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Nanoparticle exposure due to pyrotechnics during a football match

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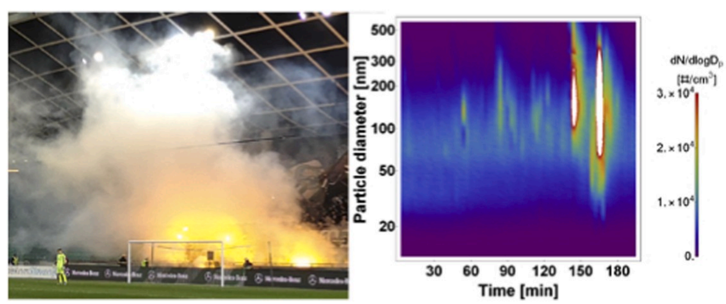
HIGHLIGHTS

- The use of pyrotechnics at sport events poses harm to the audience and athletes.
- The use of pyrotechnics resulted in a release of large amounts of nanoparticles.
- Heavy metals such as Cu, Sr, and Ba were detected.
- The total concentration increased up to 1200% during the pyrotechnic event.
- Athletes were more exposed due to their higher respiration rate.

GRAPHICAL ABSTRACT



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ABSTRACT

Use of pyrotechnic articles is a common, though forbidden practice at football matches. While the fans, especially the members of the ultras groups, view pyrotechnic displays as a part of their culture, these devices can be dangerous for the public. In addition to the risks of burning, hand flares and similar items release toxic combustion by-products, including particulate matter – inhaling of which is harmful both for the spectators and players. In order to assess the amounts and composition of the aerosol particles released at a typical event, we performed a study using both a scanning mobility particle sizer and a low-pressure cascade impactor. The number concentration of nanoparticles was clearly correlated with the burning events. Several elements were identified in the collected samples, including heavy metals, while the majority of the sample consisted of amorphous carbon. The nanoparticle number concentration increased up to 12-fold immediately after the beginning of the flares burning, with the largest contribution of particles 155 nm in diameter. The cumulative dose the players inhaled during the match was around 7×10^8 particles/kg, which is 300% higher than the dose one would get in a low-pollution environment. We discuss the results in view of similar pyrotechnical events, especially fireworks.

1. Introduction

Football is the most popular sport worldwide, with matches attracting hundreds of thousands of spectators to the stadia every week, and up to tens of thousands to individual matches at the largest venues. Among the football fans, there are special types of football support groups, called the ultras (Kennedy, 2013). The ultras are typically associated with the football culture of southern Europe (the term “ultra”

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originating from Italy), but such groups exist across the entire continent and beyond (Kennedy, 2013; Tsoukala et al., 2016). During the matches, the ultras support their teams with chants, banners, elaborated choreographies, and often with the use of hand flares, smoke bombs, and other pyrotechnic devices. Pyrotechnics at sport events are a complex and controversial topic. While they are sometimes discussed within the scope of football hooliganism (Tsoukala et al., 2016; Brechbühl et al., 2017; Perasović and Mustapić, 2018; Scholz and Vespalec, 2019) and are banned from the stadia, the use of pyrotechnic articles is often seen by ultras groups as an intrinsic part of their culture (Brechbühl et al., 2017; Herd, 2017; Almgren et al., 2018). This goes even to the extent that they would be risking a confrontation with the police and heavy fines in order to bring them to the stadium. A survey among members of such groups showed that even two thirds of them perceive firing of pyrotechnics as the least serious among various violent incidents (Scholz and Vespalec, 2019). On the other hand, the Union of European Football Associations (UEFA), the European football's highest governing body, frowns upon the use of pyrotechnics and has recently commissioned a scientific study on associated health and safety risks (Smith, 2016). According to a senior UEFA official, the use of pyrotechnics is reported at over 25% of matches in UEFA competitions. The UEFA's position is that pyrotechnics behave like explosives and that there is no such thing as safe use of pyrotechnics in such confined spaces (stadia) (Independent scientific). According to the Slovenian Football Association (private communication), hand flares were used at 54 matches in the 2018/19 season (35 of them at the Slovenian First Football League matches).

Some of the dangers associated with the use of pyrotechnics in stadia include burns, physical injuries, hearing loss, and toxic combustion by-products production (SO_2, NO_2, NO_x, as well as solid oxides of metals and more complex compounds) (Smith, 2016). Typically, pyrotechnic articles are designed to burn to completion (the composition includes their own oxidants) and can therefore not be extinguished by conventional means during burning. Some devices even burn under water. Furthermore, some articles are designed to explode, thus potentially causing both blast damage and fragment throw (Smith, 2016). The dangers increase when devices perform "abnormally", which is more likely in cheap or home-made articles or those that are poorly handled (Smith, 2016). The use of pyrotechnics exposes both spectators and football players to the polluted air. The players themselves are particularly prone to risk due to the airborne pollutants, as they are physically highly active during the match. Midfielders can run over 10 km during the course of the match and even the goalkeepers can cover 4 km (Reilly and Thomas, 1976), and they play matches at least on a weekly basis.

In the past two decades, the adverse health effects of particulate matter in the air have become widely known (Curtis et al., 2006; Vu et al., 2017; Pöschl, 2005; Khan et al., 2019; Zeng et al., 2019; Rizza et al., 2019; Naeher et al., 2007; Remškar et al., 2015; Brook et al., 2002; Peters et al., 1997). Some of the most recognized sources of particular matter include industrial activities, wood (Naeher et al., 200; Herich et al., 2014) and coal burning, as well as traffic, in particular diesel engines (Jathar et al., 2020; Ning et al., 2013; Rakowska et al., 2014). Significant amounts of nanoparticles are also emitted during some traditional entertainment activities, such as use of sparklers (Remškar et al., 2015), incense sticks (Višić et al., 2018), or fireworks (ten Brink et al., 2019). The PM_{10} particles (*i.e.* particulate matter 10 μm or less in diameter) and heavy metals have been associated with a series of medical conditions. Among those, especially prominent are pulmonary, such as asthma, bronchitis, pneumonia, decreased lung function, higher levels of chronic cough; and cardiovascular, such as heart problems, strokes and peripheral vascular disease, cardiac arrhythmias, arterial vasoconstriction, increased plasma viscosity. Additional adverse effects on health are higher risk of lung cancer, neurological and psychiatric problems, higher rates of mortality, reproductive and developmental problems (Curtis et al., 2006; Vu et al., 2017; Pöschl, 2005; Zeng et al., 2019; Naeher et al., 2007; Brook et al., 2002; Khan et al., 2018). Children are especially at risk when exposed to air pollution, as their lungs are still developing and may not reach their full capacity in adulthood (Gauderman, 2015).

In previous studies of aerosols and particulate matter on football stadia, Faber et al. (2013) analysed aerosol particles emitted in a football stadium during a match in Germany using high-resolution aerosol mass spectrometry. They recorded emissions from cigarette smoke and cooking at the food stalls; however, they recorded no pyrotechnics. In a related study, Veres et al. (2013) further demonstrated that spectators on a stadium also release substantial amounts of volatile organic compounds (VOC). Lichter et al. (2017) studied effects of air pollution (PM_{10}) on productivity of professional soccer players and reported that air pollution limits physical ability of the players and also induces behavioural adjustments with the aim to reduce pollution-induced physical strain. Using pyrotechnics at stadiums can cause fires such as the one at the Euroborg soccer stadium in Groningen in The Netherlands (van Belle et al., 2010). The study assessed the exposure to toxic fumes emitted when polyamide chairs are set on fire. The study found that CO, NO_x, HCN, NH_3, and other volatile compounds are formed during the burning of the polyamide chairs, and that the exposure to NO_x is the main health risk for the spectators. While there is only a small number of studies dealing with sport events in particular, several studies have focused on the aerosol particles released by fireworks at various types of celebrations. These can be linked to pyrotechnics on stadia, although each situation has its own peculiarities. Despite the fact that the fireworks are one of the most uncommon sources of air pollution, it has been recently clearly demonstrated that they are responsible for the increase of concentrations of both solid particles (including metals and organic compounds) and toxic gases (Singh et al., 2019). Drewnick et al. (2006) detected a 10-fold increase of mass concentration of sub-micron particles following the 2005 New Year's fireworks in Mainz, Germany. The daily PM_{10} concentration exceeded 50 $\mu g/m^3$ even the following day. The analysis of the physical and chemical properties of the particles (elements, ions, organic compounds, black carbon) and the granulation spectra of particles collected in Milan, Italy, in July 2006 when Italy won the FIFA World Cup (Vecchi et al., 2008) showed massive increases in concentrations of certain elements in nanoparticles: 120-fold for Sr, 22-fold for Mg, 12-fold for Ba, 11-fold for K, and 6-fold for Cu. Andradottir and Thorsteinsson (2019) reported a 104-fold increase in Cu, 96-fold in Sr, and 27-fold in Ba due to fireworks in Reykjavik. Croteau et al. (2010) used a combustion chamber to estimate the emission levels from ground-level pyrotechnics. They report that 5–13% of the combusted mass is converted to particles, and they estimate high emission factors for metals: 23–45 g/kg for K, 1–7 g/kg for Mg, 0.05–7 g/kg for Cu, and 0.03–6 g/kg for Ba, where the units are grams of particles per kg of combusted pyrotechnic material.

Wang et al. (2007) reported a six-fold increase of $PM_{2.5}$ during the Beijing Lantern festival. Joly et al. (2010) report a 50-fold increase of $PM_{2.5}$ during the international fireworks competition in Montreal (2007), not only in the centre of the event but in the surrounding area with the radius of 2 km as well. At the central locations, the top concentrations exceeded the background levels even 1000 times. The concentrations of K, Cl, Al, Mg, and Ti were elevated. This study demonstrated that the people in the vicinity of the fireworks are exposed to both high levels of $PM_{2.5}$ and chemical elements in nanoparticle forms with known adverse health effects. In a study by Bisht et al. (2013), PM_{10}, $PM_{2.5}$, black carbon, CO, and NO were measured at multiple stadiums during the Commonwealth Games in Delhi. The study showed that the concentration of $PM_{2.5}$ particles inside the stadiums were around 18% lower than outside the stadiums while the concentration of PM_{10} particles were similar. The source of the particles was attributed to combustion of fossil fuel, biomass burning, industrial activities and unfavourable meteorological conditions.

The above and other studies were typically time-limited and focused on particular events. On the other hand, Seidel and Birnbaum (2015) followed the average 1 h and 24 h $PM_{2.5}$ concentrations on 315 sites in the United States on the 4th of July celebrations in the years 1999–2013.

They found that the concentrations on the night before jumped everywhere, however the quantitative characteristics of the increases were dependent on various factors, such as the local weather conditions, distance from the source, the quality of the sensors, etc.

In this paper, we present a case study of nanoparticles emitted by pyrotechnics during a football match in Slovenia. The two teams, NK Olimpija Ljubljana and NK Maribor, have a long-standing rivalry, with teams being supported by their ultras groups, Green Dragons and Viole Maribor, respectively. This particular match was chosen for this case study as it had a high probability of pyrotechnics usage during the event, since the two ultras groups top the chart of hand flare-related incidents in Slovenian football. Nanoparticle time-dependant size distribution was measured in relation to the sporadic uses of pyrotechnic articles. The samples were collected by low-pressure cascade impactor for further investigations by electron microscopy and energy-dispersive X-ray spectroscopy.

2. Materials and methods

The experiment was carried out during the Slovenian First Football League match, NK Olimpija Ljubljana vs. NK Maribor, taking place at the Stožice stadium in Ljubljana on March 16, 2019 (Prva liga match report.). The stadium, inaugurated in 2010, is an open-roof stadium with a seating capacity of 16 038. The match started at 20:30 and lasted the standard 90 min with a 15-min half time break and included some minutes of extra time at the end of each half. Experimental equipment was positioned on the same level as the football pitch, on the western side behind the commercial banners, roughly at the central line. This particular position was chosen in order to be at the same distance from both supporter groups, seated at the opposite seating areas at North (Green Dragons, GD) and South stand (Viole Maribor, V), behind the goal area.

Two experimental setups were used for measurements on the stadium. For chemical analysis, we used Dekati® low-pressure cascade impactor (DLPI; Dekati Ltd.). The multi-stage DLPI collects particles from air on 13 stages. Each stage was covered with an aluminium substrate on which the sample was collected. Each stage has a defined cut diameter D_{50}, determined as the particle diameter with the 50% collection efficiency, and ranges from 30 nm up to 10 μm (Marjamäki et al., 2000).

The nanoparticle (NP) total concentration and time-dependant size distribution was measured with a Scanning Mobility Particle Sizer (SMPS, electrostatic classifier model 3080; TSI Co., Shoreview, MN, USA) equipped with desiccator, soft X-ray neutralizer, long differential mobility analyser (DMA, model 3081; TSI), and a water condensation particle counter (WCPC; model 3785; TSI). The electrical mobility diameter of counted NP was from 13 nm to 572.5 nm. The normalized concentration, $dN/dlogD_p$, measured with the SMPS, is the total concentration (dN) in within the measured range divided by the difference between the logarithms ($dlogD_p$) of the lower and upper diameter of the counted particles. This makes the normalized concentration independent on the channel width of the instrument (Hinds, 1999). The normalized concentration in this form is used so that results from instruments with different channel widths can be compared. The measurements were performed as successive size scans, with a 3-min duration of a single scan. The measurements with SMPS started 63 min before the beginning of the match and ended 22 min after the match finished, when all spectators have already left the stadium, in order to measure the background concentration. DLPI was collecting the samples only during start-end period of the match, as this was the time the pyrotechnics were used.

To get a reasonable estimation of the background pollution levels, the data from an automatic monitoring station by the Slovenian Environment Agency (situated in Ljubljana Bežigrad, about 2 km from the stadium) were analysed, including the PM_{10} concentration, as well as the values for SO_2 NO, NO_2, NO_x, and CO, together with the temperature and relative humidity.

Chemical analysis was made with a FEI HeliosNanolab 650 scanning electron microscope (SEM) using energy-dispersive X-ray spectroscopy (EDS). All the samples were coated with approximately 5 nm of gold with the aim to prevent charging effects during electron irradiation. To determine the presence of different elements, an area about the size of 100×100 μm^2 was selected on the deposited material in each DLPI stage where the EDS analysis was performed.

The pyrotechnics used during the match were composed of an unknown mixture of commercial and homemade articles (due to the nature of the event, a detailed identification of the sources was not possible). NK Olimpija's colours are green while those of NK Maribor are purple, which was reflected in the choice of the colours of the hand flares of the two teams. In total, according to the officials, the estimated number of hand flares was 20 on GD side and 60 on V side. Other potential sources of particulate matter included cigarette smoke from spectators, dust on the spectator area being lifted due to people moving, and traffic sources from Ljubljana bypass, which runs within 100 m from the football stadium. Mid-March of 2019 was still within the heating season, so sources related to individual households burning wood should not be excluded.

3. Results

3.1. Event progress

The SMPS measurements started 1 h before the beginning of the match in order to capture the background not related to the pyrotechnics, allowing us to estimate the contribution of sources such as cigarette smoke, wood burning, and traffic. Table 1 shows the key events during the experiment. The spectators started arriving *en masse* about 20 min before the match begun and have mostly left the stadium 15 min after the match has concluded. According to the official record, the number of spectators was 9500 (Prva liga match report.).

During the match, significant use of pyrotechnics was recorded on three occasions. The first batch of hand flares was lit by GD and burned approximately 3 min. The second event started roughly simultaneously by both groups (5 min of burning in total), and the third occurrence was initiated by V and followed by GD a couple of minutes later (8 min). Some representative photographs of the event are shown in Fig. 1. In addition, cigarette smoke was present during the entire event with some variations in intensity, more intensely around the beginning of the match and during the half time. Apart from hand flares, nothing particularly significant happened during the match. Two yellow cards were issued, not correlated with the times of the flare burning, and no goals were scored, ending the match with 0:0.

According to the automatic monitoring station, the temperature in the evening was 12 °C and the relative humidity was 70%.

3.2. Particle size distribution measurements

Fig. 2 shows a two-dimensional size distribution colour plot, given as normalized concentration ($dN/dlogD_p$) (a), and the total nanoparticle

Table 1
Relevant events during the measurement and their timestamps.

Real time (hh:mm)	Time (min)	Event
19:30	0	SMPS measurement starts
20:30	63	Match starts, DLPI measurement starts
20:48	81	Hand flares, GD
21:17	108	End of first half
21:33	126	Start of second half
21:48	141	Hand flares, V and GD
22:09	162	Hand flares, V and GD
22:21	174	Match ends
22:25	177	DLPI measurement ends
22:42	195	SMPS measurement ends

Fig. 1. Photographs taken during the match. a) Yellow flares were used on the North stand at 20:48. b) Red flares and smoke bombs used on the South stand at 21:48. c) Green flares used on the North stand at 22:09. d) At some times, the entire stadium was filled with smoke from the flares and smoke bombs. The photograph was taken at 21:54. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

concentration (TC, number of NPs/cm^3 = #/cm^3) (b), versus time.

In Fig. 2 a), the 2D distribution colour plot shows that the majority of the detected nanoparticles have diameters between 20 nm and 200 nm. The periods when the torches were lit (at 81 min, 141 min, and 162 min) are clearly visible from the size distribution plot as the signal increases from background levels of approximately 7.5 × 10^3 #/cm^3 to 2 × 10^4 #/cm^3 and higher, for particles 80 nm in diameter. The signal oversaturation (white) at around 140 min and 160 min, i.e. in time periods of the highest NPs concentration, is presented in an appropriate contrast for other important events during the match to be visible. An increase of more than 1800% of the normalized concentration value, reaching 1.4 × 10^5 #/cm^3 for particles 155 nm in diameter, was measured at 165 min.

The background signal was measured for 63 min before the match. The TC, shown in Fig. 2 b), was 5400–6300 #/cm^3, with the average value of 5700 #/cm^3. As previously mentioned, the background pollution originated from of sources due to the nearby traffic, heating (time-independent on the time scale of the event), and cigarette smoke. One increase in TC was detected approximately 10 min before the match started, as visible from Fig. 2 b)-label A, with the TC returning to the background level within minutes.

The first set of hand flares was lit 81 min after the measurement started and reached the maximal NP concentration in the subsequent measurement 3 min later (label B). The TC increased for 200% compared to the background levels. After this event, the TC was decreasing back to the background level. Nevertheless, a slight increase in TC (15%) was detected during the half time (label C), likely caused by cigarette smoke. The largest peaks, at 143 min (label D) and 165 min (label E), followed two hand flare lighting events. The first one, where the flares were lit from both supporters' groups, raised TC levels 5.5 times (550%), while the increase from the second one was 12-fold (1200%). This last event occurred 12 min before the end of the match, and TC started to continuously decrease, reaching the background level only 10 min after the end of the match.

Fig. 3 a) shows the contributions of NPs of selected diameters (50, 100, 200, 400, and 575 nm) to the size distribution of all NPs. The largest contribution came from 200 nm to 100 nm particles. Concentration of 50 nm particles steadily decreased with time but was raised for 4 and 4.5 times, at 162 and 168 min, following the events D and E. These small particles reached the detector with a delay after the burning

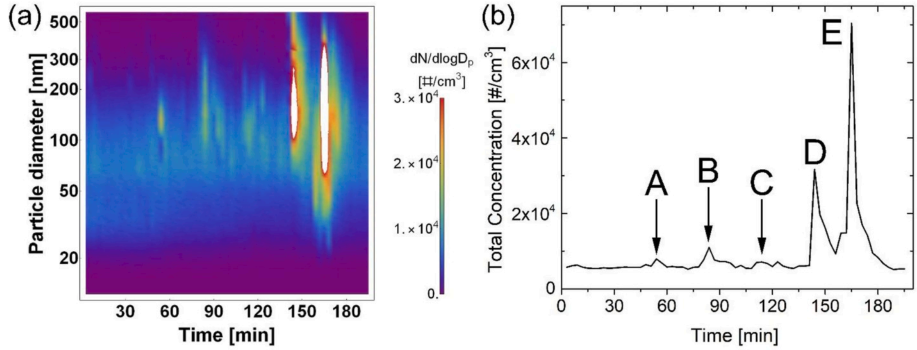

Fig. 2. (a) Normalized concentration (dN/dlogD$_p$ [#/cm^3]) of NPs given as diameter in log scale vs time; (b) Total NP concentration (TC) before (0–60 min), during (63–174 min), and after (177–195 min) the football match. Labels A-E denote the peaks in air pollution.

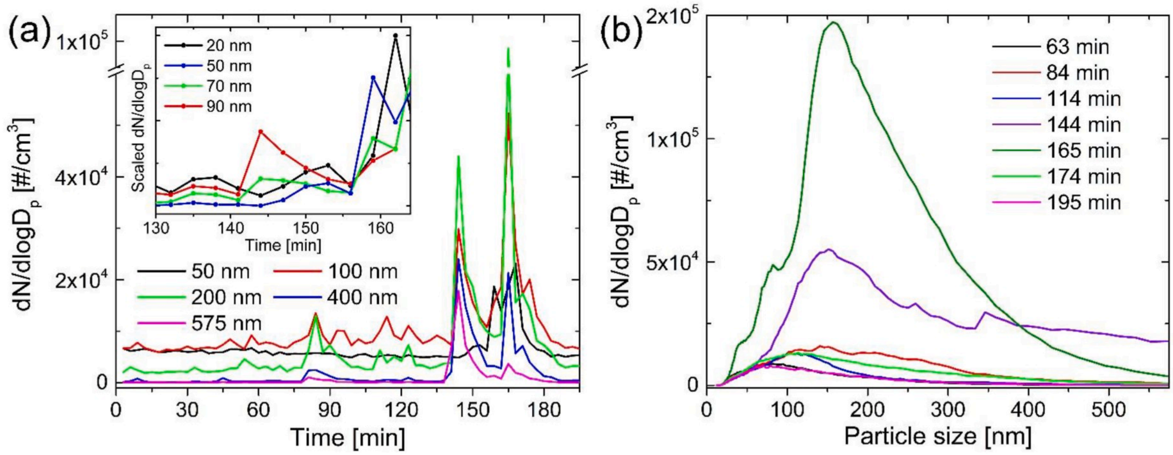

Fig. 3. (a) Contributions to the total size distribution due to particles of 50 nm, 100 nm, 200 nm, 400 nm, and 575 nm in diameter. (b) Normalized concentration spectra vs. particle diameter for selected time points corresponding to important events during the football match. Inset in (a) the scaled normalized concentrations of nanoparticles 20 nm, 50 nm, 70 nm, and 90 nm in diameter (the whole spectra was normalized to the interval [0,1]) vs. time for the last two flare burning events.

events. As shown in inset of Fig. 3 a), the delay in the arrival for the event D was 18 and 15 min for 20 nm and 50 nm particles, respectively. The delay for the event E was 6 min. For sizes larger than 100 nm, a peak at 84 min appeared, corresponding to the event B. The peaks corresponding to the last two flare burning events, D and E, appeared at the first subsequent measurements, at 144 and 165 min. Fig. 3 b) depicts the particle size distribution for some selected time points during the match listed in Table 1. The lowest normalized concentration was at 63 min and 195 min, corresponding to the last measurement before the match started and the very last measurement. These two measurements appear to be almost identical, with a broad peak situated around 80 nm. The first flare burning caused the peak at 84 min, which resulted in a very broad particle size distribution in the range from 30 nm to 350 nm. During the halftime, represented by the 114 min line, the size distribution became narrower with the peak maximum at 110 nm. The flares burning at 144 min resulted in a substantial increase of the overall signal, with one broad peak at 150 nm, and significant number of large-diameter particles. The lighting of the last set of pyrotechnic articles at 162 min resulted in the maximal signal at 165 min, with the largest contribution of particles, 155 nm in diameter and substantial contribution of smaller particles, visible as two small bumps at 40 and 80 nm seen also in Fig. 2 a) and 3 a). Pyrotechnic events are usually associated with an increase in the nanoparticle concentration, which is accompanied with a decrease in the mean particle size as most of the celebrations that are presented in the literature take place in big cities where the air is usually polluted with accumulation mode particles due to traffic, wood burning and industry (Yadav et al., 2019). In our case, the background mean particle size was below 100 nm due to relatively clean air, so during the pyrotechnic event the mean particle size increased. The match ending at 174 min resulted in a dramatic decrease of air pollution, with the broad maximum in the size distribution around 100 nm in diameter.

All three events where pyrotechnic articles were used resulted in the release of large number of nanoparticles with diameters smaller than 200 nm as well as large amounts of nanoparticles with bigger diameter, which were formed in the air due to agglomeration. On the contrary, the nanoparticles released by human behaviour at the start, halftime, and the end of the match, were on average smaller than 100 nm. This pollution most likely occurred due to cigarette smoke and dust raising from the spectators' movement. Only 3 min after the match ended, the TC lowered to the double of that of initial background, while after 15 min, it became approximately the same as at the beginning of the measurements.

Considering the experimental results, we may estimate the cumulative dose of particles the players were exposed to during the match. Following the methodology in ref. (Slezakova et al., 2019), the dose can be calculated as

$$D = \frac{BR \times C \times t}{BW},$$

where *BR* is the breathing rate, *BW* is the body weight, *C* is the median concentration of the particles, and *t* is the exposure time. The *BR* value depends on the age and the type of activity – it is higher for more intense physical activities. If we consider an average football player who is between 20 and 30 years old, weights 77 kg, and is vigorously physically active during the match, we can estimate the cumulative dose of 6.8×10^8 # kg^{-1}, which is around 300% larger than the dose one would get in a low-pollution environment (Slezakova et al., 2019).

According to the automatic monitoring station, the average background PM_{10} concentration was 24 μg/m^3, which is lower than the average annual value for Ljubljana in 2018, 35 μm/m^3 (Dolšak and Gorjup, 2019). The values of SO_2 NO, NO_2, NO_x, and CO were also lower than the annual average.

3.3. Chemical analysis

The shape and chemical composition of the collected particles were analysed by means of SEM and EDS. The samples were collected from the DLPI stages with nominal cut diameters D_{50} from 29 nm to 4 μm. The stages with the cut diameters of 6.8 μm and 10 μm collected negligible amount of particles.

In Fig. 4, representative SEM images of all DLPI stages 1, 4, 6, and 8 are shown. The sizes of the collected particles range from a few micrometres down to few tens of nanometres. Most of the organic matter is composed of carbon nanoparticles with a diameter of a few 10 nm. Interestingly, the chemical compositions of the samples collected at different stages do not vary significantly. Small particles are found on all DLPI stages, sometimes agglomerated. Some of the large particles, most of them having irregular shapes, were found in a cuboid or micro/nanowire shape, with particle diameters in the micrometre range. As they appear in stages where only nanometre-scale particles are collected, we conclude that these structures do not originate from the outside sources but have instead grown in situ on the stages. It is likely that the metal halide salts, such as NaCl, $MgCl_2$, KCl, and other water-soluble compounds found in pyrotechnics, were dissolved in air moisture and recrystallized inside the DLPI during the measurement.

Based on the EDS analysis, one can see that most of the material is composed of carbon-based compounds (Tables 2 and 3). Carbon is

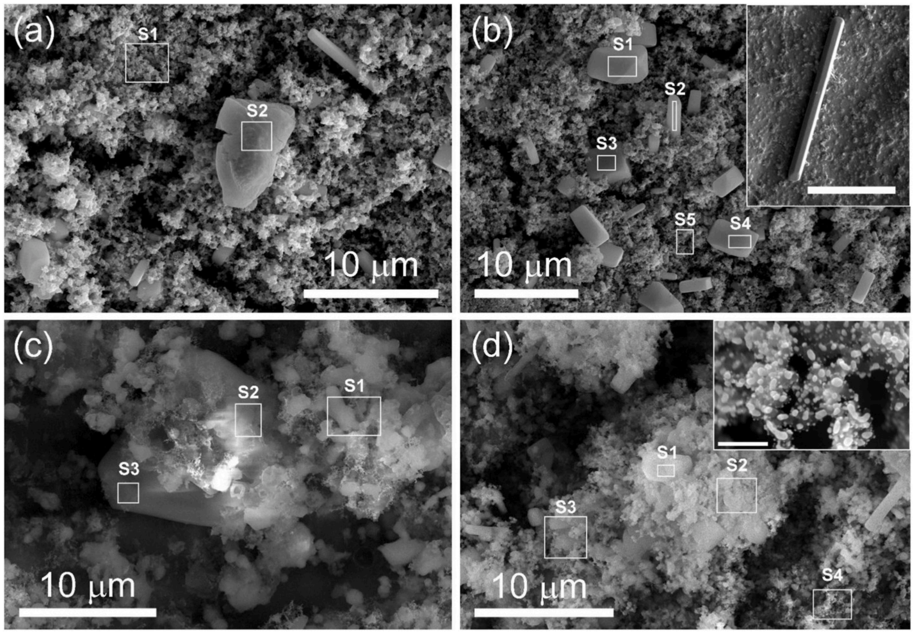

Fig. 4. Representative SEM images of the DLPI stages with white squares marking the regions of EDS analysis: a) – b) the stage 4. The inset in b): a micro wire consisting of potassium, nitrogen, and oxygen from stage 1. The scale bar: 5 μm; c) stage 8; d) stage 6. Inset: carbon-rich nanoparticles with diameters of a few 10 nm. The inset scale bar: 200 nm.

always present in the air due to the exhaust from cars and wood combustion and is also used in pyrotechnics as a fuel in the form of charcoal (Cao et al., 2017; Patrón et al., 2017). Metals such as sodium, magnesium, potassium, calcium, strontium, and barium were found scattered thorough all samples in combination with oxygen and chlorine (Tables 2 and 3). Metal halide salts, metal oxides, and carbonates were likely formed during the exothermic reaction in the pyrotechnic device where the oxidants (oxygen, potassium nitrate, potassium perchlorate, barium nitrate) react with fuels (charcoal, sulphur, resins), colouring agents (sodium nitrate (yellow), strontium carbonate (red), copper oxide (blue), barium nitrate (green), calcium carbonate (orange)) and colour enhancing agents (PVC, Saran) (Cao et al., 2017). The oxidants and colouring agents explain the origin of detected metals and oxygen, while chlorine arises from the colour enhancing agents. Small concentrations of fluorine were also detected, indicating a formation of metal halide salts with fluorine. The source of fluorine can be the fluoropolymeric pyrotechnic casing or colour enhancing agents (Nauflett et al., 1999). Micrometre-sized particles of KNO_3, KHS, and KCl were observed, as shown in Figs. 5–7. These particles usually have almost cuboid shapes typical for their crystal symmetry. The content of aluminium could not be determined because the substrate on which the particles are collected is made of aluminium foil. A prior analysis of the aluminium foil demonstrated that the foil also contains small amounts of iron.

Silicon, phosphorous, sulphur, copper, and iron were detected in minor concentrations. The source of these elements could be the fuel (sulphur), colouring agents (copper), or some additives to produce sparks (iron) and to enhance the pyrotechnic device performance (silicon, phosphorous) (Cao et al., 2017). Mapping of a flake identified as potassium hydrosulphide is shown in Fig. 6, and of a cube identified as potassium chloride in Fig. 7.

Table 3 shows the presence of different elements in each DLPI stage. For each DLPI stage an area of 100 × 100 μm^2 was selected on the deposited material where the EDS analysis was performed. Additionally, point spectra and mappings were performed at higher magnification. All the elements that were detected are presented in Table 3. Carbon, oxygen, and potassium are present on all stages. Most of the samples also contain sodium, magnesium, sulphur, and chlorine. Silicon and barium are also commonly found while the other elements listed in Table 3 are found only in a few samples.

Particles in the shape of microwires were identified as potassium nitrate, as seen in Fig. 5. The source of these particles could be potassium nitrate used as an oxidant.

Table 2
EDS analysis of different DLPI stages. All values are in atomic %.[a] Spec. relates to the sampling sites, for example, a-S1 refers to Fig. 4 (a) spectrum S1.

Spec.[a]	a-S1	a-S2	b-S1	b-S2	b-S3	b-S4	b-S5	c-S1	c-S2	c-S3	d-S1	d-S2	d-S3	d-S4
C	78.65	77.02	56.21	71.28	59.20	53.92	77.63	58.03	49.49	37.35	41.26	42.28	47.96	57.33
O	11.14	16.25	4.58	9.15	5.57	6.53	10.42	20.98	12.66	36.10	41.24	41.66	25.45	17.83
F											0.66	1.00	0.61	0.66
Na	0.26	0.10					0.21	1.22	17.95	0.59	0.32	0.46	0.85	0.92
Mg	1.34	0.24	0.30	0.58	0.23	0.39	1.79	2.07	0.90	2.61	5.11	8.82	7.06	4.53
Si	0.22	0.26					0.28	0.21	0.15					0.37
P								0.18		0.18				
S	0.57						0.80	0.19		0.17		0.25	0.41	0.41
Cl	3.63	0.37	18.83	9.08	16.93	19.14	4.38	2.60	14.80	0.67	5.13	2.01	5.52	4.44
K	3.35	0.31	19.67	9.49	17.75	19.65	3.77	1.64	2.22	15.93	4.90	1.32	4.05	4.25
Cu													0.15	0.16
Sr	0.18						0.22	0.20			0.17	0.64	0.51	0.24
Ba	0.16							0.24	0.05	0.08	0.15	0.47	0.89	0.69

Table 3
Elements found with EDS in different DLPI stages: x marks the presence of an element.

Element	D50 [mm]	C	N	O	F	Na	Mg	Si	P	S	Cl	K	Ca	Cu	Sr	Ba
Stage																
1	0.029	x	x	x								x				
2	0.057	x	x	x			x			x		x				
3	0.097	x		x		x	x			x	x	x				
4	0.163	x		x		x	x	x		x	x	x			x	x
5	0.274	x		x		x	x	x		x	x	x			x	x
6	0.397	x		x	x	x	x	x		x	x	x		x	x	x
7	0.633	x		x	x	x	x			x	x	x		x	x	x
8	0.98	x		x		x	x	x	x	x	x	x		x	x	x
9	1.64	x	x	x		x	x	x	x	x	x	x	x			x
10	2.45	x	x	x		x	x	x	x	x	x	x	x			x
11	4.08	x	x	x		x	x	x	x	x	x	x	x			x

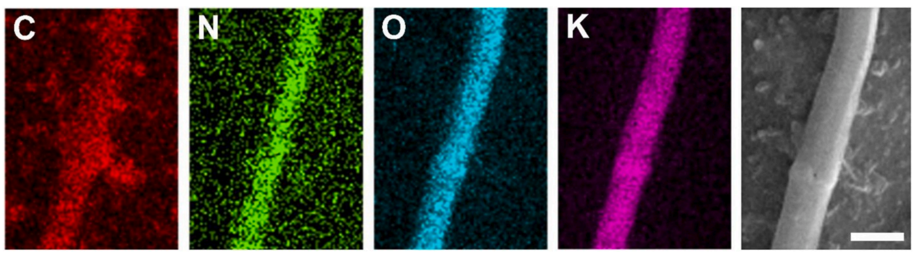

Fig. 5. SEM image and EDS mapping of a micro-wire. Associated EDS maps of selected elements (C (red), N (light green), O (blue) and K (purple)) are compared with the SEM image of the mapping area (the far right image). The scale bar is 1 μm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

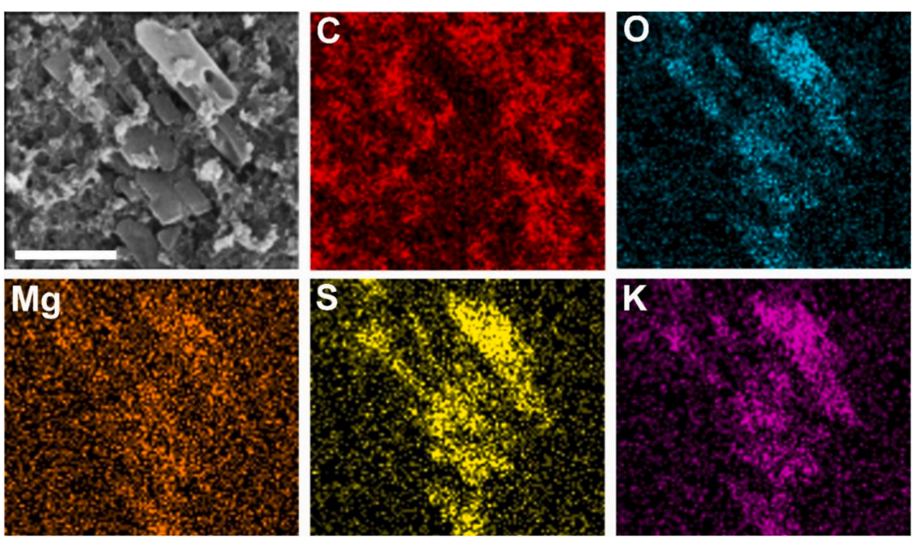

Fig. 6. SEM image and EDS mapping of a potassium hydrosulphide flake: C (red), O (blue), Mg (orange), S (yellow) and K (purple)). The scale bar: 2.5 μm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

4. Discussion

We discuss the results of the measurements in view of similar experiments conducted during pyrotechnic events. In our study, as seen in Fig. 2 a), the majority of the particles have the diameter in the range between 20 and 200 nm. This is consistent with the distributions observed in other studies (Zhang et al., 2019; Joshi et al., 2016). This size interval overlaps with the so-called Aitken mode particles (extending from 10 to 100 nm in diameter) and the accumulation mode particles (extending from 100 to 300 nm in diameter). Aitken mode particles in our case are formed during the condensation of hot vapours in combustion processes. These particles subsequently act as the nuclei for the condensation of low-vapour pressure gaseous species, causing them to grow. The lifetime of these particles is short, as they coagulate into accumulation mode particles which can stay in the atmosphere for longer periods of time.

Looking at the time it took the particles to reach the detector (Fig. 3), we found out that the larger particles arrived first. This is just the opposite from what was previously reported in the literature for fireworks (Yadav et al., 2019). The delayed detection of sub-100 nm particles in our case could be attributed to their slow mobility along a certain direction, as a consequence of scattering with air molecules and other NPs and the shape and size of the stadium, where the particles were confined. Usually fireworks take place high above the ground where the atmospheric conditions are different than those in a stadium, affecting the particle mobility. A different setting also resulted in a quick

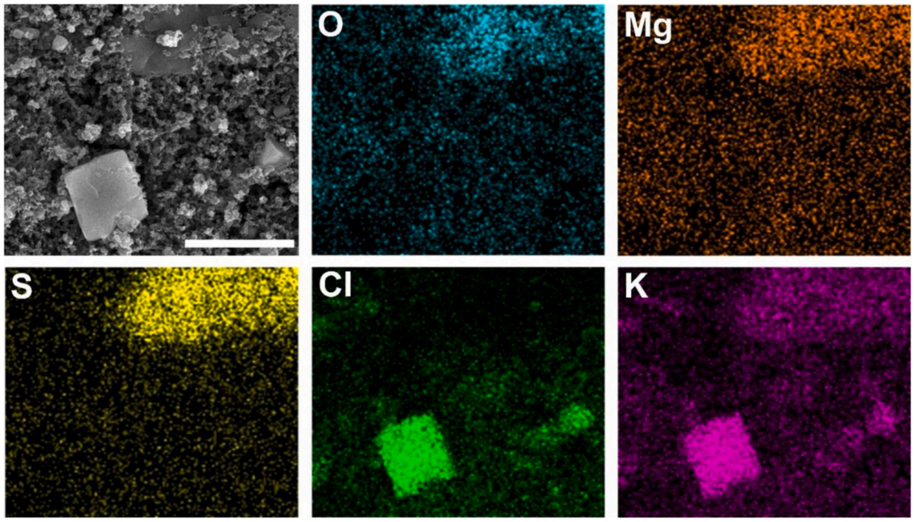

Fig. 7. SEM image and EDS mapping of a KCl particle: O (blue), Mg (orange), S (yellow), Cl (green) and K (purple). The scale bar is 5 μm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

decrease in concentration after the end of the event, as the concentration returned to background levels within 15 min. In fireworks, on other hand, the particle concentrations can remain high even hours after the event (Drewnick et al., 2006).

The chemical composition of the collected sample was consistent with the expectations – a large share of amorphous carbon is attributed to the fuel in the pyrotechnics (charcoal), while the confirmed elements correspond to the colouring agents, colour enhancing agents, and other components of the pyrotechnic devices such as the casing (Cao et al., 2017). It was shown (Croteau et al., 2010) that emission factors for metals, such as K, Mg, Cu, and Ba, commonly found in ground-level pyrotechnics, exceeded occupation exposure guidelines (ACGIH, 2016).

Since this case study was a one-off event, we can only discuss the health effects through a comparison with related studies of inhaling particulate matter. While most studies focus on health effects of a long-term exposure, certain studies look at acute exposure as well, with a special focus on sport performances, in scenarios which resemble the situation on the stadium. There are two important factors influencing the inhalation exposure at sport activities. The increased ventilation rate may lead up to a 6- to 10- fold increase in number of particles deposited in the airways (Qiu et al., 2019), and changes the way of breathing from nasal to mouth, bypassing the nasal filtration system that would otherwise reduce the lung exposure (Slezakova et al., 2019; Giles and Koehle, 2014; Brocherie et al., 2015).

The majority of the studies concluded that the benefits of practicing outdoor sport activities in a low-pollution environment will far exceed the adverse effects of air pollution for general population (Slezakova et al., 2019; Giles and Koehle, 2014; Qin et al., 2018). The conclusions are different for a high-pollution environment, where exercise could impair health functions and athletic performance (Cutrufello et al., 2011). In a study where the participating athletes were inhaling the PM exhaust from a gasoline engine during intense exercise, their performance was lower than in the control group. The effect was attributed partially to impaired vasodilation in the peripheral vasculature (Cutrufello et al., 2011). In a study of people cycling near a major bypass road, the percentage of blood neutrophils increased significantly 30 min after exercise (Jacobs et al., 2010). In a recent meta-study (Qin et al., 2018), nearly two thirds of the analysed studies showed that the combination of exercise and air pollution could impair health function and athletic performance, as well as increase the incidence of cardiopulmonary deterioration (Cutrufello et al., 2011; Rundell and Caviston, 2008).

While the nanoparticle dose received by the spectators during the event may be small in comparison to the dose accumulated by living in an urban environment with pollution from traffic and other anthropogenic sources, the studies of reduced performance of athletes is of higher concern, as fractional differences in performance are often crucial for winning or losing the match. In addition to football players, the accompanying personnel also receives larger doses than one-off spectators as they are present at large number of matches throughout the season. Furthermore, the composition of nanoparticles generated by pyrotechnics differs from that generated by heating or traffic as there are more heavy metals present, coming from colouring agents. Some metallic nanoparticles, such as Cu and Sr detected here, are considered to be an environmental hazard. Studies showed that they are potentially toxic, and can remain in the environment for a long time, whereby they can pose as a health threat for prolonged period of time (Yang et al., 2019; Pongpiachan et al., 2018). Opposite to the wishes of the ultras to support their team with fireworks, they are actually hampering their performance. Further research on this topic is thus required.

5. Conclusion

Air pollution was measured during a football match, when fans, especially members of the ultras groups, were burning hand flares and other pyrotechnic articles to support their teams. Time-dependent analysis of size distribution of particles in local atmosphere has evidenced a substantial release of fine and ultrafine particles with up to 200 nm in diameter. When flares were lit from both supporters' groups, the total concentration of particles initially raised 5.5 times (550%), while the second such event caused even 12-fold (1200%) increase in air pollution by particulate matter. Chemical analysis of the samples collected by the low-pressure impactor unveiled that most of the particulate matter consisted of amorphous carbon, elements linked to the oxidizing agents, fuel, and colour-enhancing agents typical for commercial and/or home-made pyrotechnic articles used by football team fans, especially the ultras groups. Some of the detected metals pose an environmental hazard. Air circulation in open-air stadium caused the concentration of particles to halve in approximately 3 min and to return to near-background levels in about 15 min.

As the experimental equipment was positioned roughly in the middle between both stands with the support groups, the results can be viewed as a representative exposure to air pollution a general spectator is exposed to, however, the concentrations and exposure were clearly significantly higher in close proximity to the pyrotechnics. The estimated cumulative dose the players inhaled during the match was about 300% higher than the dose one would get in a low-pollution environment. In view of related work, the acute exposure to nanoparticles may hamper the performance of elite athletes, which is highly relevant when it comes to competitive sport. Simultaneous measurements at different locations at the stadium, which would improve the assessment of immediate and long-term effects of exposure on spectators and players, falls under the scope of future work.

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.

CRediT authorship contribution statement

Luka Pirker: Formal analysis, Writing - original draft. **Anton Gradišek:** Writing - original draft. **Bojana Višić:** Formal analysis, Writing - original draft. **Maja Remškar:** Writing - original draft.

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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.2020.117567.

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