J Radioanal Nucl Chem (2016) 309:309–315
DOI 10.1007/s10967-015-4673-5

Exposure and inhaled dose of susceptible population to chemical elements in atmospheric particles

Susana Marta Almeida[1] · **Carla A. Ramos**[1] · **Marina Almeida-Silva**[1]

Received: 20 August 2015 / Published online: 31 December 2015

Abstract The objective of this study was to assess the exposure and inhaled dose of susceptible populations to chemical elements in atmospheric particles. Particles were sampled in schools, fitness centers and elderly care centers and k_0-INAA was used to determine their chemical composition. Results show that besides the similar element concentrations measured in the outdoor levels of all microenvironments, the inhaled dose experienced by each one of the vulnerable population groups was significantly different, indicating that measurements of outdoor air concentrations do not provide an accurate estimation of the population exposure to particles and specific elements and of the respective dose.

Keywords k_0-INAA · Schools · Elderly care centers · Fitness centers · Particles · Elements

Introduction

The health effects of air pollution have been subject of intense study in recent years. Exposure to pollutants such as airborne particulate matter (PM) has been associated with increases in mortality and hospital admissions due to respiratory and cardiovascular diseases [1]. It is estimated that approximately 3 % of cardiopulmonary and 5 % of lung cancer deaths are attributable to PM globally [2]. According to the World Health Organization (WHO) urban air pollution, especially PM, causes significant health problems throughout Europe, reducing the life expectancy of residents of more polluted areas by over 1 year [2]. The WHO's specialized cancer agency, the International Agency for Research on Cancer (IARC), has recently classified outdoor air pollution and one of its major components, PM, as carcinogenic to human beings [3]. PM effects have been seen at very low levels of exposure and there is no evidence of a safe level of exposure or a threshold below which no adverse health effects occur. This is due to the fact that PM is a complex mixture of microscopic particles enriched with different chemicals derived from both anthropogenic and natural sources [4]. From a mechanistic perspective, it is highly plausible that the chemical composition of PM would better predict health effects than other characteristics such as PM mass or size.

The potential of harmful health effects of air pollution has been estimated by comparing outdoor levels with air quality guidelines and with health outcomes. However, this logic has been changed by a number of recent developments in both air pollution and scientific knowledge. Since people spend 90–95 % of their time indoors, individual's exposure to PM is dominated by indoor air pollution, which is partly outdoor air pollution that has penetrated indoors and partly pollution from indoor sources. However, data available for risk assessment of indoor air pollution are scarce and often insufficient. Information is available for the indoor air concentrations of some well-known pollutants, but is lacking for others which effects are unclear such as the chemical components of indoor PM that are currently poorly characterized.

The objective of this work was to assess the indoor air concentrations of PM chemical elements in schools, elderly care centers and fitness centers in order to estimate the

---

✉ Susana Marta Almeida
smarta@ctn.ist.utl.pt

[1] Centro de Ciências e Tecnologias Nucleares (C2TN), Instituto Superior Técnico, Universidade de Lisboa, EN 10, ao km 139.7, 2695-066 Bobadela LRS, Portugal

exposure of susceptible groups and the respective inhaled dose. Children, elders and sport practitioners are at great risk. Children breathe higher volumes of air relative to their body weight and their tissue and organs are growing [5]. Elders susceptibility originates from natural and pathological ageing and related processes. Ageing has been associated with a decline in immune defences and respiratory function, and predisposition to respiratory infections [6]. Sport practitioners can be at risk when they are practicing exercise in polluted environments due the fact that the increase in the minute ventilation increases proportionally the quantity of inhaled pollutants, most of the air is inhaled through the mouth, bypassing the normal nasal mechanisms for filtration of large particles and the increased airflow velocity carries pollutants deeper into the respiratory tract [7].

Experimental

Particulate matter was sampled in 3 schools, 3 fitness centers and 4 elderly care centers located in Lisbon, the capital city of Portugal where the traffic is the main source of atmospheric particles [8, 9]. Due to the geographic position of Lisbon and to the dominant western wind regime the expected high levels of pollutants should be uncommon. The transport of maritime air mass is usually associated with cleaner air masses from the Atlantic Ocean and with better dispersion conditions of pollutants coming from the industrial areas [10]. Nevertheless, under adverse meteorological conditions, low dispersion conditions and thermic inversions, high concentrations of air pollutants are registered. Moreover, in Lisbon, natural PM sources cause a number of PM exceedances due to natural mineral particulate sources such as high-dust Saharan air mass intrusions [11].

Three types of samplers were used to collect particles with a diameter lower than 10 µm (PM10): (1) Gent samplers, operating at 1.0 $m^3 h^{-1}$ and collecting particles in polycarbonate filters; (2) a Partisol Plus 2025 Sequential Ambient Particulate Sampler, operating at a flow rate of 1.0 $m^3 h^{-1}$ and collecting particles on Teflon filters and (3) a Leckel Medium Volume Sampler 6, operating at a flow rate of 3.5 $m^3 h^{-1}$ and collecting particles onto quartz filters. Details on sampling campaigns performed in schools, elderly care centers and fitness centers can be obtained in works developed by Almeida et al. [12], Almeida-Silva et al. [13] and Ramos et al. [14], respectively.

The filter loads were measured by gravimetry in a controlled clean room (class 10,000) at 20 °C and relative humidity of 50 % according to EN 12341: 1998 [15]. Filters were weighted on a semi-micro-balance and filter weight before and after sampling was obtained as the average of three measurements, when observed variations were less than 5 %.

Element analysis was carried out by k_0-Instrumental Neutron Activation Analysis (k_0-INAA) at C^2TN/IST [16]. The ability of k_0-INAA to analyse solid phase samples for many elements, without the need for sample dissolution or digestion and with a high degree of sensitivity and selectivity made it particularly suitable for the elemental analysis of PM filters. Filters were rolled up and put into a clean thin foil of aluminium and irradiated for 5 h at a thermal neutron flux in the Portuguese Research Reactor [17]. After irradiation, the sample was removed from the aluminium foil and transferred to a polyethylene container. For each irradiated sample, two gamma spectra were measured with a hyperpure germanium detector: one spectrum 2–3 days after the irradiation and the other one after 4 weeks.

Blank filters were treated the same way as regular samples. All measured species were very homogeneously distributed; therefore concentrations were corrected by subtracting the filter blank contents.

Previous to the sampling campaign, tests of reproducibility within the filters and between filters were taken, using parallel sampling with two similar sampling units and measuring the particle species by k_0-INAA. Results were reproducible to within 5–15 %, providing strong support for the validity of the analytical techniques. The details of the analytical control tests are given in the work developed by Almeida et al. [18].

The quality control of the techniques has been developed with the intention of using the methods to meet the demands of trace element analysis for aerosol filters. The accuracy of analytical methods was evaluated with NIST filter standards, revealing results with an agreement of 10 %. More details can be found in works dedicated to this subject [19, 20].

Results

Concentration and composition of particles

The average indoor and outdoor PM10 and element concentrations measured in schools, elderly care centers and fitness centers are presented in Table 1. Results show that the highest PM10 concentrations were measured in schools (85 ± 32 µg m^{-3}) exceeding the limit value of 50 µg m^{-3} established by the Portuguese legislation for indoor air, Portaria no. 353-A/2013 [21]. In elderly care centers PM10 average concentration was 10.9 ± 4.2 µg m^{-3} in bedrooms and 18.5 ± 6.0 µg m^{-3} in living-rooms. In fitness centers, an average concentration of 15 ± 15 µg m^{-3} was

Table 1 Inhalation rates ($V\dot{E}$), Body Weight (BW) and concentrations (average ± standard deviation) of PM10 and elements measured in schools, elderly care centers and fitness centers

		$V\dot{E}$ (Lm^{-1})	BW (kg)	Micro-environment	PM10 (μg m^{-3})	As (ng m^{-3})	Ca (ng m^{-3})	Ce (ng m^{-3})	Co (ng m^{-3})	Cr (ng m^{-3})	Fe (ng m^{-3})
Schools	Classroom	22	32	Indoor	85 ± 32	9.9 ± 12.5	15300 ± 7200		0.71 ± 0.43	10.8 ± 1.9	750 ± 570
				Outdoor	33.0 ± 7.1	0.42 ± 0.23	3800 ± 2800		0.39 ± 0.28	6.0 ± 3.3	350 ± 120
Elderly care centers	Bedroom	5.2	80	Indoor	10.9 ± 4.2	0.28 ± 0.34		0.47 ± 0.34	0.063 ± 0.047	9.2 ± 4.2	48 ± 28
				Outdoor	23.1 ± 7.3	0.64 ± 0.93		0.35	0.17 ± 0.14	9.2 ± 5.6	230 ± 160
	Living-room	4.9	80	Indoor	18.5 ± 6.0	0.33 ± 0.21		0.37 ± 0.13	0.107 ± 0.061	12.0 ± 2.2	220 ± 180
				Outdoor	19.4 ± 4.5	0.97 ± 0.11		0.42	0.200 ± 0.030	15.8 ± 5.3	380 ± 260
Fitness centers	Classroom	Holistic classes - 26	80	Indoor	15 ± 15	0.10 ± 0.09	600 ± 630		0.076 ± 0.037	1.6 ± 1.0	210 ± 180
		Aerobic classes - 47		Outdoor	32 ± 14	0.48 ± 0.42	2700 ± 2900		0.21 ± 0.16	4.5 ± 2.8	850 ± 950

		$V\dot{E}$ (Lm^{-1})	BW (kg)	Micro-environment	K (ng m^{-3})	La (ng m^{-3})	Na (ng m^{-3})	Sb (ng m^{-3})	Sc (ng m^{-3})	Sm (ng m^{-3})	Zn (ng m^{3})
Schools	Classroom	22	32	Indoor	1000 ± 580	0.70 ± 0.33	1800 ± 660	4.7 ± 3.3	0.18 ± 0.13	0.156 ± 0.082	94 ± 35
				Outdoor	263 ± 99	0.21 ± 0.13	2400 ± 820	2.27 ± 0.96	0.098 ± 0.111	0.053 ± 0.061	22.3 ± 6.8
Elderly care centers	Bedroom	5.2	80	Indoor	100+50		200 ± 160	0.50 ± 0.33	0.0040 ± 0.0029	0.0030 ± 0.0008	26 ± 14
				Outdoor	830 ± 1340		930 ± 280	1.8 ± 1.0	0.023 ± 0.016	0.019 ± 0.008	32 ± 26
	Living-room	4.9	80	Indoor	160 ± 74		440 ± 120	1.4 ± 1.2	0.018 ± 0.016	0.013 ± 0.0092	39 ± 15
				Outdoor	820 ± 800		1900 ± 1400	1.9 ± 1.4	0.030 ± 0.020	0.040 ± 0.030	30.8 ± 9.5
Fitness centers	Classroom	Holistic classes - 26	80	Indoor	100 ± 70	0.064 ± 0.050	540 ± 770	0.46 ± 0.36	0.010 ± 0.008	0.01	12 ± 9.0
		Aerobic classes - 47		Outdoor	290 ± 250	0.150 ± 0.094	1020 ± 680	2.5 ± 1.8	0.015 ± 0.012	0.018 ± 0.00044	46 ± 42

measured. This difference between micro-environments can be explained by the high intensity of the pupils' physical activity that led to re-suspension of particles and largely contribute to increased PM10 concentrations in classrooms [22]. In fitness centers, there is also a high occupancy and physical activity but people are obliged to change their shoes before entering in the classrooms that reduce the contamination of the indoor spaces by dust transported from outdoor. Moreover, as opposed to the schools that have natural ventilation, all fitness centers have mechanic ventilation and filtration of air before its supply in the building, which reduces the outdoor particles infiltration. Consequently, PM10 concentrations were significantly higher in the indoor of schools than in the outdoor, whereas in fitness centers PM10 concentration were lower in the indoors (Fig. 1).

Sodium was the only element that presented indoor/outdoor ratios lower than 1 for all the buildings typologies, indicating the non-indoor origin of this element, which is typically associated with the sea emissions. In schools it was observed the highest concentration of Fe, La, Sc and Sm, which have a soil origin, and the highest indoor/outdoor ratio for these elements. This fact confirms that the most probable cause of increased classroom PM10 concentrations is the penetration of mineral dust through the windows and the re-suspension of settled dust or suspension of soil material brought in by the children's shoes. In schools and elderly care centers higher concentrations of Zn were also measured indoors. This fact has already been observed in other studies [23] and indicates that, besides the association of this element to traffic emissions and industrial activities, there is an indoor Zn source probably associated with several products that can be applied indoors to protect steel, walls, wood surfaces, doors and windows. Ca was also detected at very high concentrations inside schools' classrooms (15 000 ± 7 200 ng m^{-3}) due to the chalk (mainly $CaSO_4$) used in the blackboards and/or the gypsum walls and plasters used as building material [24]. The elements As and Sb presented higher concentrations inside the classrooms than outdoor. These elements, typically present in outdoor of urban environments, probably penetrated by infiltration to the indoor of the buildings where the dilution conditions are reduced. The element concentrations in elderly care centers and fitness centers were lower than in schools and presented lower values indoors than outdoors. These facts indicate the importance of the filtration of particles, in air treatment units, that avoid the contamination of the indoor spaces by

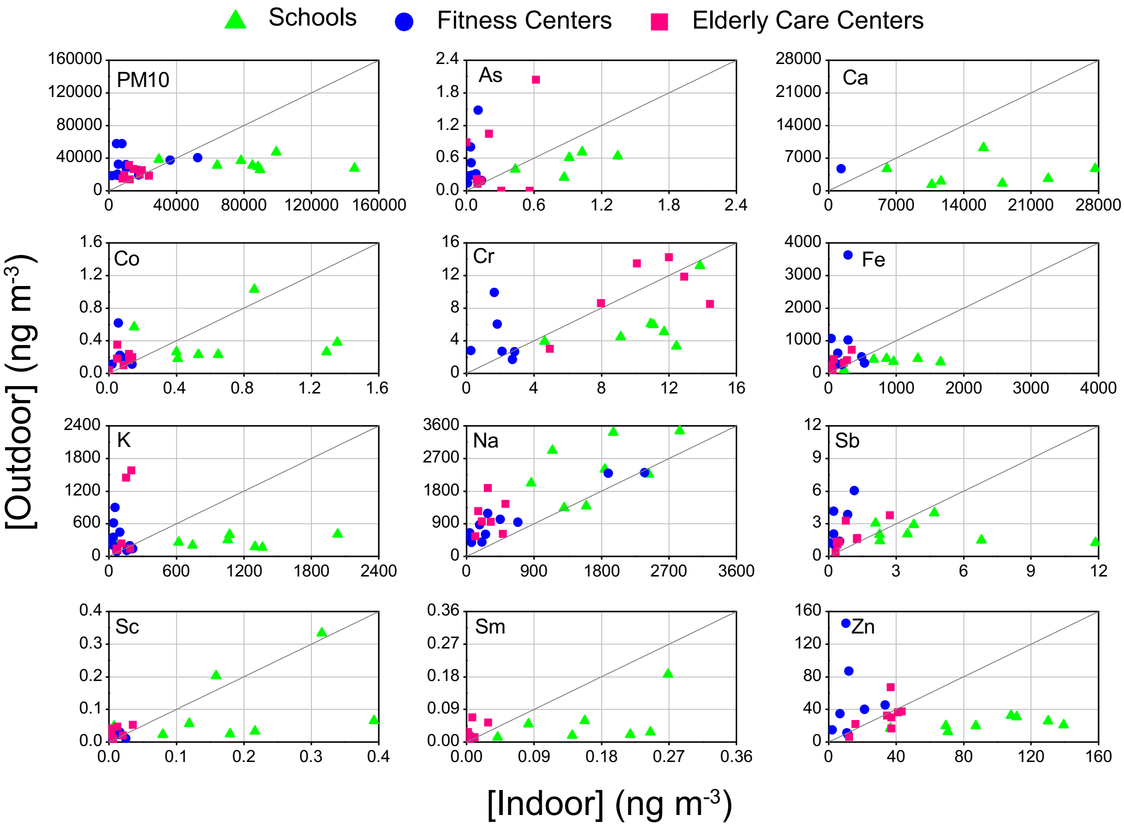

Fig. 1 Indoor and outdoor concentrations measured in Schools, Elderly Care Centers and Fitness Centers (values in ng m^{-3})

outdoor pollutants, and the highest ventilation of spaces, preconized by mechanic ventilation, to dilute the pollutants generated indoors [25].

Inhaled dose

The average inhaled dose (D) in schools, fitness centers and elderly care centers was assessed by integrating the concentration of the pollutant in each micro-environment (C), the inhalation rate ($V\dot{E}$) and the body weight (BW) according to Eq. 1.

$$D = \left(C \times V\dot{E}\right)/BW \tag{1}$$

The $V\dot{E}$ used in schools and elderly care centers were recommended by EPA [26] for different ages and distinct activities—sleep, sedentary and light intensity. Since $V\dot{E}$ has never been measured before for fitness classes, $V\dot{E}$ was estimated using heart rate (HR) measurements and a pre-established individual relation between HR and $V\dot{E}$ according the procedure described by Ramos et al. [27]. Table 1 presents the $V\dot{E}$ used in the calculation of the inhaled dose. The BW used in this study was 32 and 80 kg for children and adults, respectively, also based on US-EPA [26].

Figure 2 presents the average inhaled dose in schools' classrooms, elderly care centers and fitness centers. Results show that the highest inhaled doses were experienced by children in schools (21 ng min^{-1} kg^{-1}) and the lowest were associated with elders in the bedroom of elderly care centers (0.72 ng min^{-1} kg^{-1}). The same trend was observed for the inhaled dose of the particles chemical elements. This fact can be explained by the highest concentrations measured in the schools, the lowest BW in children and the lowest $V\dot{E}$ in elders. In elderly care centers the inhaled dose of the elements (except for Ce) was higher in living-rooms than in bedrooms. Living-rooms have more occupancy and indoor concentrations of dust particles are strongly influenced by activities and movement of occupants, which may allow the re-suspension of particles. Higher PM10 inhalation doses were determined for the aerobic classes (9.0 ng min^{-1} kg^{-1}) comparing with the holistic classes (4.9 ng min^{-1} kg^{-1}) and the same behavior was observed for the inhaled dose of chemical elements. Consequently, assessing the inhaled dose of particles' components in fitness centers is the key determinant of the impacts that these pollutants can have on the health of sport practitioners. Differences between $V\dot{E}$ and pollutant concentrations between different classes influence the inhaled dose of air pollution and therefore an accurate assessment of these two parameters is fundamental.

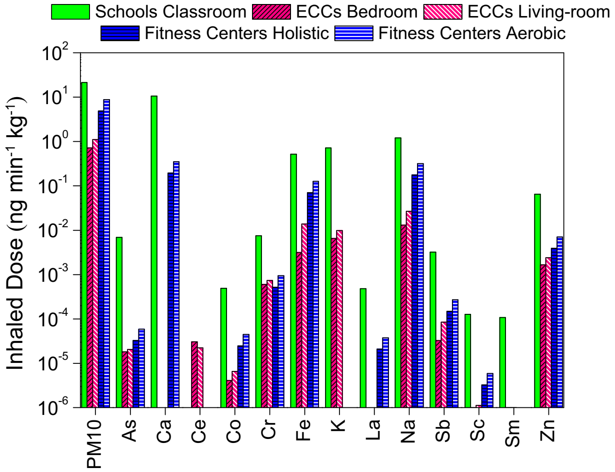

Fig. 2 Inhaled dose in Schools, Elderly Care Centers (ECCs) and Fitness Centers (values in ng min^{-1} kg^{-1})

Discussion

Results show that besides the similar element concentrations measured in the outdoor levels of all micro-environments, the inhaled dose experienced by each one of the vulnerable population groups is significantly different. Therefore, the local measurements of outdoor air concentrations do not provide an accurate estimation of the population exposure to particles and specific elements and of the respective dose. People spend the majority of their time indoors where the concentrations of pollutants depend a lot on indoor sources and type and maintenance of ventilation systems.

Moreover, $V\dot{E}$ depends on the type of activity and highly influences the inhaled dose of pollutants. In this study, the average inhaled dose per minute in each micro-environment was calculated. However, the duration of exposure in each micro-environment is another factor that should be considered for the calculation of the daily exposure and daily inhaled dose. For instance institutionalized elders spend most of their time inside elderly care centers which means that the inhaled dose measured in this study can be a good predictor of the daily inhaled dose. However, population that practice sport in fitness centers spend between 1 and 2 h per day in fitness centers and therefore the daily inhaled dose will be highly affected by other micro-environments such as home, work and transports. Children spend on average 8 h per day in school and therefore the inhaled dose in schools highly affects the daily inhaled dose.

The more intense and the longer the exposures, the greater the risk may be. From the pathophysiological point of view, being exposed for 1 h every day to a pollutant for 20 days is different from being exposed for 20 h to the same pollutant for only 1 day [28]. As a result the evaluation of the integrated exposure to air pollutants and associated dose needs to be conducted taking into account daily activity patterns in different environments in which individuals spend their time and, within each environment, the $\dot{V}E$, the length and the intensity of exposure. The assessment of the daily dose to air pollutants is a critical component of epidemiological studies associating air pollution and adverse health effects.

Conclusions

Results showed that concentrations of particles were higher in schools. The element composition of the classrooms' particles suggested that earth crustal materials, detritions of the building materials and chalk have an important role in the particles levels and characteristics. Assuming that crustal materials and combustion-related particles vary in toxicity, our findings on concentrations of particle components support the hypothesis that indoor-generated PM in schools may be less toxic compared to PM in ambient air. In fitness centers the inhaled dose of pollutants depended not only on the fitness center but also in the type of activity that occur inside the classrooms. In aerobics classes the minute ventilation increases and, consequently, inhaled dose of chemical elements was higher during this activity then in holistic classes. In elderly care centers lower inhaled doses were estimated. However, besides being more vulnerable, elders spend the majority of their time inside these centers and therefore air quality must be kept with a good quality once health effects of particles occur even at relatively low concentrations.

Acknowledgments The authors gratefully acknowledge Fundação para a Ciência e Tecnologia (FCT) for funding M. Almeida-Silva PhD grant (SFRH/BD/69700/2010), C.A. Ramos PhD grant (SFRH/BD/79277/2011) and S.M. Almeida contract (IF/01078/2013). C^2TN/IST authors gratefully acknowledge the FCT support through the UID/Multi/04349/2013 project.

References

1. Almeida SM, Silva AV, Sarmento S (2014) Effects of exposure to particles and ozone on hospital admissions for cardiorespiratory diseases in Setúbal, Portugal. J Toxicol Environ Heal A 77:837–848
2. WHO (2013) Health effects of particulate matter: policy implications for countries in Eastern Europe, Caucasus and central Asia, Copenhagen, WHO Regional office for Europe
3. IARC (2013) Press release no. 221. Outdoor air pollution a leading environmental cause of cancer deaths. IARC-WHO, 17 Oct 2013
4. Calvo AI, Alves C, Castro A, Pont V, Vicente AM, Fraile R (2013) Research on aerosol sources and chemical composition: past, current and emerging issues. Atmos Res 120–121:1–28
5. Canha N, Almeida M, Freitas MC, Almeida SM (2011) Seasonal variation of total particulate matter and children respiratory diseases at Lisbon basic schools using passive methods. Proced Environ Sci 4:170–183
6. Almeida-Silva M, Wolterbeek HTh, Almeida SM (2014) Elderly exposure to indoor air pollutants. Atmos Environ 85:54–63
7. Ramos C, Viegas C, Cabo-Verde S, Wolterbeek HT, Almeida SM (2015) Characterizing the fungal and bacterial microflora and concentrations in fitness centres. Indoor Built Environ. doi:10.1177/1420326X15587954
8. Almeida SM, Freitas MC, Repolho C, Dionísio I, Dung HM, Caseiro A, Alves C, Pio CA, Pacheco AMG (2009) Characterizing air particulate matter composition and sources in Lisbon, Portugal. J Radioanal Nucl Chem 281:215–218
9. Almeida SM, Freitas MC, Repolho C, Dionísio I, Dung HM, Pio CA, Alves C, Caseiro A, Pacheco AMG (2009) Evaluating Children exposure to air pollutants for an epidemiological study. J Radioanal Nucl Chem 280:405–409
10. Almeida SM, Silva AI, Freitas MC, Dzung HM, Caseiro A, Pio CA (2013) Impact of maritime air mass trajectories on the western european coast urban aerosol. J Toxicol Environ Heal A 76:252–262
11. Almeida SM, Freitas MC, Pio CA (2008) Neutron activation analysis for identification of African mineral dust transport. J Radioanal Nucl Chem 276:161–165
12. Almeida SM, Canha N, Silva A, Freitas MC, Pegas P, Alves C, Evtyugina MG, Pio CA (2011) Children exposure to air particulate matter in indoor of Lisbon primary schools. Atmos Environ 45(40):7594–7599
13. Almeida-Silva M, Almeida SM, Pegas PN, Nunes T, Alves CA, Wolterbeek HT (2015) Exposure and dose assessment to particle components among an elderly population. Atmos Environ 102:156–166
14. Ramos C, Wolterbeek HTH, Almeida SM (2014) Exposure to indoor air pollutants during physical activity in fitness centers. Build Environ 82:349–360
15. EN 12341: 1998 (1998) Air quality—determination of the PM10 fraction of suspended particulate matter—reference method and field test procedure to demonstrate reference equivalence of measurement methods. European Committee for Standardization (CEN), Brussels
16. Almeida SM, Freitas MC, Reis M, Pinheiro T, Felix PM, Pio CA (2013) Fifteen years of nuclear techniques application to suspended particulate matter studies. J Radioanal Nucl Chem 297:347–356
17. Fernandes AC, Santos JP, Marques JG, Kling A, Ramos AR, Barradas NP (2010) Validation of the Monte Carlo model supporting core conversion of the Portuguese Research Reactor (RPI) for neutron fluence rate determinations. Ann Nucl Energy 37:1139–1145
18. Almeida SM, Reis MA, Freitas MC, Pio CA (2003) Quality assurance in elemental analysis of airborne particles. Nucl Instrum Methods Phys Res B 207:434–446
19. Dung HM, Freitas MC, Blaauw M, Almeida SM, Dionisio I, Canha NH (2010) Quality control and performance evaluation of k0-based neutron activation analysis and the Portuguese Research Reactor. Nucl Instrum Methods Phys Res A 622:392–398
20. Almeida SM, Almeida-Silva M, Galinha C, Ramos CA, Lage J, Canha N, Silva AV, Bode P (2014) Assessment of the Portuguese k0-INAA laboratory performance by evaluating internal quality control data. J Radioanal Nucl Chem 300:581–587
21. Portaria 353-A/2013 (2013) Ministério do ambiente, ordenamento do território e energia, da saúde e da solidariedade, emprego e segurança social

22. Canha N, Almeida SM, Freitas MC, Wolterbeek HTh, Cardoso J, Pio C, Caseiro A (2014) Impact of wood burning on indoor PM2.5 in a primary school in rural Portugal. Atmos Environ 94:663–670
23. Canha N, Almeida SM, Freitas MC, Trancoso M, Sousa A, Mouro F, Wolterbeek HTh (2014) Particulate matter analysis in indoor environments of urban and rural primary schools using passive sampling methodology. Atmos Environ 83:21–34
24. Canha N, Freitas MC, Almeida SM, Almeida M, Ribeiro M, Galinha C, Wolterbeek HTH (2010) Indoor school environment: easy and low cost to assess inorganic pollutants. J Radioanal Nucl Chem 286:495–500
25. Canha N, Almeida SM, Freitas MC, Täubel M, Hänninen O (2013) Winter ventilation rates at primary schools: comparison between Portugal and Finland. J Toxicol Environ Heal A 76:400–408
26. United States Environmental Protection Agency (2011) Exposure factors handbook, 2011th edn. Office of Research and Development, Washington, DC **20460**
27. Ramos C, Reis JF, Almeida T, Alves F, Wolterbeek HT, Almeida SM (2015) Estimating the inhaled dose of pollutants during indoor physical activity. Sci Total Environ 527-528C:111–118
28. Bentayeb M, Simoni M, Baiz N, Norback D, Baldacci S, Maio S, Viegi G, Annesi-Maesano I (2012) Adverse respiratory effects of outdoor air Pollution in the elderly. Int J Tuberc Lung Dis 16(9):1149–1161