Sorption and desorption behaviour of iodine in alluvial soils of Varanasi, India

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Iodine is an essential element in *soil-plant-atmosphere-animal* continuum. In order to understand its behaviour in the soil, the sorption and desorption behaviour of iodine in alluvial soils of Varanasi was investigated in the present study. Further, the soil properties that govern iodine retention in soil, were also characterised. The rate and percentage of adsorption and desorption was higher in the University farm soil than from Kapsethi soil. The adsorption and desorption of iodine was very low and ranged between 1.4 to 4.0% for sorption and 1.35 to 4.10% for desorption. The higher sorption and desorption of iodine was due to strong correlation with particular soil properties like organic matter, clay and CaCO3.

Key words: sorption, desorption, iodine, alluvial soils

INTRODUCTION

Iodine is the least abundant element of halogen group presently aging its importance in *soil-plant-atmosphere-animal* continuum. It is a rather rare element although it can be found in nearly all media on the Earth (Delange et al. 2001). Iodine requirement of human and animals largely depends on food crops and water, consumed by people/animals because their organisms cannot synthesize iodine by themself . Food crops and water derive their iodine from soils. Iodine, present in the soil solution, can be taken up by plants in the both form of negatively charged ions of iodide and iodate. Lack of iodine in human nutrition leads to visible and invisible spectrum of health consequences collectively called iodine deficiency disorders (IDD) (Liao, 1992). To prevent its negative effect , iodine either have to be incorporated in the daily intake of individual (50-200 μg I day[-1]) or to be fortified in agricultural crops, especially in vegetables and leafy vegetables (Dai et al. 2004; Hong et al. 2009; Huanxin et al. 2003).

It is widely distributed in the nature in both, organic and inorganic forms. The chemical forms of iodine, present in the soil solution, vary from different regions and it is predominant as iodide in humid and temperate areas and iodate in alkaline soils of arid region. Its chemical forms in soil solution also vary under flooded and non flooded conditions: in the non flooded condition, IO_3^- is the dominant, most present in an insoluble form, while under flooded conditions, I^- is the dominant form of iodine (Yuita 1992). The iodine content of soils is reported to vary from 0.5-50 mg kg[-1], depending on the soil type and location. Its content in topsoil in Austria varied between 1.1-5.6 mg kg[-1] (Gerzabek et al. 1999) in Japan between 0.63-44.9 mg kg[-1] (Muramatsu et al. 1999) and between 2.7-36.9 mg kg[-1] in British soil

(Whitehead, 1973a). It's content in the soils of Tarai regions of India viz. Pantnagar, Bilashpur and Kaladungi were reported in the range from 3.65-7.54, 5.45-9.82 and 4.23-8.08 mg kg[-1], respectively (Singh et al. 2002).

The adsorption and desorption behaviour of iodine in soil is affected by several factors *i.e.* pH, organic matter, hydrous oxides of iron and aluminium, calcium carbonate, temperature and texture of the soils (Whitehead 1973, 1974, 1978). The amount of adsorbed iodine in soil increases with increasing content of organic matter, hydrous oxides of iron and aluminium and calcium carbonate. However, increase in pH and temperature reduces the amount of adsorbed iodide in soil, as reported by Whitehead (1973b) and Sheppard and Hawkins (1995). Its adsorption capacity also increases with increasing clay content in the soil, means coarse textured soils having less adsorption capacity than finer textured soils.

Iodide can be adsorbed on sesquioxides if pH is below 6.0. Iodide can also be bound to clay or to various organic matter in soils with pH lower than 6.9. At higher pH, iodide seems to be exchanged by OH[-]. Binding of iodide to organic material in the soil have been reported by several authors (Whitehead 1973, 1974, 1978, 1981; Sheppard and Hawkins 1995; Seki et al. 1984) and the retention of iodine in soil is often interpreted as binding to these organic components. In soil profiles, the highest levels of iodine are often found in the upper layers where also the organic content is higher (Fuge and Long 1989). The level of iodine is often the highest in peat soil which is probably due to its reaction with thyrosine, thiols and polyphenols in the organic part of the soil (Fuge 1987). Distribution of iodine in a soil profile shows large variation according to type of soil. These can be due to the distribution of binding molecules, but also due to the fact that iodine can show various oxidation levels depending on pH and Eh. In podsol, iodine accumulates in the organic material and in the B horizon, where it is associated with iron and aluminium oxides. In peat soil, the level of iodine is the highest in the organic layers and much lower in the

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underlying mineral soil layers. One complicating factor is that the bulk density of peat soil is about 20% of the bulk density of mineral soil. This means if iodine concentration is expressed as mg per dm^3, the differences is much smaller. The chemical species which seems to be the most efficient in binding iodine is thus organic matter and oxides of iron and aluminium (Whitehead 1973, 1974, 1978).

Sorption studies of iodine in soils and minerals have been carried out by several authors (Whitehead 1973, 1974, 1978, 1984; Yoshida et al. 1992; Tanabe et al. 2004). However, the sorption data in alluvial soil were scarce, especially the ?sorption data. In this work, the sorption and desorption behaviour of iodine in alluvial soil were determined using a range of iodide concentration from 2-10 ppm. The relative contribution of different soil properties that govern iodine retention was also studied.

MATERIAL AND METHODS

Two surface soil samples (0-0.15 m) of the established cultivated land of Alluvial soil belongs to *Entisol* soil type. Samples were taken randomly in Varanasi (25^018' N Latitude, 83^003' E Longitude, altitude 128.83 m) from BHU and Kapsethi respectively. In the agricultural research farm, the predominant cropping system is *Rice-Wheat-Green manuring* crop for the last twenty years with recommended package of practices as per ICAR (Indian Council of Agricultural Research) recommendation. On the contrary, the second representative soil was bought from Kapsethi farm with prominent cropping system r*ice-wheat/mustard-vegetable* with frequent alteration of crops depending on the fluctuation of available resources and weather in a subsistence mode. The climate is predominantly sub humid and subtropical with annual temperature between 5 and 46^0

Table 1: General soil properties

Parameters	BHU	Kapsethi
Physical properties		
Bulk density (Mg m^{-3})	1.22	1.26
Particle density (Mg m^{-3})	2.50	2.55
Water holding capacity (%)	49.95	46.28
Sand (%)	47.81	49.83
Silt (%)	30.29	30.57
Clay (%)	21.90	19.60
Soil texture	Sandy loam	Sandy loam
Electro-chemical and chemical properties		
pH	7.90	7.60
Electrical conductivity (dSm^{-1})	0.248	0.218
$CaCO^3$ (%)	0.75	0.37
Organic carbon (g kg^{-1})	3.50	3.20
Cation exchange capacity (mol kg^{-1})	1.38	1.09
Available nitrogen (kg ha^{-1})	214.21	195.59
Available phosphorus (kg ha^{-1})	20.18	18.58
Available potassium (kg ha^{-1})	225.74	201.15
Available sulphur (kg ha^{-1})	24.84	19.33
Hot water extractable iodine (mg kg^{-1})	1.81	0.64
NaOH extractable iodine (mg kg^{-1})	1.96	1.14

Table 2: Adsorption pattern of iodine in soils

Location of soil	Iodine applied (mg kg^{-1})	Equilibrium concentration Ce (mg L^{-1})	Amount of iodine adsorbed x/m (mg kg^{-1})	log C_e	log $\frac{x}{m}$
	2.0	1.79	0.08	0.253	-1.076
	4.0	3.66	0.13	0.563	-0.866
BHU	6.0	5.62	0.15	0.749	-0.818
	8.0	7.6	0.16	0.881	-0.796
	10.0	9.59	0.16	0.982	-0.785
	2.0	1.84	0.06	0.265	-1.194
	4.0	3.76	0.09	0.575	-1.078
Kapsethi	6.0	5.7	0.12	0.756	-0.921
	8.0	7.64	0.14	0.883	-0.842
	10.0	9.63	0.14	0.983	-0.830

C and with an average rainfall 1100 mm per year of (90 % of rainfall is distributed during June to September). The soil samples were air dried, ground in wooden pestle and mortar and passed through a 2 mm stainless steel sieve.

The bulk and particle density were determined using pycnometer bottle (Black 1965), water holding capacity (WHC) using Keen Rackzowski box (Black 1965), particle size distribution by international pipette methods (Black, 1965), pH used a combined electrode (Jackson 1967) & EC (1: 2.5 soil water suspension), $CaCO_3$ by rapid titration method (Piper 1951), organic matter by chromic acid wet digestion method (Walkely and Black 1934), Cation Exchange Capacity (CEC) by neutral N Ammonium Acetate (Jackson 1967), plant available nitrogen by alkaline permanganate method (Subbiah and Asija 1956), phosphorus (Olsen et al. 1954), potassium flame photometry (Jackson 1967) and sulphur by the turbidimetric method (Chesnin and Yien 1950). Total (Whitehead 1973) and water-soluble iodine (Newton 1951) content in soils was extracted and analyzed spectrophotometry (Bedi 1999).

ADSORPTION AND DESORPTION EXPERIMENT

Soil was shaken with different concentrations of KI solution and after the equilibrium period, the suspension centrifuged and filtered. There after soil was again centrifuged with same amount of distilled water for desorption and filtered. The equilibrium concentration of iodide in the supernatant solutions was determined by using spectrophotometer as method proposed by Bedi (1999). Equilibration was carried out by shaking 2.5 g oven dry soil with 25 mL of various quantities of iodide solution (0, 2.0, 4.0, 6.0, 8.0 and 10.0 mg kg^{-1}) in triplicate, in the glass tube fitted with polyethylene caps, on an end-over-end shaker. Two drops of chloroform was added to each tube in order to suppress microbial activity. The samples were shaken for 5 hour at normal laboratory temperature (14–23^{0}C). The amount of iodine (adsorbed and desorbed) was assessed in comparison with control solutions, prepared without the addition of soil (Whitehead 1973)-blind samples?

The specific adsorption of iodine by soil (C_s) and the corresponding equilibrium iodine concentration (C_e) were fitted to the following adsorption isotherm equation namely

Freundlich Equation

The amount of iodine adsorbed in soil (C_s) was calculated from the difference between the initial and equilibrium concentration (C_e). The data was fit to logarithmic from form by? the Freundlich equation:

$$\log C_s = \log K_{f_{ads}} + \frac{1}{n_{ads}} \log C_e$$

$\log K_{f_{ads}}$ and $\frac{1}{n_{ads}}$ are the constants representing the intercept and slopes of the isotherms, respectively.

The distribution coefficient, Kd was calculated as $Cs/Ce\left[or \sum (C_s.C_e)\middle/ \sum (C_e)^2\right]$ to measure the adsorption extant. The adsorption constant ($K_{f_{des}}$)normalized to soil organic carbon content, K_{oc} and to soil clay content, K_c These are important parameters that play a significant role in environmental fate assessment of iodine and were evaluated using the following equation:

$$K_{OC} = \frac{K_d x 100}{\text{Organic carbon (\%)}},$$

$$K_C = \frac{K_d x 100}{\text{Clay (\%)}}$$

For studying iodine desorption, the soil from the sorption run was resuspended and the amount desorbed from soil was calculated from the difference between the concentrations of iodine before and after equilibration in desorption experiment. The data was fit to the logarithmic form of Freundlich equation and $K_{f_{des}}$ and $\frac{1}{n_{des}}$ were obtained as intercept and slopes of the isotherm. A desorption hysteresis coefficient (h) was calculated as the ratio between the sorption and desorption isotherm slopes:

$$h = \frac{1}{n_{ads}} \middle/ \frac{1}{n_{des}}$$

The statistical difference between soils were subjected to analysis of paired t test, performed using windows based SPSS (*version 17*).

RESULTS

The characteristic of the soil under adsorption / desorption study are presented in the Table 1 revealed that there were no significant differences in the physical properties of the soils *viz*. bulk density, particle density and water holding capacity of the soils. However, BHU (Agriculture Research Farm) soil showed better physical condition in comparison to Kapsethi (Farmer's personal farm) soil. The textural class of the both soils, *i.e.* BHU and Kapsethi, were sandy loam (light textured).

Moreover, there were wide variations in the electrochemical and chemical properties of the soils (Table 1). The hot water extractable and total iodine content in BHU farm soil were comparatively higher than Kapsethi soil. The pH, EC, free $CaCO_3$, organic carbon, organic matter, CEC, available N, P, K and S were higher in the BHU farm soil than Kapsethi farmer's farm soil.
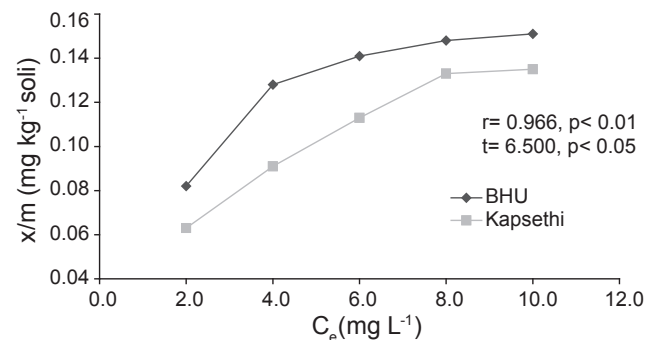

r= 0.966, p< 0.01
t= 6.500, p< 0.05

Fig. 1: Adsorption pattern of iodine in soils of

Varanasi

Adsorption of Iodine

The data of iodine adsorption (based on iodine content) in the selected two soils of district Varanasi are presented in the Table 2 and 3 and adsorption pattern in the soils of BHU (Agricultural Research Farm) and Kapsethi (Farmer's personal farm) are depicted in the Fig. 1. The adsorption isotherm was drawn between the log values of the amounts of iodine adsorbed on the soils and the amount of iodine present in the equilibrium concentration (mg L^{-1}) in soil solution, respectively at fixed volume of solvent. The adsorption of iodine initially increased with increasing iodine concentration but at later stages of higher concentration, adsorption rate decreased with increasing concentration of iodine.

Table 3: Frendlich adsorption isotherm values

Adsorption parameter	BHU	Kapsethi
$K_{f_{ads}}$	1.331	0.973
$\dfrac{1}{n_{ads}}$	0.514	0.202
$K_{D_{ads}}$	0.025	0.020
K_{OC}	7.143	6.250
K_C	0.127	0.091

Where, $K_{f_{ads}}$ = intercept, $\dfrac{1}{n_{ads}}$ = slope of adsorption, $K_{D_{ads}}$ = distribution coefficient, K_{OC} = organic matter coefficient, K_C = clay coefficient.

The adsorption data of iodine were fitted in the Freundlich adsorption equation, $\dfrac{x}{m} = K_f C^{\frac{1}{n}}$ where K_f and $\dfrac{1}{n}$ are constants, associated with the affinity of adsorbent for the adsorbate or adsorption capacity of soil at equilibrium concentrations and to the degree of curvature of the isotherm or intensity of adsorption, respectively. The Freundlich constants of iodine in the soils of BHU and Kapsethi are $K_{f_{ads}}$, $\dfrac{1}{n_{ads}}$, KD_{ads}, K_{OC} are presented in the Table 4 and Fig. 3. It was revealed from the Table 4 that the adsorption of iodine was higher in the BHU than in Kapsethi soil as the Freundlich's constant values are higher in BHU farm soil. The role of organic carbon and clay content in binding iodine is evaluated through the sorption index. The higher values of KD_{ads}, K_{OC} and K_C were observed in BHU soil comparing to Kapsethi Kapsethi soil. Iodine is held with greater tenacity within soil with higher adsorbing capacities i.e. organic mattrix (6.03 g kg^{-1}), clay content (assemblage of minerals) (219 g kg^{-1}) and free $CaCO_3$ content (7.5 g kg^{-1}), which were comparatively higher in BHU soil.
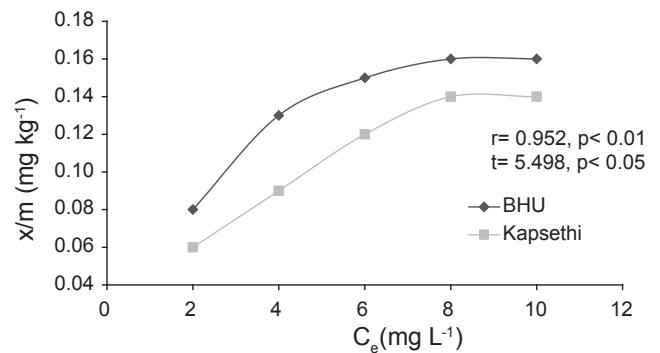

Fig. 2: Desorption pattern of iodine in soils of Varanasi

DESORPTION OF IODINE IN THE SOILS

The data of iodine desorption in the soils of Varanasi *viz.* BHU (Agricultural Research Farm) and Kapsethi (Farmer's personal farm) soil are shown in the Table 3 and 4, while desorption patterns are shown in Fig. 2. Desorption isotherm also were in L-type. The percent desorption of iodine in soils varied from 1.51 to 4.10% in the BHU soil and from 1.35 to 3.15% in the Kapsethi soil. Comparing observed soils, the percent of iodine adsorption was higher in BHU (t=5.498, p<0.05). The hysteresis "h" was calculated from adsorption

Table 4. Desorption pattern of iodine in soils

Location of soil	Iodine applied (mg kg^{-1})	Equilibrium concentration Ce (mg L^{-1})	Amount of iodine desorbe $\dfrac{x}{m}$ (mg kg^{-1})	log C$_e$	log $\dfrac{x}{m}$
BHU	2.0	0.006	0.082	-2.222	-1.086
	4.0	0.020	0.128	-1.699	-0.893
	6.0	0.028	0.141	-1.553	-0.851
	8.0	0.031	0.148	-1.5.09	-0.830
	10.0	0.033	0.151	-1.481	-0.821
Kapsethi	2.0	0.003	0.063	-2.523	-1.201
	4.0	0.012	0.091	-1.921	-1.041
	6.0	0.018	0.113	-1.745	-0.947
	8.0	0.028	0.133	-1.553	-0.876
	10.0	0.032	0.135	-1.495	-0.870

& desorption isotherms constant data and depicted in Table 4 and Fig. 3 and 4. It was revealed from the Table 4 that as the hysteresis value in BHU soil was higher than the soil of Kapsethi, the desorption capacity of iodine in the BHU was higher than in Kapsethi soil.
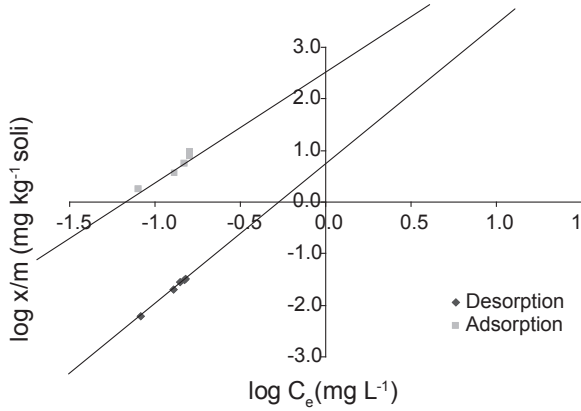

Fig. 3: Adsorption-desorption isotherms of iodine in soil of BHU (Agril. Res. Farm)

The ratio between $\frac{1}{n}$ for adsorption and desorption isotherm is a measure of the extent of hysteresis coefficient (h). When h is less than one (h<1), a positive hysteresis is observed. For example, $0.7 < h < 1$, no hysteresis could be defined, while h is greater than one (h>1) negative or irreversible adsorption is expected (Bailey and White 1970). It was observed from the data presented in the Table 5, that the value of hysteresis of iodine adsorption – desorption in both BHU and Kapsethi soil was 3.2 and 1.49, respectively. Thus, the negative hysteresis was recorded in both soils This means the adsorption and desorption of iodine in the both the soils was irreversible.
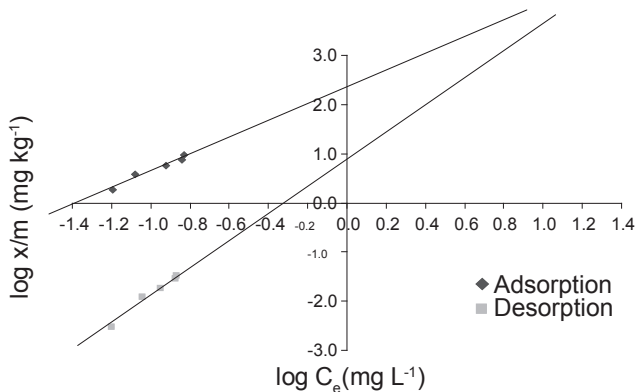

Fig. 4: Adsorption-desorption isotherms of iodine in soil of Kapsethi

DISCUSSION

Increasing rate adsorption of iodine was grater at lower concentration than at higher concentration of applied iodine as KI. Thus, the adsorption isotherm was L-type (Langmuir type) in both soils Giles et al. (1960) suggested that isotherm shape provides an indication of the adsorption mechanism operating for a given solute – solvent adsorbent system.

According to Giles, there are four types of adsorption mechanism *i.e.* solvent affinity type (S – type), Langmuir type (L – type), constant partition type (C – type) and high affinity type (H – type). In L – type, isotherm has an initial slope that is curvilinear and concave with respect to the amount of adsorbed iodine. It was confirmed by $\frac{1}{n}$ values for both soils under study *i.e.* $\frac{1}{n} < 1$. The L-type isotherm has an initial slope *i.e.* non-linear and concave with respect to the abscissa. The percent of adsorption varied from 1.6-4.0 in the BHU (Agriculture Research Farm) soil and from 1.4-3.0 in the Kapsethi (Farmer's Personal Farm). The percentage adsorption of iodine was greater at lower concentration than at higher concentration in both soils. The rate and percentage of adsorption was higher in the BHU than in Kapsethi soil (t=6.50, p<0.05). But, overall the adsorption of iodine in alluvial soil was very low (only 1.4 to 4.0%) which is in conformity with the general rule, that anion (I^-) sorption decrease as pH increases. Above pH 7, I^- is not readily adsorbed, due to loss of positively charged sites and completion with hydroxyl ions. The findings was in agreement with Whitechead (1974), who reported that the sorption of I^- in surface soils at nearly neutral pH was due to higher organic matter content and sorption increased with degree of humification. The pH of soils under study was higher than 7, thus, the level of adsorption were very low.

Table 5: Frendlich desorption isotherm values

Desorption parameter	BHU	Kapsethi
$K_{f_{des}}$	0.292	0.375
$\frac{1}{n_{des}}$	0.063	0.344
$K_{D_{des}}$	5.508	5.752
h	3.21	1.49

Where, $K_{f_{des}}$ = intercept, $\frac{1}{n_{des}}$ = slope of adsorption, $K_{D_{des}}$ = distribution coefficient, h = hysteresis

The above results of the present study indicate that iodine deficiency in these soils is manifested though the irreversible nature of iodine sorption capacity largely depends on the pH, organic carbon and clay content. The hot water soluble iodine content of these soils further moderated the adsorption properties of these anions. For higher hysteretic and higher sorption capacity of soil, iodine supplementation is needed to ensure efficient management of iodine in soil-plant system and safeguard against iodine deficiency disorders.

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