



Air quality impacts of a large waste fire in Stockholm, Sweden

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HIGHLIGHTS

- Increased PM₁₀, PM_{2.5} and black carbon when wind comes from direction of waste fire.
- Elevated concentrations of particulate heavy metals and PAHs during open burning.
- Waste fires can have a strong impact on air quality of nearby residential areas.

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ABSTRACT

Fires in waste facilities are a common occurrence. Since many waste facilities are located adjacent to densely populated areas, these fires could potentially expose large populations to the emitted pollutants. However, at the moment there are only few field studies investigating the impact of waste fire emissions on air quality since the unpredictable nature of these events makes them challenging to capture.

This study investigated the impact of a large and persistent un-prescribed fire in a waste storage facility in Stockholm county, Sweden, on the local air quality of two residential areas in close proximity to the fire. In-situ measurements of particulate matter, black carbon and nitrogen oxide concentrations were conducted both during open burning and after the fire was fully covered. In addition, filter samples were collected for offline analysis of organic composition, metal content and toxicity.

Strongly increased concentrations of PM₁₀, PM_{2.5} and black carbon were found during the open burning period, especially when the wind was coming from the direction of the fire. In addition, elevated concentrations of particulate heavy metals and polycyclic aromatic hydrocarbons were observed in the air during the open burning period. These results show that waste fires can have a strong impact on the air quality of nearby residential areas.

1. Introduction

Fires in waste facilities can lead to the emission of different toxic pollutants into the air and subsequently into soil and water, potentially causing negative effects on the environment and human health. Studies

have for example shown increased dioxin and furan concentrations in air during a landfill fire (Weichenthal et al., 2015) and increased dioxin concentrations in food samples collected in the period following a landfill fire (Vassiliadou et al., 2009). A recent study estimates that about 2% of the total emissions of particulate matter with a diameter

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below 10 μm (PM_{10}) in Poland are caused by large waste fires (Bihalo-wicz et al., 2021). Fires in waste facilities are a relatively common occurrence: In the US, approximately 839 landfill fire incidents were estimated to have occurred over a seven year period from 2004 to 2010 (Powell et al., 2015). In Sweden, 143 fires in waste facilities were detected over a period of seven years (2012–2018), including 44 fires at landfills and 12 fires at sites for intermediate storage (Ibrahim, 2020). When it comes to the location of waste handling and storage sites, the external and social costs for communities in close proximity to such a site need to be balanced with the need to optimize waste transportation, plant construction and maintenance costs. Waste handling and storage sites are therefore usually located adjacent to densely populated areas (Ibrahim, 2020). This means that depending on the meteorological conditions, emissions from waste fires into the air can lead to the exposure of large populations. The emission of pollutants from waste fires into air depends on many different factors such as the composition and amount of fuel as well as the combustion conditions. A study of simulated scrap tyre fires for example showed that the emissions of various pollutants were affected by the burn rate (Lemieux and Ryan, 2012). There are currently only few field studies investigating the emissions of air pollutants from fires in waste facilities and their impact on air quality. One case study of a three-months long landfill fire in Iqaluit, Canada, found that daily median concentrations of particulate matter with a diameter below 2.5 μm ($\text{PM}_{2.5}$), nitrogen dioxide (NO_2), and ozone (O_3) were not dramatically impacted by the fire, although spikes in the hourly average $\text{PM}_{2.5}$ levels occurred when the wind was blowing from the direction of the fire. In contrast, levels of benzene and dioxins and furans were strongly increased (Weichenthal et al., 2015). Increased levels of dioxins and furans were also found in the air of landfills during an experimental and a spontaneous landfill fire in Finland (Ruokojärvi et al., 1995). The impact of a landfill fire on $\text{PM}_{2.5}$ concentrations in the metropolitan area of Santiago, Chile was investigated based on data from continuous monitoring stations and modelling of air parcel trajectories (Morales S et al., 2018). Results give an estimate of which parts of the population were exposed to particularly high pollutant levels. A more recent study concerning the same fire found that concentrations of particle-phase polybrominated diphenyl ethers (PBDEs) were enhanced by a factor 2–4 as compared to before the start of the fire (Pozo et al., 2023). However, a risk assessment suggested that there was no direct health risk due to PBDE exposure. A recent study of a landfill fire in Delhi based on data from continuous monitoring stations showed a strong influence of the fire on local air quality, with increases in both gaseous and particulate pollutants (Sharma et al., 2023). Another study measured the concentrations of suspended particulate matter and different inorganic gases during fires at five different municipal solid waste dumps in Nigeria. High concentrations for some pollutants such as PM, carbon dioxide (CO_2), carbon monoxide (CO) and methane (CH_4) were measured during the fires (Rim-Rukeh, 2014). Two additional studies focus on tyre fires. The first study evaluated the impact of a tyre fire in a municipal landfill in Iowa City, US, on particle composition and presents fuel-based emission factors for a variety of pollutants (Downard et al., 2015). The other study investigated the concentrations of polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and polychlorinated dibenzo-p-furans (PCDD/Fs), polychlorinated biphenyls (PCBs), and toxic metals in air, soil and crop samples collected after a tyre fire in a landfill in Seseña, Spain (Nadal et al., 2016). Most of the target pollutants did not show increased concentrations in the environment surrounding the fire, with airborne levels of PAHs being the notable exception. It should be emphasized here that these samples were collected post-fire, when PM levels had already decreased compared to the maxima observed during the fire. Due to the common occurrence of fires in waste facilities and the potential hazard they pose, more measurements of emissions from waste fires of different size, fuel composition and combustion conditions are needed to better understand the potential health impacts of this pollutant source.

In this study, we investigate the impact of a waste fire at an intermediate storage site in Kagghamra, Stockholm county, Sweden on the air quality of nearby residential areas. For this, in-situ measurements of PM, black carbon (BC) and nitrogen oxides (NO and NO_2) concentrations were conducted in two residential areas close to the fire both during open burning and after the fire was covered. In addition, filter samples were collected for offline analysis of organic composition, metal content and toxicity.

2. Methodology

2.1. Sampling sites and instrumentation

The fire described in this study occurred at a waste storage facility in Kagghamra, Stockholm county, Sweden ($59^\circ 05' 53.3''\text{N}$ $17^\circ 47' 19.0''\text{E}$). The facility was originally classified as an intermediate storage site for up to 10,000 tons of demolition wood as well as wood and metal construction waste. However, over the years multiple complaints were filed against the business regarding violations of the permitted storage duration and the quantity and type of the stored waste. The exact nature and amount of fuel for the waste fire are therefore not known, but pictures of the site revealed the presence of gypsum, metal, large amounts of various types of wood and different plastics. According to the responsible fire brigade, the waste heap covered an area of 170×70 m with a height of 5–10 m at the time of the fire. The fire described in this article started on Dec 23rd 2020. The site of the fire is located close to a groundwater body as well as a fjord and a river. It was therefore deemed that the fire could not be extinguished with water due to the risk of contaminating the surrounding area. Therefore, the decision was made to extinguish the fire by covering it with sand. Work to cover the fire began on Jan 26th 2021. A first thin layer of sand was in place by Feb 10th 2021, vastly reducing the amount of emitted smoke. This marks the end of the open burning period. By the 16th of February, the extinguishing efforts were in their final phase and the sand cover was completed on Feb 19th, marking the end of the extinguishing period and the transition to the fully covered period starting from Feb 20th.

Two measurement sites were set up at about 900 m south-southwest of the fire (Söderängstorp) and 1000 m east-northeast of the fire (Tegelvreten) to capture potential population exposure, see Fig. 1. Initial measurements of $\text{PM}_{2.5}$ and PM_{10} were conducted at the Söderängstorp site between Jan 8th 2021 and Jan 10th 2021 using Lighthouse Handheld 3016 IAQ particle counters. For Jan 8th, the time resolution was 10 s, while for Jan 9th and 10th a resolution of 5 min was used. In this paper we present hourly averages using the end time as time stamp. Continuous monitoring of $\text{PM}_{2.5}$, PM_{10} , NO_2 , NO and BC was started on Jan 22nd 2021 at Söderängstorp and on Jan 25th 2021 at Tegelvreten, with measurements at both sites lasting until the early morning on May 3rd. Grimm EDM 180 PM monitors were used to measure $\text{PM}_{2.5}$ and PM_{10} , while NO_2 and NO were determined using chemiluminescent analysers (Environment S.A, model AC31M) and BC was measured using Magee Scientific aethalometers (model AE33). All online measurements had a time resolution of 15 min. Data in this paper are presented as hourly averages using the end time as time stamp. A sonic anemometer, located at Söderängstorp, was used to determine wind direction and speed. In addition to the continuous measurements, different types of PM_{10} filter samples were collected at both measurement sites. Leckel SEQ 47/50 low volume samplers were used to collect 24-h PM_{10} samples on 47 mm diameter Teflon filters (Zeflour, Pall laboratory) with a flow rate of 16.67 L/min, and sample collection starting at midnight every day. In total, 177 low-volume filters were collected (88 from Söderängstorp and 89 from Tegelvreten) between Jan 28th 2021 and April 26th 2021. In addition, 14 Teflon field blanks were collected. Selected filters from this group were used for measuring the concentrations of particulate PAHs, metals and total extractable organofluorine (TEOF). High-volume 24-h PM_{10} samples were collected at a set flow rate of 68 m^3/h between Feb 8th 2021 and Feb 17th 2021. Samples at each site were alternately

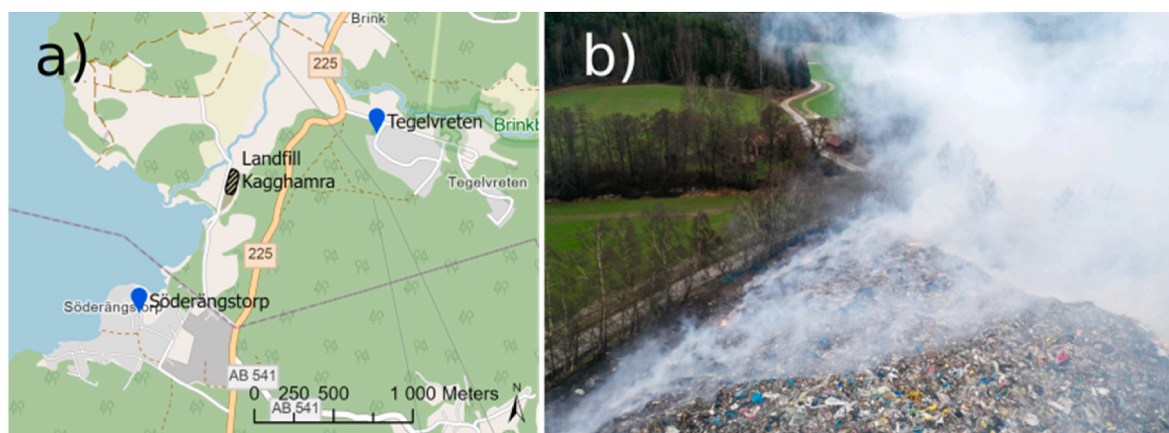


Fig. 1. (a) Location of the fire and the two stationary measurement sites Söderängstorp and Tegelvreten and (b) view of the fire (photo: Piritta Lutz).

collected on QM-A quartz fibre filters (QFF, Whatman, 203×254 mm) and Teflon-coated glass fibre filters (Pall T60A20, 8×10 in, Pallflex Corporation, Putnam, CT, USA). In total, 12 high volume filters were collected: 6 QFF (3 at each site, QFF 001–006) and 6 Teflon-coated glass fibre filters (4 at Söderängstorp and 2 at Tegelvreten, TC 001–006). In addition, 8 field blanks were collected: 2 QFF and 2 Teflon-coated glass fibre filters at each site. Due to large uncertainties with the flow rate determination, the samples collected with the high-volume sampler were only analysed qualitatively with the exception of the toxicity measurements, where the dose of particles used for exposure could be determined from the weight of the filter before and after particle removal. Half of each QFF sample was used for the analysis of per- and polyfluoroalkyl substances (PFAS) while the other half was used for non-target analysis. The Teflon-coated glass fibre filters were divided into subsections. Sections of all six Teflon-coated glass fibre filter samples were used for the extended analysis of PAHs and their derivatives, while only the filter most strongly impacted by the fire was used for determining the elemental composition of individual particles and for toxicity analysis. Finally, measurements of VOCs were performed on the morning of Feb 1st at both Tegelvreten (10 min) and Söderängstorp (20 min) using a proton-transfer-reaction time of flight mass spectrometry (PTR-ToF-MS) instrument (model PTR-TOF 1000 QB2, Ionicon Analytik) installed on the University of Oslo mobile laboratory (drift tube operating conditions: $p = 2.4$ mbar, $T = 80^\circ\text{C}$, $E = 360$ V $\Rightarrow E/N \sim 100$ Td). The PTR-MS instrument was calibrated using a dynamically diluted VOC standard (Apel-Riemer Environmental Inc., Miami, USA). For VOCs not present in the standard, calibration factors reported by Koss et al. (2018) were used. The PTR-MS raw data were analysed using the Ionicon Data Analyzer software (v1.0.0.0, Ionicon Analytik). The automated data analysis detected 131 mass spectrometric signals with enhanced intensities in fire-impacted air masses. Only compounds with concentrations $>1 \mu\text{g}/\text{m}^3$ (as measured during a mobile survey in Tegelvreten) were included in the further analysis. An overview of the instruments used in this campaign and the different analyses can be found in Table S1 of the Supporting Information (SI).

2.2. Filter analyses

A variety of filter-based analysis techniques were used to investigate how the composition of PM_{10} was impacted by the fire.

2.2.1. Heavy metals

PM_{10} samples collected on low-volume Teflon filters were used to investigate the presence of heavy metals in the particles emitted from the fire. In total, 18 filters were analysed: 8 filters from the open burning period (4 from Söderängstorp and Tegelvreten each), 8 filters from the fully covered period (4 from Söderängstorp and Tegelvreten each) and 2

field blanks (one per site). The filters from the fully covered period were pooled pairwise due to the low particle mass collected. All samples were analysed for nickel (Ni), cadmium (Cd), copper (Cu), vanadium (V), arsenic (As), lead (Pb), cobalt (Co), chromium (Cr), manganese (Mn) and zinc (Zn). Five of the samples from the open burning period were additionally analysed for mercury (Hg). For the analysis, the filters were first digested by microwave digestion in nitric acid and the samples then diluted before analysis with inductively coupled plasma quadrupole mass spectrometry (ICP-QMS). All stated values are blank subtracted. Values below the limit of quantification (LOQ, determined as 3 times the limit of detection) were treated as 0 for the calculation of summary statistics. Raw data of the measured concentrations of the different metals in solution before blank subtraction and the respective limits of detection can be found in Table S2 of the SI.

2.2.2. Elemental composition of individual particles

A JEOL 7000 F Scanning Electron microscope (SEM, 15 kV) equipped with an Energy Dispersive X-ray Spectrometer (EDS) was used for imaging and mapping of the elemental distribution in a high-volume Teflon-coated glass fibre filter from the open burning period which was strongly impacted by the fire (TC 001) and a matching field blank. The element analysis was evaluated by use of INCA program package from Oxford Instruments.

2.2.3. Non-target screening

For non-target screening, sections of all collected high-volume PM_{10} QFF were extracted following the extraction protocol for polar organic compounds from Papazian et al. (2022). In total, six filters were analysed: one filter from the open burning period which was strongly impacted by the fire (QFF 001) and five filters from the final extinguishing period. For those five filters from the extinguishing period (QFF 002–006), the wind largely did not come from the direction of the fire. Those samples will therefore be considered as samples of background air. All the resulting extracts were analysed by ultra-high-pressure liquid chromatography (UHPLC, Ultimate 3000) coupled to a High-Resolution Mass Spectrometer (HRMS) Q Exactive Orbitrap HF-X (ThermoFisher Scientific, Waltham, MA, USA) with electrospray ionization (ESI). As quality control and to enable the calculation of Retention Time Index (RTI) for the detected ions, mixtures of analytical standards were injected as described by Aalizadeh et al. (2021). By reporting the retention times of the standards, RTIs were calculated for the selected features in the samples using the website <http://rti.chem.uoa.gr/>. For each detected feature, RTI, precursor mass (m/z), and fragmentation data (mass spectrum) were collected and run through the high-resolution spectral library MassBank Europe (<https://massbank.eu/MassBank/>). Features with a similarity score higher than 70%, and a $\Delta m/z$ lower or equal to 5 ppm compared to reference spectra from the

library were classified as candidates. Predicted RTI of the candidates was retrieved from the NORMAN Substance Database (<https://www.norman-network.com/nds/susdat/index.php>). MGF files of the detected peaks were extracted from MS-DIAL and imported into SIRIUS 5.5.3 for molecular prediction (Dührkop et al., 2019) in both ionization modes. In the software, the mass accuracy was set to 5 ppm and the database used to retrieve possible formula was set as NORMAN. An example of a chromatographic feature with a spectral match and an assigned molecular formula is given in Fig. S1. Additional details regarding the extraction and analysis of the samples can be found in the SI.

2.2.4. EPA's 16 priority PAHs

The concentrations of the 16 priority PAHs as selected by the United States Environmental Protection Agency (EPA) (acenaphthene, acenaphthylene, anthracene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene, chrysene, dibenz[a,h]anthracene, fluoranthene, fluorene, indeno[1,2,3-cd]pyrene, naphthalene, phenanthrene and pyrene) as part of PM₁₀ in the air were determined according to the Swedish standard SS-EN 15549:2008. In brief, PM₁₀ low-volume Teflon filters (47 mm diameter) were first extracted in acetone using Soxhlet extraction. The samples were then diluted with water and transferred into pentane. For clean-up, the PAH extract was fractionated on a silica column and the solvent was changed to methanol. As a final step, the samples were analysed using high-performance liquid chromatography with fluorescence detection (HPLC-FLD). A total of 10 samples were analysed: 4 filters from the open burning period and the fully covered period each and two blanks. The four filters with the highest PM₁₀ mass were selected for the open burning period. Those PAH concentrations therefore present an upper limit. For the open burning period, each sample represents a single day while for the fully covered period 24 to 29 filters were pooled per sample due to low concentrations. One blank sample contained only one filter (used for blank subtraction of the samples from the open burning period) and for the blank of the fully covered period 9 filters were pooled. All stated values are blank subtracted. For blank subtraction of the pooled samples from the fully covered period, the concentrations in the pooled blank were divided by 9 and then multiplied by the number of pooled sample filters to account for the differing numbers of filters used for pooling. Values below the limit of quantification (LOQ, determined as 3 times the limit of detection) were treated as 0 for the calculation of summary statistics. Raw data of the measured concentrations of the different PAHs before blank subtraction and the respective limits of detection can be found in Table S3.

2.2.5. Extended analysis of PAHs and OPAH derivatives

From each of the 6 high-volume Teflon-coated glass fibre filters (TC 001–006) three $\phi = 30$ mm filter punches were sampled. The filter punches were inserted into 5 mL stainless steel accelerated solvent extraction (ASE) cells followed by addition of internal surrogate standards. The cells were then extracted by pressurized fluid extraction (ASE 200, Dionex Corporation, Sunnyvale, CA, USA). Toluene (HPLC grade, Rathburn, Walkersburn, UK) was used as extraction solvent and the extraction temperature and pressure was 200°C and 1500 psi (10.3 MPa), respectively. Five static extraction cycles with 5 min each were used.

The resulting toluene extracts were evaporated to 0.5 mL under a gentle stream of nitrogen gas and heating (55°C), followed by solid phase extraction (SPE) on 100 mg silica Isolute IST SPE cartridges (Biotage AB, Uppsala, Sweden). The analytes were eluted with 2 mL toluene followed by evaporation to 0.3 mL and vialling in micro-injection vials.

Instrumental analysis of PAHs and OPAHs was carried out on a coupled High-Pressure Liquid Chromatography/Gas Chromatography/Mass Spectrometer (HPLC/GC/MS)-system described in detail elsewhere (Sadiktsis et al., 2023). Briefly, 50 μ L sample was injected into the

HPLC/GC/MS-system which isolates the analytes by means of backflush HPLC (Östman and Colmsjö, 1987) followed by sample introduction into the injector port (a Gerstel® Cis-3 programmable temperature vaporizer) of the GC. GC oven program, zone temperatures and MS parameters are available in Table S4 of the SI. Analyte specific selected ion monitoring (SIM) ions are all listed in Table S5 of the SI.

Quantitation was made using relative response factors between each corresponding surrogate standard and analyte (listed in Table S5 of the SI) and using single point calibration.

2.2.6. PFAS

The other halves of the same six 203 \times 254 mm QM-A QFF mentioned in Section 2.2.3 were analysed for the presence and concentration of 29 Per- and polyfluoroalkyl substances (PFAS). Before extraction, the filters were spiked with 2000 pg of ¹³C-labelled internal standard containing a mixture of PFAS (see Table S6 in the SI). The filters were subsequently extracted following the same method as described by Sha et al. (2022).

Analysis was performed on a Thermo Fisher Q-Exactive HF Orbitrap (Thermo Fisher Scientific, Waltham, MA, USA) LC-HRMS system, equipped with a 1.7 μ m, 50 \times 2.1 mm Waters Acquity BEH C18 column and a Waters Acquity BEH C18 guard. Furthermore, the mobile phase was led through a Waters PFC isolator column after mixing. The mobile phases consisted of A: water:acetonitrile (H₂O:ACN, 95:5) and B: ACN: H₂O (95:5) with 2 mM ammonium bicarbonate.

Instrumental limits of quantification (LOQs) and detection (LODs) of the analytes were determined by calculating the difference between calibration sample concentrations and the values of the calibration curve for the lowest three detected calibration standards. The standard error ($\frac{s}{\sqrt{N}}$) of these differences was then divided by the slope of the calibration curve and multiplied by 10 and 3.3 for LOQ and LOD, respectively. If the analyte was present in the field blanks, the LOQs and LODs were calculated by multiplying the standard deviation of the blank response by 10 and 3 respectively and dividing by the slope of the calibration curve. These approaches were chosen because common approaches of quantification limit determination using signal-to-noise ratios are not very suitable for HRMS analyses due to the very low or absent noise levels due to high mass accuracy and noise removal algorithms of these instruments. An overview of all LODs and LOQs can be found in Tables S7–S10 of the SI.

2.2.7. Total extractable organofluorine

Eight 47 mm Teflon filters which remained after the analysis of PAHs and heavy metals were extracted via ultrasonication in 3 \times 10 mL methanol. The extracts were combined and evaporated to 0.5 mL under a gentle stream of nitrogen. Two laboratory blanks were prepared from unused filters treated in the same way as samples.

Measurements of TEOF were carried out using a Thermo-Mitsubishi CIC. Extracts (100 μ L) were placed in a ceramic sample boat containing glass wool and combusted in a horizontal furnace (HF-210, Mitsubishi) at 1100°C for approximately 5 min, under a flow of oxygen (400 mL/min) and argon mixed with water vapor (200 mL/min). Combustion gases were absorbed in MilliQ water using a gas absorber unit (GA-210, Mitsubishi). A 200 μ L aliquot of the absorption solution was injected onto an ion chromatograph (Dionex IntegriON HPIC, Thermo Fisher Scientific) equipped with a 2 \times 50 mm guard column (Dionex IonPac AS19-4 μ m) and 2 \times 250 mm analytical column (Dionex IonPac AS19-4 μ m) operated at 35°C. Chromatographic separation was achieved by running a gradient of aqueous hydroxide mobile phase ramping from 8 mM to 60 mM at a flow rate of 0.25 mL/min (Table S11). Fluoride was detected using a conductivity detector.

The concentration of total extractable organofluorine was quantified using a nine-point calibration curve ranging from 0 to 10 μ g/mL fluoride. Quality control samples containing 3.0 μ g/mL organofluorine and 5.6 μ g/mL fluoride were analysed alongside the samples. The average

accuracy was 94% and the relative standard deviation 6%. The laboratory blanks were in line with instrumental blanks. The LOD and LOQ were therefore calculated as 3 and 10 times the standard deviation of the response of both instrumental and laboratory blanks. Normalised to the same sample volume as the samples, the LOD and LOQ were 4.3 and 14 ng/m³, respectively.

2.2.8. Toxicity testing

THP-1 cells were cultured as previously described (McCarrick et al., 2021) and differentiated to macrophages (dTHP-1) using phorbol 12-myristate 13-acetate (50 ng/mL) in a 96 well plate for 48 h. A subsection of the open fire impacted Teflon-coated glass fibre filter (TC 001) was used for toxicity testing. To remove the particles from the filter, approximately 4 × 4 cm filter pieces were cut, weighed, and transferred to 50 mL glass bottles (Schott bottles). Next, 5 mL ethanol was added on to the filter pieces and sonication in a water bath was performed for 20 min at 30 °C. The bottles were left open in the fume hood to evaporate ethanol, until ~300 µL was left. A stock concentration of 1 mg/mL was then prepared in RPMI medium and was sonicated again for 20 min before exposure. Finally, different exposure concentrations were prepared (5, 10, 25, 50 and 100 µg/mL) from the stock and fresh suspensions were prepared for each experiment. Cytotoxicity and cytokine release was determined by using Alamar blue assay and MSD® multi-spot assay, respectively, as previously described (McCarrick et al., 2021) with a slight difference in the number of cells seeded (55 × 10⁴ cells/well). For cytokine analysis, supernatants were diluted in RPMI, followed by diluent buffer to achieve final dilutions (30 times for exposures, and 50 times for lipopolysaccharide (LPS)).

3. Results and discussion

3.1. Concentrations and composition

Concentrations of PM₁₀ and PM_{2.5}, BC, particulate metals, PFAS and PAHs as well as NO₂, NO and VOCs in air were measured at two sites, Söderängstorp and Tegelvreten, in close proximity to a waste heap fire in Kagghamra, Stockholm County, Sweden. In addition to the quantitative results, the composition of PM₁₀ was investigated further via single-

particle analysis regarding elemental composition, an extended qualitative analysis of PAHs and their oxygenated derivatives as well as non-target screening.

3.1.1. Concentrations of PM and BC

Concentrations of PM₁₀, PM_{2.5} and BC were measured during three time periods: open burning, extinguishing, and fully covered (Table 1) at Söderängstorp and Tegelvreten. Events of co-occurring high concentrations of all three pollutants were seen at the same time during the open burning period (Fig. 2 and Fig. S2). These peaks coincided with a wind direction towards the measurement sites, i.e. it is clear that the elevated concentrations originated from the fire. This effect can be seen from wind roses in Fig. 3 showing PM_{2.5} as a function of wind direction at Söderängstorp (see Fig. S3 for the corresponding plots from the Tegelvreten site). During the extinguishing period, particle mass and BC concentrations decreased drastically, and decreased even further when the fire was fully covered. For example, the daily mean PM_{2.5} concentration at Söderängstorp was 38.0 µg/m³ during the open fire period, and went down to 12.7 and 5.4 µg/m³ during extinguishing and fully covered period, respectively. Comparing the 2 PM fractions shows that PM_{2.5} is the major contributor to PM₁₀, i.e. the mass of fine particles in the air was dominating. At both Söderängstorp and Tegelvreten, fine particles contributed to approximately 90% of the particle mass concentration of PM₁₀ during the open fire and the extinguishing period. All PM concentrations measured at Tegelvreten were lower in comparison to those at Söderängstorp. This is mainly a result of the dominating wind directions during the open burning period.

Swedish environmental quality standards (EQS) for 24-h mean PM₁₀ (SFS, 2010:477) which match with the European Union air quality standards (EU AQS) (Directive, 2008/50/EC) were not exceeded during the measurement period (50 µg/m³ maximum 35 days/year). This value was exceeded on 6 days at Söderängstorp and 4 days at Tegelvreten during the measurement in the open fire period (approx. 3 weeks). However, the fire started approx. 1 month before the continuous measurements began and the brief initial measurements at Söderängstorp in early January showed PM₁₀ concentrations above the levels detected during the continuous measurements. Hence, the total number of exceedances during the open burning period could not be confirmed.

Table 1

Daily concentrations from on-line measurements of different pollutants. Data coverage for the different pollutants during the three measured time periods ranged from 80 to 100% at Söderängstorp and 90–100% at Tegelvreten. The initial measurements of PM_{2.5} and PM₁₀ at Söderängstorp in early January are excluded from this table.

Pollutant	Waste fire status					
	Open burning		Extinguishing period		Fully covered	
	Mean (range)	Median	Mean (range)	Median	Mean (range)	Median
Söderängstorp (22nd Jan 2021–3rd May 2021)						
PM ₁₀ (µg/m ³)	43 (1.7–140)	23	14 (3.7–48)	6.7	7.8 (1.5–43)	5.6
PM _{2.5} (µg/m ³)	38 (1.6–130)	21	13 (3.3–42)	6.2	5.4 (0.8–39)	3.5
BC (µg/m ³)	1.3 (0.16–7.5)	0.95	0.49 (0.13–1.8)	0.22	0.21 (0.026–1.4)	0.16
NO ₂ (µg/m ³)	8.1 (3.2–21)	6.1	6.0 (1.2–19)	4.1	2.6 (0.7–5.7)	2.6
NO (µg/m ³)	1.8 (0.7–6.6)	1.4	1.2 (0.3–4.1)	0.8	0.4 (0.1–0.9)	0.3
Tegelvreten (25th Jan 2021–3rd May 2021)						
PM ₁₀ (µg/m ³)	30 (1.8–130)	6.7	6.9 (2.6–29)	4.8	6.1 (1.3–38)	4.6
PM _{2.5} (µg/m ³)	26 (1.7–110)	6.3	6.4 (2.3–28)	4.2	4.6 (0.8–36)	3.1
BC (µg/m ³)	0.74 (0.12–2.4)	0.34	0.50 (0.25–1.7)	0.38	0.22 (0.044–1.5)	0.15
NO ₂ (µg/m ³)	3.7 (1.3–6.7)	3.8	3.0 (0.3–9.5)	2.5	1.2 (0.2–4.1)	1.1
NO (µg/m ³)	0.8 (0.2–2.5)	0.5	0.8 (0.3–1.7)	0.5	0.6 (0.2–1.2)	0.6

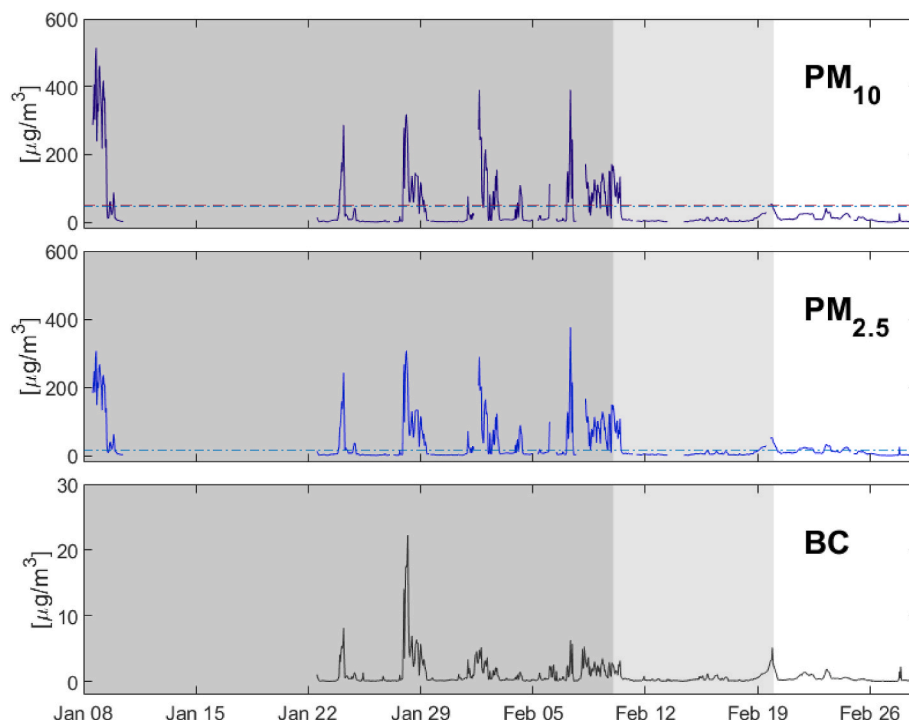


Fig. 2. Timeseries of hourly averages of PM_{10} , $PM_{2.5}$ and BC at the Söderängstorp measurement site. Dark grey, light grey and white mark the open burning period, final extinguishing period and fully covered period (only partially shown), respectively. The dashed red line marks the limit of the 24 h average PM_{10} according to the Swedish environmental quality standards for outdoor air ($50 \mu\text{g}/\text{m}^3$ for a maximum of 35 days/year), while the turquoise dash-dotted lines indicates the recommend levels of PM_{10} and $PM_{2.5}$ over 24 h according to the WHO 2021 air quality guidelines (45 and $15 \mu\text{g}/\text{m}^3$ for a maximum of 3–4 days/year, respectively).

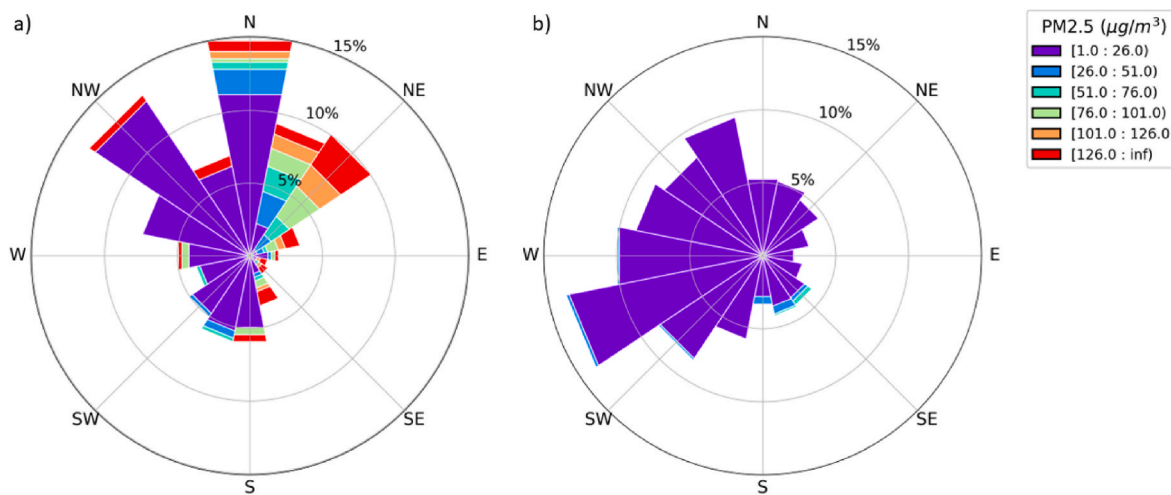


Fig. 3. Wind roses showing $PM_{2.5}$ depending on wind direction for the measurement station at Söderängstorp during the open burning period (a) vs. the fully covered period (b). The fire is located north-northeast (N-NE) of Söderängstorp. The initial measurements of $PM_{2.5}$ and PM_{10} at Söderängstorp in early January are excluded from this figure.

The World Health Organisation recommends stricter global air quality guidelines (WHO AQG) for PM concentrations (24-h mean of $45 \mu\text{g}/\text{m}^3$ for PM_{10} , $15 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ for max. 3–4 days) (WHO, 2021). The WHO AQG for PM_{10} and $PM_{2.5}$ were exceeded at the measurement sites during the Kagghamra waste heap fire in 2021.

Two other studies have also measured PM_{10} and $PM_{2.5}$ concentrations during fires at waste storage sites. In Iquluit, Canada, daily mean $PM_{2.5}$ was similar during active burning, extinguishing and when extinguished (4.61 , 2.23 , and $3.55 \mu\text{g}/\text{m}^3$) (Weichenthal et al., 2015) at four sites located 1.3 – 3.8 km away from the fire (compared to 0.9 – 1 km in our study). These concentrations are at the same or lower level

compared to the daily mean $PM_{2.5}$ during the covered period in our study. However, their study points out that the wind predominantly carried the emissions away from the measurement sites during the fire and that short-term spikes in concentration were observed when the wind was blowing towards the measurement site. Burning of a landfill in Iowa city, US, caused noticeable spikes in $PM_{2.5}$ at locations 3.2 , 4.2 and 10.5 km away from the fire. Hourly averages stayed below $80 \mu\text{g}/\text{m}^3$ (Downard et al., 2015) and were therefore much lower than the highest hourly averages observed in our study (Fig. 2). Elemental carbon concentrations at the Iowa city landfill fire ($>0.45 \mu\text{g}/\text{m}^3$) were lower than the BC concentration at open burning in our study.

3.1.2. Heavy metals

The concentrations of 11 heavy metals in air as components of PM₁₀ were determined during the open burning vs. the fully covered period at Söderängstorp and Tegelvreten (Table 2). During the open burning, the highest metal concentrations were found for Cu, Pb and Zn (in the range approx. 200–450 ng/m³). It should be noted that high concentrations of Cr and Zn were found in the blanks, leading to larger uncertainty regarding their concentrations. High concentrations of metals present in the air coincided with high PM concentrations. Most of the metals (Cd, As, Cr, and Mn) were found at concentrations <6 ng/m³. Concentrations for V and Co were even lower (<0.4 ng/m³). The concentrations of Ni and Hg were below the LOQ in all samples. In Sweden and in the EU, 4 of the heavy metals are regulated based on annual averages; EQS and EU AQS: Pb (500 ng/m³), Cd (5 ng/m³), As (6 ng/m³), Ni (20 ng/m³) (SFS, 2010:477; Directive, 2004/107/EC). The concentrations of these four metals measured at the sites next to the fire were compared with those at a regional background site (Norunda, Uppsala, Sweden 2018–2020) (SMHI). All regulated heavy metals, apart from Ni, were found at greatly elevated concentrations during days with open burning of the fire and intense smoke at Kagghamra compared to normal concentrations in regional background: 392 ng/m³ Pb (background: 0.70 ng/m³), 3.5 ng/m³ Cd (background: 0.02 ng/m³), 4.1 ng/m³ As (background: 0.13 ng/m³). After the coverage of the fire, the concentrations of all metals in the air were low. Concentrations of the regulated metals (Pb, Cd and As) were very close to those of the regional background in Sweden.

Some of the metal concentrations fluctuated greatly from day to day (Pb, Cu and Cd, and to some degree also As and Zn), which indicates that these metals were present in the burning waste (see the min–max range in Table 2). The fluctuation is most probably an effect of the changing wind directions, i.e. the filter samples were exposed to more particles and metals if the wind was blowing towards the measurement stations. For example, the maximum concentration of Pb was 1000 times higher than the minimum concentration measured during the open burning period. The concentration of the other measured metals (Co, Mn, V, Zn, Cr) did not vary as much between samples with and without a high load of particles from the fire. This may indicate that these metals did not, or

only partly originated from the fire.

Other studies have also measured metal concentrations in PM₁₀ during or after fires at waste storage sites. During a landfill fire in Iowa city, US, metal concentrations in PM₁₀ were 29 ng/m³ (Zn) and up to 4 ng/m³ (Pb) (Downard et al., 2015), which is much lower compared with those measured during the open fire period in our study. After the extinction of a landfill fire in Seseña, Spain, similar concentrations of 4 metals (As, Cd, Ni, Pb in PM₁₀) were measured irrespective of the distance to the landfill (0.5 and 4 km) (Nadal et al., 2016). Closest to the landfill, the metal concentrations in air were approximately 0.9 ng/m³ (As), 0.2 ng/m³ (Cd), 4 ng/m³ (Ni), and 5 ng/m³ (Pb) after the extinction, which is higher compared to those measured at the fully covered period in our study.

3.1.3. Elemental composition of individual particles

Measurement of the elemental composition of individual particles on a PM₁₀ filter sample impacted by the fire showed that the particles for example contained a lot of carbonaceous material, as expected at a fire, and also several different metals. A selection of the elemental mappings is shown in Fig. 4. Many particles contained Pb, in agreement with the heavy metal analyses in our study. As seen from the mapping, Pb often co-existed with Cl. Fe and a small amount of Zn, possibly existing as oxides, were present in some of the particles. Furthermore, potassium chloride (KCl) salt particles were found in the samples as concluded by the co-presence of K and Cl. Potassium chloride is commonly present in biomass burning particles and in particular in fresh emissions (Pósfai et al., 2003; Zauscher et al., 2013), and were therefore also expected in the samples impacted by the waste fire. A low amount of several other metals were also detected in the samples; Co, Ge, Mn, Ti (<1 wt%).

Blank filters mainly contained Si, O, C and F, in agreement with the used filter material of Teflon-coated (polytetrafluoroethylene) glass fibres. A low amount of Al, Ba, Ca, Mg, Na (<1 wt%) was also present in the blank filters. Hence, all these elements were also present in the analyses of the samples impacted by the fire, and could potentially therefore also be present in the collected particles. A lot of additional C was found in the collected particles, which is most likely carbonaceous particles from the fire. An SEM image of collected particles versus a blank filter can be found in Fig. S4.

3.1.4. Nontarget screening of PM₁₀

From the nontarget analysis, 7621 peaks were detected only in the filter impacted by the fire compared to the filters from the extinguishing period (3157 in positive ionization and 4464 in negative ionization mode). Twenty-nine of these had a >70% spectral match in MassBank Europe and a RTI difference of 30 or less compared to the RTI estimated for the candidate (retrieved from the NORMAN Substance Database). Nine additional peaks also have a good spectral match, but no RTI information for the corresponding candidate. However, based on the collected information (MS2 spectral match and precursor mass) they can still be considered as credible candidates. All of these candidates are identified at Level 2 in the Schymanski scale (Schymanski et al., 2014) and would require analytical confirmation with a reference standard to insure of their presence in the samples. Nevertheless, it is worth noticing that 6 of these candidates are classified as dangerous according to the Globally Harmonized System (GHS) classification of hazards (either due to their acute toxicity or their carcinogenic toxicity). These compounds are: 4-methoxyaniline (used in the packing of food), N-(2,4-dimethylphenyl)formamide (transformation product of the pesticide Amitraz, which is used in Europe and registered in the EU Pesticide database as an active substance (ECHA a)), di(2-ethylhexyl)phthalate (plasticizer), 3-ethylphenol (used as a plastic additive and in personal care products), 4-nitrophenol (industrial compound classified as persistent, mobile and toxic (Serrà et al., 2020)) which can also be formed via biomass burning (Mohr et al., 2013), and ethylparaben (a plastic additive). Details of these spectral matches are given in Table S12 of the SI.

641 features had a molecular formula assigned using SIRIUS (252

Table 2

Concentrations of analysed metals in PM₁₀ in the air during open burning and the fully covered period (results are a combination of data from Söderängstorp and Tegelvreten). Concentrations of nickel and mercury were below the limit of quantification (LOQ) for all samples and are therefore not listed. Samples below the LOQ were taken as 0 for the calculation of the summary statistics.

Pollutant (ng/m ³)	Open burning		Fully covered	
	Mean (range)	Median	Mean (range)	Median
Cd	3.5 (0.037–7.5)	2.8	0.026 (<LOQ–0.056) ^c	0.024
Cu	200 (2.1–630)	140	3.1 (1.5–4.6)	3.2
V	0.36 (<LOQ–0.92) ^a	0.40	0.50 (0.31–0.86)	0.42
As	4.1 (0.25–10)	3.3	0.19 (<LOQ–0.31) ^c	0.23
Pb	390 (1.0–1000)	310	0.77 (<LOQ–1.7) ^c	0.69
Co	0.054 (<LOQ–0.14) ^b	0.035	0.049 (0.033–0.071)	0.046
Cr	1.4 (0.83–2.9)	1.1	0.11 (<LOQ–0.42) ^d	0.00
Mn	5.3 (0.71–12)	4.6	1.8 (1.2–2.3)	1.9
Zn	450 (17–1300)	330	10 (<LOQ–23) ^e	9

^a 2 out of 8 samples below limit of quantification (2/8).

^b 4/8.

^c 1/4.

^d 3/4.

^e 2/4.

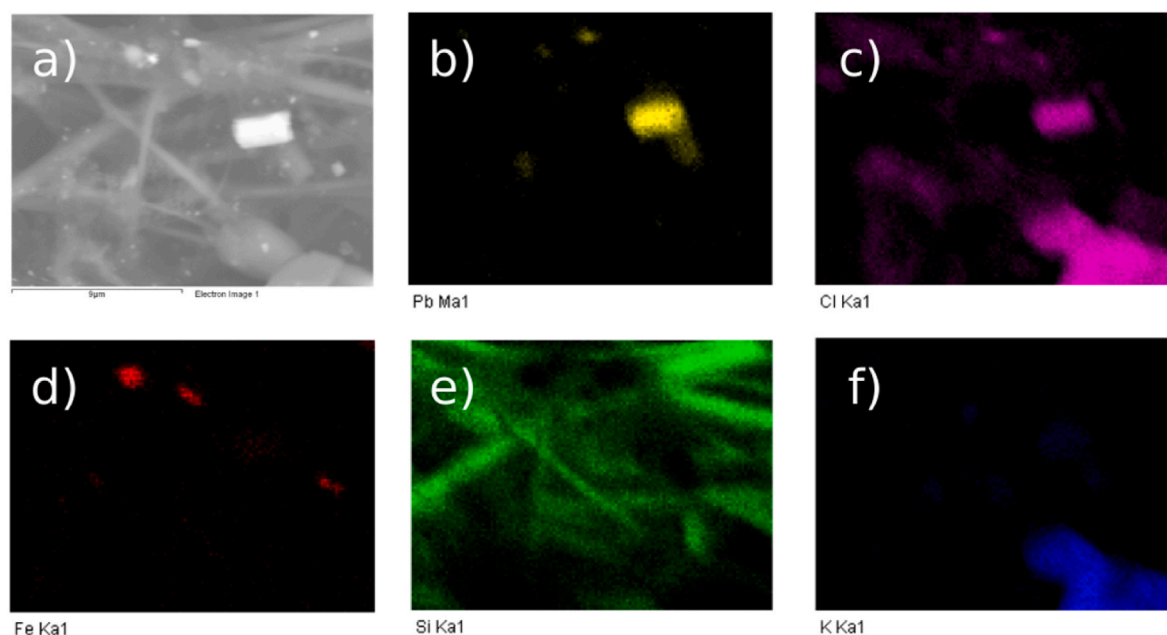


Fig. 4. (a) SEM image of particles from the fire, and corresponding mapping of the elemental distribution of (b) Pb ($M\alpha_1$), (c) Cl, (d) Fe, (e) Si, (f) K ($K\alpha_1$). Co-presence of Pb and Cl, as well as K and Cl can be observed in some particles. Presence of Si is seen where the filter fibres are.

and 389 in positive and negative ionizations, respectively). Oxidized and nitrogen-containing compounds as well as unsaturated molecules are dominating the assigned formulae (Fig. 5). This pattern in chemical classes has previously been described by Papazian et al. (2022) for $PM_{2.5}$ samples collected from polluted air.

3.1.5. EPA's 16 priority polycyclic aromatic hydrocarbons (PAHs)

The EPA's 16 priority PAHs were determined in PM_{10} samples from the open burning and the fully covered period at Söderängstorp and Tegelvreten (Table 3). High PAH concentrations coincided with high PM and BC concentrations originating from the fire. Small-scale wood

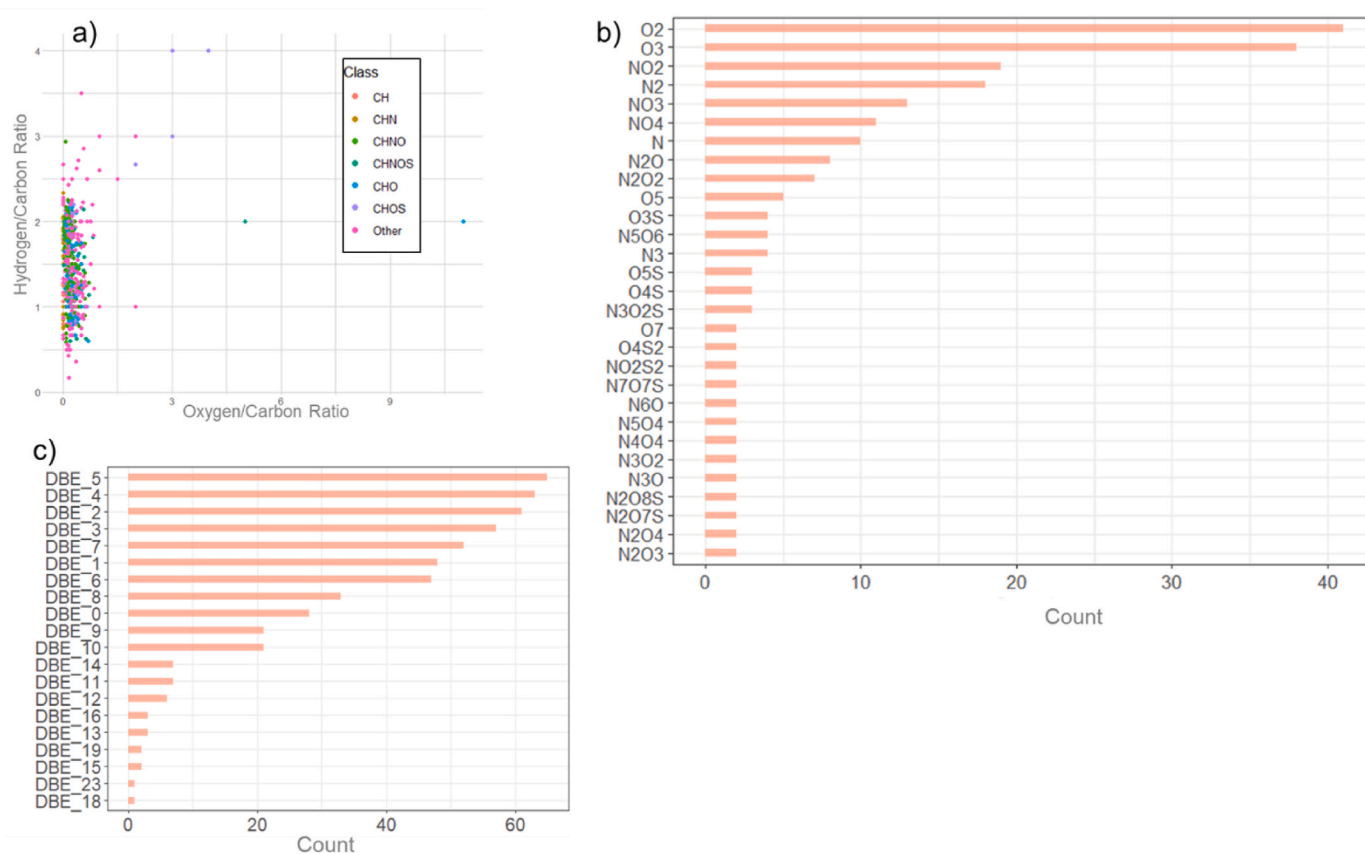


Fig. 5. (a) Van Krevelen plot of all the features with an assigned molecular formula. (b) Number of assigned formulae containing a specific functional group when more than one formula has the same functional group. (c) Distribution of the calculated double bond equivalent.

Table 3

Concentrations of PAHs in PM₁₀ in the air during open burning and the fully covered period (results are a combination of data from Söderängstorp and Tegelvreten). Samples from the fully covered period have lower detection limits due to filter pooling. Concentrations that are < LOQ for the open burning period are therefore not necessarily lower than quantified concentrations during the fully covered period. Samples < LOQ were taken as 0 for the calculation of the summary statistics.

Pollutant (ng/m ³)	Open burning		Fully covered	
	Mean (range)	Median	Mean (range)	Median
Naphthalene	<LOQ		0.040 (<LOQ–0.12) ^a	0.021
Acenaphthylene	<LOQ		0.013 (<LOQ–0.033) ^b	0.0095
Acenaphthene	<LOQ		0.0030 (<LOQ–0.0083) ^b	0.0019
Fluorene	<LOQ		0.0059 (<LOQ–0.016) ^a	0.0035
Phenanthrene	0.84 (0.25–1.9)	0.58	0.030 (<LOQ–0.063) ^a	0.028
Anthracene	0.34 (0.091–0.81)	0.23	0.0019 (<LOQ–0.0056) ^b	0.0010
Fluoranthene	13 (4.5–22)	12	0.055 (0.018–0.089)	0.056
Pyrene	12 (5.2–21)	12	0.038 (<LOQ–0.081) ^a	0.036
Benzo[a]anthracene	18 (10–28)	17	0.013 (0.005–0.022)	0.013
Chrysene	17 (9.2–25)	16	0.030 (0.012–0.047)	0.030
Benzo[b]fluoranthene	5.3 (3.0–8.3)	4.9	0.060 (0.028–0.097)	0.058
Benzo[k]fluoranthene	2.3 (1.3–3.8)	2.0	0.028 (0.013–0.045)	0.026
Benzo[a]pyrene	5.1 (3.2–8.8)	4.2	0.048 (0.024–0.078)	0.045
Dibenz[a,h]anthracene	0.55 (0.34–0.93)	0.46	0.014 (0.0067–0.023)	0.014
Benzo[ghi]perylene	3.1 (1.9–5.3)	2.6	0.076 (0.039–0.12)	0.074
Indeno[1,2,3-cd]pyrene	2.9 (1.6–4.8)	2.6	0.071 (0.035–0.11)	0.070

^a 1 out of 4 samples below limit of quantification (1/4).

^b 2/4.

burning for heating of homes may also have contributed to the PAH concentrations in the Kagghamra area. However, for the 12 PAHs which have lower volatilities and are therefore more likely found in the particle phase, concentrations drastically decreased upon covering of the fire, indicating that the waste heap fire was the major contributor to those PAHs in the air during the open burning period. The highest daily mean concentrations were found for benzo[a]anthracene (18 ng/m³) and chrysene (17 ng/m³). For the 4 most volatile PAHs (naphthalene, acenaphthene, fluorene, acenaphthylene) the concentrations were very low both during open burning and fully covered periods, often below the LOQ. Naphthalene is routinely detected in the gas phase of waste burning emissions, with emission factors in the order of g/kg (Chaudhary et al., 2022). In addition, the gas-phase concentrations of naphthalene measured during this campaign were above 1 µg/m³ (see Section 3.1.10). The low concentrations of high volatility PAHs in the particulate phase are therefore likely a reflection of their volatility rather than their absence in the waste fire emissions.

Benzo[a]pyrene is the only PAH that is regulated in the air in Sweden (and the EU); according to the EQS and EU AQS its concentration should not exceed an average of 1.0 ng/m³ annually (SFS, 2010:477; Directive, 2004/107/EC). Due to the limited measurement duration, it is not possible to determine whether this limit was exceeded. However, the daily average benzo[a]pyrene concentration during open burning (5.1 ng/m³), was greatly elevated relative to regional background concentrations in Sweden (0.02 ng/m³) (SMHI).

The concentrations of PAHs have also been measured at other waste

fires. At a landfill fire in Iowa city, US, several PAHs were measured in the PM_{2.5} fraction in the plume impacted by the fire (4.2 km from the fire). The highest concentration (of those PAHs also measured in our study) were chrysene (5.44 ng/m³ in PM_{2.5}) and benzo[b+k]fluoranthene (9.82 ng/m³ in PM_{2.5}). Most PAH concentrations at the Kagghamra fire (in PM₁₀ fraction 1 km from the fire) were 1–3 times higher, and sometimes approximately 20 times higher (for fluoranthene and pyrene) than at the Iowa city landfill fire (Downard et al., 2015). After a landfill fire in Seseña, Spain, EPA's priority PAHs were measured 0.5 km from the landfill (Nadal et al., 2016). In agreement with the Kagghamra fire, the concentrations of chrysene and benzo[a]anthracene were the highest (112 and 9.77 ng/m³, respectively), and furthermore, the concentration of the 4 most volatile PAHs were not detected. However, the heaviest PAHs (benzo[a]pyrene, benzo[ghi]perylene, and indeno[1,2,3-cd]pyrene) were not detected at Seseña, but found at relatively high concentrations at the Kagghamra fire. At a landfill fire in Iqaluit, Canada, the concentrations of benzo[a]pyrene during active burning and when extinguished were at similar levels (0.303 and 0.245 ng/m³, respectively), probably since the benzo[a]pyrene concentration was not clearly impacted by the fire (Weichenthal et al., 2015).

3.1.6. Extended analysis of PAHs and OPAH derivatives

In addition to the quantitative analysis of the 16 priority PAHs, the relative contributions of an extended set of PAHs and OPAH derivatives were determined for a selection of samples. After blank subtracting (average from three field blanks) the quantified amounts of each compound, we calculated the compositional profiles of the 45 targeted PAHs and 10 OPAHs, respectively, for the analysed PM₁₀ high-volume Teflon filter samples. The results from these calculations are shown in Table S13.

The compositional PAH profiles, shown in Fig. 6a and b shows that the open fire impacted filter sample (TC 001) had a very high (85%) relative abundance of low-molecular weight (LMW) PAH (<252 Da); consequently the relative abundance of the more toxic (Andersson and Achten, 2015) high-molecular weight (HWM) PAHs (≥252 Da) was low (15%). For the filter collected upstream the wind direction of the largely covered fire (TC 004), representing ambient background air, the LMW PAH accounted for 60%, and the HWM PAHs for 40% of the total PAH content. The other 4 filters collected during the final extinguishing period showed similar compositional profiles to the ambient background filter regardless of wind direction, indicating only minimal contribution from fire emissions during this period. In the open fire impacted filter, more volatile OPAHs were more abundant than in the background air. The compositional OPAH profiles shown in Fig. 6c showed that the major OPAH constituents in the background air were 9, 10-anthracenedione, 7H-Benz[de]anthracene-7-one, 6H-Benzo[cd]pyrene-6-one, while the OPAHs 9-Fluorenone and 9,10-anthracenedione (sum: 62%) dominated the OPAH compositional profile for the open fire impacted filter.

Noteworthy is the very high relative abundance of alkylated PAHs in the sample impacted by the open fire. In this sample the alkylated species 1-Methylphenanthrene, 2-Methylphenanthrene, 2-Methylanthracene, 9-Methylphenanthrene, 1,7-Dimethylphenanthrene, 1-Methylfluoranthene and 3-Methylfluoranthene, 1-Methylpyrene, 4-Methylpyrene, 3-Methylchrysene, and 6-Methylchrysene accounted for 43% of the total PAH content. For the background filter sample, these alkyl PAHs accounted for only 9.2% of the total PAH content. Alkyl three ring PAHs are typically not highly abundant on atmospheric particles compared to their unsubstituted parent PAH compound (Lim et al., 2021), likely because of predominately being partitioned into the gas phase due to their higher vapor pressures (Moradi et al., 2022), as well as their enhanced atmospheric reactivity compared to their unsubstituted parent PAH (Keyte et al., 2013). Elevated air concentrations of alkyl PAHs have been associated with unburned and/or combusted fossil fuels (Moradi et al., 2022).

Among the most abundant PAHs (>5% relative abundance) in

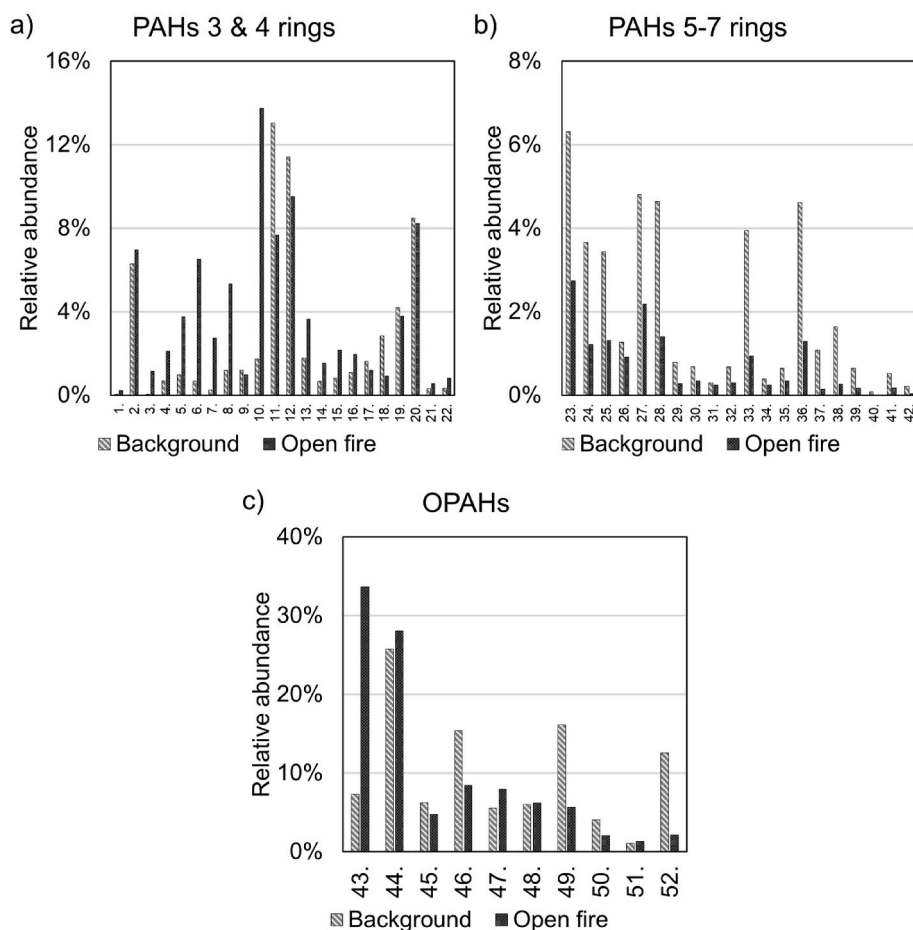


Fig. 6. (a) Compositional profiles for PAHs between 178 and 242 Da, (b) high molecular weight PAHs, between 252 and 302 Da, and (c) OPAHs in the open fire impacted filter and the ambient air background. Compound numbering: 1. Dibenzothiophene, 2. Phenanthrene, 3. Anthracene, 4. 3-Methylphenanthrene, 5. 2-Methylphenanthrene, 6. 2-Methylantracene, 7. 9-Methylphenanthrene, 8. 1-Methylphenanthrene, 9. 4*H*-Cyclopenta[*def*]phenanthrene, 10. 1,7-Dimethylphenanthrene, 11. Fluoranthene, 12. Pyrene, 13. 1/3-Methylfluoranthene, 14. 7*H*-Benzo[*c*]fluorene, 15. 4-Methylpyrene, 16. 1-Methylpyrene, 17. Benzo[*c*]phenanthrene, 18. Benzo[*ghi*]fluoranthene, 19. Benzo[*a*]anthracene, 20. Triphenylene + Chrysene, 21. 3-Methylchrysene, 22. 6-Methylchrysene, 23. Benzo[*b*]fluoranthene, 24. Benzo[*j*]fluoranthene, 25. Benzo[*k*]fluoranthene, 26. Benzo[*a*]fluoranthene, 27. Benzo[*e*]pyrene, 28. Benzo[*a*]pyrene, 29. Perylene, 30. Dibenz[*a,j*]anthracene, 31. Dibenz[*a,c*]anthracene, 32. Dibenz[*a,h*]anthracene, 33. Indeno[1,2,3-*cd*]pyrene, 34. Benzo[*b*]chrysene, 35. Picene, 36. Benzo[*ghi*]perylene, 37. Anthanthrene, 38. Coronene, 39. Dibenz[*b,k*]fluoranthene, 40. Dibenz[*a,l*]pyrene, 41. Dibenz[*a,e*]pyrene, 42. Dibenz[*a,i*]pyrene, 43. 9-Fluorenone, 44. 9,10-Anthracenedione, 45. 4*H*-Cyclopenta[*def*]phenanthrene-4-one, 46. 2-Methyl-9,10-anthracenedione, 47. 11*H*-Benzo[*a*]fluoren-11-one, 48. 11*H*-Benzo[*b*]fluoren-11-one, 49. 7*H*-Benz[*de*]anthracen-7-one, 50. Benzo[*a*]anthracene-7,12-dione, 51. 5,12-Naphthacenedione, 52. 6*H*-Benzo[*cd*]pyren-6-one.

background air were: Fluoranthene (median: 13%), Pyrene (median: 11%), sum of Triphenylene and Chrysene (median: 8.5%), Phenanthrene (median: 6.3%), and Benzo[*b*]fluoranthene (median: 6.3%). Similarly, for the open fire impacted filter the most abundant PAHs were: 1,7-Dimethylphenanthrene (14%), Pyrene (9.5%), sum of Triphenylene and Chrysene (8.2%), Fluoranthene (7.7%), Phenanthrene (7.0%), 2-Methylantracene (6.5%). Some noteworthy differences between the background air and the open fire sample, in addition to the alkyl-PAHs, and LMW/HMW PAHs mentioned above are the OPAHs 9-Fluorenone (much higher relative abundance in the open fire sample), 7*H*-Benz[*de*]anthracene-7-one, and 6*H*-Benzo[*cd*]pyren-6-one that both were substantially more abundant in the background air sample. Compositional profiles for those PAHs that were in common between the 16 PAHs measured on the low-volume filters (Section 3.1.5) and the extended analysis in this section largely agreed, with two exceptions (Fig. S5). However, it should be noted that it is difficult to directly compare the results due to differences in sampling periods and methods, as well as sample preparation and analysis.

3.1.7. Per- and polyfluoroalkyl substances (PFAS)

Per- and polyfluoroalkyl substances are synthetic chemicals used in a

wide range of industrial and consumer applications, such as food-contact materials, textiles, fire-fighting foams, cosmetics and fluoropolymers (Glüge et al., 2020). Several PFAS are regulated under REACH (ECHA b) and the Stockholm Convention (UNEP), due to concerns around their hazard to human health. A proposal to restrict the use of all PFAS within the EU was recently submitted to the European Chemicals Agency (ECHA c). Due to their ubiquity in consumer products, PFAS eventually enter the waste stream. A number of studies have reported their occurrence in landfills, solid waste, and landfill leachate (Allred et al., 2015; Bečanová et al., 2016; Björklund et al., 2021; Lang et al., 2016; Wei et al., 2019). Despite of this, little is known about the fate of PFAS during combustion. Laboratory studies on the thermal stability of fluoropolymers have reported formation of various organofluorine substances (Ellis et al., 2001, 2003; García et al., 2007; Schlummer et al., 2015) at 390–600°C. This suggest that uncontrolled burning of waste may be a source of PFAS or other organofluorine substances to the atmosphere.

Three PFAS were detected in the sample which was strongly influenced by the fire (QFF 001): perfluoropentanoic acid (PFPeA, 8.5 pg/m³) and the fluorotelomer sulfonic acids 6:2 FTS (1.29 pg/m³) and 8:2 FTS (>LOD). These PFAS were present in lower levels in the other

samples (QFF 002–QFF 006) or not detected at all. Additionally, perfluorooctane sulfonic acid (PFOS) was observed below the LOQ in one sample (QFF 004). The presence of these PFAS may indicate release by the (incomplete) combustion of various types of packaging, carpets and textiles containing fluorotelomer based coatings (Coffin et al., 2023; Lang et al., 2016). The waste fire was extinguished by covering it with sand. As such, no aqueous film forming foams (AFFFs) were applied, probably ruling these out as the source of airborne PFAS. However, because of the highly contaminated nature of the fire impacted sample, peaks were noisy and the response of the internal standard was strongly suppressed or absent altogether. Therefore, the presence or absence of most of the analytes could not be confirmed. Due to the matrix effects, the relatively low levels of the detected PFAS, uncertainties in the flow rate determination and the fact that only half of the filter was used for this analysis, estimated air concentrations of the detects stated above and in Table S14 for the other samples should be interpreted very cautiously and seen as semi-quantitative. Some analytes, for example perfluorooctanoic acid, have previously been reported in Swedish background air at levels exceeding the LODs achieved for samples less impacted by the fire in this study (SMHI). A possible reason for the low detection frequency in this study could be the sampling time resolution, which was 24 h in contrast to one month in the Swedish air monitoring program. Another possible reason was the very cold and dry weather during the sampling period.

The severe matrix effects observed here demonstrate that the most commonly used method for extraction and analysis of PM-associated PFAS does not perform well for fire impacted samples. Method development for this specific sample type is needed to confidently study PFAS emissions from combustion.

3.1.8. Total extractable organofluorine (TEOF)

Two samples displayed detectable levels of TEOF (6.4 and 4.4 ng F/m³). These were among the top three samples with the highest BC levels among the samples analysed for TEOF, indicating that the fire may have contributed to elevated levels of TEOF in air. However, TEOF was below the LOD in the sample displaying the highest BC levels. All other analysed samples also had TEOF concentrations below the LOD. The results should be interpreted with caution, as the detected levels were below the LOQ.

TEOF is a measure of the sum of all fluorine-containing organic substances which can be extracted using the chosen method. These may

include the PFAS analysed using a targeted method in this study (section 3.1.7), but also other known PFAS as well as previously uncharacterised organofluorine potentially formed during combustion. There is currently no validated or standardised method for TEOF in air samples. To our knowledge, only one previous study has reported a measure of total organofluorine in air. Lin et al. (2022) determined organofluorine indirectly, by subtracting measured fluoride concentrations from measured total fluorine levels. They observed organofluorine levels in the range 1.7–7.3 ng F/m³ in outdoor air PM in samples collected in urban and industrialised areas in Japan. For the fire-impacted samples analysed as part of this study an extraction method using methanol was selected to produce a direct measure of the TEOF. Methanol is the most commonly used solvent for targeted analysis of known PFAS associated with PM in air, but may be less suitable for more non-polar PFAS. Due to the low sample volume (24 m³), the LOD and LOQ achieved for TEOF are approximately three orders of magnitude higher than background levels (SMHI) of the PFAS covered by the targeted analysis in our study. As many data points were below the LOD and there is a paucity of other data in the literature, we cannot conclude whether the fire event contributed to elevated levels of organofluorine in air.

3.1.9. Nitrogen oxides

The daily concentrations of NO₂ and NO measured at Söderängstorp and Tegelvreten were decreasing in the order from open burning, extinguishing period to fully covered period (Table 1, Fig. 7 and Fig. S6). However, the concentration differences between the periods were relatively small, especially for NO. The Swedish EQS (and EU AQS) for NO₂ was not exceeded (24-h average of 60 µg/m³ max. 7 days/year) (SFS, 2010:477; Directive, 2008/50/EC). WHO daily recommended AQG for NO₂ is stricter; 25 µg/m³ max 3–4 days/year (WHO, 2021). However, even these stricter guideline values were never exceeded during the measurement period. Unlike the PM_{2.5} concentrations, NO₂ concentrations were relatively low during the open burning period in Kagghamra. This result is similar to the observations during the landfill fire in Iqaluit, Canada (Weichenthal et al., 2015).

3.1.10. VOCs

The main VOC classes found in the smoke include a) small linear hydrocarbons (lin.-C_xH_y, including saturated and unsaturated hydrocarbons) which cannot be quantified by PTR-MS, b) small oxygenated VOCs (C_xH_yO_z such as methanol, formaldehyde, acetaldehyde, acetic

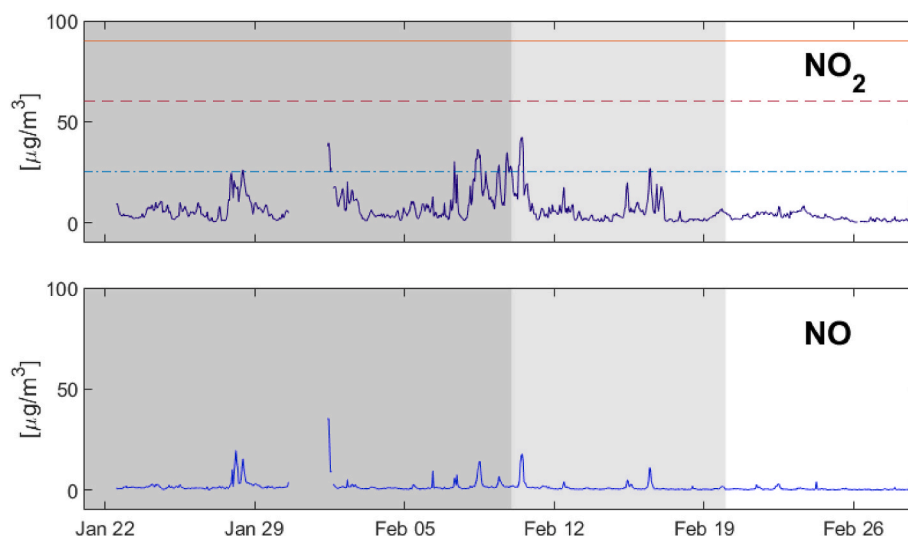


Fig. 7. Timeseries of hourly averages of NO₂ and NO at the Söderängstorp measurement site. Dark grey, light grey and white mark the open burning period, final extinguishing period and fully covered period (only partially shown), respectively. The orange solid line and the red dashed line mark the limits for NO₂ over 1 h (90 µg/m³ max 175 h/year) respective 24-h (60 µg/m³ max 7 days/year) according to the Swedish environmental quality standards for outdoor air, while the turquoise dash-dotted line indicates the recommend level over 24 h according to the WHO 2021 air quality guidelines (25 µg/m³ max 3–4 days/year).

acid, furans and phenols) and c) aromatic hydrocarbons (arom.-C_xH_y, such as benzene, toluene, xylenes/ethylbenzene, styrene, and naphthalene). A full list of the major VOCs detected in the gas phase and their concentrations can be found in Table S15. Known air toxics such as formaldehyde (26.1 and 8.5 µg/m³), 1,3-butadiene (3.1 and 2.9 µg/m³), and benzene (49.2 and 27.3 µg/m³) were detected at concentrations about one order of magnitude higher at Söderängstorp respective Tegelvreten than typically observed in a Nordic urban environment such as Oslo during wintertime (unpublished data by the University of Oslo group). While no direct health implications can be derived from short-term measurements of these air toxics, their presence at the fire indicates potential health concerns and highlights the need for more extensive investigation of their emissions from waste facility fires. Benzene, toluene, xylenes and in particular styrene were found in higher relative abundances than typically observed when natural wood fuels are burned (e.g. Koss et al., 2018). High concentrations of styrene (and methylstyrene) indicate that polystyrene was among the fire fuels.

3.2. Toxicity

The results on cytotoxicity showed no evident effect at the tested concentrations following 24 h exposure (Fig. 8a).

Similarly, the release of the four cytokines (IL-1β, IL-6, TNF-α and IL-8) showed in general no or small increased levels in comparison to the control. Although the mean of IL-6 was higher compared to the control, the increase was not significant due to high standard deviation (Fig. 8c). For TNF-α, the levels were slightly increased (2.8-fold increase) although this increase was not statistically significant (p value 0.178) (Fig. 8d). Despite the dilution of samples, the levels of IL-8 were above the standard curve and no conclusions can be drawn (data not shown). The positive control LPS exhibited a significant increase in IL-1β and TNF-α levels compared to the control. Taken together, the results show only small effects on the release of cytokines (with mainly a slight increase of TNF-α levels) following dTHP-1 cells exposure to particles at 50 µg/mL. Ultimately, additional doses and different endpoints should

have been studied, but this was not possible due to limited amount of collected particles. Given the high levels of PAHs, genotoxicity/mutagenicity would have been interesting to further explore.

4. Conclusions

Only few characterizations of waste fire emissions have been done to date, probably mainly due to the difficulties in arranging measurement campaigns at the moment fires occur. The Kagghamra waste fire lasted for nearly 2 months (23 December 2020–19 February 2021), which made it possible to perform detailed quantitative and qualitative measurements of different air pollutants.

At open burning, high concentrations of PM₁₀, PM_{2.5} and BC are co-occurring, especially when the wind direction was towards the measurement sites. WHO AQG for PM₁₀ and PM_{2.5} were exceeded during the measurement period, while the less strict 24-h mean of EQS and EU AQS were not exceeded. However, we only conducted continuous measurements for less than half of the duration of the open burning period and towards its end, while initial measurements showed particularly high concentrations of PM. It is therefore not unlikely that EQS and EU AQS can be exceeded during long ongoing fires such as the one in this study. All regulated metals (Pb, Cd, As) apart from Ni, were found at greatly elevated concentrations in air during the open burning period compared to regional background. However, as only the annual mean is regulated, the measured values in this study cannot be directly compared to the standards. The same is true for PAHs: strongly elevated concentrations of particulate PAHs were found during the open burning period for the less volatile PAHs as compared to the regional background. However, only one of the PAHs is currently regulated in the EU and the respective standard only concerns the annual mean.

The results from this study show that waste fires can have a strong impact on the air quality of nearby residential areas. To enable better assessment of the impact of future fires on health, it would be important to be prepared to rapidly mobilise air quality monitoring equipment.

Although strongly elevated concentrations of harmful pollutants

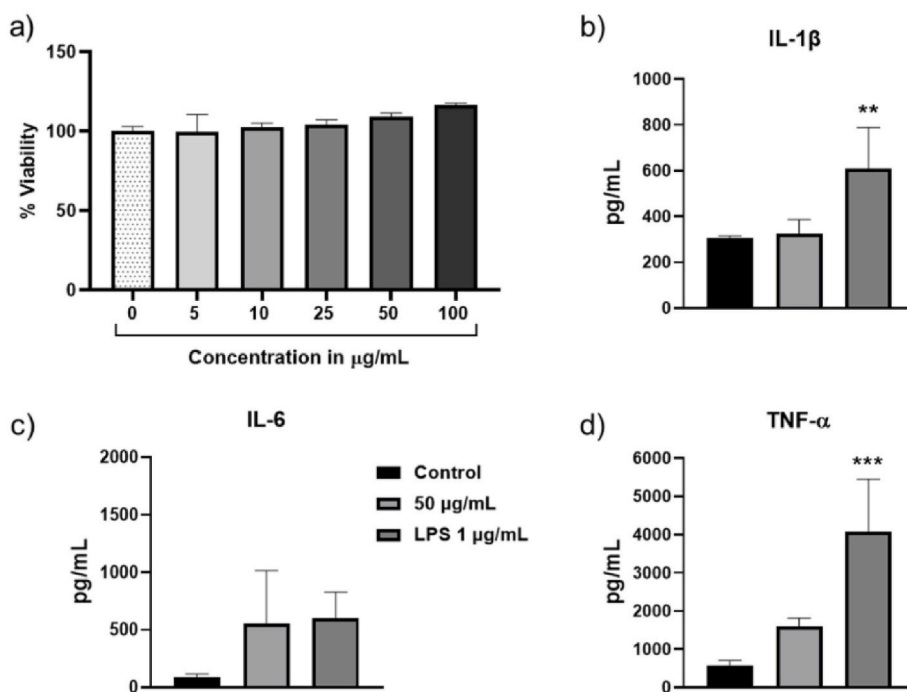


Fig. 8. Cell viability and cytokine release of dTHP-1 following exposure to the particles for 24 h. The cell viability (a) was assessed by the Alamar blue assay. The results are presented as mean ± SD of two independent experiments. Cytokine release (b, c, and d) was assessed using a multiplex electrochemiluminescence immunoassay kit. LPS (1 µg/mL) was used as a positive control and the results are presented as mean ± SD of two independent experiments with two technical replicates (n = 4).

were found in air, in-vitro testing of collected particles showed only minor toxic effects. It should be noted that toxicity study was limited in scope regarding the number of samples and measured endpoints. Future studies focusing specifically on investigating the toxicity of this type of particulate emission are therefore needed.

Credit author statement

All authors: Writing - Review & Editing. Karine Elihn: Conceptualization, Formal Analysis, Investigation, Resources, Writing - Original Draft, Visualization, Project Management. Joost Dalmijn: Investigation, Writing - original draft. Jean Froment: Investigation, Formal analysis, Writing - Original Draft, Visualization. Alexander Håland: Investigation. Jana H. Johansson: Conceptualization, Investigation, Writing - original draft. Hanna L. Karlsson: Methodology, Supervision. Jonathan W. Martin: Supervision. Tomas Mikoviny: Investigation. Michael Norman: Conceptualization, Formal analysis, Investigation, Writing - Original draft, Project administration. Felix Piel: Investigation. Ioannis Sadiktis: Formal analysis, Investigation, Resources, Visualization, Writing - Original Draft. Daniel Schlesinger: Formal analysis, Visualization. Sanna Silvergren: Investigation, Data curation, Writing - original draft, Visualization, Project administration. N. V. Srikanth Vallabani: Investigation, Formal analysis, Writing - Original Draft. Armin Wisthaler: Investigation. Sarah S. Steimer: Conceptualization, Formal analysis, Investigation, Resources, Writing - Original Draft, Visualization, Project administration.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2023.120124>.

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