



Littered cigarette butts as a source of nicotine in urban waters



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SUMMARY

The effect of nicotine from littered cigarette butts on the quality of urban water resources has yet to be investigated. This two-part study addresses the spatial variation, seasonal dynamics and average residence time of littered cigarette butts in public space, as well as the release of nicotine from cigarette butts to run-off in urban areas during its residence time. Thereby, we tested two typical situations: release to standing water in a puddle and release during alternating rainfall and drying. The study took place in Berlin, Germany, a city which completely relies on its own water resources to meet its drinking water demand. Nine typical sites located in a central district, each divided into 20 plots were studied during five sampling periods between May 2012 and February 2013. The nicotine release from standardized cigarette butts prepared with a smoking machine was examined in batch and rainfall experiments.

Littered cigarette butts are unevenly distributed among both sites and plots. The average butt concentration was 2.7 m^{-2} (SD = 0.6 m^{-2} , $N = 862$); the maximum plot concentration was $48.8 \text{ butts m}^{-2}$. This heterogeneity is caused by preferential littering (gastronomy, entrances, bus stops), redistribution processes such as litter removal (gastronomy, shop owners), and the increased accumulation in plots protected from mechanized street sweeping (tree pits, bicycle stands). No significant seasonal variation of cigarette butt accumulation was observed. On average, cigarette butt accumulation is characterized by a 6 days cadence due to the rhythm and effectiveness of street sweeping (mean weekly butt accumulation rate = $0.18 \text{ m}^{-2} \text{ d}^{-1}$; SD = 0.15 m^{-1}). Once the butt is exposed to standing water, elution of nicotine occurs rapidly. Standardized butts released 7.3 mg g^{-1} nicotine in a batch experiment (equivalent to 2.5 mg L^{-1}), 50% of which occurred within the first 27 min. In the rainfall experiment, the cumulative nicotine release from fifteen consecutive precipitation events (each 1.4 mm) was 3.8 mg g^{-1} , with 47% during the first event. According to these results, one cigarette butt may contaminate an amount of 1000 L water to concentrations above the predicted no effect concentration (PNEC) of only $2.4 \times 10^{-3} \text{ mg L}^{-1}$ (Valcárcel et al., 2011). Given the continuous littering of cigarette butts, and the rapid release of nicotine, cigarette butts are assessed to be a relevant threat to the quality of urban waters and consequently to drinking water.

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1. Introduction

One of the most common elements of urban litter, a cigarette butt, contains a complex mixture of toxins, including the highly water-soluble nicotine. Extremely addictive, nicotine is consumed in massive amounts around the world: 20% of the population over 15 years of age smoke on average 16 cigarettes per day (Giovino et al., 2012). With 73% of the world's population over the age of 14 years (United Nations, online), worldwide an estimated 16 billion cigarettes are consumed daily. With an estimated urbanization

of 60% in 2030 (United Nations, online) about 10 billion cigarettes will then be smoked in urban areas every day.

Cigarette butts are the most commonly littered item in urban areas (Bator et al., 2011; Schultz et al., online), making up 22–46% of visible litter, as reported in numerous litter audits worldwide (Schneider et al., 2011; Seco Pon et al., 2012; Moriawaki et al., 2009; Schultz et al., online). Patel et al. (2013) observed that in cities, 76% of cigarettes smoked in public were littered, rather than disposed in appropriate receptacles. The butts are then transported by urban waters to other ecosystems such as coastal areas, where they are consistently the most numerous element of litter collected (International Coastal Clean-up, 2013; Ariza et al., 2008). The persistence of cigarette butt filters, made of non-biodegradable cellulose acetate is a concern for wildlife

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(Stanley et al., 1988). Cigarette butts are thus a concern in terms of its environmental, public health, social, and economic consequences (Marais and Armitage, 2004b; Pon et al., 2012; Schultz et al., 2013; Schneider et al., 2011).

Cigarette butts are not evenly distributed in the urban environment. Distribution of butts is linked to the location of sales and consumption. The highest concentrations in the US are associated with bars, convenience stores, liquor stores, cafés, gas stations, grocery stores, restaurants, and traffic signals (Marah and Novotny, 2011). A preference of cigarette smoking has been observed in conjunction with alcohol, coffee, meals, and during breaks, (Van Gucht et al., 2010). Cigarette butts are discarded at places of transition from an outdoor to an indoor environment where smoking is not tolerated, such as entrances to buildings, vehicles or public transportation stations, where the authors found a cigarette butt concentration of 102 m^{-2} in one puddle (see Fig. 1). Non-smoking legislation, in effect in Berlin since 2008, prohibits smoking in the indoor workplace, inside public transit stations and platforms, restaurants, and bars. Since then, it is more common to see an accumulation of smokers outside of restaurant entrances and offices. This phenomenon may well have caused an increase of littered cigarette butts to the urban environment.

The quality of urban run-off is linked to urban litter (Marais et al., 2004a), which in turn is recognized as a major threat to urban water quality (Heinzmann, 1998).

1.1. Nicotine toxicity

Cigarette butts contain a mixture of substances with toxic effects to organisms, most notably heavy metals, polycyclic aromatic compounds, ethyl phenol, and nicotine (Micevska et al., 2006; Moerman and Potts, 2011; Moriwaki et al., 2009). The human toxicological and health effects of nicotine have been extensively studied (Brčić Karačonji, 2005). Nicotine is easily absorbed through the skin, lung alveoli, small intestine, and urinary bladder. It passes easily through the placenta to the fetus.

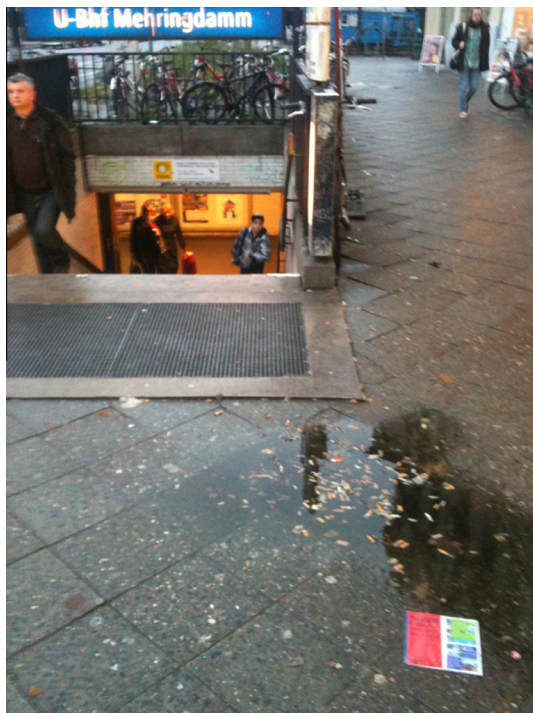


Fig. 1. Typical situation at the entrance of an underground station in detail. Cigarette butt count in the 1.44 m^2 area in the puddle tallied 151. (Photo: T. Nehls, taken on 27.11.2012 in Berlin, Germany).

Strong teratogenic and genotoxic effects have been observed. Nicotine is associated with cardiovascular disease and acts on the central nervous system. Acute toxicity causes death due to paralysis of respiratory muscles or respiratory failure. Nicotine is known to cause liver damage in fish (Konar, 1970), and effects planarians in a similar way as in mammals (Rawls et al., 2011). The effects of chronic sub-lethal concentrations of nicotine and its metabolites in the aquatic environment are not well understood. Based on the EC50 for *Daphnia* of 0.2 mg L^{-1} (Savino and Tanabe, 1989), the calculated value of the predicted no effect concentration (PNEC) for nicotine is $2.4 \times 10^{-3} \text{ mg L}^{-1}$ (Valcárcel et al., 2011).

1.2. Nicotine in water resources

Nicotine, and its most important metabolite, cotinine, are important emerging pollutants, widely detected in water resources worldwide in studies of pharmaceutically active compounds (Stuart et al., 2012; Benotti and Brownawell, 2007; Focazio et al., 2008; Valcárcel et al., 2011). Littered cigarette butts have not been examined to date as a source of nicotine in urban waters.

Nicotine has been in use as an insecticide since the 15th century. Because of its toxicity to aquatic organisms, its application has been severely limited in the US, Canada and Europe. Today, nicotine is on the toxic release inventory (TRI) in United States, thus its release into the environment must be reported to state authorities (Environmental Protection Agency, 1996). The European Union classifies tobacco waste as toxic and hazardous when the nicotine content exceeds 0.5 mg g^{-1} dry weight (Civilini et al., 1997). The nicotine content of cigarettes varies according to brand and country of consumption. Between 1998 and 2005, the mean concentration of nicotine in the tobacco rods sold by major manufacturers increased by 9% from 17.1 mg g^{-1} to 18.7 mg g^{-1} in the USA (Connolly et al., 2007).

Nicotine has been identified as one of the three most significant pharmaceuticals with regard to the potential eco-toxicological and toxicological impacts of waste-water treatment plant (WWTP) effluent (Muñoz et al., 2008). In an extensive study of emerging pollutants in Madrid, Spain, nicotine was detected in all river samples downstream from WWTP, with concentrations of up to $1.9 \times 10^{-3} \text{ mg L}^{-1}$ (Valcárcel et al., 2011). Leaking septic tanks have been linked to nicotine concentrations of up to $8.1 \times 10^{-3} \text{ mg L}^{-1}$ in UK groundwater (Stuart et al., 2012). Nicotine concentrations ranging from 2.5 to $6 \times 10^{-4} \text{ mg L}^{-1}$ were observed in estuary water in the US in the proximity of a WWTP (Benotti and Brownawell, 2007).

Nicotine is also found in drinking waters worldwide. An international study in 30 cities reported an average nicotine concentration of $1.9 \times 10^{-5} \text{ mg L}^{-1}$ (Boleda et al., 2011). In Madrid, Spain, nicotine was detected in 3 of 5 of tap water samples at levels higher than $4 \times 10^{-6} \text{ mg L}^{-1}$ reaching up to $1 \times 10^{-4} \text{ mg L}^{-1}$ (Valcárcel et al., 2011). In Miami, USA, concentrations of nicotine of $3 \times 10^{-3} \text{ mg L}^{-1}$ have been observed, thus clearly above the PNEC (National Research Council, 1977).

Even in bottled mineral water taken from pristine aquifers, far from anthropogenic sources or sites of tobacco cultivation or production, nicotine was detected in concentrations of up to $1.5 \times 10^{-5} \text{ mg L}^{-1}$ (Alonso et al., 2012).

It is important to understand sources, pathways and transformations of nicotine at the numerous natural and technical urban water interfaces (Gessner et al., 2014) and its relevance as an urban water contaminant. As littered cigarette butts may be one of these sources, the aims of the study are:

- (i) To analyze the spatial and temporal variation of littered cigarette butt accumulation in Berlin, Germany, and to determine average butt weights and residence times, as well as.

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