

Determination of potentially toxic metals and natural radionuclides in airborne pollens produced different urban environments in Turkey and health risk assessment

Şeref Turhan, Talip Çeter, Ergin Murat Altuner, Serhat Karabıcak, Selin Çeter, Oktay Biyıklıoğlu, Şeymanur Aktaş & Aslı Kurnaz

To cite this article: Şeref Turhan, Talip Çeter, Ergin Murat Altuner, Serhat Karabıcak, Selin Çeter, Oktay Biyıklıoğlu, Şeymanur Aktaş & Aslı Kurnaz (2025) Determination of potentially toxic metals and natural radionuclides in airborne pollens produced different urban environments in Turkey and health risk assessment, International Journal of Environmental Health Research, 35:5, 1296-1313, DOI: [10.1080/09603123.2024.2391460](https://doi.org/10.1080/09603123.2024.2391460)

To link to this article: <https://doi.org/10.1080/09603123.2024.2391460>



Published online: 13 Aug 2024.



Submit your article to this journal [↗](#)



Article views: 142



View related articles [↗](#)



View Crossmark data [↗](#)



RESEARCH ARTICLE



Determination of potentially toxic metals and natural radionuclides in airborne pollens produced different urban environments in Turkey and health risk assessment

Şeref Turhan^a, Talip Çeter^b, Ergin Murat Altuner^b, Serhat Karabıçak^b, Selin Çeter^b, Oktay Biyıkloğlu^b, Şeymanur Aktaş^b and Aslı Kurnaz^a

^aDepartment of Physics, Faculty of Science, Kastamonu University, Kastamonu, Türkiye; ^bDepartment of Biology Faculty of Science, Kastamonu University, Kastamonu, Türkiye

ABSTRACT

Air pollutants are associated with potentially toxic metals (PTMs) and natural and/or artificial radionuclides, which can pose a major threat to human and environmental health. Pollens can be utilized as a bioindicator to determine the level of air pollution in urban areas. In this study, the concentrations of PTMs and natural radionuclides in 35 airborne pollen samples of 22 species belonging to Pinaceae, Cupressaceae, Araucariaceae, Betulaceae, Salicaceae, and Oleaceae families grown in different urban areas in Turkey were determined using an energy-dispersive X-ray fluorescence spectrometry. For the first time, non-carcinogenic and radiologic health risk assessments for adults were done, estimating hazard index (HI) and annual effective dose (AED), respectively. The concentrations of Fe, Mn, Zn, Ti, Sr, Cu, Ni, Co, Cr, V and Pb analyzed in airborne pollen samples varied from 52.1 to 3078.0, 26.1 to 159.6, 15.6 to 199.7, 9.1 to 282.2, 1.0 to 128.4, 5.0 to 40.1, 5.4 to 23.6, <LD to 11.3, 1.6 to 26.6, <LD to 11.1 and <LD to 5.7 mg/kg, respectively. The average concentrations of K (2%), Th (1.3 mg/kg) and U (1.0 mg/kg) were converted into the activity concentrations (in Bq/kg) of ⁴⁰K (595.8), ²³²Th (5.2) and ²³⁸U (11.8), respectively. All HI and AED values revealed a very low non-carcinogenic and radiological health risk due to exposure to all PTMs and radionuclides in the pollen samples studied.

ARTICLE HISTORY

Received 2 April 2024
Accepted 7 August 2024

KEYWORDS

Toxic element; airborne pollen; health index; natural radionuclides; annual effective dose

Introduction

One of the possible important consequences of the rapid and uncontrolled growth of population, urbanization, industry, and economy and the appearance of new technologies is an increase in emissions of atmospheric pollutants (APs) in the ambient air (Robichaud 2020). It is well known that APs can have adverse effects and harm human and environmental health. APs include a mixture of solid particles, liquid droplets, and gases from a variety of natural and anthropogenic sources such as carbon monoxide (CO), sulfur oxides (SO₂ and SO₃), nitric oxides (NO and NO₂), ozone (O₃), particulate matter (PM₁₀ and PM_{2.5}), which contains bioaerosols such as pollen, bacteria, fungal spore, viruses, etc., volatile organic compounds (VOC), potentially toxic metals (PTMs), natural and artificial radionuclides (Capone et al. 2023). High concentrations of PMs in the atmosphere, especially airborne allergens derived from plant pollen, can increase and exacerbate allergic respiratory symptoms and diseases (Oduber et al. 2019; Gisler 2021;

Capone et al. 2023). In the last few decades, respiratory diseases such as allergic rhinitis and asthma have been increasing rapidly in adults and children (Bobrowska-Korzeniowska et al. 2021; Madaniyazi and Xerxes 2021; DeWeger et al. 2021; Luschkova et al. 2022; Capone et al. 2023).

Although PTMs are natural elements of the Earth's crust, there has been a large (approximately ten-fold) increase in the accumulation of PTMs in environmental samples (soil, water, sediment, plants, air, food, etc.) due to anthropogenic activities such as mining, smelting, industry, agriculture (pesticides, insecticides, fertilizers, etc.), fuel burning, transportation, waste disposal, etc (Nriagu 1996; Łokas et al. 2019; Abbasi and Mirekhtiary 2020; Abbasi et al. 2020, 2022; Torres et al. 2023; Yoon et al. 2023; Miletić et al. 2023). In addition to their harmful effects on the environment, PTMs, which are very stable and do not biodegrade, can accumulate in human organs and cause serious damage in case of long-term exposure (Huang et al. 2019; Miletić et al. 2023). Accumulation of PTMs in organs causes various acute and chronic diseases affecting the body system, such as respiratory diseases, cardiovascular, nervous system damage, immune system, endocrine, slowing of growth development, skeletal, etc (Tan et al. 2016; Doležalová et al. 2019; Miletić et al. 2023).

Atmospheric radionuclides originate from both natural and anthropogenic sources. Natural sources include cosmogenic and terrestrial radionuclides. Terrestrial radionuclides contain radioactive potassium (^{40}K) and the radioactive series of uranium (^{238}U) and thorium (^{232}Th) (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008; Zakaly et al. 2024). It is well known that atmospheric accumulation of anthropogenic radionuclides occurred as a result of nuclear accidents and a series of major nuclear weapon tests and atmospheric explosions (Łokas et al. 2019). The primary source of artificial radionuclides in Turkey is the fallout from the Chernobyl accident (Turhan Ş, Köse and Varinlioğlu 2007). Humans are exposed to ionizing radiations (alpha-, beta-, and gamma-rays) emitted from these radionuclides in two different ways (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008; Abbasi and Mirekhtiary 2019) internal and external. Internal exposure occurs when the radionuclide enters the body via ingestion through the digestive tract, inhalation into the respiratory airways, percutaneous absorption through the skin, or contamination from a wound. External is the exposure of the entire body to gamma-ray radiation emitted by terrestrial or cosmic radionuclides. As a result of these two exposures, ionizing radiation interacts with biological tissues or cells and causes diverse damages (Nautiyal et al. 2021, Kefalati et al. 2021). The severity of these damages depends mainly on the radiation dose absorbed by body organs or tissues (Nautiyal et al. 2021). The most significant damage is DNA double-strand breaks, and DNA damage can cause genomic instability, apoptosis, and mutations in the cell (Nautiyal et al. 2021).

Numerous APs interact with pollen and can cause direct effects on pollen viability, fertility, and germination rate, properties of the pollen surface, the release of allergens/proteins from pollen, physical and chemical properties of pollen grains and its allergenic potential (Capone et al. 2023). Pollen or pollen grains also accumulate potentially toxic elements or PTMs or heavy metals and serve as indicators of air pollution (Kalbande et al. 2008; Vasilevskaya 2022). Therefore, monitoring the pollen in the air of urban areas is of interest in terms of ecological studies and investigating the level of environmental pollution caused by anthropogenic activities such as vehicle transportation, industrial emissions, etc (Main 2003; Vasilevskaya 2022). The number of plant species involved in monitoring studies is high and varies from herbaceous plants to different tree species whose pollen may or may not be allergenic (Vasilevskaya 2022). To date, many studies have been published on the interaction of APs (sulphur oxides, nitrogen, ozone, PMs, PTMs or heavy metals, and polyaromatic hydrocarbons) resulting from anthropogenic activities with different pollens and how they affect them (Xiong and Peng 2001; Wang et al. 2009; Guedes et al. 2009; Mohsenzadeh et al. 2011; Yousefi et al. 2011; Beck et al. 2013; Sénéchal et al. 2015; Kaur et al. 2016; Azzazy 2016; Robichaud 2020; Vasilevskaya 2022; Capone et al. 2023). However, a few studies have used airborne pollen as an indicator of environmental pollution caused by PTMs or heavy metals

(Bessonova 1992; Calzoni et al. 2007; Kalbande et al. 2008). According to our literature research, there is no detailed study investigating the urban air PTM and radionuclide pollution of any city in Turkey using airborne pollens.

Having all this in mind, the purpose of this study is (i) to determine the concentrations of PTMs (Fe, Mn, Zn, Ti, Sr, Cu, Ni, Co, Cr, V, and Pb) and natural radionuclides (K, Th, and U) and the corresponding activity concentration of ^{40}K , ^{232}Th and ^{238}U in 35 pollen samples of 22 species belonging to Pinaceae, Cupressaceae, Araucariaceae, Betulaceae, Salicaceae, and Oleaceae families naturally distributed and grown in parks, gardens, and recreation areas in different locations of Turkey using an energy dispersive X-ray fluorescence (EDXRF) spectrometry and (ii) to assess the non-carcinogenic human health risk associated with these PTMs through inhalation, ingestion, and dermal contacts estimating average daily dose, health quotient, and hazard index, and radiological health risk due to internal exposure estimating annual effective dose. The novelty of this study is the determination of PTMs and radionuclide levels in airborne pollen used as bioindicators for the first time in Turkey using the EDXRF spectrometric technique and the assessment of the health risks arising from these metals and radionuclides. The data obtained as a result of this study can be used to evaluate air metal pollution.

Materials and methods

Sample collection and preparation

A total of thirty-five fresh pollen samples from twenty-two species belonging to the Pinaceae, Cupressaceae, Araucariaceae, Betulaceae, Salicaceae, and Oleaceae families, which are grown in parks, gardens and recreation areas of various provinces of Turkey, were collected in field studies carried out between February and October in different years. The locations of the collected pollen samples are shown in Figure 1. For the study, male cones and male catkins collected from trees along with their branches were laid on clean acetate papers at room temperature and allowed to mature and release pollen for 3–4 days. The pollen samples obtained were passed through a series of sieves appropriate to the pollen size of the taxa to separate them from non-pollen parts. For pollen purity, preparations were prepared according to the method given by Wodehouse (1935), and pollen purity was ensured to be at least 99%. The sieved pollen samples were kept in a -20°C deep freezer until analysis. Some data from pollen samples are given in Table 1.

Potentially toxic metal analysis

Analysis of PTMs in the pollen samples was done by using an energy-dispersive X-ray fluorescence (EDXR) spectrometry (Spectro Xepos, Ametek) with a thick binary Pd/Co alloy anode X-ray tube (50 kV, 60 W), the properties of which were detailed in previous studies (Turhan et al. 2020, 2022; Altıkulaç et al. 2022; Altıkulaç and Turhan 2023). The EDXRF spectrometry has many different excitation conditions that guarantee the best detection of all elements from Na to U (Turhan et al. 2020). The spectral resolution of the system is lower than 155 eV. The EDXRF spectrometry has twelve automatic sampling devices and software to analyze samples at the same time. It uses sophisticated calibration techniques such as “no-standard” calibration, often based on the basic parameters method. NIST SRM 2709 reference material was utilized for quality assurance of the EDXRF spectrometry (Turhan et al. 2020). Each pollen sample was placed in an automatic sampler, and the analysis procedures were completed by counting for approximately one hour. The XRF spectrum of each sample was evaluated with the help of the software installed in the system. The overall uncertainty of Fe, Mn, Zn, Ti, Sr, Cu, Ni, Co, Cr, V, Pb, K, Th and U was found to be 0.3, 0.7, 0.5, 1.4, 0.5, 1.9, 3.0, 9.5, 1.8, 8.7, 4.6, 0.1, 7.7 and 10.0% respectively. The detection limits (DLs) of Fe, Mn, Zn, Ti, Sr, Cu, Ni, Co, Cr, V, Pb, Th and U were determined as 1.0, 1.0, 0.5, 2.0, 1.4, 0.5, 0.5, 3.0, 1.0, 0.8, 0.9, 0.4 and 0.2 mg/kg, respectively.

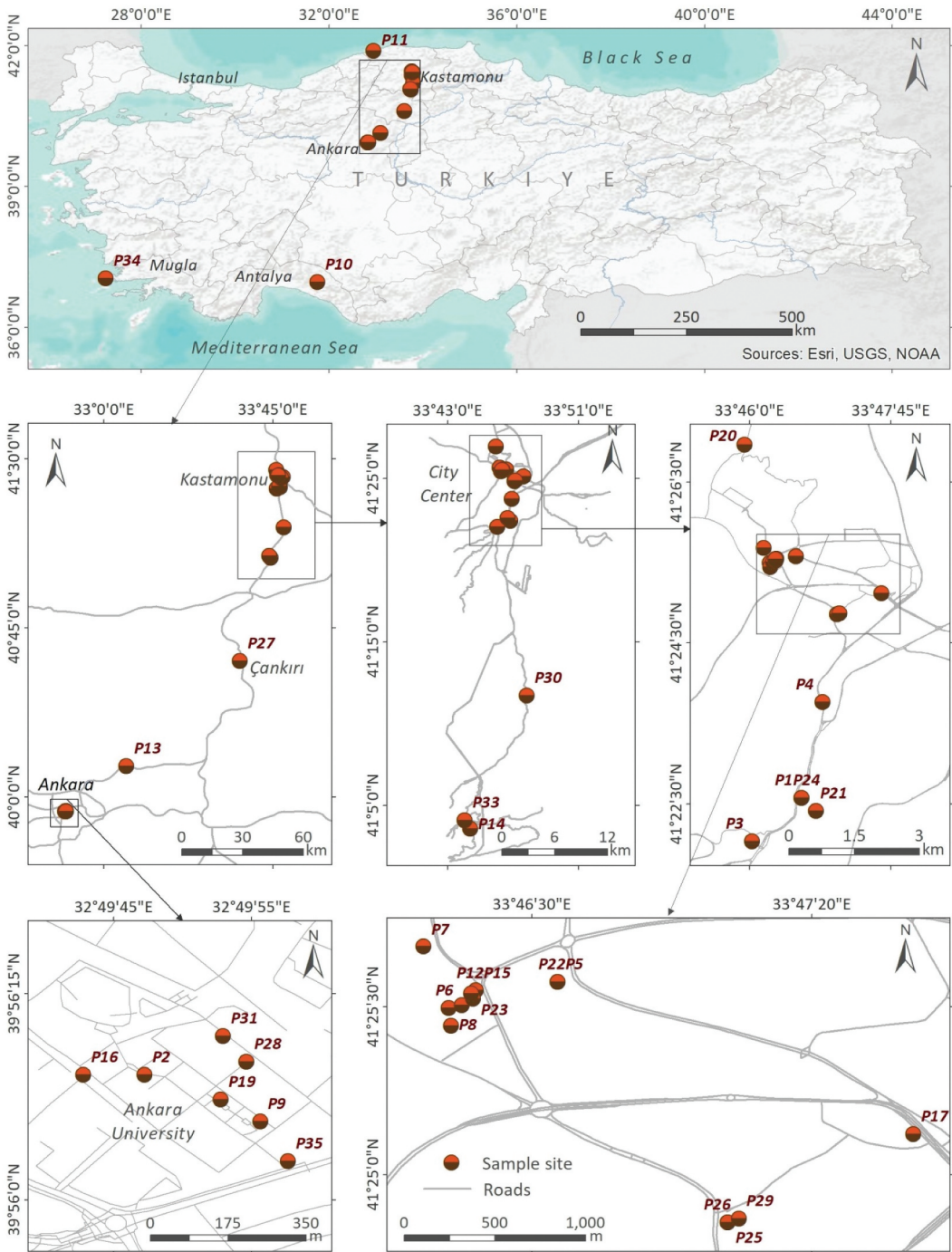


Figure 1. Map of locations of pollen samples.

Activity concentration of radionuclides

The elemental concentration of K (in %), Th (in ppm), and U (in ppm) analyzed in each pollen sample were converted into the activity concentration (A) of ^{40}K , ^{232}Th and ^{238}U using the following formula:

Table 1. Some data of collected pollen samples.

Sample code	Family	Taxon name	Male flower maturation time	Collection date
P1	Pinaceae	<i>Cedrus libani</i>	4 mos (July-Oct)	21.10.2012
P2	Pinaceae	<i>Cedrus libani</i>		04.10.2012
P3	Pinaceae	<i>Cedrus atlantica</i>	4 mos (July-Oct)	21.10.2012
P4	Pinaceae	<i>Cedrus deodora</i>	5 mos (June-Oct)	21.10.2012
P5	Betulaceae	<i>Betula pendula</i>	9 mos (Aug-Apr)	14.04.2012
P6	Betulaceae	<i>Betula pendula</i>		03.04.2016
P7	Pinaceae	<i>Pinus nigra</i>	2 mos (Apr-May)	07.05.2016
P8	Pinaceae	<i>Pinus sylvestris</i>	2 mos (Apr-May)	28.04.2018
P9	Pinaceae	<i>Pinus sylvestris</i>		26.04.2016
P10	Pinaceae	<i>Pinus graffiti</i>	2 mos (Apr-May)	15.05.2013
P11	Pinaceae	<i>Pinus brutia</i>	2 mos (Apr-May)	18.05.2012
P12	Pinaceae	<i>Pinus pinea</i>	6 mos (Nov-May)	15.05.2015
P13	Salicaceae	<i>Populus nigra</i>	7 mos (Nov-Apr)	08.04.2013
P14	Salicaceae	<i>Populus nigra</i>		07.04.2018
P15	Salicaceae	<i>Populus nigra</i>		01.05.2012
P16	Salicaceae	<i>Populus nigra</i>		14.04.2012
P17	Salicaceae	<i>Populus alba</i>	7 mos (Nov-Apr)	04.03.2013
P18	Cupressaceae	<i>Juniperus horizontalis</i>	10 mos (July-June)	19.03.2013
P19	Cupressaceae	<i>Juniperus horizontalis</i>		20.03.2013
P20	Cupressaceae	<i>Juniperus virginica</i>	6 mos (Sep-Mar)	26.02.2013
P21	Cupressaceae	<i>Juniperus sabina</i>	6 mos (Sep-Mar)	21.03.2013
P22	Cupressaceae	<i>Juniperus oxycedrus</i>	6 mos (Sep-Mar)	04.05.2012
P23	Cupressaceae	<i>Cupressus arizonica</i>	4 mos (Dec-Mar)	04.04.2012
P24	Cupressaceae	<i>Cupressus arizonica</i>		04.04.2012
P25	Cupressaceae	<i>Cupressus arizonica</i>		04.04.2012
P26	Cupressaceae	<i>Cupressus sepveriansis</i> ssp. <i>horizontalis</i>	4 mos (Dec-Mar)	30.03.2013
P27	Cupressaceae	<i>Cupressus sepveriansis</i> ssp. <i>horizontalis</i>		30.02.2013
P28	Cupressaceae	<i>Thuja orientalis</i>	4 mos (Dec-Mar)	03.03.2013
P29	Cupressaceae	<i>Thuja orientalis</i>		02.03.2013
P30	Cupressaceae	<i>Thuja orientalis</i>		08.03.2013
P31	Betulaceae	<i>Corylus avellana</i>	8 mos (May-Jan)	18.02.2018
P32	Betulaceae	<i>Corylus avellana</i>		21.03.2012
P33	Oleaceae	<i>Fraxinus excelsior</i>	2 mos (Feb-Apr)	11.03.2019
P34	Pinaceae	<i>Abies nordmanniana</i> subsp. <i>equi-trojani</i>	4 mos (Mar-June)	21.05.2012
P35	Araucariaceae	<i>Araucaria sp</i>	4 mos (July-Oct)	16.10.2021

$$A(\text{Bq/kg}) = \frac{C \times \lambda \times N_{\text{AV}} \times h}{M \times F} \quad (1)$$

where C is the elemental concentration of element; λ is the decay constant (1/s) of each radionuclide; N_{AV} is Avogadro's number (6.023×10^{23} atoms/mol); h is the atomic abundance (%) of each radionuclide in nature; M is the atomic mass (kg/mol) and F is a factor with a value of 100 for ^{40}K and 1,000,000 for ^{238}U and ^{232}Th .

Assessment of non-carcinogenic health risk

Health risk assessment can be defined as the risk characterization of potential unfavorable health effects of human exposure to pollutants (Tan et al. 2016; Miletic et al. 2023). Health risks of pollutants often contain non-carcinogenic and carcinogenic risks. Health risk assessment is a tool for evaluating the degree and likelihood of adverse health effects in individuals exposed to toxic substances in polluted environments (USEPA United States Environmental Protection Agency 2014a; Sultana et al. 2023). Chronic daily intake (CDI in mg/kg/d) associated with inhalation, ingestion, and dermal contact was calculated for adults as follows (Tan et al. 2016; Sultana et al. 2023; Miletic et al. 2023):

$$\text{CDI}_{\text{Inh}} = \frac{C \times R_{\text{Inh}} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BM} \times \text{AT}} \quad (2)$$

$$CDI_{\text{Ing}} = \frac{C \times R_{\text{Ing}} \times EF \times ED \times 10^{-6}}{BM \times AT} \quad (3)$$

$$CDI_{\text{Derm}} = \frac{C \times SA \times AF \times ABS \times EF \times ED \times 10^{-6}}{BM \times AT} \quad (4)$$

where C is the PTM concentration in the pollen samples (mg/kg); R_{Inh} and R_{Ing} are the inhalation and ingestion rate of pollen given as $20 \text{ m}^3/\text{d}$ and $100 \text{ mg}/\text{d}$, respectively (USEPA United States Environmental Protection Agency 2011; Miletić et al. 2023); EF is exposure frequency ($300 \text{ d}/\text{y}$); ED is exposure duration to pollen (24 y) (Miletić et al. 2023); BM is the average body mass of adults (70 kg) and AT is the average exposure time ($EF \times ED$); PEF is the particle emission factor ($1.36 \times 10^9 \text{ m}^3/\text{kg}$) (Miletić et al. 2023); SA is skin surface area available for contact (5700 cm^2) (Miletić et al. 2023); AF is the adherence factor represented the number of PTMs adhered to the skin ($0.07 \text{ mg}/\text{cm}^2/\text{d}$) (Miletić et al. 2023) and ABS is the dermal absorption factor (0.001) (Miletić et al. 2023). The estimation of non-carcinogenic risk refers to a determination of the effect of PTMs in pollen on non-carcinogenic effects in humans. Hazard quotient (HQ) and hazard index (HI) are used to estimate the non-carcinogenic risk (Miletić et al. 2023). HQ_i is the ratio of the CDIs of the PTMs given in Equations (2)-(4) to their respected reference doses (R_fD) for exposure routes as follows (Tan et al. 2016; Sultana et al. 2023; Miletić et al. 2023):

$$HQ_{i,\text{Inh}} = \frac{CDI_{i,\text{Inh}}}{R_fD_{i,\text{Inh}}} \quad (5)$$

$$HQ_{i,\text{Ing}} = \frac{CDI_{i,\text{Ing}}}{R_fD_{i,\text{Ing}}} \quad (6)$$

$$HQ_{i,\text{Derm}} = \frac{CDI_{i,\text{Derm}}}{R_fD_{i,\text{Derm}}} \quad (7)$$

$$HQ_i = HQ_{i,\text{Ing}} + HQ_{i,\text{Inh}} + HQ_{i,\text{Derm}} \quad (8)$$

where R_fD_{Inh} , R_fD_{Ing} and R_fD_{Derm} are the reference dose values of V, Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb taken as 9×10^{-3} , 3×10^{-3} , 140×10^{-3} , 7×10^{-1} , 2×10^{-2} , 20×10^{-3} , 4×10^{-2} , 300×10^{-3} and $3.5 \times 10^{-5} \text{ mg}/\text{kg}/\text{d}$, 7×10^{-3} , 2.86×10^{-5} , 1.43×10^{-5} , 7×10^{-3} , 5.7×10^{-6} , 2.06×10^{-2} , 4.02×10^{-2} , 300×10^{-3} and $3.52 \times 10^{-3} \text{ mg}/\text{kg}/\text{d}$ and 7×10^{-5} , 6×10^{-5} , 1.84×10^{-3} , 2×10^{-3} , 1.6×10^{-2} , 5.4×10^{-3} , 1.2×10^{-2} , 3×10^{-1} and $5.24 \times 10^{-4} \text{ mg}/\text{kg}/\text{d}$, respectively USEPA (United States Environmental Protection Agency (2004); Miletić et al. (2023). Thereafter, all HQ_i values for each PTM in a pollen sample are summed to obtain the HI as follows:

$$HI = \sum_{i=1}^n HQ_i \quad (9)$$

where n is the number of PTMs. If the $HQ > 1$ or $HI > 1$, there is a potential risk that PTMs may have a detrimental effect on health; If $HI < 1$, the effect of PTMs is negligible (Miletić et al. 2023).

Assessment of radiological risk

The radiological risk for people exposed to the internal exposure caused by alpha-, beta- and gamma-ray emitted from the radionuclides in the ingested pollen samples was assessed by estimating the annual effective dose (AED) as follows (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008; Altamemi et al. 2021):

$$\text{AED}(\mu\text{Sv/y}) = \sum_i A_i \times \text{DCF}_i \times \text{AIP} \quad (10)$$

where A_i is the activity concentration of the radionuclide i (Bq/kg) given in Equations 1; DCF_i is the dose conversion coefficient of the radionuclide i (0.0062, 0.22 and 0.044 $\mu\text{Sv/Bq}$ for ^{40}K , ^{232}Th and ^{238}U , respectively) (ICRP 119 2012) and AIP is an annual intake of pollen (1.64×10^{-4} kg/y).

Positive matrix factorization (PMF) model

EPA PMF version 5.0 software was used to develop the PMF receptor model (USEPA United States Environmental Protection Agency 2014b) to identify the sources of PTMs in pollen samples, as described previously by Jiang et al. (2023). As indicated by studies conducted by Zhang et al. (2018), Cheng et al. (2020), and Jiang et al. (2023), p. 20 base runs were conducted in random seed mode, ranging from 3 to 6 factors, to obtain the optimal solution. Subsequently, two factors were tested for better results and accepted as sources of PTMs in pollen samples.

Statistical analysis

In this study, the Shapiro-Wilk and Bartlett tests were used to investigate normal distribution and variance homogeneity in the data. Since most of the data did not present normal distribution or variance homogeneity, logarithmic transformation was applied to the data.

The re-evaluation of the logarithmically transformed data showed considerable progress in normal distribution and variance homogeneity, but a small subset of the data still demonstrated deviations from normal distribution or lacked homogeneity of variances.

If the transformed data presented normal distribution and variance homogeneity, a one-way analysis of variance (ANOVA) was conducted to explore potential differences among groups, followed by post-hoc Tukey tests to identify specific groups showing significant differences, in the event of a significant result ($p < 0.05$) from the ANOVA. On the contrary, the Kruskal-Wallis test, a non-parametric alternative of ANOVA, was used, and for the cases where the Kruskal-Wallis test yielded a significant p-value ($p < 0.05$), subsequent pairwise Wilcoxon rank-sum tests were performed to identify the specific groups exhibiting significant differences.

In addition, Pearson correlation coefficients were calculated to elucidate any correlations among the concentrations of PTMs in the pollen samples. All statistical analyses were conducted by R studio version 2023.06 (R Core Team 2024).

Results and discussion

PTM concentration

Concentrations and some descriptive statistical data of PTMs and elemental concentrations of K, Th, and U and the corresponding activity concentrations of ^{40}K , ^{232}Th , and ^{238}U analyzed in each pollen sample are given in Table 2. The frequency distribution of the concentration of PTMs is shown in Figure 2. According to Figure 2, the log concentration distributions of Ti, V, Cr, Mn, Fe, Zn, and Sr exhibit a normal distribution, while Co, Ni, Cu, and Pb have a non-normal distribution. However, based on the statistical analysis, it was found that Cr did not exhibit a normal distribution ($p = 0.03$), while Pb demonstrated a normal distribution ($p = 0.05$).

From Table 2, the average concentrations of PTMs analyzed in pollen samples are ranked as $\text{Fe} > \text{Mn} > \text{Zn} > \text{Ti} > \text{Sr} > \text{Cu} > \text{Ni} > \text{Co} > \text{Cr} > \text{V} > \text{Pb}$ (Figure 3). The concentrations of As, Cd, and Hg in pollen samples were below the detection limit of 0.5, 2.0 and 1.0 mg/kg, respectively.

The trend of occurrence of PTMs in pollen samples of three six plant families is shown below: $\text{Fe} > \text{Mn} > \text{Zn} > \text{Ti} > \text{Cu} > \text{Ni} > \text{Co} > \text{Cr} > \text{Sr} > \text{V} > \text{Pb}$ for Pinaceae; $\text{Fe} > \text{Zn} > \text{Mn} > \text{Ti} > \text{Cu} > \text{Ni} > \text{Sr} > \text{Cr} > \text{V} > \text{Co} > \text{Pb}$ for Betulaceae; $\text{Fe} > \text{Zn} > \text{Mn} > \text{Cu} > \text{Ti} > \text{Sr} > \text{Ni} > \text{Co} > \text{Cr} > \text{V} > \text{Pb}$ for

Table 2. The concentration and some descriptive statistics data of PTMs in pollen samples.

Sample code	Elemental concentration (mg/kg)														Activity concentration (Bq/kg)			
	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Sr	Pb	Th	U	K(%)	⁴⁰ K	²³² Th	²³⁸ U	
P1	27.5	2.2	4.6	118.8	150.5	4.1	9.7	8.4	58.0	3.1	1.8	1.6	<LD	2.1	640.9	6.5	<LD	
P2	64.2	2.6	7.5	145.8	423.3	11.3	11.7	14.0	45.1	3.8	2.5	1.3	<LD	1.8	531.0	5.3	<LD	
P3	35.7	1.9	5.2	63.5	198.9	10.9	10.8	7.8	40.2	1.9	2.3	1.8	0.8	2.2	651.5	7.3	9.9	
P4	76.8	5.1	6.0	57.8	506.3	8.2	11.4	14.4	60.5	3.3	1.9	1.5	<LD	2.2	656.0	6.1	<LD	
P5	35.5	2.7	4.6	59.6	359.9	4.3	8.0	22.1	126.0	11.7	1.4	1.3	1.1	1.9	576.4	5.3	13.6	
P6	27.1	4.7	3.2	78.0	194.5	<LD	8.7	27.4	145.5	7.8	<LD	1.1	<LD	1.8	545.6	4.5	<LD	
P7	10.7	1.5	3.4	61.9	54.1	6.0	8.1	11.9	87.6	1.7	1.4	1.4	1.2	2.0	615.2	5.7	14.8	
P8	17.0	<LD	2.7	76.7	82.5	10.7	8.2	8.6	82.9	1.3	1.4	1.6	1.3	2.1	633.0	6.5	16.0	
P9	23.3	1.9	4.3	59.5	144.1	10.2	9.6	7.7	81.0	2.5	2.2	2.2	0.7	2.4	740.4	8.9	8.6	
P10	14.3	2.8	3.9	75.9	57.1	3.8	8.6	7.5	63.6	3.6	1.6	1.1	0.9	3.0	905.0	4.5	11.1	
P11	18.2	1.0	3.9	53.0	87.2	7.3	8.5	9.5	66.6	1.1	1.8	1.3	0.8	2.2	650.9	5.3	9.9	
P12	11.9	1.0	3.4	52.8	52.1	4.0	8.2	5.0	54.6	1.0	1.3	1.2	1.1	2.7	811.2	4.9	13.6	
P13	15.4	<LD	2.2	63.0	129.3	4.0	6.0	35.5	199.7	7.3	<LD	0.8	0.7	3.5	1045.4	3.2	8.6	
P14	9.1	<LD	1.6	56.8	63.1	7.9	8.4	22.1	81.5	5.5	1.6	2.3	1.3	2.1	620.9	9.3	16.0	
P15	51.5	2.0	3.5	83.2	481.4	4.0	8.1	35.2	160.7	6.0	1.5	1.4	1.2	3.0	903.8	5.7	14.8	
P16	40.9	2.0	4.5	90.4	404.4	3.4	10.2	28.5	178.7	11.3	1.5	1.2	0.9	4.4	1341.7	4.9	11.1	
P17	17.9	1.1	2.3	72.3	121.0	<LD	6.7	32.3	116.5	21.4	<LD	1.0	<LD	2.8	836.1	4.1	<LD	
P18	180.6	8.5	9.7	108.3	1992.0	4.2	9.6	15.3	40.9	121.7	2.5	0.9	1.9	1.4	428.5	3.7	23.5	
P19	26.3	1.4	3.3	77.1	254.7	6.4	6.8	11.5	34.9	32.6	1.6	2.0	0.9	1.6	470.8	8.1	11.1	
P20	216.2	10.1	14.7	159.6	1772.0	4.2	10.7	18.6	55.7	128.4	5.7	0.6	2.0	1.0	295.7	2.4	24.7	
P21	70.5	2.6	5.8	67.7	662.5	<LD	7.5	11.2	28.9	44.8	1.2	1.0	0.7	1.5	455.1	4.1	8.6	
P22	282.1	11.1	12.5	98.1	3078.0	7.3	12.7	11.3	47.7	54.9	2.8	1.5	0.7	1.8	559.2	6.1	8.6	
P23	39.5	3.4	4.4	28.7	365.6	4.2	6.0	9.2	15.6	40.5	1.2	0.6	0.6	0.8	255.1	2.4	7.4	
P24	139.9	5.7	9.5	90.5	1423.0	4.3	10.1	10.7	26.5	70.1	4.3	2.1	1.4	0.9	264.6	8.5	17.3	
P25	80.4	4.3	5.9	31.8	603.3	4.3	9.2	10.7	25.4	39.6	2.2	1.3	0.4	1.1	332.8	5.3	4.9	
P26	40.0	2.1	3.9	48.6	342.4	<LD	6.5	8.1	24.1	25.5	<LD	0.8	0.7	1.6	480.5	3.2	8.6	
P27	43.6	2.3	3.5	47.7	334.9	4.2	6.4	8.2	24.2	25.3	1.3	0.8	<LD	1.6	475.7	3.2	<LD	
P28	43.4	2.8	6.7	66.5	431.6	4.2	8.5	5.5	37.1	9.0	<LD	0.7	0.9	1.6	472.0	2.8	11.1	
P29	45.0	2.9	4.9	68.5	384.9	4.2	8.9	7.2	39.3	10.9	2.0	1.6	0.7	1.7	504.7	6.5	8.6	
P30	32.3	3.1	4.2	78.2	280.3	<LD	7.5	7.0	32.6	19.6	1.3	0.9	0.9	1.7	509.3	3.7	11.1	
P31	45.8	3.9	3.4	120.4	483.0	4.2	8.2	40.1	101.0	5.5	<LD	1.1	0.8	1.8	536.8	4.5	9.9	
P32	102.5	5.7	26.6	96.7	838.0	<LD	23.6	38.8	113.2	16.1	2.3	1.4	<LD	1.5	465.4	5.7	<LD	
P33	16.1	1.5	2.8	97.8	170.9	<LD	5.4	15.1	98.0	4.6	<LD	0.8	0.7	1.5	456.6	3.2	8.6	
P34	20.3	1.4	3.1	26.1	144.7	8.9	9.6	12.0	66.0	2.6	2.0	1.4	0.4	2.1	639.1	5.7	4.9	
P35	29.0	1.8	4.2	82.3	244.3	4.3	8.1	17.1	36.9	15.2	2.3	1.4	<LD	1.8	545.9	5.7	<LD	
Average	55.7	3.3	5.6	76.1	494.7	5.8	9.0	15.9	71.3	21.7	2.0	1.3	1.0	2.0	595.8	5.2	11.8	
Median	35.7	2.6	4.2	72.3	334.9	4.3	8.5	11.5	58.0	9.0	1.8	1.3	0.9	1.8	545.9	5.3	11.1	
SD	61.0	2.5	4.6	29.7	638.7	2.5	3.0	10.2	46.7	30.8	1.0	0.4	0.4	0.7	220.2	1.8	4.8	

(Continued)

Table 2. (Continued).

Sample code	Elemental concentration (mg/kg)													Activity concentration (Bq/kg)			
	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Sr	Pb	Th	U	K(%)	⁴⁰ K	²³² Th	²³⁸ U
SE	10.3	0.4	0.8	5.0	108.0	0.4	0.5	1.7	7.9	5.2	0.2	0.1	0.1	0.1	36.7	0.3	0.8
Kurtosis	2.4	1.9	3.2	0.9	2.7	1.1	3.3	1.2	1.2	2.4	2.5	0.5	1.2	1.3	1.3	0.5	1.2
Skewness	5.9	3.2	12.6	1.2	8.0	-0.2	15.4	0.2	0.8	6.0	7.7	-0.1	1.7	2.9	2.9	-0.1	1.7
Min	9.1	< LD	1.6	26.1	52.1	< LD	5.4	5.0	15.6	1.0	< LD	0.6	< LD	0.8	255.1	2.4	< LD
Max	282.1	11.1	26.6	159.6	3078.0	11.3	23.6	40.1	199.7	128.4	5.7	2.3	2.0	4.4	1341.7	9.3	24.7

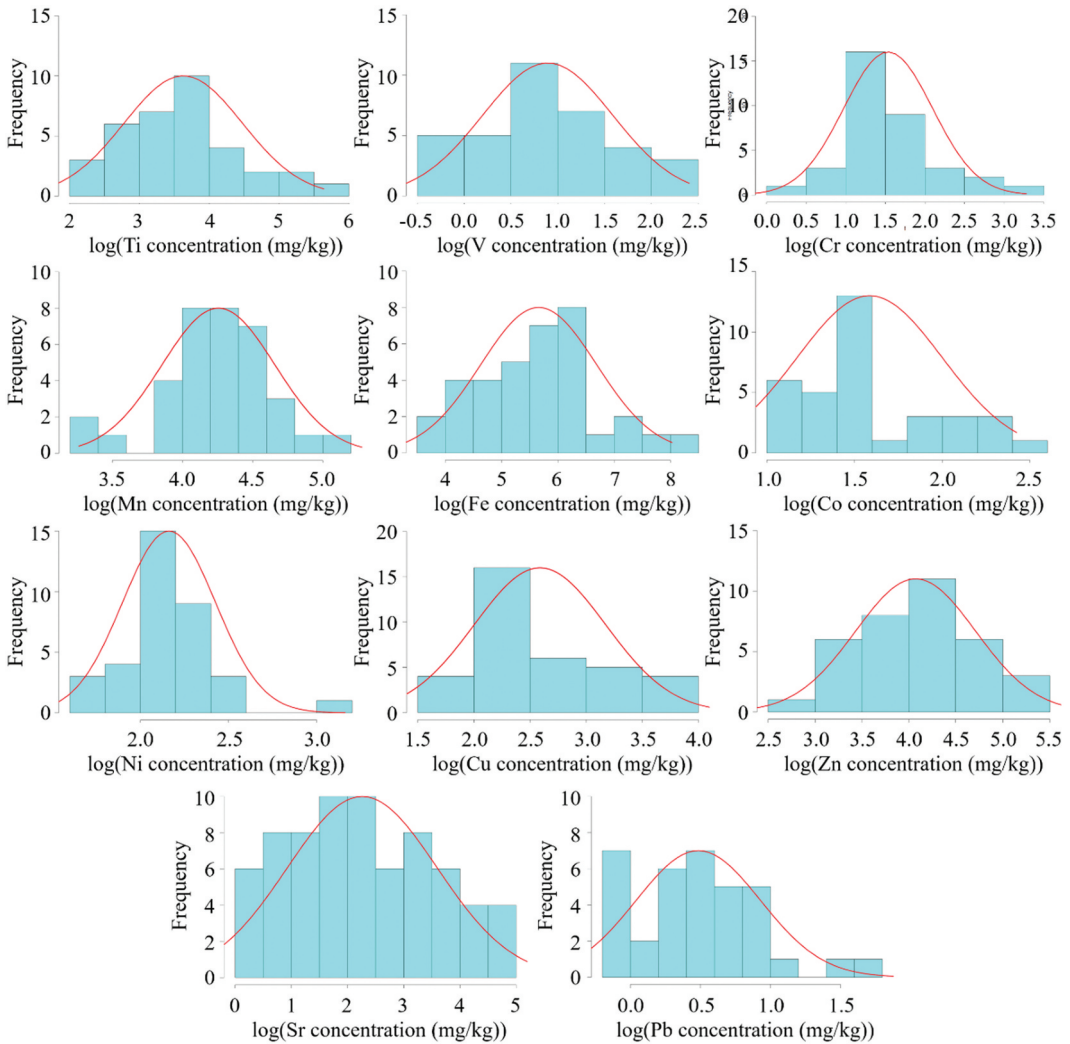


Figure 2. The frequency distribution of the logarithmic transformed concentration of PTMs.

Salicaceae; Fe > Ti > Mn > Sr > Zn > Cu > Ni > Cr > Co > V > Pb for Cupressaceae; Fe > Zn > Mn > Ti > Cu > Ni > Sr > Cr > V (Pb and Co < LD) for Oleaceae and Fe > Mn > Zn > Ti > Cu > Sr > Ni > Co > Cr > Pb > V for Araucariaceae. Since the number of samples in Oleaceae and Araucariaceae is insufficient for statistical analysis, the concentration of PTMs in Pinaceae, Betulaceae, Salicaceae, and Cupressaceae families are compared statistically.

According to the statistical analysis, Ti and Fe concentrations in Cupressaceae are statistically different from those in Pinaceae and Salicaceae ($p < 0.05$). Regarding the V concentration, it is observed that Betulaceae and Cupressaceae are different from Pinaceae and Salicaceae ($p < 0.05$). The Cr concentration in Salicaceae is observed to be different from Pinaceae, Betulaceae, and Cupressaceae. On the other hand, the Cu concentrations in Pinaceae and Cupressaceae are similar ($p > 0.05$), and they are different from the Cu concentrations in Betulaceae and Salicaceae ($p < 0.05$). In terms of both Zn and Sr concentrations, it is noted that Betulaceae and Salicaceae exhibit similarity ($p > 0.05$), differing from Pinaceae and Cupressaceae ($p < 0.05$). Additionally, Pinaceae and Cupressaceae also demonstrate dissimilarity ($p < 0.05$).

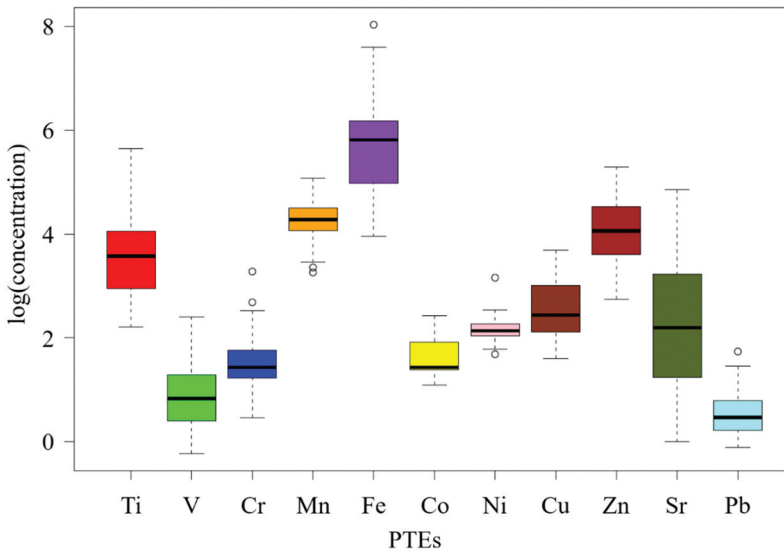


Figure 3. The box plot for the logarithmic transformed data.

According to the statistical analysis, there are no statistical differences for Mn, Co, Ni, and Pb in the Pinaceae, Betulaceae, Salicaceae, and Cupressaceae families.

The trend of occurrence of PTMs in pollen samples of five provinces is shown below: Fe > Mn > Zn > Ti > Sr > Cu > Ni > Co > Cr > V > Pb based on the average of 23 samples from Kastamonu; Fe > Mn > Zn > Ti > Sr > Cu > Ni > Cr > Co > V > Pb based on the average of 7 samples from Ankara; Fe > Zn > Mn > Ti > Cu > Ni > Sr > Co > Cr > V > Pb based on the average of 3 samples from Çankırı; Fe > Mn > Zn > Ti > Cu > Sr > Ni > Co > Cr > Pb > V for Muğla (Bodrum) and Fe > Zn > Mn > Ti > Cu > Ni > Co > Cr > Pb > Sr > V for Antalya (Akseki). Based on the statistical analysis, it was found that there was no statistically significant difference in the concentrations of PTMs in all provinces ($p > 0.05$).

From Table 2, the Fe concentrations in the pollen samples varied from 52.1 (P12 from the Cide district of Kastamonu) to 3078.0 (P22 from Kastamonu city center) mg/kg with an average value of 494.7 mg/kg. According to average Fe concentration, provinces ranked as Kastamonu > Ankara > Çankırı > Muğla > Antalya. All Fe concentrations are significantly lower than the Earth's crust average concentration (ECAC) of 46,500 mg/kg (Yaroshevsky 2006). The Mn concentrations in the pollen samples varied from 26.1 (P34 from Bodrum district of Muğla) to 159.6 (P20 from the garden of the Faculty of Science of Ankara University Campus) mg/kg with an average value of 76.1 mg/kg. According to average Mn concentration, provinces ranked as Ankara > Muğla > Kastamonu > Çankırı > Antalya. All Mn concentrations are significantly lower than the ECAC of 1000 mg/kg (Yaroshevsky 2006). The Zn concentrations in the pollen samples varied from 15.6 (P23 from Kuzyekent neighborhood of Kastamonu) to 199.7 (P13 from Kastamonu University Kuzyekent Campus, Vocational School Garden) mg/kg with an average value of 71.3 mg/kg. According to average Zn concentration, provinces ranked as Çankırı > Ankara > Kastamonu > Antalya > Muğla. All Zn concentrations are lower than the ECAC of 83 mg/kg (Yaroshevsky 2006). The Ti concentrations in the pollen samples varied from 9.1 (P14 from the Akyurt district of Ankara) to 282.1 (P22 from Kastamonu city center) mg/kg with an average value of 55.7 mg/kg. According to average Ti concentration, provinces ranked as Ankara > Kastamonu > Çankırı > Muğla > Antalya. All Ti concentrations are significantly lower than the Earth's crust average concentration (ECAC) of 4500 mg/kg (Yaroshevsky 2006). The Sr concentrations in the pollen samples varied from 1.0 (P12 from the

Cide district of Kastamonu) to 128.4 (P20 from the garden of the Faculty of Science of Ankara University Campus) mg/kg with an average value of 21.7 mg/kg. According to average Sr concentration, provinces ranked as Ankara > Kastamonu > Muğla > Çankırı > Antalya. All Sr concentrations are lower than the Earth's crust average concentration (ECAC) of 340 mg/kg (Yaroshevsky 2006). The Cu concentrations in the pollen samples varied from 5.0 (P12 from the Cide district of Kastamonu) to 40.1 (P31 from Kırık dam construction site in Kastamonu) mg/kg with an average value of 15.9 mg/kg. According to average Cu concentration, provinces ranked as Ankara > Çankırı > Muğla > Kastamonu > Antalya. All Cu concentrations are lower than the Earth's crust average concentration (ECAC) of 47 mg/kg (Yaroshevsky 2006). The Ni concentrations in the pollen samples varied from 5.4 (P33 from Vocational School Garden of Kastamonu University) to 23.6 (P32 Ankara University Faculty of Science Block C garden) mg/kg with an average value of 9.0 mg/kg. According to average Ni concentration, provinces ranked as Ankara > Çankırı > Antalya > Kastamonu > Muğla. All Ni concentrations are significantly lower than the ECAC of 58 mg/kg (Yaroshevsky 2006). Co concentrations analyzed in six pollen samples were found below the detection limit. The Co concentrations analyzed in thirty pollen samples varied from 3.4 (P16 from Vocational School Garden of Kastamonu University) to 11.3 (P2 from Ankara University Tandoğan Campus, next to the cafeteria) mg/kg with an average value of 5.8 mg/kg. According to average Co concentration, provinces ranked as Antalya > Ankara > Kastamonu > Çankırı > Muğla. All Co concentrations are lower than the ECAC of 18 mg/kg (Yaroshevsky 2006). The Cr concentrations in the pollen samples varied from 1.6 (P14 from Akyurt district of Ankara) to 26.6 (P32 Ankara University Faculty of Science Block C garden) mg/kg with an average value of 5.6 mg/kg. According to average Cr concentration, provinces ranked as Ankara > Kastamonu > Çankırı > Muğla > Antalya. All Cr concentrations are lower than the Earth's crust average concentration (ECAC) of 83 mg/kg (Yaroshevsky 2006). V concentrations analyzed in three pollen samples were found below the detection limit. The V concentrations analyzed in thirty-three pollen samples varied from 1.0 (P11 and P12 from Antalya (Akseki) and Cide district of Kastamonu Vocational School Garden of Kastamonu University, respectively) to 11.1 (P22 from Kastamonu city center) mg/kg with an average value of 3.3 mg/kg. According to average V concentration, provinces ranked as Ankara > Kastamonu > Çankırı > Muğla > Antalya. All V concentrations are significantly lower than the ECAC of 90 mg/kg (Yaroshevsky 2006). Pb concentrations analyzed in seven pollen samples were found below the detection limit. The Pb concentrations analyzed in twenty-nine pollen samples varied from 1.2 (P21 and P22 from Kastamonu city) to 5.7 (P20 from the garden of the Faculty of Science of Ankara University Campus) mg/kg with an average value of 2.0 mg/kg. According to average Pb concentration, provinces ranked as Ankara > Muğla > Kastamonu > Antalya > Çankırı. All Pb concentrations are significantly lower than the ECAC of 16 mg/kg (Yaroshevsky 2006).

Potassium consists of three natural isotopes: two stable forms ^{39}K (93.3%) and ^{41}K (6.7%), and a very long-lived radioisotope ^{40}K (0.0117%; half-life: 1.28×10^9 y). From Table 2, the activity concentrations of ^{40}K in the pollen samples varied from 255.1 (P23) to 1341.7 (P16) Bq/kg with an average value of 595.8 Bq/kg. According to the average ^{40}K activity concentration, families were ranked as Salicaceae (679.5 Bq/kg) > Pinaceae (531.0 Bq/kg) > Araucariaceae (545.9 Bq/kg) > Betulaceae (531.0 Bq/kg) > Oleaceae (456.6 Bq/kg) > Cupressaceae (423.4 Bq/kg). All activity concentrations of ^{40}K , except for four pollen samples, are higher than the worldwide average value of 412 Bq/kg (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008). The activity concentrations of ^{232}Th in the pollen samples varied from 2.4 (P20) to 9.3 (P14) Bq/kg with an average value of 5.2 Bq/kg. According to the average ^{232}Th activity concentration, families ranked as Pinaceae (6.0 Bq/kg) > Araucariaceae (5.7 Bq/kg) > Salicaceae (5.4 Bq/kg) > Betulaceae (5.0 Bq/kg) > Cupressaceae (4.6 Bq/kg) > Oleaceae (3.2 Bq/kg). All activity concentrations of ^{232}Th , except for four pollen samples, are lower than the worldwide average value of 45 Bq/kg (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008). U concentrations analyzed in eight pollen samples were found below the detection limit. The

activity concentrations of ^{238}U analyzed in twenty-eight pollen samples varied from 4.9 (P25) to 24.7 (P20) Bq/kg with an average value of 11.8 Bq/kg. According to average ^{238}U activity concentration, families ranked as Salicaceae (12.7 Bq/kg) > Cupressaceae (12.1 Bq/kg) > Betulaceae (11.7 Bq/kg) > Pinaceae (11.1 Bq/kg) > Oleaceae (8.6 Bq/kg). All activity concentrations of ^{238}U are lower than the worldwide average value of 32 Bq/kg (UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation 2008).

Pearson correlation analysis was employed to illustrate pairwise correlations and investigate potential sources of PTMs in pollen samples. Concurrently, the PMF model was employed to unveil common sources, if any, contributing to the presence of PTMs in the samples, as mentioned previously by Jiang et al. (2023). The analysis revealed two factors as the sources of PTMs.

According to the results, Factor 1 and Factor 2 contributed 25.10% and 74.90%, respectively, to the PMF model (Figure 4a).

Figure 4b demonstrates the contribution of each PTM to the identified factors in the PMF model. Figure 4c illustrates the Pearson correlation coefficients for pairs of PTMs and their relationship to the factors identified in the PMF model. The color of the lines connecting each PTM to the factors indicates the percentage contribution of each PTM to the factors.

The model indicates that Zn, Cu, Mn, Co, and Ni exhibit significant contributions to Factor 1. Previous studies associated Zn with coal combustion, oil combustion, wood combustion, and industrial applications (Haque et al. 1982). Al-Masri et al. (2006) proposed that Cu originates from sources such as oil combustion and wood smoke. Furthermore, Mn is suggested to derive from the combustion of fossil fuels (Howe et al. 2004). Al-Khashman (2004) recommended that Co can be found in the air due to the corrosion of metallic components in vehicles, such as engine wear, thrust bearings, and brush wear. In addition, Ni can be present in the air as a result of processes such as coal combustion, as well as the burning of fuel and diesel oil (Von Burg 1997). Thus, it is possible to propose that Factor 1 could represent air pollution.

The results showed that Ti, V, Cr, Fe, Sr, and Pb contributed to Factor 2. Previous studies suggested that both Ti (Maina et al. 2016; Kumar et al. 2022), V (Hernandez and Rodriguez 2012), and Cr (Kierczak et al. 2021) can originate from natural sources. Thus, they can be accumulated in the soil. Fe can be found in the soil as a result of production, use of hydrocarbons, mining, and other industrial activities (Emeh et al. 2019). The source of Sr Earth's crust and this PTM is found in soil due to mining, combustion, and

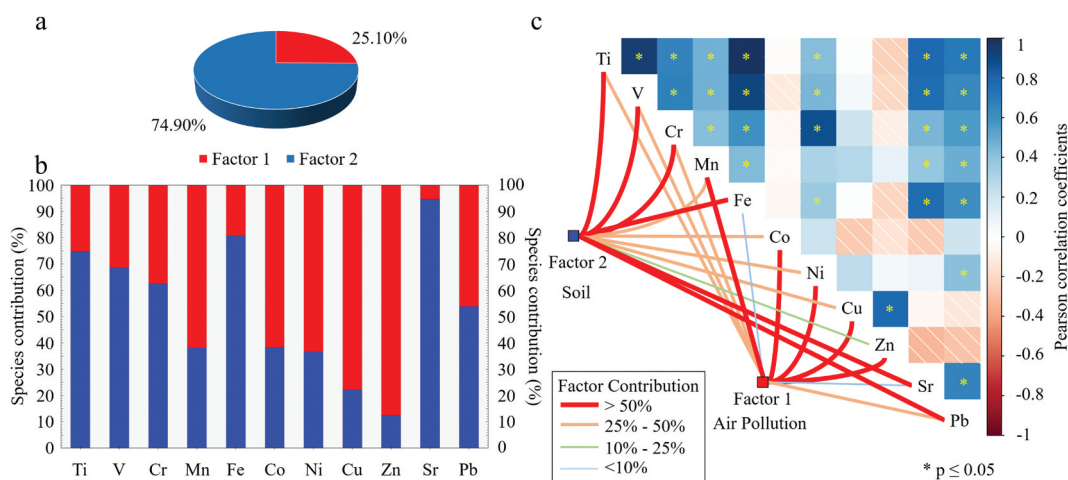


Figure 4. Source assignments for PTMs. (a) factor contributions (%) to the PMF model, (b) PTM contributions (%) to each factor, (c) the Pearson correlation coefficients for PTM pairs and the relation of each PTM to the factors of the PMF model.

Table 3. Impact of potentially toxic metals in pollen under different exposure routes on human health risks.

PTM	CDI (mg/kg d)			HQ _i		
	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal
Ti	1.2×10^{-8}	8.0×10^{-5}	3.2×10^{-7}	–	–	–
V	7.0×10^{-10}	4.8×10^{-6}	1.9×10^{-8}	1.0×10^{-7}	5.3×10^{-4}	2.7×10^{-4}
Cr	1.2×10^{-9}	8.0×10^{-6}	3.2×10^{-8}	4.1×10^{-5}	2.7×10^{-3}	5.3×10^{-4}
Mn	1.6×10^{-6}	1.1×10^{-4}	4.3×10^{-7}	1.1×10^{-3}	7.8×10^{-4}	2.4×10^{-4}
Fe	1.0×10^{-7}	7.1×10^{-4}	2.8×10^{-6}	1.5×10^{-5}	1.0×10^{-3}	1.4×10^{-3}
Co	1.2×10^{-9}	8.3×10^{-6}	3.3×10^{-8}	2.1×10^{-4}	4.2×10^{-4}	2.1×10^{-6}
Ni	1.9×10^{-9}	1.3×10^{-5}	5.1×10^{-8}	9.2×10^{-8}	6.5×10^{-4}	9.5×10^{-6}
Cu	3.3×10^{-9}	2.3×10^{-5}	9.0×10^{-8}	8.3×10^{-8}	5.7×10^{-4}	7.5×10^{-6}
Zn	1.5×10^{-8}	1.0×10^{-4}	4.1×10^{-7}	5.0×10^{-8}	3.4×10^{-4}	1.4×10^{-6}
Sr	4.6×10^{-9}	3.1×10^{-5}	1.2×10^{-7}	–	–	–
Pb	4.3×10^{-10}	2.9×10^{-6}	1.2×10^{-8}	1.2×10^{-7}	8.3×10^{-2}	2.2×10^{-5}
				$HI_i = \sum HQ_i$	1.4×10^{-3}	9.0×10^{-2}
				$HI = \sum HI_i$	9.4×10^{-2}	2.5×10^{-3}

using phosphate fertilizers in agriculture (Burger and Lichtscheidl 2019; Kebonye and Eze 2019). Lastly, Bird (2011) proposed that the sources of Pb in the environment can be linked to industrial anthropogenic activity and coal combustion. Thus, Factor 2 predominantly represents contributions from natural sources, soil, and pollution in the soil due to fertilizers and combustion.

Health risk assessment

Non-carcinogenic health risk results assessed for adults are presented in Table 3. The values of the CDI_{Inh} , CDI_{Ing} and CDI_{Derm} calculated for all PTMs varied from 4.3×10^{-10} to 1.0×10^{-7} mg/kg d, 2.9×10^{-6} to 7.1×10^{-4} mg/kg d and 1.2×10^{-8} to 2.8×10^{-6} mg/kg d, respectively. The average CDI_{Inh} , CDI_{Ing} and CDI_{Derm} values of the PTMs are ranked as Fe > Mn > Zn > Ti > Sr > Cu > Ni > Co > Cr > V > Pb. The values of the HQ_{Inh} , HQ_{Ing} and HQ_{Derm} estimated for PTMs varied from 5.0×10^{-8} to 1.1×10^{-3} , 3.4×10^{-4} to 8.3×10^{-2} and 1.4×10^{-6} to 1.4×10^{-3} , respectively. The average HQ_{Inh} , HQ_{Ing} and HQ_{Derm} values of the PTMs are ranked Mn > Co > Cr > Fe > Pb > V > Ni > Cu > Zn; Pb > Cr > Fe > Mn > Ni > Cu > V > Co > Zn; Fe > Cr > V > Mn > Pb > Ni > Cu > Co > Zn. The highest and lowest HQ_i values were 8.3×10^{-2} for ingestion of Pb and 5.0×10^{-8} for inhalation of Zn, respectively. All HQ values are lower than the risk limit of 1. The average values of the HI_{Inh} , HI_{Ing} and HI_{Derm} estimated for PTMs were 1.4×10^{-3} , 9.0×10^{-2} and 2.5×10^{-3} , respectively. Concerning average HI_i , the three exposure routes are ranked as ingestion > inhalation > dermal contact. The estimated HI value for all PTMs and three exposure routes is 0.094, which is below the risk limit of 1.

The AED values varied from 0.04 to 0.16 nSv/y with an average of 0.086 nSv/y.

Conclusion

It is a known fact that the air quality of urban areas of provinces decreases due to vehicle exhaust and dust pollution. Thus, vegetation and street plants in urban areas are exposed to air pollution. Pollen from flowers is exposed to air pollution and absorbs potentially toxic trace metals. In this study, for the first time, potentially toxic trace metal and natural radionuclide concentrations in the pollen produced by twenty-two species of the Pinaceae, Cupressaceae, Araucariaceae, Betulaceae, Salicaceae, and Oleaceae families grown in parks, gardens, and recreation areas in different locations of five provinces of Turkey were determined by the EDXRF spectrometry. Concentrations of eleven potentially toxic trace metals analyzed in pollen samples revealed that, except for zinc, they were well below the average concentrations in the Earth's crust. The interesting result obtained in the study is that the average activity concentration of potassium is greater than

the world average. With this study, pollen can also be used as a biological indicator of urban air pollution, and it has been shown that pollen has the potential to bioaccumulate potentially toxic trace metals and radioelements.

Human health risk assessment is a critical approach to estimating the remarkable risks due to the potentially toxic trace metal pollutants when in contact with the human body. In this study, human health risk assessment was performed using the deterministic model recommended by USEPA. Risk assessment results showed that HQ and HI values were all within safe levels, reflecting that potentially toxic trace metals do not pose a non-carcinogenic risk to adults' health. The average annual effective dose caused by the internal irradiation exposed to adults due to ingestion of examined pollen samples was estimated as 0.086 nSv, which is significantly lower than the worldwide average annual ingestion dose of 290 μ Sv (290×10^3 nSv) reported by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) (2008).

Acknowledgements

The authors thank Dr. Celalettin Duran for the map.

Disclosure statement

No potential conflict of interest was reported by the author(s).

Funding

The author(s) reported there is no funding associated with the work featured in this article.

Authors' contributions

Çeter, Karabıçak, Çeter, Bıyıklıoğlu, and Aktaş collected the samples and prepared them for analysis. Turhan and Kurnaz followed the analysis process. Altuner has done the statistical analyses, and Turhan evaluated the data and wrote the manuscript. All authors read and finalized the manuscript.

References

- Abbasi A, Kurnaz A, Mirekhtiary Sf TŞ. 2020. Radiation hazards and natural radioactivity levels in surface soil samples from dwelling areas of North Cyprus. *J Radioanal Nucl Chem.* 324(1):203–210. doi: [10.1007/s10967-020-07069-w](https://doi.org/10.1007/s10967-020-07069-w).
- Abbasi A, Mirekhtiary F. 2020. Heavy metals and natural radioactivity concentration in sediments of the mediterranean sea coast. *Mar Pollut Bull.* 154:1–7. doi: [10.1016/j.marpolbul.2020.111041](https://doi.org/10.1016/j.marpolbul.2020.111041).
- Abbasi A, Mirekhtiary SF. 2019. Risk assessment due to various terrestrial radionuclides concentrations scenarios. *Int J Radiat Biol.* 95(2):179–185. doi: [10.1080/09553002.2019.1539881](https://doi.org/10.1080/09553002.2019.1539881).
- Abbasi A, Zakaly HMH, Algethami M, Abdel-Hafez SH. 2022. Radiological risk assessment of natural radionuclides in the marine ecosystem of the northwest mediterranean sea. *Int J Radiat Biol.* 98(2):205–211. doi: [10.1080/09553002.2022.2020359](https://doi.org/10.1080/09553002.2022.2020359).
- Al-Khashman OA. 2004. Heavy metal distribution in dust, street dust and soil from the work place in Karak industrial estate, Jordan. *Atmos Environ.* 38(39):6803–6812. doi: [10.1016/j.atmosenv.2004.09.011](https://doi.org/10.1016/j.atmosenv.2004.09.011).
- Al-Masri MS, Al-Kharfan K, Al-Shamali K. 2006. Speciation of Pb, Cu and Zn determined by sequential extraction for identification of air pollution sources in Syria. *Atmos Environ.* 40(4):753–761. doi: [10.1016/j.atmosenv.2005.10.008](https://doi.org/10.1016/j.atmosenv.2005.10.008).
- Altamemi RAA, Turhan Ş, Kurnaz A. 2021. Natural and anthropogenic radioactivity in some vegetables and fruits commonly consumed in the western black sea Region of Turkey. *Radiochim Acta.* 109(12):935–942. doi: [10.1515/ract-2021-1100](https://doi.org/10.1515/ract-2021-1100).
- Altıkulaç A, Turhan Ş. 2023. Assessment of the levels of potentially toxic elements contained in natural bentonites collected from quarries in Turkey. *ACS Omega.* 8(23):20797–20986. doi: [10.1021/acsomega.3c01773](https://doi.org/10.1021/acsomega.3c01773).

- Altıkulaç A, Turhan Ş, Kurnaz A, Gören E, Duran C, Hançerlioğulları A, Uğur FA. 2022. Assessment of the enrichment of heavy metals in coal and its combustion residues. *ACS Omega*. 7(24):21239–21245. doi: [10.1021/acsomega.2c02308](https://doi.org/10.1021/acsomega.2c02308).
- Azzazy M. 2016. Environmental impacts of industrial pollution on pollen morphology of eucalyptus globulus Labill. (Myrtaceae). *J Appl Biol Biotechnol*. 4:57–62.
- Beck I, Jochner S, Gilles S, McIntyre M, Butters JTM, Schmidt-Weber C, Berendt H, Ring J, Menzel A, Traidl-Hoffman C. 2013. High environmental ozone levels lead to enhanced allergenicity of birch pollen. *PLOS ONE*. 8(11):1–7. doi: [10.1371/journal.pone.0080147](https://doi.org/10.1371/journal.pone.0080147).
- Bessonova VN. 1992. The state of pollen, as an indicator of environmental contamination by heavy metals. *Russ J Ecol*. 4:45–50.
- Bird G. 2011. Provenancing anthropogenic Pb within the fluvial environment: developments and challenges in the use of Pb isotopes. *Environ Int*. 37(4):802–819. doi: [10.1016/j.envint.2011.02.007](https://doi.org/10.1016/j.envint.2011.02.007).
- Bobrowska-Korzeniowska M, Jerzyńska J, Polańska K, Kaleta D, Stelmach I, Kunert A, Stelmach W. 2021. The effect of air pollution on the respiratory system in preschool children with contribution of urban heat islands and geographic data—the aim of the study and methodological assumptions. *Int J Occup Med Environ Health*. 34(4):453–460. doi: [10.13075/ijomeh.1896.01651](https://doi.org/10.13075/ijomeh.1896.01651).
- Burger A, Lichtscheidl I. 2019. Strontium in the environment: review about reactions of plants towards stable and radioactive strontium isotopes. *Sci Total Environ*. 653:1458–1512. doi: [10.1016/j.scitotenv.2018.10.312](https://doi.org/10.1016/j.scitotenv.2018.10.312).
- Calzoni GL, Antognoni F, Pari E, Fonti P, Gnes A, Speranza A. 2007. Active biomonitoring of heavy metal pollution using *Rosa rugosa* plants. *Environ Pollut*. 149(2):239–245. doi: [10.1016/j.envpol.2006.12.023](https://doi.org/10.1016/j.envpol.2006.12.023).
- Capone P, Lancia A, D'Ovidio MC. 2023. Interaction between air pollutants and pollen grains: effects on public and occupational health. *Atmosphere*. 14(10):1–16. doi: [10.3390/atmos14101544](https://doi.org/10.3390/atmos14101544).
- Cheng W, Lei S, Bian Z, Zhao Y, Li Y, Gan Y. 2020. Geographic distribution of heavy metals and identification of their sources in soils near large, open-pit coal mines using positive matrix factorization. *J Hazard Mater*. 387:1–14. doi: [10.1016/j.jhazmat.2019.121666](https://doi.org/10.1016/j.jhazmat.2019.121666).
- DeWeger LA, Bruffaerts N, Koenders MMJF, Verstraeten WW, Delcloo AW, Hentges P, Hentges F. 2021. Long-term pollen monitoring in the Benelux: evaluation of allergenic pollen levels and temporal variations of pollen seasons. *Front Allergy*. 2:1–12. doi: [10.3389/falgy.2021.676176](https://doi.org/10.3389/falgy.2021.676176).
- Doležalová WH, Mihočová S, Chovanec P, Pavlovský J. 2019. Potential ecological risk and human health risk assessment of heavy metal pollution in industrial affected soils by coal mining and metallurgy in Ostrava, Czech Republic. *Int J Environ Res Public Health*. 16(22):1–29. doi: [10.3390/ijerph16224495](https://doi.org/10.3390/ijerph16224495).
- Emeh C, Igwe O, Onwo ES. 2019. Potential effect of environmental pollution on the degree of dissolution of iron and aluminium oxides in lateritic soils. *Environ Earth Sci*. 78(8):1–15. doi: [10.1007/s12665-019-8259-3](https://doi.org/10.1007/s12665-019-8259-3).
- Gisler A. 2021. Allergies in urban areas on the rise: the combined effect of air pollution and pollen. *Int J Public Health*. 66:1–2. doi: [10.3389/ijph.2021.1604022](https://doi.org/10.3389/ijph.2021.1604022).
- Guedes A, Ribeiro N, Ribeiro H, Oliveira M, Noronha F, Abreu I. 2009. Comparison between urban and rural pollen of *Chenopodium alba* and characterization of adhered pollutant aerosol particles. *J Aerosol Sci*. 40(1):81–86. doi: [10.1016/j.jaerosci.2008.07.012](https://doi.org/10.1016/j.jaerosci.2008.07.012).
- Haque MA, Subramanian V, Gibbs RJ. 1982. Copper, lead, and zinc pollution of soil environment. *Crit Rev Env Sci Tec*. 12(1):13–68. doi: [10.1080/10643388209381693](https://doi.org/10.1080/10643388209381693).
- Hernandez H, Rodriguez R. 2012. Geochemical evidence for the origin of vanadium in an urban environment. *Environ Monit Assess*. 184:5327–5342.
- Howe P, Malcolm H, Dobson S. 2004. Manganese and its compounds: environmental aspects. World Health Organization.
- Huang S, Shao G, Wang L, Wang L, Tang L. 2019. Distribution and health risk assessment of trace metals in soils in the golden triangle of southern Fujian province, China. *Int J Environ Res Public Health*. 16(97):1–17. doi: [10.3390/ijerph16010097](https://doi.org/10.3390/ijerph16010097).
- ICRP 119. 2012. Compendium of dose coefficients based on ICRP publication 60. Ottawa: Elsevier.
- Jiang W, Meng L, Liu F, Sheng Y, Chen S, Yang J, Mao H, Zhang J, Zhang Z, Ning H. 2023. Distribution, source investigation, and risk assessment of topsoil heavy metals in areas with intensive anthropogenic activities using the positive matrix factorization (PMF) model coupled with self-organizing map (SOM). *Environ Geochem Health*. 45(8):6353–6370. doi: [10.1007/s10653-023-01587-8](https://doi.org/10.1007/s10653-023-01587-8).
- Kalbande DM, Dhadse SN, Chaudhari PR, Wate SR. 2008. Biomonitoring of heavy metals by pollen in urban environment. *Environ Monit Assess*. 138(1–3):233–238. doi: [10.1007/s10661-007-9793-0](https://doi.org/10.1007/s10661-007-9793-0).
- Kaur M, Sharma A, Kaur R, Katnoria JK, Nagpal AK. 2016. Palynological studies of some roadside plants under exposure to traffic stress. *Aerobiologia*. 32(2):245–254. doi: [10.1007/s10453-015-9394-2](https://doi.org/10.1007/s10453-015-9394-2).
- Kebonye NM, Eze PN. 2019. Zirconium as a suitable reference element for estimating potentially toxic element enrichment in treated wastewater discharge vicinity. *Environ Monit Assess*. 191(11):1–15. doi: [10.1007/s10661-019-7812-6](https://doi.org/10.1007/s10661-019-7812-6).
- Kefalati M, Masoudi SF, Abbasi A. 2021. Effect of human body position on gamma radiation dose rate from granite stones. *J Environ Health Sci Eng*. 19(1):933–939. doi: [10.1007/s40201-021-00660-7](https://doi.org/10.1007/s40201-021-00660-7).

- Kierczak J, Pietranik A, Pędziwiatr A. 2021. Ultramafic geoecosystems as a natural source of Ni, Cr, and Co to the environment: a review. *Sci Total Environ.* 755:1–15. doi: [10.1016/j.scitotenv.2020.142620](https://doi.org/10.1016/j.scitotenv.2020.142620).
- Kumar A, Barbhuiya NH, Singh SP. 2022. Magnéli phase titanium sub-oxides synthesis, fabrication and its application for environmental remediation: current status and prospect. *Chemosphere.* 307:1–19. doi: [10.1016/j.chemosphere.2022.135878](https://doi.org/10.1016/j.chemosphere.2022.135878).
- Łokas E, Zaborska A, Sobota I, Gaca P, Milton JA, Kocurek P, Cwanek A. 2019. Airborne radionuclides and heavy metals in high Arctic terrestrial environment as the indicators of sources and transfers of contamination. *The Cryosphere.* 13(7):2075–2086. doi: [10.5194/tc-13-2075-2019](https://doi.org/10.5194/tc-13-2075-2019).
- Luschkova D, Traidl-Hoffmann C, Ludwig A. 2022. Climate change and allergies. *Allergo J Int.* 31(4):114–120. doi: [10.1007/s40629-022-00212-x](https://doi.org/10.1007/s40629-022-00212-x).
- Madaniyazi L, Xerxes S. 2021. Outdoor air pollution and the onset and exacerbation of asthma. *Chronic Dis Transl Med.* 7(2):100–106. doi: [10.1016/j.cdtm.2021.04.003](https://doi.org/10.1016/j.cdtm.2021.04.003).
- Main CE. 2003. Aerobiological, ecological and health linkage. *Environ Int.* 29(2–3):347–349. doi: [10.1016/S0160-4120\(03\)00012-6](https://doi.org/10.1016/S0160-4120(03)00012-6).
- Maina DM, Ndirangu DM, Mangala MM, Boman J, Shepherd K, Gatari MJ. 2016. Environmental implications of high metal content in soils of a titanium mining zone in Kenya. *Environ Sci Pollut R.* 23(21):21431–21440. doi: [10.1007/s11356-016-7249-1](https://doi.org/10.1007/s11356-016-7249-1).
- Miletić A, Lučić M, Onjia A. 2023. Exposure factors in health risk assessment of heavy metal(loid)s in soil and sediment. *Metals.* 13(7):1–28. doi: [10.3390/met13071266](https://doi.org/10.3390/met13071266).
- Mohsenzadeh F, Chehregani A, Yousefi N. 2011. Effect of the heavy metals on developmental stages of ovule, pollen, and root proteins in *Reseda lutea* L. (Resedaceae) *Biol Trace Elem Res.* 143(3):1777–1788. doi: [10.1007/s12011-011-9009-x](https://doi.org/10.1007/s12011-011-9009-x).
- Nautiyal A, Mondal T, Goel A, Dey SK, Mitra D. 2021. Biological effects associated with internal and external contamination of diagnostic nuclear medicine sources: an in vitro study. *Indian J Nucl Med.* 36(3):288–292. doi: [10.4103/ijnm.ijnm_17_21](https://doi.org/10.4103/ijnm.ijnm_17_21).
- Nriagu JO. 1996. A history of global metal pollution. *Science.* 272(5259):223–223. doi: [10.1126/science.272.5259.223](https://doi.org/10.1126/science.272.5259.223).
- Oduber F, Calvo AI, Blanco-Alegre C, Castro A, Vega-Maray AM, Valencia-Barrera RM, Fernández-González D, Fraile R. 2019. Links between recent trends in airborne pollen concentration, meteorological parameters and air pollutants. *Agric For Meteorol.* 264:16–26. doi: [10.1016/j.agrformet.2018.09.023](https://doi.org/10.1016/j.agrformet.2018.09.023).
- R Core Team. 2024. R: A language and environment for statistical computing. Vienna, Austria: R Foundation for Statistical Computing; [accessed 2024]. <https://www.R-project.org/>.
- Robichaud A. 2020. An overview of selected emerging outdoor airborne pollutants and air quality issues: the need to reduce uncertainty about environmental and human impacts. *J Air Waste Manag Assoc.* 70(4):341–378. doi: [10.1080/10962247.2020.1723738](https://doi.org/10.1080/10962247.2020.1723738).
- Senéchal H, Visez N, Charpin D, Shahali Y, Peltre G, Biolley JP, Lhuissier F, Couderc R, Yamada O, Malrat-Domenge A, et al. 2015. A review of the effects of major atmospheric pollutants on pollen grains, pollen content, and allergenicity. *Sci World J.* 2015(1):1–29. doi: [10.1155/2015/940243](https://doi.org/10.1155/2015/940243).
- Sultana Z, Rehman MYA, Khan HK, Malik RN. 2023. Health risk assessment associated with heavy metals through fractionated dust from coal and chromite mines in Pakistan. *Environ Geochem Health.* 45(5):1617–1633. doi: [10.1007/s10653-022-01285-x](https://doi.org/10.1007/s10653-022-01285-x).
- Tan SY, Praveena SM, Abidin EZ, Cheema MS. 2016. A review of heavy metals in indoor dust and its human health-risk implications. *Rev Environ Health.* 31(4):447–456. doi: [10.1515/reveh-2016-0026](https://doi.org/10.1515/reveh-2016-0026).
- Torres P, Llopis AL, Melo CS, Rodrigues A. 2023. Environmental impact of cadmium in a volcanic archipelago: research challenges related to a natural pollution source. *J Mar Sci Eng.* 11(1):1–18. doi: [10.3390/jmse11010100](https://doi.org/10.3390/jmse11010100).
- Turhan Ş, Garad AMK, Hañçerlioğulları A, Kurnaz A, Gören E, Duran C, Karataşlı M, Altıkulaç A, Savacı G, Aydın A. 2020. Ecological assessment of heavy metals in soil around a coal-fired thermal power plant in Turkey. *Environ Earth Sci.* 79(134):1–15. doi: [10.1007/s12665-020-8864-1](https://doi.org/10.1007/s12665-020-8864-1).
- Turhan Ş, Tokat S, Kurnaz A, Altıkulaç A. 2022. Distribution of elemental compositions of zeolite quarries and calculation of radiogenic heat generation. *Int J Environ Anal Chem.* 109(19):7851–7862. doi: [10.1080/03067319.2020.1839439](https://doi.org/10.1080/03067319.2020.1839439).
- Turhan Ş, Köse A, Varinlioğlu A. 2007. Radioactivity levels in some wild edible mushroom species in Turkey. *Isot Environ Health Stud.* 43(3):249–256. doi: [10.1080/10256010701562794](https://doi.org/10.1080/10256010701562794).
- UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). 2008. Sources and effects of ionizing radiation. New York (USA): United Nations Publication: (2010).
- USEPA (U.S. Environmental Protection Agency). 2004. Risk assessment guidance for superfund. Volume 1: human health evaluation manual (part E, supplemental guidance for dermal risk assessment). In: EPA/540/R/99/005, office of superfund remediation and technology innovation, U.S. Environmental protection agency. Washington (DC): U.S. Environmental Protection Agency; p. 1–156.
- USEPA (U.S. Environmental Protection Agency). 2011. Exposure factors handbook (final report). Chapter 5: soil and dust ingestion. Washington (DC): U.S. Environmental Protection Agency.

- USEPA (U.S. Environmental Protection Agency). 2014a. Regional screening levels (RSLs)-generic tables (May 2016). Washington (DC): U.S. Environmental Protection Agency.
- USEPA (U.S. Environmental Protection Agency). 2014b. Regional screening levels (RSLs)-generic tables (May 2016). Washington (DC): U.S. Environmental Protection Agency 2011.
- Vasilevskaya N. 2022. Pollution of the environment and pollen: a review. *Stresses*. 2(4):515–530. doi: [10.3390/stresses2040035](https://doi.org/10.3390/stresses2040035).
- Von Burg R. 1997. Toxicology update: nickel and some nickel compounds. *J Appl Toxicol*. 17(6):425–431. doi: [10.1002/\(SICI\)1099-1263\(199711/12\)17:6<425::AID-JAT460>3.0.CO;2-R](https://doi.org/10.1002/(SICI)1099-1263(199711/12)17:6<425::AID-JAT460>3.0.CO;2-R).
- Wang X, Gong S, Nakamura S, Kurihara K, Suzuki M, Sakamoto K, Miwa M, Lu S. 2009. Air pollutant deposition effect and morphological change of cryptomeria japonica pollen during its transport in urban and mountainous areas of Japan. *WIT Trans Biomed Health*. 11:77–89.
- Wodehouse RP. 1935. *Pollen grains*. (NY): Hafner Publishing Company.
- Xiong ZT, Peng YH. 2001. Response of pollen germination and tube growth to cadmium with special reference to low concentration exposure. *Ecotoxicol Environ Saf*. 48(1):51–55. doi: [10.1006/eesa.2000.2002](https://doi.org/10.1006/eesa.2000.2002).
- Yaroshevsky AA. 2006. Abundances of chemical elements in the Earth's crust. *Geochem Int*. 44(1):48–55. doi: [10.1134/S001670290601006X](https://doi.org/10.1134/S001670290601006X).
- Yoon S, Kim DM, Yu S, Batsaikhan B, Kim T, Yun ST. 2023. Characteristics of soil contamination by potentially toxic elements in mine areas of Mongolia. *Environ Geochem Health*. 46(1):1–18. doi: [10.1007/s10653-023-01812-4](https://doi.org/10.1007/s10653-023-01812-4).
- Yousefi N, Chehregani A, Malayeri BE, Lorestani B, Cheraghi M. 2011. Investigating the effect of heavy metals on developmental stages of anther and pollen in *Chenopodium botrys* L. (*Chenopodiaceae*). *Biol Trace Elem Res*. 140(3):368–376. doi: [10.1007/s12011-010-8701-6](https://doi.org/10.1007/s12011-010-8701-6).
- Zakaly HMM, Abbasi A, Almousa N, Savaşan A. 2024. Naturally occurring radioactive materials (NORM) concentration and health risk assessment of aerosols dust in Nicosia, North Cyprus. *J Radioanal Nucl Chem*. 333(3):1073–1082. doi: [10.1007/s10967-023-09346-w](https://doi.org/10.1007/s10967-023-09346-w).
- Zhang X, We S, Sun Q, Wadood SA, Guo B. 2018. Source identification and spatial distribution of arsenic and heavy metals in agricultural soil around Hunan industrial estate by positive matrix factorization model, principle components analysis and geo statistical analysis. *Ecotoxicol Environ Saf*. 159:354–362. doi: [10.1016/j.ecoenv.2018.04.072](https://doi.org/10.1016/j.ecoenv.2018.04.072).