature of alkalinity are:

1) $3\,Al_2(SO_4)_3 \cdot 18H_2O + NaF + 9Na_2CO_3 \rightarrow [5Al(OH)_3 \cdot Al(OH)_2F] + 9Na_2SO_4 + NaHCO_3 + 8CO_2 + 45H_2O$

2) $3\,Al_2(SO_4)_3 \cdot 18H_2O + NaF + 17NaHCO_3 \rightarrow [5Al(OH)_3 \cdot Al(OH)_2F] + 9Na_2SO_4 + 17CO_2 + 54H_2O$.

Table 2. Approximate alum dose (mg/l) required to obtain permissive limit (1 mg F/l) of fluoride in water at various alkalinity and fluoride levels

Test water fluorides (mg F/l)	Test water alkalinity (mg $CaCO_3$/l)							
	125	200	300	400	500	600	800	1000
2	145	220	275	310	350	405	470	520
3	220	300	350	405	510	520	585	765
4	*	400	415	470	560	600	690	935
5	*	*	510	600	690	715	885	1010
6	*	*	610	715	780	935	1065	1210
8	*	*	*	*	990	1120	1300	1430
10	*	*	*	*	*	*	1510	1690

* To be treated after increasing the alkalinity with lime or sodium carbonate.

Table 3. Domestic defluoridation – approximate volume of 10% alum solution (ml) required to be added in 40 l test water to obtain permissive limit (1 mg F/l) of fluoride in water at various alkalinity and fluoride levels

Test water fluorides (mg F/l)	Test water alkalinity (mg $CaCO_3$/l)							
	125	200	300	400	500	600	800	1000
2	60	90	110	125	140	160	190	210
3	90	120	140	160	205	210	235	310
4		160	165	190	225	240	275	375
5			205	240	275	290	355	405
6			245	285	315	375	425	485
8					395	450	520	570
10							605	675

Table 4. Plant data for populations up to 200 on the basis of 40 LPCD defluoridated water

Population served	Daily supply (m^3)	Plant Dia. (m)	Recommended HP for motor
50	2	1 30	1.0
100	4	1.85	2 0
200	8	2 60	2.0

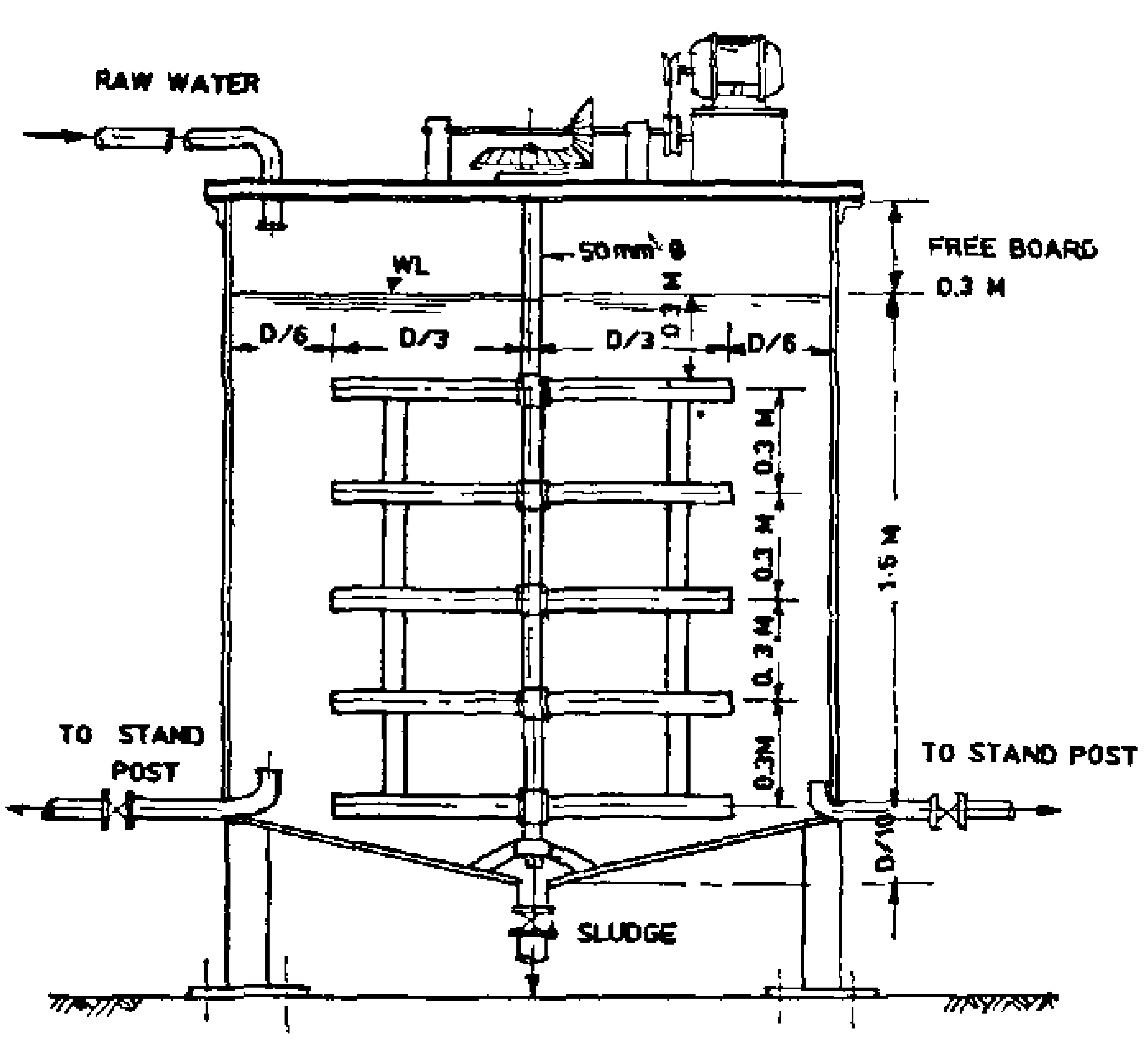

Figure 1. Fill-and-draw defluoridation plant for small community basis: 40 LPCD domestic water (for population up to 200). D = Diameter of plant.

Sedimentation following chemical mixing and flocculation, permits the resultant flocs loaded with fluorides, turbidity, bacteria, and other impurities to settle, thus reducing the concentration of suspended solids that must be removed by rapid sand filters. The filtered water collected in the storage water tank is rechlorinated, if necessary, with bleaching powder before distribution.

Nalgonda technique is effective even when the fluoride concentration is above 20 mg F/l, the dissolved solids above 1500 mg/l and hardness above 250 mg/l. The technique can be adapted for domestic as well as community water supply. Fill-and-draw or continuous operation systems can be installed for defluoridation of community water supply.

Domestic defluoridation

Defluoridation at domestic level can be achieved in a container (bucket) of 60 l capacity fitted with a tap 3–5 cm above the bottom of the container for withdrawal of treated water after precipitation and settling. The raw water taken in the container is mixed with adequate amounts of aluminium sulphate (alum) solution, lime or sodium carbonate and bleaching powder depending upon its alkalinity and fluoride content. Alum solution is added first and mixed well with water. Lime slurry is then added, the contents are stirred slowly for 20 min and allowed to settle for nearly one hour. The supernatant, which contains permissible amount of fluoride, is withdrawn through the tap for consumption. The settled sludge is discarded. Approximate volumes of 10% alum solutions for defluoridation of 40 l of water are given in Table 3.

Fill-and-draw defluoridation plant for small communities

The defluoridation plant for communities up to 200 population comprises a hopper-bottom cylindrical tank with a depth of 2 m and is equipped with a hand-operated or power-driven stirring mechanism (Figure 1). Raw water is pumped or poured into the tank and the required amounts of alum, lime or sodium carbonate and bleaching powder are added while stirring. The contents are stirred slowly for ten min and allowed to settle for two hours. The defluoridated supernatant water is withdrawn and supplied through standposts. The settled sludge is discarded. The plant data for populations up to 200 with a per capita water supply of 40 LPCD are summarized in Table 4.

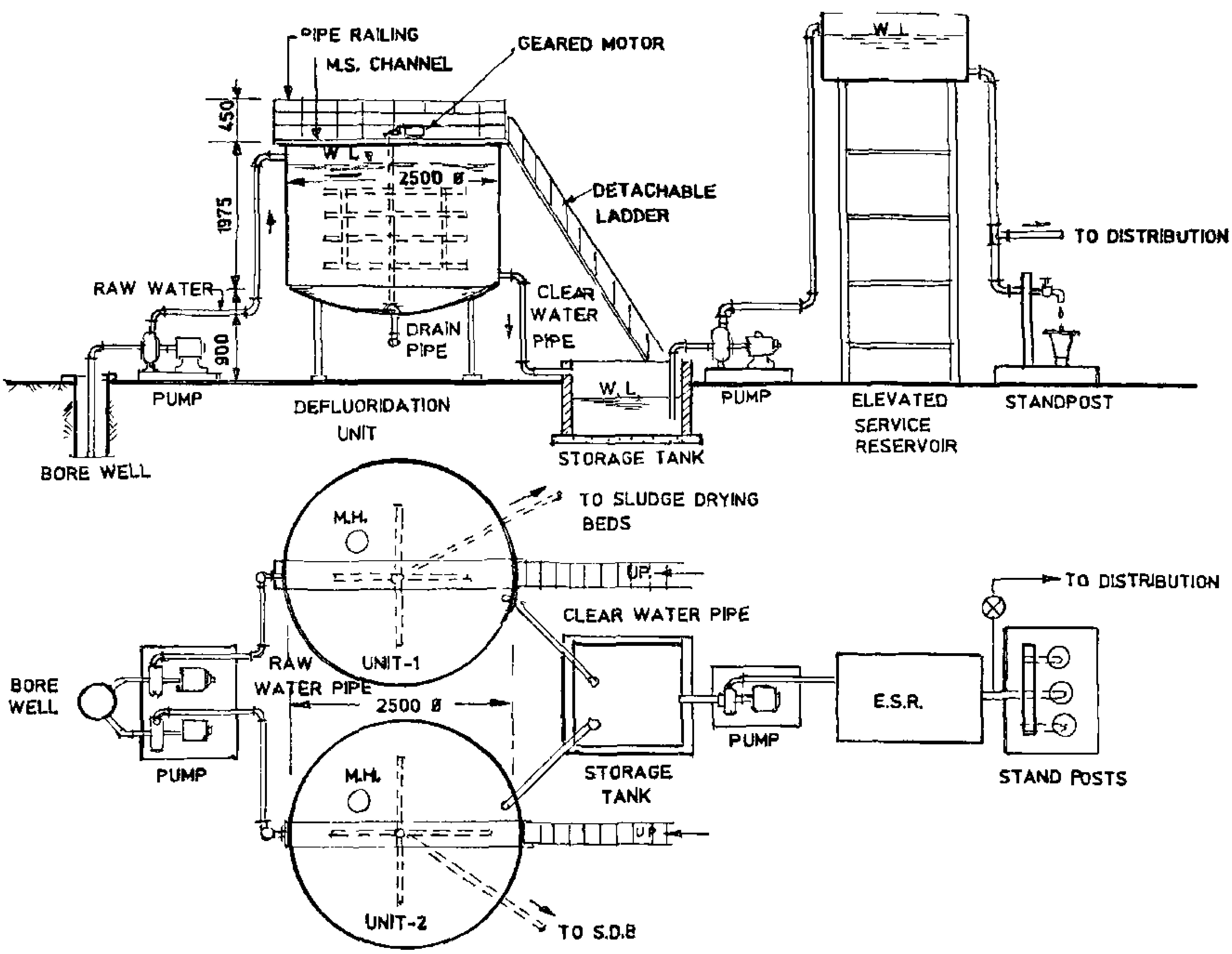

Figure 2. Fill-and-draw defluoridation plant for rural water supply.

Table 5. Raw and treated water characteristics

Village	Raw water pH	Raw water Alkalinity (mg $CaCO_3$/l)	Raw water Fluoride (mg F/l)	Treated water pH	Treated water Alkalinity (mg $CaCO_3$/l)	Treated water Fluoride (mg F/l)
Tartur (AP)	7.5	380	3.45	6.8	186	1.00
Fazilpur (Haryana)	8.1	450	2.50	6.9	320	0.90
Tavadia (Gujarat)	8.1	370	3.80	6.8	197	0.90
Badarpur (Gujarat)	8.1	780	3.20	6.6	380	0.92
Sheshpur (Gujarat)	8.0	200	6.20	6.7	68	0.92
Ranchodpura (Gujarat)	8.3	550	2.70	7.0	354	1.00
Samadhiyala (Gujarat)	8.2	640	4.00	7.0	367	1.00

Fill-and-draw defluoridation plant for rural water supply

The main components of fill-and-draw type defluoridation plant for rural water supply are depicted in Figure 2. The raw water from the source is pumped to the reactor which is made of HDPE, ferro-cement or RCC, and is circular in shape with dished bottom. The top of the reactor is provided with a sturdy lid and a manhole for inspection and for facilitating addition of chemicals into the reactor. The stirring mechanism consists of a motor, a reduction gear and mixing paddles attached to the shaft. Following the addition and mixing of appropriate doses of chemicals, the water in the reactor is allowed to settle. The settled water is drawn through an outlet valve into a sump well. The

Table 6. Cost estimates for defluoridation plants

Village	Plant capacity (m^3/day)	Capital cost (Rs)	Total annual cost of operation (Rs)	Total operational cost (Rs/m^3)	Running cost (Rs/m^3)
Tartur (AP)	40	212,000	90,500	6.20	1.52
Fazilpur (Haryana)	80	600,000	197,100	6.75	1 00
Tavadia (Gujarat)	40	405,000	155,000	5 31	1.53
Badarpur (Gujarat)	40	362,000	148,600	5.09	1.63
Sheshpur (Gujarat)	20	344,400	106,300	14.56	2.70
Ranchodpura (Gujarat)	180	874,700	305,500	4.65	1.39
Samadhiyala (Gujarat)	120	696,200	267,200	6.10	2.10

settled sludge after drying on the drying beds is disposed off.

In 1986 the Government of India launched the Technology Mission on Drinking Water and Related Water Management for supplying safe drinking water to rural population. Following water quality assessment of sources of water supply to rural areas, villages with the problem of excessive fluorides in water were identified in the states of Andhra Pradesh, Gujarat, Karnataka and Tamil Nadu. Seven fill-and-draw type defluoridation plants at Tartur, Fazilpur, Tavadia, Badarpur, Sheshpur, Ranchodpura and Samadhiyala have been installed based on Nalgonda Technique developed by NEERI. The performance of these plants is summarized in Table 5.

Cost of defluoridation

The capital and O&M costs for the seven defluoridation plants installed in different states of India are presented in Table 6. The O&M cost includes depreciation, interest on capital, maintenance, repairs, salaries of personnel and cost of chemicals and power.

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