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ORIGINAL PAPER

Assessment of the ultra-trace mercury levels in selected desert plants

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Abstract Conventional methods that assessed the mercury (Hg) levels were not only an outcome of atmospheric pollution, but also the possibility of Hg contamination from the sample collection to laboratory analyses. Our studies used the direct mercury analyzer that measured Hg rapidly and precisely at ultra-trace concentrations with detection limit of 0.0015 ng g^{-1} on six favored desert plants and their surrounding soil in Kuwait. Analysis revealed elevated Hg concentrations in *Tamarix chinensis* Lour., and *Salsola imbricate* Forssk., among the chosen desert plants, especially during summer than in winter, thus labeling the qualities of a bio-indicator to Hg pollution. The overall parts-wise analysis on the six selected plants showed the elevated mean Hg concentrations in the leaves (0.89 ng g^{-1}) followed by root (0.51 ng g^{-1}) and stem (0.39 ng g^{-1}) in the desert plants. Reasons attribute to the capability of these plant parts to absorb, accumulate, and assimilate Hg at varying concentrations. The overall mean Hg concentration was high in soil (2.24 ng g^{-1}) in comparison with the mean Hg concentrations in the desert plants (0.60 ng g^{-1}) irrespective of the two seasons. Translocation and bioaccumulation factors indicated low uptake of Hg translocation in the plant parts from the soil. Furthermore, the mean Hg concentration was found high in samples collected from Governorates (GIII) in comparison with the samples collected from other Governorates indicating the effect of pollution from various sources. The present study characterizes the selected plants as bio-indicators and also validates the impact of regional and seasonal variations to Hg pollution at ultra-trace levels in the arid ecosystem.

Keywords Bio-indicators · Flora · Soil · Metal pollution · Translocation

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Introduction

The source of mercury (Hg) in nature includes earthquakes, dust deposition, soil erosion, and volatilizations from the ocean. Rocks, sediments, water, and soils contain low levels of mercury. In the natural environment, scientists found high mercury levels in some minerals on earth and springs (USGS 2009). Metallic-Hg evaporates slowly to air exposures. The geo-accumulation index and enrichment factor indicated the dust-originated mercury from anthropogenic sources (Lu et al. 2009). Particulate deposition on many edible vegetables and desert plants in excess produced physiological, pathological damage, and reduced yield (Freer-Smith et al. 2004; USEPA 2009; Ling et al. 2010; Morales et al. 2011; Suruchi and Khanna 2011).

Bio-monitors are very effective for tracing maps of metal contamination in the urban arid environment (Baker and Brooks 1989). Leaves are efficient accumulators of air particulates (Ma et al. 2001; Chung et al. 2005; Takashi et al. 2005; Morales et al. 2011). Freer-Smith et al. (2004) reported foliar contamination was severe at roadsides and similar pollution with particulate matter occurs in urban and suburban cities of various countries. Many shrubs and trees of drought and heavy metals tolerance that grew in semi-arid and arid regions revealed deposition of more particulates than other plants and found suitable for bio-monitoring and phyto-remediation studies (Al-Farraj and

Al-Wabel 2007; Prabha and Li 2007; Zhuang et al. 2007; Kadukova et al. 2008). Freer-Smith et al. (2004) observed two probable ways of damage in plants due to Hg uptake. They are elemental or gaseous Hg that is directly toxic, damaging sensitive cell membranes or they act as metabolic regulators or plant hormones and disrupt normal patterns of growth and development. Eboh and Thomas (2005), Onweremadu et al. (2007), Rafat et al. (2009), Keane et al. (2001), Converse et al. (2010) showed metal contents with quantitative measures of soil, air, and wastewater contamination at sites where plants grew. This assessed the plant tissue growth to that of the environmental metal content. Rafia et al. (2006), Katnoria et al. (2008) showed the effect of mercury in the experimental set-up as well as from soil to the growth inhibition in various plants.

The state of Kuwait recorded around 400 species of desert plants apportioned in six Governorate areas. Among these, 15–20 species were commonly spread over six Governorate areas of Kuwait (Greenwell 2008). Over the recent years, many desert plants in Kuwait were found at the verge of extinction. Reasons attribute to the rapid industrialization, exploitation by cattle, partial, or complete trampling due to construction and camping activities, impact of oil pollution through living medium, and rooting out plants for food and fuel (KUNA 2010), besides the increasing inorganic pollutants in the living media due to anthropogenic sources and dust storm. Kuwait experiences northwest and southeast wind direction during most part of the year. The wind speed is high during summer (12–14 mph) than during winter (9–10 mph) during normal weather (Windfinder 2011). At times, the wind speed has been reported to reach 30–35 mph causing dust storms (Arab Times 2011). High wind speed causing raising dust carries organic and inorganic pollutants, especially during summer. One among the pollutants of recent interest is the uptake of mercury (Hg) in the desert plant parts (leaves, stem, and root) through air and soil. These plants were the main source of food for the grazing cattle and accumulation of Hg in these cattle posed risk to humans. The source of Hg concentrations accumulated in the various parts of desert plants was suspected as a result of wind action (especially when the prevailing wind was from the northwest with a wind speed ranging between 1.5 and 2.5 ms^{-1} that resulted in the Hg increase between 1 and <8 ng m^{-3}), fugitive dust deposition, the possibilities of absorbing residual Hg in the un-reclamation soil leached from few chlor-alkali plants, partial translocation and bioaccumulation process, besides evaporation of soil-bound mercury during warm periods (Baker and Brooks 1989; Ma et al. 2001; Liu et al. 2002; Charron and Harrison 2003; Nahida et al. 2005; Takashi et al. 2005; Al-Awadhi et al. 2008). We conducted research on air pollution during the years 2009–2011 considering the global scenario on Hg pollution, frequent dust storms, and rapid urbanization in Kuwait in addition to the Hg concentration determination at ultra-trace levels in desert plants with unparalleled precision. Our investigations determined: (a) the mercury (Hg) concentrations in desert plants and soil using direct mercury analyzer (DMA-80: compliant to EPA 7473 method) with lowest possible detection limit of 0.015 ng g^{-1} Hg in comparison with conventional methods (EPA, 245.1, 7471A) and analyzers (Hydra-IIc, Ohio Lumex) with higher detection limits ≥0.5 ng g^{-1} (Dufault et al. 2009; Kolesnikov et al. 2010), (b) the distribution and the effect of Hg levels in plants from the six Governorates of Kuwait during summer (April–September, 2009–2011) and winter seasons (October–March, 2009–2011), (c) to label selected desert plants as bio-indicators or accumulators to Hg pollution using translocation (TF) and bioaccumulation factors (BF), and (d) an overview on the impact of environmental variables to Hg distribution.

Materials and methods

Sampling sites

Among the various kinds of desert plants in Kuwait, we chose commonly distributed six desert plants that were suspected for Hg accumulation through air and soil in four areas each, from six Kuwait Governorates (GI-GVI) (Fig. 1). Each Governorate in Kuwait showed similar geographical distribution of these plants but varied with the anthropogenic sources. The features of Kuwait Governorate areas (17,820 km^2) are as follows: (1) GI (Jahra: 29°20′13.2″N, 47°39′28.8″E) situated at the North of Kuwait mainly with desert and industrial, and sparingly populated residential areas (thermal, power, desalination, and water treatment plants are cited in this area), (2) GII (Al-Asimah/Kuwait City 29°21′57.6″N, 47°58′40.8″E) is the central Kuwait zone with industrial, residential areas and significant for its business centers, (3) GIII (Hawalli 29°19′58.8″N, 48°01′44.4″E) is a commercial center with moderately populated residential areas, (4) GIV (Farwaniya 29°16′37.2″N, 47°57′32.4″E) is a densely populated residential area, (5) GV (Mubarak Al-Kabeer: 29°15′25.2″N, 48°03′25.2″E) has moderately populated residential areas and also noted for many recreational centers, and (6) GVI (Ahmedi 29°4′37.2″N, 48°05′2.4″E) is the Kuwait's southern region with oil fields and oil wells (Fig. 1).

Meteorological data

Kestrel-4200 (Nielsen-Kellerman, USA) determined the mean 24-h meteorological data for the summer (April–

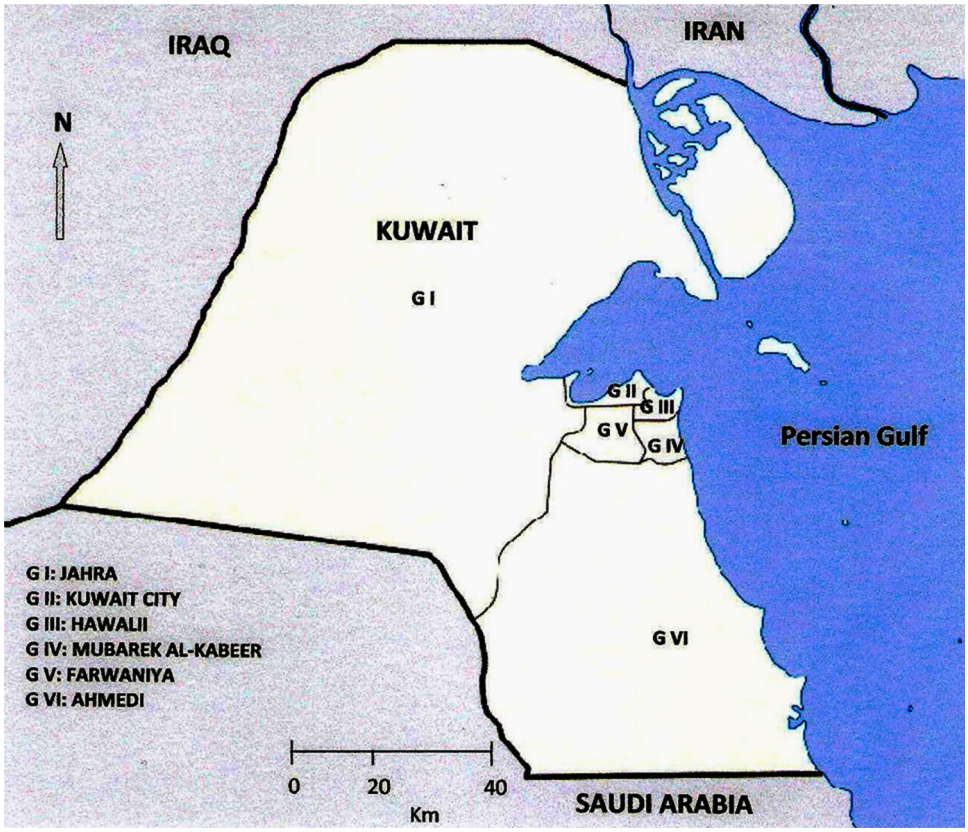

Fig. 1 Sampling sites of Kuwait Governorates

September) and winter (October–March) seasons. Meteorological data such as wind direction, wind speed, relative humidity, temperature, and dew point were recorded.

Sample collection

Desert plants (ten replicates × six species × four areas × six Governorates × twice sampling in two seasons = 5,760 Plants year^{-1}), such as *Malva parviflora* L., *Fagonia bruguieri* DC., *Salsola imbricate* Forssk., *Tamarix chinensis* Lour., *Cornulaca aucheri* Moq., and *Tribulus terrestris* L., were collected from six Kuwait Governorate areas during the years 2009–2011 (Fig. 1). Samples (17,280) were thoroughly rinsed in deionized distilled water to remove the dust and soil. They were collected in sterile polyethylene labeled (Fischer brand, USA) zipper bags (34 cm × 30 cm × 0.3 mm), stored in Thermo Cole box, and transported to the laboratory. They were stored at −4 °C before analysis. In sterilized Petri-dish (9 cm), the thawed plant parts (leaves, stem, and roots) were cut into small pieces (5 cm) using a sterile surgical blade (#11), weighed (0.2 g), and analyzed for mercury in the direct mercury analyzer (DMA-80, Milestone, Italy).

Using a soil auger, (15″L × 3–1/4″ diameter, AMS, Inc.), soil replicates (50 g) near the surrounding plants at 0.1 m depth were collected and stored in polyethylene container. Samples were oven dried (GallenKamp II, USA) at 45 °C until dryness. They were ground to obtain a homogenized powder and sieved using a 1 mm mesh, of which 0.2 g was analyzed for mercury concentrations (Keane et al. 2001).

Digestion of soil samples

Dried soil samples (0.2 g) collected from the surrounding plants were predigested with 4 ml HNO_3 (v/v, Aristar grade) overnight. HNO_3 was further added to make up the volume to 5 ml (recommended volume in the digester) and digested in a microwave digester (Ethos1, Milestone, Italy). This novel microwave digester edges over other conventional microwave digester in possessing a visually controlled monitoring feature with complete sample digestion (99 ± 1 % efficiency) in customized time, volume, and pressure validating perfection in technology. The digested soil samples were analyzed for Hg analysis in the DMA-80. Quality assurance using standard reference material (SRM-2709 San Joaquin Soil) from National Institute of Standard Technology (NIST, USA) through microwave digestion facilitated 100 % recovery of Hg from the soil samples when compared to 98.4 % ±0.3 Hg recovery of undigested soil samples by direct analysis in the DMA-80.

Determination of mercury in desert plants and soil

The DMA-80 uses three principles, namely: (a) thermal decomposition to eliminate cumbersome sample preparation, handling and the use of hazardous chemicals, (b) catalyses to remove N_2, SO_2, halogen, and interfering compounds, and (c) amalgamation to trap Hg and release Hg through a carrier gas. The Hg flow is aligned to a tri-cell-positioned optical path spectrophotometer which measures Hg by atomic absorption at 253.65 nm wave length and compliant to EPA 7473 method. Thus, the three principles of DMA-80 in combination facilitate Hg analysis: (a) in elemental or gaseous forms, (b) in samples of solid, liquid and gaseous state at greater precision, (c) with low detection limits and standard error (0.0015 ng g^{-1}–1,000 μg g^{-1} and ±0.001 ng g^{-1}) compared with detection limits (≥0.5 ng g^{-1}, ICP-MS 1–5 μg g^{-1} and ±5 μg g^{-1}) of other instruments such as HydraIIc, Ohio-lumex (Dufault et al. 2009; Kolesnikov et al. 2010). In the present study, replicates (10 numbers) of the selected six species of plant parts (leaves, stem, and roots) and the digested soil samples (0.2 g) were analyzed in the DMA (DMA-80, Milestone, Italy). Quality assurance using standard reference material (SRM-1547 Peach leaves, NIST, USA) showed 99.99 % recovery of plant samples and hence analyzed directly in the DMA-80 without microwave digestion. Analytical data were transformed to logarithmic concentrations to acquire precision results over wide-ranged samples.

Translocation (TF) and bioaccumulation factor (BF) of Hg in desert plants

Baker and Brooks (1989) revealed TF > 1 in plants with effective translocation from root to the stem. Hg translocation in the desert plants from stem to root was measured using translocation factor (TF):

$$\text{TF} = \text{Hg concentrations in the stem}/\text{Hg concentrations in roots}$$

Ma et al. (2001) classified 'hyper-accumulators,' 'accumulator,' and 'excluders or bio-monitor' to those samples which accumulated metals >10 μg g^{-1}, >1, and <1, respectively. Using this classification, Hg bio-accumulation factor (BF) in the desert plants was determined by calculating the ratio of Hg concentration in the aerial parts (stem and leaves) to that of the soil.

$$\text{BF} = C_p/C_s$$

where C_p and C_s are Hg concentrations in aerial parts of the plant and in soil (μg g^{-1}), respectively.

Quality control and assurance described by Keane et al. (2001) were carried out by using the Hg standards (ICP-grade), blanks, and SRM. Recovery of the samples to that of the standard reference material (SRM 2976) was 98.5 %, thus validating the precision of the instrument.

Results and discussion

Among the 370 desert plant species in Kuwait, in-depth studies were conducted on six desert plants, namely *M. parviflora* L., *F. bruguieri* DC., *C. aucheri* Moq., *T. terrestris* L., *S. imbricate* Forssk., and *T. chinensis* Lour., that were abundantly distributed in the six Kuwait Governorates and suspected for Hg absorption in their plant parts (leaves, stem, and root). The mean mercury (Hg) log concentrations in the desert plant parts ranged between 0.16 and 2.12 ng g^{-1} (Fig. 2). Among the six desert plants, *T. chinensis* Lour., revealed high Hg concentration that could be attributed to the absorption properties of the roots, foliage absorption from the ambient air besides, deducing the probable observations of Nahida et al. (2005), Prabha and Li (2007) and Zhuang et al. (2007) of high Hg exposure in shrub plants with more biomass (leaves) compared to the Hg levels in the plants with lower biomass.

Hg concentrations were high during summer than in winter irrespective of the different species. Frequent dust storms and wind direction causing Hg bound particulate deposition in desert plants during summer, Hg dissolution in soil from the chlor-alkali plant stationed near industrial-residential area, and evaporation of soil-bound mercury in warm periods could be related to the high Hg concentrations during summer than in winter. The above observation was in line with the findings of Liu et al. (2002), Al-A-wadhi et al. (2008), and Lu et al. (2009). In addition to the above, earlier studies (Converse et al. 2010; Morales et al. 2011) showed that an increase in photosynthetic activity could probably elevate the absorption capabilities of the available Hg by plants during summer in contrast to activities of plants in winter.

Parts-wise analysis revealed high Hg concentrations in leaves > roots > stem in *T. chinensis* Lour., > *S. imbricate* Forssk., > *T. terrestris* L., > *C. aucheri* Moq., > *M. parviflora* L., > *F. bruguieri* DC., respectively (Fig. 2). Reasons attribute to their absorption of gaseous or elemental Hg from ambient air or from soil through roots. The roots of these plants were reported to aid in translocation of Hg from soil to stem, and hence, Hg concentration was comparatively found higher than stem. This observation was in line with the findings of Rafia et al. (2006), Katnoria et al. (2008), Suruchi and Khanna (2011). In some plants, stem aids only in transport of Hg from roots to leaves, and hence, accumulation of Hg in stem is low. These observations were found supportive to that of the earlier studies (Al-Farraj and Al-Wabel 2007; Kadukova et al. 2008).

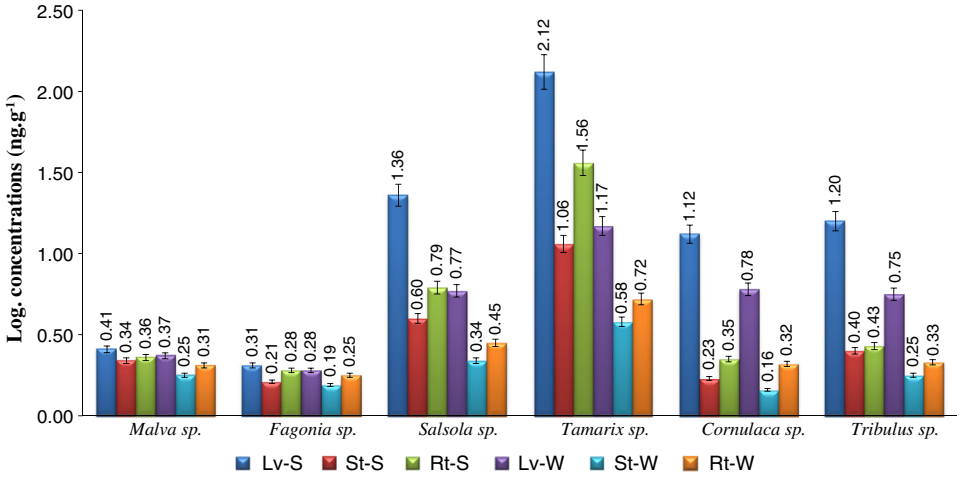

Fig. 2 Plants parts-wise mercury levels (ng g^{-1} log. conc.) during summer and winter. *Lv* leaves, *St* stem, *Rt* root, *S* summer, *W* winter

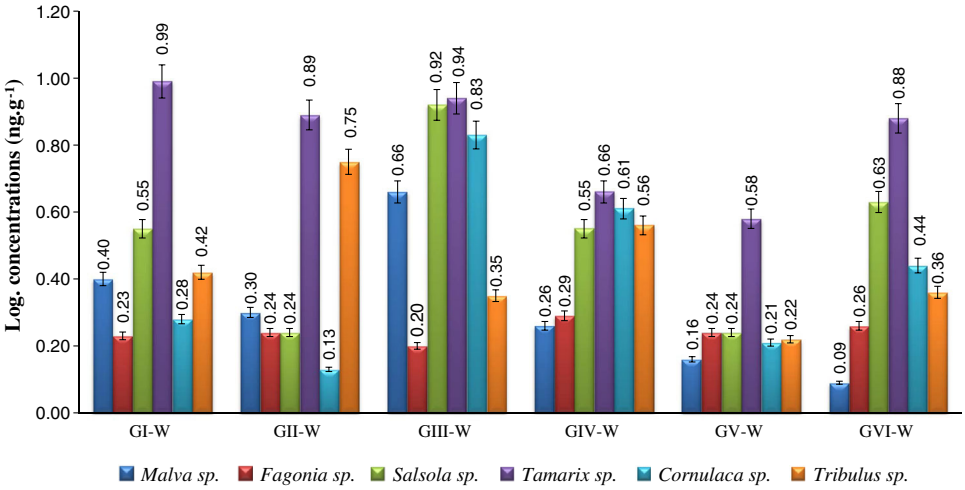

Fig. 3 Governorates-wise mercury levels (ng g^{-1} log. conc.) in desert plants during winter. *W*: winter, *GI-GVI* Kuwait Governorates

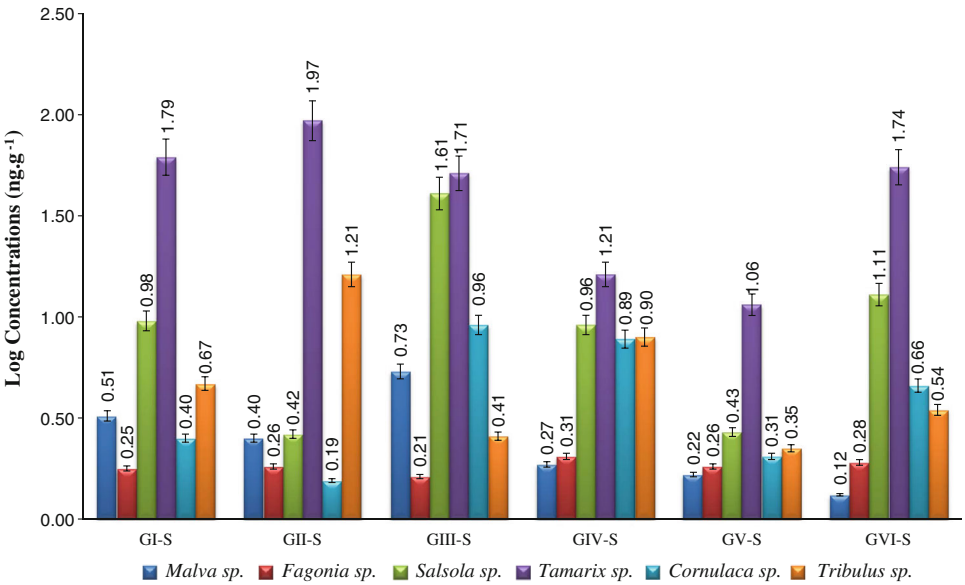

Fig. 4 Governorates-wise mercury levels (ng g^{-1} log. conc.) in desert plants during summer. *S* summer, *GI-GVI* Kuwait Governorates

Table 1 ANOVA on Hg concentrations in desert plants parts, Governorates, season-wise analyses, and surrounding soil

Source of variation	SS	*df*	*F*	*P* value	*F* ratio	Significance
A. Desert plants species versus parts-wise Hg analysis						
Plant species	3.45	5	13.76	0.0016	2.60	*
Parts (LSR)	2.37	5	9.47	0.0036	2.60	*
Error	1.25	25				
Total	7.07	35				
B. Governorate versus season-wise analysis						
Governorates (GI-GVI)	0.190	5	24.90	0.0015	5.05	*
Seasons (S, W)	0.210	1	139.46	0.0076	6.60	*
Error	0.007	5				
Total	0.407	11				
C. Species versus Governorates-wise analysis						
Plant species	06.90	5	18.84	0.0072	2.38	*
Governorates (GI-GVI)	02.53	11	3.15	0.0023	1.96	*
Error	04.02	55				
Total	13.45	71				
D. Soil analysis Governorate versus season-wise						
Governorates (GI-GVI)	0.025	5	205.48	0.008	5.05	*
Seasons (S, W)	0.003	1	126.75	0.0009	6.60	*
Error	0.001	5				
Total	0.029	11				
E. Desert plant parts (leaves, stem, root) versus soil						
Species	1.33	5	5.55	0.004	2.90	*
Parts (LSR) and soil	13.04	3	90.69	7.69×10^{-10}	3.29	*
Error	0.72					
Total	15.09					

LSR leaves, stem roots; *GI-GVI* Kuwait Governorates; *S, W* summer, winter; *SS* sum of squares; *df* degree of freedom; *F* calculated value; *F ratio* table value

* Significant

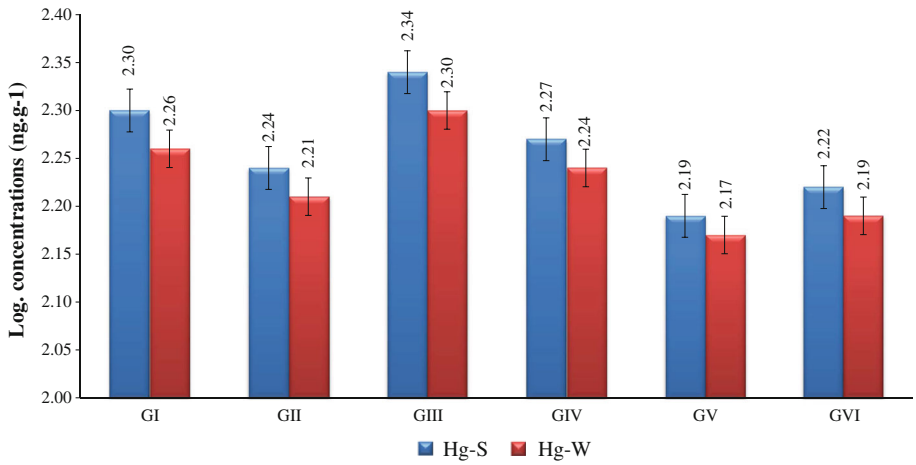

Fig. 5 Governorates-wise mercury levels (ng g^{-1} log. conc.) in soil surrounding the desert plants. *S* summer, *W* winter, *GI-GVI* Kuwait Governorates

The overall Governorate wise analysis on these desert plants showed high Hg concentrations sequence of GIII > GI > GIV > GII > GVI > GV during summer and winter seasons of Kuwait (Figs. 3, 4). Industrial wastewater percolation in the soil from chlor-alkali plants, domestic wastewater discharges, and influence of Hg vapor in the ambient air released from anthropogenic activities attribute to the high Hg concentration in GIII and GI. This supports the views of Charron and Harrison (2003), Rafat et al. (2009). Governorate wise analysis showed comparatively higher Hg concentrations in *T. chinensis* Lour., in GII and GIII because of this plant's inorganic and organic bound

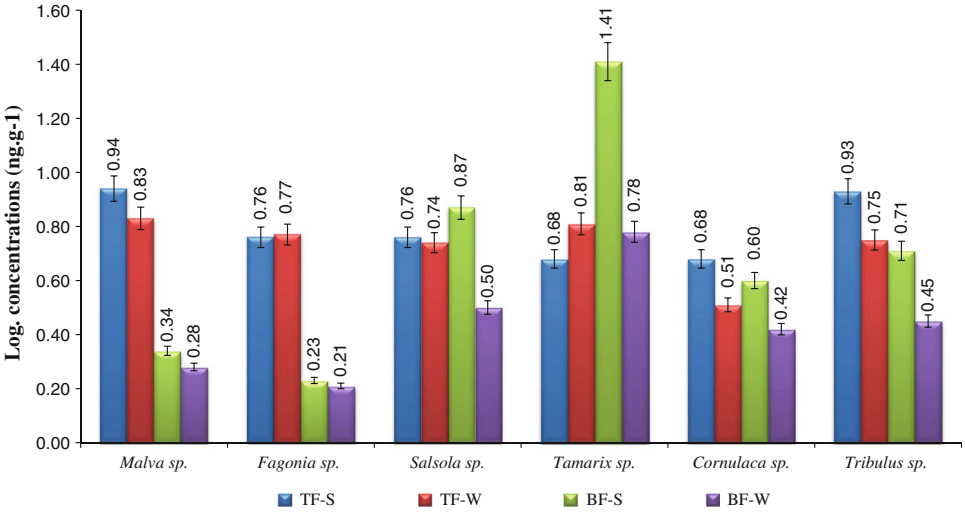

Fig. 6 Seasonal-wise translocation and bioaccumulation factors (ng g^{-1} log. conc.) for desert plants in Kuwait. *TF* translocation factor, *BF* bioaccumulation factor, *S* summer, *W* winter

soil Hg absorbing capabilities of their roots, foliage susceptibility to Hg vapor from the ambient air in addition to their abundant apportionment of plants in this Governorate compared to its distribution elsewhere the other Governorates. Statistical tests by ANOVA showed significant differences among Hg concentrations in the six desert plants during both seasons (summer and winter) and in the respective Kuwait Governorate areas (Table 1A–C).

Comparatively, Hg concentration was high in soil collected from different Governorate areas when compared to the Hg concentrations in the desert plants. Hg concentrations sequence in soil samples revealed similar Governorate wise sequence to that of the desert plants (Fig. 5). Eolian dust deposition, frequent sandstorms, and industrial activities that discharge wastes into the soil without proper treatment attributes to the high Hg levels in the soil samples. Such observations were found in line with the earlier studies of Freer-Smith et al. (2004), Chung et al. (2005), Onweremadu et al. (2007), Rafat et al. (2009), USEPA (2009), and Morales et al. (2011). ANOVA showed significant differences between Hg concentrations in the soil during the two seasons and in the respective Kuwait Governorate areas (Table 1D). Statistical test by ANOVA revealed significance between the exposure of Hg from soil to the different parts of each desert plants (Table 1E).

It is interesting to note that translocation and bioaccumulation factors were low (TF < 1) in most of the sampled desert plants, probably indicating seldom or low absorption of Hg from roots to the other aerial plant parts (Fig. 6). However, BF in desert plants and soil was found lower than the permissible limits (0.5 mg l^{-1}) of CEPA (2009) and USGS (2009). This observation agreed with the earlier findings of Baker and Brooks (1989), Ma et al. (2001), and Takashi et al. (2005) labeling plants with TF < 1 as bioindicators of pollution and TF > 1 as 'hyper' and 'accumulators' of pollution. BF was >1 in the case of *T. chinensis* Lour., especially during summer. Reasons may attribute to the following: (a) the Hg exposure from ambient air to the larger biomass of this plant in comparison with the other smaller sampled desert plants, as a result of frequent dust storms in Kuwait, (b) its halophytic nature, (c) the nature of roots that absorbs Hg from the surrounding soil to a certain extent which is present in organic speciation (sulfates, chlorides) and, (d) the Hg absorption from the surrounding soil containing Hg accumulated fallen *T. chinensis* Lour., leaves.

Conclusion

The present study revealed the state-of-the-art-technology in determining Hg concentrations in plants and soil samples at ultra-trace-level precision (0.0015 ng g^{-1} detection limits), rapidly and with seldom contamination using the DMA-80 unlike other instruments that has $\geq$0.5 ng g^{-1} detection limits. In an overall view, Hg concentrations in plants and soil samples were lower than the statutory permissible limits. Analysis on these desert plant parts revealed partial or slow process of Hg translocation and bioaccumulation compared with earlier studies in other trace metals in foliar plants. However, the recent trend in urbanization and construction activities with increasing population in Kuwait is anticipated to increase the Hg concentrations in the near future. The present study reveals that desert plants, such as *T. chinensis* Lour., and *S. imbricate* Forssk., could be characterized as a tool for biomonitoring studies. Further, taxonomical studies could be undertaken to label some of these desert plants for possible phytoremediation and curb Hg pollution in countries with arid ecosystem.

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