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Characterization of organic aromatic compounds in soils affected by an uncontrolled tire landfill fire through the use of comprehensive two-dimensional gas chromatography–time-of-flight mass spectrometry

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ABSTRACT

Discarded vehicle tires have become an increasing concern worldwide due to the enormous amount of wastes generated and the increasing evidence of health problems associated to their disposal and accidental combustion. Previous studies conducted involving either simulated or open uncontrolled tire fires have identified aromatics belonging to two main classes, volatile organic compounds and polycyclic aromatic compounds (PAHs), as the most relevant chemicals generated in these burning processes. As a consequence, and due to their recognized toxicity, most studies reported up to now have mainly focused on these two categories of compounds being information concerning the possible occurrence of other aromatic classes rather limited.

In this study, the enhanced separation power and structural confirmation capabilities provided by comprehensive two-dimensional gas chromatography–time-of-flight mass spectrometry (GC × GC – ToF MS) has been used, for the first time, for the non-targeted analysis of soils impacted by a tire fire and an ash collected at the scene of the fire. In total, 118 volatile and semi-volatile aromatic compounds have been differentiated. Among them, 104 compounds have been either positively or tentatively identified. PAHs with 3–5 rings and their alkyl-derivatives were the most numerous and relevant classes in the investigated samples. A significant number of sulfur, oxygen- and nitrogen-containing PAHs were also detected in the samples. The application of a script function to the raw GC × GC – ToFMS data allowed the fast filtering and automatic recognition of compounds containing halogens in their structure. This part of the study evidenced that only a limited number of regulated persistent organic pollutants were present in the investigated samples. However, it also revealed the presence of emerging organophosphorous flame retardants, whose levels in tire fire impacted soils are reported for the first time.

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1. Introduction

The disposal of discarded vehicle tires is nowadays recognized as a growing problem and cause of concern worldwide. The annual global production of tires was estimated to overpass 1.5 billion units in 2015, while that of used tires approached 17 million tons per year. At present, in the European Union (EU), 355 millions of

tires are manufactured per year. In the same time period, 3.2 million tons of used tires are generated (data for the EU including also Norway and Switzerland). The European Tyre and Rubber manufacturers' association (ETRMA) estimates that 78% of these tires (corresponding to around 250 million units) are either recycled or recovered [1,2]. However, despite these efforts, a significantly increasing number of used tires are discarded in landfills or illegal dumps. In the EU, dumping of used tires in landfills is specifically forbidden by the 1999 EU directive on Landfill of Waste [3] due to their potential to contaminate the surrounding environment, in particular surface water, groundwater, soil, air, and because of the negative effects on human health [4].

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Tires composition varies depending on its use, i.e. the type of vehicle for which they were manufactured, but in general they can be considered complex cross-linked structures of natural and synthetic rubbers that contain different types of carbon black, fillers such as clay and silica, and chemicals and minerals added to allow or accelerate vulcanization [5]. This composition made tires resistant to chemical-, biological- and photo-degradation, as well as resistant to high temperatures. These features, highly appreciated during tires life-time, made them also difficult to recycle. Recycling alternatives include material recovering by grinding or shredding into new products, use in civil engineering applications (e.g., roads, railways, and landfill construction); and energy recovery by combustion or pyrolysis at high temperatures [1,6]. However, most of these processes are considered laborious, difficult and, in general, rather expensive [4]. They also required specific equipment that is not available in all locations. This situation made that, in many cases, discarded tires were simply stockpiled or illegally dumped. The increasing number of tire stockpiles and illegal dumps worldwide is also raising the occurrence and magnitude of uncontrolled tires fires. The huge amount of heat generated in these fires make them difficult to extinguish, so they can keep active for days [7] and even months [4,8]. Previous studies conducted on real open tire fires [7,9,10] or on simulated scrap tire fires [4] have evidenced that a variety of hazardous organic and inorganic compounds can be generated during these combustion processes. The nature and amount of chemicals generated depend on the tire quantity and typology, and on the combustion conditions [4,7,9,10]. In open fires, the emitted compounds can then be dispersed in the surrounding environment through the atmosphere, ground waters and soil. However, their capacity to disperse (and so contaminate) the environment is affected by a number of factors, including the topography of the fire environment, the meteorological conditions and the approach used to extinguish the fire [4,9].

The organic compounds identified in tire fires, both under open uncontrolled burning conditions and on small-scale simulated experiments, are primarily aromatics belonging to two main classes, volatile organic compounds (VOCs) and polycyclic aromatic compounds (PAHs) [4]. As a consequence, and due to the accepted hazardous nature of some of these analytes, these two families of compounds have received most of (in not exclusive) attention in previous studies reported in the literature [7,10]. Wang et al. [9] identified 165 PAH compounds in smoke wipe samples collected from glass and metal surfaces exposed to the smoke of a fire generated in a commercial tire products outlet facility using GC–quadrupole MS (GC–qMS) operated in both the scan and single ion monitoring (SIM) mode for compound identification and quantitation, respectively. The largest concentrations corresponded to the EPA priority PAHs and other pyrogenic PAHs (in particular, the 4–7 rings PAH isomers), while alkylated PAHs were detected at significantly lower levels. These authors also reported on the presence of a number of O-, S- and N-containing PAHs (O-PAHs, PASHs and N-PAHs) in the investigated wipe samples. Essentially similar findings were reported by Downard et al. [7] in a more recent study concerning a much larger uncontrolled open-burning of shredded tire in a landfill, where 4–5 rings PAHs were also identified as the most abundant PAHs. In that study, which also involved GC–qMS for the analytical determination, the presence of O-PAHs and N-PAHs was also reported. However, data concerning the possible presence of PASHs was missing.

Due to the target approaches used in the previously mentioned studies, limited data concerning the presence of other regulated aromatic persistent organic pollutants (POPs), such as polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) and polychlorinated biphenyls (PCBs), in uncontrolled open tire fires is available in the literature [11]. To the best of our knowledge, no data regarding on the possible formation of toxic polybrominated diphenyl

ethers (PBDEs) and emerging flame retardants/pollutants, such as halogenated norbornenes and organophosphorus flame retardants (OPFRs), have been published up to now.

Comprehensive two-dimensional gas chromatography (GC × GC) is recognized worldwide as a powerful separation technique [12]. Due to its enhanced resolution power and improved sensitivity, GC × GC is particularly suitable for the characterization of complex mixtures, especially when used in combination with a detector providing structural information, such as a mass spectrometer. However, up to now, the feasibility of this technique for the non-targeted characterization of this type of complex fire affected mixtures has not been evaluated.

On 14 May 2016, an uncontrolled fire took place in the largest illegal tire landfill in Spain and one of the largest in Europe. The landfill had an extension of 117,000 m² and was located in the border of two Spanish Autonomous Communities (Castilla-La Mancha, 70%) and Madrid (30%), and close to two important population nuclei, Seseña (Toledo) and Valdemoro (Madrid). The landfill was used for the illegal dumping of discarded tires since 2002 and declared illegal in September 2015. When the fire started, it was estimated that 70,000–90,000 tons of tires were accumulated in the landfill. On 2 June, the fire was declared as extinguished in the zone of Castilla-La Mancha. Five days later, it was considered completely extinguished in the zone of Madrid. At that moment, only 15,000 tons of tires remained unburnt [13]. To our knowledge, up to now, only a preliminary study on the levels of selected toxic metals, the 16 EPA PAHs, and the toxic dioxin-like PCBs and 2,3,7,8-PCDD/Fs in soils collected in the Seseña area a few days after fire extinction has been reported in the literature [13]. In that paper, relative low levels of PCBs and PCDD/Fs were detected as compared to those reported by other authors in areas impacted by tire fires [11]. In fact, the authors concluded that, among the investigated chemicals, only PAHs showed levels high enough to be considered cause of concern for the inhabitants of the closest urbanizations.

This study evaluates the potential of GC × GC combined with time-of-flight mass spectrometry (GC × GC – ToF MS) for the non-targeted characterization of the volatile and semi-volatile aromatic compounds present in seven soils collected in the surrounding tire fire area of Seseña at the beginning and after the burning episode. A sample of ash was also investigated. Analytes detected in these samples were identified and their abundances and profiles compared to tentatively assess their possible variation with the distance to the fire and the burning time. Apart from PAHs and VOCs, special attention was paid to the identification of halogen-containing aromatic compounds. Levels of halogenated flame retardants, including PBDEs, halogenated norbornenes, and OPFRs in these types of samples were reported for the first time.

2. Materials and methods

2.1. Standards and reagents

PAH Mix 9 (US EPA 16), containing the 16 EPA priority PAHs in acetonitrile, and PAH mix 183, containing the 15 + 1 EU PAHs in isooctane, were acquired from Dr. Ehrenstorfer Laboratories (Augsburg, Germany). Method 1614 Surrogate Stock Solution (PAR Solution) containing tri-BDE-28, tetra-BDE-47, penta-BDE-99, pentaBDE-100, hexa-BDE-153, hexa-BDE-154, hepta-BDE-183, and deca-BDE-209 were purchased from Wellington Laboratories Inc. (Guelph, ON, Canada). Method 1614 Labeled Surrogate Stock Solution containing the mass labeled PBDEs (¹³C-BDE-28, ¹³C BDE-47, ¹³C BDE-99, ¹³C BDE-100, ¹³C BDE-154, ¹³C BDE-153, ¹³C BDE-183, and ¹³C BDE-209) was purchased from Cambridge Isotope Laboratories Inc. (Andover, MA). *Syn*- and *anti*-isomers of dechlorane plus (DP) were purchased from Wellington Laboratories Inc., Dec

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