Arsenic distribution in soil aggregate fractions of two different cropping fields in Nepal

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Arsengehalte in Aggregatfraktionen zweier unterschiedlicher Ackerböden in Nepal

1 Introduction

Arsenic (As) in soil can pose a risk to plant health and the food chain. Its origin can be natural (Robertson, 1989) or deriving from anthropogenic activities (FOLKES et al., 2001, ROYCHOWDHURY et al., 2005). The bioavailability of arsenic depends on its speciation and on soil conditions, e.g. soil texture, soil structure, type of oxides, types of clay minerals, organic matter content and pH (OTTE et al., 1991; BECH et al., 1997; BALASOIU et al., 2001). Iron oxides which are known to be involved in arsenic adsorption in soils (WAYCHUNAS et al., 1993), mainly present in the clay fraction. Moreover, other soil components, such as carbonate minerals and manganese oxides can also contribute to As retention (MAHIMAIRAJA et al., 2005).

Several studies on metal distribution in soil aggregates

Zusammenfassung

Das Grundwasser im Nawalparasi-Distrikt in Nepal ist stark mit Arsen kontaminiert, das durch Bewässerung in die Böden gelangt. Aggregatfraktionen sind wesentliche Bodenbestandteile, die verschiedene Elemente enthalten, welche von Pflanzen aufgenommen werden. Daher wurden die Arsengehalte unterschiedlicher Aggregatfraktionen und ihre Korrelation mit Fe, Mn, Al und Ca in zwei unterschiedlich genutzten landwirtschaftlichen Ackerböden (Reis- und Gemüseanbau) untersucht. Die untersuchte Bodengesamtfraktion war < 2 mm. Innerhalb dieser Fraktion wurden 4 unterschiedliche Aggregatgrößen durch Ultraschallbehandlung und Siebung aufgetrennt. Das meiste Arsen kommt in den überwiegend vorhandenen kleinen Aggregaten vor. Hingegen waren die Arsengehalte in den großen Aggregaten am höchsten. Der Elementgehalt jeder Aggregatfraktion wurde nach Mikrowellenaufschluss gemessen. Beim Boden unter Reisanbau konnte keine deutliche Korrelation zwischen Arsen einerseits und Fe, Mn, Al und Ca andererseits gefunden werden. Beim Boden mit Gemüseanbau zeigte Arsen eine klare Korrelation mit Fe, Mn und Ca in den kleinen Aggregatfraktionen (≤ 0,63 mm). Die Arsenverteilung in Böden wird daher durch die Verteilung unterschiedlicher Aggregatfraktionen beeinflusst.

Schlüsselworte: Arsen, Bodenaggregatfraktionen, Reisanbau, Gemüseanbau.

Summary

The groundwater of Nawalparasi District of Nepal is severely contaminated with arsenic which can add in to soils during irrigation. The aggregate fractions are a major part of soil which holds various elements which were uptaken by plants. Therefore, arsenic contents in different aggregate fractions and their correlation with Fe, Mn, Al and Ca were studied in two agricultural cropping fields (rice and vegetable). The investigated soil particle size fraction was < 2 mm. Within this fraction, 4 different aggregate sizes were separated by sonification and sieving. Most amount of arsenic was found in the more abundant smaller aggregates; however, arsenic concentrations were highest in larger aggregates. The element content of each aggregate fraction was measured after microwave digestion. Under rice no distinct correlation of arsenic with Fe, Mn, Al and Ca was found. Under vegetables As showed a significant correlation with Fe, Mn and Ca in the smaller aggregate fractions (≤ 0.63 mm). Hence, the arsenic distribution in soil is affected by the distribution of soil aggregate fractions.

Key words: Arsenic, soil aggregate fractions, rice field, vegetable field.

were conducted by WILCKE and AMELUNG (1996), WILCKE and KAUPENJOHANN (1997) and WILCKE et al. (1998). Most plant roots grow on aggregate surfaces. Therefore, plant roots mainly depend on the chemical conditions of the aggregate surfaces rather than on the average properties of the bulk soil (WHITELEY and DEXTER, 1983). This implies that for the assessment of pollutants in relation to soil functioning, the distribution of elements and their reactions at the aggregate level is important. Many studies have been carried out focussing on the arsenic contamination of bulk soils, As in soil solution and in the rhizosphere, but the arsenic distribution in different aggregate size fractions has not been investigated yet. Therefore, this publication focuses on the arsenic distribution and its correlation with further elements contained in different aggregate fractions under different cropping conditions.

2 Materials and methods

2.1 Study site and sample collection

Soils were sampled from two agricultural cropping areas: rice fields (flooded) and vegetable fields (non-flooded) of the Nawalparasi district, Nepal (Figure 1).

These soils have been used for the cultivation of rice, maize, wheat and several varieties of vegetables for more than 30 years.

Surface soil samples (0–10 cm) were collected from 8 sites having different arsenic contents, ranging from 6 to 12.5 mg kg^{-1} (mean ≈ 8 mg kg^{-1}, n = 8) and 6.4 to 16.7 mg kg^{-1} (mean ≈ 9 mg kg^{-1}, n = 8) of rice and vegetable fields respectively. Five replicate samples were collected from each site. A first collection of samples was carried out in the last week of August 2005 (rice field). A second collection took place in the last week of February 2006 (vegetable field). The samples were air dried, manually ground in a mortar, sieved through a 2 mm sieve and packed in individual polyethylene bags. The soils were classified as Calcic Fluvisols, according to the World Reference Base for Soil Resources (FAO, 2006). The general soil properties are shown in Table 1.

2.2 Separation of aggregate size fractions

The Ö NORM-L-1072 method was followed for separation of soil aggregate fractions. The soil particle size fractions < 2 mm were obtained by sieving. 50 g of soil with 200 ml of de-ionized water were put in a plastic cup and dispersed with a Bandelin Sonoplus 2200 ultrasound equipped with a cylindrical probe VS 70 T with a diameter of 12.7 mm. The sonification took place for 1 minute at 20 kHz; the vibration amplitude was 25 μm; the energy value was 7.8 J.ml-1 and the insertion depth 10 mm. To minimize the potential of re-aggregation of the dispersed fractions, the suspension was washed through a stainless
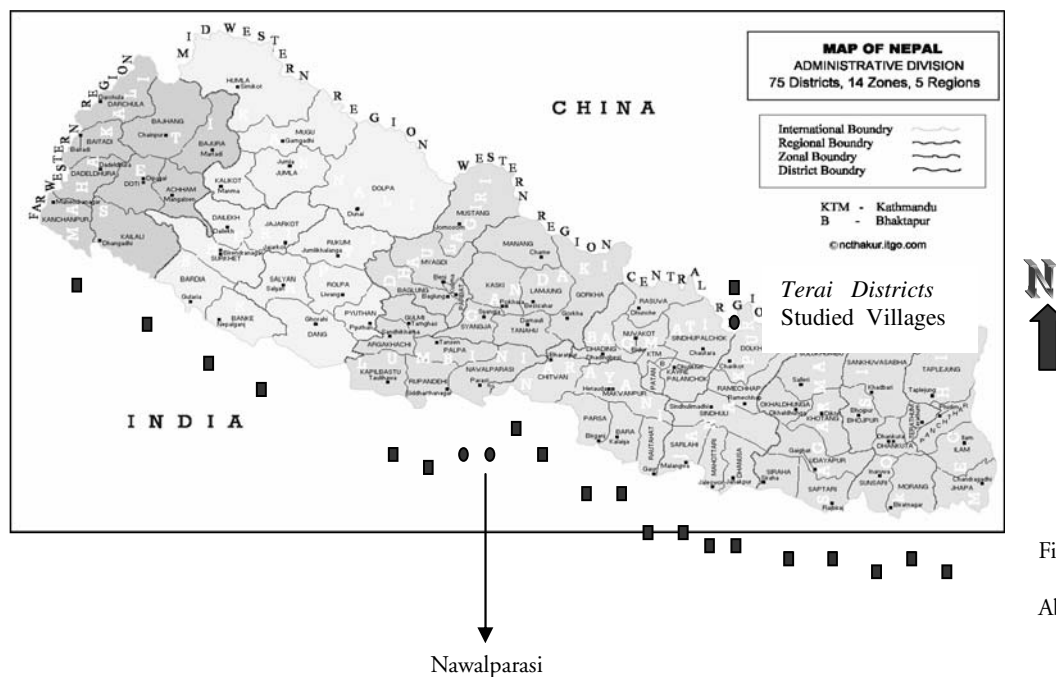

Figure 1:   Map of Nepal showing study area

Abbildung 1: Karte von Nepal mit Untersuchungsgebiet

Table 1:   General soil characteristics
Tabelle 1: Allgemeine Bodeneigenschaften

| Field | n | pH H$_2$O (1:2.5) | Texture % | | | Aggregate fraction% | | | | Eh (mV) | Total As mg kg-1 (mean values) | T OC% | CEC (cmolc kg-1) |
			Clay	Silt	Sand	< 0.063 (mm)	0.063 –0.25 (mm)	0.25 –0.63 (mm)	0.63 –2 (mm)				
Rice	8	8.4±0.2	7.5	37	55.9	68	30	1	1	80	8	0.9	8.6
Vegetable	8	8.4±0.2	8.0	36	58.9	64	32	2	2	200	9	1.0	9.6

Eh = Redoxpotential, TOC = total Organic Carbon, CEC = Cation Exchange Capacity

steel sieve for 2 min. Size limits for the aggregate fractions were < 0.063 mm, 0.063–0.250 mm, 0.250–0.630 mm and 0.630–2.0 mm, which were collected separately. The aggregates were denoted RF1, RF2, RF3, and RF4 (rice field) and VF1, VF2, VF3 and VF4 (vegetable field) for aggregate sizes: < 0.063 mm, 0.063–0.250 mm, 0.250–0.630 mm and 0.630–2.0 mm, respectively. After oven drying at 105 °C for 48 hours, the aggregate fractions were kept in a desiccator for cooling. Then the weight of each fraction was measured and expressed in percent (Table 1).

2.3 Chemical analysis and quality assurance

2.3.1 Measurement of pH and Redoxpotential (Eh)

Soil pH was measured in water at a ratio of one part of soil to 2.5 parts of water by weight. A soil solution was prepared with water and kept it for 2 hours for equilibration after shaking it. The equilibrated soil solution was shaken once again and measured the pH with a pH meter. Before the measurement of pH of soil solution sample, the pH meter was calibrated with the buffer solution of pH 4 and 7. The redoxpotential (Eh) was measured in the same soil solution by using Ag/AgCl electrode. Three replicates were run for each sample.

2.3.2 Determination of texture

The determination of soil texture was done by Pipette method which determines the physical proportions of three sizes of primary soil particles as determined by their settling rates in an aqueous solution using a pipette. Ten grams of air dried soil sample was kept in a plastic vessel and 50 ml of 0.5 % Sodium Hexametaphosphate (HMP) solution as a dispersing agent. The solution was placed on reciprocating horizontal shaker for 16 hours. Then, the solution was transferred to a measuring cylinder of 1000 ml, and added distilled water to make volume 1000 ml and allowed to settle. The temperature of the solution was 22±1 °C. There-

fore, 10 ml of solution was taken out after 10 min. of settled to determine the silt and clay content in soil, with a special pipette set in such way that to suck the sample from a certain depth and kept it on pre-weighed porcelain which was kept in oven until to be completely dry. The dried residue was kept in a desiccator for cooling until room temperature. A 2nd sample was taken after 2 hours to determine the clay content in soil and followed the same procedure as in 1st sample collection. The content of sand, silt and clay in soil by percentage was calculated.

2.3.3 Determination of Total Organic Carbon (TOC)

Total Organic Carbon (TOC) was determinate with a Carlo Erba System (CAN 1500), with dray flash combustion method. The pyrolysis gas was separated and determined by GC-TCD according to Austrian standard method ÖNORM L-1080-89.

2.3.4 Determination of Cation Exchange Capacity (CEC)

The CEC was worked out appropriate to Austrian standard method ÖNORM L-1086-89. 50 ml of 0.1 M BaSO$_4$ solution was added to 10 g of soil and shaken for 3 hours for the equilibration of soil solution. The solution was filter with Whatman filter paper (45 mm). The soil is basic in nature, therefore, Mg^{++}, K$^+$, Na$^+$ and Ca^{++} were measured in the filtrate. The CEC was calculated by adding the charge of those elements.

2.3.5 Determination of As, Al, Ca, Fe and Mn

The sample preparations and analytical procedures were accompanied by blanks and spiked standards. Samples were analyzed in three independent replicates. Aggregate samples of 0.5 g were weighed into teflon vessels. 5 ml of nitric acid (65 %) was added, followed by 1 ml of hydrogen peroxide (30 %) and the samples were digested in a microwave vessel. The digested samples were transferred into 100 ml volumetric flasks and filtered. The filtrate was analyzed for total

As by an Atomic Adsorption Spectrophotometer (AAS) and for total Fe and Mn by a Flame Atomic Adsorption Spectrophotometer (FAAS). Total Al and Ca were measured by ICP-AES. A standard sample of the respective elements was analyzed with every 10 samples for quality assurance.

The laboratory of the Institute of Soil Research of BOKU, Vienna, has an internal and an external quality control system for chemical soil analysis. The Eurosoil ES2a was used for total As, 4.14 mg g^{-1} and ES7 for Al, 23 271 mg kg^{-1} (± 1 257); Ca, 1995 mg kg^{-1} (± 198); Fe, 34 714 mg kg^{-1} (± 940); and Mn, 916 mg kg^{-1} (± 35.8), as an internal quality control reference (GAWLIK et al., 1999; WEISSTEINER et al., 1999). The quality of the internal reference material is controlled by external quality assessment 4 times per year. Average recoveries of the elements were > 85 % of the certified values.

3 Results

3.1 Contents of As, Al, Ca, Fe and Mn in the aggregate fractions

The weight distribution of aggregate fractions of rice field soils was 68 %, 30 %, 1 % and 1 % for RF1, RF2, RF3 and RF4, respectively (Table 1). Similarly, the weight distribution of aggregates of the vegetable field soils was 64 %, 32 %, 2 % and 2 % for VF1, VF2, VF3 and VF4, respectively (Table 1).
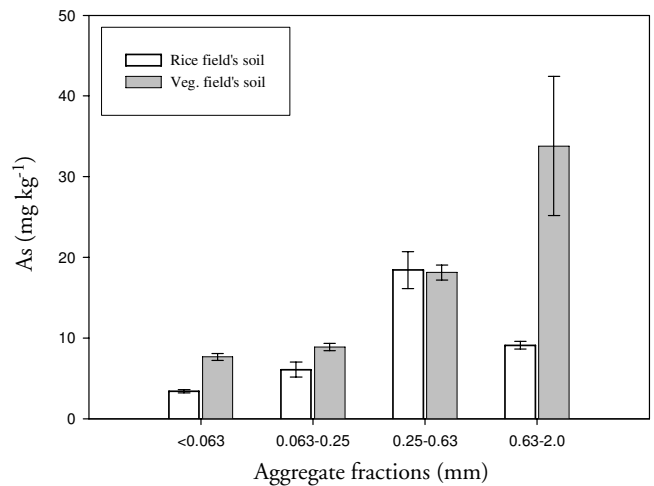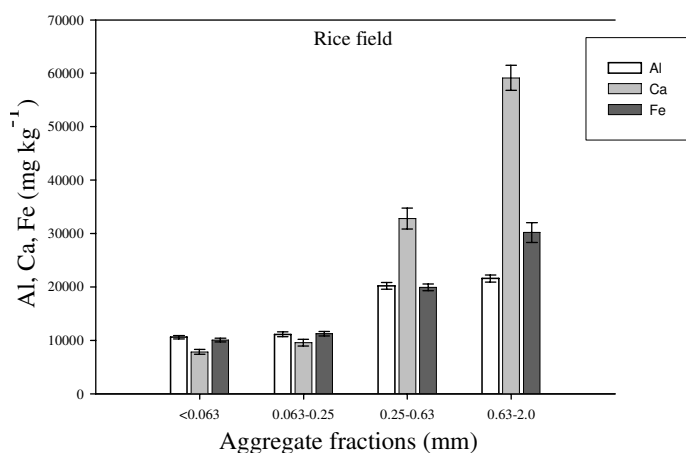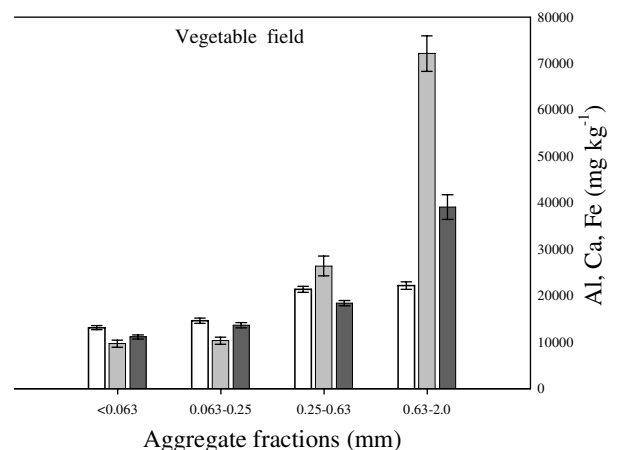

Figure 2:   Arsenic contents in different soil aggregate fractions (n = 8, mean concentration ± S.E.)

Abbildung 2: Arsengehalte in unterschiedlichen Bodenaggregatfraktionen (n = 8, mittlere Gehalte ± S.E.)

Figure 2 describes As concentration in different aggregate fractions of rice and vegetable fields. The aggregate fractions of the rice fields contained 3, 6, 18, and 9 mg As kg^{-1} in RF1, RF2, RF3 and RF4, respectively. Arsenic in the aggregate fractions of the vegetable fields was measured to be 8, 9, 18 and 33 mg As kg^{-1} for VF1, VF2, VF3 and VF4, respectively.

Results of the Fe, Al and Ca concentrations in the aggregate fractions for the two cropping fields (Figure 3a and b)

3a

3b

Figure 3:   Contents of Fe, Al and Ca (mean value ± S.E.) in different soil aggregate fraction sizes
Abbildung 3: Gehalte an Fe, Al und Ca (Mittelwerte ± S.E.) in verschiedenen Bodenaggregatfraktionsgrößen

showed that these elements were mainly contained in the largest aggregate fractions (RF4 and VF4). In the RF4 fraction, the mean concentrations were 30 203, 21 596 and 59 119 mg kg^{-1} for Fe, Al and Ca, respectively (Figure 3a). Similarly, the VF4 fraction contained 39 044, 22 161 and 72 087 mg kg^{-1} of Fe, Al and Ca, respectively (Figure 3b). The element concentration in the F1 and F2 fractions of both fields showed no significant difference ($p > 0.05$). Concentrations of Fe, Al and Ca in RF1 were 10 043, 7 846 and 10 628 mg kg^{-1}, respectively (Figure 3a), and concentrations for Fe, Al and Ca in the VF1 fraction were 11 142, 9 664 and 13 125 mg kg^{-1}, respectively.

The results show that the soils are rich in calcium, which is consistent with their pH of 8.4. Total Mn distribution for the two fields (not shown in the figure) followed the trend observed for Fe, Al and Ca. However, the contents of Mn were lower than those of Fe, Al and Ca. The mean concentrations of Mn in the RF1 and RF4 fractions were 237 and 1 952 mg kg^{-1}, respectively and in the VF1 and VF4 fractions they were 264 and 3 229 mg kg^{-1}, respectively.

Calculated correlations of As with the measured elements (Fe, Mn, Al and Ca) contained in different soil aggregate fractions, for both rice and vegetable fields, are presented in Figures 4 and 5. In the rice fields, As showed a good correlation with Fe (r = 0.665, p = 0.05) and Mn (r = 0.654, p = 0.05) in the smallest aggregate fraction (Figure 4a). There was no good correlation between As and the elements contained in larger soil fractions (Figure 4b–d).
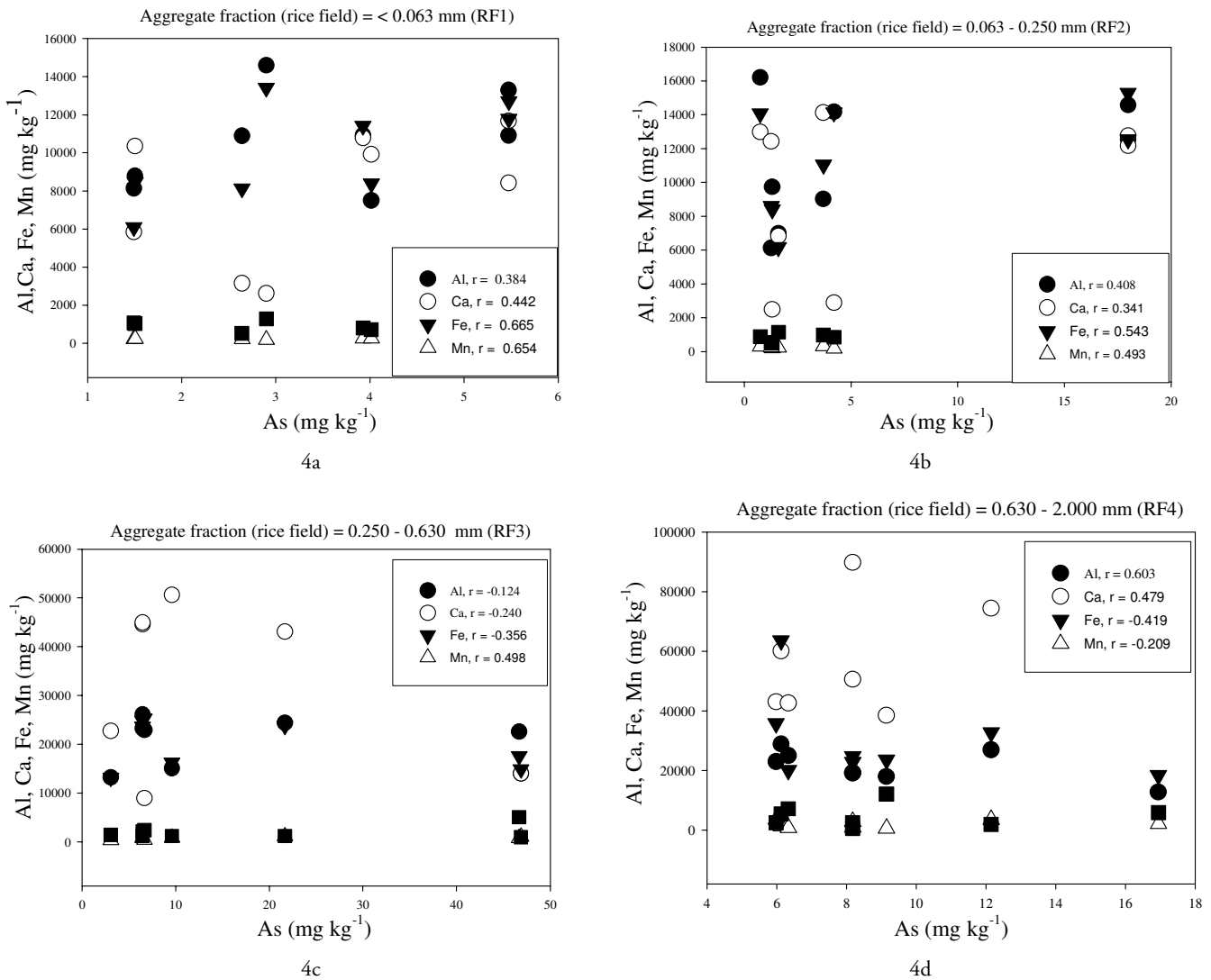

4a

4b

4c

4d

Figure 4:   Correlation between As and Fe, Mn, Al and Ca in aggregate fractions of the rice fields (Significance level, p = 0.05)
Abbildung 4: Korrelation zwischen As und Fe, Mn, Al und Ca in Bodenaggregatfraktionen der Reisanbauflächen (Signifikanz bei p = 0,05)

In the soil of the vegetable fields, the correlation of As with the elements in the smallest aggregate fraction was high for Ca (r = 0.912, *p* = 0.05), followed by Fe (r = 0.851, *p* = 0.05), Mn (r = 0.850, *p* = 0.05) and Al (r = 0.581, *p* = 0.05) (Figure 5a). The correlations of As with Fe, Mn and Ca decreased with increasing aggregate sizes (Figure 5a–d). But no such correlation was found for Al. Negative correlations of As with Fe and Mn and poor correlations with Al and Ca in the largest aggregate fraction were observed for Fe (r = –0.039, *p* = 0.05), Mn (r = –0.165, *p* = 0.05), Al (r = 0.390, *p* = 0.05) and Ca (r = 0.405, *p* = 0.05) (Figure 5d).

Although the concentrations of As, Fe, Al and Ca increased with increasing aggregate sizes (Figure 2 and 3), the percentage of these elements by weight was found higher in

smaller aggregate fractions in both rice and vegetable fields (Figure 6). The aggregate fractions of the rice fields contained 55, 43, 1, and 0.5 % of As (of the total As in bulk soil) in RF1, RF2, RF3 and RF4, respectively (Figure 6a). Arsenic percentage in the aggregate fractions of the vegetable fields was calculated as 56, 32, 5, and 8 % for VF1, VF2, VF3 and VF4, respectively (Figure 6b).

4 Discussion

Significant differences (*p* < 0.05) in the content of arsenic were also observed between soils of the rice field and those of the vegetable field, indicating that both, the kind of plant
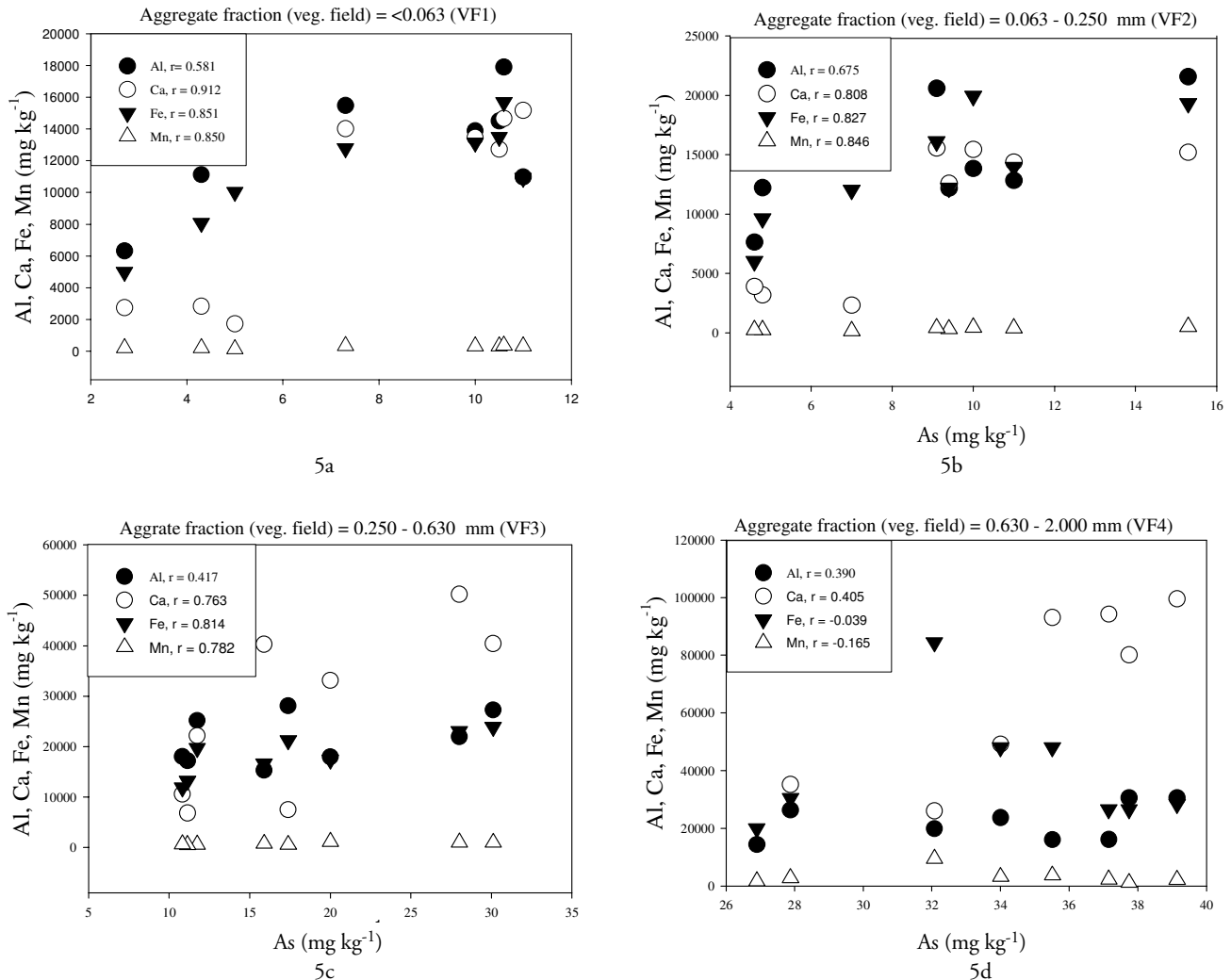

Figure 5: Correlation between As and Fe, Al, Mn and Ca in soil aggregate fractions of the vegetable fields (Significance level, p = 0.05)
Abbildung 5: Korrelation zwischen As und Fe, Mn, Al und Ca in Bodenaggregatfraktionen der Gemüseanbauflächen (Signifikanz bei p = 0,05)
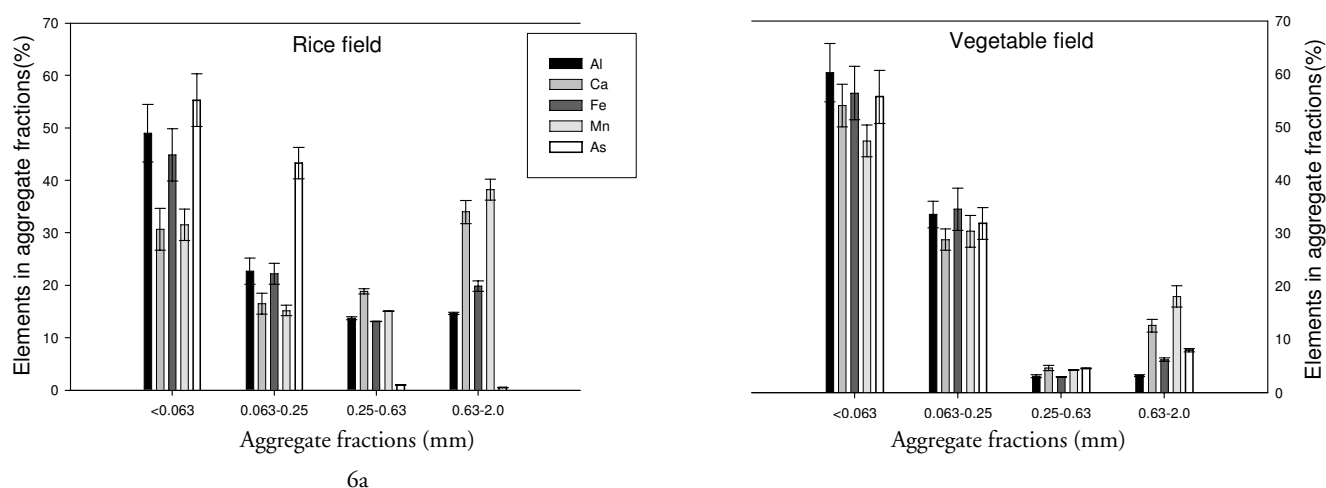
6a

Figure 6:   Distribution of elements in aggregate fractions in % by weight
Abbildung 6: Verteilung von Elementen in den Bodenaggregatfraktionen in Gewichts-%

cropping and the aggregate distribution clearly influence the arsenic content in soil. This may be also due to of different Redoxpotential in the soils collected from the fields (Table 1). The rice fields are in the flood plain, and the mean value of soil redoxpotential was measured with 80 mV which is comparatively lower than that of the vegetable field soil with 200 mV. The solubility and mobility of arsenic was found higher under reductive conditions (Fitz and Wenzel, 2002). TAKAHASHI et al. (2004) found that 70 % of the arsenic was present as As (III) in an experimental paddy field under flooded conditions. Under such conditions, the arsenic content is higher in soil solution than in the bulk soil. According to several past studies, arsenic sorption in soils is primarily related to the content of Fe and Al oxides and other elements e.g., Ca, Mn (FULLER et al., 1993; WILKIE and HERING, 1996; MAHIMAIRAJA et al., 2005; FAYIGA et al., 2007). But, only total As, Fe, Mn, Al and Ca in different aggregate fractions have been observed in our study. Figure 4 and 5 depicts the correlation of As with Fe, Mn, Al and Ca in different soil aggregate fractions of the rice and vegetable field where As showed a good correlation with Fe, Mn, Al and Ca content in aggregate fractions of vegetable soil having of higher redoxpotential (200 mV).

Effect on previous air-drying is in any case a stress to soil aggregates but the main process on the soil aggregates starts when a next wetting cycle starts. Swelling and shrinking caused by different water content is the driving force for break down of soil aggregates. The smaller aggregate fractions provide a large number of charge surface compare to large aggregate fractions. This may be a reason for presence of higher percentage by weight of As, Fe, Mn, Al and Ca in smaller aggregate sizes in both rice and vegetable fields (Figure 6). This implies that the elements percentage was mostly affected by the weight of soil aggregate fractions. Consequently an effect was found on the distribution amount of arsenic in different soil aggregate fractions (Table 1 and Figure 6). Arsenic showed a similar correlation with Fe and Mn in RF1, which could indicate that the elements have a similar potential for stabilizing As in rice soils under low Redoxpotential conditions. Arsenic showed the strongest correlation with Ca in the smallest fraction (VF1) than with Fe, Mn, and Al (Figure 5a) due to mostly of abundant of Ca in those fractions. Furthermore, Ca has a strong binding capacity towards anion in alkaline soils. DAHAL et al. (2007, unpublished data) found that Ca-As fraction (20 %) was more abundant followed by Fe-As (14 %), Mn-As (2 %) and Al-As (1 %) in the bulk soil of vegetable fields which were analyzed extracting with 0.25 M $H_2S_2O_4$ for As, 0.2M (NH_4) 2 $C_2O_4.H_2O$ for Fe and Al, and 0.1 mol.l-1 $NH_2OH.HCl$ for Mn.

ADRIANO (2001) also reported that in calcareous soils arsenic is preferentially sorbed by calcium minerals and less with iron and aluminum. FAYIGA et al. (2007) found that 51–70 % of the total arsenic uptake by *Pteris vittata* appeared from Ca-As among Fe-As, Al-As and Water soluble-As fractions. This means that As bound as Ca-As is more mobile than in presence of other element combinations. Therefore, agricultural plants grown on the As-contaminated calcareous soils might be affected by arsenic. The low correlation of arsenic with Fe, Mn and Al is probably due to

the fact that these elements are more concentrated in the inner part of the aggregates (CORNU et al., 2006; WILCKE et al., 1999). This indicates that the adsorption and de-sorption of arsenic in soils is influenced by those elements which accumulate preferentially on aggregate surfaces (WILCKE and KAUPENJOHANN, 1998). This also supports the hypothesis that the main source of arsenic in these soils is the arsenic contaminated irrigation water (DAHAL et al., 2007). Under higher redoxpotentials, the correlation of As with Fe Mn, and Ca decreased with increasing aggregate sizes (Figures 5a–d). The reason could be that smaller aggregate fractions have an enough external surface for arsenic adsorption.

5 Conclusions

In summary, our results indicate that the distribution of aggregate size fractions affects on the distribution of Fe, Al, Mn and Ca which consequently effects on the As dynamics in soil.

Acknowledgements

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