# Mathematical and Experimental Approach for Removal of Fluoride, Arsenic, Lead by Non-Conventional (NLP, PLP, ALP) Absorbents

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**Abstract** - In the present study various types of leaves powder such as Neem Leaf Powder (NLP), Peepal Leaf Powder (PLP), Amla Leaf Powder (ALP) are used for the removal of fluoride, lead and arsenic from synthetic solutions. NLP, PLP, ALP leaves after experiments showed greater adsorption potential and strong affinity toward the ions. The experimental values regarding adsorption of different ions are extrapolated using MATLAB software to formulate corresponding quadratic equations. These equations can be used to interpolate the values of ions removal for any amount of adsorbent. The extrapolated values by MATLAB software helps, in theoretical calculations of the values without experiments, which in turn useful for the pilot project as well as to scale up the material at industrial level. The study of Adsorption kinetics clearly shows that the percentage removals of all three ions are increased by increasing adsorbent.

Keywords: Adsorption, experimental mathematical approach, leaves powders, Removal.

# **INTRODUCTION**

Water is a marvelous and unique substance which is essential for life. For the agronomist, water is necessary for food crops. Water is an important chemical compound as a cooling agent from engineering point of view. Water is a medium as well as climate factor for the biologist and meteorologist respectively. Its availability with good quality and adequate quantity is very important for human life and other purposes. In general, the quality of water is equally important as the quantity. Therefore, water quality is considered as an important factor to judge environment changes which are strongly associated with social and economic development

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## **1.1 FLUORIDE**

Fluoride is naturally abundant from 0.065% to 0.09% by weight in the earth's crust. Fluoride ion is extremely reactive. It is the 13th most abundant element on earth. In halogen group of minerals and are natural constituents of the environment the fluorides are one of them. Fluorides are originated soil of the earth's crust by the solvent action of water on the rocks. Fluoride never encountered in nature as it the most electronegative of all chemical elements and is exit in element form. [2]. Geological sources are the main sources of fluoride contamination which is the main source of drinking water contamination. [3, 4]

Fluoride contamination is injurious to health when taken in excessive amount above permissible limits of 1.0 mg/L, it leads to causes mottling of teeth in mild

cases, dental or skeletal fluorosis involve chronic problems, neurological damage and bone damage are sever problems [5].

# **1.2 AARSENIC**

Arsenic is very poisonous in nature and commonly referred to as king of poisons because it is powerful, prudent, odorless and tasteless; hence it can be easily camouflaged in food [6]. Arsenic is metalloid in nature mainly available in nature in local bedrocks. It comes into the soil through weathering of rocks and minerals, their leaching through anthropogenic sources. There are many source of Arsenic, inorganic arsenic sources are geological, while organic arsenic is found in abundance in marine life.

Surface water gets contaminated when geothermal fluids, rich in arsenic come in contact with it [7]. Arsenic may exist in different forms. The stability, solubility and toxicity of arsenic vary among its various forms. The inorganic forms of arsenic, namely arsenate and arsenite are more persistent and bio available; hence they are more hazardous as compared to their organic counterparts [3].

## **1.3 LEAD**

Industrial water act has present and future hazards for the environment by producing pollutants such as heavy metal contamination. Heavy metal contamination act as most crucial pollutants in surface and ground water [8]. The industrial wastewater contains toxic metals are Cd, Zn, Pb and Ni. Nowadays, due to population explosion there is continuous increase in heavy metal emissions into the environment and it is essential to control them [9,10] Toxic elements like lead causes many serious disorders like, anemia, kidney disease, nervous disorders, and even death [11].

# **OBJECTIVES OF THE STUDY**

The objective of the study is to extrapolate experimental values in theoretical calculations by using MATLAB software of all adsorption of different ions for removal process, such of the values which in turn useful for the pilot project as well as to scale up the material at industrial level.

## MATERIALS AND METHODS

# 1.1. PREPARATION OF LEAF POWDER (NLP, PLP AND ALP):

The preparation of the adsorbent, NLP, PLP and ALP included the various steps such as:

Firstly, Green neem leaves were collected from tree and were brought to the laboratory in plastic bags. The leaves were washed repeatedly with water to remove dust and then dry at room temperature in a shade. The dried leaves were ground and sieved to 50. The washed powder was dried for several hours at room temperature; then the powder was preserved in glass bottles for use as an adsorbent.

# **1.2. PREPARATION OF IONS SOLUTION**

In present study the removal of all selected ions by adsorption method was conducted in synthetic single ion solution. For this purpose, stock solution of desirable concentration for every ion was prepared. From this solution different standard solutions of 50 to 300 mg/L were prepared by dilution. 50 ml of each of these solutions was used in every experiment. All chemicals were used of A. R. Grade.

Effect of nature of adsorbent: The adsorption on all types of adsorbents was investigated. In each case a fixed amount (1.0 gm) was placed in a conical flask. An ion(s) solution was then added to each adsorbent followed by the pH adjustment by adding 0.01M NaOH of 0.1 m HCl. The flask was then capped and shaken for 1hr. at  $30^{\circ}C\pm 2^{\circ}C$ . This procedure was followed for all ions selected in the present investigation.

# 1.3. DETERMINATION METHODS OF IONS CONCENTRATION

Determination of Fluoride (F-) ion: JENWAY ion meter and a fluoride selective electrode (JENWAY ISE: 924305) equipped single junction calomel electrode was employed for the measurement of fluoride ion concentration.

## **Determination of Lead (Pb<sup>2+</sup>) ion:**

Atomic absorption spectrophotometer AA-6300 SHIMADZU was used for determination of lead. Lead Hollow Cathode Lamp at 283.3 nm and acetylene – air flame was used for this purpose.

# **Determination of Arsenic** (As<sup>3+/5+</sup>) ion:

The determination of arsenic in water samples was conducted by AAS with Flow Injection - Hydrides Generation (FI-HG) AAS system. AA-6300 SHIMADZU AAS, double beam atomic absorption spectrophotometer was also used for the determination of arsenic.

## **RESULTS AND DISCUSSION 1.4 Effect of Absorbent Doses On Fluoride Experimental Procedure**

The study shows effect of NLP, PLP and ALP dosage on removal of fluoride. At initial fluoride concentration of 5mg/L, pH 5.0, temperature $30^{\circ}\pm 2^{\circ}$ C, contact time 2hr., shaking time 1hr, 0.2-1.0gm is used.

Before reaching equilibrium, the contact time was ended. Results are seen in table 1.0, 2.0, 3.0 and figure 1. Results shows that by increasing adsorbent dose, percentage of fluoride removal is increased. The highest adsorption in case of NLP is 79.49%, in case of PLP is 81.89% and 82.76% in case of ALP. The adsorption capacity for ALP > PLP > NLP. Similar results have been reported by Researchers. [11 and 12]

Table 1

	Fluoride onto NLP					
A	mount of fluc	ride adsorbed at	t varying ad	sorbent		
		dosage				
(Ini	tial concentra	tion: 5mg/L, Tem	perature: 30°	$C \pm 2^{\circ}C$ ,		
	pH: 5.0, Con	tact time: 2hr., Sh	naking time:	1hr.)		
S. No.	Amount of Adsorbent (w)	Equilibrium Concentration (C <sub>e</sub> )	Adsorbed amount (q <sub>e</sub> )	Removal (R)		
	gm	mg/L	mg/g	%		
1	0.2	3.17	9.19	36.73		
2	0.4	2.96	5.09	40.76		
3	0.6	2.28	4.53	54.47		
4	0.8	1.90	3.88	61.93		
5	1.0	1.02	3.98	79.49		

## Table 2. Fluoride onto PLP

Amount of fluoride adsorbed at varying adsorbent dosage
(Initial concentration: $5mg/L$ , Temperature: $30^{\circ}C \pm 2^{\circ}C$ ,
pH: 5.0, Contact time: 2hr., Shaking time: 1hr.)

S. No.	Amount of Adsorbent(w)	Equilibrium Concentration (Ce)	Adsorbed amount(qe)	Removal (R)
	gm	mg/L	mg/g	%
1	0.2	3.07	9.68	38.71
2	0.4	2.89	5.27	42.13
3	0.6	2.18	4.71	56.50
4	0.8	1.79	4.01	64.28
5	1.0	0.90	4.09	81.89

Table 3. Fluoride onto ALP

Amount of fluoride adsorbed at varying adsorbent dosage
(Initial concentration: $5 \text{ mg/L}$ , Temperature: $30^{\circ}\text{C} \pm 2^{\circ}\text{C}$ ,
pH: 5.0, Contact time: 2hr., Shaking time: 1hr.)

S. No	Amount of Adsorben t (w)	Equilibrium Concentratio n (Ce)	Adsorbe d amount (qe)	Remova l (R)
	gm	mg/L	mg/g	%
1	0.2	2.97	10.18	40.71
2	0.4	2.59	6.04	48.37
3	0.6	2.17	4.72	56.64
4	0.8	1.53	4.33	69.29
5	1.0	0.87	4.13	82.76

## **Mathematical Formulation**

The experimental values of NLP, PLP and ALP dosage on fluoride removal is extrapolated using MATLAB software to formulate corresponding quadratic equations relating to –

- Amount of Adsorbent (x) and Equilibrium Concentration (y)
- Amount of Adsorbent (x) and Adsorbed amount (y)
- Amount of Adsorbent (x) and Removal (y)

These equations can be used to interpolate the values of fluoride removal for any amount of adsorbent.



Figure 1. Effect of different adsorbent dosage on Fluoride removal by NLP, PLP and ALP

Table 4.	Fluoride	onto NLI	2
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S.S No.	Amount of Adsorbe nt (w) gm	Equilibrium Concentration (Ce) mg/L	Adsorbed amount (qe) mg/g	Removal (R) %
Mat Ec	hematical quations	y=-1.85x*x- 0.45*x+3.354	y=14.83x*x- 23.62*x+12. 97	y=37.16x*x+8.75*x+3 3.07
1	0.3	3.05	7.22	39.04
2	0.5	2.66	4.87	46.73
3	0.7	2.12	3.70	57.40
4	0.9	1.44	3.73	71.04

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Table	Table 5. Fluoride onto PLP							
Amo	Amount of fluoride adsorbed at varying adsorbent dosage							
SNo.	Amount of Adsorb ent (w) gm	Equilibrium Concentrati on (Ce) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %				
Mathematical Equations		y=- 1.96x*x- 0.36*x+3. 24	y=15.78x*x- 25.16*x+13.7 0	y=38.91x*x+7.5 6*x+35.04				
1	0.3	2.95	7.57	40.80				
2	0.5	2.57	5.06	48.54				
3	0.7	2.02	3.82	59.39				
4	0.9	1.32	3.83	73.36				

### Table 6. Fluoride onto ALP

Amount of fluoride adsorbed at varying adsorbent dosage

S. No	Amount of Adsorben t (w) gm	Equilibrium Concentratio n (C <sub>e</sub> ) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %
Ma E	thematical quations	y=-1.39x*x- 0.95*x+3.21	y=15.73x*x- 25.78*x+14.4 2	y=28.57x*x+18.22*x+36. 04
1	0.3	2.79	8.10	44.07
2	0.5	2.38	5.46	52.29
3	0.7	1.86	4.08	62.79
4	0.9	1.22	3.95	75.57

# **1.5 Effect of Absorbent Doses On Lead Experimental Procedure**

The study shows effect of NLP, PLP, ALP dosage on removal of lead. At initial fluoride concentration of 0.1 mg/L, pH 6.5, temperature  $30^{\circ}\pm 2^{\circ}$ C, contact time 2hr., shaking time 1hr , 0.2-1.0gm is used. Before reaching equilibrium, the contact time was ended. Results are seen in table 7.0, 8.0, 9.0 and figure 2. Results shows that by increasing adsorbent dose, percentage of lead removal is increased. The highest adsorption in case of NLP is 85.2%; in case of PLP is 81.5% and 83.6% in case of ALP. There was no significant variation in % removal by all adsorbents. A similar result was observed with other adsorbent [13]



Figure 2. Effect of different adsorbent dosage on lead removal by NLP, PLP and ALP

#### Table 7. Lead onto NLP

	Amount of lead adsorbed at varying adsorbent dosage					
(	Initial concentration: 0.1mg/L,	Temperature: $30^{\circ}C \pm 2^{\circ}$	C, pH: 6.5 , Contact time: 2hr., ha	king time: 1hr.)		
S. No.	Amount of Adsorbent (w)	Equilibrium Conc(Ce)	Adsorbed amount(qe)	Removal(R)		
	gm	mg/L	mg/g	%		
1	0.2	0.09	0.03	7.4		
2	0.4	0.08	0.02	24.1		
3	0.6	0.04	0.89	53.2		
4	0.8	0.02	0.09	78.1		
5	1.0	0.01	0.09	85.2		

#### Table 8. Lead onto PLP

	Amount	t of lead adsorbed at varying adsorb	ent dosage	
(Ir	nitial concentration: 0.1mg/L, Te	emperature: $30^{\circ}C \pm 2^{\circ}C$ , pH: 6.5 , C	ontact time: 2hr., Shaking tii	me: 1hr.)
S. No.	Amount ofAdsorbent (w)	EquilibriumConc(Ce)	Adsorbed amount (qe)	Removal (R)
units	gm	mg/L	mg/g	%
1	0.2	0.09	0.05	10.6
2	0.4	0.08	0.05	21.4
3	0.6	0.05	0.09	48.2
4	0.8	0.03	0.09	69.1
5	1.0	0.01	0.09	81.5

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Fable 9. Lead ont	o ALP			
	Amount of	f lead adsorbed at varying ad	lsorbent dosage	
(Initial co	oncentration: 0.1mg/L, Ten	nperature: $30^{\circ}C \pm 2^{\circ}C$ , pH: 6.	5, Contact time: 2hr., Shaking	g time: 1hr.)
S. No.	Amount of Adsorbent (w)	Equilibrium Concentration (Ce)	Adsorbed amount (qe)	Removal (R)
units	gm	mg/L	mg/g	%
1	0.2	0.09	0.07	12.4
2	0.4	0.09	0.04	14.2
3	0.6	0.07	0.07	38.1
4	0.8	0.03	0.09	64.3
5	1.0	0.01	0.09	83.6

# Mathematical Formulation:

The experimental values of NLP, PLP and ALP dosage on lead removal is extrapolated using MATLAB software to formulate corresponding quadratic equations relating to –

- Amount of Adsorbent (x) and Equilibrium Concentration (y)
- Amount of Adsorbent (x) and Adsorbed amount (y)
- Amount of Adsorbent (x) and Removal (y)

These equations can be used to interpolate the values of lead removal for any amount of adsorbent.

## Table 10. Lead onto NLP

	Amount of lead adsorbed at varying adsorbent dosage				
S. No.	Amount of Adsorbent (w) gm	Equilibrium Concentration (C <sub>e</sub> ) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %	
Mathe	matical Equations	y=0.035x*x- 0.15*x+0.12	y=-2.94x*x+3.63*x-0.65	y=-41.78x*x+154.94*x-24.98	
1	0.3	0.07	0.17	17.74	
2	0.5	0.05	0.43	42.04	
3	0.7	0.03	0.45	63.00	
4	0.9	0.01	0.23	80.62	

# Table 11. Lead onto PLP

S. No.	Amount of Adsorbent (w) gm	Equilibrium Concentration (C <sub>e</sub> ) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %
Mathen	natical Equations	y=-0.017x*x- 0.083*x+0.11	y=-0.071x*x+0.14*x+0.018	y=-4.82x*x+100.53*x-12.04
1	0.3	0.08	0.05	17.68
2	0.5	0.06	0.07	37.02
3	0.7	0.04	0.08	55.96
4	0.9	0.02	0.08	74.53

## Table 12. Lead onto ALP

	Amount of lead adsorbed at varying adsorbent dosage				
S. No.	Amount of	Equilibrium Concentration	Adsorbed amount (q <sub>e</sub> )	Removal (R) %	
	Adsorbent (w) gm	$(C_e) \text{ mg/L}$	mg/g	× /	
Mathe	matical Equations	y=-0.10x*x+0.018*x+0.094	y=0.089x*x- 0.062*x+0.07	y=66.60x*x+16.32*x+3.42	
1	0.3	0.08	0.05	14.31	
2	0.5	0.07	0.06	28.23	
3	0.7	0.05	0.07	47.47	
4	0.9	0.03	0.08	72.05	

# **1.6 Effect of Absorbent Doses On Arsenic Experimental Procedure**

The study shows effect of NLP, PLP, ALP dosage on removal of arsenic. At initial fluoride concentration of 0.1mg/L, pH 6.5, temperature30°± 2°C ,contact time 2hr., shaking time 1hr, 0.2-1.0gm is used. Before reaching equilibrium, the contact time was ended. Results are seen in table 13.0, 14.0, 15.0 and figure 3. Results shows that by increasing adsorbent dose, percentage of arsenic removal is increased. The highest adsorption in case of NLP is 82.1%, in case of PLP is 81.0% and 80.6% in case of ALP. There was no significant variation in % removal by all adsorbents. Further we concluded that increasing adsorption dosage provides more available active sites for fixed arsenic concentration. As compared to all used adsorbent dosage, the adsorption capacity for NLP was higher. Similar results were reported by [14].



Figure 3. Effect of different adsorbent dosage on arsenic removal by NLP, PLP and ALP

## Table 13

-						
	Arsenic onto NLP					
Am	ount of arsenic	adsorbed at vary	ying adsorben	t dosage		
(Init	ial concentratio	n: 0.1mg/L, Ten	nperature: 30°	$^{\circ}C \pm 2^{\circ}C$ ,		
	pH: 6.5, Cont	act time: 2hr., Sl	naking time: 1	hr.)		
S. No.	S. Amount of Equilibrium Concentration Adsorbed Remove No. Adsorbent(w) (Ce) (R)					
	gm	mg/L	mg/g	%		
1	0.2	0.09	0.07	12.3		
2	0.4	0.08	0.07	25.4		
3	0.6	0.05	0.08	46.2		
4	0.8	0.02	0.09	78.1		
5	1.0	0.01	0.09	82.1		

Amount of arsenic adsorbed	l at varying adsorbent
dosage	

(Initial concentration: 0.1mg/L, Temperature: 30°C ± 2°C, pH: 6.5, Contact time: 2hr., Shaking time: 1hr.)				
S. No.	Amount of Adsorbent (w)	Equilibrium Concentration (Ce)	Adsorbed amount (qe)	Removal (R)
units	gm	mg/L	mg/g	%
1	0.2	0.01	0.09	17.3
2	0.4	0.02	0.08	29.1
3	0.6	0.04	0.08	42.2
4	0.8	0.07	0.09	64.1
5	1.0	0.01	0.09	81.0

#### Table 15. Arsenic onto ALP

Amount of arsenic adsorbed at varying adsorbent dosage
Initial concentration: $0.1 \text{mg/L}$ , Temperature: $30^{\circ}\text{C} \pm 2^{\circ}\text{C}$ ,
pH: 6.5, Contact time: 2hr., Shaking time: 1hr.)

S. No.	Amount of Adsorbent (w)	Equilibrium Concentration (Ce)	Adsorbed amount (qe)	Removal (R)
units	gm	mg/L	mg/g	%
1	0.2	0.09	0.01	14.1
2	0.4	0.08	0.02	23.4
3	0.6	0.07	0.03	39.7
4	0.8	0.03	0.07	67.2
5	1.0	0.01	0.09	80.6

### **Mathematical Formulation:**

The experimental values of NLP, PLP and ALP dosage on arsenic removal is extrapolated using MATLAB

# Software to formulate corresponding quadratic equations relating to –

- Amount of Adsorbent (x) and Equilibrium
- Concentration (y)
- Amount of Adsorbent (x) and Adsorbed amount (y)
- Amount of Adsorbent (x) and Removal (y)

These equations can be used to interpolate the values of arsenic removal for any amount of adsorbent.

## Table 16. Arsenic onto NLP

An	Amount of arsenic adsorbed at varying adsorbent dosage				
S. No.	Amount of Adsorb ent (w) gm	Equilibrium Concentration(Ce ) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %	
Matł Eq	nematical uations	y=0.0x*x- 0.11*x+0.11	y=0.00x*x+0.0 30*x+0.062	y=- 12.67x*x+111.3 6*x-12.42	
1	0.3	0.07	0.071	19.84	
2	0.5	0.05	0.077	40.09	
3	0.7	0.03	0.083	59.32	
4	0.9	0.011	0.089	77.54	

Table 17. Arsenic onto PLP						
Amount o	Amount of arsenic adsorbed at varying adsorbent dosage					
Amount of Adsorbent (w) gm	Equilibrium Concentration (Ce) mg/L	Adsorbed amount(q <sub>e</sub> ) mg/g	Removal (R) %			
Mathematic al Equations	y=- 0.23x*x+0.30 *x-0.05	y=0.053x*x- 0.059*x+0.0 98	y=33.92x*x+40.48*x+ 7.52			
0.3	0.01	0.08	22.71			
0.5	0.04	0.08	36.24			
0.7	0.04	0.08	52.48			
0.9	0.03	0.08	71.43			

### Table 18. Arsenic onto ALP

SS. No.	Amou nt of Adsor bent (w) gm	Equilibrium Concentration (C <sub>e</sub> ) mg/L	Adsorbed amount (q <sub>e</sub> ) mg/g	Removal (R) %
Mathe Equ	ematical ations	y=- 0.089x*x+0.002*x +0.094	y=0.089x* x- 0.002*x+0 .006	y=34.64x*x+46.82* x+1.66
1	0.3	0.08	0.01	18.82
2	0.5	0.07	0.02	33.73
3	0.7	0.05	0.04	51.41
4	0.9	0.02	0.07	71.86

### **CONCLUSION AND RECOMMENDATION**

NLP, PLP, ALP leaves after experiments showed greater adsorption area and strong affinity toward the ions. The experimental values of fluoride, arsenic and lead dosage on NLP, PLP and ALP removal is extrapolated using MATLAB software to formulate corresponding quadratic equations. These equations can be used to interpolate the values of ions removal for any amount of adsorbent. The extrapolated values by MATLAB software helps, in theoretically calculating the values without experiments, which in turn useful for the pilot project as well as to scale up the material at industrial level.

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