Applied Radiation and Isotopes 69 (2011) 1586–1591

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

Air pollution at an urban traffic tunnel in Lisbon, Portugal—an INAA study

M. Almeida-Silva, N. Canha, M.C. Freitas *, H.M Dung, I. Dionísio

Instituto Tecnológico e Nuclear, URSN, E.N. 10, 2686-953 Sacavém, Portugal

ARTICLE INFO

Available online 15 January 2011

Keywords:
Air pollution
Urban
Traffic Tunnel
Chemical elements
INAA
Enrichment factors

ABSTRACT

In this study, the results of chemical concentrations inside and outside of a Lisbon (Portugal) traffic tunnel were compared, during one week. They were obtained by Instrumental Neutron Activation Analysis (INAA). The tunnel values largely exceed the Air Ambient legislated values and the Pearson Correlations Coefficients point out to soil re-suspension/dispersed road dust (As, Ce, Eu, Hf, Fe, Mo, Sc, Zn), traffic-markers (Ba, Cr), tire wear (Cr, Zn), break wear (Fe, Zn, Ba, Cu, Sb), exhaust and motor oil (Zn) and sea-spray (Br, Na). On all days these elements inside the tunnel were more enriched than outside; significant statistical differences were found for Co ($p=0.005$), Br ($p=0.008$), Zn ($p=0.01$) and Sb ($p=0.005$), while enrichment factors of As and Sc are statistically identical. The highest values were found for As, Br, Zn and Sb, for both inside and outside the tunnel.

1. Introduction

In the past 30 years, outdoor levels of some pollutants are decreasing in many cities of Europe and US, due to emission controls on vehicles, heating, power generation and industry. However in developing countries the outdoor air pollution is worsening due to the increasing number of vehicles and industries. The major outdoor air pollutants are particulates, O_3, CO, SO_x, H_2S, acid gases (HF, HCl), NO_x, Pb and other metals, volatile organics, solvents, pesticides, methane, bioaerosols and radionuclides (Freitas, 2009; Freitas et al., 2009). Tunnels have been built, in developing cities, to facilitate mobility of people within the cities. An efficient tunnel construction has the potential to reduce traffic congestion. On the other hand, since ventilation outlets collect vehicle exhausts and release it all in one or two locations, road tunnels in urban areas give rise to problems including localized air pollution (Ma et al., 2004). So, understanding emissions from traffic includes identification of the sources, which is also crucial for designing control measures (Sternbeck et al., 2002).

In this study, the results of chemical concentrations inside and outside of a Lisbon (Portugal) traffic tunnel were compared. They were determined by Instrumental Neutron Activation Analysis (INAA) (Freitas et al., 2008; Dung et al., 2008) and their sources were assessed.

2. Experimental

2.1. Sampling site and equipment

From 9 to 21 October 2008, at rush traffic hours and weekend, different fractions of air particulate matter were collected inside a traffic tunnel in Lisbon, using quartz filters in different selectors. Particles were collected within the periods 08:00 and 10:00 and 17:00 and 19:00 from Monday to Saturday except on Tuesday, which was within 08:45–09:45 and 17:45 and 18:45. The collected fractions were $PM_{10-2.5}$, $PM_{2.5-1}$, $PM_{1-0.5}$ and $PM_{0.5}$, which correspond, respectively, to particles with aerodynamic diameter between 2.5 and 10 μm, 1 and 2.5 μm, 0.5 and 1 μm and below 0.5 μm. Daily collection time varied between 0.46 and 3.95 h (it depended on the size collected fraction, less for the coarser particles, more for the finer ones) and the collected air volumes varied between 31.19 and 267.81 m^3. Low and high volume air samplers were used.

2.2. Sample analysis

The collected filters and blank filters, turned available by Aveiro University, Portugal, were irradiated at the Portuguese Research Reactor (RPI-ITN; nominal power: 1 MW) for analyses by k_0-INAA (De Corte, 1987). Irradiation time was 5 h at a neutron flux density of $8\times10^{12}\,cm^{-2}\,s^{-1}$, at an irradiation position with 103.4 ± 1.3 thermal-to-epithermal neutron flux ratio and an $1/E^{1+\alpha}$ epithermal neutron flux distribution shape factor of -0.0351 ± 0.0004. Each sample was rolled up and put into an aluminum foil for irradiation, removed from the foil after irradiation and put in a polyethylene container for measurement. Samples were measured 7 h after 3–5 days and after 3–4 weeks, with an ORTEC® automatic sample

* Corresponding author. Tel.: +351 219946130; fax: +351 219941039.
E-mail address: cfreitas@itn.pt (M.C. Freitas).

0969-8043/$ - see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2011.01.014

changer, equipped with an ORTEC® coaxial hyperpure germanium detector (1.90 keV FWHM—full width at half maximum at 1.33 MeV of ^{60}Co; 30% relative efficiency). One Al-0.1% Au alloy disk with a thickness of 125 μm and a diameter of 0.5 cm was used as comparator for the k_0-methodology, being irradiated with the samples. A total of 4 replicates were analyzed for each type of blank filter.

Quality control was asserted by analyzing NIST-SRM® 2783 "Air Particulate on Filter Media" ($PM_{2.5}$ on a polycarbonate filter membrane) following the same procedure as for the samples. NIST-SRM® 1633a "Coal Fly Ash" was also irradiated together with the samples, weighing 20–30 mg each replicate in a total of 9 replicates, with measuring times of 60 and 150 min after 3–5 days and after 3–4 weeks, respectively.

One Partisol Plus Sequential Air Sampler was placed at the center of Lisbon (38°44′ N–9°8′ W), from 1st January to 31st December 2007 (Almeida et al., 2009; Edwards et al., 2009; Freitas et al., 2009), aiming to collect $PM_{2.5}$. The sampler used 47 mm diameter Teflon® filters, which collected particles for periods of 24 h at 16.7 l/min; the mass of particles was gravimetrically determined in a controlled clean room (class 10000). The balance had a sensitivity of 0.1 μg. The filters were analyzed by k_0-INAA. Procedures of sample preparation, irradiation, decay and measurement of loaded and blank filters, were similar to the ones described above for the quartz filters. Quality control was asserted as described above.

2.3. Statistics

Enrichment factor analysis provides a preliminary picture of whether the indoor particulate matters come from the crust or from other natural or anthropogenic activities (Chao and Wong, 2002). Enrichment factors (EF) values were calculated relative to Fe using Mason soil data (Mason and Moore, 1982). The EFs were calculated as:

$$EF_X = (X/\mathrm{Ref})_{\mathrm{aerosol}}/(X/\mathrm{Ref})_{\mathrm{crust}}$$

where EF_X is the enrichment factor of species X and Ref is the reference element.

Source categories for aerosols constituents were identified by means of Pearson Correlation Coefficients using STATISTICA software. All statistics in this work are considered at 95% confidence level.

3. Results and discussion

Fig. 1 shows the ratios between the results obtained in this work for NIST 1633a coal fly ash and the certified values of the latter. The agreement is considered good taking into account that the uncertainties are at 67% confidence level.

Table 1 shows the mass concentrations obtained in the sampling periods, in the tunnel and in the environment. The results obtained in the environment by Partisol did not exceed the $PM_{2.5}$ limit value of 25 μg/m^3 defined in European Union (EU, 2007). However, the results showed that the recommendations of the World Health Organization (WHO, 2010) and the United States Environmental Protection Agency (USEPA, 2010) were both exceeded with levels of 10 and 15 μg/m^3, respectively. This evidence could be a concern for human health.

Table 2 shows the Pearson Correlation Coefficients obtained for the chemical elements obtained by INAA in the tunnel filter samples. The coefficients with statistical significance at 95% confidence level are shown in bold. The correlations point out to similar studies (Ayrault et al., 2010; Zechmeister et al., 2006), that Sb was the most highly enriched element in filters collected in Paris (France) and Vienna (Austria), respectively. Sb and Zn were the most strongly enriched elements in a study in Gothenburg, Sweden (Sternbeck et al., 2002), and both were pointed out

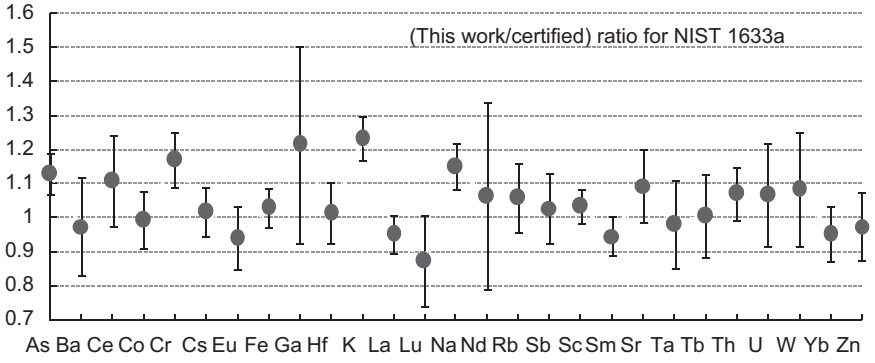

Fig. 1. Ratio between the results obtained in this work for NIST 1633a coal fly ash and the certified values of the latter. Uncertainty is at 67% confidence level.

Table 1
Mass concentrations collected in the filters. Morning and afternoon belong to the tunnel, Partisol to Lisbon center.

Mass concentration (μg m^{-3})	Fractions	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
Morning	$PM_{10-2.5}$	91.0	226	160	142	128	51.7
	$PM_{2.5-1}$	92.3	157	121	104	99.7	51.8
	$PM_{1-0.5}$	71.6	296	94.8	73.8	78.6	32.2
	$PM_{0.5}$	286	558	316	326	306	115
Afternoon	$PM_{10-2.5}$	147	279	139	114	132	69.9
	$PM_{2.5-1}$	71.7	247	104	59.7	81.2	51.0
	$PM_{1-0.5}$	55.4	206	82.6	33.5	77.4	40.0
	$PM_{0.5}$	430	608	446	414	343	214
Partisol	$PM_{2.5}$	21.0	20.7	21.1	20.5	21.2	21.1