



Polycyclic aromatic hydrocarbons, phthalates, parabens and other environmental contaminants in dust and suspended particulates of Algiers, Algeria

Lyes Rabhi^{1,2} · Abdelkader Lemou^{1,2} · Angelo Cecinato³ · Catia Balducci³ · Nabila Cherifi² · Riad Ladji² · Nouredine Yassaa^{1,4}

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Abstract

Chemicals such as polycyclic aromatic hydrocarbons (PAHs), phthalateesters, parabens, siloxanes and squalene, all of them classified as endocrine-disrupting chemicals (EDCs), have been reported to occur in all environmental compartments. The effects of EDCs on development, reproduction, growth, metabolism and obesity constitute a real public health issue. A list of EDCs (> 40 compounds) were characterised in total suspended particulates (outdoor soot: 92 samples) collected in July–September 2016 in an Algiers urban area; besides, settled indoor dusts (36 samples) were collected between November and December 2016 in schools, homes, manufacture and hospital of the same province. The analytical procedure consisted of ultrasonic bath extraction, column chromatography separation into fractions of different polarity and gas chromatographic-mass spectrometric processing. The total loads of phthalates ranging from 6.0 to 347 ng/m² (median, 26 ng/m²) were determined in indoor dust and 4.6 to 11.6 ng/m³ (median, 7.9 ng/m³) in outdoor soot; meanwhile, PAHs ranged from 12.1 to 108 ng/m² (median, 36 ng/m²) in indoor dust and ranged from 5.6 to 7.7 ng/m³ (median 5.7 ng/m³) in outdoor soot. Finally, illicit substances such as cocaine, cannabinol and Δ^9 -tetrahydrocannabinol were found to range from 0.5 to 11 pg/m³, 4.6 to 9.2 pg/m³ and 11 to 81 pg/m³, respectively, in outdoor soot.

Keywords Settled dust · Polycyclic aromatic hydrocarbons · Phthalates · Parabens · Siloxanes · Squalene · Nonylphenol · Psychotropic substances

Highlights

Phthalates, PAH and nonylphenol were measured in indoor dust and outdoor particulates from Algiers.

The median concentrations of phthalates and PAH were ca. 26 and 36 ng/m², respectively, indoors and 7.8 and 5.7 ng/m³ outdoors.

The median concentration of nonylphenol reached 16.5 and 1.6 ng/m³ respectively for indoors and outdoors.

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✉ Nouredine Yassaa
n.yassaa@cder.dz

¹ Laboratoire d'Analyse Organique Fonctionnelle, Faculté de Chimie, Université des Sciences et de la Technologie Houari Boumediene, BP 32, El-Alia, Bab Ezzouar, 16111 Algiers, Algeria

² Unité de recherche en analyses et développement technologiques en environnement-centre de recherche scientifique et technique en Analyses physico-chimiques UR-ADTE- CRAPC, BP 384, Zone industrielle RP, 42004 Bou Ismail, Tipaza, Algeria

³ Institute of Atmospheric Pollution Research (CNR-IAA), National Research Council of Italy, via Salaria, km29.3, P. O. Box10, 00015 Monterotondo, RM, Italy

⁴ Centre de Développement des Energies Renouvelables, CDER, BP 62, Route de l'Observatoire, Bouzaréah, 16340 Algiers, Algeria

Introduction

The Earth's atmosphere contains a long list of pollutants, in gaseous as well as in particulate forms, which come out from both human activity and natural phenomena (Akyüz and Çabuk 2010; Faber et al. 2015). Therefore, when carried to the human mouth or in food, suspended particulate matter and settled dust can be inhaled or ingested. After entering the human body, both types of particulates are able to give rise to human health hazards, e.g. decreased lung function, increased respiratory symptoms, cardiovascular diseases and lung cancer (Davidson et al. 2005; Anderson et al. 2012). According to the World Health Organization (WHO 2016), the estimates of deaths due to air pollution reached 4.3 million in 2012 for households, where women and children are particularly exposed; meanwhile, outdoor pollution was estimated to cause three million deaths worldwide both in cities and rural areas. Cumulatively, deaths due to air pollution in 2012 exceeded seven millions (WHO 2017).

Endocrine-disrupting chemicals (EDCs) are characterized by chemical structures interfering with normal hormone functions (Kabir et al. 2015). EDCs can be grouped according to origin, as natural and artificial hormones, as drugs with hormonal side effects, as industrial or household chemicals and as processed side products. Inhalation of such chemicals from the atmosphere is recognised as the major source of health problems. The key role of several EDCs substances is currently suspected in the development of hormone-dependent cancers (breast, uterine, prostate and testicular cancers), though the data currently available do not confirm this link. The EDC list affecting atmosphere includes a number of chemicals, both natural and anthropogenic. This list includes phthalates, parabens, alkylphenols and bisphenols and psychotropic substances.

Phthalate esters (PAEs) are among the most frequently detected and abundant in the environment; PAEs are classified as persistent organic pollutants (POPs) (Gao and Wen 2016) and are ubiquitous in the environment, though only in recent years, attention has been paid to their occurrence in the atmosphere (Khedidji et al. 2017a; Kang et al. 2017). PAEs find a wide range of applications, e.g., in cosmetics and personal care products and in various consumer, household and industrial products as plasticisers (Tran and Kannan 2015). On the other hand, they elicit reproductive and developmental toxicities in laboratory tests on animals (Tran and Kannan 2015; Tran et al. 2016). It has been reported that PAE exposure induces endocrine disruption, adverse respiratory effects and reproductive and developmental toxicities (Lin et al. 2011; Buck Louis et al. 2013).

Alkyl esters of *p*-hydroxybenzoic acid, usually known as parabens, are widely used as antimicrobial agents in food products, pharmaceutical preparations, cosmetics and toiletry consumer products (Piao et al. 2014; Larsson et al. 2014). Exposure to parabens may disrupt the endocrine human

system, fighting pineal, pituitary, thyroid, parathyroid, adrenal glands, thymus, pancreas, ovaries in females and testes in males. Hence, parabens may induce harmful effects on human health (Soni et al. 2005). Nonylphenol (NOP) is widely used as a mixture of isomers in lubricating oils, detergents, stabiliser of emulsifiers in rubber and intermediate in the production of ethoxylate surfactants (Cecinato et al. 2017). NOP is well known as endocrine-disrupting compound (EDC) (Lu and Gan 2014). Though present in the atmosphere preferably in gas phase, NOP has been detected in dust and soot (Rudel et al. 2003; Cecinato et al. 2017). Squalene is a non-volatile triterpene with six double bonds, making this compound one of the most unsaturated lipids. It is naturally appearing and continually produced by the human body shed (Popa et al. 2015). Squalene has been detected previously in dust by Weschler et al. (2011).

The psychotropic substances (PSs) or psychoactive drugs refer to both legal and illegal compounds. Nicotine and caffeine are both able to cause adverse effects on health; however, no restrictions exist with regard to their release into the air and consumption (Cecinato et al. 2014a). As for illegal substances (cocaine, heroin, amphetamine and three cannabinoids (CBs), namely cannabidiol, cannabidiol, Δ^9 -tetrahydrocannabinol), Viana and coworkers (Viana et al. 2010, 2011) reported that their occurrence is presumably related to consumption (cocaine, heroin, cannabidiol, cannabidiol and Δ^9 -tetrahydrocannabinol) or to handling (amphetamines). Some illicit PSs exist predominantly as particulate (cocaine, heroin and cannabinoids), whilst caffeine, nicotine and amphetamines are expected to partition between gaseous and particulate phases (Cecinato et al. 2012a).

Polycyclic aromatic hydrocarbons (PAHs) are suspected or ascertained carcinogens and mutagens (IARC 2005). There are two types of anthropogenic sources, i.e. petrogenic that include crude and refined petroleum and pyrogenic coming from the combustion of fossil fuels and biomass (Isobe et al. 2007). The PAH hydrophilicity and mobility decrease as the number of rings increases (Iqbal et al. 2008). A compound stands out among other PAH isbenzo(a)pyrene (BaP), due to its strong carcinogenicity (Srogi 2007). In ambient air, PAHs are predominantly associated with small particles, which drive them to accumulate in the respiratory system and induce harmful health effects (Ladji et al. 2014).

The occurrence of phthalates esters, parabens, polycyclic aromatic hydrocarbons, licit and illicit drugs in the environment has been investigated previously not only in Algeria (Khedidji et al. 2013, 2017a, b; Boudehane et al. 2016; Assami et al. 2017), but also all over the world (Cecinato et al. 2014a, b; Viana et al. 2010, 2011).

Whilst PAHs have been intensively investigated in Algiers atmosphere during the last two decades (Cecinato et al. 2012a, b), phthalate ester and drugs were assessed only recently. The major PAH sources were gasoline and diesel vehicle

emissions, in addition to the biomass and fossil fuel burning (Ladji et al. 2007, 2009). By contrast, cocaine was always absent, whilst cannabidiol cannabinol, namely D9-tetrahydrocannabinol was detected (Boudehane et al. 2016).

The goal of the present study was to evaluate the levels of phthalates, parabens, licit and illicit PSs, nonylphenol and polycyclic aromatic hydrocarbons in populated Algiers urban location (Algeria), in relation to demographics, lifestyle, housing and workplaces (Yassaa et al. 2001a, b, c, d, 2005; Yassaa and Cecinato 2005; Khedidji et al. 2017b). In addition, a look was given to non-polar fraction including *n*-alkanes, squalene and siloxanes, to draw further semi-quantitative insights about the distinct pollution sources.

Experimental

Sampling sites and procedures

Two types of particle collection were carried out in our study, the former outdoors, and dedicated to atmospheric aerosol (one only sampling point, 92 samples) and the latter indoors for settled dust (12 sampling points, 36 samples). The first approach was active sampling (through aspiration on inert membrane), whilst the second was passive (dust settled on a dedicated, pre-cleaned surface was manually recovered by means of a couple of ultra-pure glass fibre filters).

The pumping system for aerosols (total suspended particulate (TSP)) was located in Bourouba, a suburb of Algiers placed in the southeast side of the city. Bourouba was characterised by heavy traffic and frequent traffic jams on weekdays (Saturday to Thursday, because Friday is holiday). TSP was collected daily from July to September onto polytetrafluoroethylene membranes (PTFE, 0.45- μ m pore size, 47-mm outer diameter), by means of a pump equipped with TSP inlet and running at 1 m³/h. Each sampling started at 7:00 h and lasted 24 h. Instead, for indoor dust, 12 micro-environments (individual rooms) were considered, which were distributed among two schools, two houses, one hospital and a detergent manufacture (see Table 1). Except for manufacture plant, the sites lay in metropolitan Algiers. The schools were located in the residential districts of Rouïba and Sidi Moussa. Forty-nine children and nine employees attended at school in Rouïba with a rhythm of opening of 8 h, whilst in Sidi Moussa, the amounts of pupils and employees were 657 and 30, respectively, and the opening hours were 10, starting at 7:30 h. The two houses were in Bourouba and Bab Ezzouar. The hospital was located in El Harrach suburb; this structure gets the peculiarity of receiving all road victims from the east of the country. Three surgery rooms, two offices, one resuscitation room, one reception and one store room of medical equipment were investigated there. All sites were at the ground floor except a fourth-floor office; the hospital employees were more than a thousand. The

manufacturing plant was located in west side of Boumerdes. The collection protocol of dust comprised the pre-cleaning of the surface (horizontal, at 1.8–2.2 m above ground, 1 m²), free exposure to particle settlement and recovery of dust after 15 days by means of two white filter papers in cellulose (diameter = 125 mm) produced by Sartorius and purchased from Prophylab (Algiers), scrubbed in sequence; afterwards, TSP and soot samples were wrapped with aluminium foils and stored at low temperature (~ 4 °C) until analysis.

Sample extraction and clean up

Chemical characterisation of organic aerosol and dust was performed by applying a procedure described extensively elsewhere (Romagnoli et al. 2014), introducing only slight changes. Briefly, each sample was spiked with 25 μ l of acetonitrile containing a mix of internal reference compounds, i.e. perdeuterated *n*-C₁₄, *n*-C₁₆, *n*-C₂₀, *n*-C₂₄, *n*-C₃₀ (*n*-alkanes and squalene), acenaphthene-D₁₀, phenanthrene-D₁₀, fluoranthene-D₁₀, benz[a]anthracene-D₁₂, benzo[a] pyrene-D₁₂ and dibenz[a,h]anthracene-D₁₄, (PAHs), dipropyl phthalate (DPrP), di phenyl phthalate (DPhP), cocaine-D₃, Δ^9 -tetrahydrocannabinol D₃ and caffeine-¹³C₃ (polar compounds). Samples were extracted by ultrasonic bath (22 min, three times) with 12 mL of a dichloromethane, acetone and methanol mixture (DAM, 50:30:20 in volume); the extracts were reduced close to dryness under ultra-pure nitrogen and transferred to the top of a chromatographic column for fractionation and purification (silica, deactivated with 2% of water, 1.0 g; i.d. = 6 mm, *h* = 200 mm). The target compounds were eluted by using three solvents in series. The first fraction (*n*-heptane, 5 mL) comprised non-polar compounds, the second (dichloromethane/*n*-hexane 20:40, 5 mL) comprised low-polar compounds including PAHs and the third (DAM mixture, 6 mL) allowed to recover high-polar substances (phthalates, parabens and PSs). Eluents were evaporated under nitrogen and extracts back dissolved with chloroform before instrumental analysis.

Instrumental analysis and method effectiveness

All sample extract fractions were processed by means of a Trace GC Ultra gas chromatograph equipped with AS-3000 autosampler and Trace DSQ quadruple mass spectrometer (all from Thermo, Rodano MI, Italy) and managed by the dedicated Excalibur software. Chemical determinations were performed by using a DB5-MS type column (*L* = 30 m, i.d. = 250 μ m, film thickness = 0.25 μ m). The oven temperature was programmed from 70 °C (held 1.5 min) to 160 °C at 15 °C/min rate (held 2 min), then up to 280 °C at 4 °C/min (20 min); the transfer liner to MSD was set at 280 °C. The MS operating conditions were as follows: source temperature, 250 °C; filament current, 200 mA, detection modality:

Table 1 Description of individual sites and places of dust and particles in Bourouba and its surroundings (Algiers metropolitan area)

| Located | Site type | Abbreviations | Smokes | Traffic | Sampling period |
|------------------------------------|----------------------------|---------------|--------|--------------|------------------------------|
| Bourouba | Urban city | SPM | Yes | Yes (high) | June–September 2016 |
| Bourouba | Home | R1 | Yes | Yes (high) | 10 December–25 December 2016 |
| Bordj el kifane | Home | R2 | No | Yes (medium) | |
| Sidi Moussa | School | S1 | No | Yes (medium) | 27 November–11 December 2016 |
| Rouïba | School | S2 | No | No (low) | 20 November–02 December 2016 |
| west of Boumerdes (khamis kanchla) | Detergent production plant | M1 | Yes | Yes (medium) | 20 November–05 December 2016 |
| El Harrach, Zmirli hospital | Office 1 | H1 | No | Yes (medium) | 23 November–09 December 2016 |
| | Office 2 | H2 | No | Yes (medium) | |
| | Holle of the hospital | H3 | No | Yes (low) | |
| | Resuscitation room | H4 | No | Yes (medium) | |
| | Store room | H5 | No | Yes (low) | |
| | Surgery room | H6 | No | Yes (low) | |
| Bou Ismail | Laboratory | RC | No | Yes (medium) | 15 December–29 December 2016 |

electron impact, 70 eV, selected ion monitoring mode (three ion traces for analytes, two ion traces for reference compounds). Target compounds were identified by comparing the retention times and intensity ratios of characteristic ion currents with authentic standards. The detection limits (LODs) were set at three times the signal to noise ratios, whilst quantification limits (LOQs) at 3.3 times the corresponding LODs. LODs ranged from 0.4 ng/mL (*n*-C₁₈) to 0.8 ng/mL (*n*-C₃₅) for alkanes, from 0.1 ng/mL (phenanthrene, fluoranthene) to 0.5 µg/mL (dibenz[a,h]anthracene) for PAHs; besides, LODs were better than 1 ng/mL for all polar compounds. The quantitative determination was achieved by applying the isotopic dilution method and referring each analyte to the closest deuterated/¹³C labelled congener within the chromatogram. The instrument response curves were drawn by plotting the peak area ratios of native compounds vs. the respective internal standards, and five concentration levels of analytes were processed in triplicate, keeping constant the reference compounds. Different concentration ranges were tested for each group, according to the substance amounts in the samples. Blanks were analysed for both kinds of matrices. It was necessary to correct the concentrations measured of light PAHs (up to phenanthrene), light *n*-alkanes (up to C₂₀) and phthalates (diethyl, diisobutyl and ethylhexyl homologues) due to the compound occurrence in blanks. The total recovery of the method lied in the range 87 ± 15% with the worst results associated to parabens. Examples of chromatograms (total ion current (TIC) and selected ion mass (SIM)) for *n*-alkanes, squalenes and silanes are given in Fig. 1.

Results and discussion

The percentage profiles of three polarity fractions ((1) *n*-alkanes, siloxanes and squalene; (2) polycyclic aromatic

hydrocarbons; (3) phthalates, psychotropic substances and nonylphenol in dust and soot) are shown in Fig. 2. In general, the relative contributions of chemicals in dust were in the following order: nonylphenol (mean 234 ng/m², range 5.3–2239 ng/m²) > phthalates (mean 89 ng/m², range 6.0–347 ng/m²) > PAHs (mean 42 ng/m², range 12–107 ng/m²); meanwhile, in soot particulate matter (SPM), the abundances were as follows: *n*-alkanes (mean 35 ng/m³; 32–128 ng/m³) > phthalates (mean 8.0 ng/m³; 4.6–12. ng/m³) > PAHs (mean 6.3 ng/m³, range 5.7–7.7 ng/m³).

n-Alkanes, siloxanes and squalene

The non-polar fraction of organic aerosols was determined in indoor air at hospital (H1 to H6), school (S1 and S2), home (R1, R2) and chemistry laboratory (RC) and detergent manufacturing plant (M1); besides, analyses were made of soot in Bourouba (see Table 2). The total *n*-alkanes ranged from ca. 1.0 to 30 µg/m²/day for settled dust, the maximum and the minimum recorded respectively at school (S1 location) and hospital (H3). The *n*-alkane loads measured were comparable with those previously observed at Ouargla city (4–41 µg/m²; see Boudehane et al. 2016).

In the soot, the mean of total alkanes reached ~ 35 ng/m³, which was comparable with those recorded during the last decade in Algeria. In Bouira province, the measure was 20–60 ng/m³; (Khedidji et al. 2017a), but lesser in Algiers (108–204 ng/m³; 42–94 ng/m³) (Ladji et al. 2009); (14–206 ng/m³; Yassaa et al. 2001c).

To approach the soot source characterisation, both carbon preference index (CPI) and maximum homologue concentration [*C*_{max}] were investigated. CPI was expressed as the ratio of total odd carbon number *n*-alkanes vs. total even carbon number of *n*-alkanes, starting from C₂₄–C₂₅ homologues (Brown et al. 2002); *C*_{max} was the carbon number for which

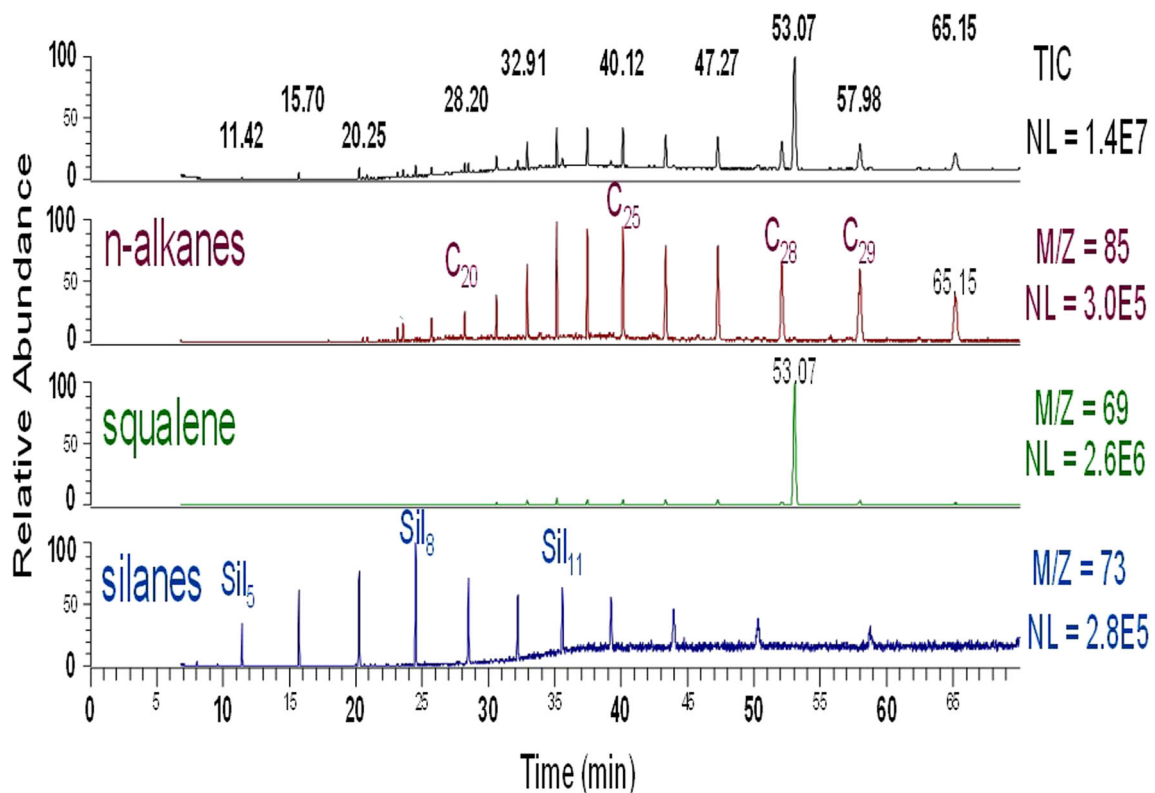


Fig. 1 Examples of chromatograms (total ion current (TIC) and selected ion mass (SIM)) of n-alkanes, squalenes and silanes

the maximum concentration was detected. To sum up, the higher rates of CPI (> 3) or C_{max} (C_{29} , C_{31} imply strong biogenic input, whilst the lower rates (≈ 1 and C_{19} up to C_{25} , respectively) are typical of anthropogenic sources. In our study, the dust CPI ranged 1.3–3.4; in general, small differences were found between locations at school, home and

hospital (1.3–1.8), unlike at manufacturing plant (3.4), suggesting natural sources with more biogenic influence (Brown et al. 2002); meanwhile, C_{max} , in dust corresponded to C_{27} – C_{29} and, according to (Cecinato et al. 1999), was indicative of important but not predominant biogenic emission. For SPM (Fig. 3), the CPI was equal to ~ 1.53 suggesting that they were

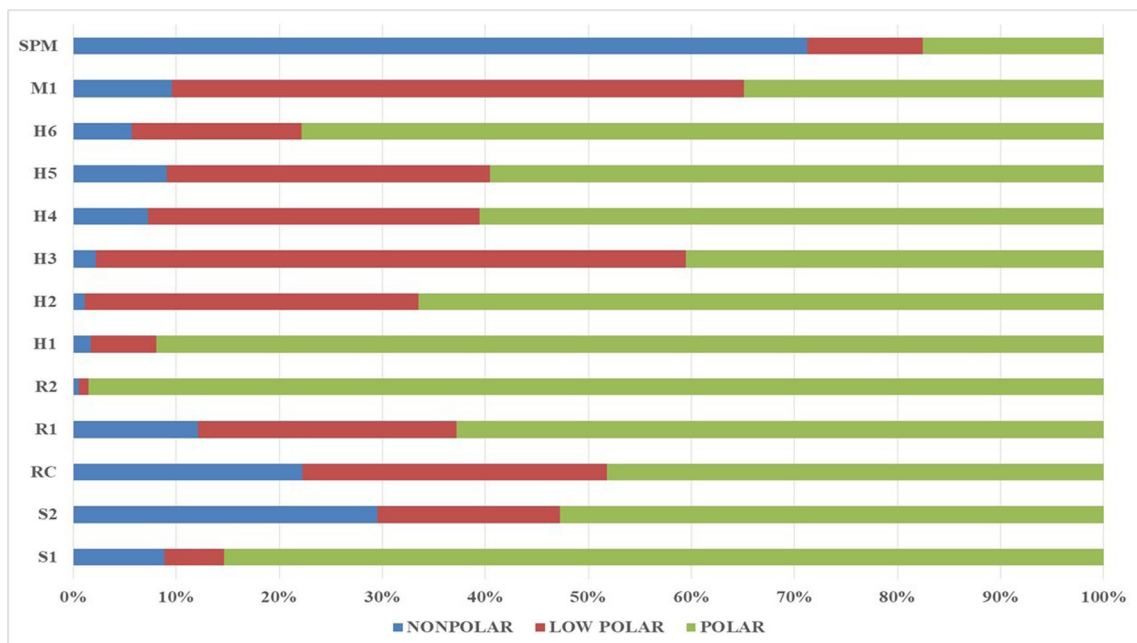


Fig. 2 Organic fraction in indoor dust and outdoor samples

Table 2 Average concentration of non-polar organic compounds in dust and SPM in in Bourouba and its surroundings (Algiers metropolitan area)

| | Samples | Alkanes | Cmax | CPI | Squalene | Siloxanes | Siloxanes/ <i>n</i> -alkanes |
|-------------------------------|-----------|---------|------|------|----------|-----------|---------------------------------|
| TSP (ng/m ³) | July | 37.5 | 27 | 1.65 | 0.01 | 2.1 | 0.06 |
| | August | 35.6 | 27 | 1.46 | 0.01 | 10 | 0.29 |
| | September | 32.4 | 28 | 1.50 | 0.01 | 2 | 0.07 |
| | Mean | 35.2 | 27 | 1.53 | 0.01 | 4.7 | 0.13 |
| DUST (ng/m ² /day) | S1 | 29.9 | 29 | 1.57 | 0.00 | 9 | 0.30 |
| | S2 | 11.9 | 27 | 1.37 | 0.00 | 8 | 0.67 |
| | RC | 20.4 | 27 | 1.30 | 0.20 | n.d | 0.00 |
| | R1 | 16.3 | 29 | 1.56 | 0.30 | 3 | 0.18 |
| | R2 | 4.7 | 29 | 1.48 | 0.25 | 1.3 | 0.28 |
| | H1 | 5.3 | 29 | 1.60 | 0.11 | n.d. | 0.00 |
| | H2 | 3.1 | 29 | 1.78 | 0.02 | n.d | 0.00 |
| | H3 | 1.0 | 29 | 1.42 | 0.05 | n.d | 0.00 |
| | H4 | 5.8 | 29 | 1.79 | 0.01 | 0.8 | 0.14 |
| | H5 | 6.4 | 28 | 1.68 | 0.00 | 5 | 0.78 |
| | H6 | 9.9 | 27 | 1.42 | 0.15 | X | 0.02 |
| | M1 | 7.0 | 29 | 3.43 | 0.00 | n.d | 0.00 |

TSP total suspended particulate, CPI carbon preference index, C_{max} maximum homologue concentration, *n.d.* not detected, *X* detected but not determined due to blank interference

mainly derived from the incomplete combustion of fossil fuels whilst C_{max} at C27 corresponded to *n*-alkanes arising from vehicular exhaust and road dust.

Siloxanes and squalene are rarely investigated, and a few authors reported the occurrence of linear siloxanes in indoor and outdoor environments (e.g. 0.2 up to 10 ng/m³ and 0.01–10 ng/m²/day, respectively, in SPM and dust; see (Lu et al. 2010; Tran et al. 2015; Yucuis et al. 2013)), whilst lack of

information remains about squalene. Besides, these compounds were out of our primary interest in this study (this is the reason why we did not carry out the accurate determination of the respective loads in the samples). Nonetheless, it looked of concern to proceed to semi-quantitative estimate of both siloxanes and squalene, because both SPM and dust showed to contain important and widely ranging amounts of these chemicals that were attempted through comparison with

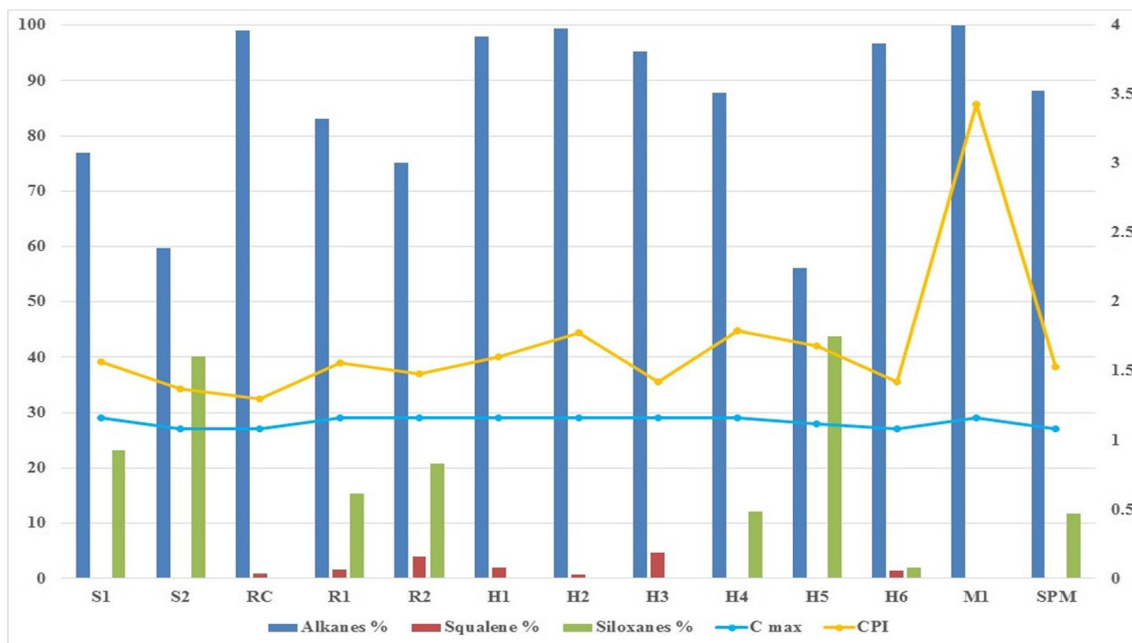
**Fig. 3** Composition profiles of alkanes, siloxanes, squalenes and their carbon preference index CPI in indoor dusts and airborne particle SPM

Table 3 Average of polycyclic aromatic hydrocarbons concentrations (ng/m³) recorded in TSP in downtown Algiers. Jun to September 2016

| Compounds | Abbreviation | Mass <i>m/z</i> | July | August | September | Mean ± SD |
|--------------------------------|--------------|-----------------|------|--------|-----------|-------------|
| Acenaphthene | ACE | 154 | 1.68 | n.d | n.d | 1.68 ± 0.00 |
| Acenaphthylene | ACY | 152 | 1.05 | 0.76 | 0.56 | 0.79 ± 0.25 |
| Anthracene | ANT | 178 | 0.03 | 0.04 | 0.03 | 0.03 ± 0.09 |
| Fluorene | FL | 166 | n.d | 0.14 | 0.01 | 0.07 ± 0.09 |
| Methyl Phenanthrene/anthracene | Me-P/A | 192 | 0.17 | 0.17 | 0.17 | 0.17 ± 0.00 |
| Phenanthrene | PHE | 178 | 0.06 | 0.13 | 0.10 | 0.10 ± 0.04 |
| Benz[a]anthracene | BaAnt | 228 | 0.05 | 0.08 | 0.17 | 0.10 ± 0.06 |
| Chrysene | CHR | 202 | 0.10 | 0.15 | 0.27 | 0.17 ± 0.09 |
| Fluoranthene | FLT | 202 | 0.06 | 0.09 | 0.12 | 0.09 ± 0.03 |
| 5-Methylchrysene | Me-CHR | 242 | 0.15 | 0.27 | 0.42 | 0.28 ± 0.14 |
| Methyl Fluoranthene/pyrene | Me-F/P | 216 | 0.05 | 0.09 | 0.12 | 0.09 ± 0.04 |
| Pyrene | PY | 202 | 0.07 | 0.08 | 0.12 | 0.09 ± 0.03 |
| Benzo[b]fluoranthene | BbF | 252 | 0.25 | 0.44 | 0.72 | 0.47 ± 0.24 |
| Benzo[j]fluoranthene | BjF | 252 | 0.13 | 0.21 | 0.34 | 0.23 ± 0.10 |
| Benzo[k]fluoranthene | BkF | 252 | 0.15 | 0.24 | 0.36 | 0.25 ± 0.11 |
| Benzo[a]pyrene | BaP | 252 | 0.12 | 0.20 | 0.42 | 0.24 ± 0.15 |
| Benzo[e]pyrene | BeP | 252 | 0.25 | 0.42 | 0.65 | 0.44 ± 0.20 |
| Cyclopenta(c,d)pyrene | CPPyr | 226 | 0.02 | 0.04 | 0.10 | 0.05 ± 0.04 |
| Dibenz[a,h]anthracene | Dbant | 278 | 0.05 | 0.06 | 0.12 | 0.08 ± 0.04 |
| Perylene | PERY | 252 | 0.05 | 0.07 | 0.12 | 0.08 ± 0.03 |
| Benzo[ghi]perylene | Bpery | 276 | 0.81 | 1.16 | 1.55 | 1.17 ± 0.37 |
| Indeno[1.2.3-cd]pyrene | IP | 276 | 0.38 | 0.73 | 1.25 | 0.79 ± 0.44 |
| ΣPAHs | | | 5.67 | 5.59 | 7.47 | 7.47 ± 1.21 |
| ΣLMW | | | 3.46 | 2.02 | 2.10 | 2.53 ± 0.81 |
| ΣHMW | | | 2.21 | 3.58 | 5.63 | 3.80 ± 1.72 |
| ΣLMW/ΣHMW | | | 1.57 | 0.56 | 0.37 | 0.84 ± 0.64 |
| BaP/BaP + BPE | | | 0.33 | 0.32 | 0.39 | 0.35 ± 0.04 |
| BaA/(BaA + CHR) | | | 0.55 | 0.57 | 0.60 | 0.58 ± 0.03 |
| IP/(IP + BghiP) | | | 0.32 | 0.39 | 0.45 | 0.38 ± 0.06 |

n.d. not detected

n-alkanes, present in the same non-polar fraction of extracts. Indeed, siloxanes find application in a wide variety of consumer products (Lu et al. 2010; MacLeod et al. 2013; Tran et al. 2015, 2017) and have been reported to be reproductive and endocrine toxicants (Genualdi et al. 2011; Tran et al. 2015, 2017). Whilst squalene is produced by living organisms and is a major component of lipids present in human skin (Fu et al. 2013).

As for SPM, siloxanes peaked in August whilst *n*-alkanes remained quite constant, so the ratios ranged from ca. 6% up to 29%. Squalene was all the time much less (≈ 0.01 ng/m³). In dusts, squalene accounted for 5% of *n*-alkanes at homes, less than 2% at the hospital and was absent at schools and manufacturing plant; by contrast, siloxanes were more important at schools than at homes, whilst in the hospital, the presence of siloxanes was widely dependent on the room use, ranging from ~ 0 to 70% of alkanes (H5). As for siloxanes,

the indoor and outdoor concentrations were low compared to those reported by Tran and coworkers (Tran et al. 2015) in Hanoi, Hatinh, Hungyen and Thaibinh in Vietnam (179 ng/g), Cartagena in Colombia (198 ng/g), Jeddah in Saudi Arabia (262 ng/g), Albany in the USA (882 ng/g) and Athens, Erateini and Komotini in Greece (1490 ng/g).

The occurrence of squalene indoors was in agreement with the staying of (many) people, in which siloxanes could be related to activities made there.

Siloxanes which are synthetic organosilicon compounds and used in a wide variety of personal care and other consumer products are highly volatile (Lu et al. 2010). Table 2 shows ratio of siloxanes to *n*-alkanes in different investigated environments. The ratio in soot peaked in August (0.29) and remained constant in July and September. In dust, it ranged from 0.02 to 0.78 and was absent at hospital office and manufacturing.

Table 4 Concentration of polycyclic aromatic hydrocarbons (ng/m²/day) in indoor dusts collected in Algiers city

| Compounds | S1 | S2 | RC | R1 | R2 | H1 | H2 | H3 | H4 | H5 | H6 | M1 |
|-----------------|------|------|------|-------|------|------|-------|-------|------|------|-------|------|
| ACE | n.d | 1.56 | 0.08 | 0.24 | n.d | n.d | 50.02 | n.d | n.d | n.d | 10.34 | n.d |
| ACY | n.d | n.d | 2.01 | 6.20 | 6.29 | 7.26 | 34.89 | 21.30 | 5.93 | 6.55 | 6.78 | n.d |
| ANT | 0.37 | 0.39 | 0.83 | 1.14 | 0.45 | 0.47 | 0.85 | 0.96 | 0.65 | 0.78 | 1.31 | 1.97 |
| FL | 9.32 | n.d | n.d | n.d | n.d | n.d | 2.44 | n.d | n.d | n.d | n.d | 4.51 |
| Me-PHE/ANT | 2.48 | 1.12 | 4.2 | 17.5 | 2.72 | 3.5 | 7.2 | 9.1 | 4.0 | 6.3 | 8.1 | 2.29 |
| PHE | 2.49 | 0.58 | 1.89 | 3.74 | 0.90 | 0.80 | 1.60 | 0.73 | 1.61 | 0.82 | 1.62 | 5.69 |
| BaAnt | 0.55 | 0.55 | 1.04 | 0.87 | 0.62 | 1.30 | 0.62 | 0.91 | 0.98 | 0.85 | 0.46 | 1.58 |
| CHR | 0.83 | 0.46 | 1.05 | 0.49 | 0.65 | 0.40 | n.d | 0.69 | 0.72 | 0.72 | 0.63 | 1.45 |
| FLT | 1.78 | 0.74 | 4.24 | 2.44 | 0.88 | 1.21 | 1.14 | 3.29 | 1.84 | 1.71 | 1.47 | 3.63 |
| Me-CHR | 1.16 | 0.74 | 2.39 | 14.90 | 2.16 | 5.08 | 1.57 | 4.57 | 2.04 | 3.04 | 2.73 | 1.84 |
| Me-FLU/PYR | 0.92 | 0.55 | 2.95 | 4.60 | 1.51 | 4.73 | 1.16 | 12.46 | 1.51 | 3.55 | 1.69 | 1.57 |
| PY | 1.41 | 0.78 | 6.75 | 1.85 | 0.81 | 1.00 | 1.01 | 1.71 | 1.38 | 1.33 | 0.92 | 2.92 |
| BbF | 0.60 | 0.63 | 0.83 | 0.77 | 0.56 | 0.83 | 0.71 | 0.82 | 1.35 | 1.73 | 0.67 | 1.20 |
| BjF | 0.59 | 0.64 | 0.84 | 0.78 | 0.54 | 0.86 | 0.72 | 0.97 | 1.31 | 1.54 | 0.81 | 1.33 |
| BkF | 0.77 | 0.72 | 1.21 | 0.95 | 0.50 | 1.31 | 0.91 | 1.34 | 2.90 | 3.62 | 0.81 | 2.22 |
| BaP | 0.45 | 0.55 | 0.81 | 0.66 | 0.53 | 0.74 | 0.61 | 0.74 | 0.76 | 0.70 | 0.53 | 1.75 |
| BeP | 0.71 | 0.70 | 0.96 | 0.99 | 0.65 | 1.21 | 0.83 | 0.78 | 1.61 | 1.98 | 0.55 | 1.68 |
| DBant | 0.03 | 0.10 | 0.12 | n.d | n.d | 0.16 | n.d | n.d | n.d | 0.39 | 0.10 | 0.31 |
| PERY | n.d | n.d | 0.59 | 0.51 | 0.49 | 0.53 | 0.51 | 0.71 | 0.56 | 0.55 | 0.50 | 0.85 |
| BPery | 0.29 | 0.55 | 0.57 | 0.10 | 0.08 | 0.33 | 0.30 | 0.10 | 0.68 | 0.65 | 0.10 | 1.64 |
| IP | 0.47 | 0.77 | 0.77 | 0.20 | 0.29 | 0.55 | 0.45 | 0.28 | 0.99 | 1.24 | 0.23 | 1.87 |
| PAHs | 25.2 | 12.1 | 34.1 | 58.9 | 20.6 | 32.3 | 107.5 | 61.5 | 30.8 | 38.0 | 40.3 | 40.4 |
| ΣLMW/ΣHMW | 0.51 | 0.38 | 0.97 | 1.05 | 0.62 | 9.90 | 1.11 | 0.72 | 0.67 | 2.37 | 0.65 | 0.51 |
| BaP/BaP + BPE | 0.39 | 0.44 | 0.46 | 0.40 | 0.45 | 0.38 | 0.43 | 0.49 | 0.32 | 0.26 | 0.49 | 0.51 |
| BaA/(BaA + CHR) | 0.40 | 0.55 | 0.50 | 0.64 | 0.49 | 0.77 | 1.00 | 0.57 | 0.58 | 0.54 | 0.42 | 0.52 |
| IP/(IP + BghiP) | 0.62 | 0.58 | 0.57 | 0.68 | 0.78 | 0.62 | 0.60 | 0.73 | 0.59 | 0.66 | 0.69 | 0.53 |

PAHs

Most routine PAH monitoring have been targeted on 3–6-ring congeners. In this study, 22 and 21 PAHs were quantified, respectively, in outdoor SPM and indoor dust (Tables 3 and 4).

Alkyl derivatives of PAHs (methyl phenanthrenes, anthracenes, fluoranthenes, pyrenes and chrysenes) accounted for ca. 7% of the total, so our concern was focussed on the percent composition on the basis of aromatic ring number. In SPM, 4/5-ring PAHs together accounted for ~60% of the total PAHs and 2/3-ring compounds for the remaining~40%. By contrast, in dust, ~80% was comprised of 2/3-ring PAHs.

Benzo(a)pyrene is usually chosen as single indicator of the whole PAH group and marker of total exposure (and consequent health risk associated) to suspended particulate carcinogenicity. In our study, the average concentration of BaP was equal to 0.24 ng/m³ in SPM, namely lesser than the 1-ng/m³ limit proposed by the WHO as air quality indicator (WHO 2000). The BaP average at the outdoor site was lower than those previously reported elsewhere in Algeria (Khedidji et al. 2013; Ladji et al. 2014). As for dust, the total PAHs ranged

between 12 and 107 µg/m²/day, found at S2 and H2 locations, respectively. Thus, despite hospital hosted citizens particularly sensitive to toxicants action, this site was more PAH-affected than the other ones, whilst a better situation was recorded at school (children are another highly sensitive population segment).

To draw information about the background of PAH sources in dust and soot, we applied the PAH diagnostic ratios approach (Ravindra et al. 2008; Tobiszewski and Namieśnik 2012), through calculating the following rates: ΣLMW/ΣHMW, BaP/(BaP + BPE), BaA/(BaA + CHR) and IP/(IP + BghiPE). ΣLMW/ΣHMW ratio is the sum of two/three ring PAHs vs. sum of 4/5-ring congeners; it has been used to distinguish petrogenic from pyrogenic sources (Zhang et al. 2008). ΣLMW/ΣHMW rate varied between 0.8 and 9.9 in dust, whilst it was ~1.1 in SPM, which indicates the two types of source were alternatively prevailing in the various environments. In fact, the low rates (<1) indicated petrogenic sources, e.g. including oil split and oil petroleum products uses, whilst the rates >1 were typical of pyrogenic sources, namely incomplete combustion of fossil fuels and wood.

Table 5 Highly polar organic compounds in the airborne particulates of Bourouba (ng/m³)

| SPM | abbreviation | Mass <i>m/z</i> | July | August | September | Sum | Mean | Median |
|------------------------------------|--------------|-----------------|-------|--------|-----------|--------|--------|--------|
| Methyl paraben | M-P | 152 | 0.08 | 0.06 | 0.05 | 0.19 | 0.06 | 0.06 |
| Ethyl paraben | E-P | 166 | 0.19 | 0.18 | 0.19 | 0.57 | 0.19 | 0.19 |
| Propyl paraben | P-P | 180 | 0.09 | 0.11 | 0.10 | 0.30 | 0.10 | 0.10 |
| Sums parabens | SPB | | 0.36 | 0.35 | 0.34 | 1.05 | 0.35 | 0.35 |
| Caffeine | CAF* | 194 | 54.31 | 169.16 | 515.00 | 738.47 | 246.16 | 169.16 |
| Cocaine | COC* | 303 | 0.50 | 11.39 | 9.30 | 21.18 | 7.06 | 9.30 |
| Tetrahydrocannabinol | THC* | 314 | 9.23 | 4.61 | n.d | 13.84 | 6.92 | 6.92 |
| Cannabinol | CBL* | 310 | n.d | 10.80 | 80.84 | 91.64 | 45.82 | 45.82 |
| Heroin | HER | 369 | n.d | n.d | n.d | n.d | n.d | n.d |
| Nonylphenol | NOP | 220 | 1.37 | 1.75 | 1.62 | 4.74 | 1.58 | 1.62 |
| <i>N,N</i> -diethyl-meta-toluamide | DEET | 191 | 0.02 | 0.08 | 0.05 | 0.14 | 0.05 | 0.05 |
| Bis 2 ethyl hexyl adipato | DEHA | 370 | 0.01 | 0.02 | 0.07 | 0.11 | 0.04 | 0.02 |
| Butyl benzyl phthalate | BBP | 312 | 0.01 | 0.03 | 0.03 | 0.06 | 0.02 | 0.03 |
| Di decyl phthalate | DIBP | 446 | 0.36 | 0.92 | 1.79 | 3.08 | 1.03 | 0.92 |
| Di iso butyl phthalate | DdeP | 278 | 0.07 | 0.07 | 0.14 | 0.28 | 0.09 | 0.07 |
| Di(2-ethylhexyl) phthalate | DEHP | 390 | 1.74 | 3.11 | 3.79 | 8.63 | 2.88 | 3.11 |
| Diethyl phthalate | DEP | 222 | 0.10 | 0.13 | 0.05 | 0.27 | 0.09 | 0.10 |
| Dibutyl phthalate | DBP | 278 | 1.98 | 3.20 | 4.92 | 10.11 | 3.37 | 3.20 |
| Di nonyl phthalate | DnoP | 418 | 0.20 | 0.19 | 0.39 | 0.78 | 0.26 | 0.20 |
| Di n octyl phthalate | DocP | 390 | 0.09 | 0.11 | 0.41 | 0.61 | 0.20 | 0.11 |
| Sums of phthalates | ΣPHT | | 4.55 | 7.76 | 11.51 | 23.82 | 7.94 | 7.74 |

*pg/m³

BaP/(BaP + BeP) rates ranged 0.26–0.51 in dust and reached 0.36 in SPM. According to the ratio values reported in literature (Oliveira et al. 2011), the rates ~0.50 meant fresh emission, which was observed only in M1, whilst rates < 0.3 suggested important photolysis of BaP and then the ageing of particles; thus, air amounts in the micro-environment investigated were more or less renewed along the time of sampling. The BaA/(BaA + CHR) ratio rate > 0.35 both in dust and SPM confirmed the presence of emission related to vehicle exhausts (Akyüz and Çabuk 2010). Finally, literature data (Yunker et al. 2002) indicate that the IP/(IP + BghiP) ratios < 0.20 imply petrogenic sources with prevalence of gasoline fuelled cars; rates between 0.20 and 0.50 are typical of petroleum combustion (and diesel trucks), and rates > 0.50 suggest grass, wood and coal combustion. In our case, the mean values were as high as ~0.40 in SPM whilst ranged 0.53–0.78 in dust, supporting the existence of distinct PAH sources for the two kinds of matter.

Parabens, phthalates and psychotropic substances

Total concentrations of 18 individual polar compounds (8 phthalate ester, 4 psychotropic substances, 3 parabens and nonylphenol as mix of isomers) occurring in SPM and dusts are reported in Tables 5 and 6, respectively. Phthalates were ubiquitous in SPM, though 10 out of 13 homologues were

identified and three were never found (dimethyl, dinonyl and didecyl phthalates); the maximum values (0.36–1.8 ng/m³ with a median 0.92 ng/m³, 1.7–3.8 ng/m³ with a median 3.1 ng/m³ and 2.0–4.9 ng/m³ with a median 3.2 ng/m³) were identified in diisobutyl, diethylhexyl and dibutyl phthalates, respectively. The concentrations of other phthalates (benzylbutyl, diethyl and dioctyl homologues) ranged between 0.02 and 0.26 ng/m³ (median 0.08 ng/m³). In dusts, 12 phthalates esters were identified together with four psychotropic substances and *N,N*-diethyl-meta-toluamide (DEET) and nonylphenol (see Table 6). In this case, didecyl and butylbenzyl phalates were found also, and the five most abundant homologues (DEHP, BBP, DIDP, DBP and DNOP) together accounted between 25 and 90% of the total phthalates. High concentrations were found in H1 (DIDP = 138 µg/m²/day), S2 (DEHP = 81 µg/m²/day, DBP = 80 µg/m²/day) and H2 (DNOP = 74 µg/m²/day). The total phthalates ranged from 6.0 to 347 µg/m²/day; the maximum values were found at hospital (H1, H2) and school (S1). The results obtained in this investigation between 0.85 and 31.54 ng/m²/day were low compared to those reported in the literature (e.g. from 71 to 1362 ng/m³ in Paris, France (Moreau-Guigon et al. 2016), from 143 to 2600 ng/m³ in Albany, USA (Tran and Kannan 2015)).

With regard to psychotropic substances affecting SPM, caffeine was the most abundant one, ranging between 55 and

Table 6 Psychotropic substances, nonylphenol and phthalates in different micro-environment in indoor dust (ng/m²/day)

| Samples | S1 | S2 | RC | R1 | R2 | H1 | H2 | H3 | H4 | H5 | H6 | M1 |
|-------------|--------|-------|-------|--------|---------|--------|--------|-------|-------|-------|--------|-------|
| Caffeine* | 10.53 | 12.16 | 9.91 | 12.59 | 4.22 | 113.42 | 22.46 | n.d | 28.39 | 31.49 | 13.34 | 8.61 |
| Cocaine* | n.d | n.d | n.d | 2.51 | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d |
| THC* | 0.16 | n.d | n.d | 0.15 | n.d | 1.08 | n.d | n.d | n.d | n.d | 9.73 | n.d |
| Cannabinol* | 0.06 | n.d | n.d | n.d | n.d | 0.18 | n.d | n.d | n.d | n.d | n.d | n.d |
| Heroin | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d | n.d |
| NOP | 16.54 | 5.52 | 26.75 | 111.27 | 2239.36 | 7.25 | 17.34 | n.d | 7.49 | 10.46 | 131.05 | 5.25 |
| DEET | n.d | n.d | n.d | n.d | n.d | 1.07 | 3.95 | n.d | n.d | n.d | n.d | n.d |
| DEHA | 12.90 | 0.36 | n.d | 0.38 | n.d | 2.04 | n.d | 0.33 | n.d | n.d | <LOQ | n.d |
| BBP | 36.55 | n.d | n.d | <LOQ | n.d | 47.96 | 59.65 | n.d | n.d | 0.19 | <LOQ | n.d |
| DDEP | 0.94 | 0.14 | 0.58 | 0.13 | <LOQ | 1.90 | 13.06 | 0.25 | 0.10 | n.d | 0.51 | 0.16 |
| DEHP | 81.23 | 4.92 | 10.12 | 9.22 | 0.60 | n.d | 0.87 | 2.68 | 4.39 | 3.86 | 9.34 | 4.60 |
| DEP | 1.56 | n.d | n.d | 0.51 | <LOQ | n.d | n.d | 0.81 | 0.45 | 4.66 | 0.40 | <LOQ |
| DIBP | 41.35 | 2.62 | 2.99 | 1.40 | 1.36 | n.d | 3.31 | 16.77 | 2.18 | 15.05 | 6.97 | 1.42 |
| DIDP | 65.62 | 4.96 | <LOQ | 3.28 | 1.37 | 137.73 | 5.91 | 1.43 | 1.10 | <LOQ | 4.10 | 1.43 |
| DINP | 7.46 | 1.36 | n.d | 1.14 | 0.54 | 63.36 | 1.78 | 0.66 | 0.27 | <LOQ | 1.87 | 0.25 |
| DMP | n.d | n.d | n.d | n.d | n.d | 2.04 | n.d | n.d | n.d | n.d | n.d | n.d |
| DBP | 80.87 | 1.94 | 1.32 | 4.67 | 1.36 | 47.79 | 13.45 | 17.72 | 11.25 | 6.48 | 3.43 | 3.38 |
| DNOP | 5.57 | 1.48 | 3.71 | 0.41 | 0.47 | 33.30 | 74.27 | 2.85 | 2.35 | n.d | 11.15 | 0.31 |
| DOCP | 12.87 | 0.77 | 0.22 | 0.21 | 0.28 | 5.21 | 9.17 | <LOQ | 0.12 | n.d | 0.28 | <LOQ |
| ΣPHT | 346.93 | 18.53 | 18.94 | 21.40 | 5.97 | 341.31 | 181.46 | 43.50 | 22.21 | 30.32 | 38.05 | 11.55 |
| Mean | 31.54 | 2.06 | 3.16 | 1.95 | 0.85 | 37.92 | 20.16 | 4.83 | 2.47 | 4.33 | 4.23 | 1.65 |
| Median | 12.90 | 1.48 | 2.16 | 0.51 | 0.60 | 33.30 | 9.17 | 1.43 | 1.10 | 3.86 | 3.43 | 1.42 |

*pg/m²/day

515 pg/m³, followed by cannabinol (11–81 pg/m³), cocaine (< 1 to 11 pg/m³) and Δ⁹-THC (4.6 to 9.2 pg/m³). Δ⁹-THC was previously found in Spain (Barcelona, 27 pg/m³; Madrid, 44 pg/m³; Postigo et al. 2009), Italy (Rome, 44 pg/m³; Balducci et al. 2009) and Algiers (190 pg/m³; Khedidji et al. 2017a); almost all reported values exceeded those of this study. On the other hand, cocaine was detected for the first time in Algiers, though its occurrence in air was much lesser than in Madrid and Barcelona (480 and 204 pg/m³, respectively; Viana et al. 2010). Caffeine levels in outdoor soots (mean 0.25 ng/m³) were much lower than during previous studies conducted in Bouira province (mean 0.72 ng/m³; Khedidji et al. 2017a). As for dusts, cocaine could be identified and quantified only at R1 (2.5 μg/m²/day) and CBs (THC + CBD) in S1, R1, H1 and H6 (0.2–9 pg/m²/day), and caffeine occurred everywhere at low concentration (4–113 pg/m²/day).

Data about the occurrence of parabens in air are lacking for Algiers and limited in worldwide. We investigated three homologues, i.e. methyl, ethyl and propyl parabens. The three homologues accounted for ca. 16, 55 and 29% of the total; this latter ranged from 0.33 to 0.44 ng/m³ (median 0.36 ng/m³). Nonylphenols were only 1.4–1.8 ng/m³ in SPM, whilst they reached 5.3 ng/m²/day in dust at manufacturing plant and exceeded 2.2 ng/m²/day at home.

Nonetheless, the volatility of both parabens and NOP suggests that higher concentrations occurred in the air, and further investigations will better clarify the total contents of these chemicals in Algiers interiors.

Conclusions

The occurrence of over 40 contaminants were characterised in 12 indoor dusts as well as in total suspended particulate of Algiers. The polar compounds cumulatively accounted ~ 59% of the total substances investigated in dust, whilst the non-polar ones were predominant in outdoor SPM.

Nonylphenol and phthalate esters occurrence changed widely with the micro-environment and were among the most abundant compounds measured; by contrast, *N,N*-diethyl-*m*-*ta*-toluamide was scarce and not always detected.

According to analysis of diagnostic PAH ratios and CPI/*C*_{max} indexes for alkanes, the main sources of organic dust were pyrogenic source and overall vehicle emissions and of organic fraction in SPM were still pyrogenic sources and mixed combustion, although some minor contribution could come from natural sources. As for psychotropic substances,

illicit drugs such as THC, cannabinol and cocaine were detected both indoors and outdoors.

Due to environmental problem emerging from endocrine-disrupting chemicals compounds, and to general lack of knowledge about their occurrence in the atmosphere in general, and overall in Algeria, more extensive investigations are needed about exposure of population to them and the consequent risk for population.

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