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E-waste dismantling as a source of personal exposure and environmental release of fine and ultrafine particles

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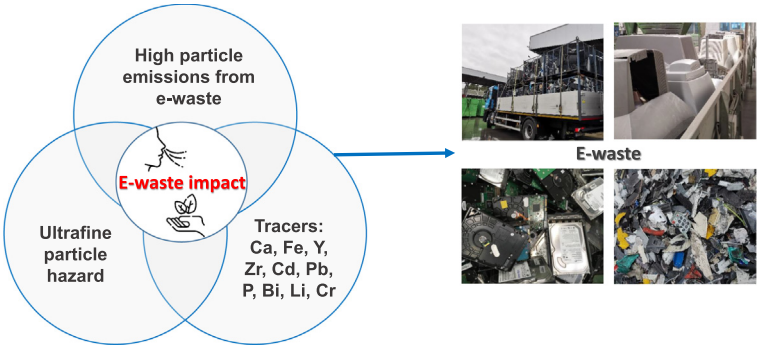
HIGHLIGHTS

- Electronic waste is one of the fastest growing waste streams in the world.
- Electronic-waste dismantling releases fine and ultrafine particles to outdoor and indoor air.
- Mechanical cutting of TV screen frames generated ultrafine particle emissions.
- Key tracers: Ca (plastic fillers), Fe (wiring), Y, Zr, Cd, Pb, P, Bi (cathode TV), Li, Cr (shredding).
- Aerosols generated did not evidence cytotoxic effects but generated reactive oxygen species.

GRAPHICAL ABSTRACT



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ABSTRACT

Electronic waste (WEEE; from TV screens to electric toothbrushes) is one of the fastest growing waste streams in the world. Prior to recycling, e-waste components (metals, wood, glass, etc.) are processed by shredding, grinding and chainsaw cutting. These activities generate fine and ultrafine particle emissions, containing metals as well as organics (e.g., flame retardants), which have high potential for human health impacts as well as for environmental release. In this work, release of fine and ultrafine particles, and their exposure impacts, was assessed in an e-waste recycling facility under real-world operating conditions. Parameters monitored were black carbon, particle mass concentrations, ultrafine particles, and aerosol morphology and chemical composition. Potential health impacts were assessed in terms of cytotoxicity (cell viability) and oxidative stress (ROS) on <2 μm particles collected in liquid suspension. Environmental release of WEEE aerosols was evidenced by the higher particle concentrations monitored outside the facility when compared to the urban background (43 vs.11 μgPM2.5/m^3, respectively, or 2.4 vs. 0.2 μgCa/m^3). Inside the facility, concentrations were higher in the top than on the ground floor (PM2.5 = 147 vs. 78 μg/m^3, N = 15.4 * 104 vs. 8.7 * 104/cm^3, BC = 12.4 vs. 7.2 μg/m^3). Ventilation was a key driver of human exposure, in combination with particle emissions. Key chemical tracers were Ca (from plastic fillers) and Fe (from wiring and other metal components). Y, Zr, Cd, Pb, P and Bi were markers of cathode TV recycling, and Li and Cr of grinding activities. While aerosols did not evidence cytotoxic effects, ROS generation was detected in 4 out of the 12 samples collected, associated to the ultrafine fraction. We conclude on the need for studies on aerosol emissions from WEEE facilities, especially in Europe, due to their demonstrable environmental and human health impacts and the rapidly growing generation of this type of waste.

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1. Introduction

The regulation of Waste Electrical and Electronic Equipment (WEEE) industry plays a key role in the environmental and circular economy agenda of the EU (Bruno et al., 2021). This EU regulation mandates that Member States must undertake reuse and recycling policies to increase the amount of waste reintroduced in the supply chain. As a result, WEEE generation and recycling is an exponentially growing research theme across the EU as well as globally (Anandh et al., 2021; Baldé et al., 2017).

In addition to the evident benefits in terms of environmental sustainability and waste reduction, WEEE recycling poses risks as a source of indoor and ambient air pollutants due to its impact on human exposure (Gangwar et al., 2019). The main activities taking place in WEEE recycling facilities (i.e., manual dismantling and mechanical grinding and shredding) may result in emissions of respirable fine particle and volatile species (e.g., VOCs, flame retardants; Baldé et al., 2017; Tansel, 2017) to which workers can be exposed. Inhalation is the main exposure route, although dermal exposures have also been reported (Stubbings et al., 2019; Wang et al., 2020). The literature is conclusive on the health impacts of exposure to ultrafine, fine and coarse particles (Héroux et al., 2015; Lelieveld et al., 2015; Oberdorster, 2000; Sisani et al., 2022), and specifically for flame retardants (Stubbings et al., 2019), evidencing that exposure to emissions from WEEE should be minimised. Finally, in addition to direct emission of air pollutants to indoor air at WEEE recycling facilities, this activity is also a source of ambient air pollutants given that recycling plants are typically open spaces with few (if any) physical barriers limiting air exchange with outdoor air. Adverse health effects for populations living in the vicinity of WEEE recycling plants have already been reported (Gangwar et al., 2019).

More than 50% of e-waste (electronic waste) in high-income countries (HICs) is shipped to low- and middle-income countries (LMICs), especially to Asia and Africa (Perkins et al., 2014), for recycling and disposal (Kumar et al., 2017). As a result, studies addressing particle emissions from WEEE recycling activities are available from different Asian locations (among others, Ahmed et al., 2015; Kim et al., 2019; Qiao et al., 2019; Wang et al., 2020). Conversely, studies in European facilities are still relatively scarce (Buiarelli et al., 2019; Simonetti, 2021; Julander et al., 2014; Papaoikonomou et al., 2018). This is because, although HIC have potential and infrastructure to recycle e-waste, a significant amount of WEEE is legally or illegally transported to LMICs having lax or no regulation (Ghimire and Ariya, 2020), to handle e-waste with little or no safety precautions and knowledge. This places both the environment and public health at serious risk. In addition to the scarcity of data from EU WEEE plants, the studies available focus mainly on emissions and exposure to flame retardants present in e-waste, with limited assessment of the metal components in WEEE, their sources and potential impacts on particle toxicity. Namias (2013) suggested that e-waste contains up to 60 metals including copper, gold, silver, palladium and platinum, in addition to Fe, Cd, Pb, among others.

This work aimed to fill this research gap by monitoring and characterising indoor and ambient particle emissions from a WEEE recycling facility in the vicinity of Barcelona (Spain), considered representative of EU standards and regulations for this industry. Results on the metal tracers of this activity are presented here, while those on organic species (e.g., brominated flame retardants, plasticisers) are reported elsewhere (Balasch et al., 2022). The final goal was to explore the implications in terms of human exposure, environmental release and particle toxicity from this rapidly growing industrial sector.

2. Materials and methods

2.1. Particle emission and exposure scenarios

As stated above, the purpose of this work was to generate data on dust emissions and potential impacts on human exposure. It was not designed as an occupational health assessment, and as a result it does not follow occupational health protocols (e.g., regarding personal sampling or sampling at breathing height). In this framework, ultrafine, fine and coarse particle emissions were monitored and sampled in a WEEE recycling plant in the vicinity of Barcelona, Spain. The plant, fully compliant with the current EU standards and regulations for this industry, receives and recycles a broad range of waste electrical and electronic equipment such as large and small household appliances (from slot machines to TVs and electrical toothbrushes), computer and telecommunications equipment, lightning equipment, toys or sport and leisure equipment, among others. WEEE processing in the plant covered activities from manual classification to mechanical shredding, crushing and circular saw cutting, in order to dismantle the different components (metals, wood, glass, etc.) prior to recycling. All processing stages were mechanical operations; neither chemical nor thermal treatments were used in the dismantling process.

Particle sampling was carried out during two weeks in October and November 2020. Two different emission scenarios were monitored:

- *October* (*week 1*): the emission scenario monitored was the top floor of the plant, where cathode-ray tube TVs and computer screens were dismantled. In this floor air exchange and renewal was estimated to be limited, even though it was not monitored, due to the absence of mechanical or natural (windows/doors connecting with outdoor air) ventilation. The main air exchange occurred through an opening (roughly 1.5 × 2.5 m) in one of the walls through which the screens were introduced from the ground floor. The samples were collected each day during the afternoon shift, from 14:00 h to 22:00 h.
- *November* (*week 2*): ground floor, where manual selection and grinding of general WEEE (plastic, electrical cables, gaming machines, etc.) took place. Although it was also not equipped with a mechanical system, the natural ventilation on this floor was significantly higher in comparison to the top floor, as it was directly connected to the entrance where trucks unloaded the e-waste, by open gates (10 × 15 m, approximately). In addition, the surface area of the ground floor was approximately double that of the top floor. The use of diesel forklifts in this plant was one source to be considered in terms of ultrafine particle emissions, as the forklifts were operating continuously across the ground floor. The samples were collected each day during the afternoon shift, from 14:00 h to 20:00 h.

The monitoring instrumentation was deployed in the worker area (WA) in each of the indoor scenarios, aiming to capture worker exposure while avoiding any interference with the industrial activity. Finally, in addition to these indoor locations, a set of homologous instrumentation was deployed in parallel in an outdoor space representing background concentrations (BG), which was also influenced by vehicular traffic (diesel trucks and forklifts offloading the waste). At all locations, the particle monitors and samplers were placed side by side on a table at approximately 1.5 m above the ground. All instruments were connected to electrical power and they each sampled air through individual conductive tubing (<20 cm in length). In the WAs, the tables were located at approximately 2 m from the workers. In the outdoor space, the table was located close to the entry gate to the plant. While it was acknowledged that the spatial variability of dust concentrations was expected to be high, especially on the ground floor, monitoring in different locations across the plant was unfortunately not feasible due to the logistics and activities of the plant itself.

2.2. Particle monitoring

Particle number (N) and mass concentrations, size distribution, and mean diameter (Dp) were monitored with online instruments over 24 h periods:

- Miniature diffusion size classifier DiscMini (TESTO AG), reporting particle number concentrations and mean particle diameter between 10 and 700 nm, with a 1 min time resolution.
- Light-scattering laser photometer DustTrak TM DRX (TSI Model 8533), for total and size-segregated particle mass concentrations in the range

250–3200 nm with 1 min time resolution.
- Optical particle size (OPS, TSI Model 3330) for total and size-segregated particle mass concentrations in the range 0.3–10 μm across 16 channels with 1 min time resolution.
- MicroAeth® AE51 Black Carbon aerosol monitor, monitoring black carbon (BC) concentrations with a 1 min time resolution.

2.3. Sample collection

Aerosol samples were collected on different substrates for the following offline determinations:

- Particle chemical composition: during the work shifts in each floor, PM10 (particle size matter of 10 μm) and PM2.5 (particle size matter of 2.5 μm) samples were collected on quartz microfiber filters (37 mm PTFE filters) using Personal Environmental Impactors (PEM, SKC®). In addition, three particle size fractions (>2.5 μm, 2.5–0.25 μm, <0.25 μm) were collected, the coarser stages on polycarbonate 25 mm filters and the last stage on Teflon 37 mm PTFE filters, using Personal Cascade Impactors (PCIS, SKC®) connected to portable SKC Leland pumps (9 L/min). The inorganic particle chemical composition was determined by ICP-MS and ICP-AES after an acid digestion procedure of the samples (Querol et al., 2001). Halogenated flame retardants (HFRs) and organophosphate esters (OPEs) were also analysed, reported in Balasch et al., 2022
- Particle toxicity: twelve 30-minute samples were collected using an SKC BioSampler® connected to a sonic-flow BioLite + pump (12.5 L/min) (López et al., 2021). Particles <2 μm were collected in liquid suspension in Dulbecco's Modified Eagle's Medium (DMEM; Sigma, Steinheim, Germany). The suspensions were stored at 2 °C after sample collection. Sampling time must be short (30 min) to minimise evaporation and avoid potential fungal growth in the medium. This, however, limits the comparability between the toxicity results and the mean chemical composition (sampled over 6 or 8 h shifts), as will be discussed below.
- Particle morphology: particles were collected on Transmission Electron Microscopy (TEM) grids (Quantifolil® Au grids with 1 μm diameter holes - 4 μm separation of 200 mesh) placed in sampling cassettes (SKC INC., USA, inlet diameter 1/8 in. filter diameter 25 mm) following the sampling setup described by Tsai et al., 2008 and Ribalta et al., 2019. The cassettes were connected to an SKC Leland pump (3 L/min). Samples were collected during the afternoon shift (14:00 h to 22:00 h in the top floor and 14:00 to 20:00 h on the ground floor).

The gravimetric analysis was performed according to the European directive EN1234-1. It included pre-sampling and post-sampling weighing of each filter (including blanks). The pre-weighing and post-weighing were carried out following the same procedure: conditioning temperature 20 °C and relative humidity 50%. Each sample was weighed three times (24-hour interval in between) and the average value was recorded. The filters were weighed using a microbalance (Mettler-Toledo, model: XP105 with electrostatic charge detection, Switzerland).

2.4. Cell viability and generation of reactive oxygen species

The oxidative potential (OP) of fine particles describes their ability to generate oxidative and inflammatory effects in bronchial cells (Daellenbach et al., 2020; Imai et al., 2008; Kelly, 2003; Pietrogrande et al., 2021; Sisani et al., 2022). The oxidative stress generated by exposure to aerosols can be estimated using acellular and cellular methods (Sisani et al., 2022). typical acellular methods are based on dithiothreitol (DTT) consumption or antioxidant depletion assays (using ascorbic acid, AA, or glutathione, GSH), while cellular tests are considered more representative of the actual processes and interactions taking place during cell exposure to aerosols. In cellular tests, exposure may be simulated through submerged cell cultures or by air-liquid interaction (ALI) (Bessa et al., 2020a; Bessa et al., 2020b). In this work, the human alveolar cell line A549 was used to assess the toxicity of the samples. It was maintained at 37 °C, 5% CO_2, in a humidified incubator, and, before performing the assays, cells were allowed to reach 80% confluence in 75 cm^2 culture flasks (Corning, NY, USA) containing culture medium DMEM (Sigma, Steinheim, Germany) supplemented with 10% fetal bovine serum (Sigma, Steinheim, Germany), 50 U/mL of penicillin and 50 μg/mL streptomycin (Gibco, Paisley, Scotland, UK). Two different assays were performed on the aerosol samples collected:

- Cell viability assay: 5 ∗ 104 cells were plated in 96-well plates and incubated 24 h before exposure to 100 μL of sampling medium or fresh medium (control cells) for 24 h. Alamar Blue (AB, Thermo Scientific™) and 5-carboxyfluorescein diacetate acetoxymethyl ester (CFDA-AMMolecular Probes, Invitrogen, Spain) where the dyes used to estimate metabolic activity and membrane impairment, respectively. The fluorescence of dyes was read at the excitation/emission pairs of 530/590 (AB) and 485/530 nm (CFDA-AM), in a Tecan Infinite M Plex plate reader (Männedorf, Switzerland). The percentage of viable cells was calculated by dividing the relative fluorescent units (RFUs) of exposed cells by RFUs of control cells, multiplied by 100.
- ROS generation was estimated by the oxidation 2′,7′-dichlorofluorescin (H2DCF-DA) (Sigma, Steinheim, Germany), which is deacetylated by membrane esterases and then oxidized by intracellular ROS, becoming fluorescent (LeBel et al., 1992). Briefly, 5 ∗ 10^4 cells were seeded in 96-well plates and allowed to attach for 24 h. The culture medium was replaced by 20 μM H2DCF-DA diluted in DPBS (1:10) supplemented with 10 mM glucose (DPBS-Glu) and the plate incubated for 30 min at 37 °C, 5% CO_2. Cells were washed with PBS and exposed to sampling medium, fresh medium (control) or 5 μM 3-morpholinosydnonimine (SIN-1; positive control). The emitted fluorescence of DCF was measured at ex/em pairs of 485/528 nm, after 15, 30, 60 and 120 min of exposure to sampling medium. Generation of ROS was expressed as fold induction (RFUs of exposed cells divided by RFUs in control cells).

3. Results and discussion

3.1. Exposure metrics and concentrations

The results on particle concentrations from the online instruments are summarised in Table 1 and shown in Fig. 1 as density plots. As discussed in the Materials and methods section, measurements were carried out under real-world operating conditions, representative of the facility's usual activities. The concentrations of all of the parameters were higher in the top floor (Table 1), probably resulting from the lower ventilation in this area of the plant. As described above, there were no open windows connecting the top floor with outdoor air. Furthermore, the meteorological conditions during the second campaign favoured particle scavenging outdoors and therefore reduced potential infiltration of ambient aerosols (Chen et al., 2020), as it rained during 2 out of 4 days of the second week. Mean daily fine particle concentrations (PM2.5) were relatively higher in the top floor (147 μg/m^3) than on the ground floor worker area (78 μg/m^3), while the background concentrations were relatively similar (37–43 μg/m^3). The indoor/outdoor (I/O) ratio for PM2.5 was higher for the top (3.4) than for the ground floor (2.1), which would indicate the influence of higher PM2.5 emissions and/or lower ventilation in the top floor. It is not possible with the available data to discern which factor was dominant in the top floor, whether the higher emission or the lower ventilation. Conversely, for the rest of the parameters monitored (PM10, N, BC), I/O ratios were very similar for both floors (2.6–2.7, 3.9–4.0 and 4.4–4.9, respectively). On the ground floor, where natural ventilation was significant given that the gates were fully open at all times, the high I/O ratios (ranging between 2.1 for PM2.5 to 4.4 for BC) confirm the impact of the WEEE recycling activities on air pollutant concentrations and, subsequently, on personal exposure. Thus, natural ventilation alone does not seem sufficient to maintain low levels of exposure to pollutants in the facility.

The density plots in Fig. 1 were used to understand the concentrations as well as the potential variety of emission sources impacting particle

Table 1
Maximum, minimum, mean and standard deviation (SD) of the different parameters (black carbon (BC), number concentration (N), diameter (dP), mass concentration (PM2.5 and PM10)) sampled in both campaign (October and November) in both scenarios (worker areas: top and ground floor) and back ground (BG).

		BC (μg/m^3)				N(/cm^3)				Dp (nm)				PM2.5 (μg/m^3)				PM10 (μg/m^3)			
		Max	Min	Mean	SD	Max	Min	Mean	SD	Max	Min	Mean	SD	Max	Min	Mean	SD	Max	Min	Mean	SD
October 2020	Top floor	35,105	2911	12,357	4060	586,084	34,958	153,652	73,442	51	25	37	6	1769	49	147	116	4804	90	357	317
	BG	29,165	13	2518	2664	622,254	2455	39,197	62,554	89	10	43	22	290	17	43	28	1770	703	131	139
November 2020	Ground floor	24,268	1903	7156	4105	316,202	20,496	86,547	46,124	48	20	32	5	901	33	78	83	2280	63	172	210
	BG	6981	37	1623	1320	216,426	3620	21,757	27,926	76	15	46	14	182	22	37	22	872	18	65	100

concentrations. BC concentrations, for example, were higher on average on the top than on the ground floor (12.4 vs. 7.2 μg/m^3), but the density profiles were highly similar on both floors with unimodal distribution, which may suggest the impact of a single, relatively uniform emission source. The most plausible interpretation is that the main source of BC was diesel engines, both from the forklifts operating on the ground floor and from

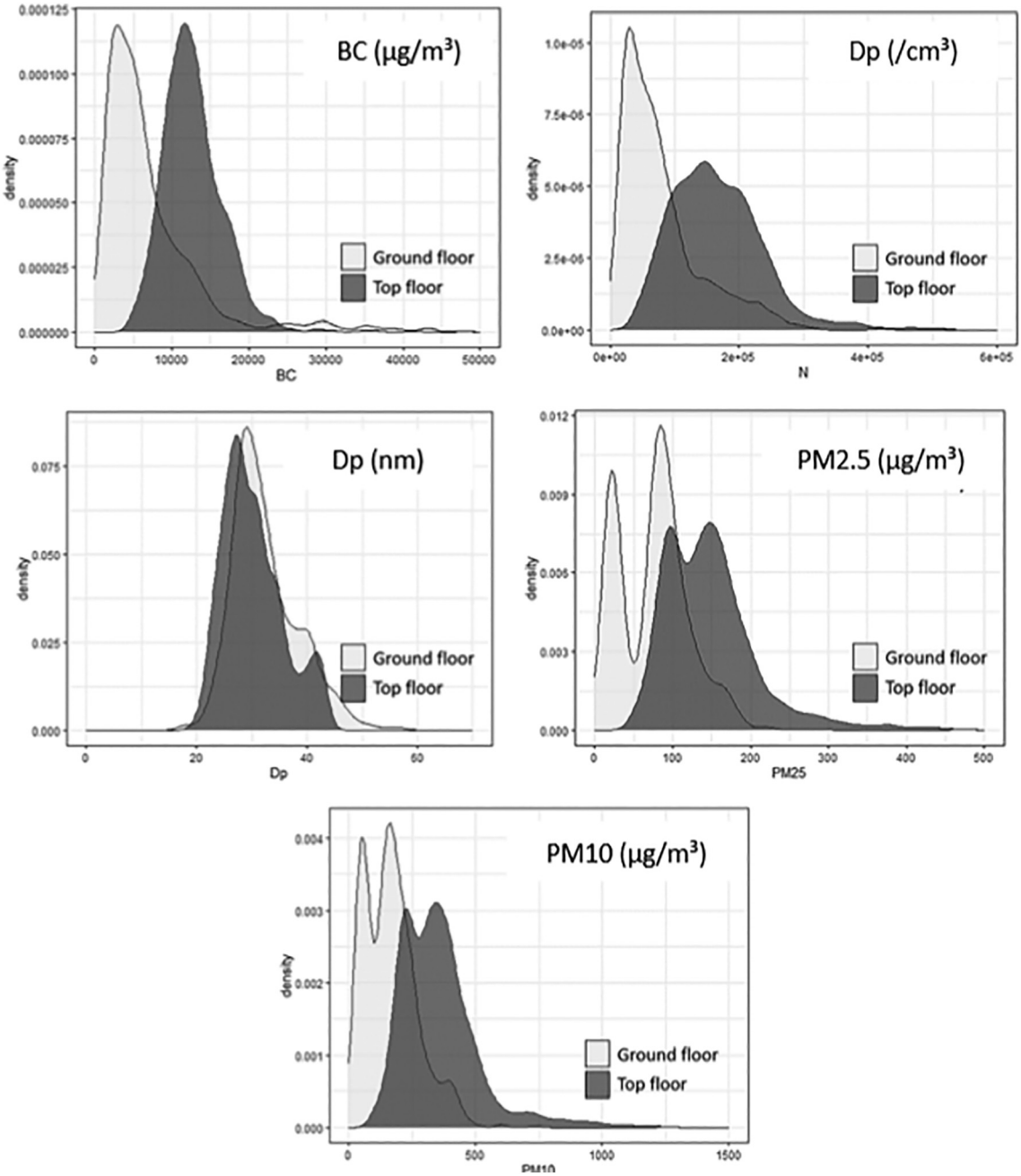

Fig. 1. Density plots of the different worker areas (Top floor and ground floor) of both campaigns (October and November), BC: Black Carbon (μg/m^3); N: Number concentration (/cm^3); Dp: Diameter (nm); PM2.5 (μg/m^3); PM10 (μg/m^3).

the trucks delivering the WEEE, which due to the effect of natural thermal convection and air exchange between both floors, caused them to concentrate on the top floor. Aside from this, a certain influence of monitoring artefacts caused by iron interference (Derimian et al., 2008) could not be discarded.

Particle number (N) concentrations, on the other hand, showed different density profiles on both floors and the highest differences in concentrations between floors. The top floor data showed higher concentrations on average than the ground floor ($15.4 * 10^4/m^3$ vs. $8.7 * 10^4/cm^3$), as was observed with BC, but with a wider distribution, which suggests a greater variety of sources. These were probably due to the emissions generated during mechanical cutting of the TV screen frames (metallic). On the ground floor, concentrations were lower and less variable (narrower density plot), suggesting a single source with relatively constant emissions (probably, trucks and diesel forklifts, as in the case of BC). In terms of particle diameter, mean Dp was very similar on both floors (32–37 nm; characteristic of diesel soot particles; Tritscher et al., 2011), also with a mostly unimodal distribution, with only a small secondary peak in the top floor probably linked to the incidental formation of new particles (Salmatonidis et al., 2018; Viana et al., 2017), due to the emissions of volatile compounds from the mechanical cutting saw.

Finally, PM10 and PM2.5 showed bimodal density distributions, suggesting the influence from at least two different sources with different concentrations, or a single source with two different emission profiles. The variety of activities taking place in the recycling plant supports this interpretation.

In order to understand the magnitude of the concentrations of the different metrics monitored in the exposure scenarios, the results were compared in terms of PM2.5 and N with representative urban background (UBG) concentrations (monitored in Barcelona city, Palau Reial EU-reference station) (Fig. 2). The concentrations inside the plant (in the worker areas, WA) were always higher than in the outdoor air at the WEEE recycling facility (background, BG). As expected, the concentrations in BG were, in turn, higher than in UBG due to the influence of fugitive emissions from the process and emissions from the forklifts and the trucks offloading the WEEE. This confirmed that the different activities in this e-waste recycling plant, considered representative of EU standards, generated distinct particle emissions with potential to impact ambient air quality at local scale as well as human health. In terms of indoor air, particle concentrations monitored (maximum 8-hour PM4 recorded = 1485.92 $\mu g/m^3$) did not exceed current limit values (3 mg/m^3 as time-weighted average).

In addition, a literature review was carried out to identify studies reporting air quality assessments at WEEE recycling facilities, as a basis for establishing a comparative scenario to analyses the results obtained in this study. Detailed information can be found in Table 2. The review accounts for studies published between 2006 and 2020 in peer-reviewed journals, based on research databases Scopus and ScienceDirect. The search terms included “air quality”, “e-waste”, “PM” and “heavy metals”. Activities assessed mainly comprised dismantling, mechanical workshops (shredding, milling), and burning (possibly, uncontrolled) to recover valuable materials. Most of the studies have been performed in China, followed by Ghana and India, with studies in European facilities being rather scarce (Buiarelli et al., 2019; Julander et al., 2014; Papaoikonomou et al., 2018), as discussed above.

PM mass concentrations in different size ranges are the most frequently reported parameter, with TSP (total suspended particles) ranging between 124 and 5066 $\mu g/m^3$, PM10 between 173 and 1674 $\mu g/m^3$, and PM2.5 between 11 and 2774 $\mu g/m^3$ (Table 2) worldwide. Concentrations reported in European facilities were, unexpectedly, at the middle/upper end of the range. This could be due to the fact that recycling activities in Asian and African countries are frequently performed in open spaces and with scarce monitoring, under non-existent or very poor regulatory frameworks and protocols. Conversely, European facilities are enclosed (indoors) and monitored under EU regulations. A similar pattern is observed for flame retardant levels, which are higher in Europe and other HICs compared to LMICs (Ding et al., 2018; Nguyen et al., 2019; Tao Wang et al., 2018). In any case, concentrations are largely variable and dependent on the type of activity, proximity to the source and ventilation conditions, with mechanical activities mainly affecting coarse particles while fine particles are more influenced by combustion processes (Table 2). In our study, hourly PM2.5 concentrations recorded in the working area and during working hours were in the range 17–404 $\mu g/m^3$, showing high variability, and with a mean value (147 $\mu g/m^3$) in the range of those more commonly reported in literature. To the authors' knowledge, little is known about the behavior of quasi-ultrafine and ultrafine particles in these environments, with only one study in Italy reporting on $PM_{0.1}$ mass concentrations, which ranged between 6.6 and 25.9 $\mu g/m^3$ (Buiarelli et al., 2019). Another study reported particle number (N) concentrations in US, recording mean concentrations of $25 * 10^4/cm^3$, associated with shredding activities (Ceballos et al., 2020). N concentrations in our study were slightly lower, with mean hourly concentrations during work shifts in the range $2.2 * 10^4$–$15.4 * 10^4/cm^3$ (Table 1) while reaching 5-min maximum concentrations recurrently $>20 * 10^4/cm^3$. As a result, it may be concluded that the concentrations recorded at the WEEE recycling facility under study are comparable, even if slightly lower, than those reported for other European studies in terms of particle mass (PM2.5, PM10) and number concentrations.

3.2. Chemical fingerprint of WEEE emissions

The PM < 0.25, PM0.25–2.5 and PM > 2.5 samples collected on the top and ground floors were analysed in terms of major and trace inorganic species. For the sake of simplicity and comparability with concentrations at a nearby urban environment, values reported in the main text will correspond to the PM2.5 fraction. Organic and elemental carbon (OC and EC) were also determined on a limited number of samples (two 24 h-PM10 samples on the top floor and two on the ground floor) due to the low availability of samples collected on quartz microfiber filters. In both floors, the key

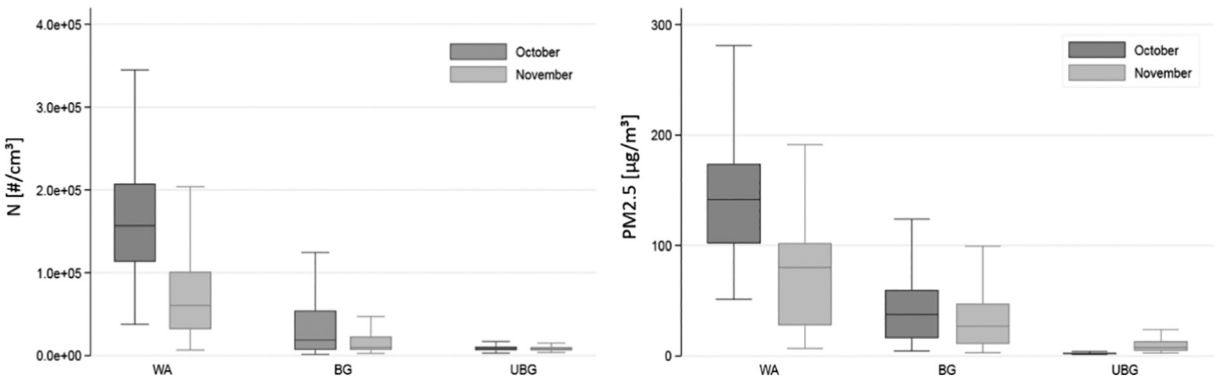

Fig. 2. Number concentrations and PM2.5 box plots comparing the different measurement areas in both campaigns (October and November).

Table 2
Literature review of other studies on WEEE recycling plants.

Study	Country	Activities	Pollutant	Concentration	Comments regarding heavy metals
(Kim et al., 2015)	US	(1) A chain-shredder, (2) a hammermill, and (3) a cathode tube disassembly line used solely to disassemble and recover components from monitors and televisions	PM2.5–10 PM0.1–2.5	174–612 μg/m^3 49–596 μg/m^3	Compared to coarse PM measurements from a regional near-roadway, Pb and Ni were enriched 170 and 20 times, respectively
(Gangwar et al., 2019)	India	E-waste burning	PM10	243.310 ± 22.729 μg/m^3	Emission from open air burning resulted in high exposure of Pb, Cr, Ni, Cu and Zn to the residents of the area
(Zheng et al., 2016)	China	Open burning and other e-waste recycling operations (not specified)	PM2.5	11–160 μg/m^3	Higher values of Pb and Cd compared to the reference site
(Bungadaeng et al., 2019)	Thailand	Open burning processes. Compiling and sweeping	PM2.5–10 PM2.5	441 ± 496 μg/m^3 2774 ± 4713 μg/m^3	
(Fang et al., 2013)	China	Mechanical workshops	PM10 PM2.5	326.3–394.5 μg/m^3 252.6–290.8 μg/m^3	Pb and Cu were the most enriched metal in the $PM_{2.5}$ and PM10 samples, respectively
Ceballos et al., 2020	US	Shredding	$N > 10$ nm PM2.5	250 #/cm^3 171 μg/m^3	
Bi et al., 2010	China	Printed Circuit Boards (PCB) recycling workshop	TSP	1129–1688 μg/m^3	Cd, Pb, Zn, Cu, Sb and Ni mainly caused by the burning of PCBs. High OC (381.9–562.6) and EC (26.3–42.4 μg/m^3) concentrations
(Deng et al., 2006)	China	Open burning and other e-waste recycling operations (not specified)	TSP PM2.5	124 ± 44.1 μg/m^3 62.12 ± 20.5 μg/m^3	Cr and Zn were the most enriched metals in TSP and PM2.5, followed by Cu, Pb, Mn and As. All metals exhibited a predominant occurrence in the fine particulate fraction
(Xue et al., 2012)	China	PCB recycling workshop	TSP PM10	282.6 μg/m^3 202.0 μg/m^3	TSP and PM10 enriched by Cr, Cu, Cd, Pb
(Kwarteng et al., 2020)	Ghana	Open burning and other e-waste recycling operations (not specified)	PM10 PM2.5	214 μg/m^3 88 μg/m^3	
Papaoikonomou et al., 2018	Greece	Dismantling and temporary storage	PM10	382.4 ± 104.8 μg/m^3	As and Pb concentrations in the indoor samples were 140 and 40 times, respectively, higher than those measured in the ambient air of nearby city. CRT (cathode ray tubes) processing release Sr, Pb, Cd and As
Buiarelli et al., 2019	Italy	Dismantling activities	PM0.1 PM0.1–2.5 PM2.5 PM10 TSP	17.6 (6.6–25.9) μg/m^3 477.7 (147.1–704.6) μg/m^3 495.2 (153.7–730.5) μg/m^3 1674.4 (599.6–3077.9) μg/m^3 5066.8 (2309.7–9682.0) μg/m^3	
Ding et al., 2018	China	Not specified (rural e-waste recycling area covering an area of about 330 km^2)	PM2.5	182 ± 62.0 μg/m^3	
Nti et al., 2020	Ghana	Collecting, dismantling and open air burning of electrical cables to recover copper	PM2.5 PM10	61.18–70.69 μg/m^3 173.49–214.43 μg/m^3	
Julander et al., 2014	Sweden	Indoor work (i.e., tasks involving handling of goods, for example, sorting of incoming and outgoing goods, truck driving, cleaning, supervision of work), outdoor work (e.g., mainly inspection tasks and transportation of goods), and office work (i.e., computer work with no time in the production buildings)			V, Sb, In, Pb and Hg showed significant correlations between air and blood/urine. Significantly higher levels of Cr, Co, Pb, In and Hg was found in recycling workers. Rare metals such as In and Sb must be monitored in these settings.
Wang et al., 2009	China	Dismantling and recovery processes			Cd, Cu and Pb were likely found to be originated from e-waste related activities
Kuntawee et al., 2020	Thailand	Not specified			House dust and airborne levels of Cr, Hg, Ni, and Pb were higher in the homes and environment of subjects involved in e-waste activities
(Wittsiepe et al., 2017)	Ghana	Not specified			Significantly high median concentrations of Pb, Cd, Cr and Ni
(Ngoc Ha et al., 2009)	India	Dismantling, extraction of valuable metals			Concentrations of Cr, Mn, Co, Cu, In, Sn, Sb, Tl, Pb and Bi in air from the e-waste recycling facility were relatively higher than in city
(Ceballos et al., 2017)	US	Electronic dismantling, automated CRT cutting, electronics shredding, batteries sorting			Overexposure to Pb and Cd

tracers of e-waste (Julander et al., 2014; Kuntawee et al., 2020; Papaoikonomou et al., 2018) and industrial activities (e.g., diesel combustion, dust re-suspension, (Gulia et al., 2019)) were detected. Tables 3 and 4 show the mean concentrations of trace and major elements, respectively. The presence of key tracers was corroborated with TEM images (Fig. 3).

As observed for the online parameters described above, element concentrations in the top floor were higher than in the ground floor (Tables 3 and 4 and Figs. 4 and 5). This was linked to the poor ventilation in the top floor and the more favourable weather conditions during the week when sampling was carried out on the ground floor Ca (7.0–15.6 μg/m^3 range in the top floor and 3–6.4 μg/m^3 range in the ground floor) and Fe (4.5–10.3 μg/m^3 range in the top floor and 1.9–5.4 μg/m^3 range in the ground floor). Ca is a known mineral filler of plastics (Thenepalli et al., 2015), indicating the contribution of this material to PM_x in the whole facility. Another sign of the impact of plastics handling is probably the high daily OC concentrations recorded, in the range 28–84 μg/m^3. Similarly, EC concentrations were relatively high (between 5 and 19 μg/m^3), probably linked to the diesel-powered forklifts operating in the facility, as well as from the trucks delivering the waste at the gate of the facility. In spite of expected sources of carbonaceous aerosols at the different sampling

locations, it should be noted that EC concentrations could be overestimated due to potential interference from iron oxides during the analysis, as described for BC above.

Regarding trace inorganic species, especially high concentrations were recorded for Zn (105–2569 μg/m^3), Pb (167–1797 μg/m^3) and Ti (105–2570 μg/m^3) in both working areas. Ba, Y, Mn, Sr, Cu, and P were also found in relatively high concentrations, with maxima in the top floor (624, 576, 270, 419, 618 and 263 μg/m^3, respectively), compared to the ground floor (139, 43, 196, 75, 201 and 155 μg/m^3, respectively). All of these elements are typically found in air emissions from this type of industry (Table 2). To further explore the compositional variability of the particles and link it to potential emission sources, a correlation analysis was carried out (Table S1 in Supporting Information). Results showed a high degree of correlation between most of the metals and tracers, probably owing to the mix of residues being processed in the plant. Conversely, this analysis also highlighted specific tracers (Cd, As, Bi, P) due to their lack of correlation with the majority of the metals, which identified them as markers of specific sources inside the plant (e.g., the P-powder emitted during dismantling of TV screens on the top floor). Thus, this correlation analysis supported the identification of single elements as markers of the emissions in the plant, which also coincided with the data reported in the literature (Table 2).

In order to understand the influence of ventilation, two tracers were used as indicators of outdoor infiltration and airtightness of the top floor: SO_4^{2-}, due to its long-range transport origin, and V, tracer of fuel combustion emissions (e.g., maritime, oil refineries), not located in the vicinity of the WEEE facility. The ratios top/ground floor for both indicators were in the range 1.5–3.5, allowing us to conclude that compounds with ratios top/ground floor > 4 could be considered as specific tracers of cathode TV recycling, the main activity taking place on the top floor. Based on this analysis (Figs. 4 and 5), Y, Zr, Cd, Pb and Bi may be identified as tracers of cathode TV recycling. According to the literature, Y is a component of cathode ray TV tubes (Innocenzi et al., 2013; Lecler et al., 2015). As regards the grinding activities on the ground floor, the main tracers could be Li and Cr (Fig. 5 and Table 4), found in higher concentrations on the ground than on the top floor.

Aside from the relative concentrations between both floors, the airborne concentrations (and therefore, exposure) of heavy metals on both floors, as well as in outdoor air at the WEEE facility (BG), were higher than usual concentrations in the urban background (UBG) (Figs. 4 and 5). The highest ratios in PM2.5 between BG and UBG, indicating environmental release from the WEEE activities, were obtained for Ca (ratio = 19), Fe (9), Mg (14), Li (16), P (22), Ti (58), Cu (10), Sr (64), Y (965), Ba (30) and Pb (23). Consequently, these tracers can be considered indicators of particle release and therefore of the impact of this type of industry on the environment.

Finally, in terms of size fractions, concentrations of all compounds were clearly higher in the range >2.5 μm than in <2.5 μm aerosols at the three monitoring sites (Figs. S1 and S2). Ratios top/ground floor were similar for particles <0.25 μm and >2.5 μm. The elements with the highest abundance in the finest fraction on the top floor were As and Ti, while on the ground floor these were As, Cu, Mn and Cr.

Although the aim of this study was not to assess occupational exposure, the inhaled dose of PM2.5 and of key trace metals released from e-waste recycling were estimated, based on the methodology described by Maceira et al. (2019) and applied in the companion article on flame retardants in the same e-waste plant (Balasch et al., 2022). This methodology assumes an average body weight of 70 kg and a volume of air inhaled of 19.92 m^3/day. Results showed that the inhaled PM2.5 dose was 14.0 μg/kg body weight during working hours on the top floor and 5.6 μg/kg body weight on

Table 4
: Maximum and minimum levels in PM2.5 of major elements (μg/m^3) in both campaigns in the worker area (WA) and in the back ground (BG) and mean annual levels of major elements in Palau Reial cabin that corresponds the levels in PM2.5 of the urban back ground (UBG).

(μg/m^3)	Top floor (October 2020)				Ground floor (November 2020)				PM2.5 major elements average 2020 in Palau Reial station
	WA		BG		WA		BG		
	Max	Min	Max	Min	Max	Min	Max	Min	
SiO_2	24.8	12.5	5.4	2.0	10.6	3.8	3.5	1.1	0.6
Al_2O_3	8.3	4.2	1.8	0.7	3.5	1.3	1.2	0.4	0.2
Ca	15.6	7.0	3.8	2.3	6.4	3.0	2.4	0.8	0.2
Fe	10.3	4.5	1.3	1.1	5.4	1.9	0.9	0.5	0.1
K	2.1	1.4	0.9	0.8	1.7	1.0	0.9	0.3	0.1
Mg	2.2	1.0	1.3	0.3	0.7	0.3	0.9	0.1	0.0
Na	3.7	1.4	0.9	0.5	0.9	0.3	0.6	0.1	0.2
$SO_{4^{2-}}$	7.4	3.6	1.8	1.3	3.6	1.0	2.4	0.3	1.2

Table 3
Maximum and minimum levels in PM2.5 of trace elements (ng/m^3) in both campaigns in the worker area (WA) and in the back ground (BG) and mean annual levels of trace elements in Palau Reial cabin that corresponds the levels in PM2.5 of the urban back ground (UBG).

ppb	Top floor (October 2020)				Ground floor (November 2020)				PM2.5 trace elements average 2020 in Palau Reial station
(ng/m^3)	WA		BG		WA		BG		
	Max	Min	Max	Min	Max	Min	Max	Min	
Li	10.3	1.8	1.2	<0,01	24.5	0.6	1.7	<0,01	0.06
P	**263.3**	**164.9**	**264.2**	**21.4**	**155.1**	**71.1**	**77.9**	**4.2**	7.60
Ti	**2569.6**	**300.1**	**1886.0**	**98.4**	**195.3**	**105.2**	**60.4**	**30.3**	5.89
V	10.9	5.4	2.0	1.3	4.3	0.6	0.8	<0,01	1.02
Cr	35.8	<0,01	15.2	<0,01	48.2	2.7	3.0	<0,01	1.09
Mn	**270.6**	**92.9**	**32.2**	**26.0**	**195.9**	**56.8**	**24.4**	**15.4**	4.05
Co	17.8	7.6	1.3	<0,01	3.4	1.7	<0,01	<0,01	0.07
Ni	61.2	22.7	18.4	8.3	23.9	6.0	<0,01	<0,01	1.06
Cu	**617.5**	**99.0**	**76.9**	**16.8**	**201.0**	**45.0**	**31.1**	**17.2**	3.95
Zn	**2371.6**	**1195.4**	**351.5**	**174.8**	**1053.8**	**275.8**	**341.6**	**103.2**	34.70
As	5.1	1.5	1.3	<0,01	5.4	<0,01	<0,01	<0,01	0.24
Rb	8.1	3.6	1.5	<0,01	3.3	<0,01	0.7	<0,01	0.23
Sr	**419.0**	**174.2**	**38.1**	**32.3**	74.7	40.3	16.5	14.2	0.77
Y	**576.0**	**215.8**	**18.0**	**12.8**	42.5	28.0	11.8	6.1	<0,01
Zr	50.7	23.0	10.3	2.4	7.7	5.9	0.7	<0,01	2.37
Cd	31.4	10.3	0.8	<0,01	4.0	0.8	<0,01	<0,01	0.07
Sn	76.9	31.8	12.7	5.9	23.0	10.1	3.1	1.8	0.89
Sb	40.6	18.6	4.1	2.8	10.3	6.3	2.6	<0,01	0.51
Ba	**624.4**	**302.9**	**93.9**	**41.0**	**138.9**	**75.6**	**31.0**	**25.3**	2.86
Pb	**1796.9**	**830.4**	**117.0**	**76.0**	**286.4**	**166.7**	**45.6**	**38.7**	2.80

Elements with higher concentrations are highlighted in bold.

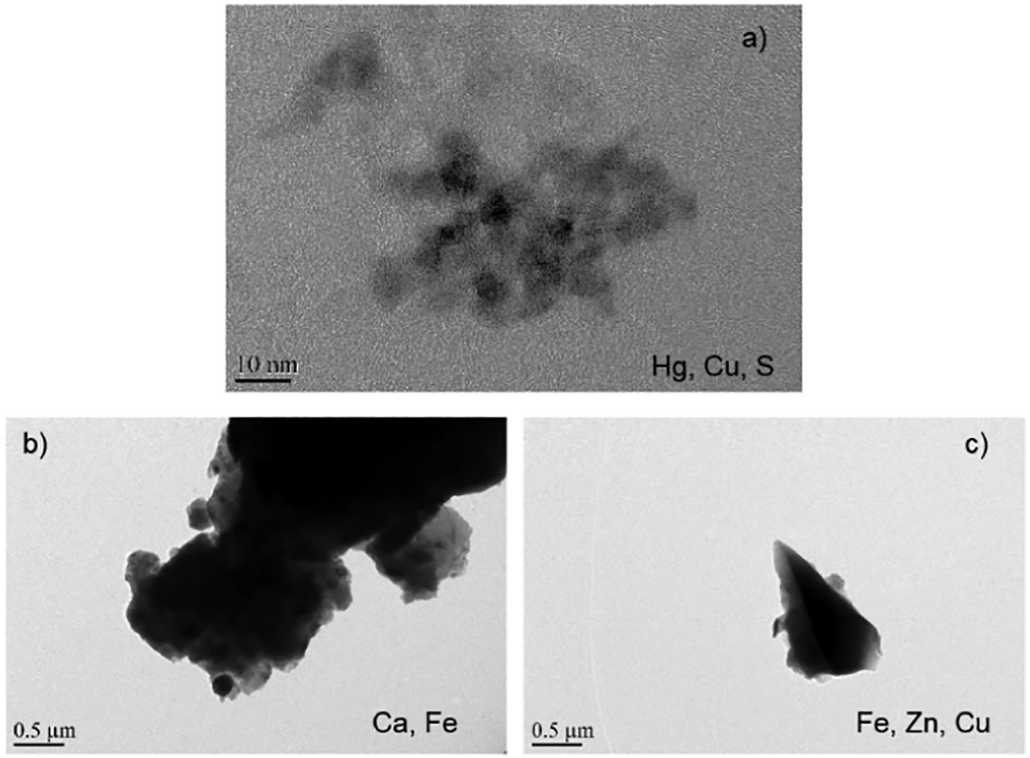

Fig. 3. Particles collected on TEM grids in the top floor (top) and in the ground floor (bottom).

the ground floor. For selected tracer metals, estimated inhaled doses were 17 and 4 ngTi/kg body weight, 0.3 and 0.1 ngCd/kg, 34.8 and 4.7 ngPb/kg, and 0.4 and 0.03 ngBi/kg, on the top and ground floors respectively.

The high concentrations described here emphasize that the main concern regarding e-waste handling is related to the toxicity of many of their components. This is why studies investigating on the concentrations of airborne polychlorinated biphenyls (PCBs), brominated flame retardants (BFRs) and heavy metals, together with their health implications, have sharply increased in recent years (Kim et al., 2019; Nti et al., 2020; Poole and Basu, 2017;Balasch et al., 2022) Focusing on heavy metals, the presence of high concentrations of Pb, Ni, Zn, Sb, Ti, Cr, Cu, Hg, Cd, Mn, As and Co in air has been frequently reported in literature (Table 2), with values exceeding those recorded at near urban environments by factors higher than 100. This agrees with factors obtained in this study for the different species analysed, ranging between 5 and 3000 for PM2.5, when comparing working areas with the urban background. Results in this study are also in line with the available information on material composition in different types of electrical and electronic equipment (Ghimire and Ariya, 2020), with releases from cathode ray tubes as one of the best-characterized sources (Ceballos et al., 2020; Lecler et al., 2015; Papaoikonomou et al., 2018), being mainly traced by Pb, Sr, Cd, Y and As, all of them were found in very high concentrations in this study (Table 4 and Fig. 5). The literature also reports that high exposures to heavy metals have even been detected at workers' homes, and also in samples of blood, urine and hair (Julander et al., 2014; Kuntawee et al., 2020; Wang et al., 2009). On the other hand, organic and elemental carbon (OC, EC) concentrations from WEEE facilities have been rarely reported in literature and mainly associated with uncontrolled burning activities, finding elevated concentrations, up to 382–563 µg/m^3 of OC and 26–42 µg/m^3 of EC (Bi et al., 2010; Ding et al., 2018), higher than those recorded in this study.

3.3. Inhalation exposure impacts

Inhalation exposure impacts were assessed in terms of cytotoxicity and generation of reactive oxygen species (ROS) on PM2 aerosols collected in

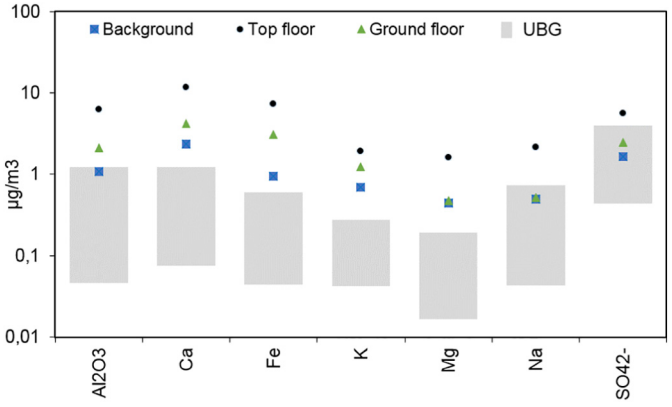

Fig. 4. Concentrations of major elements (µg/m^3) recorded at the different sampling sites (top floor, ground floor and BG) in comparison with the range of values at Palau Reial urban background (UBG). Note that values in the x axis correspond to a logarithmic scale.

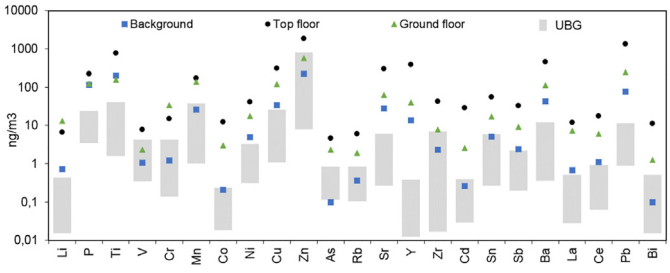

Fig. 5. Concentrations of trace elements (ng/m^3) recorded at the different sampling sites (top floor, ground floor and BG) in comparison with the range of values at Palau Reial urban background (UBG). Note that values in the x axis correspond to a logarithmic scale.

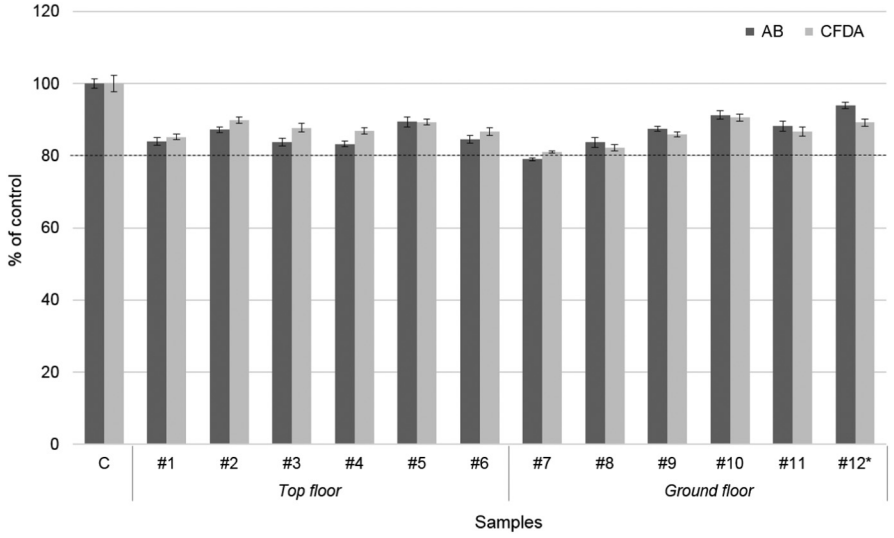

Fig. 6. Percentage of cell viability measured with Alamar Blue and CFDA-AM in A549 cells exposed to samples collected with Biosampler in both campaigns during 30 min. Results are expressed as percentage of control cells as mean ± SD of at least three different assays.

liquid suspension. Fig. 6 shows the cell viability of filtered samples (0.2 μm). The results were similar for filtered (<0.2 μm) and unfiltered (<2 μm) samples. None of the samples evidenced significant cytotoxicity using AB and CFDA; cell viability was always >80%.. The only sample close to the 80% threshold was sample #7, collected from the ground floor. Overall, these results did not evidence any strong cytotoxic potential for the samples analysed, but longer exposure times in more physiologically relevant models (He et al., 2021) might help to better characterize the toxic potential of these samples in future experiments.

Therefore, to understand the reactivity of the aerosols sampled when in contact with human lung cells, the samples were tested for oxidative stress generation. Clear differences were observed between ROS generation in the unfiltered (Fig. 7) and filtered (Fig. 8) samples. This assay measures fluorescence at different times (15, 30, 60 and 120) to detect the generation of reactive oxygen species (ROS). In the case of the filtered samples (i.e., samples containing only ultrafine particles, <0.2 μm) a significant induction of ROS was observed after only 15 min of exposure ROS generation The response obtained for the filtered samples (Fig. 8) was higher than for the unfiltered ones, confirming the larger hazard potential of ultrafine aerosols in comparison to fine particles (Oberdorster, 2000).

The samples with induced the highest levels of ROS (3-fold) generation were 1, 2 (both from the top floor; sampled in October 2020), 7 and 12 (ground and top floor, respectively; sampled in November 2020). Compared to other studies, this response in the range of that observed for bisphenol A, nonylphenol, and octylphenol in placenta cells (Pérez-Albaladejo et al., 2017). Lower response (1.5-fold) was detected for samples 9, 10 and 11. Samples 3, 4, 5, 6 and 8 (unfiltered) showed a response below control levels, suggesting an interference of the particles with the biological system. Results evidence an apparent absence of pattern in terms of ROS generation by samples collected from the top and ground floors, which limits the interpretation of the potential sources impacting aerosol reactivity. Thus, it was concluded that the variability of the WEEE entering the plant is probably the source of this heterogeneity in the particle reactivity assay. Different types of e-waste were processed each day in the facility (from gaming machines to fridges, televisions or toys), which meant that the type and intensity of the activities also varied from day to day. A larger number of samples, from a more repetitive process, would be necessary to understand the source of aerosol reactivity and to correlate it with others parameters such as size-resolved particle chemical composition.

In sum, it may be concluded that, even though exposure to ultrafine, fine and coarse aerosols from the different floors in the facility did not evidence cytotoxic effects, ROS generation was statistically significant for 4 out of the 12 samples collected, especially for ultrafine particles.

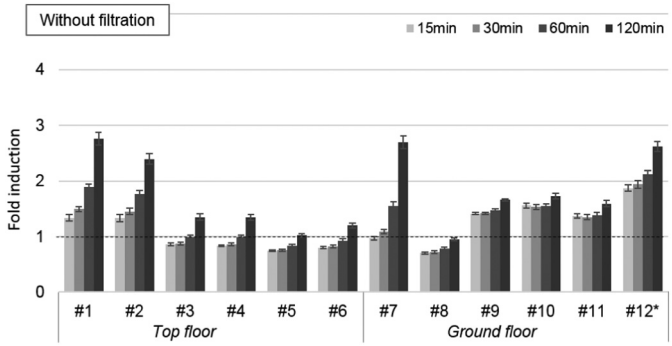

Fig. 7. ROS production in A549 cells after 15, 30, 60 and 120 min exposure to samples collected with Biosampler in both campaigns during 30 min. Values are expressed as fold induction as mean ± SD of at least six replicates in three different plates assayed. Dotted line represents ROS production in control cells. *Statistically significant differences from control.

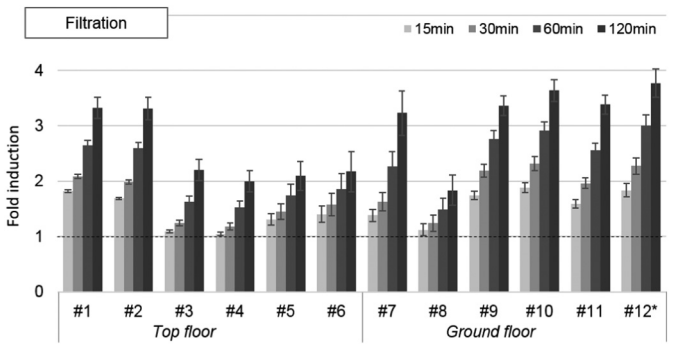

Fig. 8. ROS production in A549 cells after 15, 30, 60 and 120 min exposure to samples, filtered at <0.2 μm, collected with Biosampler in both campaigns during 30 min. Values are expressed as fold induction as mean ± SD of at least six replicates in three different plates assayed. Dotted line represents ROS production in control cells. *Statistically significant differences from control.

4. Conclusions

Waste Electrical and Electronic Equipment (WEEE) recycling generates indoor and outdoor emissions of fine and ultrafine particles, potentially hazardous for human health and the environment. In parallel to the global market, the environmental and health implications of this industrial sector are expected to grow exponentially in the coming years. This work characterized the exposure and environmental release of fine and ultrafine aerosols, with a focus on their metal content and potential for generation of reactive species when inhaled. The main conclusions extracted were:

- Monitoring of (PM10, PM2.5, BC, N and Dp) evidenced the release of fine and ultrafine particles from WEEE dismantling activities to outdoor and indoor air. Thus, e-waste dismantling activities, even if only manual or mechanical operations are involved (no thermal or chemical processes), have significant potential to impact exposure and air quality at local scale.
- Ventilation was a key driver of aerosol concentrations inside the facility, with highest concentrations in the top floor. Natural ventilation alone was not sufficient to maintain low levels of exposure to air pollutants on the ground floor.
- Ultrafine particle concentrations on the top floor were impacted by mechanical cutting of TV screen frames (metallic). High BC concentrations were also recorded on the top floor, and were attributed to the influence of the diesel forklifts operating on the ground floor.
- Key chemical tracers of the emissions generated by the facility were Ca (used plastic fillers) and Fe (from wiring and other metal components). Y, Zr, Cd, Pb, P and Bi were identified as tracers of cathode TV recycling, while Li and Cr were considered tracers of the grinding activities on the ground floor.
- The reactivity of the aerosols collected was assessed in terms of cytotoxicity and generation of reactive species. Ultrafine, fine and coarse aerosols did not evidence cytotoxic effects. ROS generation was statistically significant for 4 out of the 12 samples collected, especially for ultrafine particles. The variability in the WEEE being processed (largely variable from between days) limited the identification of significant trends. A larger number of samples, collected from more repetitive processes, would be necessary for this.
- The results from the facility under study, considered representative of EU standards, showed particle mass and number concentrations comparable (even if slightly lower) to those scarcely reported for other EU facilities. This data scarcity is especially notable for assessments of the metal components in WEEE, their sources and potential impacts on particle toxicity.
- Even though occupational exposure limits were not exceeded, particle concentrations were significant enough to be taken into consideration. Mitigation measures for nanoparticle emissions may be recommended for this specific WEEE recycling plant. First, source elimination by replacing diesel forklifts with electric ones, with proven high efficiency (Salmatonidis et al., 2019). Secondly, due to the high emissions in the top floor, the use of mechanical general ventilation systems would be recommended for exposure reduction in the TV dismantling area. Environmental release of aerosols could be addressed by filtering exhaust emissions and by improving the enclosure of the facility.

CRediT authorship contribution statement

M. López: Fieldwork; Formal analysis, Methodology, Data analysis, Writing - original draft

C. Reche: Methodology, Fieldwork, Data analysis

E. Pérez-Albaladejo: Methodology; review of initial draft

C. Porte: Conceptualization, Writing - review of initial draft

A. Balasch: Conceptualization, Writing - review of final draft

E. Monfort: Writing - review of initial draft

E. Eljarrat: Conceptualization, Writing - review of final draft

M. Viana: Conceptualization, Fieldwork; Formal analysis, Methodology, Writing - review of initial draft

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.154871.

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