Science of the Total Environment 754 (2021) 142026

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Fine and coarse particulate matter, trace element content, and associated health risks considering respiratory deposition for Ergene Basin, Thrace

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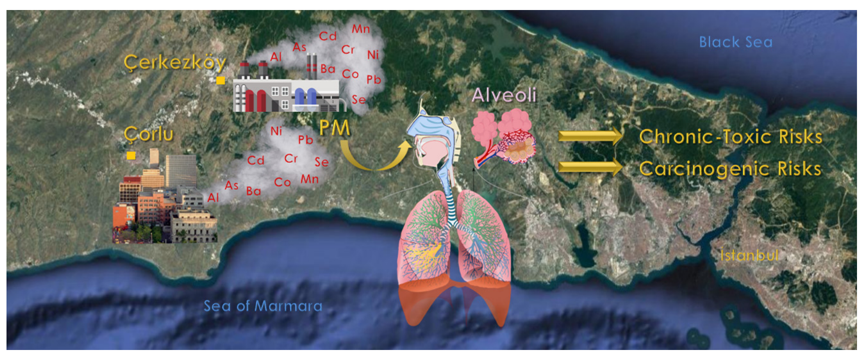
HIGHLIGHTS

- Carcinogenic risk for all PTEs was below the acceptable level except for Cr.
- Exposure to Cr via PM inhalation pathway is important for public health.
- Minor differences in health risks in urban and industrial settings except Cr & Mn
- Similarity in risk levels implies urban pollution being as significant as industrial.
- Considering respiratory deposition pulled cumulative risks below acceptable level.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 April 2020
Received in revised form 24 August 2020
Accepted 26 August 2020
Available online 27 August 2020

Editor: Jianmin Chen

Keywords:
$PM_{2.5}$
$PM_{2.5-10}$
Inhalation
Exposure
Turkey

ABSTRACT

Ergene Basin is located in Thrace, Turkey, where industries are densely populated. This study aimed to determine exposure of people living in Ergene Basin (Çorlu and Çerkezköy) to fine and coarse PM, and its potentially toxic element (PTE) content by considering variation in respiratory airway deposition rates with daily activities and PM particle size by employing deposition models of International Commission on Radiological Protection and Multiple Path Particle Dosimetry. Fine and coarse PM samples were collected daily for a year at points in Çorlu and Çerkezköy representing urban and industrial settings, respectively. A questionnaire survey was conducted in the study area to obtain time-activity budgets, and associated variation was included in the health risk assessment by considering time-activity-dependent inhalation rates. The studied PTEs were Al, As, Ba, Cd, Cr, Co, Mn, Ni, Pb, and Se. The mean fine and coarse PM concentrations were measured as 23 and 14 μg/m^3 in Çorlu, and 22 and 12 μg/m^3 in Çerkezköy, respectively. The only PTE that exceeded acceptable risk in terms of total carcinogenic risk was Cr. Non-carcinogenic risks of all the PTEs including Cr were below the threshold. The use of deposition fractions in the health risk assessment (HRA) calculations was found to prevent overestimation of health risks by at least 91% and 87% for fine and coarse PM, respectively, compared to the regular HRA. Minor differences in risk between Çorlu and Çerkezköy suggest that urban pollution sources could be at least as influential on human health as industrial sources.

1. Introduction

Atmospheric particulate matter is a complex and heterogeneous mixture of organic and inorganic substances, which vary considerably in terms of chemical composition and size. They can be suspended for

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https://doi.org/10.1016/j.scitotenv.2020.142026
0048-9697/

a long time and travel long distances in the atmosphere. Particulate matter (PM) is conventionally classified into two categories based on the size of the particles as $PM_{2.5}$ and PM_{10}. Generally, $PM_{2.5}$ is formed as a consequence of fossil fuel combustion, transformation of nitrogen and sulfur oxides, organics, and high temperature processes while PM_{10} occurs as a result of weathering and soil suspension, construction, coal and oil combustion, industrial dust resuspension (Querol et al., 2013). Because their sources differ, chemical composition of the two categories is generally different and varies spatially. $PM_{2.5}$ with mainly anthropogenic element content reflects anthropogenic sources, whereas PM_{10} with mainly crustal element content reflects natural sources (Sofuoglu et al., 1998; Paode et al., 1998).

Size of the particles is directly related to their potential to cause health problems since it is an important determinant of pulmonary deposition efficiency and oxidative stress (Nel, 2005; Strak et al., 2012; Velali et al., 2016). According to human respiratory tract model of International Commission on Radiological Protection (ICRP), most of the PM_{10} is to be held at head airways and tracheobronchial region of the respiratory system while $PM_{2.5}$ reach the alveolar region where absorption of inhaled trace elements into blood occurs (ICRP, 1994). Oxidative stress is considered to be an important pathway to PM exposure-associated diseases (Liu et al., 2018; Brook et al., 2010; Nel, 2005; Weichenthal et al., 2013). $PM_{2.5}$ is considered to create more oxidative DNA damage than PM_{10} because of the preponderant trace element content which induces more metal-catalyzed reactive oxygen species (Choi et al., 2004; Dai et al., 2002; Galaris and Evangelou, 2002; Ghio et al., 2002; Jeyapaul and Jaiswal, 2000). $PM_{2.5}$ can cause decreased lung function, chronic obstructive pulmonary disease, asthma, and may alter the immune responses of the lungs (Feng et al., 2016; Habre et al., 2014; Wu et al., 2013; Ogino et al., 2014; Duan et al., 2013). PM_{10} is related with increased frequencies of bronchitis, chest illness, chronic cough, and mortality (Medina et al., 2004; Choi et al., 2004; Pope et al., 1995). Furthermore, a recent study emphasized that hospital admissions were increased with the increased PM concentrations, and that there may be connections that have not been observed in previous studies such as genitourinary diseases, skin diseases, and problems in the digestive system in children (Zhu et al., 2019), while PM is categorized as a carcinogen by IARC (IARC, 2016).

Relationship of PM with mortality and morbidity in people is widely known (Deng et al., 2015; Kim et al., 2018; Pope and Dockery, 2006; Samet et al., 2000). Estimation of internal dose of the particles to be deposited in the human lung is essential for particle toxicology and health effects. Aerosol deposition models for the whole lung can be roughly classified into five different categories as semi-empirical regional compartment models, trumpet models, deterministic single-path models, deterministic multiple-path models, and stochastic multiple-path models (Hofmann, 2011). Semi-empirical regional compartment models are primarily based on fitting experimental data to mathematical expressions while the other models are generally termed as “mechanistic models” since they are founded on a mechanistic understanding of physiological and physical mechanisms, among which multiple-path models are more realistic because the airway geometry they use reflects the asymmetric branching structure of the lungs (Hofmann, 2011). Although various models about lung deposition exist in the literature, there are a limited number of studies combining deposition fractions and health risk assessment of PM, which is an imperative issue for prevention of over-estimated health risks (Arı, 2020; Betha et al., 2014; Betha and Balasubramanian, 2014; de la Torre et al., 2018; Huang et al., 2016; Lai et al., 2017; Lyu et al., 2017; Niu et al., 2015; Olawoyin et al., 2018; Othman et al., 2018; Sharma and Balasubramanian, 2018; Sharma and Balasubramanian, 2020; Wang et al., 2019; Xu et al., 2019; You et al., 2017). Multiple-Path Particle Dosimetry (MPPD) and ICRP models are the most frequently used models among the studies besides the model by Volckens and Leith (2003).

Ergene Basin is located in Thrace, Turkey, where textile, chemistry, and metal industries densely populated due to investment allowances and inducements. Although the economic development of the region started after 1980's, there has been an intensive development in the industry after 1990, especially in Tekirdağ province. There are 14 organized industrial zones. About 70% of the industrial assets in the region are in Tekirdağ, 14.0% in Edirne, and 15.4% in Kırklareli provinces. Çorlu and Çerkezköy are the prominent districts of Tekirdağ province in terms of industrial development (Thrace Region Plan (Rep.), 2017). With the increase in industrialization and population, pollution in Ergene Basin has increased significantly. In response, a basin protection plan with regards to water quality has been established. Although there are detailed water action plans for the basin, there is no action plan for air pollution. There are no studies on fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) PM pollution in Ergene Basin. Thereby, in this study we aimed to determine exposure of people living in Ergene Basin (Çorlu and Çerkezköy) to PM and its potentially toxic element (PTE) content through fine and coarse PM by considering variation in respiratory airway deposition rates with daily activities and particle size. We employed two aerosol deposition models to compare the effect of deposition fractions on health risk assessment: the aerosol deposition model of ICRP which is a semi-empirical regional compartment model that uses symmetric lung geometry, and an asymmetric generation model, the MPPD model. The previous studies that incorporated deposition fractions in health risk assessment did not take different activity levels, hence variable inhalation rates that also affect PM exposure into consideration. In this study, we conducted a questionnaire survey to obtain time-activity budgets, and included associated variation in the assessment. Here we present the first individual health risk assessment study that accounted both size-dependent PM deposition onto respiratory airways rather than using only external concentrations and time-activity-dependent inhalation rates.

2. Material and methods

2.1. Study location

Ergene Basin is located in the European part of Turkey, Thrace, which is bordered by Bulgaria and Greece, and Sea of Marmara. The population of the area increases every year because Eastern Thrace conforms the hinterland of fast developing Istanbul metropolitan area providing some of its industrial and energy needs. Historically the region depends on agriculture, especially western and southern parts, now facing population decrease, while population of eastern parts increase along with the industrial development, specifically in Çorlu and Çerkezköy reaching 260,437 and 157,931 in 2017 that is 16% and 20% growth of the population in the last ten years (TSI, 2017). Therefore, Çorlu and Çerkezköy districts were chosen as the study locations in the Ergene Basin. Further information on the climate of the region is provided in Supplementary Material (SM) along with a map showing the area, topography, and the study locations (Fig. S1). Selection of PM sampling locations were based on criteria recommended by the USEPA (1997) that included local meteorology, geology, land use, and the known major PM emission sources. Sampling point in Çorlu (41°09′28.8″N 27°48′40.6″E) is in its sports complex in the town center surrounded by residential areas that include small commercial establishments away from the major sources. Sampling point in Çerkezköy (41°19′06.1″N 27°58′48.4″E) was located next to the Marmara Clean Air Monitoring Station, 5 km away from the City of Çerkezköy next to its Organized Industrial Zone and a town called Kapaklı with a population of about 120,000. The monitoring station is categorized as industrial by the Ministry of Environment and Urbanization. Large industries in Çerkezköy include manufacturing / production of household appliances, pharmaceuticals, rubber-plastics, textile, carpet, metals, cable, chemicals, hydraulic and agricultural machinery, automotive parts while those in Çorlu include chemicals, metals, paper, milk and products, and textile. Çorlu represents the urban setting while Çerkezköy was chosen for its industrial characteristics.

2.2. Sampling and PTE analysis

PM samples were collected daily for 24 h on Whatman polytetrafluoroethylene (PTFE) membrane filters with a diameter of 46.2 mm and 2 μm pore diameter with Partisol Plus dichotomous samplers (Rupprecht and Patashnick Co., Inc.) from June 2015 to May 2016. The filters were stored in a desiccator for 24 h before they were weighed. After weighing, the filters were placed in airtight plastic petri dishes. Flow rates of the sampling were maintained at 1 m^3/h.

PM samples collected on the PTFE membrane filters were analyzed with inductively coupled plasma mass spectrometry (ICP-MS) for Al, As, Ba,Cd, Cr, Co, Mn, Ni, Pb, and Se. Prior to the PTE analysis, the filters were dissolved in a microwave unit using 4 mL of HNO_3 (60%), 0.5 mL of HF (40%). After digestion samples were diluted with deionized water in a 25 mL volumetric flask and thereafter analyzed with a Perkin-Elmer ICP-MS.

2.3. Quality assurance and quality control (QA/QC)

Precision was estimated as the relative standard deviation of replicate sample analysis ($n = 7$). Recovery efficiency of the extraction method was determined by analysis of blank filters spiked with a standard reference material (SPS-SW2 Batch 135, Spectrapure Standards, Oslo, Norway) treated as samples ($n = 5$). Accuracy is reported as the absolute percent difference between the spiked and measured concentrations. Precision and accuracy are listed for each analyte in Table S3. The average (±Std.Dev.) precision and accuracy of the method were 7.0 ± 2.1% and 9.0 ± 3.2%, respectively. Method detection limits (MDL) were calculated as mean blank value +3SD. The MDLs for Al, As, Ba, Cd, Cr, Co, Mn, Ni, Pb, and Se were 0.01, 0.014, 0.10, 0.02, 0.01, 0.001, 0.1, 0.02, 0.01, and 0.2 ng/m^3, respectively.

2.4. Exposure – risk assessment

An individual exposure-risk assessment for PTEs in PM was conducted using measured concentrations at the two sampling stations and data collected by a questionnaire survey. The questionnaire was self-administered by 94 participants (22 in Çerkezköy and 72 in Çorlu) who volunteered out of 100 randomly selected people to determine their daily activities. It also inquired about other exposure related variables to be used in estimation of their exposure to PTEs via airborne PM. Participants were asked to record their responses daily for a week in winter. Participants' daily activities were divided into five groups in accordance with USEPA (2011) as sleep or nap, sedentary and passive activities (such as sitting or lying quietly), light intensity activities (e.g. houseworks such as cooking, washing dishes, ironing), moderate intensity activities (e.g. walking, gardening), and high intensity activities (e.g. aerobics, running) since their inhalation rates depend on consumed energy.

Health risk assessment was carried out by using the information from the questionnaires and the measured PTE concentrations. On average, people spend 74 to 79% of their time indoors (Gungormus et al., 2014; Yılmaz Civan et al., 2015). However, due to great variety of indoor microenvironments, it generally is not possible to perform PM sampling and PTE analysis not even at one point in homes of the participants due to large volumes of air needed for PTE quantification, in addition to the time, personnel, and budget considerations. Therefore, as a conservative approach, concentrations measured at the two ambient air stations were used for all estimations with the assumption of 100% penetration of PM into the buildings (Gungormus et al., 2014). For taking the difference in respiratory deposition rates in coarse and fine particulate matter, and prevention of overestimation in exposure and health risks, the computational model of MPPD version 3.04 (ARA, n.d.), and the simplified equations of the ICRP model (Hinds, 1999) were used. Deposition fractions of inhaled fine and coarse particulate matter in the three main parts of the human respiratory tract, i.e., head airways (HA), tracheobronchial region (TB), and alveolar region (AR) were calculated with the ICRP and the MPPD models. Deposition fraction is defined as the fraction of inhaled particles that deposit in the lung. The dominant mechanisms for particle deposition are diffusion, sedimentation, and impaction. Deposition fraction calculations in the MPPD model based on these mechanisms can be found elsewhere (Anjilvel and Asgharian, 1995).

The MPPD model (ARA, n.d.) provides eight options for human lung morphometry. In this study, stochastic lung airway morphometry is adopted. Unlike the ICRP model, which uses a symmetric lung geometry model and does not reflect asymmetry of the lungs, MPPD model is based on stochastic lung airway morphometry model (Koblinger and Hofmann, 1990) that represents an asymmetric and more realistic geometry of the human lung to obtain more realistic deposition results. Default values of functional residual capacity (3300 mL), upper respiratory tract volume (50 mL), breathing frequency (12 per minute), and tidal volume (625 mL) which provided by the model itself for adults were used in the estimations of deposition fractions. Particles were assumed to be spherical with a density of 1.0 g/cm^3. Among the breathing conditions of nasal, oral, oronasal, endotracheal available in the model, nasal breathing was chosen, and all of the particles were assumed to enter the respiratory tract through the nose.

Deposition efficiencies of size-specific particles were also estimated with simplified equations of the ICRP model in the HA (Eq. (2)) based on inhalable fraction (Eq. (1)), TB (Eq. (3)), and AR (Eq. (4)).

$$IF_i = 1-0.5\left(1-\frac{1}{1+0.00076D_{p,i}^{2.8}}\right) \tag{1}$$

$$DF_{HA,i} = IF_i \times \left(\frac{1}{1+\exp\left(6.84+1.183\ln D_{p,i}\right)}+\frac{1}{1+\exp\left(0.924-1.885\ln D_{p,i}\right)}\right) \tag{2}$$

$$DF_{TB,i} = \frac{0.00352}{D_{p,i}} \times\left[\exp\left(-0.234\left(\ln D_{p,i}+3.40\right)^2\right)+63.9\exp\left(-0.819\left(\ln D_{p,i}-1.61\right)^2\right)\right] \tag{3}$$

$$DF_{AR,i} = \frac{0.0155}{D_{p,i}} \times\left[\exp\left(-0.416\left(\ln D_{p,i}+2.84\right)^2\right)+19.11\exp\left(-0.482\left(\ln D_{p,i}-1.362\right)^2\right)\right] \tag{4}$$

where IF_i is the inhalable fraction, $D_{p,i}$ (μm) is the diameter of particle, $DF_{HA,i}$, $DF_{TB,i}$, $DF_{AR,i}$ are the deposition fractions of particle in the head airways, tracheobronchial region, and alveolar region, respectively.

Median diameters of fine and coarse particles in atmospheric mass size distribution of urban particulate matter (0.6 and 5 μm, respectively) were assumed as representatives of fine and coarse particulate matter (Sofuoglu et al., 1998; Kuzu et al., 2013), and used for the calculations of deposition fractions in both deposition models. Ratios of inorganic arsenic to total arsenic species for fine and coarse particulate matter were assumed as 0.77 and 0.78, respectively (Tziaras et al., 2015). The ratio of hexavalent chromium to total chromium species was assumed to be 20% for fine and coarse particulate matter (Torkmahalleh et al., 2013).

PTE exposures were estimated using Eq. (5).

$$CDI = \frac{C \times DF_i \times IR}{BW} \times \frac{ED \times EF}{AT} \tag{5}$$

where CDI is the chronic daily intake (mg/kg-day), C is the PTE concentration in the air (mg/m^3), DF_i is the deposition efficiency of particles in each region, IR is the inhalation rate (m^3/min), ED is the exposure duration (years), EF is the exposure frequency (min/year), BW is the body weight (kg) reported by the participant in the questionnaire, and AT is the averaging time (days). The second term in the equation is unity for chronic–toxic risk assessment, while ED and AT were assumed as 30 years and 75 × 365 days (the average lifespan is reported to be 75 years in Turkey; TSI, 2017), respectively, for the carcinogenic risk assessment. In this context, activity times were calculated based on the information obtained from the questionnaires. PTE exposures were calculated using specific inhalation rates for each activity, gender, and age of the participants (USEPA, 2011). Times that could not be related with a specific activity during the day were identified as non-activity times. For non-activity times, median inhalation rates were used according to the age and gender of the person, and the exposures were calculated subsequently. Total exposures of each participant were found by summing non-activity and activity exposures.

Carcinogenic risk associated with inhalation exposure was calculated with Eq. (6).

$$R = CDI \times SF \tag{6}$$

where R is the cancer risk, and SF is the slope factor of the PTE (mg/kg-day)$^{-1}$. Values of SF were calculated from the available inhalation unit risk values (IUR; (μg/m^3)$^{-1}$) (SM Table S2).

The hazard quotient (HQ) was calculated to estimate chronic–toxic risk using Eq. (7).

$$HQ = CDI/RfD \tag{7}$$

where RfD is the reference dose (mg/kg-day). RfD values were calculated from the reference concentrations (RfC; mg/m^3) provided by different sources as shown in SM Table S2. Cumulative non-carcinogenic and carcinogenic risks were also calculated as sum of the HQs and sum of the carcinogenic risks of all PTEs, respectively, assuming simple additivity for the sake of estimating a total risk level.

2.5. Statistical tests

Normality of the PTE concentrations was tested with the Anderson-Darling normality test. For the groups with normal distribution, F-test was applied to test the equality of the variances and accordingly two-tailed *t*-tests assuming equal or unequal variances were performed to infer on equality of the means. Statistical differences on the means between groups with non-normal distributions were determined with Mann-Whitney *U* test. The significance level of 0.05 was used for all tests except Anderson-Darling normality test which was 0.005. All statistical tests were performed with Minitab 19 software (Minitab Inc., USA).

3. Results and discussion

3.1. Questionnaire data

Randomly recruited 94 people (22 from Çerkezköy and 72 from Çorlu) participated in this study. All of the participants were adult individuals whose age and gender information are given in Table 1. Body weights of the male Çorlu participants ranged from 60 kg to 110 kg with an average of 82 kg. Female Çorlu participants' body weights ranged from 45 kg to 100 kg with a mean value of 69 kg. Male participants from Çerkezköy had body weights ranging from 65 kg to 115 kg with an average of 82 kg. Female Çerkezköy participants' body weights ranged 51 kg to 92 kg with a mean value of 72 kg. The average daily activity durations are also given in Table 1. For the participants from Çorlu, daily average sleep or nap, sedentary and passive, light intensity, and moderate intensity activities were respectively 433, 73, 146, and 29 min. The shortest activity time belonged to high intensity activities with an average of 4 min. The participants in Çerkezköy have a higher duration of high intensity (7 min) and sedentary and passive (102 min) activities, while durations for the other activities were as follows: 445 min of sleep or nap,149 min of light intensity, and 24 min of moderate intensity activities, respectively.

Table 1
Participant characteristics and daily average activity times.

	Çorlu		Çerkezköy	
	N	%	N	%
Gender				
Female	31	43	7	32
Male	41	57	15	68
Total	**72**	100	**22**	100
Age				
18–21	0	0	2	9
21–31	16	22	5	23
31–41	18	25	4	18
41–51	22	31	3	14
51–61	7	10	4	18
61–71	4	6	3	14
71–81	2	3	1	5
81+	3	4	0	0

Daily averages of activities (min)	Çorlu	Çerkezköy
Sleep or nap	433	445
Sedentary and passive	73	102
Light intensity	146	149
Moderate intensity	29	24
High intensity	4	7

3.2. Particulate matter and PTE concentrations

Table 2 provides descriptive statistics of measured fine and coarse PM and PTE concentrations for the sampling period. Fine and coarse PM concentrations in Çorlu district ranged from 2.31 to 156 μg/m^3 and 1.30 to 355 μg/m^3, respectively. For Çerkezköy, fine and coarse PM concentrations varied between 2.63 and 119 μg/m^3 and 1.29 to 49.0 μg/m^3, respectively. Even though average PM_{10} concentrations were lower than annual limit of Turkish national ambient air quality standards (40.0 μg/m^3), the daily limit value (50.0 μg/m^3) was exceeded 54 times in Çorlu and 67 times in Çerkezköy during the sampling period (AQAAMD, 2008). Turkish national ambient air quality standards does not set a limit for $PM_{2.5}$ but the average annual concentrations in both districts were greater than the limit value of 10 μg/m^3 recommended by the World Health Organization (WHO, 2006). All coarse-PM PTE concentrations except for As were significantly higher in Çerkezköy than Çorlu. However, for fine-PM, significantly higher Al, Cr, Co, and Pb concentrations were observed in Çorlu ($p < 0.05$). There was not any statistical difference for coarse-PM Co concentrations and fine-PM Mn and Ni concentrations between Çorlu and Çerkezköy districts ($p > 0.05$).

PTE and PM concentrations are compared with the other provinces of Turkey in Table 3. Fine PM concentrations were similar to other provinces except Bursa which has almost two times the concentrations of this study. However, coarse-PM concentrations were lower compared to the other provinces. Al content of fine PM from both Çorlu and Çerkezköy districts were exceptionally high at least 1.5 times of the other provinces yet Al content of the coarse PM was lower in this study. Cr and Se were the other PTEs that were relatively higher among the provinces of Turkey. The mean concentration of Se in summer which is 2.76 ng/m^3 in Çerkezköy increased significantly to 4.11 ng/m^3 in winter ($p < 0.05$). There were also a statistically significant increase in Cr (1.28 ng/m^3), As (0.11 ng/m^3), and Pb (3.57 ng/m^3) concentrations from summer to winter which may indicate the effect of fossil fuel combustion in Çerkezköy ($p < 0.05$).

When we compare the concentrations of fine PM with those measured in other countries (also in Table 3), it is seen that the concentrations are about the same level in the cities of Palermo and Barcelona, and higher than that in Navarra, Spain and Fukue Island, Japan. Although the Al and Cr concentrations in $PM_{2.5}$ are much higher than those of other countries, As concentrations were found quite the contrary, such as one-fourth of Barcelona, one-fifth of Palermo, and one-eighth of Zhengzhou. Coarse-PM PTE concentrations were found at

Table 2
Annual descriptive statistics of fine and coarse PM ($\mu g/m^3$) and PTE (ng/m^3) concentrations in Çorlu and Çerkezköy.

	Çorlu										Çerkezköy									
	Fine PM					Coarse PM					Fine PM					Coarse PM				
	Min	Max	Mean	Median	SD	Min	Max	Mean	Median	SD	Min	Max	Mean	Median	SD	Min	Max	Mean	Median	SD
PM	2.31	156	23.4	20.6	13.5	1.30	355	13.7	9.65	22.0	2.63	119	21.7	18.2	14.1	1.29	49.0	12.4	9.89	8.33
Al	208	1962	844	752	440	30.8	1953	391	277	345	83.0	1722	565	487	337	36.8	1225	346	280	249
As	0.05	0.90	0.52	0.52	0.18	0.01	0.92	0.43	0.43	0.15	0.06	0.87	0.55	0.57	0.17	0.14	0.91	0.39	0.38	0.13
Ba	3.08	26.0	14.6	14.6	5.38	0.62	26.3	10.7	10.8	5.22	1.35	35.8	16.2	16.4	6.76	1.26	29.3	11.8	12.3	4.80
Cd	0.05	2.49	0.62	0.47	0.46	0.00	1.69	0.38	0.27	0.35	0.19	1.51	0.83	0.83	0.25	0.01	3.23	0.59	0.43	0.54
Cr	6.98	20.8	14.4	14.5	2.80	0.55	18.3	8.64	9.14	3.25	5.42	29.0	13.2	12.8	3.45	3.04	26.7	12.4	12.0	4.29
Co	0.03	0.99	0.35	0.29	0.25	0.01	0.79	0.14	0.10	0.14	0.00	0.91	0.14	0.08	0.15	0.01	0.61	0.16	0.11	0.14
Pb	2.12	23.2	13.2	13.3	5.25	0.81	24.2	8.77	8.47	4.62	0.17	55.5	10.8	8.78	7.73	0.11	59.4	11.7	11.8	6.74
Mn	2.08	28.9	11.5	10.6	6.17	0.12	9.54	2.24	2.01	1.57	0.60	75.5	12.3	9.38	10.6	0.18	16.6	5.71	4.42	3.89
Ni	0.36	4.61	1.95	1.93	0.98	0.06	3.20	1.49	1.54	0.76	0.13	4.52	1.86	1.82	1.07	0.10	5.74	1.90	2.00	0.91
Se	0.61	4.68	3.18	3.32	0.97	0.68	3.95	1.98	1.70	0.79	0.80	7.82	3.45	3.55	1.06	0.52	4.97	2.60	2.73	0.80

least four times that of those in London and Cr and Mn concentrations were even higher than those in Zhengzhou. Comparison of PM levels at an industrial site in Singapore (You et al., 2017) with Çerkezköy, which represents the industrial setting of our study, shows that even though concentrations of coarse PM are lower in Çerkezköy, content of PM were found to be richer in terms of many PTEs such as Al, Ba, Cr, Pb, and Se, whereas the Mn content is one half that of Singapore and the Ni content is one-sixth. Olawoyin et al. (2018) studied heavy metal contamination and related health risks in Chemical Valley Sarnia, Ontario, Canada. Sarnia city hosts 40% of the industries in the country and therefore named as the "Chemical Valley Sarnia" (CVS). Annual median fine-PM concentration in the CVS was reported to be 87 ± 8 $\mu g/m^3$. Median elemental concentrations for As, Cd, Cr, Pb, and Ni were 26.8, 23.7, 6.83, 154, and 68.7 ng/m^3 for fine-PM sampled in the closest area to CVS. Even though the other PTE concentrations were higher than ours, Cr concentrations were one half that of in Çerkezköy but with large standard deviation (4.5 ng/m^3) in CVS makes upper-bound level of one-SD band (11.3 ng/m^3) comparable to the lower-bound level of one-SD band (9.35 ng/m^3) in Çerkezköy.

3.3. PTE exposure and health risks

Deposition fractions of different particle sizes according to ICRP and MPPD models are presented in the SM Figs. S2 and S3. According to ICRP model, deposition fractions of fine PM for HA, TB, and AR were 13.4%, 1.0%, and 9.4%, and 86%, 4.5%, and 5.8% for coarse PM, respectively. Total deposition fraction of fine PM was 24% while it was 97% for coarse PM. In the MPPD model, deposition fractions of fine PM for HA, TB, and AR were 6.6%, 4.5%, and 7.0%, and 74.6%, 9.0%, and 13.3% for coarse PM, respectively. Total deposition fraction was 18% for fine PM and 97% for coarse PM. When the two models are compared, it can be seen that MPPD gives lower HA and higher TB deposition fractions than those of ICRP. It is also noteworthy that the AR deposition fraction value for coarse PM is 13% according to the MPPD model while it is 5.8% with the ICRP model. It can be seen in Figs. S2 and S3 that particles between 0.1 and 1 μm have the lowest AR deposition fractions according to the both models. The airway surfaces of HA and TB are covered with a mucus layer that is propelled by ciliary action to the gastrointestinal tract (Hinds, 1999). However, AR does not have a protective mucus layer because of its gas exchange function (Harrison and Yin, 2000), thus particles accumulated in this region are cleared slowly over months or years. Consequently, PM exposure into AR poses significant risks to human health (Lippmann et al., 1980). Interestingly, it can be observed that the Median Mass Aerodynamic Diameter (MMAD) value assumed for coarse PM penetrated deeper in the lungs than MMAD of fine PM according to MPPD model.

PTE exposures from inhalation of fine and coarse particulate matter were calculated for 94 participants by using the deposition fractions, measured average PTE concentrations, and personal information obtained from the questionnaires. Descriptive statistics for the estimated exposures are presented in Table 4. Average CDIs of PTEs reflect the deposition fractions. The lowest CDIs were estimated for TB where the lowest deposition fractions were calculated. Distributions of carcinogenic and non-carcinogenic risks among HA, TB, and AR are shown in Figs. 1 and 2, respectively. While the proportion of AR appears to be importantly increased in the fine PM compared to that of the coarse PM with regards to ICRP model, it is reversed for the MPPD model due to higher deposition fractions of coarse PM. Considering the sum of the carcinogenic risks of HA, TB, and AR, all PTEs remained within the acceptable risk limit ($<10^{-6}$) except for Cr that remained $<10^{-5}$. Table 3 demonstrates that concentrations of Cr in Çorlu and Çerkezköy are above the other provinces of Turkey. All non-carcinogenic risks are below the threshold (1.0) as can be seen from Fig. 2, along with the highest non-carcinogenic risks belonging to Mn and Al, respectively. Also, it is noteworthy that the chronic-toxic risks associated with Cr are lower than the other PTEs except Se. This points out that exposure

Table 3
PM (μg/m^3) and PTE (ng/m^3) concentrations (Average ± SD) reported in the literature in Turkey and other countries.

	Reference	Study area	Country	Characteristic	Season	PM	Al	As	Ba	Cd	Cr	Co	Pb	Mn	Ni	Se
$PM_{2.5}$	This study	Çorlu/Tekirdağ	Turkey	Urban	Annual	23.4 ± 13.5	844 ± 440	0.52 ± 0.18	14.6 ± 5.38	0.62 ± 0.46	14.4 ± 2.80	0.35 ± 0.25	13.2 ± 5.25	11.5 ± 6.17	1.95 ± 0.98	3.18 ± 0.97
		Çerkezköy/Tekirdağ	Turkey	Industrial		21.7 ± 14.1	565 ± 337	0.55 ± 0.17	16.2 ± 6.76	0.83 ± 0.25	13.2 ± 3.45	0.14 ± 0.15	10.8 ± 7.73	12.3 ± 10.6	1.86 ± 1.07	3.45 ± 1.06
	Pekey et al. (2010)	Kocaeli	Turkey	Urban	Summer	23.5 ± 9.7	87 ± 58	2.0 ± 2.0	–	–	4.0 ± 3.0	–	47 ± 46	49 ± 49	2.0 ± 2.0	–
					Winter	21.8 ± 12.8	154 ± 122	4.0 ± 3.0	–	–	9.0 ± 4.0	–	72 ± 66	28 ± 24	3.0 ± 1.0	–
	Tecer et al. (2012)	City Center/Zonguldak	Turkey	Urban	Annual	29.6 ± 15.4	94 ± 258	–	–	–	3.8 ± 1.8	–	11.9 ± 7.4	8 ± 6	3.0 ± 1.2	–
	Summak et al. (2017)	İstanbul	Turkey	Urban	Annual	31.25	360.4	–	–	2.7	6.6	3.1	24.4	–	11.1	–
	Koçak et al. (2007)	Erdemli/Mersin	Turkey	Rural	Annual	9.7 ± 5.9	–	–	–	–	1.8 ± 3.5	–	–	1.8 ± 1.7	1.6 ± 1.3	–
	Kendall et al. (2011)	Nilüfer/Bursa	Turkey	Urban	Annual	53	–	4	20	1.3	9.4	–	32	22	7.7	1.9
	Öztürk and Keleş (2016)	City Center/Bolu	Turkey	Urban	Annual	20 ± 15	255 ± 259	0.93 ± 0.52	9 ± 1	–	–	–	16 ± 10	14 ± 11	–	–
	Minguillon et al. (2012)	Barcelona	Spain	Urban	Annual	20	–	2	5.2	0.4	3.4	–	10	38	7.1	–
	Dongarra et al. (2010)	Palermo	Italy	Urban	Annual	23.5 ± 7.0	411 ± 308	2.5 ± 1.1	45 ± 26	–	24 ± 20	0.2 ± 0.1	15 ± 8.9	4.5 ± 3.8	13 ± 8.6	–
	Wang et al. (2019)	Zhengzhou	China	Urban	Annual	–	420 ± 119	3.79 ± 1.5	18.0 ± 5.6	17.9 ± 8.6	6.47 ± 1.0		18.8 ± 12	13.5 ± 4.0	3.18 ± 1.6	2.76 ± 1.0
	Aldabe et al. (2011)	Iturrama/Navarra	Spain	Urban	Annual	15.38	–	0.16	12.08	0.05	2.39	0.99	2.29	2.57	1.31	0.23
	Aldabe et al. (2011)	Plaza de la Cruz/Navarra	Spain	Traffic	Annual	17.42	–	0.11	6.09	0.13	1.55	0.42	3.36	2.29	0.89	0.23
	Hidemori et al. (2014)	Fukue Island	Japan	Rural	Spring	16.5 ± 9.1	72 ± 46	0.7 ± 0.5	1.6 ± 1.5	0.1 ± 0.1	–	–	3.9 ± 2.6	3.7 ± 3.2	–	–
	Visser et al. (2015)	North Kensington/London	UK	Urban	Winter	–	17.1	–	2.7	–	0.6	–	0.4	1.7	0.1	–
$PM_{2.5–10}$	This Study	Çorlu/Tekirdağ	Turkey	Urban	Annual	13.7 ± 22.0	391 ± 345	0.43 ± 0.15	10.7 ± 5.22	0.38 ± 0.35	8.64 ± 3.25	0.14 ± 0.14	8.77 ± 4.62	2.24 ± 1.57	1.49 ± 0.76	1.98 ± 0.79
		Çerkezköy/Tekirdağ	Turkey	Industrial		12.4 ± 8.33	346 ± 249	0.39 ± 0.13	11.8 ± 4.80	0.59 ± 0.54	12.4 ± 4.29	0.16 ± 0.14	11.7 ± 6.74	5.71 ± 3.89	1.90 ± 0.91	2.60 ± 0.80
	Tecer et al. (2012)	City Center/Zonguldak	Turkey	Urban	Annual	24.9 ± 23.5	495 ± 666	–	–	–	3.7 ± 1.6	–	7.3 ± 5.0	12 ± 10	2.9 ± 1.0	–
	Koçak et al. (2007)	Erdemli/Mersin	Turkey	Rural	Annual	26.7 ± 26.4	–	–	–	–	3.9 ± 5.3	–	–	5.8 ± 7.5	2.1 ± 1.8	–
	Öztürk and Keleş (2016)	City Center/Bolu	Turkey	Urban	Annual	31 ± 22	291 ± 346	0.93 ± 0.63	12 ± 6	–	–	–	18 ± 14	12 ± 8	–	–
	Wang et al. (2019)	Zhengzhou	China	Urban	Annual	–	196 ± 73	4.66 ± 0.4	15.2 ± 2.3	21.0 ± 7.4	9.86 ± 4.1	–	12.5 ± 7.3	8.28 ± 3.6	6.50 ± 3.9	3.49 ± 1.3
	Visser et al. (2015)	North Kensington/London	UK	Urban	Winter	–	30.8	–	4.3	–	1.2	–	0.4	2.4	0.4	–
	You et al. (2017)	Singapore	Singapore	Bus stop	Autumn	10.27	540.67 ± 17.48	–	8.77 ± 0.26	–	3.28 ± 0.26	–	10.26 ± 0.19	2.71 ± 0.17	9.33 ± 0.39	0.35 ± 0.02
	You et al. (2017)	Singapore	Singapore	Highway	Autumn	10.93	1229 ± 28.11	–	34.13 ± 0.67	–	19.88 ± 0.98	–	19.38 ± 0.04	26.32 ± 0.57	9.19 ± 0.39	2.03 ± 0.03
	You et al. (2017)	Singapore	Singapore	Industrial	Autumn	13.77	240.53 ± 8.38	–	6.27 ± 0.31	–	6.42 ± 0.13	–	3.65 ± 0.01	9.55 ± 0.18	11.94 ± 0.39	0.58 ± 0.02
PM_{10}	Pekey et al. (2010)	Kocaeli	Turkey	Urban	Summer	59.9 ± 18.2	399 ± 156	2.0 ± 1.0	–	–	10.0 ± 6.0	–	78 ± 64	121 ± 83	1.0 ± 1.0	–
					Winter	102.3 ± 50.7	655 ± 489	7.0 ± 7.0	–	–	23.0 ± 10.0	–	159 ± 129	188 ± 153	5.0 ± 1.0	–
	Summak et al. (2017)	İstanbul	Turkey	Urban	Annual	45.53	821.1	–	–	4	10	4.1	36.9	–	15.5	–
	Bozkurt et al. (2018)	Düzce	Turkey	Urban	Summer	53.2 ± 14.7	4500 ± 1500	0.8 ± 0.1	5.4 ± 1.0	0.3 ± 0.2	11.9 ± 2.5	0.3 ± 0.1	5.3 ± 1.2	13.6 ± 5.3	6.7 ± 1.8	0.7 ± 0.3
					Winter	86.4 ± 56.0	5300 ± 5600	1.8 ± 1.0	21.4 ± 17.1	0.6 ± 0.3	14.7 ± 6.4	0.7 ± 0.4	21.1 ± 16.2	29.0 ± 28.1	11.8 ± 4.4	0.9 ± 0.6
	Gaga et al. (2012)	City Center/Eskişehir	Turkey	Urban	Summer	65.1 ± 21.0	494.1 ± 204.7	1.2 ± 1.2	8.8 ± 5.5	0.5 ± 0.5	13.4 ± 15.1	0.4 ± 0.5	5.8 ± 5.1	12.4 ± 5.2	3.1 ± 1.8	0.6 ± 0.2
					Winter	55.9 ± 19.1	451.4 ± 585.0	3.2 ± 2.1	4.6 ± 4.6	0.9 ± 1.4	4.0 ± 3.3	0.2 ± 0.2	8.7 ± 7.5	9.2 ± 10.8	1.3 ± 1.2	0.6 ± 0.5
	Lyu et al. (2017)	Zhuzhou	China	Urban	Annual	120–215	9200	19	71	5.4	42	–	159	108	39	–
	Aldabe et al. (2011)	Bertiz/Navarra	Spain	Rural	Annual	15.23	–	0.1	20.03	0.05	1.32	0.12	2.88	2.2	0.9	0.27
	Aldabe et al. (2011)	Iturrama/Navarra	Spain	Urban	Annual	25.91	–	0.21	18.91	0.04	2.81	1.2	3.33	6.88	2.21	0.43
	Aldabe et al. (2011)	Plaza de la Cruz/Navarra	Spain	Traffic	Annual	28.23	–	0.13	10.3	0.12	2.33	0.47	4.33	7.07	0.98	0.54

to Cr via inhalation of PM may be more significant for public health considering carcinogenic effects than chronic-toxic effects.

Carcinogenic and non-carcinogenic risks of PTEs for the AR are demonstrated in Figs. 3 and 4, respectively. Carcinogenic risks of PTEs in fine fraction ranged from 1.7×10^{-9} to 1.5×10^{-6}. Although Cr was still the highest carcinogenic risk PTE in Çerkezköy and Çorlu, the risk levels were below the acceptable risk limit for 95% of the participants due to low deposition fraction into AR. For the coarse fraction, carcinogenic risk of PTEs varied between 1.1×10^{-9} and 1.3×10^{-6}. Co, As, and Cd are the other PTEs with higher carcinogenic risks following Cr. Due to the higher deposition fractions, the carcinogenic risks of PTEs in coarse PM estimated with the MPPD model were found to be two times higher than those calculated with ICRP. The higher non-carcinogenic risks belonged to Mn and Al as shown in Fig. 4. HQs of PTEs in fine fraction ranged from 4.5×10^{-6} to 2.4×10^{-2} while their range was 2.4×10^{-6}–1.3×10^{-2} for coarse fraction. The mean and median values of non-carcinogenic risks were found very close except for Mn and Al. This is probably due to their high standard deviations in PM concentrations as can be seen in Table 2. Çerkezköy and Çorlu represented the industrial and urban settings, respectively, therefore higher health risks may be expected in Çerkezköy, but only slight differences were seen with Çorlu. According to the ICRP model, median non-carcinogenic and carcinogenic risks of Co in Çorlu were 4.1×10^{-3} and 8.4×10^{-8} for exposure to fine PM while in Çerkezköy these values were one-third of those in Çorlu, 1.4×10^{-3} and 2.9×10^{-8}, respectively. Even though health risks estimated for fine PM with the MPPD model were slightly lower than those of ICRP, non-carcinogenic and carcinogenic risks for Co in Çerkezköy are still one-third of those in Çorlu. PTEs that had a higher chronic-toxic risk in Çerkezköy compared to Çorlu were only Mn and Cr.

We compared the daily inhalation intake values, carcinogenic and non-carcinogenic risks of PTEs with those reported in the literature. CDIs of PTEs into AR were found to be at most 20% of the study performed for an urban site of China near non-ferrous metal smelting facilities where the concentrations of PM_{10} ranged from 120 to 215 μg/m^3 (Lyu et al., 2017). Among the PTEs, CDIs of Ba and Cd were the closest to those in the industrially polluted city of China. Even though the concentrations of PTEs and deposition fractions in this study are lower than those of Lyu et al. (2017), non-carcinogenic risks of Cr, Co, and especially Mn are higher in this study. This is probably because they used oral reference doses for the estimation of inhalation daily intakes instead of calculation of reference doses from reference concentrations, and low inhalation rate (0.45 m^3/h or 10.8 m^3/day), which show the importance of using accurate inhalation rates. Most of the things that people do in daily life are light or moderate intensity activities (Jette et al., 1990) and the average inhalation rate of an adult in light intensity activities is 15.3 m^3/day for women and 18.7 m^3/day for men (USEPA, 2011). Inaccurate use of inhalation rates may cause underestimation of actual risks; therefore time-activity information is very important in risk assessment studies in terms of activity patterns of the participants, and estimating health risks accordingly. Also, considering arsenic and cobalt, the use of oral reference doses instead of inhalation ones will result in 70-fold and 150-fold lower inhalation non-carcinogenic risks, respectively.

Wang et al. (2019) conducted a similar study in a Chinese metropol city where PTE concentrations ranged between 2.76 and 420 ng/m^3 and annual average $PM_{2.5}$ concentration was 78 μg/m^3. MPPD model was used for the calculations of deposition fractions. HQ values of Al (3.1×10^{-3}), Ba (5×10^{-3}), and Mn (3.3×10^{-2}) were found to be very close to the values estimated in this study. However, carcinogenic risk values of As (4.5×10^{-6}), Cd (9.7×10^{-6}), Cr (1.6×10^{-4}), Pb (3.3×10^{-7}), and Ni (2.2×10^{-7}) were well above the levels reported in this study. Huang et al. (2016) estimated potential health risks for residents around an e-waste recycling zone in South China by considering deposition fractions with ICRP modeling. CDIs of As, Cd, Cr, Co, Pb, Mn, Ni, and Se for AR varied between 2×10^{-8} and 4.2×10^{-6} mg/kg-day which is at least three times the CDIs in this study. This is plausible considering high PTE concentrations in their study area (0.83–150 ng/m^3). Most of the PTEs except Mn had HQs lower than the threshold, and on the contrary, majority of PTEs except Ni had carcinogenic risks above the acceptable risk limit. Similar to this study, the carcinogenic risk of Cr was above the acceptable limit, although the hazard quotient value was lower than the threshold.

Sharma and Balasubramanian (2018) investigated potential health risks during the 2015 haze episode in Singapore caused by landscape fires in Indonesia. They conducted PM sampling for seven days, out of which two were light-haze days, three were moderate-haze days, and two were severe-haze days. During the episode, probability distribution functions reflecting variations of $PM_{2.5}$ concentrations showed that mean concentrations for Al, Cr, Mn, Co, Ni, As, Se, Cd, Ba, and Pb were 1271, 29.8, 6.09, 0.13, 6.78, 4.36, 1.58, 0.16, 10.5, and 4.53 ng/m^3, respectively. All of the PTEs except for Al, As, Cr, and Ni, had lower concentrations compared to those of our study. ICRP model was used for estimation of deposition doses in the human lung. Similar to our results, Cr had the highest total non-carcinogenic (1.22×10^{-3}) and carcinogenic risk (1.46×10^{-6}) but exceeded the acceptable risk limit. Total carcinogenic risk levels for As (1.20×10^{-7}), Cr (1.46×10^{-6}), and Ni (2.99×10^{-8}) are similar to those estimated in this study. Another study conducted concerning potential health risks of PM in Singapore (You et al., 2017) measured PM concentrations alongside a highway, at an industrial site, and at a bus stop. As shown in Table 3, compared to their results obtained at an industrial site, coarse-PM of Çerkezköy was richer in terms of Al, Ba, Cr, Pb, and Se. However, probably due to equations of deposition models (Chan et al., 1980; Cheng, 2003; Cohen and Asgharian, 1990; Kim and Fisher, 1999; Kim and Iglesias, 1989; Zamankhan et al., 2006; Zhang et al., 2008) they have used, non-carcinogenic and carcinogenic risks estimated in their study were much higher than those in this study. For the industrial site estimated HQs for Cr, Mn, Ni, Pb, and Se were found to be 0.14, 0.98, 0.68, 0.026, and 0.000024, respectively. Estimated carcinogenic risks for Cr, Ni, and Pb were 9.8×10^{-4}, 1.1×10^{-6}, and 4.5×10^{-8}, approximately 134, 10, and 1.30 times the total carcinogenic risks estimated for Çerkezköy with the ICRP model.

3.4. Cumulative risks

Cumulative carcinogenic and non-carcinogenic risks of PTEs estimated with HRA with ICRP modeling and MPPD modeling in comparison with regular HRA (without considering deposition fractions) are presented in Figs. 5 and 6, respectively. Cumulative carcinogenic risks for fine and coarse PM were estimated as 1.1×10^{-5} and 6.6×10^{-6} in Çorlu and 9.4×10^{-6} and 8.7×10^{-6} in Çerkezköy, respectively, with regular HRA, and were found to be well above the acceptable risk level. However, considering deposition fractions, ICRP model showed that cumulative risks in the alveolar region were 91% lower for fine PM and 94% lower for coarse PM. The MPPD model, on the other hand, results in 93% lower cumulative risks for fine PM and 87% for coarse PM in the alveolar region. On average, regular HRA was found to cause an overestimation of health risks by 92% and 90% for fine and coarse PM, respectively. Cumulative carcinogenic risks for AR remained below the acceptable risk level, except for cumulative carcinogenic risk of coarse PM in Çerkezköy estimated with MPPD model which was found to be 1.2×10^{-6}. In addition, cumulative carcinogenic risk of fine PM in Çorlu estimated with ICRP model for AR stayed at the acceptable risk limit with 1.0×10^{-6}.

Cumulative non-carcinogenic risks with regular HRA were found in the range of 0.136–0.357, and were below the threshold. On the other hand, when deposition fractions in alveolar region are considered, this range narrowed to 0.008–0.033 for HRA with ICRP model and 0.018–0.025 for HRA with MPPD model. No major differences were observed between Çerkezköy and Çorlu in terms of cumulative non-carcinogenic risks for AR.

Table 4
Average chronic daily PTE intakes of participants in fine and coarse PM, mg/kg-day.

		HA[a]				TB[b]				AR[c]			
		Fine		Coarse		Fine		Coarse		Fine		Coarse	
		ICRP	MPPD	ICRP	MPPD	ICRP	MPPD	ICRP	MPPD	ICRP	MPPD	ICRP	MPPD
Al	Çorlu	2.1×10^{-5}	1.0×10^{-5}	5.9×10^{-5}	5.1×10^{-5}	1.6×10^{-6}	7.1×10^{-6}	3.1×10^{-6}	6.2×10^{-6}	1.5×10^{-5}	1.1×10^{-5}	3.9×10^{-6}	9.1×10^{-6}
	Çerkezköy	1.4×10^{-5}	7.0×10^{-6}	5.7×10^{-5}	4.9×10^{-5}	1.1×10^{-6}	4.8×10^{-6}	3.0×10^{-6}	5.9×10^{-6}	1.0×10^{-5}	7.5×10^{-6}	3.8×10^{-6}	8.7×10^{-6}
As	Çorlu	1.1×10^{-8}	5.2×10^{-9}	5.5×10^{-8}	4.8×10^{-8}	8.1×10^{-10}	3.6×10^{-9}	2.9×10^{-9}	5.8×10^{-9}	7.4×10^{-9}	5.6×10^{-9}	3.7×10^{-9}	8.5×10^{-9}
	Çerkezköy	1.1×10^{-8}	5.3×10^{-9}	5.0×10^{-8}	4.3×10^{-8}	8.2×10^{-10}	3.6×10^{-9}	2.6×10^{-9}	5.2×10^{-9}	7.5×10^{-9}	5.6×10^{-9}	3.3×10^{-9}	7.7×10^{-9}
Ba	Çorlu	3.7×10^{-7}	1.8×10^{-7}	1.8×10^{-6}	1.5×10^{-6}	2.8×10^{-8}	1.2×10^{-7}	9.3×10^{-8}	1.9×10^{-7}	2.6×10^{-7}	1.9×10^{-7}	1.2×10^{-7}	2.7×10^{-7}
	Çerkezköy	4.1×10^{-7}	2.0×10^{-7}	1.9×10^{-6}	1.7×10^{-6}	3.2×10^{-8}	1.4×10^{-7}	1.0×10^{-7}	2.0×10^{-7}	2.9×10^{-7}	2.2×10^{-7}	1.3×10^{-7}	3.0×10^{-7}
Cd	Çorlu	1.7×10^{-8}	8.3×10^{-9}	6.6×10^{-8}	5.7×10^{-8}	1.3×10^{-9}	5.7×10^{-9}	3.4×10^{-9}	6.9×10^{-9}	1.2×10^{-8}	8.8×10^{-9}	4.4×10^{-9}	1.0×10^{-8}
	Çerkezköy	2.1×10^{-8}	1.0×10^{-8}	9.7×10^{-8}	8.4×10^{-8}	1.6×10^{-9}	7.2×10^{-9}	5.0×10^{-9}	1.0×10^{-8}	1.5×10^{-8}	1.1×10^{-8}	6.4×10^{-9}	1.5×10^{-8}
Cr	Çorlu	7.3×10^{-8}	3.6×10^{-8}	2.9×10^{-7}	2.6×10^{-7}	5.6×10^{-9}	2.5×10^{-8}	1.5×10^{-8}	3.1×10^{-8}	5.1×10^{-8}	3.9×10^{-8}	2.0×10^{-8}	4.6×10^{-8}
	Çerkezköy	6.7×10^{-8}	3.3×10^{-8}	4.1×10^{-7}	3.5×10^{-7}	5.2×10^{-9}	2.3×10^{-8}	2.1×10^{-8}	4.3×10^{-8}	4.7×10^{-8}	3.5×10^{-8}	2.7×10^{-8}	6.3×10^{-8}
Co	Çorlu	1.0×10^{-8}	4.9×10^{-9}	2.2×10^{-8}	1.9×10^{-8}	7.7×10^{-10}	3.4×10^{-9}	1.2×10^{-9}	2.3×10^{-9}	7.1×10^{-9}	5.3×10^{-9}	1.5×10^{-9}	3.4×10^{-9}
	Çerkezköy	3.5×10^{-9}	1.7×10^{-9}	2.6×10^{-8}	2.3×10^{-8}	2.7×10^{-10}	1.2×10^{-9}	1.4×10^{-9}	2.7×10^{-9}	2.4×10^{-9}	1.8×10^{-9}	1.8×10^{-9}	4.0×10^{-9}
Pb	Çorlu	3.4×10^{-7}	1.7×10^{-7}	1.6×10^{-6}	1.4×10^{-6}	2.6×10^{-8}	1.1×10^{-7}	8.4×10^{-8}	1.7×10^{-7}	2.4×10^{-7}	1.8×10^{-7}	1.1×10^{-7}	2.5×10^{-7}
	Çerkezköy	2.7×10^{-7}	1.3×10^{-7}	1.9×10^{-6}	1.7×10^{-6}	2.1×10^{-8}	9.3×10^{-8}	1.0×10^{-7}	2.0×10^{-7}	1.9×10^{-7}	1.4×10^{-7}	1.3×10^{-7}	3.0×10^{-7}
Mn	Çorlu	2.8×10^{-7}	1.4×10^{-7}	3.8×10^{-7}	3.3×10^{-7}	2.1×10^{-8}	9.4×10^{-8}	2.0×10^{-8}	4.0×10^{-8}	2.0×10^{-7}	1.5×10^{-7}	2.5×10^{-8}	5.9×10^{-8}
	Çerkezköy	3.1×10^{-7}	1.5×10^{-7}	9.4×10^{-7}	8.1×10^{-7}	2.4×10^{-8}	1.1×10^{-7}	4.9×10^{-8}	9.8×10^{-8}	2.2×10^{-7}	1.6×10^{-7}	6.2×10^{-8}	1.4×10^{-7}
Ni	Çorlu	4.8×10^{-8}	2.4×10^{-8}	2.6×10^{-7}	2.3×10^{-7}	3.7×10^{-9}	1.6×10^{-8}	1.4×10^{-8}	2.8×10^{-8}	3.4×10^{-8}	2.5×10^{-8}	1.8×10^{-8}	4.1×10^{-8}
	Çerkezköy	4.7×10^{-8}	2.3×10^{-8}	3.1×10^{-7}	2.7×10^{-7}	3.6×10^{-9}	1.6×10^{-8}	1.6×10^{-8}	3.3×10^{-8}	3.3×10^{-8}	2.5×10^{-8}	2.1×10^{-8}	4.8×10^{-8}
Se	Çorlu	8.0×10^{-8}	4.0×10^{-8}	3.4×10^{-7}	2.9×10^{-7}	6.2×10^{-9}	2.7×10^{-8}	1.8×10^{-8}	3.5×10^{-8}	5.6×10^{-8}	4.2×10^{-8}	2.3×10^{-8}	5.2×10^{-8}
	Çerkezköy	8.8×10^{-8}	4.3×10^{-8}	4.3×10^{-7}	3.7×10^{-7}	6.7×10^{-9}	3.0×10^{-8}	2.2×10^{-8}	4.5×10^{-8}	6.2×10^{-8}	4.6×10^{-8}	2.9×10^{-8}	6.6×10^{-8}

[a] HA: head airways.
[b] TB: tracheobronchial region.
[c] AR: alveolar region.

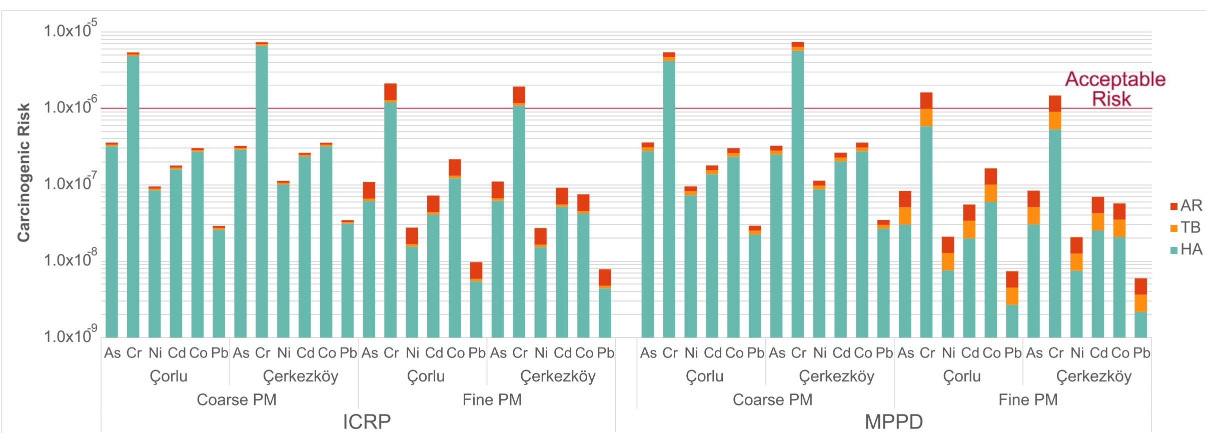

Fig. 1. Estimated carcinogenic risks for HA, TB, and AR.

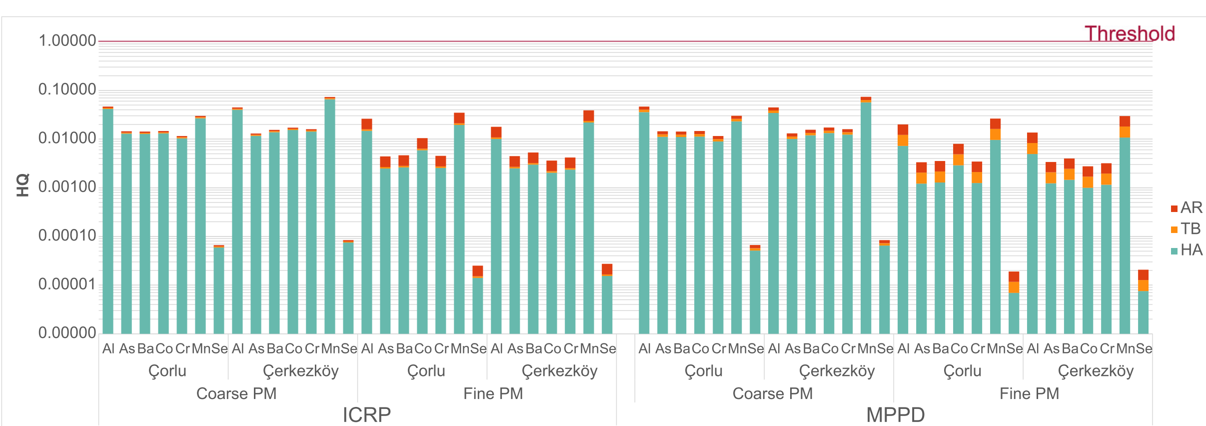

Fig. 2. Estimated non-carcinogenic risks for HA, TB, and AR.

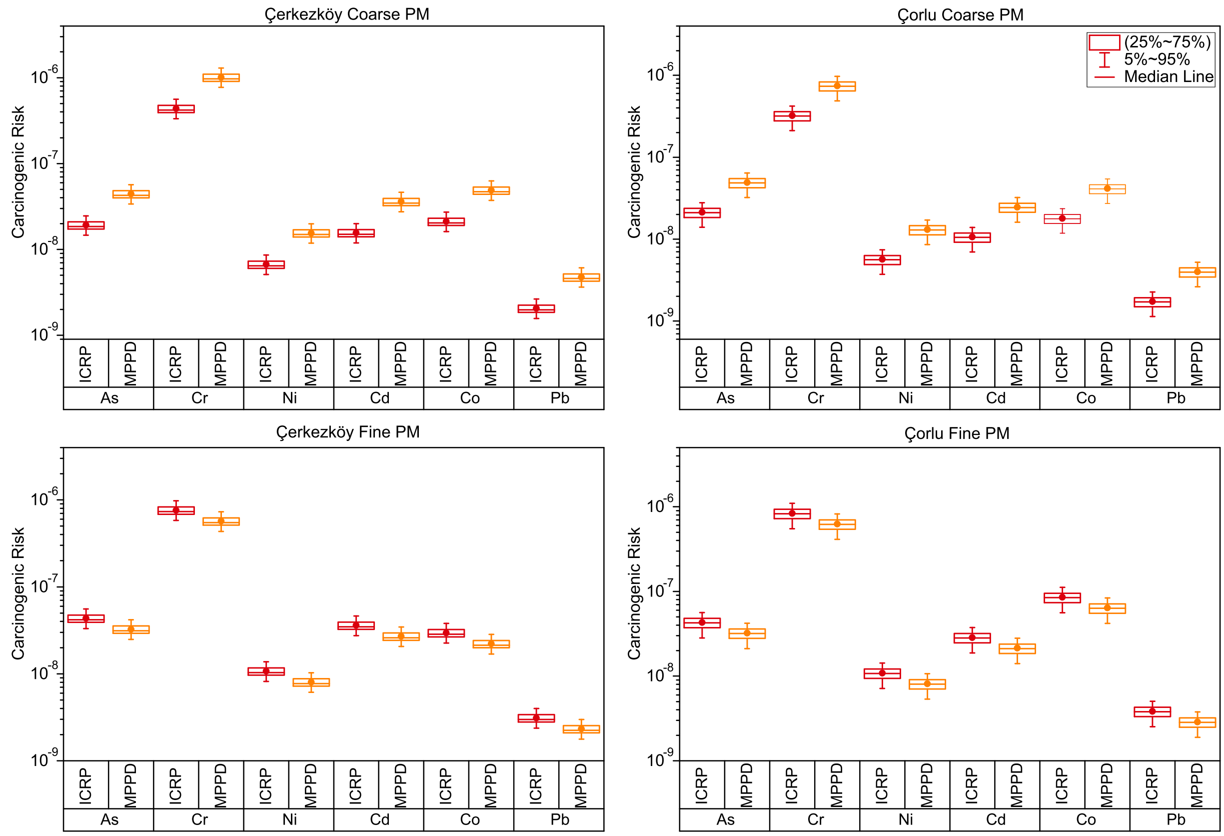

Fig. 3. Distributions of carcinogenic risks in Çorlu and Çerkezköy participants for AR.

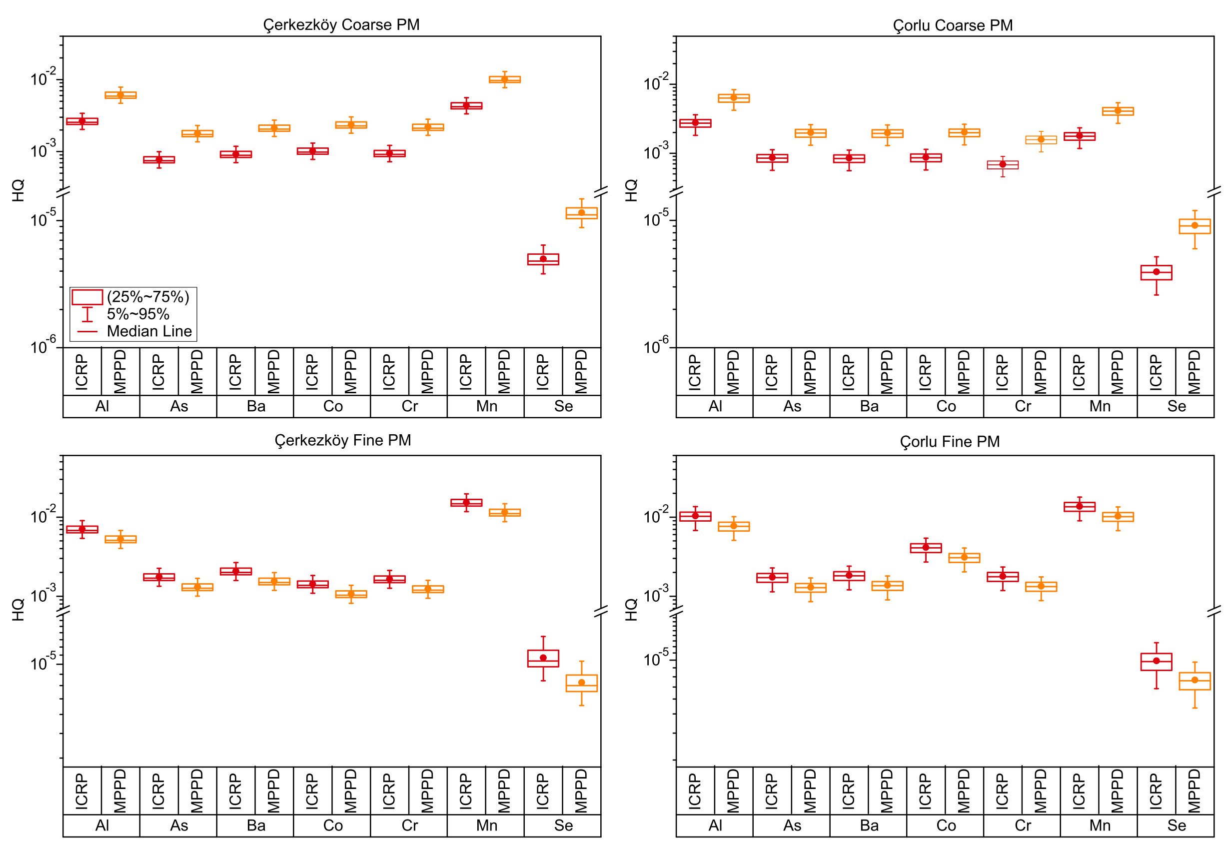

Fig. 4. Distributions of non-carcinogenic risks in Çorlu and Çerkezköy participants for AR.

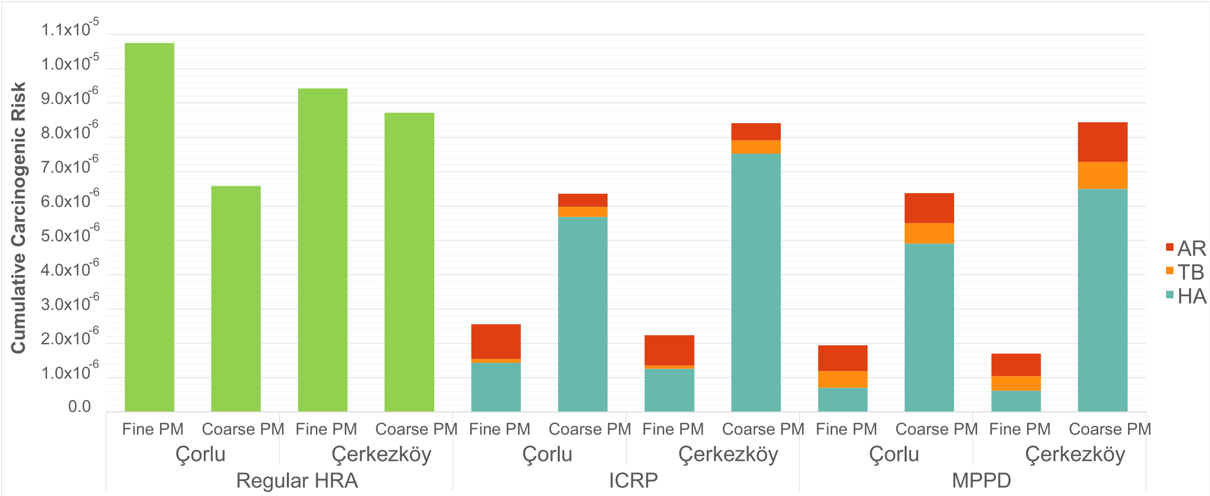

Fig. 5. Cumulative carcinogenic risks of PTEs estimated with regular HRA, HRA with ICRP modeling, and HRA with MPPD modeling.

3.5. Limitations of the study

Questionnaire survey participants were requested to provide time-activity information for seven days. The averages were considered to be sufficient for employment in chronic exposure-risk estimations (Kavcar et al., 2006; Gungormus et al., 2014). However, a week may be considered to be short for this purpose due to the seasonality of human activities as people tend to spend more time outdoors in summer or spring time with higher breathing rates. Therefore, the time spent indoors and sleep/nap or sedentary activities may be above the annual average because the survey was conducted in winter, while the time spent outdoors and consequently moderate and high intensity activities may be below an annual average. Another limitation is the small and gender-disproportionate size of the questionnaire survey at one of the sampling locations (Çerkezköy). Although equal number of volunteers (100) were randomly recruited, the response rate was low in Çerkezköy (22%, 7 females and 15 males) while it was 72% in Çorlu with a more realistic female-male distribution (43–57%). These issues in Çerkezköy impacted the population representativeness of the participant sample, and comparability of the risk estimations at the two locations, whereas this was not an issue for the collected PM samples.

We used ambient air PTE concentrations measured at two stations for health risk estimations of all participants, assuming 100% penetration of PM into the buildings. There are many studies showing that indoor and outdoor PM concentrations are correlated. Braniš et al. (2005) have found 95% and 92% correlation between outdoor and indoor $PM_{2.5}$ levels in winter and summer, respectively. They stated that effect of human activities were stronger on indoor PM concentrations in winter compared to summer but only when the outdoor PM levels were low. Zwoździak et al. (2014) found that the indoor PM_1 concentrations in a school were mostly due to infiltration of ambient air. Huang et al. (2015) investigated indoor-outdoor relationship of fine PM for urban residences in Beijing, and concluded that ambient-origin $PM_{2.5}$ dominantly contributed to residential indoor $PM_{2.5}$ exposure.

There was also a lack of measured PM mass size distribution for the study area. Our assumption of mass median diameters of fine and coarse PM has a direct effect on the PM deposition efficiencies in the lungs. Although we provide literature support to the assumed mass median diameters of 0.6 and 5 μm, respectively for fine and coarse fractions, varying these levels would result in variation in the estimated risks due to changing lung deposition efficiencies. Therefore, measurement of a further segregated particle size distribution would be useful to reduce the associated uncertainties.

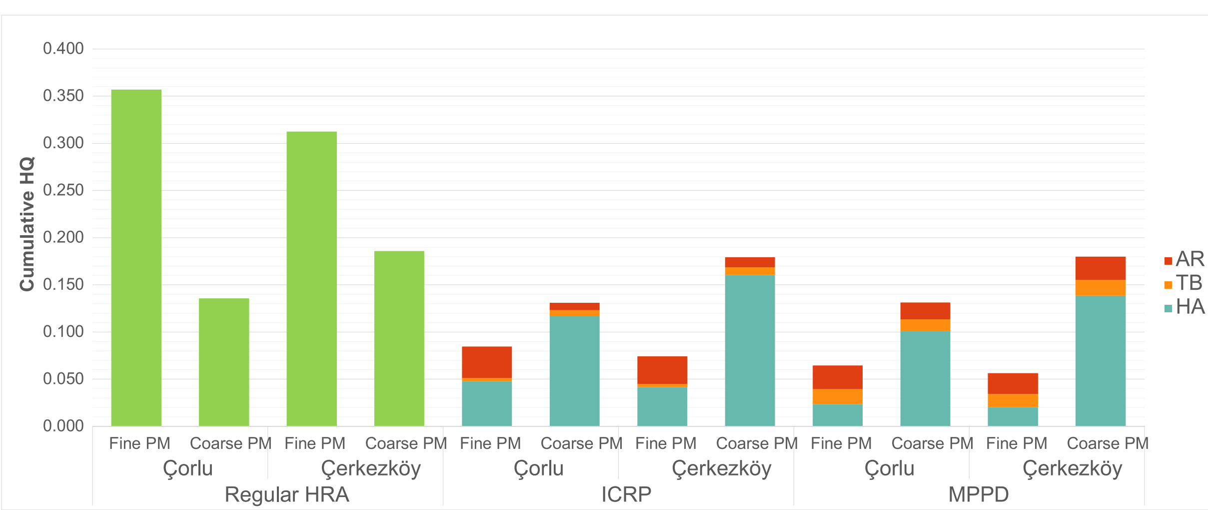

Fig. 6. Cumulative non-carcinogenic risks of PTEs estimated with regular HRA, HRA with ICRP modeling, and HRA with MPPD modeling.

4. Conclusions

Exposure of people living in two popular cities (Çorlu and Çerkezköy) of Ergene Basin to PM and its PTE content were investigated in this study. Compared to other provinces of Turkey, fine-PM concentrations were similar but coarse-PM concentrations were lower. Al, Cr, and Se were the PTEs at relatively higher concentrations among the other provinces of Turkey. Cr was the only PTE that exceeded acceptable risk in terms of total carcinogenic risk of three deposition regions (HA, TB, and AR). However, carcinogenic risk of Cr for AR was below the acceptable risk limit. Non-carcinogenic risks of all the PTEs including Cr were below the threshold value. The results suggest that Cr exposure through anthropogenic PM may be important for locations with dense industrial activity, for which further research is necessary to determine its actual speciation and control its main sources. Although Çerkezköy represents the industrial setting, therefore, higher health risks may be expected, only minor differences have been observed with Çorlu in terms of health risks, which suggest that urban pollution could be at least as impacting human health as industrial sources. Taking respiratory deposition into account, even with dichotomous particle-size data, results in avoiding overestimation of health risks, yet, size segregation at a higher resolution would be more definitive, along with inclusion of semi-volatile organic compounds that would broaden the picture for health risks associated with PM content.

CRediT authorship contribution statement

Begum Can-Terzi: Writing - original draft, Formal analysis, Visualization. **Merve Ficici:** Investigation, Formal analysis. **Lokman Hakan Tecer:** Conceptualization, Funding acquisition, Project administration, Resources, Methodology. **Sait C. Sofuoglu:** Funding acquisition, Methodology, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This research was supported by the Scientific and Technological Research Council of Turkey (TUBITAK) Grant#114Y576.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.142026.

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