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# The Trend of Trace Elements (Cd, Ni, Pb) from $PM_{2.5}$ and $PM_{10}$ Aerosols and its Effect on Human Health in Bucharest, Romania

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***Abstract****. The comprehensive investigation of the elemental characteristics in fine and coarse particles at Bucharest was carried out. The daily samples of $PM_{2.5}$ and $PM_{10}$ particulate matter were collected at eight monitoring stations for a one-year period, and concentrations of Cd, Ni, Pb elements were analyzed. The results show that PMs and trace elements were present in high concentrations in Bucharest. The annual concentrations of $PM_{10}$ and $PM_{2.5}$ were 31.57 $\mu g/m^3$ and 21.30 $\mu g/m^3$, respectively. In Bucharest, the average concentration ratio of fine ($PM_{2.5}$) and coarse particulates ($PM_{10}$) was 0.67. Trace elements concentration carried by the $PM_{2.5}$ was higher than the concentration detected from $PM_{10}$. The Cd, Ni, and Pb accumulation was higher by 55.16%, 37.46% and 29.14% in fine particles than in coarse particles. The annual mean trace element concentration from $PM_{2.5}$ and $PM_{10}$ was in the case of Cd 0.22/0.24 ng/$m^3$, for Ni 3.28/3.14 ng/$m^3$ and for Pb 5.61/6.44 ng/$m^3$, respectively. The highest Spearman correlation was found between Cd and Ni with correlation coefficient of 0.62 in $PM_{2.5}$ and 0.48 in $PM_{10}$, which suggests that they share common sources. The health risk indexes were estimated for both adults and children thanks to the trace elements from the particulate matter (PM), and the results revealed that inhalation was the major exposure pathway in both cases.*

***Keywords****: $PM_{2.5}$, $PM_{10}$, trace elements (Cd, Ni, Pb), health effect*

## 1.Introduction

Particles (PM) with an aerodynamic diameter less than 10 μm lead to many premature deaths worldwide and contribute to an increase in cardiovascular disease [1,2]. In addition fine particles ($PM_{2.5}$, particles ≤ 2.5 μm in aerodynamic diameter) carries greater danger to human health than the coarse particles ($PM_{10}$)[3]. Trace elements (TEs) generally represent a small fraction of atmospheric particles (PM), but they nevertheless have a significant impact on the environment and human health [4,5]. Toxic trace elements such as As, Ni, Cd, Pb are considered as carcinogens with potential health risks [6]. There are three possible routes of exposure to toxic elements: ingestion, inhalation, and skin absorption, and have an accumulation potential in the biological systems, especially in the fatty tissues [7]. Therefore, the elemental characterization of PM particles of different sizes is of great concern. Cd and Ni compounds in particulate matter, mainly originate from coal and fuel oil combustion processes, metallurgical industry, and road transport, respectively [8]. A number of statistical techniques, including correlation matrix, and health effects calculation were utilized to identify the influences of diverse emission activities. The results provide useful insights for developing effective emission reduction strategies. The trace elements composition in $PM_{10}$ has been studied by a number of studies worldwide, in Romania by [9-15] in Europe by [4, 16–18] and in other continents by [7, 19–22].

## 2. Materials and methods

Daily trace element concentrations (Cd, Ni, Pb) were determined in $PM_{2.5}$ and $PM_{10}$, which were sampled in Bucharest, the capital of Romanian, between January 2018 and December 2018. Bucharest is the most populated city in Romania has over 2 million inhabitants covering a surface of 228 $km^2$ with coordinate: latitude 44.4268 °N and longitude: 26.1025 °E. Bucharest has a humid continental climate with an average temperature of 12°C

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[23]. The main $PM_{2.5}$ and $PM_{10}$ pollution in Bucharest comes from intense traffic and industrial activities (CETs) emissions [24]. The daily $PM_{2.5}$, $PM_{10}$ and trace elements concentration data were obtained from the National Air Quality Monitoring Network. Regarding the monitoring stations ($B_1$-$B_8$), the trace elements in the case of $B_1$ were determined from $PM_{2.5}$ and at the remaining stations the data were obtained from $PM_{10}$fraction.

The temporal variation of daily concentrations of $PM_{2,5}$ and $PM_{10}$ were statistically analyzed, hence the most important correlations were identified. Spearman correlation coefficient (Ri386 3.5.3.) was used to determine the correlation between PM and trace elements (Cd, Ni, Pb) concentrations.

To determine the exposure levels of trace elements via inhalation, ingestion and dermal absorption the health risk model developed by the US Environmental Protection Agency (EPA) was used [25]. The quantitative assessment of the cancer risk (CR) was carried out for adults and children. Risk characterization was quantified separately for carcinogenic and non-carcinogenic effects. The non-carcinogenic risk was assessed by the hazard quotient (HQ). HQ and carcinogenic risks (CR) posed by heavy metals in $PM_{10}$ via ingestion, inhalation and dermal contact were calculated. Values below 1E-06 for individual chemicals and pathways show negligible cancer risks.

$$CDI_{ingest}=(C*IngR*EF*ED*CF)/(BW*AT) \quad (1)$$

$$EC_{inh}=(C*ET*EF*ED)/AT \quad (2)$$

$$DAD_{derm}=(C*SA*AF*EV*ABS*EF*ED*CF)/(BW*AT) \quad (3)$$

where, C: metal concentrations in $PM_{10}$ ($\mu g/m^3$); IngR- ingestion rate (200 mg/day for children and 100 mg/day for adults); EF- exposure frequency (250 days $year^{-1}$); ED- exposure duration (6 years for children and 24 years for adults); BW- the average body weight (15 kg for children and 70 kg for adults); AT- averaging time (for non-carcinogens, AT=ED×365 days; for carcinogens, AT=70×365=25,550 days); CF- conversion factor ($10^{-6}$ kg $mg^{-1}$); SA- surface area of the skin that contacts the airborne particulates (3300 $cm^2$ for adults and 2800 $cm^2$ for children); AF- skin adherence factor for the airborne particulates (0.2 mg $cm^{-2}$ for both adults and children); ABS: dermal absorption factor (0.001 for Cd and 0.01 for other elements); ET- exposure time (24 h $day^{-1}$); $AT_n$: average time (for non-carcinogens, AT=ED×365 days ×24 h/day; for carcinogens, $AT_n$=70 year×365 days/year×24h). All parameters used in the calculation of CDI, DAD, and EC are shown in Table 1 including the SFo, RfDo, RfC, GIABS and IUR values [25].

**Table 1.** Parameters used in cancer non-carcinogen risk assessment

| | SFo | RfDo | GIABS | IUR | RfCi |
|---|---|---|---|---|---|
| | $(mgkg^{-1}day^{-1})^{-1}$ | $(mgkg^{-1}day^{-1})^{-1}$ | | $(\mu gm^{-3})^{-1}$ | $(mgm^{-3})$ |
| Cd | | $1.00E^{-03}$ | 0.025 | $1.80E^{-03}$ | $1.00E^{-05}$ |
| Ni | | $1.10E^{-02}$ | 0.040 | $2.60E^{-04}$ | $2.00E^{-05}$ |
| Pb | $2.80E^{-01}$ | $3.50E^{-03}$ | 1 | $1.20E^{-05}$ | |

Where, RfDo- oral reference dose (mg $kg^{-1}$ $day^{-1}$); RfCi- inhalation reference concentrations ($mgm^{-3}$); SFo- oral slope factor (mg $kg^{-1}$ $day^{-1})^{-1}$); GIABS- gastrointestinal absorption factor; IUR: inhalation unit risk ($mgm^{-3})^{-1}$). The SFo and/or IUR values for Cd (diet), Ni (refinery dust) and Pb (acetate) were used. The quantitative assessment of the cancer risk (CR) was carried out for adults and children living in Bucharest. In the Bucharest region, the chemical components considered for the evaluation included: Cd, Ni, and Pb. Risk characterization was quantified separately for carcinogenic and non-carcinogenic effects.

$$HQ_{ing}=CDI/RfDo \quad (4)$$

$$HQ_{inh}=EC/(RfCi*1000\ \mu g\ mg^{-1} \quad (5)$$

$$HQ_{derm}=DAD/(RfDo*GIABS) \quad (6)$$

$$CR_{ingest}=CDI*SFo \quad (7)$$

$$CR_{inh}=IUR*EC \quad (8)$$

$$CR_{derm}=DAD*(SFo/GIABS) \quad (9)$$

The non-carcinogenic risk was assessed by the hazard quotient (HQ). HQ and carcinogenic risks (CR) posed by heavy metals in $PM_{10}$ via ingestion, inhalation and dermal contact were calculated using the above-mentioned equations.

## 3.Results and discussions

The ambient atmospheric concentrations of $PM_{2.5}$, $PM_{10}$ and trace elements at the Bucharest region are summarized in Table 2. Using descriptive statistics the mean, minimum, median, maximum, $25^{th}$- and $75^{th}$ percentile values, and confidence intervals were calculated. During the studied period, the average concentration of $PM_{10}$ in Bucharest was 31.57±1.33 μg/m$^3$ which is 57.85% higher than the WHO's acceptable limit (20 μg/m$^3$ WHO, 2005). In the case of $PM_{2.5}$, the annual concentration was also 113% higher than the admissible level (10 μg/m$^3$). Overall, the Pb and Ni contributions were the highest.

According to the WHO guidelines the maximum annual average safe level of heavy metals is 0.5 μg/m$^3$ for Pb, 5 ng/m$^3$ for Cd, and 20 ng/m$^3$ for Ni, respectively.

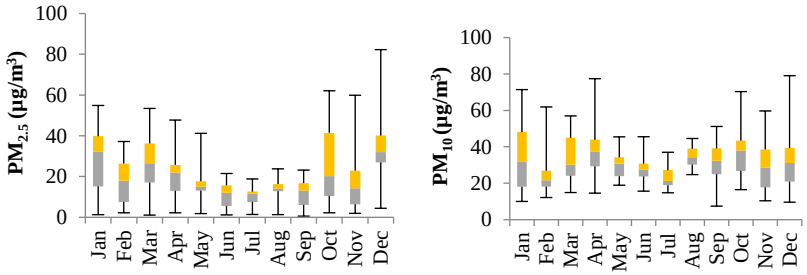
In Bucharest, the average concentration ratio of $PM_{2.5}$ and $PM_{10}$ was 0.67, meaning that for the same concentrations of $PM_{10}$ and $PM_{2.5}$, trace element enrichment was significantly higher in $PM_{2.5}$ than in $PM_{10}$. The Ni, Cd and Pb accumulation in fine particles was higher than in coarse particles by 55.16%, 37.46%, and 29.14%, respectively.

**Table 2.** Trace elements concentration from $PM_{2.5}$ and $PM_{10}$

| D.U. | | | mean | min | P25 | P50 | P75 | Max | Count | 95%CI |
|---|---|---|---|---|---|---|---|---|---|---|
| Cd | [ng/m$^3$] | $PM_{2.5}$ | 0.22 | 0.00 | 0.02 | 0.08 | 0.29 | 2.55 | 338 | 0.18-0.26 |
| | | $PM_{10}$ | 0.24 | 0.00 | 0.10 | 0.17 | 0.31 | 1.61 | 364 | 0.21-0.26 |
| Ni | [ng/m$^3$] | $PM_{2.5}$ | 3.28 | 0.00 | 0.96 | 2.62 | 5.06 | 12.53 | 338 | 2.98-3.58 |
| | | $PM_{10}$ | 3.14 | 0.16 | 1.90 | 3.06 | 4.12 | 7.91 | 365 | 2.98-3.28 |
| Pb | [ng/m$^3$] | $PM_{2.5}$ | 5.61 | 0.00 | 1.00 | 4.00 | 8.00 | 33.00 | 311 | 4.96-6.25 |
| | | $PM_{10}$ | 6.44 | 0.40 | 3.71 | 5.85 | 8.71 | 19.71 | 364 | 6.06-6.8 |
| PM | [μg/m$^3$] | $PM_{2.5}$ | 21.30 | 0.59 | 12.22 | 17.03 | 28.28 | 82.03 | 357 | 19.90-22.70 |
| | | $PM_{10}$ | 31.57 | 7.45 | 22.51 | 29.64 | 38.69 | 79.07 | 365 | 30.24-32.90 |

**Temporal variations**

The temporal variations of trace element concentrations in $PM_{2.5}$ and $PM_{10}$ at Bucharest are presented separately (Figure 1). Overall, the trace elements presented similar temporal patterns in both sizes. On the other hand, time series analyzes revealed drastic concentration variations for Cd, Ni, and Pb, mainly higher during warm period and significantly lower in the cold period (December-February) which is strongly correlate with the traffic intensity (higher in summer). In early spring thanks to the stronger wind speed and lower vegetation favored the suspension of soil/road dust. In the case of particulate matter, annual evolution shows the highest peak concentrations in winter and a minimum level in summer, thanks to the meteorological variables [26-29].



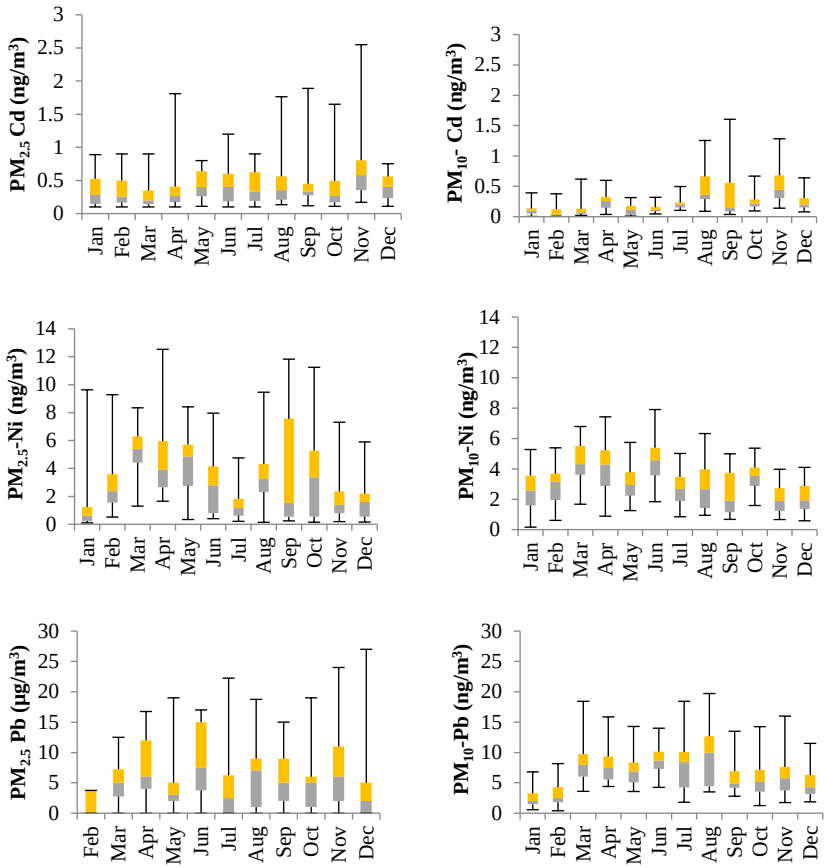



**Figure 1.** Monthly variation of PMs and trace elements concentration

## Spearman correlation

Spearman correlation analysis was used to investigate the correlation between all the trace elements from $PM_{2.5}$ and $PM_{10}$ (Figure 3). The monthly average concentrations were used for the correlation analysis and P-values of 0.05 (5%) were considered statistically significant. Trace elements originated from the fine particulars show a higher correlation level than those from the coarse particulate. The Spearman correlation between monthly means of PM's and trace element concentration in 2018 has been studied separately for the $B_1$ monitoring station ($PM_{2.5}$) and the $B_{2\text{-}8}$ ($PM_{10}$) respectively. In the $B_1$ monitoring station, a higher correlation was found between the Cd and Ni (r=0.62). A moderate positive correlation was detected between the Pb and Ni (r=0.45). A moderate negative correlation was identified between Ni and $PM_{2.5}$ (r=-0.42) and between Pb and $PM_{2.5}$ (r=-0.45) as well, which suggests that the source origin of trace elements differs from $PM_{2.5}$ sources.

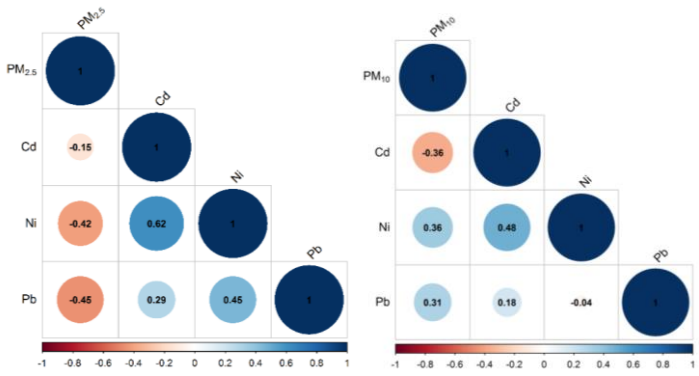



**Figure 2.** Spearman correlations between the trace elements in $PM_{2.5}$ and $PM_{10}$

Furthermore, at $B_{2-8}$ monitoring stations, the correlation level between the trace elements and $PM_{10}$ concentration was less significant than in $PM_{2.5}$. Between the Cd and Ni, the correlation coefficient was 0.48 which suggests common sources. In contrast to $PM_{2.5}$, positive moderate correlation was found between the $PM_{10}$, Ni (r=0.36) and Cd (r= 0.31).

## Health risk assessment by toxic metals in PM $_{2.5}$ and $PM_{10}$

### Non-carcinogenic risk assessment

Inhalation is the primary route of direct exposure to particulate bound trace elements. The HQ values for the three exposure pathways are presented in Table 3. The HQ values for dermal and ingestion exposure for both groups, adults, and children, in Bucharest regions, were lower than the safe limit (=1). In the case of ingestion and dermal absorption, significant differences were observed between children and adults thanks to their behavior and physiological activities[30]. The HQ value for Pb could not be calculated for the reason that US EPA does not establish the threshold value (RfCi) for different exposure routes. Via inhalation, the HQ value for Ni in $PM_{2.5}$ was higher than the safe limit (=1) in both cases, adults (2.70E+00) and children (2.70E+00). Regarding the HQ value from $PM_{10}$ similar tendency was obtained (2.65E+00), indicating that Ni might pose a non-carcinogenic risk to adults and children.

**Table 3**. Hazard Quotient (HQ) non-carcinogen cases for metals in $PM_{2.5}$ and $PM_{10}$ via ingestion, inhalation and dermal contact for children and adults

| HQ | | Ingestion | | Inhalation | | Dermal | |
|---|---|---|---|---|---|---|---|
| | | Children | Adults | Children | Adults | Children | Adults |
| $PM_{2.5}$ | Cd | 2.05E-06 | 2.20E-07 | 3.69E-01 | 3.69E-01 | 2.71E-07 | 4.92E-08 |
| | Ni | 2.73E-06 | 2.92E-07 | 2.70E+00 | 2.70E+00 | 2.25E-06 | 4.09E-07 |
| | Pb | 1.77E-05 | 1.90E-06 | | | 5.84E-07 | 1.06E-07 |
| $PM_{10}$ | Cd | 2.23E-06 | 2.39E-07 | 4.02E-01 | 4.02E-01 | 2.95E-07 | 5.36E-08 |
| | Ni | 2.68E-06 | 2.87E-07 | 2.65E+00 | 2.65E+00 | 2.21E-06 | 4.01E-07 |
| | Pb | 1.78E-05 | 1.91E-06 | | | 5.88E-07 | 1.07E-07 |
| HI | | 4.52E-05 | 4.84E-06 | 6.12E+00 | 6.12E+00 | 6.19E-06 | 1.13E-06 |

The HI (sum of all the elements under consideration) value increased to 6.12E+00, indicating the accumulative non-carcinogenic health risk due to inhalation exposure of the mixture of elements.

### Cancer risk assessment

For the investigation of carcinogen risks in $PM_{2.5}$ and $PM_{10}$ three trace elements (Cd, Ni, Pb) were taken into consideration, because these are classified as carcinogen/probably carcinogenic/possibly carcinogenic to human health according to IARC (International Agency for Research on Cancer).

The carcinogen risk via inhalation for adults, in all examined trace elements, was lower than the acceptable limit (1E-06). Adults had a higher probability of experiencing carcinogenic risk compared to children. The HI value increased to 1.52E-05 indicating the accumulative cancer risk due to inhalation exposure of the mixture of elements (Table 4.).

In order to figure out the concentrations level, the values obtained in Bucharest from the $PM_{2.5}$ and $PM_{10}$ were compared with those measured from different parts of the world (Table 5.).

**Table 4**. Cancer risk (CR) results obtained for carcinogenic elements in $PM_{2.5}$ and $PM_{10}$ via ingestion, inhalation and dermal contact for children and adults

| CR | | Ingestion | | Inhalation | | Dermal | |
|---|---|---|---|---|---|---|---|
| | | Children | Adults | Children | Adults | Children | Adults |
| $PM_{2.5}$ | Cd | 1.07E-09 | 4.60E-10 | 5.70E-07 | 2.28E-06 | 1.42E-10 | 1.03E-10 |
| | Ni | 4.37E-09 | 1.87E-09 | 1.20E-06 | 4.81E-06 | 3.61E-09 | 2.62E-09 |
| | Pb | 1.49E-09 | 6.37E-10 | 1.15E-07 | 4.59E-07 | 4.91E-11 | 3.57E-11 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | Cd | 1.17E-09 | 5.00E-10 | 6.20E-07 | 2.48E-06 | 1.54E-10 | 1.12E-10 |
| $PM_{10}$ | Ni | 4.29E-09 | 1.84E-09 | 1.18E-06 | 4.72E-06 | 3.54E-09 | 2.57E-09 |
| | Pb | 1.50E-09 | 6.41E-10 | 1.15E-07 | 4.62E-07 | 4.94E-11 | 3.59E-11 |
| HI | | 1.39E-08 | 5.95E-09 | 3.80E-06 | 1.52E-05 | 7.54E-09 | 5.48E-09 |

**Table 5**.$PM_{10}$ (μg/m$^3$) and trace elements concentration (ng/m$^3$) in different countries around the world

| | **Present study** | | **India** | | **USA** | | **Taiwan** | | **Spain** | |
|---|---|---|---|---|---|---|---|---|---|---|
| | $PM_{2.5}$ | $PM_{10}$ | $PM_{2.5}$ | $PM_{10}$ | $PM_{2.5}$ | $PM_{10}$ | $PM_{2.5}$ | $PM_{10}$ | $PM_{2.5}$ | $PM_{10}$ |
| Cd | 0.22 | 0.245 | 6.6 | | 0.177 | | 0.7 | | 0.32 | |
| Ni | 3.28 | 3.14 | 29.3 | | 8.67 | | 9.84 | | 2.29 | |
| Pb | 5.61 | 6.44 | 85.2 | | 3.61 | | 21.2 | | 13.14 | |
| PM | 21.3 | 31.57 | | 216 | | 57.59 | | 52.4 | | 41.96 |
| Source reference | | | | [27] | | [28] | | [3] | | [29] |

Comparatively, it can be said that the Cd concentration (0.22/0.24 ng/m$^3$) is much higher than in the USA, Spain, and Taiwan, respectively. The Ni concentration (3.28/3.14 ng/m$^3$) is higher than in Spain. The Pb concentration in the Bucharest region (5.61/5.44 ng/m$^3$) is between the USA and Spain (lower in the USA and higher in Spain).

## 4.Conclusions

The time-series analysis results show that the annual mean concentration of $PM_{2.5}$ and $PM_{10}$ was 21.30 μg/m$^3$ and 31.57 μg/m$^3$, respectively. These concentrations exceeded the annual mean standard established by the European Union ($PM_{2.5-10}$ μg/m$^3$ and $PM_{10}$- 20 μg/m$^3$). Although the $PM_{10}$ concentration has decreased since the implementation of the European environmental legislation, the daily limit values exceeded the maximum permissible levels almost continuously; therefore Bucharest is under the infringement procedure regarding the air quality. Briefly, all trace elements in fine particles ($PM_{2.5}$) were higher enriched than in coarse particles ($PM_{10}$). The mean metal concentration decreased in the following order: Pb>Ni>Cd. The seasonal variation in PM concentration was distinguishable; the highest concentration was observed during the winter season, followed by autumn, spring, and summer, respectively. Regarding the trace element distribution over the years, the maximum peak was found in the warm season and minimum in the cold season, thanks to the traffic intensity. Health hazard assessment identified inhalation as the leading pathway of particle-bound trace element exposure to humans. The multi-elemental hazard index (HI) indicated greater health risks in case of mixture of elements.

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