



Airborne emissions of asphalt/wax blends for warm mix asphalt production



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ABSTRACT

During the asphalt heating process in the various stages of hot mix asphalt (HMA) production and paving operations a complex mixture of hydrocarbon aerosols, vapors and gases, generally composed of products of combustion, volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs), is emitted into the atmosphere. The reduction of these airborne asphalt emissions is a continuous issue and challenge for asphalt industry and road agencies. Thus, a number of new sustainable and cleaner technologies, generally referred to as warm mix asphalts (WMA), that significantly reduce the manufacturing and application temperatures, had been developed. Despite the great number of reported WMA methodologies, organic waxes are widely used for reducing the binder viscosity and for enhancing the asphalt mixture workability already at lower temperatures (90–140 °C).

The manuscript presents a detailed laboratory investigation, based on headspace gas chromatography-mass spectrometry (HS-GC/MS) and photoionization detection (PID), which aimed to assess the content of VOCs and PAHs in the asphalt emissions, taking into account the effective contribution of binder or binder/wax blend. The characterization of asphalt fume composition has allowed the identification of more than 200 volatile compounds, highlighting at the same time a hierarchy of their appearance in relation to the temperature. The detection of the less volatile compounds was feasible only at temperatures higher than 160 °C, temperature range in which the HMA is generally produced. The study of the binary asphalt/wax mixtures highlighted the twofold way in which the waxes perform in reducing emissions: on the one hand they act as flow improvers (reduction of production and paving temperatures) and on the other as fume suppressants.

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1. Introduction

Hot mix asphalt (HMA) is a paving material, consisting of asphalt binder and mineral aggregates, which is generally manufactured in stationary or mobile blending plants at 170–190 °C and laid down at 140–160 °C, with significant percentage of energy consumption and release of pollutant fumes (Almeida-Costa and Benta, 2016; Rubio et al., 2013). The temperature dependent emissions generated during asphalt binder heating processes, which could have an impact on the ecosystem and on the human health (IARC, 2013; Chong et al., 2014), are defined as complex mixtures (saturated and aromatic hydrocarbons) of aerosols, vapors

and gases, generally composed of products of combustion (CO₂, NO_x and SO_x), carbon monoxide (CO), volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs) (Thives and Ghisi, 2017). Although most of the VOCs and PAHs are removed during the manufacturing process, some residues remain in the asphalt and are emitted during hot storage, transportation and road paving (Boczkaj et al., 2014). A variety of analytical techniques for sample collection, identification and quantification are available for evaluating asphalt emissions both at plant (Jullien et al., 2010) and at laboratory scale (Gasthauer et al., 2008). However, relatively little is known about detailed characterization of VOCs in asphalt fumes, since their content varies and depends on crude oil and asphalt composition (Boczkaj et al., 2014). Asphalt contains a complex mixtures of aliphatic compounds, cyclic alkanes, aromatic hydrocarbons, PAHs and heterocyclic compounds (Lesueur, 2009). Several well established strategies for the characterization of asphalt composition are based on solvent separation procedures

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(Bissada et al., 2016), even though recently some safer and simpler approaches have been developed to find composition–microstructure relationships (Autelitano et al., 2017; Gong et al., 2017).

The reduction of airborne asphalt emissions related with production, mixing, transportation and paving operations is a continuous issue and challenge for asphalt industry and road agencies. The HMA plants require approval from local authorities which is given according to the pollutant thresholds, generally expressed as annual average emissions of combustion gases, total organic carbon (TOC), total particulate matter (TPM), VOCs and PAHs (Paranhos and Petter, 2013; Ventura et al., 2015). Limits for occupational exposure to asphalt emissions have been set in more than 50 countries, often considering the 8-h time-weighted average (TWA) concentration of TPM or PAHs (Kriech and Osborn, 2014).

Recently, a line of research has examined the possibility of using fume suppression agents (flame retardants, activated carbon, expanded graphite and polymers) as additives for HMA in order to reduce asphalt emissions (Qian and Wang, 2012; Xiao et al., 2010). In parallel, a growing international pressure on the reduction of fossil fuels consumption and the emission of greenhouse effect gases has led the scientific and technical communities to develop a number of new sustainable and cleaner technologies for asphalt materials, generally referred to as warm mix asphalts (WMA), that significantly reduce the manufacturing and application temperatures (Capitão et al., 2012; Rubio et al., 2012). WMA is generally produced in a temperature range from 90 to 140 °C, with a temperature decrease up to about 30 °C with respect to HMA. Although some minor drawbacks and concerns (premature rutting, increased in-service moisture susceptibility and binder/aggregate adhesion problems) from the use of WMA were found, a number of potential benefits (environmental, economic and paving) were consistently identified (Zaumanis, 2014). Firstly, the reduced production and placement temperatures translates into lower airborne emissions: a significant decrease of combustion and greenhouse gases (10–60%) and asphalt aerosols/fumes (30–50%), in terms of VOCs and PAHs, has been observed both at plant and at site construction (Capitão et al., 2012). But, every country had developed different sampling and testing protocols for point source emissions at HMA plants, so direct comparisons of measurements are difficult. Furthermore, the lower mix temperature provides to reduce the worker exposure to pollutant compounds and to improve the working environment (D'Angelo et al., 2008). In addition to the immediate environmental effects and economic advantages (reduction of fuel and energy usage), the development of WMA could also yield technical benefits, such as the ability to pave at cooler temperatures, to haul the mixtures longer distances maintaining the adequate workability to place and compact, and to incorporate higher percentage of reclaimed asphalt pavement (RAP) (Kheradmand et al., 2013). Moreover, WMA can be produced without modifying the industrial manufacturing process, providing asphalt pavements with mechanical performance equal to or better than HMA (Sol-Sánchez et al., 2016).

Despite the great number of reported WMA technologies, they can be basically grouped in three main classes: organic waxes (viscosity reductants), chemical additives (surfactants, emulsifiers, aggregate coating enhancers and anti-stripping additives) and foaming technologies (Capitão et al., 2012). Specifically, thanks to the compatibility between asphalt and waxes and their melting/crystallizing characteristics, several studies have been conducted to evaluate the influence of these products on the asphalt properties (Pérez-Martínez et al., 2014; Qin et al., 2014). This WMA approach gave rise to three technologies which differ in three different types of wax: Fisher-Tropsch wax, synthetic fatty acid amide and lignite wax (Montan) (Rubio et al., 2012). Essentially, waxes, generally

defined “flow improvers”, are mainly used for reducing the binder viscosity at typical asphalt production and compaction temperatures, depending on their melting temperature (Polacco et al., 2012). Moreover, waxes can be used to develop ternary asphalt/polymer/wax systems which combine the mechanical and workability properties of their constituents to produce technologically advanced warm mix polymer modified asphalts (WMPMA) (Rossi et al., 2013).

Thus, the aim of this study was the characterization of the airborne emissions of both asphalt and asphalt/wax formulations in order to assess the role of both temperature and wax addition on VOCs and PAHs generation. Unlike previous studies which focused on the reduction of plant and on-site emissions considering the asphalt mixture as a whole, the manuscript presents a detailed laboratory investigation aimed at identifying and isolating the effective contribution of both binder and binder/wax blend. According to this perspective, a great novelty lies in the fact that two different potentialities of waxes, linked to the reduction of asphalt emissions, have been investigated: the capacity to act as flow improvers (reduction of production and paving temperatures) as well as fume suppressants.

2. Materials

Three 70/100 penetration neat asphalt binders (A1, A2 and A3), produced in different refineries, were used. Asphalt A2 was manufactured using crude oil from Adriatic basin (Mediterranean Sea), whereas asphalts A1 and A3 were obtained by Middle-Eastern crude oils. Table 1 summarizes the penetration grade (EN 1426:2015) and the softening point (EN 1427:2015) of the neat binders.

Two different waxes, referred in the text as M and S, were added (3% by weight) to the three binders to produce asphalt/wax blends. The asphalt/wax mixtures were labelled using an alphanumeric acronym consisting of three characters: the first letter represents the neat asphalt, whereas the number and the second letter indicate respectively the content and the type of the added wax. Specifically, M is an amidic-modified hydrocarbon wax (Montan wax by Romonta GmbH with fatty acid amide) and S is a hydrocarbon paraffinic wax (Fisher Tropsch wax by Sasol Wax). Montan wax (lignite wax) contains a combination of non-glyceride long-chain carboxylic acid esters, free long-chain organic acids, long-chain alcohols, ketones, hydrocarbons and resins. Fischer-Tropsch wax consists mainly of saturated, straight-chain hydrocarbons without functional groups (Rubio et al., 2012). Both waxes showed a melting temperature equal to 110 °C (Polacco et al., 2012). The preparation of wax-modified asphalts was as follows. An asphalt sample of 300 g was heated in a ventilate oven at 160 °C for 30 min and subsequently placed on a heating plate. Pre-weighted wax was added to asphalt and the blend was mixed for 15 min at 160 °C.

3. Methods

Analysis of asphalt emissions was carried out following a two-step experimental program. A preliminary evaluation of asphalts macro-chemical composition, that is the assessment of saturates,

Table 1
Specification of the neat asphalts.

Property	Unit	Test result		
		A1	A2	A3
Penetration (25 °C)	0.1 mm	84	75	86
Softening point	°C	46.4	49.6	46.2

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