




# Influence of electronic cigarette vaping on the composition of indoor organic pollutants, particles, and exhaled breath of bystanders

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## Abstract

The changes of particles and organic pollutants in indoor atmospheres as consequence of vaping with electronic cigarettes have been analyzed. Changes in the composition of volatile organic compounds (VOCs) in exhaled breath of non-smoking volunteers present in the vaping environments have also been studied. The exposure experiments involved non-vaping ( $n = 5$ ) and vaping ( $n = 5$ ) volunteers staying 12 h together in a room ( $54 \text{ m}^2$ ) without external ventilation. The same experiment was repeated without vaping for comparison. Changes in the distributions of particles in the 8–400 nm range were observed, involving losses of nucleation-mode particles (below 20 nm) and increases of coagulation processes leading to larger size particles. In quantitative terms, vaping involved doubling the indoor concentrations of particles smaller than  $10 \mu\text{m}$ ,  $5 \mu\text{m}$ , and  $1 \mu\text{m}$  observed during no vaping. The increase of particle mass concentrations was probably produced from bulk ingredients of the e-liquid exhaled by the e-cigarette users. Black carbon concentrations in the indoor and outdoor air were similar in the presence and absence of electronic cigarette emissions. Changes in the qualitative composition of PAHs were observed when comparing vaping and non-vaping days. The nicotine concentrations were examined separately in the gas and in the particulate phases showing that most of the differences between both days were recorded in the former. The particulate phase should therefore be included in nicotine monitoring during vaping (and smoking). The concentration increases of nicotine and formaldehyde were small when compared with those described in other studies of indoor atmospheres or health regulatory thresholds. No significant changes were observed when comparing the concentrations of exhaled breath in vaping and no vaping days. Even the exhaled breath nicotine concentrations in both conditions were similar. As expected, toluene, xylenes, benzene, ethylbenzene, and naphthalene did not show increases in the vaping days since combustion was not involved.

**Keywords** Volatile organic compounds · Electronic cigarettes · Indoor air pollution · Atmospheric particles · Exhaled breath · Nicotine · Formaldehyde · Polycyclic aromatic hydrocarbons · Vaping · Black carbon

## Introduction

Electronic cigarettes (e-cigarettes) deliver nicotine to the pulmonary system avoiding tobacco burning. They contain mixtures of glycerin and/or propylene glycol that are vaporized by electric heating which releases much less toxic compounds than conventional tobacco smoking (Marco and Grimalt 2015; Logue et al. 2017; Polosa et al. 2017). The use of these

devices has increased considerably which requires full assessment of the products and by-products released. In this context, measurements of indoor air concentrations and indicators of secondhand inhalation may be relevant for the development of specific legal protection guidelines.

Exposure of bystanders to secondhand vaping in indoor atmospheres has been considered in previous studies involving concentration measurements of air particulate matter (PM) of different particle size fractions (Czogala et al. 2014; Schober et al. 2014; Maloney et al. 2015; Soule et al. 2017; Chen et al. 2017). The indoor aerosol size distributions generated by vaping (Schripp et al. 2013; Schober et al. 2014) as well as the composition of polycyclic aromatic hydrocarbons (McAuley et al. 2012; Schober et al. 2014; O'Connell et al. 2015) have also been considered in these approaches. Other studies have been concerned with the nicotine concentrations

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(McAuley et al. 2012; Schripp et al. 2013; Czogala et al. 2014; Ballbè et al. 2017; Schober et al. 2014; O'Connell et al. 2015; Maloney et al. 2015; Chen et al. 2018).

Changes in the indoor concentrations of formaldehyde as consequence of vaping have been measured (Schripp et al. 2013; Schober et al. 2014; O'Connell et al. 2015; Maloney et al. 2015). Some studies have also included the determination of total volatile organic compounds (VOCs) (McAuley et al. 2012; Chen et al. 2017) or analysis of the composition of these mixtures in indoor atmospheres with vaping activity (Schripp et al. 2013; Schober et al. 2014; O'Connell et al. 2015). In some cases, mathematical models have been used for risk assessment of secondhand vapors (Colard et al. 2015; Logue et al. 2017). The results of several of these studies have been compiled in some review reports (Pisinger 2015; Abidin et al. 2017; Kauneliene et al. 2018).

This previous work outlines the need for integrated assessments of bystander exposure to the pollutants released upon vaping activity. This approach has been addressed in the present study. Changes in indoor gas-phase and particle-phase organic pollutants as consequence of vaping have been determined by comparison of the qualitative and quantitative composition in vaping and non-vaping days in a closed room (54 m<sup>2</sup>, 146 m<sup>3</sup>) without direct contact with external air. Air was sampled during 12 h (8 a.m. to 8 p.m.), while ten volunteers were staying inside. The mass concentrations of particles with aerodynamic diameters smaller than 1, 2.5, and 10 µm, PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, respectively, as well as the submicron aerosol distributions have also been considered. In addition, the VOCs in exhaled breath of non-vaping volunteers staying in these indoor atmospheres were analyzed in both vaping and non-vaping conditions. The overall approach provides a comprehensive description of a real case of indoor bystander exposure to secondhand vaping in which these vaping inputs are compared with background pollution levels.

## Materials and methodology

### Sampling design

The room where the measurements were performed was located in the ground floor and did not contain devices that could generate particles or VOCs. Tobacco consumption was strictly prohibited anytime. The windows were closed before, during, and after the experiments. A garden separated these windows from a quiet street by more than 100 m. On 1 day (Saturday, March 12, 2016), no vaping or cigarette smoking was performed. On another day (Saturday, March 19, 2016), half of the volunteers were vaping *ad libitum* during the 12-h period with the vaping mixtures summarized in Table 1. The bystanding volunteers were not smokers

**Table 1** Composition of the liquid mixtures used in the e-cigars of the vaping volunteers

e-Liquid	Power (W)	Nicotine (mg/mL)	Proportion glycerin/propylene glycol
#1	50	3	70/30
#2	70	3	80/20
#3	45	6	50/50
#4	20	3	40/60
#5	15	12	30/70

neither vapers (Table 2). They were the same on both days, which minimized possible differences attributable to individual metabolic variations and increases the representativity of the results despite the small number ( $n = 5$ ).

During the exposure period, atmospheric particles of different size ranges and equivalent black carbon (eqBC) concentrations were analyzed at a time resolution of 5 min. VOCs, PM, and its organic composition were sampled during the 12 h of the experiment and analyzed afterwards. The exhaled air from the volunteers that were not using the e-cigarettes was analyzed for VOCs. This approach provided a first assessment on the influence of use of electronic cigarettes in indoor atmosphere environments and effects of secondhand vaping.

Twelve hours before and after the experiments, the sampling equipment for PM was operating in the room to collect data for reference.

### Analysis of volatile organic compounds in the indoor air

The VOCs analyses followed the methods described in Marco and Grimalt (2015). Indoor VOCs were concentrated by adsorption in stainless steel cartridges (89-mm length, 0.64-cm external diameter) packed with 200 mg of Tenax TA 60/80 mesh (Supelco, Sigma-Aldrich, St. Louis, USA) for the analysis of nicotine and 150 mg Carbotrap C 20–40 mesh, 150 mg of Carbotrap 20–40 mesh, and 150 mg of Carbotrap X 40–60 mesh (Supelco, Sigma-Aldrich) for the analysis of VOCs of lower vapor

**Table 2** Data of the passive smokers participating in the study

Volunteer	Gender	Age	Smoking	Alcohol consumption	Residence
#1	M	60	No	Moderate	Barcelona
#2	M	27	No	Sporadic	Barcelona
#3	F	25	No	Sporadic	Barcelona
#4	M	45	No	Sporadic	Barcelona
#5	F	45	No	Moderate	Barcelona

pressure than nicotine. The adsorption cartridges were conditioned with helium (grade 5N, 100 mL/min) at 320 °C for 2 h and then at 335 °C for 30 min before each use.

Sampling was performed by connection of the cartridges to a low flow sampling pump (Universal PCXR8, SKC, PA, USA) provided with a dual adjustable low flow tube holder (224-26-02, SKC). One channel was maintained at a constant air inlet flow of 40 mL/min (sampled volume 9.6 L) and the other at 25 mL/min (6 L) for collection of nicotine and VOCs, respectively. Cartridges (2 × 12 and 2 × 19) were collected in the first and second days, respectively. The cartridges were placed 1.60 m above the floor. The pump was calibrated before and after each air intake with a Defender 510-L primary flow calibrator (Bios International, NJ, USA). Indoor air was sampled every 4 h in duplicate during the experiments.

The cartridges were analyzed in a Thermal Desorber Unity Series 2/Ultra 50:50 Multi-tube Autosampler (Markes International Ltd., UK) coupled to a gas chromatograph with a mass spectrometry detector. The VOCs were desorbed at 300 °C for 5 min with helium (5 N) at 40 mL/min. All desorbed VOCs were reconcentrated in the cold trap of graphitized carbon without flow diversion (General Purpose Trap; Markes International Ltd., UK) at −20 °C. Subsequently, the trap was heated at 300 °C for 5 min while passing a helium flow of 7.5 mL/min to introduce the VOCs to a transfer line—fused silica deactivated and uncoated capillary (length 1 m, internal and external diameters 0.25 and 0.35 mm, respectively) heated at 200 °C. The split flow during this operation was 6 mL/min which involved a total split ratio of 5:1.

The transfer line introduced the compounds into a gas chromatograph 7890 (GC, Agilent Technologies Inc., Santa Clara, USA) coupled to an Inert XL MSD 5975C Mass Spectrometer. The chromatograph was equipped with a DB-5MS UI capillary column (length 60 m, internal diameter 0.32 mm, film thickness 1 µm, Agilent J&W GC columns). Helium was the carrier gas at a flow of 1.5 mL/min (constant flow mode). The gas chromatograph oven program started at 40 °C maintaining this temperature for 10 min, then increased to 150 °C with a gradient of 5 °C/min and to 210 °C at 15 °C/min (final holding time 10 min). The VOCs were transferred from the chromatograph to the mass spectrometer by a transfer line heated at 280 °C. The mass spectrometer source and quadrupole were heated at 230 °C and 150 °C, respectively. This detector was operated in the electron impact mode and scanned between 30 and 380 amu.

The VOCs were identified from the retention time and library mass spectra (NIST2009, Mass Spectral Search Program, version 2.0f). Quantification was performed by the external standard method. The limits of detection ranged between 0.05 and 0.65 ng. Repeatability ranged between 5.9 and 23%. Recoveries ranged between 92 and 114% (Marco and Grimalt 2015).

## Analysis of formaldehyde and other carbonyl compounds in the indoor air

Air was collected at a constant sampling rate of 200 mL/min using a low flow sampling pump (Universal PCXR8, SKC) equipped with a single adjustable low flow tube holder (224-26-01, SKC). The pump was calibrated before and after each air sampling as described above. Packed glass cartridges with two sections of silica gel impregnated with 2,4-dinitrophenyl hydrazine (2,4-DNPH) with built-in ozone scrubber (226-120, SKC) were used to collect formaldehyde and other carbonyl compounds. The indoor air was sampled (48 L) every 4 h and in duplicate, at a 1.60-m height from the floor. All three sections were separated by glass wool. After sampling, the cartridges and field blanks were closed with polypropylene plugs and stored at ~4 °C protected from sunlight. The cartridges were analyzed by eluting each of the sections of silica gel-DNPH with 3 mL of acetonitrile, discarding the section of the ozone scrubber. The 2,4-DNPH derivatized carbonyls were analyzed by reverse-phase liquid chromatography (1260 Infinity Quaternary LC System, Agilent Technologies) with an ultraviolet detector operated at 360 nm (1260 Infinity Diode Array Detector, Agilent Technologies).

## Analysis of volatile organic compounds in exhaled air

The exhaled breath of the non-vaping volunteers was sampled using Bio-VOC devices (Markes International Ltd., Llantrisant, UK). These volunteers breathed in and out deeply three times, then held their breath for 20 s and blown into the Bio-VOC cartridge through a disposable cardboard tube until all air from the lungs was discharged to collect the alveolar air. The final air remaining in the Bio-VOC was transferred to a cartridge filled with Tenax TA by pushing it with the threaded piston through the cylinder body of the Bio-VOC. This action was repeated four times so that all the VOCs present in the exhaled air were accumulated in the same cartridge. With this procedure, a total volume of 600 mL of exhaled air was collected. On the day without vaping, exhaled breath was collected at 11 a.m. and 7 p.m. On the vaping day, it was collected at 8 a.m., 12 a.m., 4 p.m., and 8 p.m.

## Analysis of particles in indoor air

Particle sizes and quantities in the range of 8–400 nm were analyzed by means of a scanning mobility particle sizer (SMPS, TSI model 3082). PM<sub>1</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> were analyzed by a GRIMM Monitor EDM 107. A MicroAeth AE51 was used for determination of eqBC levels. The temporal resolution of the devices was 5 min.

PM with an aerodynamic diameter lower than 2.5 µm (PM<sub>2.5</sub>) was collected on pre-heated quartz microfiber filters (Pall, 47 mm) with a low volume sampler (LVS, Derenda).

Sample filters were Soxhlet extracted in a mixture of (2:1, v/v) dichloromethane/methanol (60 mL; Merck) for 8 h. Before the extraction, 25  $\mu$ L of an internal standard containing d7-levoglucosan (Cambridge Isotopic Laboratories) and a mixture of deuterated PAHs (PAH mix 9; Dr. Ehrenstorfer) were added. Insoluble particles were removed from the extracts by filtration through glass fiber filters using a syringe. Then, they were concentrated to 0.5 mL under a gentle stream of nitrogen. Extract aliquots of 25  $\mu$ L together with 25  $\mu$ L of bis-(trimethylsilyl)trifluoroacetamide + trimethylchlorosilane (99:1) (Supelco) and 10  $\mu$ L of pyridine (Merck) were evaporated to dryness under a gentle nitrogen flow. The solution was heated at 70 °C during 1 h for derivatization of the saccharides, acids, and polyols to the equivalent trimethylsilyl esters. Before instrumental analysis, 25  $\mu$ L of an internal standard of 1-phenyldodecane was added. The remaining extract was concentrated to nearly dryness under nitrogen and redissolved in 0.5 mL of (9:1 v/v) hexane/dichloromethane (Merck). This solution was purified by column chromatography packed with 1 g of aluminum oxide (Merck) previously activated at 120 °C for 12 h. The analytes were eluted with 4 mL of (9:1 v/v) hexane/dichloromethane and 4 mL of (1:2 v/v) hexane/dichloromethane, respectively. The fractions were collected together and concentrated under a gentle nitrogen flow to 50  $\mu$ L. An internal standard of 1-phenyldodecane was added before the instrumental analysis.

The extracts were analyzed with a gas chromatograph coupled to a Thermo Trace GC Ultra-DSQ II mass spectrometer equipped with a 60-m capillary column (HP-5MS 0.25 mm  $\times$  25  $\mu$ m thick film). The oven temperature started at 60 °C (holding time 1 min) and increased to 120 °C at 12 °C/min and to 310 °C at 4 °C/min (final holding time 10 min). The temperatures of injector, ion source, quadrupole, and transfer line were 280 °C, 200 °C, 150 °C, and 280 °C, respectively. Helium was used as carrier gas at 0.9 mL/min. The mass spectrometer operated in full scan ( $m/z$  50–650) and electronic impact ionization mode (70 eV).

Levoglucosan and nicotine were identified by comparison of the retention times with standards and the  $m/z$  204 and 84 mass fragments, respectively. The quantification was done with external calibration lines. The concentrations were corrected by the recoveries of the d7-levoglucosan ( $m/z$  206) surrogate standard. The limit of quantification was 0.06 ng/ $m^3$ .

The PAHs were identified by retention time in these specific mass fragmentograms: phenanthrene ( $m/z$  178), anthracene ( $m/z$  178), fluoranthene ( $m/z$  202), pyrene ( $m/z$  202), benz[a]anthracene ( $m/z$  228), chrysene ( $m/z$  228), benzo[b+ j+k] fluoranthene ( $m/z$  252), benzo[e]pyrene ( $m/z$  252), benzo[a]pyrene ( $m/z$  252), indeno[1,2,3-cd]pyrene ( $m/z$  276), and benzo[ghi]perylene ( $m/z$  276)). The quantification was performed by the external standard method. The concentrations were corrected by the PAH mix 9 surrogate standard.

The limits of quantification were 0.1 ng/ $m^3$  for the saccharides and nicotine and 0.02 ng/ $m^3$  for the PAHs (Fontal et al. 2015).

## Results

### Particles

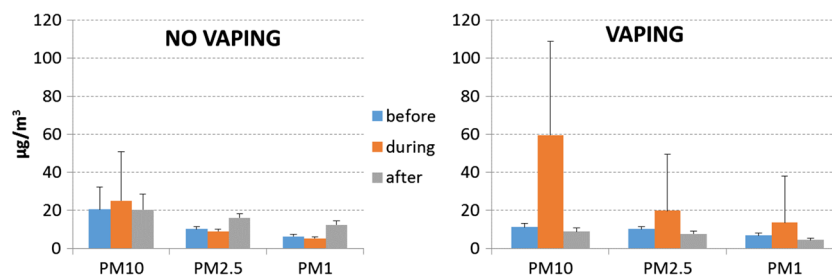
Histograms showing the PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> concentrations on days with and without vaping are shown in Fig. 1. The concentrations measured before and after each experiment with volunteers are also shown. On the day without vaping, the mean indoor air mass concentrations of PM<sub>10</sub> (25  $\mu$ g/ $m^3$ ), PM<sub>2.5</sub> (10  $\mu$ g/ $m^3$ ) and PM<sub>1</sub> (6  $\mu$ g/ $m^3$ ) were not different from the concentrations measured before the experiment with the passive volunteers (Fig. 1). PM<sub>2.5</sub> and PM<sub>1</sub> showed some mass concentration increases, 16  $\mu$ g/ $m^3$  and 12  $\mu$ g/ $m^3$ , respectively, after the experiment.

On the vaping day, the PM mass concentrations before and after the experiment were similar to those of the day without vaping volunteers. During the experiment, higher mean concentrations of all particle sizes were observed, 60  $\mu$ g/ $m^3$ , 20  $\mu$ g/ $m^3$ , and 14  $\mu$ g/ $m^3$  for PM<sub>10</sub>, PM<sub>2.5</sub>, and PM<sub>1</sub>, respectively, than on the non-vaping day. The PM<sub>10</sub> concentrations correlated with PM<sub>2.5</sub> and PM<sub>1</sub> ( $R^2 = 0.44$ ;  $p > 0.001$ ), although the correlations were much stronger between PM<sub>1</sub> and PM<sub>2.5</sub> ( $R^2 = 0.99$ ;  $p > 0.001$ ).

The average particle number concentrations and size distributions of the nanoparticles between 8 and 400 nm recorded on the days with and without vaping are shown in Fig. 2. The distributions measured before and after the presence of the volunteers are also shown. During the experiment with volunteers without vaping, the mean number particle concentration of  $2.8 \times 10^5$  #/ $cm^3$  was 40 times higher than the concentrations before and after both experiments ( $6.9 \times 10^3$  #/ $cm^3$ ). Moreover, in the presence of non-vaping volunteers, the average size range was smaller (14 nm) than before and after performing the experiment (32–62 nm). In the absence of vapor in the air, the formation of very small particles by nucleation processes prevails, and is probably favored by precursors introduced by the non-vaping volunteers (Kulmala and Kerminen 2008; Vartiainen et al. 2006). However, the PM mass concentrations did not increase during the experiment, indicating that these new particles do not add substantially to the mass load.

In contrast, in the presence of vaping volunteers, the average nanoparticle size was larger (32 nm) and the particle distribution was not substantially different from those observed before and after the experiment. In this case, there is a two times increase of particle number concentration ( $9.6 \times 10^3$  #/ $cm^3$ ), compared to before and after the experiment ( $5.2 \times 10^3$  #/ $cm^3$ ).





**Fig. 1** Mean concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, and PM<sub>1</sub> in the atmosphere of the room before (0:00–8:00 h) and during (8:00–20:00 h) vaping (March 19, 2016) and in a reference day without vaping (March 12,

2016). The PM mass concentrations were also measured in the next day following the experiments (8:00–20:00 h) in the absence of any personnel in the room

The eqBC concentrations on days without vaping and vaping were 650 ng/m<sup>3</sup> and 910 ng/m<sup>3</sup>, respectively.

### Polycyclic aromatic hydrocarbons and organic tracer compounds in the particles

The concentrations of PAHs and organic compounds present in the particles collected during the experiments are shown in Table 3. The PAHs were generally found in higher concentration on the vaping day, but benz[a]anthracene, chrysene, and the benzo[fluoranthenes] showed the lowest concentrations on this day. In some cases, e.g., pyrene, the difference involved an order of magnitude. Concerning benzo[a]pyrene, the concentrations during vaping, 0.042 ng/m<sup>3</sup>, were higher than in the absence of this activity, 0.026 ng/m<sup>3</sup>. Total PAH concentrations involved 0.53 and 1.5 ng/m<sup>3</sup>, without vaping and vaping, respectively. In any case, these concentrations were low in comparison to other indoor environments.

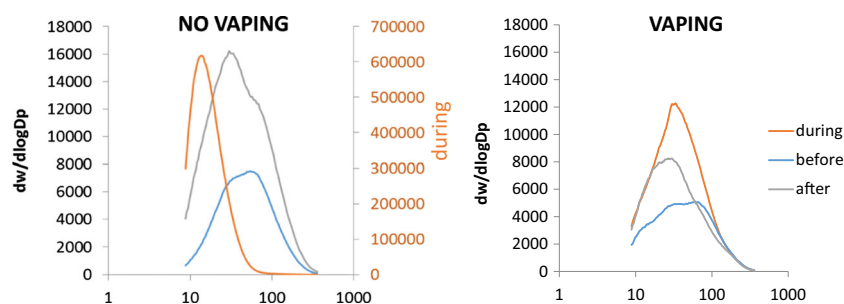
Levoglucosan, a tracer of biomass combustion (van Drooge et al. 2012), was present at low concentrations on the days with and without vaping, 35 ng/m<sup>3</sup> and 53 ng/m<sup>3</sup>, respectively (Table 3). Glycerin was also identified in the particulate matter. As expected, higher concentrations were found on the vaping day, 3300 ng/m<sup>3</sup>, than on the non-vaping day, 170 ng/m<sup>3</sup>. Nicotine was also found in the particulate fraction with higher concentrations on the vaping than on the non-vaping days, 16 ng/m<sup>3</sup> and 0.1 ng/m<sup>3</sup>, respectively.

### Volatile organic compounds

The concentrations of indoor VOCs on the days with and without vaping are represented in Fig. 3 and reported in full in the supplementary material. Most compounds showed the same concentrations on both days. However, formaldehyde, isoprene, nicotine, 2,3-dimethylpentane, 2-butanone, 1,2,4,5-tetramethylbenzene, and bromodichloromethane showed higher concentrations on the vaping day than on the non-vaping day in all analyzed samples. Ethanol, (R)-(+)-limonene, n-heptane, (1S)-(-)-β-pinene, and (1S)-(-)-α-pinene also showed higher concentrations on the vaping day but with some overlap with non-vaping day concentrations.

Some VOCs showed higher concentrations on the non-vaping day but with some overlapping with the vaping day concentrations, e.g., methylene chloride, n-tridecane, styrene, 1,2-dichloroethane, and 1,2-dichlorobenzene.

The mean concentrations, standard deviations, and minimum and maximum concentrations of selected VOCs in indoor air during the vaping and non-vaping days are shown in Table 4. Higher average formaldehyde concentrations were observed on the vaping day than on the previous day, 14 µg/m<sup>3</sup> and 7.4 µg/m<sup>3</sup>, respectively. The nicotine also showed higher average concentrations during vaping, 0.20 µg/m<sup>3</sup>, than on the day without vaping, 0.13 µg/m<sup>3</sup> (Table 4). The differences in mean concentrations of toluene, xylenes, benzene, ethylbenzene, and naphthalene corresponding to the day with vaping and without vaping were not significant.



**Fig. 2** Histograms of the average distribution of the particles in the range of 8 to 400 nm before (0:00–8:00 h), during (8:00–20:00 h), and after (20:00–0:00 h) the presence of the volunteers staying in the room (the experiments) with (March 19, 2016) and without vaping (March 12,

2016). The right y-axis (orange) on the day without vaping corresponds to particle distributions during the experiment with volunteers present in the room

**Table 3** Concentrations (ng/m<sup>3</sup>) of the main organic compounds associated with particles present in the indoor atmosphere

	No vaping March 12, 2016	Vaping March 19, 2016
Glycerin	170	3300
Nicotine	0.1	16
Levogluconan	52	35
Phenanthrene	0.035	0.058
Fluoranthene	0.059	0.15
Pyrene	0.077	0.77
Benz[a]anthracene	0.035	0.019
Crysene	0.045	0.027
Benzo[b+j+k]fluoranthenes	0.10	0.081
Benzo[e]pyrene	0.034	0.049
Benzo[a]pyrene	0.026	0.042
Indeno[123-cd]pyrene	0.06	0.079
Benzo[ghi]perylene	0.066	0.260

### Volatile organic compounds in exhaled air

The VOC concentrations in the exhaled breath of the non-vaping volunteers during vaping and no vaping are represented in Fig. 4 and reported in full in the supplementary material. Nearly all compounds showed the similar concentrations on both days. Ethanol and ethyl acetate showed higher concentrations on the vaping day but with some overlap with non-vaping day concentrations. 1,4-Dichlorobenzene showed higher exhaled breath measurements on the vaping day than on the non-vaping day.

The mean concentrations, standard deviations, and minimum and maximum concentrations of selected compounds are shown in Table 5. The concentrations of benzene, toluene, ethylbenzene, dimethylbenzene, and naphthalene in exhaled breath of the volunteers were very similar, irrespectively of vaping or no vaping which is consistent with the absence of combustion in the vaping process. Concerning nicotine, no significant differences were observed during vaping and no vaping.

### Discussion

In general, the vaping activity under the condition of the present experiment did not imply contaminant increases, and in the cases in which these increases were observed, the concentrations were low.

### Particles

The PM concentrations increased in the presence of vaping volunteers due to the occurrence of vapor components such as

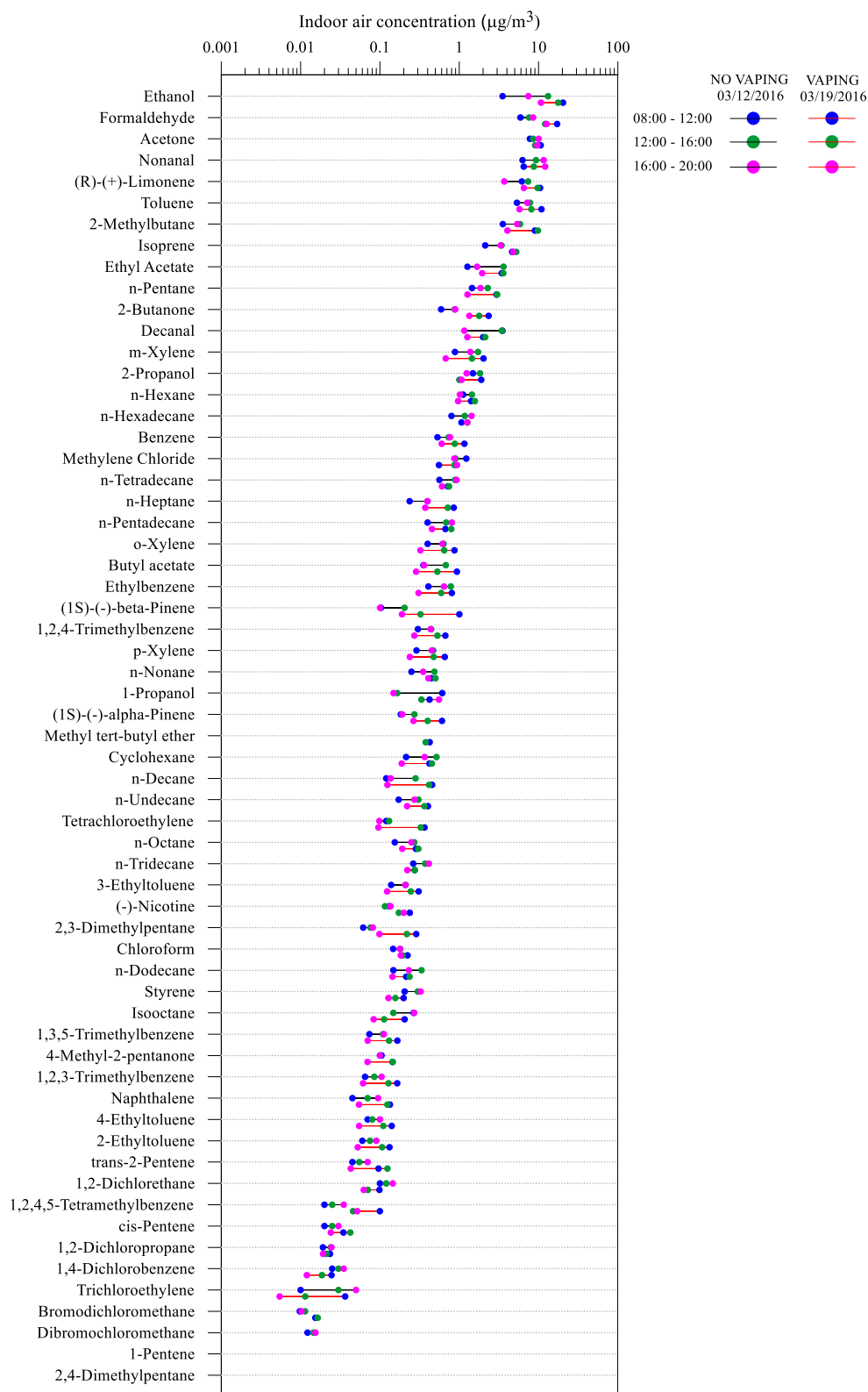
glycerin and/or propylene glycol. The observed PM mass concentrations in this study are comparable with those observed in an experiment with nine volunteers in a closed off room (Schober et al. 2014). Nevertheless, in other studies, with similar experimental setting, much higher concentrations were found in the fine fractions during e-cigarette vaping. In one study, the indoor PM<sub>2.5</sub> concentrations increased from 32 to 152 µg/m<sup>3</sup> (Czogala et al. 2014). In another with second-generation e-cigarettes involving 2-h room session, the PM<sub>2.5</sub> concentrations increased from 18 to 1454 µg/m<sup>3</sup> (Melstrom et al. 2017). The PM<sub>1</sub> concentrations, in a study using third-generation e-cigarettes, increased from 20 µg/m<sup>3</sup> to values between 50 µg/m<sup>3</sup> and 1000 µg/m<sup>3</sup> (Protano et al. 2018). These very high PM concentrations in relatively small rooms were also observed in a larger setting during e-cigarette use (Soule et al. 2017), and in a vaping convention, the observed average PM<sub>10</sub> concentrations were 1800 µg/m<sup>3</sup> (Chen et al. 2017). The doubling of the PM mass concentrations observed in the present study can be related to the emission of particles from the exhaled vapors after puffing which probably may be related to the frequency of vapor puffing, among other conditions, such as the operating power of the e-cigarette (Protano et al. 2018; Sleiman et al. 2017). In any case, real-life vaping studies have shown that e-cigarettes emit notably lower PM<sub>2.5</sub> than conventional smoking (Fernandez et al. 2015).

The mean PM<sub>10</sub> concentration with vaping volunteers in the present study was higher than the 24-h limit value of 50 µg/m<sup>3</sup> for external atmospheres (EU 2008). However, the observed values were within the range of other indoor air environments. The average levels of PM<sub>10</sub> and PM<sub>2.5</sub> in indoor atmospheres of schools in Portugal were 130 µg/m<sup>3</sup> and 82 µg/m<sup>3</sup>, respectively (Madureira et al. 2015). In Barcelona, the average PM<sub>2.5</sub> levels in schools were 34 µg/m<sup>3</sup>, and in bars with and without tobacco smoke, the average PM<sub>10</sub> levels were 150 µg/m<sup>3</sup> and 70 µg/m<sup>3</sup>, respectively (Pey et al. 2013).

Similar particle number concentration and size distribution to those of Fig. 2 were observed in the presence of vaping volunteers in comparable experiments (Schober et al. 2014; Schripp et al. 2013; McAuley et al. 2012), although in those studies, secondary peaks in the particle size range above 100 nm were found, and this was not observed in the present study. The difference could be related to the presence of non-vaping volunteers on the reference day of the present study.

Interestingly, nanoparticle number concentrations in the 8–400 nm range increased more than 50 times in the experiment with no vaping, compared to vaping, especially regarding the nucleation-mode particles (under 20 nm). This difference could be related to formation of nanoparticles by nucleation, a process that is likely favored by precursors introduced by the non-vaping volunteers. The nucleation mechanism may have been depleted in the vaping atmosphere as consequence of the higher particle and vapor concentrations which enhanced coagulation processes (Andreae 2013).

**Fig. 3** Representation of the indoor air concentrations of volatile organic compounds measured on the days without and with vaping. Compounds in the abscissas having no values were analyzed but found below quantification limit



The eqBC concentrations on days without vaping and with vaping volunteers present in the room were  $650 \text{ ng}/\text{m}^3$  and  $910 \text{ ng}/\text{m}^3$ , respectively. These concentrations are slightly lower than those observed in weekdays in indoor air of

schools in Barcelona where motorized traffic is an important BC source and indoor and outdoor concentrations were highly correlated (Rivas et al. 2014). In fact, the increase of the levels between the days with no vaping and vaping was directly

**Table 4** Summary of the concentrations of the main volatile organic pollutants found in the atmosphere ( $\mu\text{g}/\text{m}^3$ )

	No vaping <i>N</i> = 6				With vaping N = 6				WHO limits <sup>*</sup>
	March 12, 2016				March 19, 2016				
	Min	Max	Mean	SD	Min	Max	Mean	SD	
Formaldehyde	5.7	10	7.4	1.7	11	20	14	3.3	100
Toluene	5.0	8.0	6.8	1.3	5.4	11	8.3	2.3	300
m-Xylene	0.83	1.8	1.3	0.39	0.67	2.1	1.4	0.61	200
Benzene	0.53	0.78	0.68	0.12	0.59	1.2	0.88	0.25	
Ethylbenzene	0.39	0.80	0.61	0.17	0.30	0.82	0.57	0.23	
o-Xylene	0.39	0.65	0.55	0.12	0.32	0.89	0.62	0.25	200
p-Xylene	0.27	0.48	0.40	0.09	0.23	0.66	0.46	0.19	200
(-)-Nicotine	0.11	0.15	0.13	0.01	0.16	0.24	0.20	0.03	
Naphthalene	0.04	0.10	0.06	0.02	0.04	0.15	0.10	0.04	10

\*WHO (2010)

related to the outdoor levels of 900 and 1060  $\text{ng}/\text{m}^3$ , respectively, indicating that eqBC was not emitted during exhalation of vapor from e-cigarettes, which is in agreement with other studies (Rupert et al. 2017). In a bar in Barcelona with and without tobacco smoke, the average levels of soot were 4400 and 1400  $\text{ng}/\text{m}^3$ , respectively (Pey et al. 2013) which was considerably higher than those recorded in the present study.

### Polycyclic aromatic hydrocarbons and organic tracer compounds

Increases of 30–90% PAH concentrations have been reported during vaping in relation to non-vaping in a room of 18  $\text{m}^2$  (Schober et al. 2014). No significant increases were observed in other studies (O’Connell et al. 2015). In the present study, the total PAH concentrations on the vaping day were higher than on the non-vaping day, 1.5  $\text{ng}/\text{m}^3$  and 0.53  $\text{ng}/\text{m}^3$ , respectively. However, some PAHs such as benz[a]anthracene, chrysene, and the benzofluoranthenes were in lower concentration during vaping (Table 3). In any case, the PAH concentrations observed on both days are about 100 times lower than those in the study of Schober et al. (2014). On the other hand, the observed concentrations in the present study are similar to the average concentrations of total PAHs in schools of Barcelona, 1.7  $\text{ng}/\text{m}^3$ , but lower than the average concentrations of these hydrocarbons in bars and restaurants in Europe, between 20 and 840  $\text{ng}/\text{m}^3$  (Bolte et al. 2008; Harrison et al. 2009). In a bar from Barcelona, in the presence and absence of tobacco smoke, the total PAH concentrations were 13  $\text{ng}/\text{m}^3$  and 2  $\text{ng}/\text{m}^3$ , respectively (Pey et al. 2013).

Concerning benzo[a]pyrene, the concentration during vaping, 0.042  $\text{ng}/\text{m}^3$ , was higher than in the absence of this activity, 0.026  $\text{ng}/\text{m}^3$ . Again, the concentrations of benzo[a]pyrene observed in the present study are low in comparison to other studies. The average concentrations of this

hydrocarbon in internal atmospheres of schools from Barcelona were 0.09  $\text{ng}/\text{m}^3$  (Mortamais et al. 2017), and the average values in internal atmospheres of European homes were 0.01–0.65  $\text{ng}/\text{m}^3$  (Chuang et al. 1991; Minoia et al. 1997; Fischer et al. 2000; Kingham et al. 2000; Fromme et al. 2004; Gustafson et al. 2008; Harrison et al. 2009), and in North American homes, they ranged between 0.05 and 0.44  $\text{ng}/\text{m}^3$  (Chuang et al. 1991; Mitra and Ray 1995; van Winkle and Scheff 2001; Naumova et al. 2002). Compared to outdoor air quality standards, the detected benzo[a]pyrene concentrations in the room in the present study are much lower than the target value of 1  $\text{ng}/\text{m}^3$  (EU 2008).

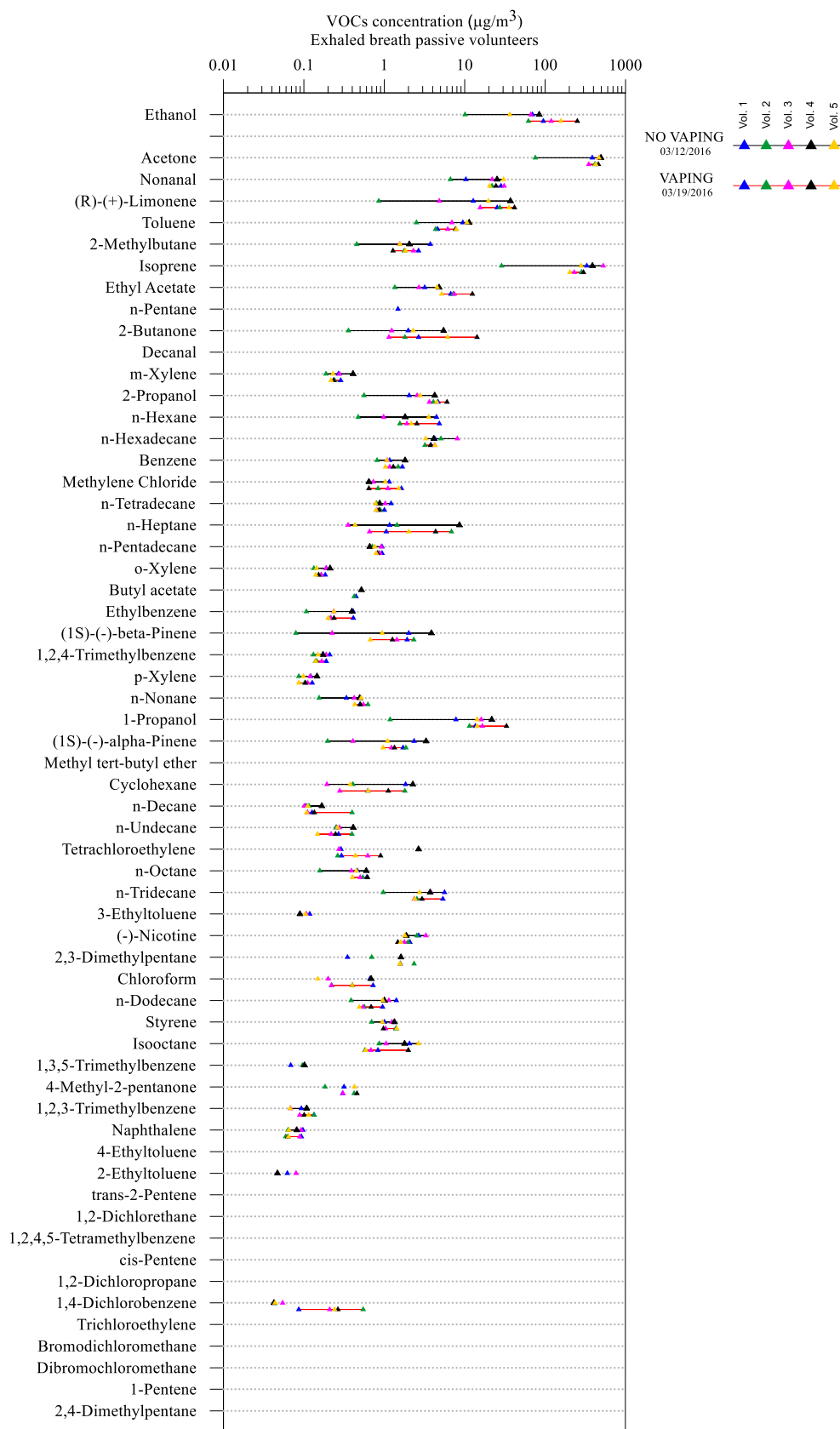
No significant difference in indoor levoglucosan levels was observed between the vaping and non-vaping days. The observed values, 35–52  $\text{ng}/\text{m}^3$ , were typical for outdoor concentrations in winter in Barcelona (van Drooge et al. 2012). In a bar in Barcelona, levoglucosan concentrations of 800  $\text{ng}/\text{m}^3$  and 90  $\text{ng}/\text{m}^3$  were observed in the presence and absence of tobacco smokers, respectively (Pey et al. 2013).

### Volatile organic compounds in indoor air

The VOCs concentrations observed in the present study (Table 4; Fig. 3) were consistent with comparable studies where VOCs were measured in the absence and presence of e-cigarette vapor (Schober et al. 2014). The concentrations of all analyzed VOCs were very low, both on the vaping and non-vaping days. These low concentrations are consistent with previous studies in which emissions from e-cigarettes were measured (Kim and Kim 2015; Marco and Grimalt 2015).

In this sense, the toluene concentrations, 6.8–8.3  $\mu\text{g}/\text{m}^3$ , were much lower than those generally found in indoor atmospheres of European buildings, 20–74  $\mu\text{g}/\text{m}^3$  (Jantunen et al. 1998). Indoor concentrations of this hydrocarbon at levels of





**Fig. 4** Representation of the volatile organic compounds present in the exhaled breath of non-vaping volunteers present in the room on the days with and without vaping. Compounds in the abscissas having no values were analyzed but found below quantification limit. The compounds are represented in the same Fig. 3 abscissa scale for easier comparison of indoor and exhaled breath concentrations

65  $\mu\text{g}/\text{m}^3$  or below are common (WHO 1989; Maroni et al. 1995). The WHO limits of toluene concentration in internal atmospheres are defined as 300  $\mu\text{g}/\text{m}^3$ . This level is defined to avoid health damage due to non-carcinogenic or carcinogenic effects (WHO 2010) and is much higher than those found in the present study (Table 3).

The indoor benzene concentrations of the present study, 0.68–0.88  $\mu\text{g}/\text{m}^3$ , were also below those found in European indoor environments 2.6–5.8  $\mu\text{g}/\text{m}^3$  (Batterman et al. 2007; Sexton et al. 2007; Dodson et al. 2008; Jia et al. 2008) and much lower than those described in some cities such as Milan or Athens, 10–13  $\mu\text{g}/\text{m}^3$  (European Commission 2005). Indoor benzene concentrations in bars and restaurants have been described in the ranges of 5.1–79  $\mu\text{g}/\text{m}^3$  and 1.1–23  $\mu\text{g}/\text{m}^3$ , respectively (Baek et al. 1997; Leung and Harrison 1998; Kim et al. 2001; Guo et al. 2004; Bolte et al. 2008). The concentration limits of benzene in internal atmospheres defined by WHO are 17, 1.7 and 0.17  $\mu\text{g}/\text{m}^3$ , for a cancer every 10,000, 100,000, and 1,000,000 inhabitants throughout life (70 years), respectively.

The cumulative m- and p-xylene concentrations, 1.7–1.8  $\mu\text{g}/\text{m}^3$ , were lower than those found in indoor atmospheres of European buildings, 7.8–37  $\mu\text{g}/\text{m}^3$  (Table 4; Saarela et al. 2003). The total xylenes, 2.3–2.5  $\mu\text{g}/\text{m}^3$ , are also very low in comparison with the recommended concentration values by

WHO (2010), 200  $\mu\text{g}/\text{m}^3$ . The o-xylene concentrations, 0.55–0.62  $\mu\text{g}/\text{m}^3$ , were also lower than those generally found in indoor atmospheres of new buildings in Europe, 8.9–32  $\mu\text{g}/\text{m}^3$  (WHO 1989).

The concentrations of naphthalene, 0.06–0.10  $\mu\text{g}/\text{m}^3$ , were also lower than those found in European buildings, 1–2  $\mu\text{g}/\text{m}^3$  (Edwards et al. 2005). These concentrations are much lower than the maximum concentrations prescribed by the WHO (2010) to avoid human health damage, 10  $\mu\text{g}/\text{m}^3$ .

The concentrations of formaldehyde, 7.4–14  $\mu\text{g}/\text{m}^3$ , were lower than those found in an indoor vaping study with nine volunteers, 30  $\mu\text{g}/\text{m}^3$  (Schober et al. 2014) in which no concentration increases were observed in the presence of e-cigarette vaping. Low concentrations of this compound overlapping background were also observed in two industrial hygiene pilot e-cigarette passive vaping studies (Maloney et al. 2015). The release of formaldehyde from e-cigarette vaping has been attributed to thermal alteration of propylene glycol by voltage increases in the vaporizer (Sleiman et al. 2017). Vaporizing device aging may also result in higher air formaldehyde concentrations (Logue et al. 2017). The formaldehyde concentrations of the present study were also low in comparison with those described in other indoor environments, e.g., 22–170  $\mu\text{g}/\text{m}^3$ , also in the UK (Raw et al. 2004), Germany (Bekanntmachung des Umweltbundesamtes 2008), Helsinki (Jurvelin et al. 2001), and Austria (Kirchner et al. 2007). Concentrations of 3.1–46  $\mu\text{g}/\text{m}^3$  have been observed in German schools (Fromme et al. 2008) and 1.5–50  $\mu\text{g}/\text{m}^3$  in European nursery classrooms (Kotzias et al. 2009). The concentrations observed in the present study are much lower than 100  $\mu\text{g}/\text{m}^3$  that is the WHO (2010) reference level to avoid human damage.

## Nicotine

In the present study, the average gas-phase nicotine measurements were 0.20  $\mu\text{g}/\text{m}^3$  and 0.13  $\mu\text{g}/\text{m}^3$  on the vaping and non-vaping days, respectively, and 16 ng/ $\text{m}^3$  and 0.1 ng/ $\text{m}^3$  for the nicotine in the particulate phase, respectively. Accordingly, the largest increase of nicotine in ambient air from vaping is present in the particulate phase. Comparison of the present results with previous studies should consider whether gas or particulate phase nicotine was measured. The gas-phase measured nicotine concentration in the present study was lower than those observed in a comparable room experiment with nine volunteers and e-cigarettes (0.6–4.4  $\mu\text{g}/\text{m}^3$ ; Schober et al. 2014) and similar to the indoor air concentration in homes of persons using e-cigarettes (0.13  $\mu\text{g}/\text{m}^3$ ; Ballbè et al. 2017). These concentrations were also lower than the concentrations measured in homes with conventional tobacco smokers (2.2  $\mu\text{g}/\text{m}^3$ , Kraev et al. 2009). Reported concentrations in offices, restaurants, bars, and homes in which there was at least one conventional tobacco smoker were 4.1,

**Table 5** Summary of the concentrations ( $\mu\text{g}/\text{m}^3$ ) of the main VOCs detected in the exhaled air of the volunteers present in the room during the absence and presence of vaping

	No vaping				With vaping			
	March 12, 2016				March 19, 2016			
	N = 10				N = 19			
	Min	Max	Mean	SD	Min	Max	Mean	SD
Toluene	2.4	13	8.2	4.0	1.3	15	6.1	3.5
(–)-Nicotine	0.88	3.4	2.4	0.76	1.3	3.7	1.8	0.64
Benzene	0.62	2.7	1.2	0.57	0.96	2.5	1.4	0.43
Ethylbenzene	0.07	0.48	0.28	0.13	0.08	0.46	0.26	0.11
m-Xylene	0.11	0.51	0.27	0.12	0.12	0.35	0.25	0.06
o-Xylene	0.07	0.24	0.17	0.05	0.08	0.23	0.16	0.04
p-Xylene	0.05	0.15	0.11	0.03	0.05	0.15	0.11	0.03
Naphthalene	0.04	0.13	0.08	0.03	0.04	0.17	0.08	0.03

6.5, 19.7, and 4.3  $\mu\text{g}/\text{m}^3$ , respectively (Siegel 1993). Concentrations of 125  $\mu\text{g}/\text{m}^3$  in indoor air were observed during a vaping convention (Chen et al. 2017).

Previous studies reported that increases of the nicotine concentrations in the e-liquid as well as indoor vaporizing e-cigarette consumers result in higher indoor ambient air concentrations (Schober et al. 2014; Logue et al. 2017). The nicotine concentrations in the e-liquids of the present study ranged between 3 mg/mL and 12 mg/mL (Table 1) which is lower than 20 mg/mL, the maximum nicotine concentration set by the European Union's Tobacco Product Directive (EU 2014).

On the other hand, the particulate-phase nicotine concentrations during vaping, 16  $\text{ng}/\text{m}^3$ , were very similar to levels observed in the urban outer atmospheres of Barcelona (van Drooge and Grimalt 2015) and higher than in the absence of vaping, 0.1  $\text{ng}/\text{m}^3$ . The higher difference observed in the particulate phase than in the gas-phase nicotine concentrations between vaping and non-vaping days probably reflects the association of a portion of the released nicotine to the aerosols generated during vaping.

### Volatile organic compounds in exhaled breath

The exhaled air of non-vaping volunteers does not show significant VOCs increases, including nicotine, when they shared the room with vaping volunteers. The strong similarity of the concentrations observed in all individuals further assesses the reliability of the results despite the limited number of volunteers. Vaping does not involve any combustion process so that these compounds are not inhaled when no smokers are located besides the vapers. Studies of exhaled e-cigarette aerosol showed a lack of increase of bystander exposure for phenolics and carbonyls above the levels observed in exhaled breaths of air (Long 2014).

The small concentrations of nicotine in the exhaled breath of the non-vapers both in the vaping and non-vaping days are consistent with a mathematical modeling on bystander exposure to nicotine (Colard et al. 2015). The low concentrations observed in the 2 days of the present study may be explained by the background concentrations of nicotine in urban areas (van Drooge and Grimalt 2015) which involve a baseline exposure of all citizens to this compound.

### Conclusions

The observed changes in airborne organic pollutants in the present experimental approach involving a room with the absence of ventilation in days with vaping and without vaping were generally small. Most VOCs showed the same concentrations on both days. However, isoprene, 2,3-methylpentane, 2-butanone, 1,2,4,5-tetramethylbenzene,

bromodichloromethane, formaldehyde, and nicotine showed higher concentrations on the vaping than on the non-vaping days.

The changes in nicotine concentrations were examined separately in the gas and in the particulate phases showing that the most substantial change was the increase of nicotine in the particle phase during vaping, probably due to condensation to the aerosol generated. Nicotine monitoring studies should also consider the particulate phase for assessment of the vaping (and smoking) airborne impacts.

Changes in the distributions of particles in the 8–400 nm range were observed, involving losses of nucleation-mode particles (below 20 nm) and increases of coagulation processes leading to larger size particles. In quantitative terms, vaping involved doubling the indoor concentrations of  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , and  $\text{PM}_{10}$  observed during no vaping. Black carbon concentrations in the indoor and outdoor air were similar in the presence and absence of electronic cigarette emissions. Changes in the qualitative composition of PAHs were observed when comparing vaping and non-vaping days.

In any case, the concentration increases of nicotine, formaldehyde, and several PAHs were small when compared with those described in other studies of indoor atmospheres from urban zones and were also low if compared with health regulatory thresholds.

No significant change is observed when comparing the concentrations of exhaled breath in vaping and no vaping days. Even in the case of nicotine, the exhaled breath concentrations in both conditions were similar. As expected, toluene, xylenes, benzene, ethylbenzene, and naphthalene did not show increases in the vaping days because no combustion was produced.

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