Pollution characteristics and health risk assessment of heavy metals in dry atmospheric deposits from Sfax solar saltern area in southeast of Tunisia

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Abstract

Concentrations of six heavy metals such as Fe, Pb, Cu, Cd, Ni and Zn in dry atmospheric deposits weekly collected through 20 sample sites from Sfax solar saltern during two successive sampling campaigns, selected from a long time monitoring, have been examined in order to evaluate their contamination levels as well as their human health risk; such concentrations (expressed in mg/kg) have shown that Fe varied from 7006.24 to 7856.45, Pb from 8.64 to 344.45, Cu from 9.33 to 298.67, Cd from 0.16 to 85.24, Ni from 6.02 to 150.02, and Zn from 12.23 to 624.57. During the study period, average concentrations (mg/kg) have been 7315.99, 103.08, 82.34, 15.93, 46.21 and 142.39, for Fe, Pb, Cu, Cd, Ni and Zn, respectively. Except for Fe, the other concentrations in dry atmospheric deposits have recorded the highest level during the first campaign especially in the sites which are close and more exposed to emissions of local pollutant industries, as well as nearby main road, resulting from high exposure frequencies. Statistical approaches, such as principal component analysis and hierarchical cluster analysis have been applied to all data, revealing an affinity between analyzed metals resulting from their origins, and confirming the influence of exposure frequencies on the spatial variability of heavy metal concentrations. Human health risk assessment has revealed that ingestion of heavy metals is the main exposure pathway for the local population. Computed Hazardous Quotient and Hazardous Index have been higher for children than for adults, thus indicating that the former may be subjected to potentially higher health risk than the latter especially during the first campaign. Calculated carcinogenic risks through ingestion and dermal contact, as well as the total carcinogenic risk for the selected heavy metals, have shown that cancer could occur more probably through ingestion than dermal contact, for children than adult, and during the first campaign (during C1: average values CR_{ing} = 8.72×10^{-4} and CR_{der} = 1.40×10^{-6} for child; average values CR_{ing} = 5.61×10^{-5} and CR_{der} = 2.84×10^{-6} for adult) than the second one (during C2: average values CR_{ing} = 1.59×10^{-4} and CR_{der} = 2.54×10^{-7} for child; average values CR_{ing} = 1.02×10^{-5} and CR_{der} = 5.19×10^{-7} for adult). The total calculated carcinogenic risk through all the sites have infrequently signaled high to very high carcinogenic risk in the first campaign (average CRA = 8.73×10^{-4} for child and CRA = 5.89×10^{-5} for adult) and occasionally exceeded the safe range for the local population in the second one (average CRA = 1.59×10^{-4} for child and CRA = 1.07×10^{-5} for adult).

Keywords Dry atmospheric deposits · Heavy metals · Exposure frequencies · Statistical approaches · Human health risk · Sfax · Solar saltern

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Introduction

In recent years, atmospheric pollution has become a serious problem in most cities around the world given its direct impact on our health and the different ecosystems [1–4]. Particulates and gaseous pollutants emitted into the atmosphere have been found to cause a serious threat due to their quick and uncontrolled spreading. This has raised more concern about the impact of particulates given their association with other chemical and microbial pollutants [2, 5–7, 103, 104].

These particulates could be removed from the atmosphere basically through wet and dry depositions process. In semi-arid and arid areas, dry deposition represents the main pathway for removing particulates from the atmosphere through gravitational settling depending on turbulent vertical transport ([8]; [9–11]). Accordingly, the dry deposition has been more significant at the semi-arid and arid regions because of infrequently and irregularity of precipitation event yielded low average annual precipitation in such regions [12–14]. Atmospheric deposition process has been identified as an important source of toxic substances, specifically heavy metals, in areas close to urban and/or industrial outlets where negative impacts on soils, sediments, aquatic environments, as well as on human health have been confirmed (; [15–17]).

Therefore, people living within areas of intensive industrial activities and high traffic density have usually been subjected to different health risks such as cancer, lung disease, DNA damage and central nervous system demolition, especially among children being more sensitive than adults due to their exposure to heavy metals [18–20]. Such exposure could be through ingestion, inhalation and dermal contact of airborne atmospheric particulate deposits containing heavy metals. Consequently, a bulk of studies have been focused on studying heavy metals found in dust and atmospheric depositions, as well as their impact on human health and the different ecosystems [6, 21–28, 102].

In the last decades, a number of studies have been concerned with pollution level and health risks basically associated with heavy metals founds in dust collected from urban and suburban road, household, airborne, nursery schools, campus bus station, Yerevan's tree and date palm leaves ([25, 29–35][24, 36–41][95][96, 97, 99, 105]). Other studies have been focused on atmospheric deposition with particular attention to their concentration, flux, spatial and temporal distribution and sources (; [10, 42–44]). But the subject still requires more investigation dual with human health risks assessment of atmospheric deposition and their heavy metal contents, particularly, presented in high concentrations [26, 45–47].

Sfax, considered as a highly industrialized and urbanized city in Tunisia, raised the concern of a number of scholars who have conducted studies. Some of them have been focused on identifying pollutants from the main industrial outlets. Other ones have been focused on examining principal constituents of aerosols (PM₁₀), based on the contribution of different enrichment sources as well as the impact of local and regional meteorology. Other studies have been focused on evaluating the spatial and temporal variability of the deposited particulate fluxes ([42, 43, 48, 49]). But further research is still required, particularly in terms of human health risk assessment of heavy metals in the atmospheric particulate deposition in Sfax. The current study is therefore focused on the southern part of Sfax, given that various

pollution sources have been implanted in its coastline, including, phosphoric acid plant (SIAPE), huge amounts of phosphogypsum, other man-made plants, pronounced shipping activities, commercial and fishing harbor, urban wastes, municipal wastewater treatment plant without neglect the contribution of road traffic (Fig. 1).

Hence, there are three main objectives of this study: (1) to determine the heavy metals Iron (Fe), Lead (Pb), Copper (Cu), Cadmium (Cd), Nickel (Ni) and Zinc (Zn) concentrations in dry atmospheric deposits collected from Sfax solar saltern during two selected campaigns; (2) to notify the influence of the exposure frequencies to the main industrial emissions on the spatial variability of such concentrations (3) to evaluate human health risk, during such campaigns, on children and adults caused by exposure to heavy metals, founded in dry atmospheric deposits, through the different exposure pathway.

Study area

The Sfax solar saltern, located in the southern coast of Sfax, is in the central east of Tunisia (latitude 34°39'N and longitude 10°42'E) on the border of Sfax solar saltern there are intensively industrialized and urbanized areas (Fig. 1). Sfax has a semi-arid Mediterranean climate, influenced partly by its moderate topography and partly by its maritime exposure. The region has been identified as well-ventilated, with usually low to moderate wind speed which could occasionally exceed 5 m/s.

Sfax solar saltern, located in the southern coastal area of the city, as well as the neighboring industrial activities have been thoroughly described in previous studies, including Bahloul et al. [42, 43, 50].

Materials and methods

Sampling and samples analysis

Dry atmospheric deposits have been collected from twenty sites abbreviated by the technical staff of Sfax solar saltern as PI2, TS2, TS35, TS36, TS38, S6, TS3, TS16, TS20, TS25, PM2, R2, R3, PI1, TS7, TS10, TS12, TS13, TS32, and TS42 (Fig. 1). In consistency with the objectives of this study, these sites have been selected in a bid to cover the majority of the areas, thus allowing for a better understanding of the impact of pollutants sources on the dry atmospheric deposits' enrichment in terms of heavy metals. In fact, samples from such sites have been collected by placing a 20 cm diameter bucket of high-density polyethylene on elevated support (1.5 m) above the ground surface in order to minimize contamination from local resuspended particles generated by high wind speed and/or mechanic engine movements between ponds. The use of bucket has been highly recommended based

Fig. 1 Sfax Solar saltern and nearby industrial sources location and map of sampling sites

on its efficiency revealed in previous studies such as Zheng et al. [51], Al-Momani et al. [22] and Sobhanardakani [26]. In the current study, dry atmospheric deposit samples have been collected over two successive campaigns, namely C1 from 17 February to 23 February 2013 and C2 from 24 February to 03 March 2013. Such campaigns have been selected from other ones, serving for a long-time monitoring of dry atmospheric deposits in such area, based on their similar meteorological parameters. In fact, Table 1 showed daily average of each selected parameters namely temperature ($^{\circ}\text{C}$), relative humidity (%), accumulated precipitation (mm), atmospheric pressure (mbar) and wind speed (m/s) and its weekly average of such selected parameters during selected period. During C1, temperatures have been ranged between 14 and 16 $^{\circ}\text{C}$ yielding an average value of 15 $^{\circ}\text{C}$. During C2, temperatures have been ranged between 9 and 17 $^{\circ}\text{C}$ yielding an average value of 13 $^{\circ}\text{C}$. For the relative humidity, average values recorded during C1 and C2 have reached 49% and 51%, respectively. No precipitations have been recorded during such selected campaigns. Recorded average daily values and weekly ones related to temperature, relative humidity and precipitation during C1 and C2 confirmed the dry mode of atmospheric deposits sampling. Examination of atmospheric pressure and wind speed has revealed that during both C1 and C2, the study area

has been underwent alternating short-lived cyclonic and anti-cyclonic situations associated with low to very high wind speed, reaching 8 m/s. Nevertheless, dominant wind direction has been mainly western during C1 and eastern during C2.

In consistency with the main objectives of this study, only wind directions have been significantly different between C1 and C2, yielding meaningfully different exposure frequencies of sampling sites to nearby contamination sources (Fig. 2; Table 1). Such difference will be meticulously studied for a better understand of it impact on the heavy metal concentrations in dry atmospheric deposits.

The pre-treatment and digestion of collected dry atmospheric deposits have been developed according to U.S. EPA. [52] method 3050B for Inductively Coupled Plasma-Optical Emission Spectrophotometer (ICP-OES) analysis cited by several authors including Abah et al. [53] and Sobhanardakani [26]. A specific amount of 1.00 g of each filtered sample has been transferred into a digestion vessel, where 10 ml of 1:1 nitric acid (HNO_3) has been added, mixed thoroughly and covered with a watch glass. Then, the samples have been heated to 90 $^{\circ}\text{C}$ and refluxed at this temperature for 10 min after which they have been allowed to cool for 5 min under room temperature. Afterward, 5 ml of concentrated HNO_3 has been added to each sample which has been covered and refluxed again at 90 $^{\circ}\text{C}$ for 30 min.

Table 1 Main meteorological parameters recorded during C1 and C2

Campaigns	Days	Temperature (°C)		Relative humidity (%)		Accumulated Precipitation (mm)		Atmospheric pressure (mbar)		Wind speed (m/s)	
		Daily average	Weekly average	Daily average	Weekly average	Daily average	Weekly average	Daily average	Weekly average	Daily average	Weekly average
C1 (17/02/2013–23/02/2013)	17/02/2013	14		33%		0		1017		2	
	18/02/2013	15		28%		0		1021		4	
	19/02/2013	15		68%		0		1014		3	
	20/02/2013	14	15	73%	49%	0	0	1006	1012	8	5
	21/02/2013	14		63%		0		1008		6	
	22/02/2013	16		48%		0		1012		6	
	23/02/2013	16		30%		0		1006		7	
C2 (24/02/2013–02/03/2013)	24/02/2013	11		58%		0		1011		5	
	25/02/2013	12		56%		0		1016		6	
	26/02/2013	9		39%		0		1019		3	
	27/02/2013	11	13	44%	51%	0	0	1021	1013	4	5
	28/02/2013	14		67%		0		1015		8	
	01/03/2013	17		53%		0		1002		6	
	02/03/2013	14		39%		0		1006		6	

Subsequently, the solutions have been allowed to evaporate without boiling to approximately 5 ml each one and cooled again for 5 min. Then, 2 ml of deionized water and 3 ml of 30% hydrogen peroxide (H_2O_2) have been added in every solution. The vessels have been covered and heated just enough to warm the solutions in order to launch peroxide reaction (ISO 4225, 1994). This has been continued until effervescence diminished and the solutions have been cooled. The acid-peroxide digestates have been covered with watch glasses and heated until the volume reduced to approximately 5 ml again. Afterward, 10 ml of concentrated hydrochloric acid (HCl) has been added to each solution which

has been covered and heated on a heating mantle then has been refluxed at 90 °C for 15 min. After cooling, each digestate has been filtered through Whatman filter paper (No. 41) into a 100 ml volumetric flask. Then, deionized water has been added until reaching a 100 ml volume. A volume equal to 10 ml of each digestate was taken mixed with equal volume of matrix modifier and then analyzed using ICP-OES for the levels of iron, lead, copper, cadmium, nickel and zinc [52]. Data generated from triplicate analysis have been subjected to treatment of mean, standard deviation and t-test at $P > 0.05$ error protection in order to minimize probable errors generated via experimental

Fig. 2 Wind roses related to C1, C2 and during the last 10 years referring to Sfax city

measurement. In fact, replicate analysis during experimentation usually generates multiple measurements which could be subjected to errors. So, statistical analysis could be used in order to tell us if the mean difference between the measured variables is statistically significant or not. Analysis of variance (t-tests), knowing as fairly robust (statistician's language for giving a valid answer) could be used to estimate the probability that the underlying phenomena are truly different. In the current study, differences have been deemed significant at $P < 0.05$. Average concentration of each analyzed element has been used for further interpretation because the reproducibility was at 95% confidence level. The results have shown good accuracy, with recovery rates for analyzed elements between 97.1 and 100.2 for dust samples.

Statistical approaches

Multivariate statistical analysis, including Principal Component Analysis (PCA) and Hierarchical Cluster Analysis (HCA) have been conducted using the STATISTICA 10 software, to evaluate associations among the investigated variables in the samples and identify the most common pollution sources. Previous studies have revealed that PCA and CA are the most common multivariate statistical methods used in environmental studies [54–56, 100, 101].

PCA, has been widely used to extract a smaller number of independent factors (principal components) among available data for analyzing variables relationships. PCA can reduce the number of correlated variables to a smaller set of orthogonal factors, making it easier to interpret a given multidimensional system by displaying the correlations among the original variables. PCA has been widely applied to various environmental media, to identify pollution sources and to apportion natural versus anthropogenic contributions. The components of the PCA have been transformed using a varimax rotation [33, 55].

HCA has been applied on the normalized data set using Ward's method and Pearson test as a measure of similarity degree, providing intuitive similarity relationships between any one sample and the entire data set. The results have been displayed as a created dendrogram, representing the most commonly used method of summarizing hierarchical clustering. [33, 55]. [94]

Spatial distribution maps of heavy metals concentrations in collected dry atmospheric deposits have been carried out using Geographical Information System (GIS 10.3) data software.

Health risk assessment of heavy metals

Exposure assessment via ingestion, dermal contact and inhalation

Health risk assessment models, developed by United State Environmental Protection Agency ([57, 58]) and the Dutch National Institute of Public Health and Environmental Protection [59], have been used to quantify the health risk

(carcinogenic and non-carcinogenic) for children and adults exposed to toxic heavy metals in collected particulate deposits through three main pathways. The first one is ingestion of dust particles. The second one is inhalation of dust particles through mouth and nose. The third pathway consist on dermal contact [32, 60]. The average daily dose (ADD), expressed in mg/Kg/day, for heavy metals in atmospheric particulate deposits through the three pathways have been calculated according to Exposure Factors Handbook [61] and the technical Report of USEPA [62] using the following equations:

$$ADD_{ing} = \frac{C \times Ing_R \times CF \times EF \times ED}{BW \times AT} \quad (1)$$

$$ADD_{inh} = \frac{C \times Inh_R \times EF \times ED}{PEF \times BW \times AT} \quad (2)$$

$$ADD_{ing} = \frac{C \times SA \times CF \times AF \times ABF \times EF \times ED}{BW \times AT} \quad (3)$$

Whereby the ADD_{ing} , ADD_{inh} and ADD_{dermal} are the average daily dose exposure to heavy metals through ingestion, inhalation and dermal contact, respectively.

Lifetime average daily dose of cancer elements (LADD) has been calculated using eq. (5) and later used in the assessment of cancer risk ([32, 57, 58, 62]; Shabbaj et al. 2017).

$$LADD = \frac{C \times CR \times EF \times ED}{PEF \times BW \times AT} \quad (4)$$

Whereby CR is the contact frequency and is the same Ing_R used in the calculation of ADD_{ing} ([57, 58, 62]; Shabbaj et al. 2017).

Description and values of factors used in Eqs. (2–5) are provided in Table 2.

Risk calculation

In order to evaluate health risk of heavy metal exposure within particulate deposits collected from southern coastal area of Sfax, the Hazard Quotient (HQ), Hazards Index (HI), and Carcinogenic Risk Assessment (CRA) have been applied, yielding finally a probability of effects occurring in human beings under specific exposure conditions.

HQ, HI, and CRA have been calculated on the basis of the proceeding calculation of ADD, using the following equations:

$$HQ = \frac{ADD}{RfD} \quad (5)$$

$$HI = \sum HQ_i \quad (6)$$

$$CRA = LADD \times SF \quad (7)$$

Reference dose (RfD) has been defined as an estimation of maximum allowed risks to human population through daily

Table 2 Heavy metal concentrations (mg/kg) in dry atmospheric deposits collected through twenty sites from southern coastal of Sfax

Factor	Description	Unit	Value		References
			Children	Adults	
C	Concentration of metals in dusts	mg/Kg			Present study
IngR	Ingestion rate of dust	mg/day	200	100	ESAG, 2009; [57, 58]
EF	Exposure frequency	days/year	350	350	[65]; [66]; ESAG, 2009
ED	Exposure duration	years	6	24	[57, 58], [67]
BW	Average body weight	Kg	15	70	[68]; [33]
AT	Average time	days/year	365 x ED	366 x ED	[60]; ESAG, 2009; [69]
CF	Conversion factor	kg/mg	1×10^{-6}	2×10^{-6}	[69]
InhR	Inhalation rate of dust	m ³ /day	7.63	12.8	[67]
PEF	Particular emission factor	m ³ /Kg	1.36×10^9	1.36×10^9	[57, 58]
SA	Surface area of skin exposed to dust	cm ²	1600	4350	[60]; ESAG, 2009
AF	Skin adherence factor	mg/cm ²	0.2	0.7	[70]; [71]
ABF	Absorption factor (Dermal)	unitless	0.001	0.001	[72]; [57, 58]; [73]

exposure pathways referring to children during a lifetime. RFD values (Table 3), used for assessment of non-carcinogenic risk, have been taken from the integrated risk information system [74, 76]

To assess cancer risk from carcinogenic metals, the calculated daily dose has been multiplied by the corresponding slope factor (SF) (mg/kg/day) for a particular carcinogen. SF is an upper bound probability of an individual developing cancer as a result of a lifetime exposure to an agent by ingestion or inhalation. The slope factor values of the selected metals (Table 4) have been extracted from numerous previous studies ([57, 58, 77]; Shabbaj et al. 2017).

The Hazard Quotient (HQ) has been calculated to assess the health effect resulting from metals. The ADD for the multiple exposure pathway, namely ingestion, inhalation and dermal contact have been divided by the specific reference dose (RfD) (mg/kg/day) for a particular metal to obtain HQ ([57, 58]; [47, 74]). If $ADD < RfD$, HQ value is lower than 1 ($HQ < 1$), occurrence of serious health risk is very likely, irrespective of the exposure pathways [74].

The hazard index (HI) refers to the “sum of more than one HQ for multiple substances and/or multiple exposure pathways” [69]. HI has been used to assess the cumulative non-cancer risk for children and adults by adding HQ of the different exposure pathways, namely ingestion, dermal contact and inhalation, from a

single metal ([39, 47, 74]). If $HI < 1$, there is no harmful effect on our health. If $HI > 1$, occurrence of serious health risk on our health is likely, becoming higher as HI value increase [39, 57, 58, 74].

The carcinogenic risk has been defined as the probability of a children or an adult developing any type of cancer over his lifetime in case of exposure to carcinogenic hazards. The estimated value for the carcinogenic risk is the probability that an individual will develop any type of cancer from life time exposure to carcinogenic hazards. Generally, referring to USEPA, the acceptable or tolerable risk for regulatory purposes is within the range of 10^{-6} and 10^{-4} implying the absence of significant carcinogenic risk. In the current study, description of carcinogenic levels will be developed based on Rapant et al. (2010) classification, summarized in Table 5.

Results and discussion

Spatial-temporal variation of heavy metals concentrations in dry atmospheric deposition

Heavy metal concentrations in dry atmospheric deposits have been summarized in Table 6, including average, maximum and minimum values as well as their standard deviation. The table

Table 3 Comparison of the heavy metals' contents (mg/kg) in atmospheric deposits of Sfax southern coastal, Tunisia with those reported values for other areas of the world

	Reference doses “RFD” (mg/kg/day)					
	Fe	Pb	Cu	Cd	Ni	Zn
RFDing	8.40E+00	3.50E-03	4.00E-02	1.00E-03	2.00E-02	3.00E-01
RFDinh	2.20E-04	3.52E-03	4.02E-02	1.00E-03	5.71E-06	3.00E-01
RFDderm	7.00E-02	5.25E-04	1.20E-02	1.00E-05	1.60E-02	6.00E-02

Table 4 Slope factor for selected metals (mg/kg/day)⁻¹

Selected metals	Slope factor “SF” (mg/Kg/day) ⁻¹	Reference
Fe	.	.
Pb	8,50E-3	[57, 58] [77]Shabbaj et al. 2017
Cu	.	.
Cd	6,30	[57, 58] [77] Shabbaj et al. 2017
Ni	8,40E-01	[57, 58] [77] Shabbaj et al. 2017
Zn	.	.

also shows crust average concentrations of selected metals [78, 79]. Fe has been the most abundant metal in all concentrations whereas Cd has been found in the lowest concentrations. Despite their important concentration, in the range of 7006.24 and 7856.45 mg/Kg, iron has always been found lower than the crust background value (35,900 mg/Kg). The geogenic origin of this metal could be the main reason for its high recorded concentrations [47, 80, 85]. As for the other metals Pb, Cu, Cd, Ni and Zn, maximum concentrations have largely exceeded crust average values, indicating that the pollution in urban particle deposits derive mainly from anthropogenic sources. In fact, maximum concentrations have been 5 times higher than the crust average values for zinc and around 420 times for cadmium, during C1, and one time higher for Ni and 3 times higher for lead, during C2. During the two selected campaigns, average concentrations of Pb, Cu, Cd, Ni and Zn, have varied between 26.81 and 182.70 for Pb; between 29.55 and 139.02 for Cu; between 4.40 and 27.89 for Cd; between 18.79 and 75.16 for Ni and between 29.98 and 260.42 mg/kg for Zn. The distribution of all the measured metals has been found to follow the pattern of Fe > Zn > Pb > Cu > Ni > Cd during C1 and Fe > Zn > Cu > Pb > Ni > Cd during C2. Such pattern difference could be explained by the effect of dominant wind, yielding a variations of exposure frequencies,

Table 5 Sampling sites repartition based on their distance from main industrial sources' and exposure frequencies to their plumes'

Risk level	Calculated cases of cancer occurrence	Cancer risk
I	$< 10^{-6}$	Very low
II	$10^{-6} - 10^{-5}$	Low
III	$10^{-5} - 10^{-4}$	Medium
IV	$10^{-4} - 10^{-3}$	High
V	$> 10^{-3}$	Very high

leading the inflow of atmospheric anthropogenic emissions.

A comparison of heavy metal concentrations in the atmospheric deposits of the southern coastal area of Sfax with those in other areas around the world has been summarized in Table 7. In fact, the heavy metal concentrations in dry atmospheric deposits collected from our study area have sometimes been comparable to those recorded in other areas and sometimes lower/higher. In fact, similar patterns have been found on Kandy city, Sri Lanka, for instance, slightly changed in another study on Yerevan, Armenia.

The variation of heavy metal concentrations and levels in dry atmospheric deposits collected from different areas around the world could be explained by the variety and intensity of human activities, traffic density, land use patterns, employed sampling technologies and local weather conditions [55, 88].

Spatial distribution

Based on Figs. 3 and 4, the spatial distribution of heavy metal concentrations, except iron, in collected deposits through twenty sampling sites over Sfax southern coastal area has shown that TS35, TS38, S6, R2, R3 and TS36 frequently recorded high metal concentrations. PI1, PI2, PM2, TS32, TS20, TS16, TS3, TS7 and TS25 have shown lower concentrations than those aforementioned. The lowest heavy metal concentrations have been registered at TS42, TS13, TS12, TS2 and TS10. Such spatial distribution of heavy metal concentrations in collected dry deposits has been consistent with those elaborated in the case study of heavy metal concentrations in surface sediments [50].

In fact, distribution maps for Cu, Cd and Zn concentrations have shown that the highest concentrations have been recorded in collected dry atmospheric deposits from R2, TS35, TS38 and S6 sites, during C1 (Fig. 3). Accordingly, the significant exposure frequency of such close sites to the SIAPE and lead foundry releases during this campaign explain highest founded concentrations. It has been demonstrated, elsewhere, that Cu, Cd and Zn have been mainly originated from nearby industrial activities [21]. For lead and nickel, in addition to recorded high concentrations in dry atmospheric deposits collected from R2, TS35, TS38 and S6 sites, dry atmospheric deposits collected from TS3, TS16 and rarely TS7 have shown significant concentrations. Such high concentrations in terms of Pb and Ni in dry deposits collected from TS3, TS16 and TS7 compared to other close sampling sites could be explained by the impact of high traffic activities of Gabes road as well as the impact of re-suspension of the roadside soil (enriched with Pb from past usage of leaded fuel), originating from incomplete sidewalk, in the study area atmosphere [47, 88].

During the second campaign, spatial distribution maps of heavy metal concentrations in collected dry atmospheric deposits (Fig. 4) have usually been completed referring to crust

Table 6 Values of exposure factors for heavy metals doses for children and adults

Concentrations (mg/kg)/sampling sites	C1						C2					
	Fe	Pb	Cu	Cd	Ni	Zn	Fe	Pb	Cu	Cd	Ni	Zn
TS35	7856,45	254	298,7	85,24	148,1	624,6	7836,39	45,02	58,98	15,43	37,02	49,43
TS38	7462,48	268,4	288,3	79,45	136,9	586,5	7452,41	47,71	46,53	14,23	34,22	50,38
S6	7368,51	253,4	254,6	66,74	124,3	601,2	7353,44	45,60	33,64	10,03	31,08	40,14
R2	7499,84	298,4	261,8	80,03	150	592	7481,77	47,78	65,44	17,05	37,51	66,79
PM2	7176,54	344,5	201,7	44,32	101,9	432,1	7174,48	32,50	50,42	8,33	25,47	48,02
R3	7324,90	277,8	228,3	50,45	112,3	401,3	7316,83	42,36	27,08	5,61	28,08	24,80
TS36	7126,24	154,2	198,8	45,68	99,78	365,4	7124,18	27,90	39,69	5,08	24,95	45,51
TS32	7116,08	198,7	167,5	33,43	78,54	267,4	7108,95	24,68	41,89	3,71	19,64	38,51
TS20	7029,95	155,4	88,67	12,33	68,65	203,3	7022,82	8,64	15,17	1,37	17,16	19,13
TS25	7063,02	98,45	76,45	10,21	55,49	187,6	7034,90	14,31	19,11	1,13	13,87	18,19
TS16	7682,09	301,5	123,3	8,44	70,23	123,4	7783,21	26,60	30,84	0,60	17,56	27,43
TS3	7636,30	288,4	101,3	8,33	68,60	98,65	7667,42	25,31	25,33	1,37	17,15	21,53
TS7	7676,33	258,7	113,7	8,38	65,44	167,6	7677,45	20,83	28,42	0,93	16,36	26,75
TS42	7006,24	60,54	33,23	4,32	24,07	78,54	7084,36	11,47	9,33	0,48	6,02	12,23
TS13	7013,15	56,79	51,23	3,48	30,01	70,32	7031,27	9,21	12,81	0,39	7,50	14,34
TS12	7024,82	101,3	65,45	2,78	33,22	88,43	7042,94	16,04	16,36	0,31	8,31	21,36
TS2	7016,08	123,1	34,23	1,43	25,43	68,76	7017,20	29,14	16,56	0,16	6,36	24,75
TS10	7599,87	66,92	88,45	1,04	52,32	98,02	7611,00	28,76	22,11	0,56	13,08	20,55
PI1	7411,21	11,16	10,58	0,26	6,92	14,54	7381,21	16,16	15,58	1,26	12,92	19,54
PI2	7117,22	13,21	12,64	0,94	7,56	15,27	7230,22	18,21	19,64	2,09	14,56	21,27
Average	7310,37	179,25	134,94	27,36	72,99	254,25	7321,62	26,91	29,75	4,51	19,44	30,53
Max	7856,45	344,45	298,67	85,24	150,02	624,57	7836,39	47,78	65,44	17,05	37,51	66,79
Min	7006,24	11,16	10,58	0,26	6,92	14,54	7017,20	8,64	9,33	0,16	6,02	12,23
Standard Deviation	269,15	104,47	92,53	29,74	44,15	208,48	269,44	12,69	15,61	5,38	9,77	14,48
Crust Average ([78], [79])	35,900,00	16,00	32,00	0,20	20,00	127,00	35,900,00	16,00	32,00	0,20	20,00	127,00

average concentrations. It has been demonstrated that for all analyzed metals recorded concentrations during C2 have been lower than those recorded during C1. This suggest the impact of meteorological parameters, particularly exposure frequencies resulting from dominant wind direction, in conditioning heavy metals levels in collected dry atmospheric deposits. In fact, during C2, most heavy metal concentrations have rarely exceeded crust average concentrations, excepting in the case study of cadmium. Such distribution could suggest the combined impact of nearby industrial sources, road traffic (Gabes road) and atmospheric effluents coming from urban areas on charging dry atmospheric deposits in terms heavy metal contents.

In order to better understand such difference, study of temporal variation of heavy metal concentrations in collected dry atmospheric deposits will be carried out.

Temporal variation

The investigation of temporal variation of all heavy metal concentrations in dry atmospheric deposits, excepting iron,

have shown a significant difference between C1 and C2. In fact, the average concentrations of Pb, Cu, Cd, Ni and Zn have been 4 to 8 times higher during C1 than during C2. Moreover, heavy metal concentrations in dry atmospheric deposits, collected during C1 basically from TS35, TS38, S6, R2, R3, TS36 and PM2 and to a lesser extent from TS16, TS3, and TS7, have largely exceeded those recorded during C2. However, for other sampling sites such as TS10, TS2, TS12, TS13 and TS42 and to a lesser extent TS20 and TS25 such concentrations have been slightly higher during C1 than those recorded during C2. Such variation has been likely resulting from the concomitant effect of main surrounding industrial sources, namely SIAPE, Lead foundry FP Sfax sud and SIOS-ZITEX, sites exposure to such industrial plumes and local airflow characteristics.

Meteorological parameters examination

The investigation of meteorological parameters during C1 and C2 has revealed that the study area has been underwent

Table 7 Average daily exposure dose for carcinogenic elements through ingestion and dermal contact and their Hazard Quotients and Hazard Index for adults and children

Study area	Metal concentrations in atmospheric deposits (mg/kg)						References
	Fe	Pb	Cu	Cd	Ni	Zn	
Sfax solar saltern, Southern coastal area of Sfax, Tunisia	7006.24–7856.45	8.64–344.45	9.33–298.67	0.16–85.24	6.02–150.02	12.23–624.57	Present study
Kandy City, Sri Lanka	770.80–43,922.50	30.30–772.10	25.50–347.70	6.50–386.50	8.90–306.40	136.90–3331.40	Weerasundara et al. [47]
Kermanshah province, west part of Iran	13,150.00–35,562.00	.	24.00–256.00	.	60.00–245.00	88.00–700.00	Doabi et al. [55]
Yerevan, Armenia	.	12.10–108.47	60.24–757.70	0.09–783.43	15.48–47.77	129.01–511.45	Maghakyan et al. [40]
Nanjing, China	34,200.00	.	123.00	.	55.90	394.00	Hu et al. (2011)
Industrial area of Jordan	12,100.00–15,500.00	47.9–66.20	63.1–129.20	4.46–12.24	.	499.20–713.10	Jaradat et al. [86]
Eastern areas of Mazowieckie Province (three cities: Siedlce, Mińsk-Mazowiecki, and Sokołów Podlaski), Poland	.	12.80–13,381.00	9.90–8228.80	0.10–83.90	6.90–356.10	89.70–63,077.80	Królak [87]
Madrid, Spain	19,300.00	.	188.00	.	44.00	476.00	De Miguel et al. (1997)

alternating short-lived cyclonic and anticyclonic situations associated with low to very high wind speed, reaching 8 m/s.

For C1, the first three days have been characterized by high pressure, reaching 1021 hPa, and low to moderate wind speed, ranging between 2 and 4 m/s (Table 1). Within the next days, a cyclonic situation has been shown marked by high to very high wind speed, reaching 8 m/s. The wind has blown from all directions, with the dominance of western sector (Fig. 2). Such meteorological conditions have been confirmed favorable for a significant dispersion of pollutants, disfavoring their dry deposition. Whereas, it has been proved that concentrations of such pollutants could be 10 times higher with obstacle attendance nearby of emission sources. Consequently, highest recorded heavy metals concentrations in dry atmospheric deposits, specifically collected from R2, TS35, TS38 and S6, could be explained partly by the highest exposure frequencies of such sites, reaching 44% (Table 8), to industrial plumes especially of SIAPE, associated with high wind speeds, reaching 8 m/s and partly by the location of phosphogypsum deposits, representing an obstacle, creating hence risk zones of locally increasing pollution levels, along both its upstream and downstream. Such results have been proved elsewhere [43, 49].

As for C2, examination of meteorological parameters has revealed short-lived cyclonic situations marked by moderate to high wind speed, reaching 8 m/s. Such situations have been

interspersed by short anticyclonic situation characterized by low to high wind speed, ranging between 3 and 6 m/s (Table 1). The wind has blown from all directions, with the dominance of eastern sector (Fig. 2). During this campaign, exposure frequencies of sampling sites to the industrial plumes, mainly originating from SIAPE and Lead foundry FP Sfax sud, have never exceeded 20%. Such exposure frequencies have been lower than those observed during C1 (Table 8). Being boarded by Mediterranean Sea in its eastern part, the study area has been then emanated mainly by marine elements under winds, originating principally from eastern sector [42, 43]. Such conditions could explain obtained low heavy metal concentrations in dry atmospheric deposits recorded during the second campaign.

In terms of distance between sampling sites and main industrial sources (SIAPE and Lead foundry FP Sfax sud), four areas have been determined (Table 8). This distribution has demonstrated that average concentrations for Pb, Cu, Cd, Ni and Zn in dry atmospheric deposits collected from “A1” and “A2” have been 4 to 12 times higher during C1 than during C2. This could explain that sampling sites located less than 3 Km from the main industrial sources (SIAPE and Lead foundry FP Sfax sud) and which have been significantly exposed to such industrial source’s plumes, during C1 have recorded higher average heavy metals concentrations than those which have been less exposed to such industrial plumes during C2. In the case study

Fig. 3 Spatial distribution map of heavy metals concentrations in atmospheric deposits above Sfax solar saltern during the first sampling campaign C1 (17/02/2013–23/02/2013)

of the third area “A3”, average heavy metals concentrations during C1 have been 3 to 7 times higher than those recorded during C2. For A4, average heavy metals concentrations during C2 have been slightly higher than those recorded during C1. This could be explained by the similar frequencies exposure to abovementioned industrial sources ranging between 10 and 12% during the two campaigns.

Such distribution patterns seemed to be governed by the combined effects between nearby industrial sources, exposure to their industrial plumes, and local airflow characteristics.

Chemometric approaches

The chemometric study has been carried out based on multivariate statistical analysis methods such as PCA and CA applied to all data. Such methods have been considered as

efficient tools usually used for studying the behavior of analyzed elements and particularly, in the current study, have used for studying behavior of heavy metals in sampled urban dust.

PCA applied to heavy metal concentrations in collected dry atmospheric deposits has shown highly significant correlation between Pb, Zn, Cu, Ni and Cd, but no significant one between Fe and aforementioned elements.

Representation of analyzed metals in correlation circle has shown their good repartition through the factorial axes. In fact, the first and the second factorial axis have described 68.93 and 14.17% of the total variance in the dataset, respectively, representing then the maximum of inertia (83.10%). Additionally, Pb, Ni, Cu, Cd and Zn have been regrouped and plotted in negative pole of axis 1, contrary to Fe which has been plotted in axis 2 positive pole (Fig. 5). From such repartition, two distinct groups have been identified:

Fig. 4 Spatial distribution map of heavy metals concentrations in atmospheric deposits above Sfax solar saltern during the second sampling campaign C2 (24/02/2013–02/03/2013)

- Group1 (G1), positively correlated with axis 2, has been articulated around iron, undoubtedly indicating its natural origin;
- Group2 (G2), consisted of Pb, Zn, Cu, Ni and Cd, strongly and negatively plotted over axis 1. This group has not revealed significant correlation with the first one (G1), indicating thus the anthropogenic origin of such metals.

HCA, characterized by revealing specific linkage between heavy metals through the measuring of their similarities and dissimilarities in order to determine the regrouping of heavy metals' profiles among the sampling sites and thereafter their likely origins. In the current study, HCA has been performed, on the normalized dataset, using Ward's method, Pearson's test as the measurement unit of similarity to assess the inter-

relationship between Fe, Pb, Zn, Cu, Ni and Cd (Fig. 6). In fact, obtained dendrogram has shown two main classes. The first class has been represented by Fe, indicating its natural origin. The second class has been defined by Pb, Ni, Cu, Cd and Zn, indicating likely their anthropogenic origin. Moreover, under this class some statistical affinities have been revealed, engendering two subclasses. The first one has been represented by Pb and Ni and the second one has been represented by Cu, Zn and Cd. Affinity between Pb and Ni could be explained by the impact of dust resuspension trend mainly originated from Gabes road. In fact, several studies [89–91] have already highlighted the fact that road traffic emissions are the main source of heavy metals such as Pb and Ni.

The projection of twenty sampling sites over the factorial plan 1×2 (representing the maximum of inertia = 83.10%) (Fig. 7) has shown a clear spatial distribution, thus enabling to detect the

Table 8 Reference dose (RFD) values for the multiple exposure pathways: ingestion (RFDing), inhalation (RFDinh) and dermal contact (RFDderm)

Areas	Sampling sites	Distance from main industrial sources		Directions and Exposure frequencies to main industrial sources plumes (%)					
		SIAPE	Lead foundry FP Sfax sud	SIAPE			Lead foundry FP Sfax sud		
					C1	C2		C1	C2
A1	R2, TS35, TS38, S6, PM2, R3	≤ 2000 m		WSW, SW	44%	11%	W, WNW, WSW	27%	18%
A2	TS36, TS32, TS20, TS25, TS16, TS3, TS7	]2000;3000[		WSW, SW	44%	11%	W, WSW	24%	13%
A3	TS42, TS13, TS12, TS2, TS10	≥ 3000 m		WSW, SW, SSW	48%	14%	WSW	20%	9%
A4	PI1, PI2	≤ 2000 m		NW, WNW	10%	12%	NW, WNW	10%	12%

same groups already identified in studies on spatial distribution of heavy metals.

In fact, distribution of sampling sites over axis 1 has been demonstrated inversely proportional, indicating that sites with high heavy metal concentrations in collected deposits have been strongly and negatively plotted over axis 1. In contrast, the sites with low heavy metal concentrations in collected deposits have been strongly and positively plotted over the same axis. Correspondingly, the nearest sites to industrial sources, namely TS35, R2, TS38, S6, R3 and PM2, have been negatively plotted over axis 1. In turn, the most distant sites from industrial sources, namely TS42, TS13, TS12 and TS2, and low exposed sampling sites to nearby industrial plumes, namely PI1 and PI2, have been positively plotted over axis 1, characterizing sites with low heavy metal concentrations.

HCA of twenty sampling sites, performed on the normalized dataset using Ward's method and Pearson's correlation coefficients, has shown a dendrogram composed of two main clusters

(Fig. 8). The first cluster, has demonstrated a significant statistical affinity between TS35, TS38, S6 and R2 on the one hand and between PM2, R3 and TS36 on the other hand. This cluster has regrouped sampling sites, characterizing by their proximity and high exposure frequencies to the main industrial plumes originating from SIAPE and Lead foundry FP Sfax sud, yielding high heavy metal concentration in collected deposits. The second cluster has revealed statistical affinities harvesting the identification of two sub clusters: the first one regroups TS32, TS20, TS25, TS16, TS3 and TS7 and the second one regroups TS42, TS13, TS10, TS12, TS2, PI1 and PI2. Such statistical affinities have been in consistency with previous results revealed from spatial distribution maps as well as principal component analysis studies, thus signaling the impact of nearby industrial sources and exposure frequencies to their plumes.

In the light of the foregoing, the study of potential adverse health effects caused by exposure to heavy metal contents in dry atmospheric deposits is worth to be well assessed.

Fig. 5 Projection of variables (analyzed metals) in the 1×2 factorial plane (representing 83.10% of the total variance) during the study period (threshold of significance = 0.444 for $p < 0.05$ and $n = 20$)

Fig. 6 Dendrogram showing hierarchical cluster analysis of analyzed metals with regard to Pearson method-Pearson test

Health risks assessment

Health risk assessment of trace metals exposure from dry depositions particulates

Human exposure to atmospheric particulate matter (PM) deposits bounds with varying concentrations of heavy metals has been assessed using the established model by the US EPA (2009a, 2010, 2016a). Usually, there have been three pathways of heavy metal exposures namely inhalation, ingestion and dermal contact to assess. For the purpose of this study, risk from inhalation will not be assessed, given that undifferentiated dust

from collected dry atmospheric deposits has not been investigated [40].

The average daily dose (ADD), Hazard quotient (HQ) and Hazard index (HI) of heavy metals in atmospheric dry deposits

Calculated average daily doses for metal exposures through ingestion and dermal contact pathways have been listed in Table 9. The results have shown that exposure through ingestion pathway has been more harmful to the local population (children and adult) than dermal contact during both C1 and C2 in the area. Such results have been in consistency with previous studies reported worldwide [26, 72]. Comparatively, children have been more vulnerable to heavy metals exposure than adults through ingestion pathway, contrary to dermal contact where adults have been more vulnerable than children. In addition, for all selected heavy metals founded average daily doses during C1 have been higher than those founded during C2. In fact, the highest value of ADD corresponding to the iron ingestion by children has been recorded during C1 in the most exposed area to the nearby industrial plumes namely A1. In contrast, the lowest value of ADD corresponding to the cadmium dermal contact of children has been recorded during C2 in the most distant area from the nearby industrial sources namely A3. If an average daily dose (ADD) value has been lower than the reference dose, there has been unlikely to be any adverse health effect; otherwise, if the ADD value has been higher than

Fig. 7 Projection of twenty sampling sites in the 1×2 factorial plane (representing 83.10% of the total variance) during the study period (threshold of significance = 0.444 for $p < 0.05$ and $n = 20$)

Fig. 8 Dendrogram showing hierarchical cluster analysis of sampling sites with regard to Pearson method-Pearson test

the RfD, it has been likely that the exposure pathway will cause adverse human health effects [76]. Based on Tables 3 and 9, adverse human health effects have occurred rarely during C1 but never during C2.

The ratio of the average daily dose to the reference dose can be used to estimate the hazard quotient: when $HQ < 1$, there are no adverse health effects and $HQ > 1$ indicates that there are likely adverse health effects (US EPA, 1986). Calculated HQs of analyzed heavy metals in collected dry atmospheric deposits have been shown in Table 9. Such results have revealed that HQs of all analyzed heavy metals for ingestion have been higher than those for dermal contact for both children and adults. Moreover, HQs values calculated for C1 have been higher than those calculated for C2, suggesting a potential health risk by ingestion and/or inhalation during C1 more likely than during C2. That is, highest value of HQ, greater than 1, has been recorded for children ingestion of lead during C1 in A1. Excepting HQ_{ingestion} of Pb by children, generally hazard quotients calculated for the exposure to the selected metals have been lower than 1, implying low risk to non-cancer diseases.

HI values for all metals have revealed that calculated HI during C1 have been higher than those during C2, suggesting that the former may be subjected to potentially higher health risk than the latter (Table 9). In fact, during C1, HI values for the studied metals have decreased in the order of $Pb > Cd > Ni > Cu > Zn > Fe$, and during C2 in the order of $Cd > Pb > Cu > Zn > Ni > Fe$. Such disrespectful order could be explained by the different recorded exposure frequencies between C1 and C2 responsible for a significant heavy metals input, through nearby industrial plumes, to the study area. Moreover, during both C1 and C2, HI values for children have been higher than those for adults, indicating that the former may have more potential risk due to the exposure to heavy metals in dry atmospheric deposits than the latter. Based on HI

values, Pb and Cd have exhibited higher values close to safe level, infrequently exceeding such level during C1, indicating a significant risk for children due to their exposure. Accordingly, Pb and Cd should be paid more attention for the high HI values as well. Such results have in consistency with other ones from diverse studies, revealing that Pb in urban areas has demonstrated a significant influence on children health [34, 74, 92].

Carcinogenic risk assessment

The carcinogenic risks through ingestion mode of exposure (CRA_{ing}) and using dermal contact (CRA_{der}) as well as the total carcinogenic risk for selected heavy metals (Pb, Cu, Cd and Ni) have been calculated based on slop factors (SF) values from previous studies (Table 4) ([57, 58, 77]; Shabbaj et al. 2017). Due to lack of slop factors for Fe and Zn, their carcinogenic risks have not been considered. The CRA values for children and adults exposed to these heavy metals in dry atmospheric deposits from different functional areas are presented in Table 10.

In fact, whatever the exposure pathway, calculated carcinogenic risks have been ranged between 1.04×10^{-10} , indicating very low risk, and 5.46×10^{-3} , suggesting very high risk. It has been revealed that cancer risk through the ingestion of heavy metals has been higher than through dermal contact. Such results have been in constancy with previous studies which have indicated that ingestion of dust particles has been shown the main pathway of exposure to toxic heavy metals in dust, leading to a higher risk ([32, 39, 60]; ; [26]). Comparatively, children have been shown more vulnerable to carcinogenic risks than the adults. Generally, carcinogenic risk levels have been higher during C1 than during C2. In addition, during C1 very high risks have been recorded for Ni and Cd. Contrary, during

Table 9 Carcinogenic risk assessment via ingestion exposure and dermal contact of heavy metals in dry atmospheric deposits for both children and adult population collected from urban and industrialized area of south-Sfax city, Tunisia

			ADDing		ADDdermal		HQing		HQdermal		HI = $\sum$ HQ	
			Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult
A1	C1	Fe	9,52E-02	1,02E-02	1,52E-04	3,11E-04	1,13E-02	1,21E-03	2,18E-03	4,44E-03	1,35E-02	5,65E-03
		Pb	3,55E-03	2,28E-04	5,68E-06	1,16E-05	1,01E+00	6,52E-02	1,08E-02	2,21E-02	1,03E+00	8,73E-02
		Cu	3,27E-03	2,10E-04	5,23E-06	1,07E-05	8,17E-02	5,25E-03	5,23E-04	1,07E-03	8,22E-02	6,32E-03
		Cd	8,66E-04	5,56E-05	1,39E-06	2,82E-06	8,66E-01	5,56E-02	1,39E-01	2,82E-01	1,00E+00	3,38E-01
		Ni	1,65E-03	1,06E-04	2,64E-06	5,38E-06	8,24E-02	5,30E-03	4,88E-05	9,96E-05	8,25E-02	5,40E-03
		Zn	6,90E-03	4,44E-04	1,10E-05	2,25E-05	2,30E-02	1,48E-03	1,84E-04	3,75E-04	2,32E-02	1,85E-03
	C2	Fe	5,66E-02	3,64E-03	9,06E-05	1,85E-04	6,74E-03	4,33E-04	1,29E-03	2,64E-03	8,03E-03	3,07E-03
		Pb	5,73E-04	3,68E-05	9,17E-07	1,87E-06	1,64E-01	1,05E-02	1,75E-03	3,56E-03	1,65E-01	1,41E-02
		Cu	6,01E-04	3,86E-05	9,62E-07	1,96E-06	1,50E-02	9,66E-04	9,62E-05	1,96E-04	1,51E-02	1,16E-03
		Cd	1,51E-04	9,68E-06	2,41E-07	4,91E-07	1,51E-01	9,68E-03	2,41E-02	4,91E-02	1,75E-01	5,88E-02
		Ni	4,12E-04	2,65E-05	6,59E-07	1,34E-06	2,06E-02	1,32E-03	1,22E-05	2,49E-05	2,06E-02	1,35E-03
		Zn	5,96E-04	3,83E-05	9,53E-07	1,94E-06	1,99E-03	1,28E-04	1,59E-05	3,24E-05	2,00E-03	1,60E-04
A2	C1	Fe	9,38E-02	1,00E-02	1,50E-04	3,06E-04	1,12E-02	1,20E-03	2,14E-03	4,37E-03	1,33E-02	5,57E-03
		Pb	2,66E-03	1,71E-04	4,25E-06	8,67E-06	7,59E-01	4,88E-02	8,10E-03	1,65E-02	7,68E-01	6,53E-02
		Cu	1,59E-03	1,02E-04	2,54E-06	5,18E-06	3,97E-02	2,55E-03	2,54E-04	5,18E-04	4,00E-02	3,07E-03
		Cd	2,33E-04	1,50E-05	3,73E-07	7,62E-07	2,33E-01	1,50E-02	3,73E-02	7,62E-02	2,71E-01	9,12E-02
		Ni	9,26E-04	5,95E-05	1,48E-06	3,02E-06	4,63E-02	2,97E-03	2,74E-05	5,59E-05	4,63E-02	3,03E-03
		Zn	2,58E-03	1,66E-04	4,13E-06	8,42E-06	8,61E-03	5,53E-04	6,88E-05	1,40E-04	8,67E-03	6,94E-04
	C2	Fe	5,55E-02	3,57E-03	8,88E-05	1,81E-04	6,60E-03	4,25E-04	1,27E-03	2,59E-03	7,87E-03	3,01E-03
		Pb	2,22E-04	1,43E-05	3,56E-07	7,25E-07	6,35E-02	4,08E-03	6,77E-04	1,38E-03	6,42E-02	5,46E-03
		Cu	3,66E-04	2,35E-05	5,86E-07	1,19E-06	9,15E-03	5,88E-04	5,86E-05	1,19E-04	9,21E-03	7,08E-04
		Cd	2,59E-05	1,67E-06	4,15E-08	8,46E-08	2,59E-02	1,67E-03	4,15E-03	8,46E-03	3,01E-02	1,01E-02
		Ni	2,31E-04	1,49E-05	3,70E-07	7,55E-07	1,16E-02	7,44E-04	6,86E-06	1,40E-05	1,16E-02	7,58E-04
		Zn	3,60E-04	2,31E-05	5,76E-07	1,17E-06	1,20E-03	7,71E-05	9,60E-06	1,96E-05	1,21E-03	9,67E-05
A3	C1	Fe	9,12E-02	9,77E-03	1,46E-04	2,97E-04	1,09E-02	1,16E-03	2,08E-03	4,25E-03	1,29E-02	5,41E-03
		Pb	1,04E-03	6,72E-05	1,67E-06	3,41E-06	2,99E-01	1,92E-02	3,18E-03	6,49E-03	3,02E-01	2,57E-02
		Cu	6,97E-04	4,48E-05	1,12E-06	2,27E-06	1,74E-02	1,12E-03	1,12E-04	2,27E-04	1,75E-02	1,35E-03
		Cd	3,34E-05	2,15E-06	5,34E-08	1,09E-07	3,34E-02	2,15E-03	5,34E-03	1,09E-02	3,87E-02	1,30E-02
		Ni	4,22E-04	2,71E-05	6,75E-07	1,38E-06	2,11E-02	1,36E-03	1,25E-05	2,55E-05	2,11E-02	1,38E-03
		Zn	1,03E-03	6,64E-05	1,65E-06	3,37E-06	3,44E-03	2,21E-04	2,76E-05	5,62E-05	3,47E-03	2,78E-04
	C2	Fe	5,30E-02	3,41E-03	8,48E-05	1,73E-04	6,31E-03	4,06E-04	1,21E-03	2,47E-03	7,52E-03	2,88E-03
		Pb	3,10E-04	1,99E-05	4,96E-07	1,01E-06	8,85E-02	5,69E-03	9,44E-04	1,93E-03	8,95E-02	7,62E-03
		Cu	1,97E-04	1,27E-05	3,16E-07	6,44E-07	4,93E-03	3,17E-04	3,16E-05	6,44E-05	4,96E-03	3,82E-04
		Cd	4,83E-06	3,11E-07	7,73E-09	1,58E-08	4,83E-03	3,11E-04	7,73E-04	1,58E-03	5,61E-03	1,89E-03
		Ni	1,06E-04	6,78E-06	1,69E-07	3,44E-07	5,28E-03	3,39E-04	3,13E-06	6,37E-06	5,28E-03	3,46E-04
		Zn	2,38E-04	1,53E-05	3,81E-07	7,78E-07	7,95E-04	5,11E-05	6,36E-06	1,30E-05	8,01E-04	6,40E-05
A4	C1	Fe	9,29E-02	9,95E-03	1,49E-04	3,03E-04	1,11E-02	1,18E-03	2,12E-03	4,33E-03	1,32E-02	5,51E-03
		Pb	1,56E-04	1,00E-05	2,49E-07	5,08E-07	4,45E-02	2,86E-03	4,75E-04	9,69E-04	4,50E-02	3,83E-03
		Cu	1,48E-04	9,54E-06	2,38E-07	4,84E-07	3,71E-03	2,39E-04	2,38E-05	4,84E-05	3,73E-03	2,87E-04
		Cd	7,67E-06	4,93E-07	1,23E-08	2,50E-08	7,67E-03	4,93E-04	1,23E-03	2,50E-03	8,90E-03	3,00E-03
		Ni	9,26E-05	5,95E-06	1,48E-07	3,02E-07	4,63E-03	2,98E-04	2,74E-06	5,59E-06	4,63E-03	3,03E-04
		Zn	1,91E-04	1,22E-05	3,05E-07	6,22E-07	6,35E-04	4,08E-05	5,08E-06	1,04E-05	6,40E-04	5,12E-05
	C2	Fe	9,02E-02	5,80E-03	1,44E-04	2,94E-04	1,07E-02	6,91E-04	2,06E-03	4,21E-03	1,28E-02	4,90E-03
		Pb	2,20E-04	1,41E-05	3,52E-07	7,17E-07	6,28E-02	4,04E-03	6,70E-04	1,37E-03	6,35E-02	5,40E-03
		Cu	2,25E-04	1,45E-05	3,60E-07	7,35E-07	5,63E-03	3,62E-04	3,60E-05	7,35E-05	5,66E-03	4,35E-04
		Cd	2,15E-05	1,38E-06	3,44E-08	7,01E-08	2,15E-02	1,38E-03	3,44E-03	7,01E-03	2,49E-02	8,39E-03

Table 9 (continued)

	ADDDing		ADDdermal		HQing		HQdermal		HI = $\sum$ HQ	
	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult
Ni	1,75E-04	1,13E-05	2,80E-07	5,71E-07	8,76E-03	5,63E-04	5,19E-06	1,06E-05	8,76E-03	5,74E-04
Zn	2,61E-04	1,68E-05	4,17E-07	8,51E-07	8,69E-04	5,59E-05	6,96E-06	1,42E-05	8,76E-04	7,01E-05

Table 10 Carcinogenic risk level classification (Rapant et al. 2010)

			ADDDing		ADDdermal		SF	CRing		CRder		CR	
			Ch.	Ad.	Ch.	Ad.		Ch.	Ad.	Ch.	Ad.	Ch.	Ad.
C1	A1	Fe	9,52E-02	1,02E-02	1,52E-04	3,11E-04							
		Pb	3,55E-03	2,28E-04	5,68E-06	1,16E-05	8,50E-03	3,02E-05	1,94E-06	4,83E-08	9,85E-08	3,02E-05	2,04E-06
		Cu	3,27E-03	2,10E-04	5,23E-06	1,07E-05							
		Cd	8,66E-04	5,56E-05	1,39E-06	2,82E-06	6,30E+00	5,45E-03	3,51E-04	8,73E-06	1,78E-05	5,46E-03	3,68E-04
		Ni	1,65E-03	1,06E-04	2,64E-06	5,38E-06	8,40E-01	1,38E-03	8,90E-05	2,22E-06	4,52E-06	1,39E-03	9,35E-05
		Zn	6,90E-03	4,44E-04	1,10E-05	2,25E-05							
	A2	Fe	9,38E-02	1,00E-02	1,50E-04	3,06E-04							
		Pb	2,66E-03	1,71E-04	4,25E-06	8,67E-06	8,50E-03	2,26E-05	1,45E-06	3,62E-08	7,37E-08	2,26E-05	1,53E-06
		Cu	1,59E-03	1,02E-04	2,54E-06	5,18E-06							
		Cd	2,33E-04	1,50E-05	3,73E-07	7,62E-07	6,30E+00	1,47E-03	9,45E-05	2,35E-06	4,80E-06	1,47E-03	9,93E-05
		Ni	9,26E-04	5,95E-05	1,48E-06	3,02E-06	8,40E-01	7,77E-04	5,00E-05	1,24E-06	2,54E-06	7,79E-04	5,25E-05
		Zn	2,58E-03	1,66E-04	4,13E-06	8,42E-06							
	A3	Fe	9,12E-02	9,77E-03	1,46E-04	2,97E-04							
		Pb	1,04E-03	6,72E-05	1,67E-06	3,41E-06	8,50E-03	8,88E-06	5,71E-07	1,42E-08	2,90E-08	8,90E-06	6,00E-07
		Cu	6,97E-04	4,48E-05	1,12E-06	2,27E-06							
		Cd	3,34E-05	2,15E-06	5,34E-08	1,09E-07	6,30E+00	2,10E-04	1,35E-05	3,36E-07	6,86E-07	2,11E-04	1,42E-05
		Ni	4,22E-04	2,71E-05	6,75E-07	1,38E-06	8,40E-01	3,55E-04	2,28E-05	5,67E-07	1,16E-06	3,55E-04	2,39E-05
		Zn	1,03E-03	6,64E-05	1,65E-06	3,37E-06							
	A4	Fe	9,29E-02	9,95E-03	1,49E-04	3,03E-04							
		Pb	7,89E-04	5,07E-05	1,26E-06	2,57E-06	8,50E-03	6,71E-06	4,31E-07	1,07E-08	2,19E-08	9,74E-10	1,04E-10
		Cu	6,69E-04	4,30E-05	1,07E-06	2,18E-06							
		Cd	6,88E-05	4,42E-06	1,10E-07	2,24E-07	6,30E+00	4,33E-04	2,79E-05	6,93E-07	1,41E-06	3,55E-08	3,81E-09
		Ni	3,70E-04	2,38E-05	5,92E-07	1,21E-06	8,40E-01	3,11E-04	2,00E-05	4,98E-07	1,01E-06	5,72E-08	6,13E-09
		Zn	9,79E-04	6,29E-05	1,57E-06	3,19E-06							
C2	A1	Fe	5,66E-02	3,64E-03	9,06E-05	1,85E-04							
		Pb	5,73E-04	3,68E-05	9,17E-07	1,87E-06	8,50E-03	4,87E-06	3,13E-07	7,79E-09	1,59E-08	4,88E-06	3,29E-07
		Cu	6,01E-04	3,86E-05	9,62E-07	1,96E-06							
		Cd	1,51E-04	9,68E-06	2,41E-07	4,91E-07	6,30E+00	9,49E-04	6,10E-05	1,52E-06	3,10E-06	9,50E-04	6,41E-05
		Ni	4,12E-04	2,65E-05	6,59E-07	1,34E-06	8,40E-01	3,46E-04	2,23E-05	5,54E-07	1,13E-06	3,47E-04	2,34E-05
		Zn	5,96E-04	3,83E-05	9,53E-07	1,94E-06							
	A2	Fe	5,55E-02	3,57E-03	8,88E-05	1,81E-04							
		Pb	2,22E-04	1,43E-05	3,56E-07	7,25E-07	8,50E-03	1,89E-06	1,21E-07	3,02E-09	6,16E-09	1,89E-06	1,28E-07
		Cu	3,66E-04	2,35E-05	5,86E-07	1,19E-06							
		Cd	2,59E-05	1,67E-06	4,15E-08	8,46E-08	6,30E+00	1,63E-04	1,05E-05	2,61E-07	5,33E-07	1,64E-04	1,10E-05
		Ni	2,31E-04	1,49E-05	3,70E-07	7,55E-07	8,40E-01	1,94E-04	1,25E-05	3,11E-07	6,34E-07	1,95E-04	1,31E-05
		Zn	3,60E-04	2,31E-05	5,76E-07	1,17E-06							
	A3	Fe	5,30E-02	3,41E-03	8,48E-05	1,73E-04							
		Pb	3,10E-04	1,99E-05	4,96E-07	1,01E-06	8,50E-03	2,63E-06	1,69E-07	4,21E-09	8,59E-09	2,64E-06	1,78E-07

Table 10 (continued)

	ADD _{ing}		ADD _{dermal}		SF	CR _{ing}		CR _{der}		CR	
	Ch.	Ad.	Ch.	Ad.		Ch.	Ad.	Ch.	Ad.	Ch.	Ad.
Cu	1,97E-04	1,27E-05	3,16E-07	6,44E-07							
Cd	4,83E-06	3,11E-07	7,73E-09	1,58E-08	6,30E+00	3,04E-05	1,96E-06	4,87E-08	9,93E-08	3,05E-05	2,06E-06
Ni	1,06E-04	6,78E-06	1,69E-07	3,44E-07	8,40E-01	8,86E-05	5,70E-06	1,42E-07	2,89E-07	8,88E-05	5,99E-06
Zn	2,38E-04	1,53E-05	3,81E-07	7,78E-07							
A4 Fe	4,93E-02	3,17E-03	7,89E-05	1,61E-04							
Pb	1,56E-04	1,00E-05	2,49E-07	5,08E-07	8,50E-03	1,32E-06	8,51E-08	2,12E-09	4,32E-09	1,37E-09	1,47E-10
Cu	2,00E-04	1,28E-05	3,19E-07	6,51E-07							
Cd	7,64E-06	4,91E-07	1,22E-08	2,49E-08	6,30E+00	4,81E-05	3,10E-06	7,70E-08	1,57E-07	9,95E-08	1,07E-08
Ni	9,26E-05	5,95E-06	1,48E-07	3,02E-07	8,40E-01	7,77E-05	5,00E-06	1,24E-07	2,54E-07	1,08E-07	1,16E-08
Zn	1,91E-04	1,23E-05	3,05E-07	6,22E-07							

C2, cancer risk levels have been ranged between 1.47×10^{-10} and 9.50×10^{-4} , indicating very low to medium risk. Moreover, during C1, area which has been the most exposed and nearest to nearby industrial emission, namely A1, has recorded higher cancer risk levels than other ones. Furthermore, the total carcinogenic risk levels from Cd and Ni have been higher than those from other metals, indicating infrequently serious cancerogenic risk on local children and adults. Referring to the International Agency for Research on Cancer (IARC), Cd and Ni are classified as class 1 carcinogens [93], this suggest that even though the presence of a single heavy metal does not pose a significant risk, the presence of multiple heavy metals could be harmful to human health and should arouse wide concern especially where most population could be exposed.

It is noteworthy to insist here that; the computed carcinogenic risk has shown serious risk from heavy metals contents in dry atmospheric deposits via different exposure pathways in nearby areas to the main industrial plants under meteorological conditions advantageous to heavily dry atmospheric. As for distant areas, less exposed to nearby industrial plumes, calculated carcinogenic risk levels have been shown medium to very low. While such low risk levels, the possibility of serious health effects caused by accumulation of such metals in body tissues persists (Shabbaj et al. 2017).

Conclusion

This study has been conducted to investigate the concentrations, spatial-temporal variation and health risk implication of human exposure to heavy metals namely Fe, Pb, Cu, Cd, Ni and Zn in dry atmospheric deposits weekly collected from twenty sampling sites from Sfax solar saltern during two consecutive sampling campaigns namely C1 and C2. Such campaigns have revealed generally similar meteorological

characterization, excepting wind directions yielding different exposure frequencies of such area to the nearby industrial plumes. Only iron concentrations have been always lower than crust background concentrations. As for Pb, Cu, Cd, Ni and Zn average concentrations in dry atmospheric deposits have been almost higher than crust average concentrations, thus indicating that pollution is the result of anthropogenic inputs. Heavy metal concentrations in dry atmospheric deposits collected from southern coastal area of Sfax have been generally at medium or higher levels than those recorded in other areas around the world. Spatial distribution has demonstrated that the nearest and most exposed sites to industrial plumes of local pollutant sources as well as to nearby road dust have recorded highest heavy metal concentrations in dry atmospheric deposits. The study of temporal variation has revealed that recorded concentrations of selected heavy metals during C1 have been higher than those recorded during C2, excepting for iron. Such results have exposed the significant impact of exposure frequencies of sampling sites to the nearby industrial plumes. Statistical approaches applied to analyzed metals based on PCA and HCA have revealed natural origin of Fe and anthropogenic one of Pb, Cu, Ni, Cd and Zn, and a notable statistical similarity between Pb and Ni suggesting their common origin. Accordingly, such statistical approaches applied to the twenty sampling sites have revealed a clear spatial distribution allowing signaling the importance of exposure frequencies of selected sites to the industrial plumes and their locations compared to those industrial sources.

The human health risk assessment has been conducted through computed ADD, HQ, HI and CR. Calculated ADDs have revealed that ingestion has been the main exposure pathway to heavy metals in atmospheric deposits for both children and adults. In addition, calculated ADD_{ing} and ADD_{dermal} during C1 have been higher than C2. Computed HQs and HIs for children have been higher than those for adults, indicating

thus that children may be subjected to potentially higher health risk than adults especially in areas more exposed to local industrial plumes. Additionally, HQs of selected heavy metals for ingestion have been higher than those for dermal contact for both children and adults. Moreover, HQs values calculated for C1 have been higher than those calculated for C2, suggesting a potential health risk by ingestion and/or inhalation during the former more likely than during the latter. HI values for all metals have revealed that local inhabitants may be subjected to potentially higher health risk during C1 than during C2. Based on HI values, Pb and Cd have exhibited higher values close to safe level, infrequently exceeding such level during C1, indicating a significant risk for children due to their exposure. Calculated carcinogenic risks through ingestion mode of exposure (CRAing) and dermal contact (CRAder) as well as the total carcinogenic risk for selected heavy metals have shown that cancer could occur more probably through ingestion than dermal contact for children further than adult. Moreover, carcinogenic risk levels during C1, recording infrequently very high risks, have been generally higher than during C2, revealing frequently low to medium risks. Computed total carcinogenic risk levels from Cd and Ni have been higher than those from other metals, indicating higher cancerogenic risk during C1 than C2, on local children and adults.

This study suggests that more attention should be paid to toxic heavy metals that long-term exposure to which via atmospheric particulate deposition could have adverse effects on the health of city dwellers where to-date only limited research has been undertaken. In fact, in order to reduce carcinogenic risk through ingestion and dermal contact of heavy metals contents in dry atmospheric deposits for workers at Sfax solar saltern, some preventive measures should be considered: (i) Using personal protective equipment (PPE) such as: hard hat, ear protection, gloves, respirator and coveralls for workers; (ii) Reduce the number of exposed workers through temporary exceptional technical solution when exposure to heavy metals contents in atmospheric deposits seem to be important; and (iii) Maintain a rigorous control of industrial emission sources in terms of heavy metals especially in the case of enterprises which do not respect pollutants standards limits.

Nevertheless, it is preferably to mention that limitations of the current study lie in a small number of samples on the one hand and other metals affecting human body have not been assessed, on the other hand. Also, used risk assessment model in the current study reveals some limitations: (i) limited investigation of the exposure patterns (e.g. frequency and duration); (ii) the risk of exposure to heavy metal mixtures could not be calculated and (iii) information gap about population assess and life stages (e.g. highly exposed population, vulnerable, sensitive or susceptible groups) still not considered.

Despite these facts, application of the described risk model helped us get a better vision of probable health risks to residents and workers in Sfax southern areas.

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Compliance with ethical standards

Conflict of interests The author declare that there is no conflict of interests regarding the publication of this paper.

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