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Contamination of Paddy Soil and Rice with Arsenic

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Abstract

The drinking water and food are main pathway entry of the As in humans and animals. Their intakes cause diseases *i.e.* skin cancer, vascular disorder, etc. A wide variety of the rice is cultivated in the central India. The field soil and rice cultivated in the summer season at Koudikasa village, central India were selected for the As contamination studies. The concentration (n = 20) of total-As (AsT) in the field soil, rice grain, husk, straw and root was ranged from 44 - 270, 0.17 - 0.72, 0.40 - 1.58, 2.5 - 5.9 and 204 - 354 mg/kg with mean value of 126 ± 28, 0.47 ± 0.07, 0.83 ± 0.15, 4.2 ± 0.5 and 276 ± 21 mg/kg, respectively. The total arsenic, monomethylarsenonate, dimethylarsinite and inorganic As in the rice grain are quantified.

Keywords

Arsenic, Soil, Rice, Accumulation, Speciation

1. Introduction

Arsenic is a toxic metalloid, and its long exposure causes diseases *i.e.* hypopigmentation, melanosis, keratosis, skin, bladder, lung, and prostate cancer, etc. [1]. Rice is a predominant staple food in many countries *i.e.* Asia, Pacific, America and Africa. The rice husk and straw are used as fodder for feeding cattle and fuel. The exposure of arsenic from rice has been reported a global health issue [2]-[4]. The elevated levels of As in the field soils in various regions of the World were reported [5]-[12]. Arsenic is a bioactive toxic element, accumulated in rice of several regions of the World [13]-[19]. In the present work, the contamination of As in the paddy soil, rice grain, husk, straw and root grown in the contaminated environment, Kaudikasa village, Ambagarh Chowki, central India is described. The speciation and translocation of As in various parts of the rice plants are discussed.

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2. Methods and Materials

2.1. Choice of Study Area

The Ambagarh Chowki block, Rajnandgaon district, Chhattisgarh state, India is a hot spot for the As contamination research due to the huge mineralization of As in the environment [20] [21]. The studied area falls in a tribal belt with population of ≈0.1 million over 155 villages. Among them, Koudikasa village (area ≈ 5 km^2) was selected for the proposed studies due to As contamination of the environment at the hazardous levels.

2.2. Sampling of Soil and Rice Samples

The sampling network for collection of field water, soil and rice is presented in **Figure 1**. The water samples were collected as prescribed in the literatures [22]. Twenty water samples (once in a month) in duplicate from August-November, 2012 were taken from 20 different rice fields. A total of 4 samples, from each field from the

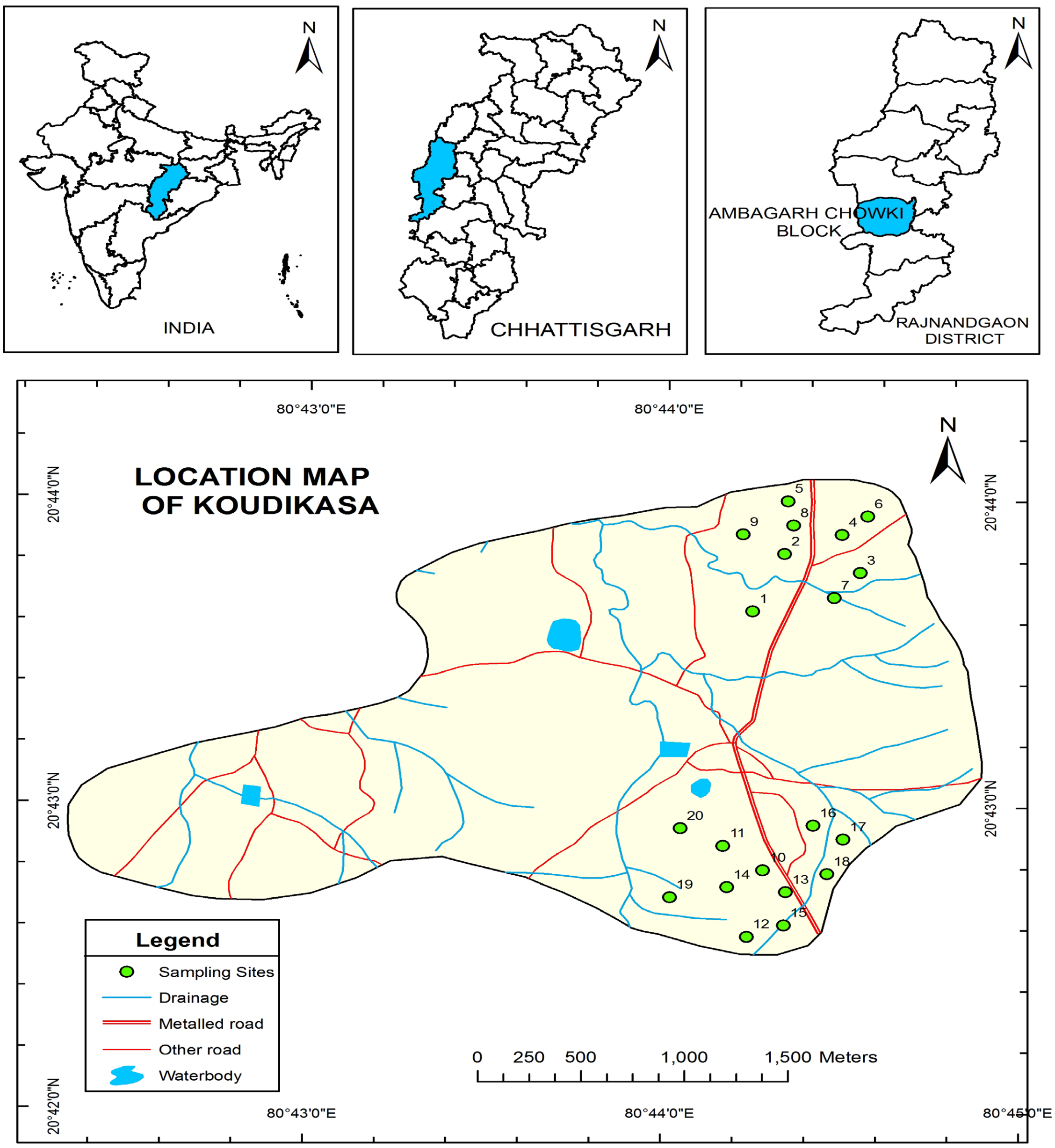

Figure 1. Sampling net-work for collection of the field soil and rice samples in Koudikasa village.

period: August-November, 2012 was taken for the investigation. They were collected directly from rice field in duplicate, and placed in 250-mL polypropylene plastic bottle. Bottles were first rinsed thrice with the water and then, completely filled with the same water. The first sample was acidified with concentrated nitric acid (0.1%, v/v) for analysis of the As. The second sample was left free for analysis of the ions. The physical parameters *i.e.* pH and electrical conductivity (EC) of the water were measured at the spot.

Twenty composite soil samples (0 - 10-cm depth) were collected after harvesting of rice paddy (December, 2009) from 20 fields of Koudikasa village as prescribed in the literature [23]. The samples were stored in polyethylene bottles and dried in open air under diffused sunlight followed by drying in oven at 50˚C for 24 hr. Rice grains were separated from the plants by hand picking in December, 2012 from the field in the polyethylene bag. They were dried in the oven at 50˚C for 24 hr and their husk was separated manually. Similarly, the straw and root of the rice plant were collected. They were washed with the deionized water several times to remove the soil particles. The dried soil, rice grain, husk, straw and root samples were ground to a fine powder with mortar and passed through a mesh sieve of <1 mm.

2.3. Analysis

The Bruker S2 Picofox TXRF portable spectrometer was used for the analysis of the elements in soil. A suspended solution was prepared by mixing 10 mg of soil sample with 10 ml of a water solution containing 1% (w/v) triton and 10 µg/mL Ga in ultrasonic bath for 15 min. For each measurement, 10 µL of sample solution was sprayed on the quartz filter with subsequent drying. The X-ray source was focused on the filter for quantification of the elements. The peak area of the signal was computed. The three replicate measurements for each sample were carried out. The content of 14 elements (*i.e.* P, K, Ca, As, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb) in each soil was analyzed. The standard soil sample (NCS DC 73382 CRM) was used for the quality control.

The As content in the soil and rice samples were analysed by the ICP-MS and AFS techniques. The samples were digested with nitric acid and H_2O_2 in Perkin-Elmer microwave system. The total, inorganic and organic As species in the rice grain samples were quantified by using method proposed by Williams *et al.* [24]. The Rice flour SRM 1568 was used for the quality control.

3. Results and Discussion

3.1. Rice Morphology

The details of the rice morphology (*i.e.* cultivation period, height of plant, grain and husk content) are summarized in **Table 1**. The cultivation period, plant height and yield of rice varies by variety and environmental conditions, ranging from 80 - 145 day, 90 - 140 cm and 33 - 57 Q/ha with mean value of 124 day, 110 cm and 44 Q/ha, respectively. Among them, the plant height was found to be partially correlated (r = 0.42) with the cultivation period. The single grain weight, rice and husk content were varied from 14 - 30 mg, 65% - 87% and 13% - 35% with mean value of 21 mg, 74% and 26%, respectively.

3.2. Characteristics of Field Water

The mean value (n = 4) of the water parameters in 20 fields is summarized in **Table 2**. The pH value of the water was found to be neutral, ranging from 6.9 - 7.6 with mean value of 7.3 ± 0.1. The moderate EC values of the water were observed, ranging from 180 - 472 µS/cm with mean value of 316 ± 34 µS/cm. The concentration of F^-, Cl^-, SO_4^{2-}, NO_3^-, Na^+, K^+ and As was ranged from 1.7 - 3.4, 10 - 43, 32 - 150, 1.9 - 7.9, 14 - 50, 2.6 - 9.8 and 0.029 - 0.090 mg/L with mean value of 2.4 ± 0.2, 20 ± 4, 78 ± 12, 4.2 ± 0.7, 32 ± 7, 5.2 ± 0.8 and 0.058 ± 0.007 mg/L, respectively. Among them, SO_4^{2-} showed the highest content in the water. The concentration of F^- and As was found to higher than the recommended value of 1.5 and 0.001 mg/L, respectively [25].

3.3. Concentration of As and Other Elements in Soils

The distribution of As and other elements *i.e.* P, K, Ca, Cr, Mn, Fe, Ni, Cu, Zn and Pb in the field soil is shown in **Table 3**. The concentration of elements *i.e.* As, P, K, Ca, Cr, Mn, Fe, Ni, Cu, Zn and Pb in the field soil (n = 20) was ranged from 44 - 270, 823 - 2653, 29,182 - 76,242, 12,173 - 116,031, 574 - 1306, 1623 - 14,419, 57,111 - 157,542, 245 - 548, 237 - 441, 444 - 817 and 9 - 79 mg/kg with mean value of 126 ± 28, 1516 ± 195, 45192 ± 5305, 35292 ± 12135, 883 ± 73, 8395 ± 1587, 94804 ± 10650, 392 ± 39, 343 ± 26, 582 ± 50 and 32 ± 10 mg/kg,

Table 1. Rice morphology in Koudikasa village.

S. No.	Type	Cultivation Period, day	Height of rice plant, cm	Yield Q/Ha	Grain type	Weight of grain, mg	Rice %	Husk %
1	IR-64	100	90	43	LS	22.6	78	22
2	Culture	80	110	43	MS	14.5	72	28
3	Shyamla	130	90	33	LS	25.0	76	24
4	G. Gurmatia	140	140	37	LB	29.6	81	19
5	Masuri	145	120	47	MS	14.3	87	13
6	Purnima	100	90	37	LS	21.6	79	21
7	Mahamaya	125	110	57	LB	28.8	83	17
8	Kalinga	100	120	37	LS	17.1	82	18
9	Luchai	145	120	42	MS	13.6	75	25
10	Safari	140	140	47	MS	16.3	72	28
11	Ek Hazar Das	115	98	50	LS	26.0	70	30
12	Sarna	140	110	50	MS	22.0	65	35
13	HMT	130	110	43	LS	20.0	65	35
14	M2	140	105	45	LS	23.0	68	32
15	Zero JR	125	100	45	MS	22.0	65	35
16	Sonan	130	100	43	LS	24.0	70	30

LS = Long slender, LB = Long Bold, MS = Medium size.

Table 2. Characteristics of field water in Koudikasa village.

Field no.	pH	EC	F^-	Cl^-	SO_4^{2-}	NO_3^-	Na	K	As
		µS/cm				mg/L			
Fi1	7.3	382	2.1	19	150	5.2	40	5.2	60
Fi2	7.4	180	2.7	25	85	3.4	17	3.9	53
Fi3	7.1	310	1.9	10	56	3.5	23	3.2	48
Fi4	7.3	221	2.0	12	32	4.3	40	6.5	65
Fi5	7.4	312	1.8	13	47	3.8	50	8.2	49
Fi6	7.1	229	2.7	31	72	7.2	14	2.6	29
Fi7	7.5	325	3.2	24	76	2.8	31	4.7	56
Fi8	7.2	310	3.4	43	92	1.9	21	4.3	90
Fi9	7.1	305	1.8	11	103	2.8	23	3.4	49
Fi10	7.2	439	2.3	18	105	5.7	34	6.2	88
Fi11	6.9	207	2.9	25	101	3.7	29	4.7	68
Fi12	7.2	472	2.1	10	69	3.9	24	3.8	62
Fi13	7.4	254	1.8	12	37	4.7	45	7.8	64
Fi14	7.5	374	1.7	13	59	4.2	46	9.8	63
Fi15	7.3	248	2.6	30	103	7.9	26	3.1	76
Fi16	7.1	389	2.5	24	83	3.1	35	5.6	49
Fi17	7.5	372	2.9	42	44	2.1	24	5.2	54
Fi18	7.6	351	2.0	10	71	3.1	25	4.1	34
Fi19	7.4	311	2.8	14	89	4.6	31	4.7	62
Fi20	7.2	319	2.2	17	92	5.2	38	7.7	41

Fi = Field.

Table 3. Chemical characteristics of field soil in Koudikasa village, mg/kg.

S. No.	As	P	K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Pb
1	148	1528	42,773	40,453	878	12,065	100,476	413	437	648	64
2	110	1425	36,673	38,177	811	14,292	104,938	436	418	606	79
3	90	1761	46,996	54,885	838	10,286	98,347	410	357	584	62
4	122	1560	35,543	37,082	922	9280	98,261	398	379	488	21
5	132	2113	49,238	97,179	961	11,212	157,542	475	441	817	64
6	44	1368	29,182	15,357	882	8077	90,405	320	279	455	46
7	186	914	60,933	15,381	673	3560	71,388	245	314	533	10
8	270	1133	76,242	13,477	963	1673	69,027	401	310	601	21
9	112	2653	56,127	116,031	1099	10,472	140,329	495	425	768	66
10	250	1235	65,635	12,173	879	2509	57,111	278	255	536	9
11	140	1219	51,707	12,236	717	3846	68,913	262	335	561	16
12	104	1318	48,651	14,209	640	5226	68,909	301	283	470	16
13	110	1268	41,638	53,294	782	14,419	95,301	466	377	526	46
14	104	1939	42,435	35,031	947	7287	87,998	355	300	491	10
15	234	1173	35,658	27,387	574	7925	72,626	256	237	444	20
16	70	1389	32,930	20,603	986	9676	107,638	425	365	547	28
17	74	1877	33,457	22,435	1306	11,632	111,356	497	359	768	25
18	56	1534	34,941	23,668	948	7722	96,383	548	283	526	12
19	91	823	42,334	31,345	836	8201	95,473	411	363	514	11
20	78	2093	40,740	25,427	1014	8534	103,649	439	341	761	14

respectively. Among them, the highest concentration of Fe was observed in all soil samples. They were found to occur in the following increasing order: Pb < As < Cu < Ni < Zn < Cr < P < Mn << Ca < K < Fe. Among them, a good correlation (r = 0.73) of the As with the K was observed, indicating their existence as K_3AsO_4 in the soil, **Table 4**. The Fe showed good correlation with the heavy metals *i.e.* Cr, Mn, Ni, Cu and Pb in the soil, indicating origin from similar sources, **Table 4**. The As concentration in the soil of studied area was found to be higher than reported in other regions of the country and World [5]-[12].

3.4. Distribution of As in Rice Grain, Husk, Straw and Root

The distribution of As in rice grain, husk, straw and root is summarized in **Table 5**. The concentration of As in the rice grain, husk, straw and root (n = 20) was ranged from 0.17 - 0.72, 0.40 - 1.58, 2.5 - 5.9 and 204 - 354 mg/kg with mean value of 0.47 ± 0.07, 0.83 ± 0.15, 4.2 ± 0.5 and 276 ± 21 mg/kg, respectively. The As content in husk was found to be higher than the rice grain, may be due to external contamination from the environment. The high yield rice varieties *i.e.* Kalinga, IR-64, G. Gurmatia, Shyamla, Ek Hazar Das, M2, etc. were found to be more sensitive to the As-accumulation. The concentration of As in the rice of the studied area was found to be higher than reported in the other region of the country and World [13]-[19]. The As content in the straw and root was found to be comparable to the values reported in the rice plants from Taiwan region of China [14].

3.5. Biological Absorption Coefficient and Concentration Factor

The biological absorption coefficient, BAC (*i.e.* plant to soil metal ratio) and concentration factor, CF (*i.e.* plant to water soluble metal ratio) of As are presented in **Table 6**. The CF and BAC values depend on the physical

Table 4. Correlation matrix for elements in field soil.

	P	K	Ca	Cr	Mn	Fe	Ni	Cu	Zn	As	Pb
P	1.00										
K	−0.12	1.00									
Ca	**0.71**	0.04	1.00								
Mn	0.41	−0.64	0.54	0.28	1.00						
Fe	**0.73**	−0.30	0.82	0.57	**0.69**	1.00					
Ni	0.55	−0.28	0.51	**0.70**	**0.64**	**0.74**	1.00				
Cu	0.44	−0.09	**0.68**	0.37	**0.69**	**0.77**	0.58	1.00			
Zn	**0.70**	0.15	0.57	**0.66**	0.34	**0.72**	0.58	**0.64**	1.00		
As	−0.36	**0.73**	−0.14	−0.35	−0.54	−0.48	−0.50	−0.29	−0.12	1.00	
Pb	0.42	−0.14	**0.67**	0.13	**0.71**	**0.63**	0.40	**0.71**	0.43	−0.19	1.00

Table 5. Distribution of As in various parts of rice in Koudikasa village, mg/kg.

S. No.	Rice type	Grain	Husk	Straw	Root
1	IR-64	0.46	1.13	5.4	354
2	Culture	0.25	0.40	2.8	204
3	Shymla	0.56	0.84	4.1	282
4	G.Gurmatia	0.62	1.11	5.2	306
5	Kalinga	0.68	1.58	5.9	348
6	Masuri	0.35	0.55	3.1	222
7	Purnima	0.27	0.47	2.8	216
8	IR-64	0.62	1.29	5.6	318
9	Ek Hazar Das	0.72	0.94	4.6	282
10	Culture	0.48	0.76	3.8	258
11	Luchai	0.50	1.36	5.7	330
12	Safari	0.17	0.46	2.5	210
13	Mahamaya	0.38	0.51	3.2	246
14	Sarna	0.51	0.72	4.7	276
15	HMT	0.48	0.68	3.8	258
16	M2	0.64	1.02	5.1	294
17	HMT	0.32	0.51	2.8	240
18	Masuri	0.22	0.44	2.7	234
19	Zero JR	0.62	0.98	4.9	330
20	Sonan	0.57	0.83	4.4	312

Table 6. BAC and CF value of rice grain, husk, straw and root.

S. No.	Rice	CF				BAC			
		Grain	Husk	Straw	Root	Grain	Husk	Straw	Root
1	IR-64	8	19	90	5898	0.003	0.008	0.036	2.4
2	Culture	5	8	53	3852	0.002	0.004	0.025	1.9
3	Shymla	12	18	85	5874	0.006	0.009	0.046	3.1
4	G.Gurmatia	10	17	80	4710	0.005	0.009	0.043	2.5
5	Kalinga	14	32	120	7104	0.005	0.012	0.045	2.6
6	Masuri	12	19	107	7656	0.008	0.013	0.070	5.0
7	Purnima	5	8	50	3858	0.001	0.003	0.015	1.2
8	IR-64	7	14	62	3534	0.002	0.005	0.021	1.2
9	Ek Hazar Das	15	19	94	5754	0.006	0.008	0.041	2.5
10	Culture	5	9	43	2934	0.002	0.003	0.015	1.0
11	Luchai	7	20	84	4854	0.004	0.010	0.041	2.4
12	Safari	3	7	40	3390	0.002	0.004	0.024	2.0
13	Mahamaya	6	8	50	3846	0.003	0.005	0.029	2.2
14	Sarna	8	11	75	4380	0.005	0.007	0.045	2.7
15	HMT	6	9	50	3396	0.002	0.003	0.016	1.1
16	M2	13	21	104	6000	0.009	0.015	0.073	4.2
17	HMT	6	9	52	4446	0.004	0.007	0.038	3.2
18	Masuri	6	13	79	6882	0.004	0.008	0.048	4.2
19	Zero JR	10	16	79	5322	0.007	0.011	0.054	3.6
20	Sonan	14	20	107	7608	0.007	0.011	0.056	4.0

and chemical composition of the field water, type of rice species and their morphology. The CF values in the rice grain, husk, straw and root was ranged from 3 - 15, 7 - 32, 40 - 120 and 2934 - 7656 with mean value 9 ± 2, 15 ± 3, 75 ± 11 and 5065 ± 1646, respectively. Relatively very low BAC values for the rice grain, husk, straw and root were observed, ranging from 0.001 - 0.009, 0.003 - 0.015, 0.015 - 0.073 and 1.0 - 5.0 with mean value of 0.004 ± 0.001, 0.008 ± 0.002, 0.039 ± 0.007 and 2.7 ± 0.5, respectively. The remarkably higher FC and BAC values with new rice variety *i.e.* Kalinga, Masuri, Ek Hazar Das and M2 were seen, may be due to smaller grain size and higher cultivation period, **Figure 2** and **Figure 3**. In addition, the FC and BAC values for different plant parts were found to be fairly correlated (r = 0.76 - 0.96). The As was found to be poorly translocated from the root to grain and its concentration was found to increase in order: grain < husk < straw << root. The As content was found 1.9 ± 0.3, 9 ± 1 and 108 ± 15 folds higher in the rice husk, straw and root with respect to the respective grain. The higher As content in the husk, straw and root of the rice species *i.e.* Masuri, IR-64, Luchai, Safari and HMT was observed.

3.6. Speciation of As in Rice

The As species concentration in four new fast growing rice grains is summarized in **Table 7**. The main As species detected in the rice extract were monomethylarsenonate (MMA) and dimethylarsinite (DMA) and As(III). Total arsenic concentration of the four rice samples was varied from 207 - 548 µg/kg. The inorganic As(III) concentration was ranged from 46% - 66%. The higher DMA fraction was observed in the rice grain containing higher As content. Amongst them, the lowest concentration of the MMA was marked. The inorganic and organic

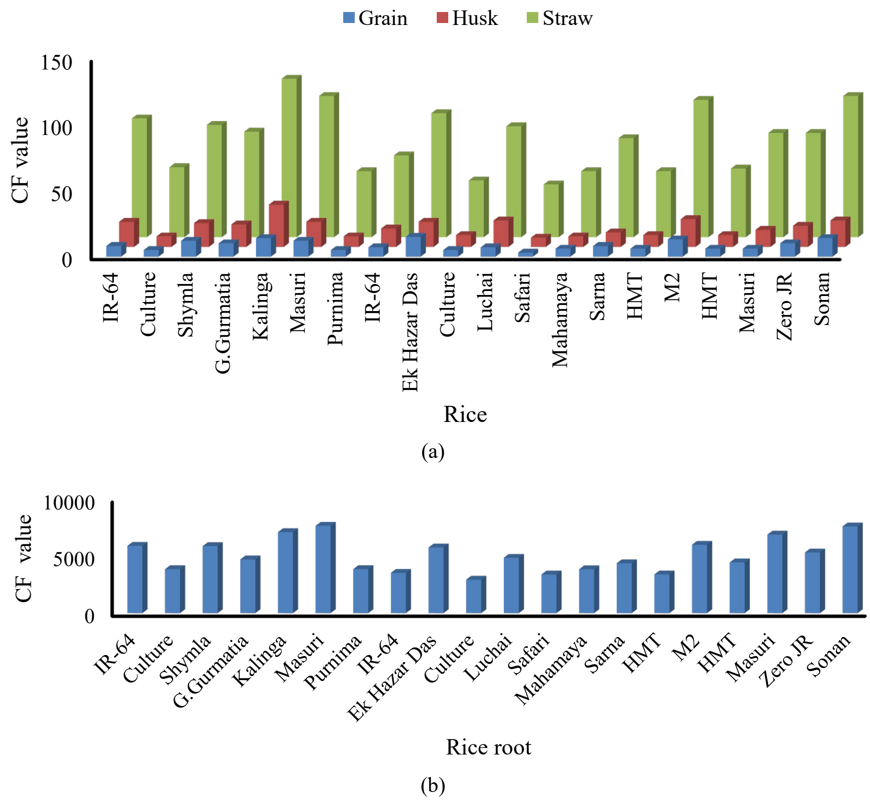

Figure 2. The FC value of rice plants in Koudikasa village.

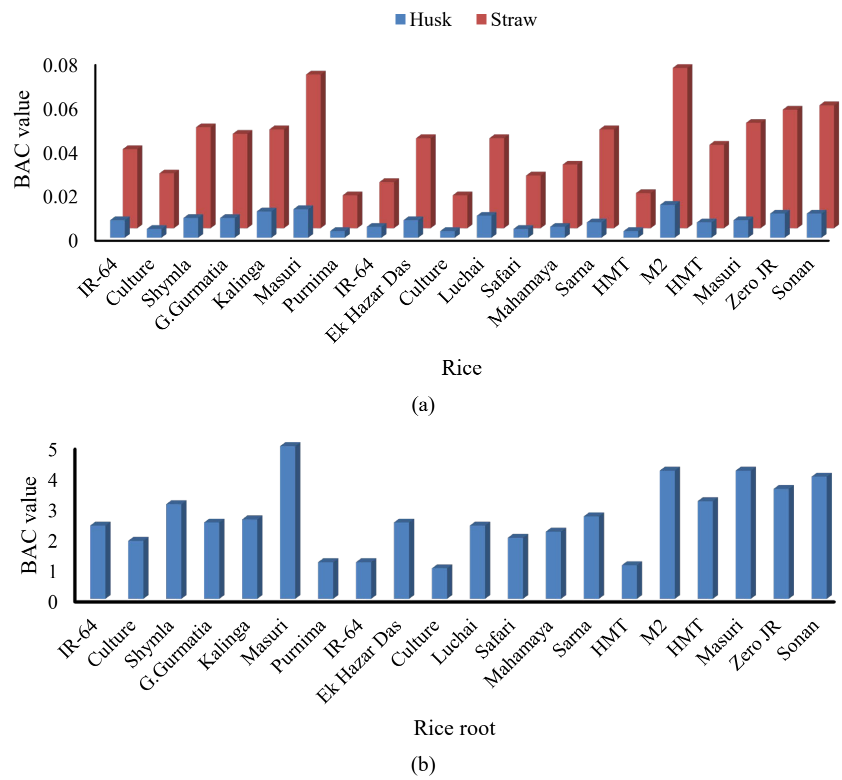

Figure 3. The BAC value of rice plants in Koudikasa village.

Table 7. Arsenic speciation in rice grain, μg/kg.

S. No.	Sample	AsT	DMA	MMA	As(III)
1	CMR rice	296 ± 21	165 ± 3	10 ± 1	97 ± 5
2	Masuri	207 ± 39	65	<3	115
3	Sonan	548 ± 14	207	16	253
4	HMT	309 ± 34	92	5	204
5	Zero JR	509 ± 29	95 ± 3	15 ± 5	270 ± 5

Certified value of As in the CMR = 290 ± 30 μg/kg.

As content was found to be fairly correlated with the AsT content in the grain. The distribution pattern of As-species in the rice grain of this region is observed similar to Bangladeshi rice [24].

4. Conclusion

The field water and soil were found to be contaminated with As at dangerous levels, may be due to geogenic origins. The high yield rice variety *i.e.* IR-64, Kalinga, Ek Hazar Das, M2, Zero JR, etc. was found to be more sensitive to the As accumulation. Several folds higher As contamination of the straw and root of the rice plants than the grain was marked. The feeding of straw to the domestic animals seems to be a potential pathway entry of As. The rice root was marked as hyper phytoextractants for accumulating As from the surface soil.

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