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Outdoor and indoor particle characterization from a large and uncontrolled combustion of a tire landfill

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HIGHLIGHTS

- BC and particle number were used as tracers of the tire fire emission plume.
- Infiltration processes have been documented at a school located 700 m downwind the fire.
- Deposited inhalable dust measured indoor and outdoor at a near school was enriched in metals and PAHs associated to tire combustion.

GRAPHICAL ABSTRACT



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ABSTRACT

A large and uncontrolled fire of a tire landfill started in Seseña (Toledo, Spain) on May 13, 2016. An experimental deployment was immediately launched in the area for measuring regulated and non-standard air quality parameters to assess the potential impact of the plume at local and regional levels. Outdoor and indoor measurements of different parameters were carried out at a near school, approximately 700 m downwind the burning tires. Real time measurements of ambient black carbon (BC) and total number particle concentrations were identified as good tracers of the smoke plume. Simultaneous peaks allowed us to characterize situations of the plume impact on the site. Outdoor total particle number concentrations reached in these occasions 3.8×10^5 particles cm^{-3} (on a 10 min resolution) whereas the indoor concentration was one order of magnitude lower. BC mass concentrations in ambient air were in the range of 2 to 7 μg m^{-3}, whereas concentrations < 2 μg m^{-3} were measured indoor. Indoor and outdoor deposited inhalable dust was sampled and chemically characterized. Both indoor and outdoor dust was enriched in tire components (Zn, sulfate) and PAHs associated to the tire combustion process. Infiltration processes have been documented for BC and particle number concentrations causing increases in indoor concentrations.

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1. Introduction

Around 5 million tons of tires per year are produced in Europe, representing 20% of the world tire production (ETRMA, 2015). Due to

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expected EU car production for the next decades it seems that tire market will keep being a healthy business for main manufacturers in this sector. On the other side of the coin there is a tire output, estimated as 1.5 billion units on a global basis (ETRMA, 2015), which subsequently mostly will fall into the category of end-of-life tires. This fact represents a serious challenge for manufacturers, users and environmental authorities that will have to deal with this problem within an incipient regulatory framework. Tire recovery and recycling have become in the last decade a valuable resource with growing potential. Although the tire recycling business is complex, storage and reuse of tires can generate many benefits linked to a wide diversity of economy sectors, from agriculture, civil engineering or sport, in a developing market continuously searching for new applications.

Spain is currently one of the EU countries with the highest recovery rates, >90% (ETRMA, 2011). But, although 200,000 tons are recycled every year, many others are lost along the way by the negligence of users and, above all, for breaching of regulations. In the last decade, many companies engaged in buying and selling used tires, bloomed in this apparently easily profit market. In 2006, the new Spanish regulations made the situation non-viable for many of them, unable to fulfil the legal requirements, giving rise to a number of uncontrolled and abandoned tire landfills. These landfills are subject to potential fires, posing a serious threat for the environment and human health.

On May 13, 2016 a very large and uncontrolled fire was started in one of these abandoned landfills in Spain. The tire landfill was located in Seseña, a small town (21,500 inhabitants) 40 km South of Madrid, the Spanish capital. According to the available data, the landfill had over 100,000 tons of tires deposited on its surface occupying a total area of 117,000 m^2. National authorities estimated that the fire, which was officially declared extinct on June 7, 2016, affected more than ¾ of the landfill area.

During the first stages of the fire, a dense column of black smoke, visible from a distance of almost 100 km, ascended above ground and was transported several hundred kilometers from the site without nearly apparent impacts on ground surface. From the very early stages, once the fire started, an emergency plan was activated with participation of a number of public bodies and authorities that included Civil Protection, Public Health, Fire Service, and the Emergency Call Center who, in a coordinated manner, managed the situation and decided the measures to be taken. These were firstly to combat and extinguish the fire, and at the same time, assessing on how to reduce the impact of emissions on population exposure. First actions were devoted to limit the fire propagation, as other measures addressed to extinguish it, such as capping with sand or other materials, were discarded by the Fire Service experts. The flaming phase of the fire was active for several days and, as the fuel was progressively consumed the combustion became more inefficient and the plume became grey and brown during the smoldering phase of the fire. The composition of the smoke plume probably also varied from the initial to the final phase. Besides, mechanical works devoted to improve the combustion efficiency and to accelerate the fire extinction, and the tire piling prior to their removal, were carried out during this second phase of the fire.

Although this type of event is not very frequent, similar experiences have been documented in other countries (Demarini et al., 1994; Wang et al., 2007; Shakya et al., 2008; Downard et al., 2015) most of them affecting scrap tires. The tire's chemical composition typically includes natural and synthetic rubber polymers, oil fillers, sulfur and sulfur compounds, phenolic resin, clay minerals, aromatic, naphthenic and paraffinic oils, fabric, petroleum waxes, pigments such as zinc oxide and titanium dioxide, black carbon, fatty acids, inert materials and fiber made from steel, nylon, polyester or rayon (Reisman, 1997; Thorpe and Harrison, 2008). The smoke plume from open burning of rubber tires is enriched in gaseous components (CO, CO_2, SO_2, VOC) and particulate matter (Lemieux et al., 2004), the latter containing a number of elements and metals associated to tire manufacturing (i.e. Zn, Pb, Fe) and other to the combustion process, like elemental carbon (EC) and organic carbon (OC) (Reisman, 1997). Within the organic fraction a special attention should be paid to some toxic, mutagenic and carcinogenic compounds, such as polycyclic aromatic hydrocarbons (PAHs) and other products of incomplete combustion (Wang et al., 2007; Lemieux et al., 2004) which can pose a serious threat to health for the affected population.

Approximately 300 m away from the Seseña tire landfill, the first line of buildings belonging to the El Quiñón neighborhood is located. With roughly 7000 inhabitants this was the most threatened community due to its proximity to the fire and prevailing winds. Some measurements of non-regulated parameters like particle number (PN) concentration and particulate black carbon (BC), as tracers of the smoke plume, were used to assess the situation and support the decision making. These measurements included additional samplings of deposited inhalable dust indoor and outdoor performed in the El Quiñón school, located roughly 700 m away from the tire landfill.

Although a number of studies have documented indoor and outdoor ambient particle matter concentrations (Alves et al., 2014; Krugly et al., 2014; Rivas et al., 2015; Amato et al., 2014) or deposited dust (Boudehane et al., 2016) at schools, none of them has dealt with tire fire emissions. The only work published up to now related to the Seseña event (Nadal et al., 2016) provides a preliminary human health risk assessment based on the first results on chemical characterization of ambient air and soil samples around Seseña. Nevertheless no indoor/outdoor analysis was performed nor suspended inhalable dust was considered in that study.

This paper presents the results of the measurements carried out at the El Quiñón (EQ) public school. Real time measurements of total PN concentrations and BC, and sampling of deposited inhalable (diameter < 10 μm) dust were carried out indoor/outdoor for several days during the fire. The deposited dust samples were analyzed in the laboratory to determine their main composition: inorganic species, metals and PAHs.

The main aim of this paper is to present the results of this experimental deployment to contribute to assess the impact of the uncontrolled fire and its consequences on health of the exposed population. It is also aimed at providing tools and methodology to characterize this impact and, ultimately, to contribute to support the decision making by responsible authorities in case of similar events.

2. Methodology

2.1. Sampling site

Measurements were performed at the EQ school, located 700 m east of the tire landfill (Fig. 1). Indoor and outdoor measurements were performed on May 22 and June 03, 2016. Instruments for air monitoring and dust sampling were placed in a classroom approximately 3-meter high above ground corresponding to a first floor. There were no scholar activities and no heating systems were operating during the period of measurements.

Ambient concentrations of PM_{10} and $PM_{2.5}$ (particulate matter with aerodynamic diameter < 10 and <2.5 μm, respectively), recorded by a mobile laboratory, markedly increased coinciding with the impact of the plume in El Quiñón. These peaks, reaching maximum hourly concentrations of 390 μg m^{-3} and 141 μg m^{-3}, respectively during the period May 16 to June 07, 2016, were occasional and never lasted more than three hours (JCCM, 2016).

2.2. Total particle number concentrations

Sub-micron particles (diameter < 1 μm), with a greater penetration capacity in the respiratory system inversely proportional to its size, were considered to be representative of the most dangerous components of the fire plume in terms of human health. To quantify these, total particle number (PN) concentrations were measured by two



Fig. 1. Google-maps of the studied area before (top) and after (bottom) the tire fire.

Condensation Particle Counters (CPCs), which were used simultaneously outdoor and indoor at the EQ school. Indoor concentrations were measured by a TSI CPC3772 having a particle size detection limit of 10 nm and with a preimpactor in the inlet that had a particle cutoff size of 740 nm for the flow rate used (1 L min^{-1}). This CPC is able to measure PN concentrations below 10^4 particles cm^{-3}. The instrument was located less than 1 m from the window. Outdoor PN concentrations were measured by a TSI CPC3775 instrument with a detection limit of 4 nm that used another preimpactor in the sampling line with a cutoff size of 1.3 μm for 0.3 L min^{-1}. The upper concentration limit for this CPC is 10^7 particles cm^{-3}. The CPC 3372 and 3375 worked with a temporal resolution of five and 1 s respectively. The outdoor air-inlet was situated at the front of the school building, avoiding the shielding by nearby buildings. The two CPCs used in this study were in the ±10% band of the reference value used in the REDMAAS inter-comparison exercises carried out in 2015 (Gómez-Moreno et al., 2015).

These two instruments were measuring simultaneously from May 26, 2016 at 2:00 pm (local time (LT)) to May 27, 2016 at 6:00 am, which allowed recording the period when, according to the meteorological conditions, the fire plume could impact the measurement site.

2.3. Ambient black carbon mass concentrations

Black carbon can be directly associated to combustion processes emissions and, as suggested by some studies (Janssen et al., 2012), seems to be a better indicator of the harmfulness of combustion particles than PM concentration. Although authors are aware that the term "equivalent black carbon" (eBC) can be more appropriate, and has been suggested (GAW/WMO, 2011) for use instead of BC for measurements derived from optical methods, for practical purposes BC will be used throughout this work.

In this experimental study, BC concentrations were simultaneously obtained, under an outdoor/indoor measurement scheme, by two aethalometers. These instruments measure the rate of change in absorption of the transmitted light at a specific wavelength due to the continuous collection of aerosol deposit on a filter.

Ambient measurements were obtained with a multi-wavelength aethalometer (Magee Sci. mod. AE33, Aerosol d.o.o) (λ = 370, 470, 520, 590, 660, 880, and 950 nm) with a cut-off size of 10 μm and a flow rate of 5 L min^{-1}. Data were recorded automatically every minute from May 17 to June 17, 2016.

Indoor measurements were obtained on May 24, 2016 with a microaethalometer (microAeth® model AE51, Magee Scientific) at λ = 880 nm and a cut-off size of 2.5 μm. The instrument was located less than 1 m from the window. The flow rate was 0.1 L min^{-1} and the sampling time 300 s. For this experiment two configurations were used in two different time periods: during the first 20 min (5:30–5:50 pm LT) measurements were taken with the classroom windows closed and during the following 10 min (5:50–6:00 pm LT) they were open.

Simultaneous measurements of 10-min average BC concentrations obtained by the two instruments, microaethalometer and aethalometer,

showed a very good correlation ($r^2 = 0.99$) evidencing that the BC content in PM_{10} can be mainly found in the $PM_{2.5}$ fraction.

2.4. Deposited inhalable dust

The field measurement deployment at EQ school included the measurement of the deposited thoracic fraction (<10 μm in aerodynamic diameter) of dust considered relevant for evaluating both the outdoor/indoor impact as well as the efficiency of some of the cleaning measures (classrooms and playground washing) carried out during the fire period on May 30, June 03, and later also on June 06, 2016. For this purpose, deposited inhalable dust was collected in the classrooms and the schoolyard ground surface, prior and after the cleaning works at the EQ School. Sampling was performed by means of a field resuspension device, commonly used for road dust collection (Amato et al., 2009, 2011) able to collect deposited particles with aerodynamic diameter below 10 μm. Particles were vacuumed for 10 min at a 28 L min^{-1} flow rate from 1 m^2 of surface and collected on a Pallflex quartz microfiber filter, previously conditioned and weighed at 20 °C and 50% RH. After sampling, filters were brought back to laboratory to be weighed under the same conditions in order to obtain dust loadings (mg m^{-2}). Once weighed, filters were destined to several analytical treatments.

As a reference site, the Miguel Delibes (MD) School, in the town of El Viso de San Juan, 22 km upwind (West) of the fire (Fig.1), was selected. Samplings were performed at the two schools on May 26 and June 03, 2016 (Table 1), 13 and 21 days respectively after the beginning of the tire fire.

One filter per site was acid digested (5 ml HF, 2.5 ml HNO_3, 2.5 ml $HClO_4$) (Querol et al., 2001) at 90 °C during 6 h, dried at 220–240 °C and recovered with 25 ml MilliQ water acidified with 1.25 ml of HNO_3, centrifuged and analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) and Mass Spectrometry (ICP-MS) for the determination of major and trace elements, respectively. Quantification limits reached 0.2 ng ml^{-1} and 0.05 μg ml^{-1} for trace and major elements, respectively, with a relative error < 10% for most elements and <20% for P, K, Na. For accuracy control, the reference material NIST, 1633b was digested and analyzed using the same protocol.

One filter per site was used for PAH analysis. The filter was extracted in closed tubes containing dichloromethane (2.5 ml) and subsequently shaken for short-time using an automatic shaker. The method of extraction and analysis was adapted from a previous method (Barrado et al., 2012; García-Alonso et al., 2015).Two re-extractions were done to ensure recovery (dichloromethane, 1 ml) and the combined extracts were slowly concentrated by a gentle stream of nitrogen (100 μL in acetonitrile). Analyses were performed by liquid chromatography equipped with fluorescence and diode array detectors. Quantification limits were found around 2.5 and 0.25 ng per injection for diode array and fluorescence detection, respectively. Among the sixteen USEPA priority compounds, only naphthalene was not considered for quantitative purposes due to its low recovery and high variability. Nevertheless, it must be pointed out that this volatile PAH is predominantly found in gas phase (Krugly et al., 2014). For establishing optimal analytical conditions and detection of selected PAHs at these minimal levels, a rigorous protocol of control for minimal contamination of vials, syringes and solvents was considered essential and applied during the whole analytical process.

Table 1
Number, date and location of deposited inhalable dust samples.

		EQ school Fire site		MD school Reference site	
		Indoor	Outdoor	Indoor	Outdoor
<10 μm	May 26, 2016	3	3	3	3
	June 03, 2016	2	3	–	–
>10 μm	May 26, 2016	2	2	2	2
	June 03, 2016	1	1	–	–

3. Results and discussion

3.1. Bulk particle number concentrations

From May 20 to June 03, 2016 the bulk PN concentration measured outdoor the EQ school reached relatively high values. During >59% of this period the concentration of particles did not fall below 1.0×10^4 cm^{-3}, with maximum hourly values of 3.2×10^5 particles cm^{-3} on May 28, 2016.

These ambient PN levels are much higher than those recorded in urban areas like Madrid (Gómez-Moreno et al., 2011), Barcelona (Reche et al., 2014) even during winter pollution episodes (Pérez et al., 2010), Milan or London (Rodríguez et al., 2007). The observed values, in the order of 10^5 particles cm^{-3}, were closer to levels registered in industrial environments (Riffault et al., 2015; Fernández-Camacho et al., 2012) ranging from 0.7 to 60×10^5 particles cm^{-3} due to the proximity to the emission sources.

The main results from the simultaneous indoor and outdoor CPCs measurements are shown in Fig. 2. Indoor PN concentrations clearly exceed the upper CPC detection limit (10^4 particles cm^{-3}) during several hours. Above this limit, the error is higher than 10% and will increase as the particle concentration increases. The measurements above this limit can give a good idea of the trend in the concentration, although the actual values would be higher than those measured. It can be observed that the indoor PN concentration is almost one order of magnitude lower than the outdoor one. PN followed a similar trend indoor and outdoor, with a temporal delay of almost 1 h for the indoor concentration, similar to that found by Diapouli et al. (2007) for a typical suburban residence in the absence of strong indoor sources. The maximum value measured outdoor was 3.8×10^5 particles cm^{-3} at 11:20 pm of May 26 whereas the maximum indoor was 3.9×10^4 particles cm^{-3} at 0:10 am of May 27, 50 min later. This means that the main source for the indoor particles was the outdoor particles and that there was a slow indoor air renewal, as can be seen from the smoother decreasing trend in the same figure. During this period there was not any passive ventilation system (i.e. windows were closed). The wind speed after 10:00 pm of May 26 was very low and can be considered as calm, so the main transport mechanism for the particles was diffusion, a phenomenon which requires time to influence indoor PN. This mechanism for air infiltration was also the one prevailing in the subsequent peaks recorded along the night. This suggests that the PN concentration indoor has a strong dependency on outdoor PN, having accordingly the same origin and patterns.

Finally, the wind direction record indicates a clear origin of the air coming directly from the west and pointing to the fire as the origin of the air mass arriving at the measuring site. Although wind was blowing from the direction of the tire fire the whole afternoon, the nocturnal peak observed in PN concentrations could be associated to the mechanical works devoted to accelerate the fire combustion carried out by the Fire Service during that night.

3.2. Black carbon measurements

Ambient BC mass concentrations at EQ during the measurement period did not exceeded 2.5 μg m^{-3}, being similar to mean values recorded in trafficked urban areas (Querol et al., 2013). These typical urban BC concentrations point to road traffic as the main source of BC in the study area, which is relatively close to a main highway (Fig. 1). Nevertheless, in specific periods, coinciding with the direct impact of the plume on the site, as suggested by wind direction records, BC concentrations experienced temporal increases in the range 4–7 μg m^{-3}. The absolute hourly maximum, 29.0 μg m^{-3}, during the measurement period was recorded on May 18, five days after the beginning of the fire.

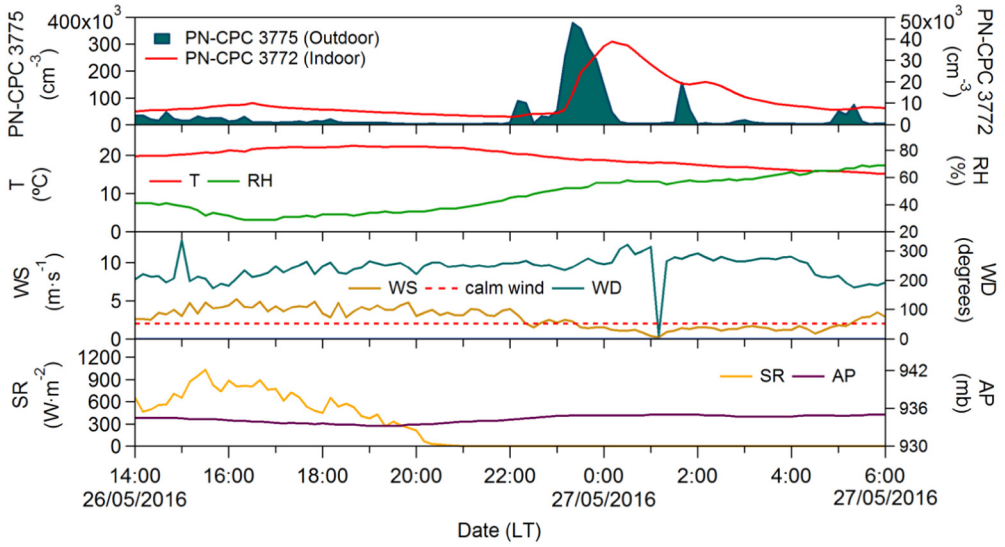


Fig. 2. Evolution of the outdoor (CPC 3775) and indoor (CPC 3772) particle number (PN) concentrations (cm^{-3}) from May 26 to May 27, 2016. The figure includes the main meteorological variables measured during the period: T = temperature, HR = relative humidity, WS = wind speed (calm WS < 2 m s^{-1}), WD = wind direction, SR = solar radiation and AP = atmospheric pressure.

Fig. 3 illustrates the continuous sampling of BC and PN during constant meteorological conditions with the wind blowing from the fire site towards the school. The similar BC and PN fluctuations in the figure are highlighted, indicating that despite constant conditions, the emission strength from the fire results in ambient air concentration variations, probably in part as a consequence of the works of extinction or acceleration of the fire combustion.

The algorithm of Sandradewi et al. (2008), updated to the results as in Zotter et al. (2016), has been used to deconvolve BC from fossil fuel (BCff) and tire fire (BCtf), in which BCtf corresponds to the organic influence originated from the tire fire at the shortest wavelengths. The relative BC concentration assigned to the tire fire origin by this model results over 80% during May 24 on the afternoon hours compared to the daily average of 30%.

Indoor BC concentrations were measured at the EQ school in two different situations (Fig. 3, colored circles). During the first period (5:30–5:50 pm LT), when the windows were closed, BC recorded an average value of 1.6 μg m^{-3} with a maximum of 1.8 μg m^{-3}. This period coincided with a situation where a previous impact of the plume on the site was evidenced, as can be seen from the BC and the PN concentrations recorded outdoor. For this period a good correlation was observed (r^2 = 0.76) between BC and PN concentrations measured outdoor. As for the particle number, the BC measurements show that even with the closed windows, particles penetrate indoor and they can exhibit even higher BC values than simultaneous outdoor measurements although with a slight delay with respect to the maximum recorded outdoor. Half an hour later, once the windows were open, an average concentration of 1.1 μg m^{-3} was recorded with a maximum value of 1.2 μg m^{-3}. This means that during this second measurement period the ambient air outdoor was cleaner, so it could be concluded that the ventilation after opening the windows had in this case a clear cleaning effect in the room.

On the other hand, as shown in Fig. 3, other maxima on ambient BC concentrations occurred late afternoon, 3 h after the previously described. These maxima occurred again with a constant and moderate wind blowing from the west, i.e. from the burning pile, producing

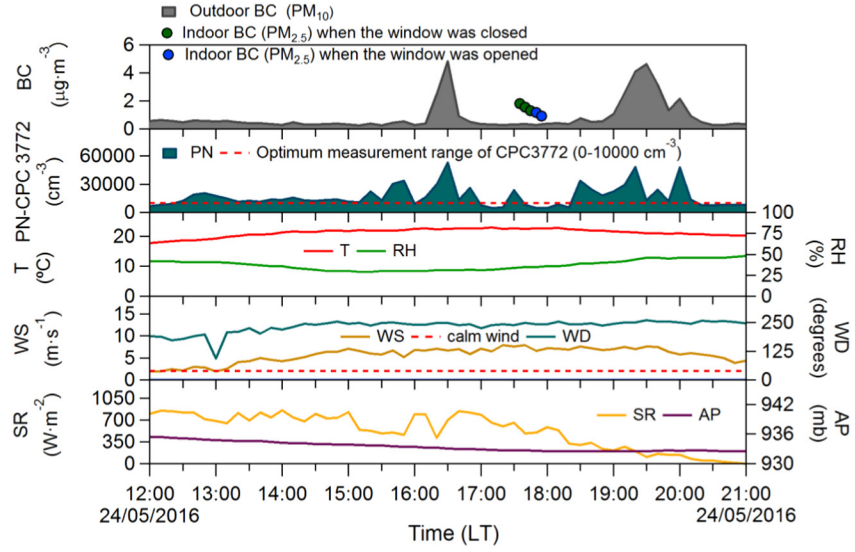


Fig. 3. Ambient air outdoor (AE33) and indoor (AE51) BC and PN (CPC 3772) concentrations on the May 24, 2016. The figure includes the main meteorological variables measured during the period: T = temperature, RH = relative humidity, WS = wind speed (calm WS < 2 m s^{-1}), WD = wind direction, SR = solar radiation and AP = atmospheric pressure.

plume impacts on the site in a discontinuous manner. This fact can be important from the point of view of estimating emission factors of the smoke plume in this type of events, as well as the exposure assessment and prevention measurements.

3.3. Deposited inhalable dust

Loadings of deposited inhalable dust varied from 4.0 to 5.9 mg m^{-2} (Table 2) at the indoor of the EQ school, both values significantly higher (>6 times) than what was found at the reference MD school (0.7 mg m^{-2}) and possibly linked to infiltration of tire fire dust. On the other hand, the outdoor loadings were high in the same range of those at the MD school (9.5–18.5 vs 13.8 mg m^{-2}), probably linked to a suburban/rural location of the MD school in an environment with high dust resuspension from local and poorly vegetated soil. Considering the EQ school only, the difference observed between May 26 and June 03, may be related to the impact of the plume after the cleaning activities on May 30 and prior to the second sampling, but also to these cleaning activities which may have dragged particles from walls to the indoor and outdoor pavements. In any case, concentrations are not overwhelming as one would expect. Inhalable deposited dust loading on roads vary within 1–20 mg m^{-2}, and can reach concentrations as high as 300 mg m^{-2} in construction areas (Amato et al., 2009, 2011).

The chemical profile of respirable (<10 μm) dust deposited outdoor shows a main mineral composition (59–61%, Table 3). Among metals, ZnO and Co show very high enrichments (ratios EQ/MD of 21:1 and 92:1, respectively) at EQ school (outdoor) when compared to MD school, reaching on the first sampling 933 and 13 μg m^{-2} and 952 and 12 μg m^{-2} on the second one, although not so high indoor (Table 3). Moreover, Zn loading increased from May 26 to June 03, probably due to the plume impact during the night of June 02 and documented by the sharp increases of SO_2 and PM_{10} levels measured by the mobile unit located near EQ (JCCM, 2016). Also, sulfate showed much higher concentration (1371 μg m^{-2}, ratio EQ/MD = 11:1) at EQ during the first sampling as well as a loading increase on the second one (2304 μg m^{-2}). Furthermore, the coarser fraction (>10 μm) shows significantly lower concentrations for these three components (Table S1), indicating a main anthropogenic (combustion) origin. Indoor, EQ/MD ratios were only 2.3, 2.0 and 4.6 respectively during the first sampling and concentrations at EQ did not vary significantly in the second one (Table 3). Zn and S have been often related to tire wear emissions (Weckwerth, 2001; Amato, 2010). Sulfur compound and oxides of Zn, Ca, Pb and Mg are added to accelerate vulcanization, yielding to concentrations of S and Zn within 1–2% of bulk tire composition (Thorpe and Harrison, 2008). The enrichment of Co is less clearly related to tire fire, given that reported concentrations of Co in tires are quite low (within 80–100 μg g^{-1}; Amato, 2010). Other elements such as Cu, Sr, Sn, Sb and Pb at the EQ were 1.3–3 times outdoor and 2–19 times indoor higher than MD school, respectively (Table 3). Copper, Sn, Sb and Pb probably originated from road traffic emissions, which are also higher at EQ school, due to the proximity of R4 y A4 highways (Fig. 1). During the second sampling, only Cu and Pb decreased significantly by 72% and 42% respectively, reaching similar concentrations to the reference MD school. For these two elements (as for As and Mo) a higher efficiency of the cleaning can be stated. Strontium instead, like CaO, is closely linked to the use of chalk in classrooms of EQ. Metals more related to mineral matter such as Ti, Rb, Zr, Sc, Ba and V were higher at the MD reference school, as the most typically mineral species (Si, Al, K and Mg, Table 3). However, after the intense cleaning activities, higher loadings were found, likely due to the

Table 2
Loadings of deposited inhalable dust in mg m^{-2}.

		EQ school Fire site		MD school Reference site	
		Indoor	Outdoor	Indoor	Outdoor
<10 μm	May 26, 2016	4.0	9.5	0.7	13.8
	June 03, 2016	5.9	18.5	–	–

Table 3
Deposited inhalable dust loadings of inorganic species (μg m^{-2}) obtained during samplings performed on May 26, 2016 and June 03, 2016 at El Quiñon (EQ) and on May 26, 2016 at Miguel Delibes (MD) sites.

μg m^{-2}	EQ school (tire fire)				MD school (reference)	
	Indoor		Outdoor		Indoor	Outdoor
	May 26	June 03	May 26	June 03	May 26	May 26
SiO_2	275	440	1207	2102	92	2501
Al_2O_3	110	176	483	841	37	1000
CaO	1375	1873	1796	3455	104	1867
Fe_2O_3	48	74	301	487	12	422
K_2O	54	82	147	211	12	342
MgO	55	88	157	283	38	423
Na_2O	35	34	59	57	10	124
P_2O_5	32	45	44	71	3	46
TiO_2	11	14	34	110	3	136
CO_3^{2-}	1080	2007	1411	3702	82	1467
Total mineral	3075	4832	5639	11,320	394	8327
% mineral	78	82	59	61	61	60
SO_4^{2-}	65	93	1371	2304	14	120
ZnO	4	3	933	952	2	44
SO_4^{2-} + ZnO	69	96	2304	3256	15	163
% SO_4^{2-} + ZnO	2	2	24	18	2	1
Li	0.06	0.1	0.31	0.51	0.01	0.39
Be	<0.01	<0.01	0.02	0.02	<0.01	0.03
Sc	<0.01	<0.01	0.03	0.03	<0.01	0.06
V	0.3	0.16	0.36	0.52	0.14	0.69
Cr	0.19	0.15	0.62	0.76	0.12	1.1
Mn	0.64	1.02	3.97	7.98	0.16	5.75
Co	0.02	0.03	12.9	12.12	0.01	0.14
Ni	0.29	0.22	0.6	0.83	0.05	0.54
Cu	1.59	0.44	5.99	6.19	0.08	1.36
Ga	0.01	0.03	0.07	0.12	<0.01	0.13
Ge	<0.01	<0.01	0.12	0.01	0.06	0.13
As	0.07	0.03	0.14	0.21	0.03	0.11
Se	<0.01	<0.01	0.01	0.01	<0.01	<0.01
Rb	0.19	0.31	0.87	1.39	0.05	2.64
Sr	1.92	2.86	8.12	24.57	0.15	4.03
Y	0.02	0.03	0.11	0.15	0.02	0.2
Zr	<0.01	0.1	0.53	0.88	0.08	1.01
Nb	0.01	0.02	0.06	0.11	<0.01	0.1
Mo	0.05	<0.01	2.18	<0.01	1.01	1.99
Cd	<0.01	<0.01	0.05	0.05	<0.01	0.02
Sn	0.05	0.05	0.35	0.34	0.02	0.2
Sb	0.01	0.01	0.35	0.39	0.01	0.12
Cs	0.01	0.01	0.05	0.08	<0.01	0.11
Ba	1.01	1.7	4.11	5.87	0.66	6.67
La	0.04	0.06	0.17	0.3	0.01	0.27
Ce	0.08	0.11	0.28	0.52	0.02	0.55
Pr	0.01	0.01	0.04	0.07	<0.01	0.06
Nd	0.03	0.04	0.42	0.56	0.01	0.25
Sm	<0.01	0.01	0.02	0.05	<0.01	0.05
Eu	<0.01	<0.01	<0.01	0.01	<0.01	0.01
Gd	0.01	0.01	0.02	0.04	<0.01	0.05
Tb	<0.01	<0.01	<0.01	0.01	<0.01	0.01
Dy	<0.01	0.01	0.02	0.03	<0.01	0.04
Ho	<0.01	<0.01	<0.01	0.01	<0.01	0.01
Er	<0.01	0.01	0.01	0.02	<0.01	0.02
Tm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Yb	<0.01	<0.01	0.01	0.02	<0.01	0.02
Lu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	<0.01	0.01	0.01	0.03	<0.01	0.03
Ta	<0.01	<0.01	<0.01	0.04	<0.01	0.02
W	0.01	0.01	0.04	0.07	0.01	0.06
Tl	<0.01	<0.01	<0.01	0.01	<0.01	<0.01
Pb	0.26	0.15	1.75	2.11	0.04	1.17
Bi	<0.01	0.01	0.01	0.02	<0.01	0.01
Th	<0.01	0.01	0.05	0.09	0.01	0.1
U	<0.01	0.01	0.03	0.04	<0.01	0.03

drag of particles from walls and/or unpaved areas and the partial removal of the sandbox at the school playground.

In our study Pb loadings were not high when compared with other sites (no literature values were found for Zn and S). Lucas et al. (2012) found an indoor deposited dust Pb mean of 8.8 μg m^{-2} in French homes, while our concentrations did not exceed 0.26 μg m^{-2}.

With respect to PAH analyses, during the first sampling PAH species directly related to the emissions from burning tires (Lemieux et al., 2004) were detected. Benzo[*g*,*h*,*i*]perylene (BghiP), chrysene (Cry), fluoranthene (Fl) and pyrene (Pyr) were measured in the highest concentrations in indoor deposited dust (Table 4). On the other hand, fluoranthene (Fl), pyrene (Pyr) and phenanthrene (Phe) were the most abundant PAHs in outdoor respirable deposited dust. The more volatile and low molecular weight PAHs, i.e. acenaphthylene (Acl), acenaphthene (Ace) and fluorene (Flu), were not detected in the samples, despite good recoveries in the applied method. Most of the PAHs detected indoor were present in low concentrations, although they were higher than those detected at the MD background reference school. Looking at absolute values, a slight increase in benz[*a*]pyrene (BaP) could be noticed in the second sampling indoor, although it cannot be considered significant as it is very close to the detection limit of the method.

A relatively recent review study on settled dust in houses in different regions of the world found median concentrations of high molecular weight PAHs (sum of four ring PAHs and larger) ranging from 0.11 μg m^{-2} in Texas to 2.9 μg m^{-2} in Azerbaijan and 162 μg m^{-2} in China (Naspinski et al., 2008). In our study we obtained a maximum PAH concentration of 0.02 μg m^{-2} for indoor and 0.13 μg m^{-2} for outdoor dust samples similar to values obtained in school sites by Boudehane et al. (2016) therefore confirming the ubiquitous nature of the PAHs observed and the low impact of the fire in this case.

Concentrations of relatively volatile compounds such as Phe and Pyr were higher than their isomers anthracene (An) and Fl, respectively, in all samples, which is usually found in urban areas that are under influence of motorized traffic (van Drooge and Grimalt, 2015). Direct collection of tire fire dust in other studies shows Phe/An and Pyr/Fl ratios which are close to unit (Wang et al., 2007; Downard et al., 2015), indicating that the observed isomeric concentrations are not exclusively related to the fire site. In fact, similar values as those found in the present study have been reported by Raffy et al. (2017), from indoor vacuumed dust samples collected in 30 French schools, and Boudehane et al. (2016), from indoor dust samples collected in a school in Algeria. In the latter, the BaP/BghiP ratio confirms, as in the present study, the predominance of vehicle exhausts as a PAH source. On the other hand, the PAHs in deposited dust could be affected by photochemical degradation after atmospheric transport, resulting in lower An and Fl levels compared to their isomers, due to the fact that these compounds are more susceptible to degradation (Galarneau, 2008).

Results from outdoor measurements show that lower PAH concentrations were observed in the second sampling with respect to the first one. Moreover, the PAHs are still higher than at the urban reference site, suggesting that the outdoor environment was under a decreasing influence of the fire. PAHs could be associated to fire, albeit it was almost extinguished during the second sampling.

Table 4
Results of PAH loads in deposited inhalable dust obtained during samplings performed on May 26 and June 03 at El Quiñon (EQ) and on May 26, 2016 at Miguel Delibes (MD) sites.

ng m^{-2}	EQ school (tire fire)				MD school (reference)	
	Indoor		Outdoor		Indoor	Outdoor
	May 26	Jun 03	May 26	Jun 03	May 26	May 26
Acenaphthylene (Acl)	<5	<5	<10	<10	<5	<10
Acenaphthene (Ace)	<0.5	<0.5	<1	<1	<0.5	<1
Fluorene (Flu)	<0.5	<0.5	<1	<1	<0.5	<1
Phenanthrene (Phe)	1.0	13	25	20	<0.5	1.6
Anthracene (An)	<0.5	1.2	1.3	<1	<0.5	<1
Fluoranthene (Fl)	1.4	3.8	27	8.2	1.8	6.5
Pyrene (Pyr)	1.4	10	64	13	6.5	24
Benz[*a*]anthracene (BaA)	1.2	<0.5	3.6	<0.5	<0.5	<1
Chrysene (Cry)	1.5	1.2	13	5.5	<0.5	<1
Benz[*b*]fluoranthene (BbF)	0.9	0.9	4.9	1.8	<0.5	<1
Benz[*k*]fluoranthene (BkF)	<0.5	<0.5	1.7	0.6	<0.5	<1
Benz[*a*]pyrene (BaP)	<0.5	0.65	1.8	<0.5	<0.5	<1
Dibenz[*a*,*h*]anthracene (DBA)	<0.5	<0.5	<1	<1	<0.5	<1
Benzo[*g*,*h*,*i*]perylene (BghiP)	1.8	1.0	3.2	<0.5	<0.5	<1
Indeno[1,2,3-*cd*]pyrene (Ind)	<5	<5	<10	<10	<5	<10

4. Conclusions

The impact of the very large and uncontrolled fire of the Seseña tire landfill on air quality and deposited inhalable dust at the El Quiñón (EQ) school has been documented through indoor and outdoor measurement and sampling of different parameters.

Ambient air measurements have been performed at the EQ school to get added information on the impact of the smoke plume on ambient air outdoor and infiltration within the school. Given their implications to children exposure (Diapouli et al., 2007; Franck et al., 2011) black carbon and ultrafine particles have been used as tracers of the tire combustion emissions and high concentrations of both have been associated to local impacts of a discontinuous and variable plume. These non-regulated pollutants complement the information provided by the air quality stations in the area.

BC has been monitored in the PM_{10} fraction outdoor and $PM_{2.5}$ indoor, although the intercomparison of both demonstrated that BC is mostly contained in the fine fraction of particles, where the plume is enriched. BC mass and PN concentrations have been found in the range of polluted urban or industrial areas. It has also been shown that even when windows are closed, there is an air exchange in which outdoor polluted air penetrates indoor by diffusion processes. It seems that opening windows for ventilation during non-impacted periods can be a proper measure to renew the air indoor. Onsite outdoor measurements should be done on a continuous basis to ensure this.

Inhalable deposited dust loadings have shown inorganic tracers of the tire combustion (Zn, sulfate and, possibly, Co) that were heavily enriched at the outdoor of EQ School. Only little proportions penetrated indoor, where concentrations were much more similar to the MD reference school, but still clearly higher. PAHs found in these samples were associated to combustion processes. Nevertheless concentrations were not very high and similar to those observed at other comparable (school) sites. More volatile PAHs were found at negligible concentrations. PAH with 4–5 fused aromatic rings reached the highest levels in outdoor measurements during the first sampling, before cleaning, in a ratio between 3 and 15 times if compared with those of MD reference school. In these samples detectable levels of higher molecular weight PAHs (5–7 ringed PAHs) were observed.

It is necessary to point out that this sampling approach on the basis of duration or choice of sampling technology can lead to additional bias. However, PAH characterization in indoor studies are still limited and more research is necessary to explore new sampling and analysis methods to achieve precise data (Oliveira et al., 2016; Boudehane et al., 2016). The study on PAH measurement in minimal mass loadings of deposited inhalable dust, as those collected in this study, can be used as preliminary consideration for fast screening of PAH measurement. Future studies comparing with classical sampling technology for exposure assessment can provide interesting alternative strategies based on advantages such as simplicity, easiness and use of minimal resources.

Exposure abatement such as cleaning classrooms and wall washing can contribute to diminish exposure to some components of deposited inhalable dust outdoor, for instance for PAHs, although the efficiency of cleaning activities for tire combustion particles should be assessed by samplings done immediately after these activities.

The possibility of using other mitigation/abatement strategies could have improved the indoor air quality in the school. For instance, Henderson et al. (2005) showed that the use of electrostatic air cleaners can reduce $PM_{2.5}$ between 63 and 88% for wildfires. In the current case, particles have some different properties that those produced in wildfires, i.e. particle size distribution and chemical composition, so the electrostatic air cleaner efficiency could be somehow different. Anyway, they could remove an important fraction of the particles and reducing $PM_{2.5}$ concentrations. Other kinds of air cleaners like fabric filters would also be possible. Otherwise, if plume keeps impacting the site regularly, some of these measures can become inefficient.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.03.148.

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