



Nano-litter from cigarette butts: Environmental implications and urgent consideration



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HIGHLIGHTS

- Cigarette butts (CGB) are the most common litter found in the environment.
- This study presents new evidence of nanoscale particles (NP) leached from CGB.
- NP contain a large part of the volume of metal released from CGB.
- Our results raised urgent questions on CGB litter and their environmental impacts.

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ABSTRACT

Cigarette butts (CGB) are equivalent to plastic litter in terms of number of pieces released directly into the environment. Due to their small size and social use, CGB are commonly found in natural systems, and several questions have been raised concerning the contaminants that are released with CGB, including metals, organic species, and nanoparticles. The aim of the present study is to investigate the release of nanoscale particles from CGB by leaching with rainwater. After seven days of passive stirring of both smoked and unsmoked CGB in synthetic rainwater, the solutions were treated and analyzed by specific nano-analytical methods. Our results demonstrate the release of $4.12 \pm 0.24\%$ (w/CGB) organic carbon in the range of 10 nm up to 400 nm and with a z-average diameter of 202.4 ± 74.1 nm. The fractal dimension (D_f) of the nanoscale particles ranges from 1.14 to 1.52 and suggests a soot (carbon)-based composition. The analysis of some metallic species (As, Pb, Cd, Cu, Ni, Cr, Co, Al, Mn, Zn, and Fe) shows that these species are essentially attached to the nanoscale particles per gram of carbon released. By considering the diffusion of the nanomaterials into different environmental compartments, our results suggest a new emerging and global contamination of the environment by cigarette butts, comparable to plastic litter, which urgently needs to be considered.

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1. Introduction

The principle litter associated with the consumption of cigarettes are smoke and cigarette butts (CGB), i.e., filter compounds and cigarette smoke (Healton et al., 2011; Marah and Novotny, 2011; Novotny et al., 2009). While the CO₂ contribution of cigarette smoke can be considered negligible compared to that emitted by other human activities, CGB, like plastic waste, raise many

environmental issues. CGB are the most common form of waste found in nature, and over 5.6 trillion cigarette butts can be found in nature each year (Healton et al., 2011). Moreover, CGB found in the environment are likely to release their components to the environment and pollute soils, water and all other environmental compartments. Several studies have shown that several hazardous pollutants are released and leached from CGB, which can have dramatic effects on the environmental and public health (Novotny et al., 2009). Moerman and Potts highlighted the release of several metals from CGB (Cd, As, Ni, Cu, Pb and Zn) with quantities ranging from few $\mu\text{g g}^{-1}$ up to several hundred $\mu\text{g g}^{-1}$ (Moerman and Potts, 2011), which has also been confirmed elsewhere (Dobaradaran

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et al., 2017; Pelit et al., 2013). However, to the best of our knowledge, no data are currently available concerning both their release and transport mechanisms. Recently, a new class of nanoscale contaminants have been considered classified as accidental nanoparticles from anthropogenic source such as nanoplastics (Koelmans et al., 2015; Ter Halle et al., 2017). Due to the recent and emerging risk of nanoparticles (Baalousha et al., 2016; Bystrzejewska-Piotrowska et al., 2009), questions have been raised about these new nanoscale contaminants and their possible release from CGB. Recently, authors have measured the air transfer of soot nanoparticles with sizes under 50 nm from the partial combustion of cigarettes into the human pulmonary system (van Dijk et al., 2011). Based on this preliminary study, it is possible to imagine the presence of nanoscale particles concentrated in CGB that could potentially leach into the environment. According to the number of CGB released into the environment, there is a considerable lack of available data on the nanoscale risk associated with CGB. The main questions, therefore, are which nanoparticles (size, composition) are concentrated in CGB and whether these nanoparticles are released into the environment. Due to their intrinsic properties, such as size and surface area, nanoscale particles are known to transport a large quantity of adsorbed species, such as metals and organic contaminants, along different environmental compartments and in living organisms (Velzeboer et al., 2014; Wang et al., 2013). The main concern with nanoscale particles is their ability to penetrate different natural barriers to deliver the species adsorbed to their surface.

The aim of this paper is to demonstrate the release of nanoscale particles (NP) from cigarette butts by leaching with rainwater, followed by their chemical and physical characterization. Different high-resolution analytical techniques, such as in situ dynamic light scattering (IS-DLS) and asymmetrical flow field-flow fractionation coupled to light scattering detection (A4F-UV-SLS), were optimized for nanoscale particle characterization to measure key parameters such as size distribution, aggregation information, fractal dimension, and elemental composition. Our results demonstrate that a large distribution of NP sizes and structures are released. The metal distribution associated with the NP fraction raised several environmental concerns that urgently need to be considered. We hope that our results will help tobacco control agencies to consider this type of environmental and health exposure from cigarette litter.

2. Materials and methods

2.1. Sample preparation

A fresh synthetic rainwater (FSRW) sample was prepared according to Davies et al. (2004). Briefly, the rainwater has an ionic strength of $0.3 \times 10^{-3} \text{ mol L}^{-1}$, and the following components comprise 1 L, which was then diluted by a factor of 1000: NaNO_3 , 4.07 g; NaCl , 3.24 g; KCl , 0.35 g; $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 1.65 g; $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 2.98 g; and $(\text{NH}_4)_2\text{SO}_4$, 3.41 g. The resulting solution had a pH of 5.2. More than 12 kg of cigarette butts (CGB) were collected over one year after immediate consumption from three different places in Rennes (Brittany, France). CGB were collected directly after consumption and placed directly in a plastic container under vacuum into a fridge (4°C). No CGB were collected on the ground to avoid bias measurement in the leaching experiment. For a single experiment, four CGB were randomly chosen, cleaned of old tobacco and placed into a 15 mL conical vial (previously rinsed by HNO_3 10% (w/v) and DI water to remove any trace of HNO_3). Then, 10 mL of FSRW was added into the conical vial. Then, the vials were placed on a rotator stirring at 30 rpm under classical light conditions for 7 full days. Then, the leached solutions were filtered at $0.45 \mu\text{m}$ and stored in the fridge. A small fraction of the solutions was kept for

total metal and organic carbon analysis. The remaining volumes were concentrated and size-fractionated by an ultrafiltration cell (Amicon[®], Millipore, France) with a 10,000 Da (10 kDa) molecular weight cut-off (MWCO) membrane made of polyethersulfone (Nadir, Alting, Metz, France). The retentates were concentrated three times and rinsed by three equivalent volumes of FSRW. The same procedure was followed for unsmoked CGB (un-CGB). The CGB leaching experiments were repeated more than 5 times, and the final volume was concentrated more than 20 times and rinsed 3–4 times by FSRW. All experiments were repeated identically 6 times for each condition in order to obtain convenient statistical results.

2.2. Reagent

The employed reagents were of analytical grade. The reagents to prepare the synthetic rainwater and the HNO_3 used for the digestion procedure were of ultra-trace pure quality (Labbox, Rungis, France). All plastic and glassware were cleaned by soaking in a 10% (w/v) HNO_3 solution and then rinsed with DI water before use. All solutions were prepared using ultrapure water (MilliQ, 18.2 mΩ cm).

2.3. Elemental analysis

Total organic carbon analysis was performed using a TOC analyzer from Shimadzu (Paris, France). The metal concentrations were determined by ICP-MS using an Agilent Technologies 7700x instrument. The samples were pre-digested with HNO_3 to avoid any interference from organic carbon during the analysis. A flux of He was injected in a collision cell to remove interferences. Quantitative analyses were performed using a conventional external calibration procedure (seven external standard multi-element solutions, Inorganic Venture, USA). Rhodium–rhenium was added on-line as an internal standard at a concentration of 300 ppb to correct for instrumental drift and possible matrix effects. Calibration curves were calculated from the intensity ratios of the internal standard and the analyzed elements. The international geostandard SLRS-4 was used to control the accuracy and reproducibility of the measurement procedure. The instrumental error of metal analysis was below 3%. The metal concentrations in the blanks were lower than the detection limits, and thus, no correction was needed.

2.4. Nanoscale particle characterization

In the present case, IS-DLS was used to detect the presence of nanoscale particles in the aqueous system. In DLS, due to the Brownian motion of nanoscale particles, time-dependent fluctuations of the scattered light intensity are measured. These fluctuations allow the determination of the dynamics of the scattering particles (Burchard, 1983). The time-dependent fluctuations are transformed into the auto-correlation function (ACF), which for nanoscale particles in a solution, decays with a relaxation rate directly correlated to the diffusion coefficient D ($\text{m}^2 \text{s}^{-1}$) and therefore, the hydrodynamic size according to the Stokes-Einstein equation is:

$$D = \frac{k_B T}{3\pi\eta d_H}$$

where d_H is the hydrodynamic diameter, η is the viscosity of the mobile phase ($\text{kg m}^{-1} \text{s}^{-1}$), T is the temperature of the medium (at room temperature, 293 K) and k_B is the Boltzmann constant ($1.38 \times 10^{-23} \text{ kg m}^2 \text{s}^{-2} \text{ K}^{-1}$). The IS-DLS instrument used in this study is a Vasco Flex from Cordouan Technologies (Pessac, France) and

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