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RESEARCH ARTICLE

Transfer of elements from paddy soils into different parts of rice plants (*Oryza sativa* L.) and the resulting health risks for the Vietnamese population

Sự vận chuyển các nguyên tố từ đất vào các phần của cây lúa (Oryza sativa L.) và đánh giá các rủi ro sức khỏe đối với người dân Việt Nam

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The uptake of elements from paddy soils into shoot, husk, and unpolished grain of rice plants was investigated in Mekong, Huong, and Red River areas in Vietnam. The transferability of most studied soil elements into plant parts decreases in the order: shoot > husk > grain. Exceptions are Mg, S, Cd, Cu, Zn, and Mo, whose transfer drops in the order: shoot > grain > husk, the transfer of P falls in the order grain > shoot > husk. The translocation of the most health relevant elements into the different plant parts is affected by soil parameters like pH, organic matter, Fe- and Mn-phases, and clay minerals. Health risk assessment approaches for the average daily rice consumption are performed for non-cancer risk (Hazard Index - HI) including the elements As, Cd, Pb, Co, Cu, Mn, Mo, and Ni as well as for cancer risk for the elements As and Pb (Incremental Lifetime Cancer Risk - ∑ILCR). All rice studied grain samples exceed the safe HI-index of below 1. 81% of the grain samples were within the level of concern ranging between 1.4 < HI < 5, 18% varied between 5 < HI < 8.4, although their corresponding soils showed only a little pollution. Cd, As, Mn, and Pb were the most important elements causing non-cancer risks for rice-consuming people. The cancer-risk values ∑ILCR were mean 2.2×10^{-3} and are considerably higher than the safe threshold of 10^{-4} to 10^{-6}. Arsenic is the dominant factor for cancer risk. Rice-eating people living in Red River and Huong River areas face mainly health risks of exposure to As and Cd in the Mekong River area in addition to Pb.

Sự di chuyển của các nguyên tố từ đất vào các bộ phận khác nhau của cây lúa được tiến hành nghiên cứu tại cùng đồng bằng sông Mekông và sông Hồng, và tại sông Hương, và sông Hồng ở Việt Nam. Sự vận chuyển của hầu hết các nguyên tố đi vào cây lúa có xu hướng giảm dần theo thứ tự: thân > vỏ trấu > hạt. Ngoại trừ sự vận chuyển của các nguyên tố Mg, S, Cd, Cu, Zn, và Mo giảm dần theo thứ tự: thân > hạt > vỏ trấu; và nguyên tố P giảm dần từ: hạt > thân > vỏ trấu. Sự vận chuyển các nguyên tố vào các bộ phận của cây bị ảnh hưởng bởi các điều kiện của đất như pH, hàm lượng chất hữu cơ, dạng Fe và Mn, và các khoáng sét. Đánh giá các rủi ro sức khỏe của người dân khi tiêu thụ gạo hàng ngày được thể hiện thông qua các chỉ số rủi ro không ung thư (HI) của các nguyên tố As, Cd, Pb, Co, Cu, Mn, Mo, và Ni; cùng với chỉ số rủi ro ung thư của As và Pb (∑ILCR). Tất cả các mẫu gạo được phân tích vượt quá chỉ số an toàn HI < 1. 81% của các mẫu có chỉ số HI nằm trong khoảng 1.4 < HI < 5 và 18% các mẫu trong 5 < HI < 8.4, mặc dù các mẫu đất tương ứng được kiểm tra đều khônghoặc rất ít thể hiện sự ô nhiễm. Các nguyên tố Cd, As, Mn, và Pb là những tác nhân quan trọng nhất gây ra các rủi ro không ung thư cho những người tiêu thụ gạo. Rủi ro ung thư ∑ILCR có giá trị trung bình 2.2×10^{-3} và cao hơn đáng kể so với ngưỡng an toàn 10^{-4} - 10^{-6}, trong đó As là một tác nhân gây ung thư nổi bật. Những người sống ở khu vực sông Hồng và sông Hương đang đối mặt với sự phơi nhiễm As và Cd; trong khi đó người dân ở khu vực sông Mekông bị phơi nhiễm thêm Pb từ gạo.

Keywords: rice; paddy soil; element transfer; health risk; Vietnam

1. Introduction

Rice is the principal energy and protein supplying source for most of the Asian population in a daily basis. However, rice can be a prominent intake of harmful elements such as As, Cd, and Pb. Rahman and Hasegawa (2011) stated that, compared to other agricultural products, rice is the food with the highest content of As. High concentration of As in rice and drinking water is recognized as one of the main reasons for serious chronic diseases affecting millions of inhabitants in Bangladesh and West Bengal, India (Bhattacharya et al., 2010; Abedin et al., 2002; Khan et al., 2009). Due to the uncomplicated uptake of Cd into rice plants, rice consumption can also cause illnesses (Rizwan et al. 2016). A 50% of rice samples, collected in Cd-polluted paddy soils in Tak Province, Thailand, exceeded

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the concentration of 0.4 mg Cd kg^{-1} and up to 90% surpassed the permissible threshold of 0.2 mg Cd kg^{-1} (Sriprachote et al. 2012; Simmons et al. 2005). Lead may also have a potential health risk in different rice-producing areas as recognized by Norton et al. (2014), Shraim (2017), and Fakhri et al. (2018). Chronic exposure to these elements causes deleterious health effects resulting in harmful impacts on heart, bones, skin, kidney, neurological disorders, and different cancer types (EFSA 2009, 2010; Smith et al. 2006; Kumarathilaka et al. 2018; Jaishankar et al. 2014).

Soils enriched with potentially toxic elements are often assumed to cause higher accumulation in plants. The growth and yield of rice plants can be negatively affected when exposed to high concentrations of trace metals (Marquez et al. 2018). Element concentrations in paddy soils are determined by their parent material, but also by anthropogenic contamination like mining and industrial operations, air pollution, agricultural practices (fertilizer and pesticides), and/or using contaminated wastewater for irrigation. However, the accumulation of an element in a plant depends on the plant species or cultivars, its concentration in soil phases, type of bonding and its concentration in interstitial solution. According to Xiao et al. (2017), the soil bioavailable concentration of Cd, Cr, and Ni and microbial activities have a major impact on their accumulation in rice grains. Some other external factors such as plant density, temperature, and light may also affect the element uptake but their effects are intricate and only poorly understood (Greger 2004). Interaction with Zn, Fe, Se, Si, and liming, limits the uptake and translocation of Cd from root to shoot (Rizwan et al. 2016; Wan et al. 2018). According to Greger (2004), the fluid-transporting process from root to other plant parts is fostered by some factors: transpiration of water, root pressure, cation exchange at cell walls of the xylem vessel, formation of complexes with amino acid (for Cu), with histidine and peptide (for Ni), and chelates with organic acids (for Zn).

Vietnam is one of the world's largest rice producers (FAO 2018). The biggest granaries of the country are the Mekong River Delta in the south and the Red River Delta in the north where abundant water boosts irrigation. In addition, annual flooding events deliver fertile suspended material that settles in the riverine areas, including rice fields. As a result, parent material of paddy soils is mainly alluvial sediment, rich in organic matter. Some of the paddy soils in Vietnam are polluted by heavy metal(loid)s such as As, Cd, Cr, Cu, Pb, and Zn from industrial and mining activities leading to the contamination of rice grains (Huong et al. 2008; Phuong et al. 2010; Ha 2011; Vinh et al. 2012). However, most of the As contamination in paddy soils and rice grains comes from natural sources, strongly influenced by redox processes as described by Seyfferth et al. (2014) and Nguyen et al. (2020a, b). Around 11% of the Mekong River soils and 92% of Red River soils exceeded the Vietnamese As limit of 15 mg kg^{-1} for agricultural soils. Different soil conditions of these areas may be a reason for toxic element enrichments within the grains.

Similar investigations on the transfer of a broad variety of toxic elements into different rice plant parts are widely lacking in the literature. Understanding the influence of soil factors on element translocation, to and within the rice plant, is crucial to mitigate their uptake and protect human health. For Vietnam, as an important rice producing, consuming and exporting country, there are some information on the daily risk exposure to toxic metal(loid) intake by eating rice (Nguyen et al. 2020a, b). However, a corresponding chronic risk assessment for the population is still missing.

The goals of this research are: 1) to evaluate the translocation of a wide range of elements from paddy soils into the rice plant parts of under the influence of soil parameters such as pH, organic material, Al-, Fe-, and Mn-oxides/hydroxides; 2) to characterize the element concentration in the differing parts of rice plants (shoot, husk, grains); 3) to get information on long-term cancer and non-cancer health risks from rice consumption in Vietnam.

2. Materials and methods

2.1. Sampling, digestion, analysis and quality control

Rice plants and their corresponding soil samples were collected along three river systems in Vietnam including the Red River Delta in the north (19 sites), the Huong River in the center (4 sites), and the Mekong River Delta in the south (78 sites). The sample locations are shown in Fig. 1 (coordinates of the sampling sites are listed in Table S2 in the supplementary material). All samples were taken within 10 days before harvesting time. The 23 rice plant samples of the Red River and Huong River areas were separated into shoot (stalk and leaves combined), husk, and unpolished rice grain. Roots and stubbles were left in the fields. The 78 rice samples of the Mekong River area were split into husks and grains. The soil samples were taken within the root zone (a depth of 10 cm). The analysis process was executed in the laboratory of the Geoscience Center of Göttingen University, Germany.

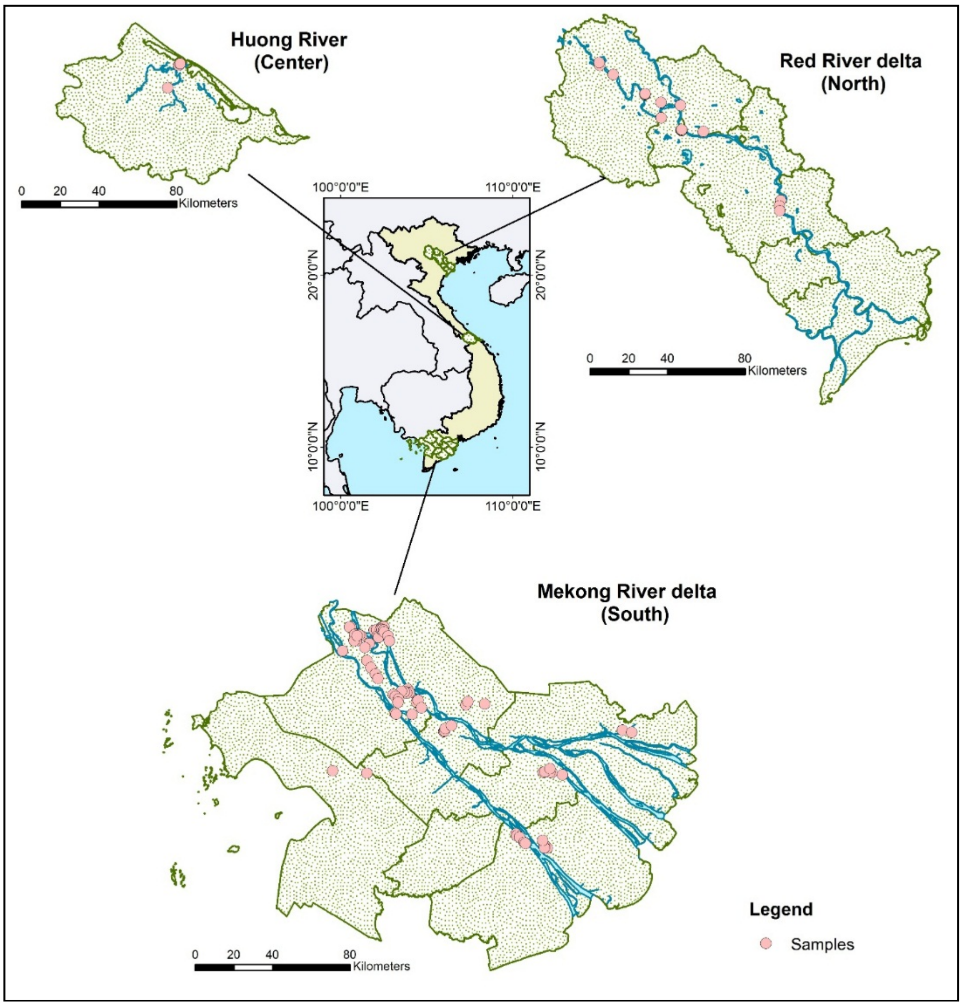

Figure 1. Sample locations in the three investigated river areas in Vietnam

The plant samples were dried at 60 ^{0}C and the soil samples at 105 ^{0}C. All samples were pulverized into grain sizes <63 µm by a Fritsch© agate ball mill before analysis. Soils' pH-values were determined in a 1:2.5 (w/v) ratio of air-dried unground soils to 0.01 M $CaCl_2$ solution by using the glass electrode ProfiLine pH/mV-Meter 197.

The Loss on Ignition (LOI), representative for organic matter and structural water in the soil samples, was determined as the percentage weight loss after heating the samples to 530°C for 24 hours. The milled plant and soil samples were completely digested in a mixture of ultrapure concentrated acids HNO_3 (65%), HF (40%) and $HClO_4$ (72%) in closed ultra clean PTFE vessels (PicoTrace®, Göttingen, Acid Sample Digestion System DAS 30). For the soil samples, about 1 ml of 37% HCl was added in the last step of the soil digestion procedure. The clear digestion solutions were then measured by ICP-OES (Inductively Coupled Plasma - Optical Emission Spectrometry) Agilent 5100 VDV and by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) Thermo Scientific iCAP Q to get the total element concentrations.

2.2. Exposure and long-term health risk calculations for rice consumption

The chronic risk exposure from rice consumption affecting human health can be evaluated on the basis of indices of Lifetime Cancer Risk and of Lifetime Non-cancer Risk (Järup, 2003; Mulware, 2013). These indices are based on the Chronic Daily Intake (CDI) in mg per kg body weight per day (USEPA 1989) which can be determined as follows:

$$CDI = (CF \times IR \times EF \times ED) / (BW \times AT)$$

where *CF* is the harmful element concentration in rice (mg kg^{-1}); *IR* is the average daily rice consumption of 0.398 kg day^{-1} for Vietnamese adults (Nguyen et al. 2020); *EF* is the exposure frequency (365 days year^{-1}); *ED* is the exposure duration (70 years); *BW* is the Vietnamese average body weight (b.w.) of 52 kg for an adult (Nguyen et al. 2020); *AT* is the average period of exposure days to hazardous element intake.

2.2.1. Chronic non-cancer risks

The chronic non-cancer risk approach is used to evaluate non-carcinogenic health effect of harmful elements from different sources. For rice, As, Cd, Pb, Mn, Co, Ni, Cu, and Mo are considered as the most critical and potentially harmful elements causing adverse health effects. The

Target Hazard Quotient (*THQ*) describes the exposure to an element and can be reckoned as follows:

$$THQ = CDI / RfD$$

RfD is the chronic Reference Dose in mg kg^{-1} b.w. day^{-1} of a harmful element and represents the maximum permissible element amount taken up from all sources (food, water, air etc.). The *RfD* values of elements for this paper are taken from previous research (Nguyen et al. 2020a, b).

The Chronic Hazard Index (*HI*) for non-cancer factors is the sum of the *THQ* for each relevant element and represents the total non-carcinogenic hazard attributable to exposure:

$$HI = \sum_{n=1}^{\infty} THQ$$

At HI ≥ 1, potential health effects should be concerned even if the exposure for every single element is below its RfD (USEPA 1989). Nordberg et al. (2015a) noticed that the HI-approach is simple but limited in its scope because it may either under- or over-estimate the risk from multiple chemical exposures.

2.2.2. Chronic cancer risks

Incremental Lifetime Cancer Risk (*ILCR*) is an index to estimate the incremental probability of an individual cancer progression over a lifetime (USEPA 1989). The ILCR of a harmful substance is computed as follows:

$$ILCR = CDI \times SF$$

where *SF* is the Slope Factor. It represents an upper estimate of increased cancer risk from a lifetime exposure to a carcinogenic substance by ingestion, inhalation or dermal contact in $[mg / (kg\ b.w.\ day)]^{-1}$ (USEPA 1989). The three elements As, Cd, and Pb are considered as key carcinogenic risk factors for low dose element intakes. However, there is insufficient information about the slope factor of oral Cd intake, therefore, Cd was excluded. In this study, the ILCR of As and Pb are estimated for eating unpolished rice with slope factors SF_{As} = 1.5 and SF_{Pb} = 0.0085 $[mg / (kg\ b.w.\ day)]^{-1}$ (OEHHA 2011).

Cumulative cancer risk (∑*ILCR*) is the sum of single lifetime cancer risks, which are restricted to the carcinogens As and Pb:

$$\sum ILCR = ILCR_{As} + ILCR_{Pb}$$

USEPA (1989), proposed a healthy safe level where ∑ILCR is below 10^{-6}. The acceptable suggested level falls in the range from 10^{-6} to 10^{-4}.

3. Results and discussion

3.1. Element distribution in parts of rice plants

After being taken up, primarily by the root surface, ions are translocated by the xylem sap to the different plant parts. During the transport, many elements are enriched at cell walls (Greger 2004; Meharg and Zhao 2012). In general, the element transferability to plant parts depends on element species, plant genotypes/cultivars, and external factors. The translocation of elements in the plant takes place by the phloem and/or xylem sap. Essential elements fulfil different biological functions such as osmoregulation. The water and nutrient mass flow within the plant is driven by stomatal aperture, energy transfer, membrane permeability and electrochemical potentials. Furthermore, some of the elements serve as cell wall and membrane stabilizers and are necessary constituents of amino and nucleic acids, proteins, enzymes, coenzymes, and chlorophyll (Marschner 2012).

The elements Ba, Na, Ca, Mn, Pb, Co, As, K, Cd, Fe, Ni, Mg, Zn, and S are more concentrated in shoot than in unpolished grains (Sh/Gr 84 to 2). Meanwhile, the Cu and Mo concentrations in shoot are approximate to those in unpolished grain. Especially, the P concentration is enriched threefold in unpolished grains in comparison to shoot. The concentration of most elements (except Ni and P) in shoot is higher than that in husk, especially for Na, Cd, Mg, and K. Most elements show higher concentrations in husk compared to those in grain, except for Cu, Cd, S, Zn, P, Mo, and Mg. Preferential transfer of some elements to grains may be explained by ion charges or the formation of organic complexes (Marschner 2012). The negative ions phosphate, molybdate and sulfate are repelled by the negatively charged cell walls allowing the more distant transport to the grains. Elements such as Cu, Zn, Mg, Ni and presumably some other metals may be transported within the plant as soluble organic complexes.

Following a general trend, most element concentrations decrease in the order: shoot > husk > grain; Cd, Mg, Zn, S, Cu, and Mo concentrations in the order: shoot > grain > husk, and P concentration in the order: grain > shoot > husk. Meng et al. (2018) found comparable results for the Cd distribution in rice plant parts.

On average, the masses portions of the plant parts to the whole aboveground plants are 0.42 of shoot, 0.46 of grain, and 0.12 of husk (Table 1). The relative mass portions of elements in different parts are shown in Fig. 2. A large load of most elements is stored in the shoot, holding more than 50% of the total element uptake. In contrast, 50% - 80% of the P, Mo, Ni, and Cu uptake is accumulated in the grain. The storage of elements in husk is below 10% except for Pb, Bi, and Sb.

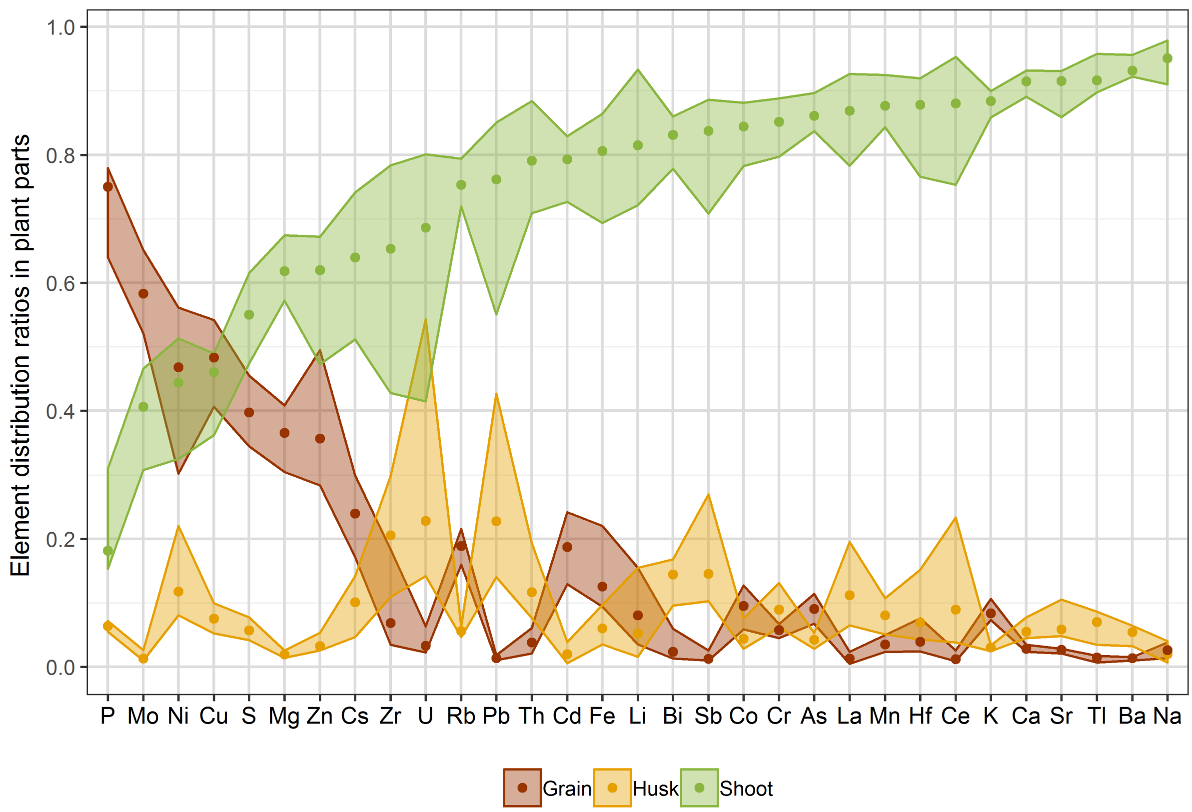

Figure 2. Mass portions of elements in plant parts in relation to the aboveground rice plant for 23 samples in northern and central Vietnam. The sum of the portions of each element in grain, husk and shoot equals 1. Dots represent median values; the colored areas cover the range of the 1st to 3rd quartile portion for each element.

There is a great difference among mean concentrations of potentially harmful elements in soils and related rice plant parts among other regions in Asia as compiled in Table 2. Arsenic concentrations in Japanese and Vietnamese soils are similar, but two times lower in soils from India, Malaysia, Thailand, and China. However, husk and grain samples from India and Malaysia contain 2 - 3 times more As. This can be explained by the very high As concentration, up to 700 µg L^{-1}, in the irrigation water (Biswas et al. 2014). Surprisingly, the As concentrations in shoots in India are about 3 times lower than those in China and Vietnam. The Cd concentrations in soils and grains in China, Japan, and Vietnam are remarkably higher than those in other countries. Concentrations of other potentially harmful elements are approximate among rice grains in China, Japan, and Vietnam. Rice grains in India and Malaysia have high contents of As, Cr and Pb. Korea shows heavy metal concentrations in soils remarkably low compared to China, Japan, and Vietnam. Nearly all elements within the different areas show decreasing concentrations in the order soil > shoot > husk > grain.

Table 1. Average physiological concentrations of element in soils and rice plant parts collected in Red River (RR), Huong River (HR) and Mekong River (MR) (mg kg^{-1}, except for LOI in wt. % and pH). Ratio represents the mass ratio of shoot, husk, or unpolished grain to the whole aboveground plant

Element	Soil (n = 101)				Shoot (n = 23)				Husk (n = 101)				Grain (n = 101)			
	RR	HR	MR	Mean	RR	HR	MR	Mean	RR	HR	MR	Mean	RR	HR	MR	Mean
Ratio	-	-	-	-	0.41	0.47	-	0.42	0.12	0.11	0.12	0.12	0.47	0.43	0.46	0.46
pH	6.2	4.4	5.0	5.2	-	-	-	-	-	-	-	-	-	-	-	-
LOI	6.5	6.4	10.3	9.3	-	-	-	-	-	-	-	-	-	-	-	-
Al	75850	74854	87130	84522	<4	<4	-	<4	<4	<4	<4	<6	<4	<4	<4	<4
Ca	5689	2180	3725	4033	5219	3915	-	4992	1099	1388	749	840	134	139	87	98
Fe	44221	34523	34773	36541	70	163	-	86	23.8	28.6	20.0	21	10.8	12.9	10.3	10.5
K	20081	19786	19680	19760	35711	22842	-	33473	3878	5184	4010	4032	2897	2945	2595	2666
Mg	8142	5772	6460	6750	2671	3013	-	2731	311	415	324	325	1384	1240	1290	1305
Mn	526	254	304	344	696	848	-	723	256	252	177	195	22	24	21	21
Na	5295	2991	4587	4657	346	541	-	380	20.1	8.6	52.5	44.7	4.2	5.5	7.6	6.9
P	865	487	780	785	1139	1493	-	1200	1308	1104	1411	1380	3545	3235	3269	3319
S	651	516	930	861	1635	2352	-	1760	690	554	625	634	1089	1051	880	926
Ti	4854	4856	4808	4819	0	0	-	0	0	0	0	0.0	0	0	0	0
As	22.5	13.6	12.6	14.8	2.7	3.2	-	2.82	0.41	0.78	0.30	0.34	0.21	0.27	0.18	0.19
Ba	417	460	394	402	93	125	-	99	19.2	21.5	7.1	10.0	1.24	1.51	0.51	0.69
Bi	0.88	0.56	0.40	0.51	0.010	0.008	-	0.009	0.0058	0.0035	0.0006	0.0017	<0.0004	0.0017	<0.0004	<0.0004
Cd	0.37	0.25	0.27	0.29	0.44	0.45	-	0.44	0.103	0.155	0.028	0.047	0.120	0.085	0.037	0.055
Ce	90	83	81	83	0.123	0.202	-	0.137	0.0714	0.0537	0.0230	0.0333	<0.0009	0.0058	<0.0007	<0.001
Co	15.7	13.3	13.2	13.8	0.17	0.79	-	0.28	0.063	0.126	0.045	0.052	0.016	0.083	0.025	0.026
Cr	69	39	94	86	0.97	0.71	-	0.92	0.38	0.39	0.17	0.22	<0.1	<0.1	<0.1	<0.24
Cu	48	27	30	34	3.52	3.51	-	3.52	2.23	2.21	1.99	2.04	3.35	3.58	3.26	3.29
Mo	0.98	1.07	0.94	0.95	0.52	0.71	-	0.55	0.10	0.06	0.17	0.16	0.64	0.69	0.41	0.47
Ni	39.9	28.5	36.3	36.4	0.37	0.85	-	0.45	0.58	0.44	0.42	0.45	0.36	0.95	0.30	0.34
Pb	50.5	29.7	28.6	33.5	0.73	0.13	-	0.63	0.90	0.74	0.44	0.54	<0.02	<0.02	0.17	0.17
Sb	2.01	1.58	2.06	2.03	0.033	0.008	-	0.029	0.0234	0.0095	0.0028	0.0070	<0.0006	<0.0006	<0.0006	<0.0007
Sn	4.81	4.56	4.15	4.32	<1.16	<0.06	-	0.94	<0.69	<0.06	<0.32	<0.38	<0.06	<0.06	<0.16	<0.14
U	3.74	4.53	4.45	4.30	0.0096	0.0042	-	0.0087	0.0062	0.0032	0.0025	0.0032	<0.0001	0.0006	<0.0001	<0.0001
Zn	110	83	90	94	40.1	62.6	-	44	11.5	9.5	15.9	14.8	23.1	26.9	19.6	20.5

Table 2. Mean concentrations of selected elements in soils and rice plant parts (mg kg^{-1}) in Vietnam compared to other Asian countries

	Country	Area	n	As	Cd	Cr	Cu	Mn	Ni	Pb	Zn
Soil	India	West Bengal	188	7.3	-	-	-	-	-	-	-
	Malaysia	Whole country	16	8.0	0.07	27	9	-	12	28	28
	Thailand	Whole country	108	6.4	0.04	25	12	-	13	20	24
	China	Yangtze River Delta	137	7.3	0.36	73	41	-	-	32	117
	Korea	Whole country	82	4.4	0.25	-	13	-	14	21	54
	Japan	Whole country	111	*	0.45	-	20	-	-	-	96
	Vietnam	This work	101	14.4	0.29	87	33	343	36	33	93
Shoot	India	West Bengal	248	0.91	-	-	-	-	-	-	-
	China	Yangtze River Delta	137	3.51	0.34	0.83	26	-	-	1.97	63.8
	Vietnam	This work	101	2.82	0.44	0.92	3.5	723	0.45	0.63	44.0
Husk	India	West Bengal	248	0.74	-	-	-	-	-	-	-
	Vietnam	This work	101	0.34	0.05	0.22	2.0	195	0.45	0.54	14.8
Grain	India	West Bengal	248/5	0.28	-	-	-	-	-	-	-
	Malaysia	Whole country	16	1.27	0.01	0.37	1.9	-	1.1	0.24	42
	Thailand	Whole country	108	<1	0.05	0.7	2	-	1.7	0.11	22.8
	China	Yangtze River Delta	137	0.13	0.06	0.19	5.2	-	-	0.10	22.8
	Korea	Whole country	82	0.15	0.02	-	4.3	-	0.35	0.11	22.6
	Japan	Whole country	111	*	0.05	-	3.3	-	-	-	15.5
	Vietnam	This work	101	0.19	0.06	<0.24	3.3	21	0.42	0.17	20.5

*India: Biswas et al. (2014); Malaysia: Zarcinas et al. (2004a); Thailand: Zarcinas et al. (2004b); China: Mao et al. (2019); Korea: Kunhikrishnan et al. (2015); Japan: Herawati et al. (2000) for Cd, Cu, and Zn (n = 111); * Kuramata et al. (2010): on soils containing 1.4 mg As kg^{-1}, the mean As concentration in 10 grain samples of rice cultivars is 0.14 mg kg^{-1}; on soils containing 8 mg As kg^{-1}, the corresponding mean As content in 10 grain samples is 2.4 mg kg^{-1}.*

3.2. Impact of soil parameter on element transfer

The influence of the soil parameters pH, LOI, Al, Fe, and Mn on the transfer of elements was evaluated and the results are displayed in Table 3. These influences can be explained by the fact that each soil parameter has a different impact on element availability. Thus, the element concentration in the plant reflects the combined effects of all soil parameters as mentioned in literature (Blume et al. 2016). Some effects of soil parameters on the transfer of elements are summarized as follows:

- The negative correlation trends of the transfer of elements Ca, Mn, P, Cd, Co, Cu, and Ni with soil pH-value may be explained by higher plant-available element concentration in soil solution at lower pH-value. In an acidic environment, H^+ ions can replace sorbed cations at the surfaces of soil phases and release them into solution. This facilitates the element uptake by the plant.

- Opposite to cations, Mo shows an increased uptake trend towards higher pH-value. Under reducing conditions, Mo is able to form complexes with organic matter, presumably with sulphur groups. Increasing soil pH-values lead to more biological destruction of organic matter, releasing Mo into the soil solution.

- The transfer of Cr and Mo are negatively correlated with soil Fe and Al. Chromium and Mo are less sorbed at low Fe and Al concentrations in soil, facilitating their plant uptake.

- The positive correlations among the transfers of As, Bi, and Sb suggest similar availability trends in the soil and comparable uptake mechanisms by the plant.

- The transfer of Mg, S, As, Bi, Cr, Mo, Ni, Sb, and Zn show negative trends with the soil Fe and Al concentrations. The sorption of these elements on Fe-oxides/hydroxides or clay minerals leads into a decrease of their bioavailable concentrations in soil solution, hence, reducing their concentrations in rice plants.

- The OM content influences positively the transfer of As. The OM may form soluble As organic complexes, facilitating the As transport into the plant. In contrast, increasing OM content leads to decreasing transfer of S, Cr, Cu, and Mo. While Sulphur is a compound of the OM, other elements may be sorbed. Under oxidizing conditions and/or at higher pH-value, organic compounds are degraded and release these elements into solution facilitating their plant uptake.

Table 3. Influences of soil parameters and soil element concentrations on transportability of elements to aboveground rice plants from the Red River and Huong River areas, listed in decreasing order of importance

Element	Soil factors	Element	Soil factors
Ca	-Ca, -pH > -Mn > +Al	Cs	-Cs
Mg	-Mg, -Fe > -Mn	Cu	-Cu > -LOI > -pH
Mn	-Mn > -pH	Mo	-Mo > -Fe, -Al, -LOI, +pH
Na	-Na, +Fe > +LOI > +Al	Ni	-Ni > -Mn, -pH, -Fe
P	-P > -pH > -Mn	Pb	-Pb
S	-LOI, -S > -Al, -Fe	Rb	-Rb > -Fe, -Al
As	-As > +LOI, -Fe,	Sb	-Sb > +pH, -Fe
Bi	-Bi > -Fe	Sr	-Sr > -Fe > -pH
Cd	-pH > -Mn	Tl	-pH, -Mn
Co	-pH, -Mn	Zn	-Fe, -Zn
Cr	-Cr, -Fe > -Al, -LOI		

+ positive correlation trends (mostly linear); - negative correlation trends (mostly exponentially decreasing)

Fe, K, Ba, Li, Sn, and U show no visible correlation with soil factors or their soil concentration.

3.3. Health risk assessment

Potentially harmful elements such as As, Cd, Pb, Co, Cu, Mn, Mo, and Ni were selected to estimate the non-cancer risk by means of Target Hazard Quotients (THQ), for single elements, and chronic cumulative Hazard Index (HI) for all selected elements (USEPA 1989; Nordberg et al. 2015a). Elements with implicit carcinogenic risk like As and Pb are evaluated by means of Incremental Lifetime Cancer Risk (ILCR) and Cumulative Cancer Risk (∑ILCR). Statistics on the exposure risk of each element and total health risks for the three river areas are compiled in Table 4. The HI and ∑ILCR for the samples of the three river areas are plotted in Fig. 3.

Table 4. Indexes for health risk assessment for harmful elements including non-cancer risk and cancer risk

Index	Area	Statistics	As	Pb	Cd	Co	Cu	Mn	Mo	Ni	HI	∑ILCR
RfD ($\times10^{-3}$)			2	1.5	1.5	0.35	200	200	40	20	-	-
CDI ($\times10^{-3}$)	Red	Min-Max	0.8-2.6	<0.2	0.02-7.4	0.03-0.24	5.5-65	88-348	1.4-9.9	0.2-7.8	-	-
		Mean	1.6	<0.2	0.92	0.12	26	170	4.9	2.8	-	-
	Huong	Min-Max	1.0-2.6	<0.2	0.33-0.96	0.48-1.02	21-36	163-202	3.7-7.0	2.9-17	-	-
		Mean	2.1	<0.2	0.63	0.63	27	181	5.3	7.3	-	-
	Mekong	Min-Max	0.6-4.3	0.02-7.1	0.06-0.88	0.06-0.88	8.4-78	101-219	0.8-7.8	0.2-23	-	-
		Mean	1.4	1.3	0.19	0.19	25	158	3.2	3.1	-	-
	Mean	Mean	1.5	1.0	0.42	0.20	25	161	3.6	3.2	-	-
THQ	Red	Min-Max	0.41-1.3	<0.1	0.05-21	0.02-0.16	0.03-0.33	0.44-1.74	0.03-0.25	0.01-039	1.4-24	-
		Mean	0.80	<0.1	2.6	0.08	0.13	0.85	0.12	0.14	4.7	-
	Huong	Min-Max	0.5-1.3	<0.1	0.93-2.7	0.32-0.68	0.11-0.18	0.81-1.01	0.09-0.18	0.15-0.86	3.7-6.1	-
		Mean	1.04	<0.1	1.86	0.42	0.14	0.91	0.13	0.37	4.9	-
	Mekong	Min-Max	0.29-2.1	0.1-4.7	0.03-4.1	0.04-0.59	0.04-0.39	0.50-1.09	0.02-0.20	0.01-1.13	1.6-8.4	-
		Mean	0.70	0.87	0.81	0.13	0.12	0.79	0.08	0.15	3.7	-
	Mean	Mean	0.73	0.69	1.19	0.13	0.13	0.81	0.09	0.16	3.9	-
ILCR ($\times10^{-4}$)	Red	Min-Max	12-39	<0.01	-	-	-	-	-	-	-	12-39
		Mean	24	<0.01	-	-	-	-	-	-	-	24
	Huong	Min-Max	15-39	<0.01	-	-	-	-	-	-	-	15-39
		Mean	31	<0.01	-	-	-	-	-	-	-	31
	Mekong	Min-Max	9-64	0.01-0.6	-	-	-	-	-	-	-	9-64
		Mean	21	0.1	-	-	-	-	-	-	-	21
	Mean	Mean	22	0.09	-	-	-	-	-	-	-	22

RfD: Reference Dose of an element represents its maximum permissible level for daily intake per kg human body weight in mg kg^{-1} b.w. day^{-1} (Nguyen et al. 2020);

CDI: Chronic Daily Intake of an element from rice consumption in mg kg^{-1} b.w. day^{-1};

THQ: Target Hazard Quotients; HI: chronic cumulative Hazard Index for non-cancer risk;

ILCR: Incremental Lifetime Cancer Risk; ∑ILCR: Incremental Cumulative Cancer Risk

3.3.1. Non-cancer risks

Chronic cumulative Hazard Indexes (HI) for the intake of the elements As, Cd, Pb, Co, Cu, Mn, Mo, and Ni from rice consumption were calculated. All samples have HI values ≥ 1.4, surpassing the safe level of 1 as suggested by USEPA (1989). A 39% of the samples show HI-values ranging between 1.4 and 3, 44% ranges between 3 and 5, and 18% ranges between 5 and 8.4. Rice consumption poses health hazards of concern with HI > 5 in 26% of the Red River samples, in 2 of 4 samples from the Huong River, and 14% of the Mekong River samples. Cadmium, As, Pb, and Mn are the most prominent harmful elements by rice consumption and contribute in 64 – 97% (average 86%) to the HI-index. In some samples, the THQs of Cd, Pb, As, and Mn are very high, reaching up to 21, 4.7, 2.1, and 1.7 respectively. The elements Ni, Cu, Co, and Mo cause a much lower risk. For lacking data, other sources for harmful element intake such as other food, drinking water or air pollution were not included in this study, but should be also considered to assess the real danger arising from these contaminants.

For arsenic, 3 of 4 samples from the Huong River, 26% of the Red River samples and 14% of the Mekong River samples have $THQ_{As} > 1$. On the average, As contributes in 22% to the HI value in the three river areas. For cadmium, 39% of the Red River and 29% and Mekong River samples show $THQ_{Cd} > 1$. Especially, samples HN10 and HN9, collected close to a brick manufactory, have THQ_{Cd} of 21 and 7 respectively. Cadmium contributes in 23% on average to the total hazard index (HI) in the three river areas. All of the Red River and Huong River grain samples have very low Pb concentrations < 0.02 mg kg^{-1} corresponding to $CDI_{Pb} < 0.2$ mg kg^{-1} b.w. day^{-1} and $THQ_{Pb} < 0.1$. Lead is responsible for less than 2% of the HI-value in these two river areas. In contrast, the Mekong River grain samples contain at least 10-times more Pb than the samples from the other river areas. Lead contributes in 3 - 69% (average 21%) to the total hazard risk in the Mekong River area. Manganese is usually not considered as a harmful element. In fact, on the average, Mn holds 24% of the HI values. Manganese surpasses THQ = 1 in 10% of the samples, but 99% of the samples have THQ > 0.5.

3.3.2. Cancer risk

The index of Incremental Lifetime Cancer Risk (ILCR) for As and Pb from rice consumption, as well as the Cumulative Cancer Risk (∑ILCR) is calculated and shown in Table 6 and Fig. 5. The ILCR depicts the probability of causing cancer, for example ILCR 10^{-4} indicates that 1 in 10,000 individuals develops cancer. All samples exceed the threshold of acceptable cancer risk which should range from 10^{-4} to 10^{-6} according to USEPA (1989). In the ∑ILCR 10^{-4} - 10^{-3}, health risk management should take action. The ∑ILCR values fluctuate from 9 x 10^{-4} to 64 x 10^{-4} (average 22 x 10^{-4}), revealing a high level of cancer risk. The mean ∑ILCR levels are 21 x 10^{-4} of the Mekong River rice, 24 x 10^{-4} of the Red River rice, and 31 x 10^{-4} of the Huong River rice. The slightly greater risk of the Huong River samples might be due to the strongly acidic condition of the soils (Nguyen et al. 2020). Of these two elements, As contributes 96% to the ∑ILCR while Pb only holds 4%. Cadmium represents an important cancer risk factor for the Red River and the Huong River rice. However, the cancer risk of Cd is mainly by inhalation while the oral intake may be overlooked.

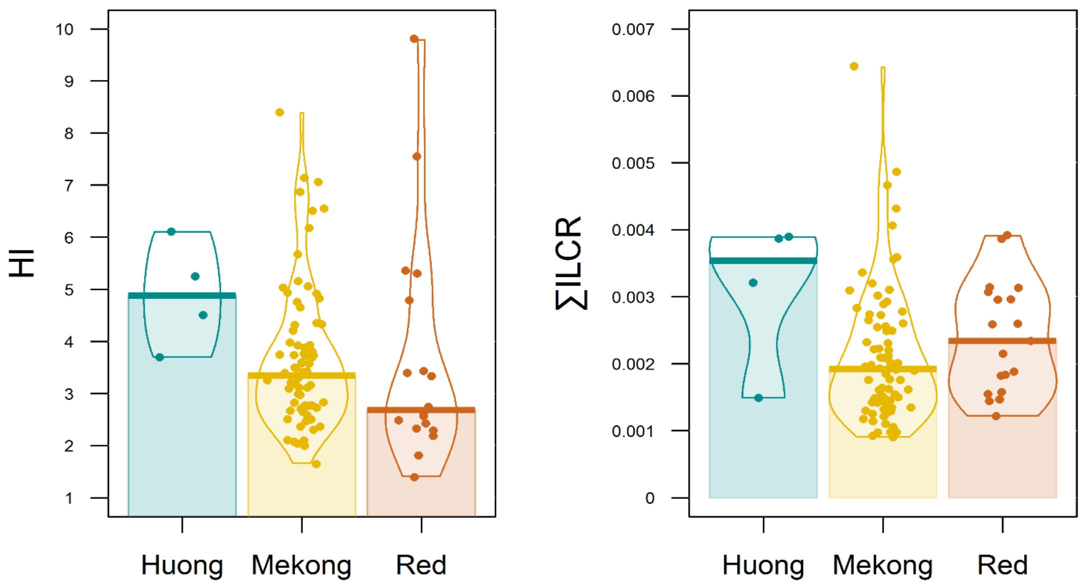

Figure 3. Target hazard index (HI) for non-cancer risk from intake of As, Cd, Pb, Co, Cu, Mn, Mo, and Ni by rice consumption and cumulative carcinogenic risk (∑ILCR) from As and Pb intake. The horizontal lines represent the medians. The extreme HI-value 24 of sample HN10 from the Red River area is not plotted.

Both, cancer risks and non-cancer risks from rice consumption, are considerably higher than the tolerable health risks levels. Our results show an urgent need to lower the uptake of potentially harmful elements into rice grains. In addition to the critical comments given above, the indexes may deliver only rough risk estimates for several reasons. The addition of single elements quotients to get the health risk seems questionable due to the lack of knowledge regarding the interaction among elements and other harmful compounds (Nordberg et al. 2015a). In addition, more sources of harmful elements such as other food sources, drinking water, air pollution among others, aggravate the situation. All calculations in this paper are done for adult lifetime exposure without considering the special stages of infancy, child, and old age. According to Liao et al. (2018), these sensible age groups may have an elevated cancer risk even with lower contaminant intake because the cancer slope factor for these groups is higher.

The system helps to get a relative risk contribution of every harmful substance and allows to compare the risk for different rice samples.

4. Conclusion

Elements are transported within the plant through charge interactions on the cell walls. As a result, concentrations of most cationic elements gradually decrease with an increasing distance from the root in the order: shoot > husk > grain. Exceptions are Cd, Mg, Zn, S, Cu, and Mo, whose concentrations decrease in the order: shoot > grain > husk. In particular, the P concentration decreases in the order grain > shoot > husk. The preferential transfer of S, Mo, and P into the grain is probably due to their anionic character and their electrostatic repulsion at the negative loaded cell walls. The easily transport of Cu, Zn and Ni into the grains may be due to the formation of soluble organic complexes in the sap.

Health risk calculations on basis of the daily intake of As, Cd, Pb, Cu, Mn, Mo, and Ni by rice consumption indicate that, all unpolished rice grains are within unsafe levels of non-cancer risk with a chronic cumulative Hazard Index (HI) ranging between 1.4 and 8.4 (with one sample even reaching 21). The risk level of HI = 1 should not be exceeded. A 18% of all samples surpass the high-risk level of HI = 5: in Huong River area in 50%, Red River area in 26%, and the Mekong River area in 14%. The elements Cd, As, Mn, and Pb are the main contributors to the HI-value contributing in 64 - 97% of the HI (average 86%). These elements should be included into any health risk study for rice consumption. Further possibilities to lower their uptake by rice grains should be explored.

The cancer risk index (∑ILCR) of As and Pb fluctuates from 9×10^{-4} to 64×10^{-4} (mean 22×10^{-4}). It is considerably higher than the acceptable cancer risk threshold between 10^{-4} and 10^{-6}. The mean ∑ ILCR values are 21×10^{-4} of Mekong River grain, 24×10^{-4} of Red River grain, and 31×10^{-4} of Huong River grain. Arsenic is the most potential carcinogenic risk factor for rice consumption in Vietnam.

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