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Evaluation of the contaminated soil and its impacts on Tobacco (Nicotiana tabacum L.) crops in Swabi, Pakistan

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Abstract

This study was conducted to find the contamination in soil and its impacts on the tobacco (Nicotiana tabacum L.) crops irrigated with industrial wastewater. For this purpose, a number of soil samples (both control and contaminated areas) and Nicotiana tabacum crop samples were collected from the Gadoon area and analyzed for major and trace elements. Results showed that the elemental contents in contaminated areas, namely Cr 65.7%, Cd 27.5%, Pb 73%, Cu 56.7%, Zn 54.3%, Co 24%, Fe 31.6%, Mn 39%, Na 49%, Ca 34% and K 42.6% were expressively higher than the control area and Chinese soil standard (CSS). While the elemental contents (mg kg-1) in Nicotiana tabacum crops, namely Cr 5.1, Cd 0.5, Pb 5.0, Ni 4.4, Cu 10.1, Mn 28.6, Na 26, and K 27. The present study depicted that, Cr, Ca, Mg may cause extreme pollution, Cu and Pb may cause moderate pollution, while Zn, Co, and Fe may cause slight pollution. In the contaminated areas, Cd, Co, Cu, Ni, Zn, K, and Na were highly accumulated, while the remaining elements were in the normal range. Furthermore, Cr may cause high ecological risks, Pb considerable, while Cd, Cu, Mg and Ca cause moderate ecological risks. The study concluded that industrial wastewater is a major threat to soil and food safety. Furthermore, it is suggested to adopt an optimum soil remediation technique for mitigating soil pollution.

Keywords: Environmental risk, Toxic trace elements, Health risk, Soil contamination, Nicotiana tabacum.

1. Introduction

Soil contamination is the foremost issue spoiled by the intensive release of toxic chemicals to the environment. The contaminated soil has negative consequences on the surrounding environment and human health (Weissmannova and Pavlovsky, 2017). The contaminants mostly comprised of hydrocarbon, persistent pollutants, organic, inorganic and toxic trace elements associated with anthropogenic/ geogenic activities (Ciesielczuk et al., 2014). Among the anthropogenic activities, the use of wastewater for irrigation further boosts the concentration of toxic trace elements in agricultural soils, which ultimately transfer to food crops and humans (Balkhair and Ashraf, 2016). Further, it's expected that the heavy accumulation/ retention of toxic trace elements (i.e., Cd, Ni, Pb, Co) in soils could also increase the soil toxicity and cause enviro-health problems.

Among the trace elements, Lead (Pb) is a toxic element and in higher concentrations (50-60 mg kg-1), it may cardiac problems, psychopathic problems and inhibits in blood production (Gowd et al., 2010; Malar et al., 2016). Cadmium (Cd) has a direct effect on enzymatic activities and causes painful Osteomalacia, kidney damages, lung, and kidney cancer (Kim et al., 2015). However, the toxic trace elements (TTE) may accumulate in the human body and can cause bronchial anemia, asthma, eczema, destruction of the thyroid gland and other genotoxic, teratogenic and carcinogenic disorders (Das et al., 2014; Hussain et al., 2018).

Besides the toxicity, the elemental contents are also essential for the human body up to some extent, but it will have some toxic effects when available in the compound/ in a higher amount. It was reported, that the high amount of trace elements are mostly available in an ionic form which can easily absorb by plants and affect the food chain and human body (Hussain et al., 2018; Rinklebe and

Shaheen, 2014). Similarly, the high intake of copper (Cu) with Pb may affect the reproductive system, thyroid gland, causing depression, anxiety, and anemia (Malar et al., 2016). The excessive intake of iron (Fe) damages the DNA chain, hepatic and cardiovascular system. Both hyper or hypo concentration of zinc (Zn) is harmful to plant growth, which reduces yield, affects photosynthesis and plants tallness (Rizwan et al., 2017). Furthermore, the regular and high ingestion of sodium (Na), potassium (K), and magnesium (Mg) and their compounds affect glucose level, liver function, hostility function and frustration (Hussain et al., 2015).

According to Kakar et al. (2020), the tobacco (Nicotiana tabacum L.) is an important and economical crop of Asian countries and is traced back to the 1600s, when locally used in cigarettes or cigars. This crop is mostly grown in Swabi areas for economic stability. Due to freshwater scarcity, peoples used industrial wastewater for irrigation purposes (Nasrullah et al., 2006; Hussain, 2014; Hussain et al., 2015). The wastewater in Gadoon areas has a high concentration of inorganic elements, which are accumulated in agriculture soil and principally transfer to plants (Hussain, 2014; Muhammad et al., 2020). Additionally, contaminated soils, toxic trace elementals, pesticides, and herbicides affect our natural environment and alter crop metabolic activities and productivity (Imfeld and Vuilleumier, 2012). Similarly, the use of industrial wastewater in Swabi (Gadoon areas) directly contaminate the agriculture land and harvested crops. Thus, the present study aimed to probe the distribution, and extent of elemental contents in soil and Nicotiana tabacum crops and its associated health risk.

2. Materials and methods

2.1. Study area and sampling

The selected area lies in the western Himalayas of northern Pakistan, stretched on the longitude 72˚32'45″ to 72˚35'45″ E and latitude 33˚5'20″ to 33˚7'20″ N (Fig. 1). The study area of Himalayas, where the rocks belong to the meta-sedimentary and igneous. Further, the mineralogical composition of igneous rock is commonly dolerite and granite. However, meta-sedimentary rocks commonly loaded with schist, quartz, and limestone (Sajid et al., 2014). The soil of the study area is derived from the Indus River and categorizes as an old alluvial plain with sandy-silty texture (Hussain et al., 2015).

Fieldwork and sampling were carried out in Gadoon Industrial and its connected agricultural zones of Swabi (Fig. 1). The representative soil i.e., 15 samples from the control area, 20 samples from contaminated areas, and 16 samples of Nicotiana tabacum were collected from each selected target and reference areas of the Gadoon industrial area and its connected area (Hussain, 2014). From extra care and accuracy, 1 kg and 0.5 kg samples of soil and plant samples (respectively) were collected from each sampling point.

The entire samples were collected randomly, where the significance was observed as reported by Dube et al. (2018) (Fig. 1). The entire samples were collected in standard grade plastic bags and transferred to the geochemistry laboratory for further processing.

2.2. Soil digestion

The representative portion of soil samples was dried in the open air and ground to 75 µm. For investigating the trace elements, a 0.5 g sample was passed through 20 ml of aqua-regia, 10 ml hydrofluoric acid, and 20 ml hydrochloric acid (2NHCl) in a sequence, while the digested contents were filtered to 50 ml plastic bottles (Hussain et al., 2014, 2015). For probing the major elements (i.e., Ca, Fe, K, Mg, Mn, and Na), 0.5 g soil samples were digested with 10 ml hydrofluoric acid, and 2ml perchloric acid at 220 °C till occurring the paste, then add 4 ml HClO4 within 2-hour interval and diluted to 50 ml plastic bottles (Chang et al., 2014). The digested samples were further analyzed through atomic absorption spectroscopy (Perkin Elmer AAS 700) for the major (i.e., Fe, Mn, Mg, Na, Ca, and K) and trace elements (i.e., Cd, Ni, Cr, Pb, Ni, Co, and Zn).

2.3. Plants digestion

The plant's samples were dried in an oven at 60 °C and then ground up to 80 mesh size for

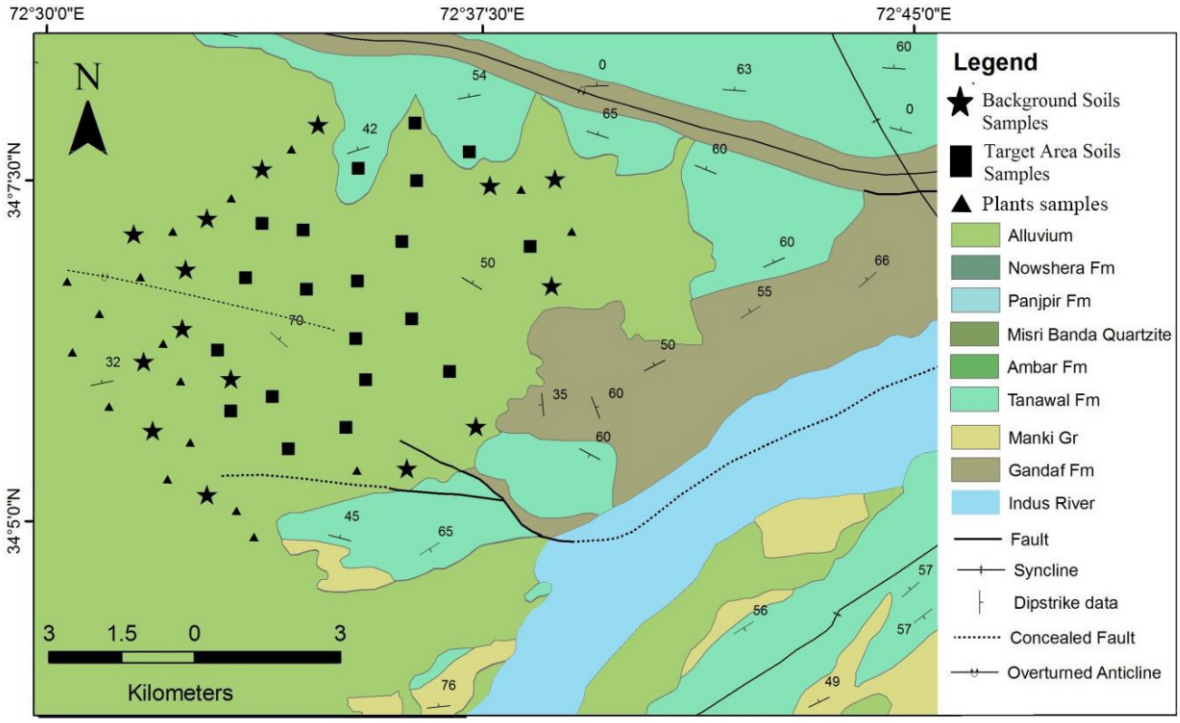

Fig. 1. Geomorphological features and sample location of the study area.

further analysis. For the acid digestion, 1.0 g Nicotiana tabacum samples were treated with 1:2:2 HF, aqua-regia and 2NHCl in a sequence. All the digested samples were diluted to 250 ml plastic bottles and analyzed for the major (Fe, Mn, Mg, Na, Ca, and K) and trace elements (Cd, Ni, Cr, Pb, Ni, Co, and Zn) through flame atomic absorption spectroscopy.

2.4. Risk assessment analysis

2.4.1. Geo-accumulation index

The geo-accumulation index (Igeo) is used to measure the level of natural and anthropogenic contaminants in the soil and was calculated through equation 1 (Eq. 1) (Khan et al., 2008).

$$Igeo = \log_2\left(\frac{C_n}{1.5 \times B_n}\right) \quad (1)$$

Where "Bn & Cn" are the elemental contents (mg kg-1) in control and contaminated areas, respectively, while 1.5 is the variation factor (Lokeshwari and Chandrappa, 2006). For assessing the areas pollution level, the following categories must be considered as; grade zero- unpolluted ($0 < \text{I-geo} \leq 1$), grade-1 slightly polluted ($1< \text{I-geo} \leq 2$), grade-2 moderate pollution ($2 < \text{I-geo} \leq 3$), grade-3 moderate polluted ($3 < \text{I-geo} \leq 4$), grade-4 severe pollution ($4< \text{I-geo} \leq 5$), and grade-5 extremely severe pollution ($\text{I-geo} > 5$) (Gupta et al., 2014).

2.4.2.Enrichment factor

Enrichment index is a geostatistical tool used to estimate the enrichment level of elements in contaminated areas as compared to control areas (Hussain et al., 2015) and expressed as:

$$EF = \frac{\left(C_m / C_n\right)}{\left(C_b / B_n\right)} \quad (2)$$

Where "Cm & Cn" represents the elements in spoiled areas, "Cn" represents the average of elements in spoiled areas. Whereas "Cb" represents elements in the control area and "Cn" represents the mean elemental level in the control area. For assessing enrichment level, the mentioned standard must be considered as: ≤1-background rank, 1 to 2 minimal enrichment, 2 to 5 moderate, 5 to 20 significant, 20 to 40 very high enrichment, >40 extreme high enrichment (Huu et al., 2010).

2.4.3. Ecological risk index

The ecological risk (Eri) is a significant strategy to assess the natural hazard/ risk to the environment (Hakanson, 1980; Zhang et al., 2007) and can be expressed as:

$$Eri = T_r \times CF \quad (3)$$

$$CF = C_n / B_n \quad (4)$$

In the above equation, "Tr" is the toxic response factor followed by Hussain et al. (2019). For assessing the ecological toxicity level, the following classes must be considered i.e., 1) Er<40 low risk, 2) 40≤Er<80 moderate risk, 3) 80≤Er<160 considerable risk, 4) 160≤Er<320 high risk and 5) Er≥320 very high risk to the ecological system (Caeiro et al., 2005; Yisa et al., 2012). However, the assessment matrix for contamination factor are; low contamination (CF<1), moderate contamination (1<CF<3), considerable contamination (3<CF<6), and high contamination (>6) (Hakanson, 1980; Farkas et al., 2007).

2.4.4. Bioaccumulation factor

Bioaccumulation factor (BCF) is used to identify and quantify the potentially transferred elements from soil to plants or from plants to other living organisms (Chang et al., 2014) and can be expressed as:

$$BCF = \frac{C_p}{C_n} \times 100 \quad (5)$$

Where "Cp" is the total concentration of elements in plants (Nicotiana tabacum plants) and "Cn" represents the concentration of elements in soils (mg kg-1), while 100 is used to represent the data in percentage.

3. Results

3.1. Trace elements in the soil

The trace elementals in the contaminated soils (contaminated with industrial wastewater) and control areas soil of Gadoon industrial and connected areas on statistical bases were summarized in Table 1, while their respective results (mg kg-1) of all the elements in the soil samples were given in supplementary tables (Table S1 and Table S2). The result shows that the elemental concentrations in contaminated areas were comparatively higher than the control areas. The elemental concentration (mg kg-1) in contaminated areas i.e., Cr (309), Cd (8.4), Pb (148.5), Ni (58.7), Cu (138.5), Zn (266), Co (32.4), Fe (1008), Mn (2084), Mg (765), Na (2500), Ca (4048), K (691.6) were higher than the control areas and Chinese soil standard level (CSS) (SEPAC, 1995) and Upper Continental Crust level (UCC) (McLennan, 2001),. The concentration of elemental contents in control areas, namely Cr 105, Cd 6.09, Pb 40.43, Ni 54.3, Cu 59.95, Zn 121.8, Co 24.6, Mn 1264, Na 1277, Ca 2667, and K 986.1 were also higher than the CSS and UCC (Table 1). The highest percent variation between the contaminated and control area was in Pb (72.77 %), while the least percent variation was in Ni 7.5 % (Table 1). The decreasing trend of percent variation was Pb > Cr > Cu > Zn > Na > K > Mn > Ca > Fe > Cd > Co > Mg > Ni. The enhanced elemental contents in the contaminated areas were due to industrial activities in the surrounding areas. The result of the current study is consistent with those reported by Karvelas et al. (2003); and Mapanda et al. (2005).

3.2. Trace elements in Nicotiana tabacum

The extent of elemental contents (mg kg-1) in Nicotiana tabacum plants i.e., Cr ranged from 2.19-9.3, Cd 0.05-1, Pb 2.2-8.9, Ni 2-7.9, Cu 7.2-14, Zn 12-21, Co 0.6-5.2, Fe 5.9-21.7, Mn 21-37, Mg 17.8-26.5, Na 12.7-40.5, Ca 25.6-89, and K 9.5-74 (Fig. 2). It is obvious, that the elemental concentration in the soil of surrounding was high, but the elemental contents in Nicotiana tabacum crops (collected from contaminated areas) were also high. The boosted elemental contents in the Nicotiana tabacum crops were due to the wastewater irrigation and contaminated soil. The long-term accumulation and retention of the element will be facing a redox reaction to convert the elements to ionic form and easily pick up by plants up to some extent. Their observe impacts on human beings will be either carcinogenic or teratogenic (Chang et al., 2014).

The study observed that the elemental contents were transferred from soil to plants. The available elemental contents in Nicotiana tabacum crops were characterized by the bioaccumulation factor (BAF) (Fig. 2). The transfer of elements (%) from soils to Nicotiana tabacum crops i.e., Cr 0.7-3, Cd 0.6-12, Pb 1.45-6, Ni 3.6-13.6, Cu 5.2-10, Zn 4.5-8, Co 1.7-16, Fe 0.6-2.2, Mn 10-18, Mg 13.5-20, Na 0.8-2.6, Ca 4.4-15 and K 13-37. However, the tendency of elemental contents is increasing (Fig. 2).

3.3. Risk assessment

3.3.1. Contamination factor

The extent of elemental contents in contaminated soil i.e., Cu, Cr, Zn, Ni, Cd, Co, Mn, Fe, and some major elements (i.e., Mg, Na and Ca) caused moderate contamination (Table 2). Lead caused considerable contamination, while K caused low contamination (Table 2). The decreasing trends of trace elements according to CF was Pb > Cr > Cu > Zn > Na > Mn > Ca > Fe > Cd > Co > Mg > Ni > K, respectively. According to the contamination factor, 10 elements out of 13 were included in the moderate contamination category, 2 elements included in considerable contamination, while one element was below the normal range (Table 2). This reveals that the elemental contamination trend in the contaminated soil is increasing from moderate to considerable contamination.

3.3.2. Geo-accumulation of trace elements

According to the geo-accumulation, Cr may cause severe pollution, Pb and Mg may cause moderate pollution, Fe causes slight pollution, while the remaining elements cause no pollution (Fig. 3). The trend of pollution in contaminated areas was Pb > Cr > Cu > Zn > Na > Mn.> Ca > Fe > Cd > Co > Mg > Ni > K. By comparing with the geo-accumulation matrix (section 2.5.1.), 7 % of elements may cause extreme pollution, 15 % may cause moderate pollution, 8% slight pollution, 31 % low pollution, while the remaining 39 % elements may cause no pollution (Fig. 3). Similarly, the high accumulation of elements in the Nicotiana tabacum crops revealed that the elemental contents might transfer from the contaminated soil that might affect the biota and human health (Sharma et al., 2007).

Table 1. The Elemental contents (mg kg-1) in contaminated and control areas soil.

Elements	**Contaminated area** *n=20*			**Control area** *n=15*			**Variation** **%**	**CSS** mg kg^{-1}	**UCC** mg kg^{-1}
	Min.	*Max.*	*Average*	*Min.*	*Max.*	*Average*			
Cr	175.0	474.3	309.2	1.80	366.2	105.9	65.74	61	83
Cd	2.50	36.50	8.40	2.90	9.45	6.09	27.52	0.097	0.10
Pb	56.3	514.6	148.5	8.75	99.87	40.43	72.77	26	17
Ni	19.8	122.2	58.73	27.87	88.00	54.30	7.54	26.9	44
Cu	13.9	498.6	138.5	11.30	142.9	59.95	56.72	22.6	25
Zn	57.3	1273	266.4	35.36	314.1	121.8	54.26	74.2	71
Co	8.90	94.70	32.35	12.85	33.65	24.60	23.95	12.7	17
Fe	684.9	1655	1008	123.5	899.5	689.9	31.56	2.94*	3.5*
Mn	945.5	5235	2084	368.3	3975	1264	39.33	0.073*	0.06*
Mg	132.5	1565	765.4	21.47	1389	596.6	22.04	0.78*	1.33*
Na	1543	4243	2500	745.2	1763	1277	48.92	1.02*	2.89*
Ca	576.9	7276	4048	275.8	5673	2667	34.11	1.54*	3.0*
K	75.00	960.0	691.6	398.4	1287	986.1	42.59	1.86*	2.8*

*- %, n - Number of samples, Min. - minimum, Max.- maximum, CSS- SEPAC, (1995), UCC - McLennan, (2001)

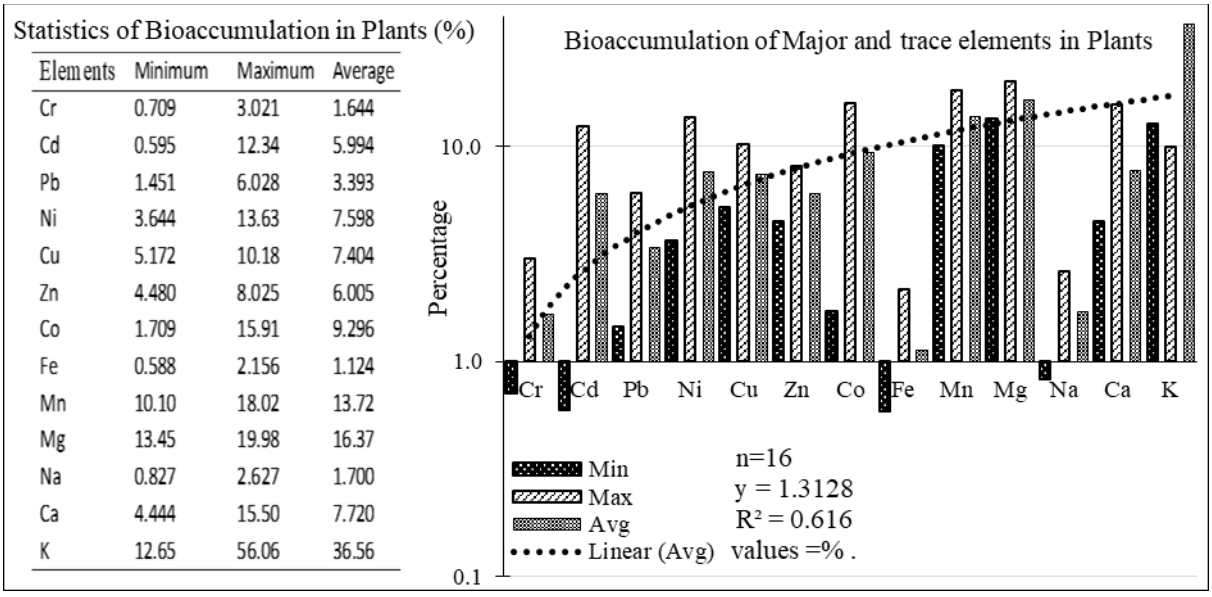

Fig. 2. Bioaccumulation of elemental contents (%) in Nicotiana tabacum crops.

Table 2. Evaluation of the soil contamination through the contamination factor.

S/N	**Parameters**	**Min** *(CF)*	**Max** *(CF)*	**Average** *(CF)*
1	Cr	97.22	1.30	2.92
2	Cd	0.86	3.86	1.38
3	Pb	6.43	5.15	3.67
4	Ni	0.71	1.39	1.08
5	Cu	1.23	3.49	2.31
6	Zn	1.62	4.05	2.19
7	Co	0.69	2.81	1.31
8	Fe	5.55	1.84	1.46
9	Mn	2.57	1.32	1.65
10	Mg	6.17	1.13	1.28
11	Na	2.07	2.41	1.96
12	Ca	2.09	1.28	1.52
13	K	0.19	0.75	0.70
Σ*CDi*		127.41	30.78	23.43

CD-degree of contamination, CF < 1=low contamination, 1<Cf<3 =moderate contamination, 3<CF<6 = considerable contamination, CF >6 high contamination

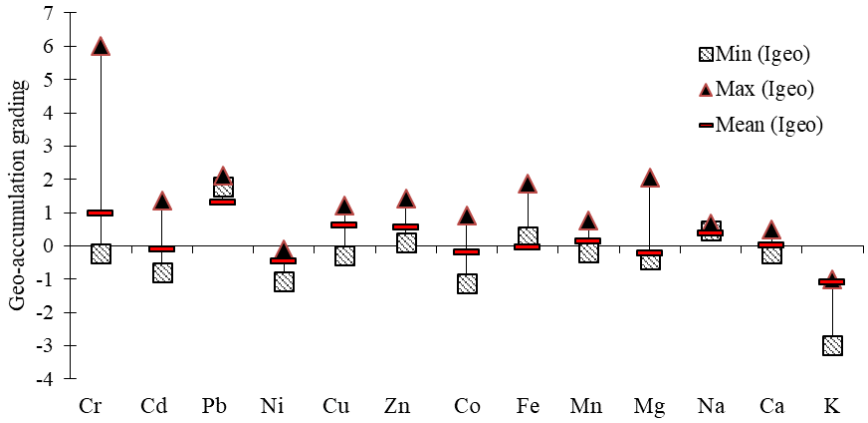

Fig. 3. Geo-accumulation and risk quantification in anthropogenic transformed soil.

3.3.3. Enrichment factor

According to enrichment factors, 46 % of elements were in background level, 15 % elements in the minimal enriched, 30 % in moderately enriched, while the remaining elements (9 %) were significantly enriched. The EF trend for trace elements was Cd > Co > Cu > Zn > Ni > Pb > Cr, while major elements were K > Na > Ca > Mn > Fe > Mg, respectively (Fig. 4). The percentile ratio of enrichment showed, that 54 % of the element were enriched and 46 % were in the normal range. This high enrichment of elemental contents in the contaminated area was due to the release of wastewater to the surrounding environment. Additionally, the elements, namely Co, Ni, Cu, Ni, Cd, K, and Na were highly enriched in the study areas, while the elements, namely Ca, Mg, Mn, Fe, Cr, and Pb were in the normal range (Fig. 4), which cause potential risk to the surrounding environment and human health. The result of elemental enrichment and methods is consistent with the result of Huu et al. (2010).

3.3.4. Ecological risk assessment

The ecological risk assessment revealed, that the element i.e., Cd may cause moderate level risk to the surrounding environment, while the other elements may cause low-level risk (Table 3). This low risk revealed that there is a native elemental concentration in both the areas (contaminated and control area), but the boosting concentration of contaminants was due to the wastewater irrigation and other industrial activities.

According to the average ecological risk assessment, 8 % of elements cause moderate ecological risk, while 92 % of elements may cause low risk. The low contamination of few elements is due to firmly bound with sediments or with water, which provides a piece of very little information in the first stage of investigation (Yang et al., 2011).

3.4. Comparison between contaminated and control areas

The dissolved elemental contents are transferred to plants through absorption and deteriorate the crop quality (Hussain et al., 2019). However, these elemental contents are strongly associated with one another as well as to the source (Abrahim et al., 2007). In the contaminated areas, a significant positive correlation was establish among the Cr-Pb (0.466), Pb-Co (0.701), Pb-Zn (0.642), Ni-Cu (0.618), Zn-Co (0.751), Fe-Mn (0.788) and Mg-Ca (0.598) (Table. 5). In contaminated areas, no significant negative correlation was found, but almost weak correlations were found among the Zn, Ca, Na, Mg, Fe, and Ni with other groups. This indicates that the elemental contents in the contaminated area were freely available rather than bind with each other. However, the anthropogenic sources are also associated with geogenic sources, which alter the content of elements creating a positive correlation. The freely available elemental contents revealed that the elements are partially or disbanded in soils and represent the anthropogenic sources (industrial wastewater) (Nobi et al., 2010; Buccolieri et al., 2006).

In the control areas, a positive correlation was observed among Cr-Pb (0.843), Cr-Ni (0.635), Cr-Fe (0.509), Pb-Ni (0.540), Pb-Fe (0.556), Ni-Co (0.999), Ni-Fe (0.610), Co-Fe (0.674), Mn-Na (0.557) and Mg-Ca (0.812) (Table 5). In the control areas, a number of non-significant positive correlations were higher than the contaminated areas. The elemental groups with more positive correlation will bond easily with each other. This also indicates that the elements are dissolved in nature rather than freely available (Nobi et al., 2010). This directed that the enrichment of elements in the control areas is geogenic instead of anthropogenic.

3.5. Distribution of elements

Geospatial distributions of elemental contents in the soil of study areas were given in Figure 5. The spatial distribution map shows a clear spatial variation among the different points with respect to elementals concentration. The interpolation of Pb distribution obviously supports the occurrence of Pb concentration in the industrial and commercial zones and decrease from the distance of industrial areas (Fig. 5a). Similarly, the concentrations of each element were found high at or nearby the industrial areas, while decreasing from the distance of the source. The geospatial

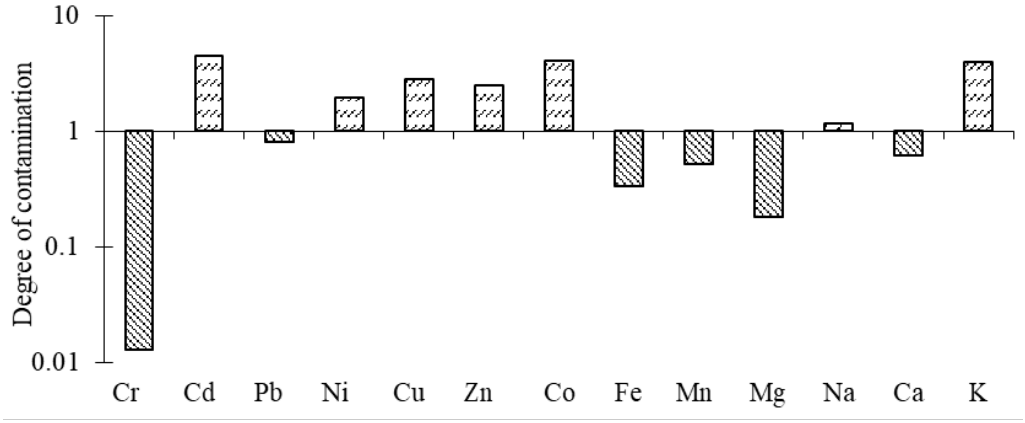

Fig. 4. The enrichment of elements in the contaminated soil of Gadoon Amazai.

Table 3. Estimation of the contamination and their risk to the ecological system.

S/N	Parameters	Min. *(Er)*	Max. *(Er)*	Average *(Er)*
1	Cr	194.44	2.591	5.838
2	Cd	25.862	115.8	41.39
3	Pb	32.171	25.76	18.36
4	Ni	3.552	6.943	5.408
5	Cu	6.150	17.45	11.55
6	Zn	1.619	4.055	2.186
7	Co	0.693	2.814	1.315
8	Fe	5.548	1.841	1.461
9	Mn	2.567	1.317	1.648
10	Mg	6.171	1.126	1.283
11	Na	2.071	2.406	1.958
12	Ca	2.091	1.282	1.518
13	K	0.188	0.746	0.701
ΣEri		283.3	184.2	94.62

Table 4. Eco-toxicity comparison and correlation through geostatistical analysis.

Elementals	Geo-accumulation index	Enrichment Factor	Ecological risk index	Contamination factor
Cr	Severely-Extreme poll.	Background level	Very high risk	High contamination
Cd	Unpolluted	High Enrichment	Considerable risk	Moderate Contamination
Pb	Moderately Poll.	Background level	Low risk	High contamination
Ni	Unpolluted	Minimal enrichment	Low risk	Moderate contamination
Cu	Unpolluted	Moderate Enrichment	Low risk	Moderate Contamination
Zn	Unpolluted	Moderate Enrichment	Low risk	Considerable Contamination
Co	Unpolluted polluted	Moderate Enrichment	Low risk	Low contamination
Fe	Slight Polluted	Background level	Low risk	Considerable contamination
Mn	Unpolluted Poll.	Background level	Low risk	Moderate contamination
Mg	Moderate polluted	Background level	Low risk	High contamination
Na	Unpolluted	Minimal enrichment	Low risk	Moderate contamination
Ca	Unpolluted	Background level	Low risk	Moderate contamination
K	Unpolluted	Moderate Enrichment	Low risk	Low contamination

Poll.=pollution

Table 5. Correlation and elemental association in contaminated and control soil of the study areas.

Control Areas		Cr	Cd	Pb	Ni	Cu	Zn	Co	Fe	Mn	Mg	Na	Ca	K		Contaminated Areas
	Cr	**1**	.466*	.356	.134	.319	.241	.415	-.378	-.238	-.033	.189	-.044	-.038	Cr	
	Cd	.366	**1**	.229	.021	.007	-.073	.058	-.156	.120	-.300	.221	-.047	.051	Cd	
	Pb	.843**	.194	**1**	.061	.034	.641**	.701**	-.313	.073	.194	.172	.029	.389	Pb	
	Ni	.635*	.464	.540*	**1**	.618**	.145	.419	-.210	-.109	.189	-.316	.248	-.058	Ni	
	Cu	-.016	-.089	.052	.079	**1**	.190	.412	-.310	-.323	-.007	-.332	.223	-.097	Cu	
	Zn	-.091	.328	.075	.192	.447	**1**	.751**	-.114	.042	.075	.016	.204	.429	Zn	
	Co	.470	.489	.424	.900**	.232	.192	**1**	-.333	-.075	.151	-.041	.091	.295	Co	
	Fe	.509	.107	.556*	.610*	.307	.116	.674**	**1**	.788**	-.334	.264	.070	-.417	Fe	
	Mn	.338	-.117	.289	-.107	.037	-.051	-.230	-.284	**1**	-.345	.292	.040	-.350	Mn	
	Mg	.089	.009	-.190	-.474	.011	-.094	-.576*	-.226	.123	**1**	-.102	.598**	.269	Mg	
	Na	.113	.086	.192	-.219	-.010	.171	-.227	-.381	.557*	.070	**1**	-.328	-.299	Na	
	Ca	-.092	-.172	-.318	-.453	-.038	.095	-.520*	-.143	.005	.812**	.097	**1**	.274	Ca	
	K	-.209	-.003	-.125	.072	-.183	.106	.200	.033	.127	-.510	.282	-.213	**1**	K	

*. Correlation is significant at the 0.05 level (2-tailed)

**. Correlation is significant at the 0.01 level (2-tailed)

distribution of Pb, Fe, Cu, and Mg supported that the contamination is decreasing far-off distance from an industrial area. The entire elements have a comparatively high distribution index in contaminated areas as compared to the control areas. The concentration of trace elements in the agricultural soil is decreasing from the source to outward (Fig. 5a-d). This proved that wastewater irrigation considerably increases the trace elements concentration in the surrounding area. The high accumulation of toxic trace elements in the soil of the connected area may influence the crops quality, and indirectly affect human health (Nobi et al., 2010; Facchinelli et al., 2001).

4. Discussions

In order to compare the relative distribution of minimum, maximum and average values on a variability basis. The R2 statistics (0.62) shows that the average variance in BAF was moderate and near to substantial (0.75). The R2 statistics have different matrix i.e., weak (0.25), moderate (0.5) and substantial (0.75) (Wong, 2013). This revealed that the elemental contents are certainly transferred from contaminated soil to plants (Fig. 2).

Similarly, based on the Pearson correlation coefficient, the highest correlation among all the elements (of Nicotiana tabacum samples) was found in-between Zn-Cr, (0.904), while the remaining elements also showing strongest correlation with different elements (Table S6).

The present study shows that the elemental contents in the contaminated area were expressively higher than the control area. This high concentration was due to anthropogenic sources as also proved through geostatistical and geospatial analysis. The accumulation of trace elements since 1986 in the surrounding environment had a great impact on agricultural soils and crops (i.e., Nicotiana tabacum crops). By observing the accumulation in soil, the concentration of the major elements is higher than the trace elements (Table I). According to elemental enrichment, the trend of elements in both the areas is the same, but the concentration of elements in the contaminated is higher than the control areas as well as UCC (McLennan, 2001) and CSS (SEPAC, 1995). The background elemental

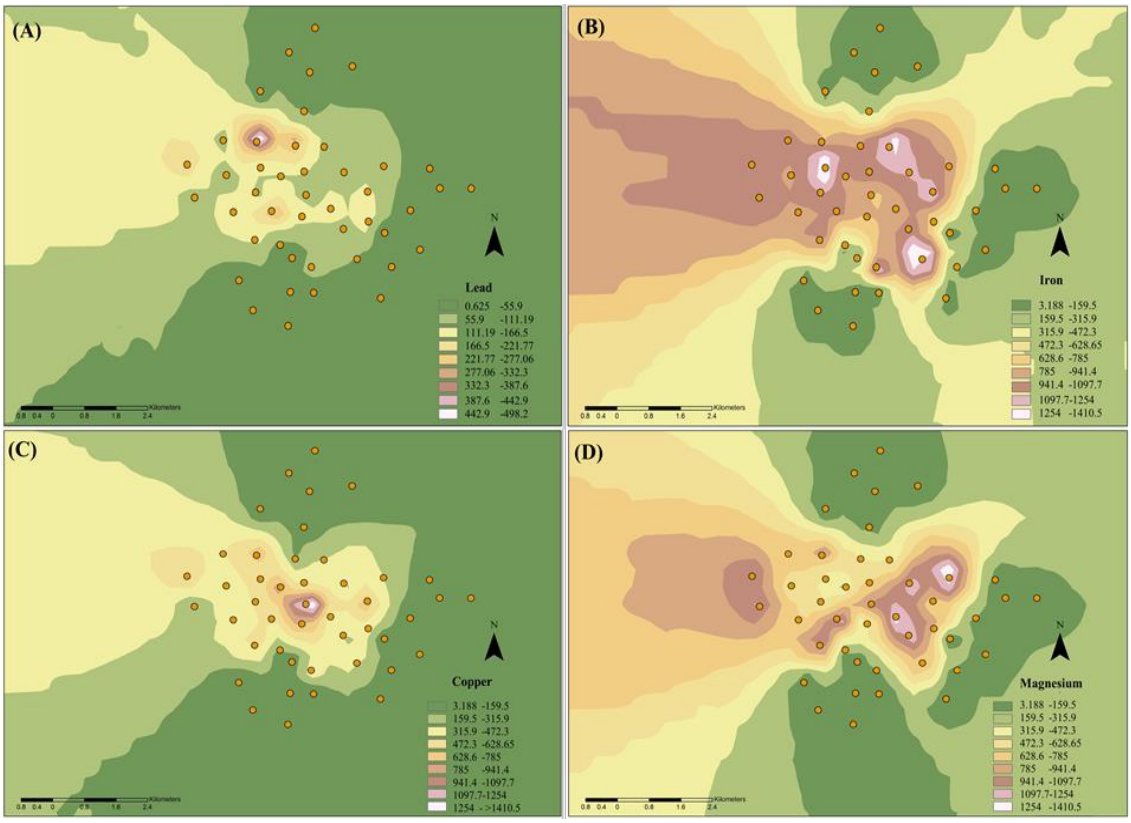

Fig. 5. Geospatial distribution and mobility of trace elements in the study areas (A-lead, B-iron, C-copper, and D-magnesium).

concentration in both contaminated and control areas revealed the geogenic occurrence of trace elements. However, the boosting concentration in contaminated areas is due to wastewater irrigation. The wastewater irrigation increasing the elemental toxicity in soil and crops (Dube et al., 2018; Li et al., 2018; Waqas et al., 2014). It was observed that the wastewater is used for irrigation on a regular basis, as there is no availability or little availability of freshwater for agricultural purposes.

The retention of elements in soil and its bioaccumulation in plants and transfer to humans received serious attention (Zhang et al., 2007). Subsequently, all the contaminated area soils have a high level of potentially toxic elements, especially Pb, Cd, Ni, and Cr elements. Some of these elements are leached to groundwater, while the major portion will be vertical transfer i.e., to the upper portion of vegetation (Burghelea et al., 2015; Galfati et al., 2011). A similar concentration was also observed in the Nicotiana tabacum plants. The decreasing trend of trace elements in Nicotiana tabacum crops was K > Mg > Mn > Co > Ca > Ni > Cu > Zn > Cd > Pb > Na > Cr > Fe, which are consistent to those reported by Hussain et al. (2019). The elemental concentrations in Nicotiana tabacum crops show dissimilarities in the trend while transferred concentration was found much higher in Nicotiana tabacum crops. The dissimilarities in trend depend on many factors, including elemental solubility, uptaking ratio and plants receiving capacity (Hussain et al., 2018). It's also clear, that each and every crop has its own receiving and bioaccumulation capacity (Ge et al., 2000).

Carcinogenic elements may occur from many sources, but the direct sources are industrial waste and mining. The carcinogenic elementals may enter to the food chains in various ways, but the most efficient source is plants and water (Ge et al., 2000). It is visible that the Nicotiana tabacum is a self-carcinogenic plant, but the hyperaccumulation of the toxic elemental in Nicotiana tabacum crops further enhance the chances of health-related problems, especially cancer (particularly the mouth and lungs), cardiovascular, chronic obstructive pulmonary diseases and psychological disorders (Talhout et al., 2011). According to WHO, 5.4 million premature deaths are attributed to Nicotiana tabacum smoking (WHO, 2011). The resting heartbeats of smokers are 2-3 times faster than non-smokers, while 2/3 of death ischemic heart disease are due to Nicotiana tabacum smoking and count about 2.5 times higher deaths in cigarette and cigar smoking in the world (Goldenberg et al., 2003; Mahmood et al., 2014). It was reported that some hazardous chemicals were found in Nicotiana tabacum products, however, the nicotine and some toxic elementals are in great concern (Dewey and Xie, 2013). In addition, the observed health problems, including heart disease, lung, and kidney problems, mentally suppressed, pediatric health problems, hypertension, and other gastric and abdominal problem are common in the local community (Hussain et al., 2015). The overall risk in the surrounding community was mostly attributed to toxic elements in the surrounding environment. Continuous ingestion and inhalation of toxic elements through different sources are responsible for the health problem in the local community (Lykkesfeldt et al., 2000).

5. Conclusions

The contaminated soils have high elemental contents as compared to the control areas, which is associated with the anthropogenic activities (wastewater irrigation). The trace elements concentration in the Nicotiana tabacum crops irrigated with wastewater was also found high. There was no significant negative correlation among the elements in contaminated areas, which indicated that the elements are freely available rather than bind with each other. It seems that the anthropogenic sources are not associated with geogenic sources, which alter the elemental contents creating a negative correlation. Geospatial distribution proved that the trace elements were decreased from the distance of the industrial zone. Cadmium, Co, Ni, Cu, K, Na, and Zn were enriched in the contaminated agricultural soil. In the contaminated areas, Cr, Ca and Mg may cause extreme level pollution; Cu and Pb may cause moderate level pollution, while Fe, Zn, and Co may cause slight pollution. Furthermore, Cr may cause high ecological risk, Pb causes considerable risk, while Cd, Cu, Mg and Ca cause moderate risks to the ecological system.

Additionally, the industrial activities and wastewater was the major source of trace elements in soil and Nicotiana tabacum crops, but the toxic elements have a greater inspiration as compared to the major elements. The intensities of toxic and carcinogenic elements in soil and Nicotiana tabacum crops were exotically higher than the control areas and may responsible for environmental and health problems in the local areas. Further, the study endorses to prohibit both Nicotiana tabacum cultivation and wastewater irrigation, while the contaminated agricultural land should be remediated in an environmentally friendly way.

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Compliance with ethical standards

The authors declared no conflict of interest in the respective research work. All the authors contributed equally.

Author's contribution

Rahib Hussain, proposed the main concept and involved in the write-up. Seema Anjum Khattak, assisted in Fieldwork, experimental design and scientific work rectification. Shehla Sattar, assisted in laboratory work. Liaqat Ali, help in fieldwork, and proofread of the manuscript. Zahid Ullah, did a technical review before submission of the manuscript. Nisar Muhammad, was involved in the preparation of figures and formatting.

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Table S1. The concentration of the major and trace elements in control areas soil (mg kg^{-1}).

Samples	Cr	Cd	Pb	Ni	Cu	Zn	Co	Fe	Mn	Mg	Na	Ca	K
Sb1	39.55	4.80	18.80	27.87	33.50	73.70	12.85	123.45	3974.78	726.99	1763.45	2954.7	1243.5
Sb2	164.9	4.8	46.8	43.0	125.0	192.6	18.55	802.9	1972.5	1389.4	1570.7	5673.3	760.0
Sb3	366.15	6.10	99.87	88.00	20.00	57.00	31.75	891.60	1858.50	537.57	1201.50	2203.6	851.0
Sb4	156.50	5.35	70.60	65.40	142.90	168.20	29.05	899.47	2117.50	187.11	1054.35	396.9	765.0
Sb5	282.60	9.45	87.85	65.30	11.30	122.00	29.40	860.15	1943.00	603.74	1541.03	2405.9	1073.5
Sb6	147.00	8.05	54.80	61.50	125.00	197.20	33.65	804.38	795.50	575.03	1380.38	2242.1	1024.0
Sb7	65.25	6.95	26.35	57.40	60.00	89.90	27.60	805.81	763.00	406.40	1343.25	1714.3	1256.0
Sb8	70.80	6.15	31.15	65.70	54.50	70.40	30.95	633.06	846.00	40.47	1325.03	275.87	1250.5
Sb9	53.10	4.30	14.80	50.90	51.00	68.50	25.70	802.95	639.00	404.75	1150.88	4022.2	1287.4
Sb10	75.05	6.15	12.05	51.30	45.00	45.90	25.75	771.49	846.00	898.43	745.21	3405.5	744.5
Sb11	1.80	6.05	34.40	68.60	65.00	314.06	29.95	734.31	835.00	21.47	1324.35	2422.0	1233.5
Sb12	50.80	5.90	8.75	51.30	52.00	55.80	21.05	623.12	755.00	916.91	1030.73	3346.7	953.5
Sb13	25.40	2.90	58.60	28.20	30.00	35.36	15.15	735.74	637.50	523.05	1239.30	2382.1	951.5
Sb14	45.95	6.65	18.50	44.00	45.00	80.65	19.05	330.47	368.27	828.63	1343.93	3026.8	398.4
Sb15	44.00	7.75	23.15	46.00	39.00	256.10	18.55	530.17	610.00	889.52	1142.10	3530.8	999.5
Min.	1.80	2.90	8.75	27.87	11.30	35.36	12.85	123.45	368.27	21.47	745.21	275.87	398.4
Max.	366.15	9.45	99.87	88.00	142.90	314.06	33.65	899.47	3974.78	1389.43	1763.45	5673.3	1287.4
average	105.92	6.09	40.43	54.30	59.95	121.82	24.60	689.94	1264.10	596.63	1277.08	2666.8	986.12

Table S2. The concentration of the major and trace elements in contaminated areas soil (mg kg^{-1}).

Samples	Cr	Cd	Pb	Ni	Cu	Zn	Co	Fe	Mn	Mg	Na	Ca	K
St1	474.30	5.45	212.9	64.20	166.7	160.7	48.45	986.7	1575.0	1105.2	2064.15	3819.9	692.25
St2	390.35	3.45	514.6	71.50	213.0	1273.5	94.70	755.0	2282.50	962.12	2644.65	3939.60	936.75
St3	398.70	36.50	342.3	48.60	13.90	205.2	36.60	908.1	3613.5	433.13	3387.15	1398.81	632.00
St4	463.20	33.50	100.7	60.80	240.0	185.5	30.45	889.5	1551.5	247.50	2481.30	5054.00	784.25
St5	301.90	5.30	80.10	45.20	130.0	140.5	19.25	1055	1303.0	389.57	1841.40	2026.50	431.50
St6	227.00	3.85	76.45	115.2	38.10	71.90	21.50	1099	2813.0	1079.3	2340.23	3665.90	605.00
St7	185.25	3.35	56.30	41.75	85.00	89.50	26.45	884.5	2071.0	467.12	2133.68	1318.10	732.75
St8	222.80	5.15	129.8	34.45	80.00	108.4	18.60	863.01	1276.5	565.13	2048.63	1828.40	679.75
St9	335.45	3.30	78.70	37.60	125.0	575.0	19.90	1000.3	1159.5	679.98	2835.00	2419.90	851.00
St10	278.90	4.60	74.80	45.50	126.5	87.35	24.95	1655.6	4325.5	265.82	4242.73	1063.30	75.00
St11	175.00	4.40	60.50	35.30	37.50	57.25	26.65	1029.6	1735.0	648.29	2284.20	2327.50	936.00
St12	269.15	2.50	69.25	19.80	67.10	61.30	8.90	1134.7	2277.0	1222.6	2957.18	6293.70	651.00
St13	267.85	4.80	72.90	44.00	80.00	324.0	20.80	1561.6	5234.9	229.35	1821.83	6783.00	580.00
St14	212.70	9.95	135.1	89.55	210.8	622.0	46.15	1333.5	2692.5	580.97	2091.83	7276.00	929.25
St15	391.95	7.45	135.6	59.70	87.50	524.0	35.50	799.37	1089.0	1331.4	2719.58	6885.90	690.50
St16	285.30	7.35	76.85	59.05	80.50	83.80	36.15	956.67	1249.0	1564.8	2619.68	6769.00	727.25
St17	318.25	6.55	80.95	122.2	498.6	144.0	43.15	684.95	1092.0	1118.7	1543.03	6503.00	456.50
St18	401.15	6.90	135.1	77.20	235.0	204.1	36.35	828.69	945.50	132.50	2783.70	576.90	693.75
St19	225.35	6.95	349.25	50.35	115.00	298.50	23.90	1030.3	2239.5	1352.8	2495.48	7098.00	960.00
St20	359.65	6.75	187.15	52.60	140.00	110.40	28.55	706.06	1146.5	930.77	2668.95	3906.00	786.75
Min.	175.00	2.50	56.30	19.80	13.90	57.25	8.90	684.95	945.50	132.50	1543.03	576.90	75.00
Max.	474.30	36.50	514.60	122.20	498.60	1273.5	94.70	1655.6	5234.90	1564.80	4242.73	7276.00	960.00
Average	309.21	8.40	148.48	58.73	138.51	266.35	32.35	1008.2	2083.60	765.35	2500.22	4047.67	691.56

Table S3. The elemental concentration of trace elements in Nicotiana tabacum crops (mg kg^{-1}).

Sample No.	Cr	Cd	Pb	Ni	Cu	Zn	Co	Fe	Mn	Mg	Na	Ca	K
P01	5.734	0.921	4.725	2.351	10.38	16.4	0.627	11.49	34.58	23.73	21.54	34.69	21.43
P02	3.21	0.361	2.947	3.158	9.174	14.7	0.9161	14.73	25.08	19.49	19.5	63.01	28.94
P03	9.342	1.037	8.946	7.958	14.1	21.3	4.848	21.73	37.48	26.47	40.53	89.43	35.27
P04	8.148	0.951	5.328	4.752	12.73	18.4	4.556	18.52	25.81	21.84	27.53	39.52	26.04
P05	2.192	0.131	2.79	7.172	8.925	12.1	0.918	6.853	29.49	23.73	18.43	27.43	31.83
P06	2.731	0.05	2.273	2.993	7.937	13.7	1.384	7.539	26.51	21.9	33.78	57.32	27.41
P07	4.67	0.421	6.947	5.338	9.117	15.48	4.163	7.511	24.19	25.73	29.48	41.44	16.07
P08	4.392	0.737	7.153	5.638	11.83	17.2	4.895	6.93	29.43	18.62	19.47	29.61	18.27
P09	2.263	0.152	2.153	2.127	7.163	11.9	0.553	5.932	33.63	17.82	15.32	60.32	12.73
P10	4.732	0.635	5.105	3.538	8.638	16.6	4.152	9.582	23.81	19.23	12.76	25.64	9.485
P11	4.289	0.173	5.492	2.984	12.73	15.8	4.548	8.529	31.96	19.59	21.83	43.29	14.87
P12	3.214	0.349	2.93	2.425	10.73	14.6	3.437	7.414	26.27	24.19	36.84	51.68	21.83
P13	6.432	0.648	4.284	3.559	11.18	17.1	4.172	15.39	28.09	21.77	33.92	34.75	74.04
P14	3.847	0.136	5.352	2.827	9.472	14.7	2.696	9.401	21.01	18.29	24.78	26.93	18.47
P15	8.372	0.427	6.28	7.605	7.904	19.1	5.147	11.83	27.35	21.83	32.84	29.72	31.32
P16	6.417	0.847	6.841	5.324	11.3	14.7	1.416	12.9	30.71	22.01	30.19	31.84	22.04
Minimum	2.192	0.05	2.153	2.127	7.163	11.9	0.553	5.932	21.01	17.82	12.76	25.64	9.485
Maximum	9.342	1.037	8.946	7.958	14.1	21.3	5.147	21.73	37.48	26.47	40.53	89.43	74.04
Average	5.084	0.503	5.036	4.435	10.25	15.94	3.001	11.33	28.55	21.69	26.22	44.54	27.42
Transfer factor of elements from soils to Tobacco crops %													
Minimum	0.709	0.595	1.451	3.644	5.172	4.48	1.709	0.588	10.1	13.45	0.827	4.444	12.65
Maximum	3.021	12.34	6.028	13.63	10.18	8.025	15.91	2.156	18.02	19.98	2.627	15.5	56.06
Average	1.644	5.994	3.393	7.598	7.404	6.005	9.296	1.124	13.72	16.37	1.7	7.72	36.56