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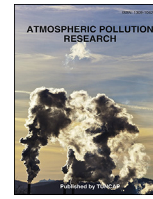

Indoor air quality during sleep under different ventilation patterns

Nuno Canha [a, b, *], Joana Lage [a], Susana Candeias [a, c], Célia Alves [b], Susana Marta Almeida [a]

[a] Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, Km 139.7, 2695-066, Bobadela LRS, Portugal

[b] CESAM - Centre for Environmental and Marine Studies, Department of Environment and Planning, University of Aveiro, 3810-193, Aveiro, Portugal

[c] Faculdade de Ciências e Tecnologia, Universidade Nova de Lisboa, Portugal

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ABSTRACT

Sleep plays a crucial role in the human welfare. This preliminary study aimed to characterise the indoor air quality (IAQ) during sleep, which has been scarcely studied, to better understand the occupant's exposure. Comfort parameters along with indoor air pollutants were assessed in one bedroom during the sleeping period of the occupant. Four scenarios of natural ventilation in the bedroom were studied regarding IAQ. The ventilation setting with door and window closed (CDCW) promoted the lowest air change rate (0.67 ± 0.28 h^{-1}) and the highest levels of carbon dioxide (CO_2), carbon monoxide (CO) and volatile organic compounds (VOCs). Irrespective of ventilation condition, particulate matter levels (PM10, PM2.5) were always high, although maximum values were recorded under CDCW. The simultaneous opening of door and window supplied the highest air change rate (4.85 ± 0.57 h^{-1}). Several pollutants were found to be in concentrations above the established Portuguese guideline for assuring IAQ, namely VOCs, formaldehyde and PM2.5, in specific ventilation settings.

1. Introduction

Indoor air has been a major focus of scientific research worldwide due to evidences of adverse health effects (Sundell, 2004) along with increase of human mortality (Kumar et al., 2005). Moreover, in developed countries, people spend more than 90% of their daily time in indoor environments: home, school, workplace and other indoor places where extra and leisure activities are developed (Almeida-Silva et al., 2014).

Scientific research within the indoor air field has been shifting to evaluate exposure of susceptible groups, based on understandable higher negative impacts that indoor air may play on them, such as children (Annesi-Maesano et al., 2013; Canha et al., 2016), sports practitioners (Ramos et al., 2014, 2015) and elderly (Almeida-Silva et al., 2015; Almeida et al., 2016). However, this research has been focused on micro-environments where people are only during daytime, often neglecting the exposure of occupants during sleeping period, which corresponds around to one third of the lifetime of a person and may have a significant contribution to the total exposure of an individual.

Moreover, sleeping environments are usually characterised by lower ventilation rates (Bekö et al., 2010), promoting pollutants' accumulation (Canha et al., 2016), along with specific exposures taking into account that the breathing area is closer to potential sources of pollutants. In bedrooms, typical specific sources, such as mattresses, emit phthalates, isocyanates and formaldehyde and contribute to the total material emissions (Kemmlein et al., 2003). Additionally, mattresses, pillows and bed linens are often heavily treated with flame-retardants and contain residual detergent components and other substances that are known to have an impact on human health (Anderson and Anderson, 2000; Hoffmann and Schupp, 2009). The mattress is also known to be a great biotope for dust mites (Wu et al., 2012), the faeces of which are a source of allergenic particles. The bed arrangements (blankets, pillows, and mattresses) are also considered as major sources of

* Corresponding author. Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, Km 139.7, 2695-066, Bobadela LRS, Portugal.

E-mail address: nunocanha@ctn.tecnico.ulisboa.pt (N. Canha).

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accumulated dust particles, which may become airborne through a process known as resuspension (Spilak et al., 2014). Besides bedding arrangements, sleeping position and human metabolism were also found to have a significant impact on the exposure of the sleeping person (Laverge et al., 2013).

Sleep is essential to help the body recover from both physical and psychological fatigue suffered throughout the day and helps restore energy to maintain bodily functions. A comfortable sleep is also necessary for high productivity during daytime. Several studies have showed the impact of sleep's quality on to the daytime sleepiness (which is a risk factor and promotes lower productivity) in different types of jobs, such as truck drivers (Catarino et al., 2014) and airline pilots (Reis et al., 2016). Athletic performance has also been shown to be negatively associated with sleep deprivation whereas sleep extension promoted its improvement (Thun et al., 2015).

Sleep quality is affected by many factors, such as health and emotional states, bedding conditions, and different environmental conditions (Pan et al., 2012), including noise levels (Halperin, 2014; Pirrera et al., 2014) and temperature (Lin and Deng, 2008; Okamoto-Mizuno and Mizuno, 2012; Lan et al., 2016). Some studies have focused on the impact of outdoor air pollution on sleep. For instance, Fang et al. investigated the association between black carbon (BC), a marker of traffic related air pollution, and sleep parameters among 3821 residents in Boston area and found that BC long-term exposure may be associated with shorter sleep duration in men and those with low socioeconomic status, but also with longer sleep duration in blacks (Fang et al., 2014). Zanobetti et al. also found that sleep efficiency (percentage of time in bed actually asleep) was reduced in relation to short-term elevations in outdoor PM10 in a cross-sectional study using objective measures of sleep (Zanobetti et al., 2010).

The sleep has a vital role in the daily welfare of people but, however, the impact of the quality of the sleeping environment has been scarcely studied (Lan and Lian, 2016; Urlaub et al., 2015). The environmental characterisation enables to understand the factors that may contribute to the degradation of sleep's quality. Strøm-Tejsen et al. found that lower levels of CO_2 during sleep improved significantly the sleep quality and the perceived freshness of the bedroom air by the occupants, along with the next day performance (Strøm-Tejsen et al., 2016). In Peru, Accinelli et al. studied 82 children with lifetime exposures to indoor fuel pollution, from which a group benefited the installation of less-polluting cooking stoves (reduction of PM2.5 concentrations by 74% when compared to traditional stoves) (Accinelli et al., 2014). Accinelli et al. found that those children (with implementation and exclusive utilisation of improved kitchen stoves) showed significant improvements in sleep and respiratory related symptoms, such as difficulty falling asleep, sore throat and morning headache (Accinelli et al., 2014). However, studies focusing on a wider characterisation of indoor air quality during sleep are lacking in the literature. Deeper studies would allow to better understanding the occupants' exposure during this representative period.

The purpose of the present study was to contribute with preliminary information to append to the still emergent and scarce databases, by assessing the indoor air quality (IAQ) during sleep. Designed as a preliminary approach, the study was carried out in one bedroom with different ventilation settings and focused on several parameters, such as carbon monoxide (CO), carbon dioxide (CO_2), formaldehyde (CH_2O), total volatile organic compounds (VOCs), particulate matter (PM10, PM2.5 and PM1) and comfort parameters (temperature and relative humidity). The different ventilation settings were studied in order to evaluate their impact on the pollutant concentrations.

2. Materials/methods

2.1. Study site

The studied bedroom belonged to an apartment located on the third floor of a residential building of an urban area (parish of Pinhal Novo) of Setúbal district, Portugal (Fig. 1). The building was built in 1999 and is located in the vicinity of an avenue with moderate traffic. The inner and outside walls of the building were built with brick and possessed an air box and thermal insulation.

The apartment had an area of 110 m^2 and two bedrooms, as shown in Fig. 2. The floors of the living room, hallway and kitchen were tiled and both bedrooms had parquet wood flooring. All the rooms were equipped with single-hung aluminium windows. The apartment did not have any air supply mechanical system. The kitchen was equipped with a cooking stove and a water heater, both gas powered. The ground floor of the building was occupied by a restaurant.

The study was conducted in bedroom C (Fig. 2), which had an

Fig. 1. Location of the apartment studied I in the urban area of Pinhal Novo (district of Setúbal, Portugal).

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Fig. 2. Left panel - Apartment floor plan: A - living room, B - kitchen, C - bedroom one (object of study), D - bedroom two. Right panel - Photo of the study bedroom (C).

area of 20 m^2 and a height of 2.8 m. The sliding window, with a total area of 1.8 m^2, half of which was openable, faced south and a main road. The floor and all the furniture were made of wood. A few electronic devices (computer, printer) and LED bulbs were regularly used. No carpets were present in the bedroom. The total area of the door was 1.6 m^2. The occupant of the bedroom during the study was a 23 years old non-smoker female. The bedding used was cotton made. It was washed before each ventilation experiment.

2.2. Studied natural ventilation modes

Four different ventilation settings during sleep were studied, namely closed door and closed window (CDCW), closed door and open window (CDOW), open door and closed window (ODCW) and open door and open window (ODOW). The monitoring campaign took place in August 2015 (summer period) on consecutive days: CDCW – days 17, 18 and 19; CDOW – days 20, 21 and 22; ODCW – days 24, 25 and 26; and ODOW – days 27, 28 and 29.

The door of the bedroom faced the hallway. This latter, with an area of 8 m^2 and a height of 2.8 m, connected different house spaces (living room, kitchen, bathroom and second bedroom), whose doors and windows were all closed.

Outdoor relative humidity (RH), temperature and atmospheric pressure were obtained from an online database of a weather station (ID IPINHALN3) located 3.75 km from the apartment (Wunderground, 2016). These meteorological parameters are displayed in Table 1.

2.3. Indoor air quality monitoring

For the monitoring of indoor air quality during sleep, three direct reading apparatus were used: 1) a Graywolf (IQ-610 probe, WolfSense Solutions, USA) to measure CO_2 (measuring range: 0–5000 ppm, accuracy: ± (3% of reading + 50 ppm)), total VOCs (measuring range: 5–20000 ppb, resolution of 1 ppb), CO (measuring range: 0–500 ppm, accuracy: ± 2 ppm < 50 ppm, ± 3% reading > 50 ppm), temperature (measuring range: −25 °C to 70 °C, accuracy: ± 0.3 °C) and RH (range: 0–100%, accuracy: ± 2% < 80%, ±3% > 80%); 2) a Formaldemeter (htV-M, PPM Technology, UK) to measure CH_2O (measuring range: 0–10 ppm, accuracy: 10% at 2 ppm); and 3) a DustTrak monitor (8530 model, TSI, USA) to measure PM10, PM2.5 and PM1 (measuring range: 0.001–400 mg m^{-3}, resolution of ±0.1% for readings of 0.001 mg m^{-3}). The sampling frequency for all monitoring devices was set at 60 s. All instruments were calibrated by certified entities, which calibrated, validated and demonstrated that every equipment was suitable for its intended purpose. For the Formaldemeter device, an *in situ* calibration was done before each monitoring period using a formaldehyde calibration standard, which takes into account the indoor temperature, supplied by the manufacturer.

All monitoring devices were placed at the centre of the bedroom, at a distance of around 1 m from the bed and at 80 cm from the floor, since this height corresponds reasonably to the breathing level of a person lying in bed. For each one of the 4 different ventilation conditions, three sleeping periods were monitored, lasting usually around 6 h each (always between 11PM and 8AM).

The Portuguese Ordinance no. 353-A/2013 (Ordinance no. 353-A/2013, 2013) establishes the limit values for indoor air pollutants along with the monitoring/sampling/analysis methods to be used (Agência Portuguesa, 2015):

(1) CO_2 – non-dispersive infra-red (NDIR);
(2) Total VOCs: a) active sampling on Tenax TA® sorbent, thermal desorption and analysis by gas chromatography using Mass Spectrometer/Flame Ionisation Detector (MS/FID); or sampling by active (b) or passive (c) methods and analysis by gas chromatography;
(3) CO: NDIR or photoacoustic spectroscopy (PAS);
(4) CH_2O: collection from air onto adsorbent cartridges coated with 2,4-dinitrophenylhydrazine (DNPH) and subsequent analysis of the hydrazones formed by high performance liquid chromatography (HPLC) with detection by ultraviolet absorption;
(5) Particulate matter (PM2.5, PM10): using reference gravimetric methods or alternative methods, such as scattered light photometry, optical aerosol spectrometer, condensation particle counter, aerosol electrometer, particle size spectrometer or time-of-flight spectrometer.

In the present study, only CO_2 and particulate matter were monitored using the methods established in the Portuguese

Table 1
Outdoor temperature, relative humidity and atmospheric pressure during the IAQ monitoring campaign under different ventilation settings (average ± standard deviation; minimum – maximum).

Ventilation Setting	Temperature (°C)	Relative Humidity (%)	Atmospheric Pressure (hPa)
CDCW	19.9 ± 0.4 [18.1–24.8]	76.7 ± 4.2 [60.0–82.0]	1017.5 ± 0.9 [1015.5–1019.5]
CDOW	22.1 ± 1.9 [19.7–27.1]	66.1 ± 9.4 [45.0–78.0]	1019.9 ± 0.7 [1018.8–1021.6]
ODCW	19.7 ± 1.2 [16.7–22.0]	76.5 ± 5.5 [60.0–85.0]	1020.1 ± 1.5 [1017.8–1022.6]
ODOW	21.3 ± 1.3 [19.5–27.0]	80.4 ± 5.3 [60.0–86.0]	1018.7 ± 1.1 [1014.4–1020.2]

legislation (Ordinance no. 353-A/2013, 2013), respectively, NDIR and laser photometer based on light scattering technology. The remaining indoor pollutants were monitored using the real time monitors described above due to their portability, noiseless and also because they enable to obtain the temporal variability during the sleeping period.

2.4. Calculation of air change rates

Air change rates (ACHs) (air change per hour, h^{-1}) were calculated for the studied sleeping periods through the use of a computerised tool that relies on the build-up and steady state phases of the CO_2 curve. CO_2 is used as tracer gas since it is readily emitted by the building occupants and is inert. The build-up phase is when the CO_2 emission level is higher than the ACHs and the steady state is the phase when a balance is reached between the CO_2 emission level and the ACHs. The method is based on a novel second-degree solution to single zone mass-balance equation. The computerised tool, developed by Hänninen (2013), is implemented as a Microsoft Excel spreadsheet. Examples of its application are available and fully described in the literature (Canha et al., 2013, 2016; Hänninen et al., 2017).

2.5. Statistical analysis

An analysis of variance of the results was performed using non-parametric statistics at a significance level of 0.050. Mann-Whitney tests and Kruskal-Wallis methods were used for binary independent groups (between two different ventilation conditions) and for multiple independent groups (between all different ventilation conditions), respectively. All these analyses were conducted using the XLSTAT 2014.1.09 software program (XLSTAT).

3. Results and discussion

Table 2 presents the IAQ guidelines established by the Portuguese legislation. Fig. 3 shows the results of the monitored parameters for the four ventilation settings during sleep, with the red line representing the limit value defined by the Portuguese legislation (Ordinance no. 353-A/2013, 2013).

3.1. Comfort parameters

The international guideline ISO 7730 (ISO 7730, 2005) establishes ranges of temperature and relative humidity in indoor environments for the occupants' comfort: 23-26 °C and 30–60%, in the summer period, respectively.

Regarding RH, all ventilation settings showed mean values within the comfort values. However, only the ventilation setting ODCW provided mean temperatures within the recommended range, namely 24.8 ± 1.1°. Due to the very hot weather outside, mean indoor temperatures around 29 °C were registered for other ventilation conditions. Thus, although air renewal by opening windows may contribute to lower indoor air pollutant concentrations, thermal discomfort can be felt, especially under heat waves.

Table 2
Limit values of indoor air pollutants established by the Portuguese Ordinance no. 353-A/2013 (Ordinance no. 353-A/2013, 2013), regarding a daily mean of 8 h.

Parameter	Limit value
PM_{10}	50 μg m^{-3}
$PM_{2.5}$	25 μg m^{-3}
VOCs	600 μg m^{-3}
CO	10 mg m^{-3}
CH_2O	100 μg m^{-3}
CO_2	2250 mg m^{-3}

3.2. Air change rates of the bedroom

Table 3 presents the mean air change rates monitored for each studied ventilation setting. The lowest ACHs were found for CDCW with a mean value of 0.67 ± 0.28 h^{-1}, while the highest ACHs were registered for the setting ODOW, with a mean value of 4.85 ± 0.57 h^{-1}. All studied ventilation settings provided significantly different ACHs between them.

Apart the ventilation setting with closed door and window that promoted the lowest ACH, the remaining ventilation conditions provided mean ACHs above the minimum value of 0.70 h^{-1}, established for bedrooms by EN 15251:2007 (EN 15251:2007, 2007).

Similar values of ACHs in bedrooms can be found in the literature. Du et al. studied the ACHs of bedrooms in 123 households of Detroit, Michigan (USA) during several seasons and found that the yearly ACH mean was 1.7 ± 1.5 h^{-1} (n = 253, with the interquartile range from 0.68 to 2.07 h^{-1}) (Du et al., 2012). This study also presented a seasonal trend regarding the ACH of bedrooms, with spring having lower mean ACHs (1.25 ± 0.87 h^{-1}, n = 76) and summer higher mean ACHs (2.12 ± 2.03 h^{-1}, n = 75).

The same seasonal behaviour was found by Liu et al. in 369 bedrooms of residences in Shanghai (China), where bedrooms in winter had significantly lower ACHs (1.13 ± 2.15 h^{-1}) than in summer (1.96 ± 2.86 h^{-1}) or autumn (2.20 ± 2.69 h^{-1}) (Liu et al., 2016).

Strøm-Tejsen et al. compared the ACHs of 14 dormitory rooms from a Danish university (from September to December). On average, the ACH was 0.17 h^{-1} in bedrooms with closed window, while in rooms with open window the values were 1.8 h^{-1}, i.e. around 10 times greater (Strøm-Tejsen et al., 2016).

Lower ACHs were found in Sweden by Bornehag et al. who studied 390 residences during the cold period and found ACHs lower than 0.5 h^{-1} in children's bedrooms of around 60% of the multi-family houses and 80% of the single-family houses registered (Bornehag et al., 2005).

The ACHs obtained in the present study are within the range reported in the literature, corroborating previous observations. Natural ventilation through an opening (e.g. window or door) promotes a more effective mixing and, consequently, higher ACHs, which are typical in summer. In contrast, keeping the windows closed and the doors shut in winter leads to low ACHs and airtightness, which can cause a rise in internal humidity and exacerbation of pollutant levels (Canha et al., 2016).

3.3. Carbon dioxide

Carbon dioxide data of day 3 under ventilation setting ODCW, as well as the records of day 1 under ventilation setting ODOW, were excluded from the statistical analysis due to erroneous probe responses. Fig. 4 shows the mean temporal variability of CO_2 during sleep for the four studied ventilation settings. All of them provided mean CO_2 values below the limit value of 2250 mg m^{-3} defined by the Portuguese legislation (Ordinance no. 353-A/2013, 2013).

The lowest mean CO_2 concentrations (1080 ± 70 mg m^{-3}, ranging between 950 and 1320 mg m^{-3}) was found for ventilation setting ODOW, whereas the highest mean level was observed for ventilation setting CDCW (1790 ± 360 mg m^{-3}, ranging between 1110 and 2390 mg m^{-3}). Ventilation settings CDOW and ODCW promoted CO_2 means of 1630 ± 310 mg m^{-3} (ranging between 1040 and 2180 mg m^{-3}) and 1150 ± 90 mg m^{-3} (ranging between 1010

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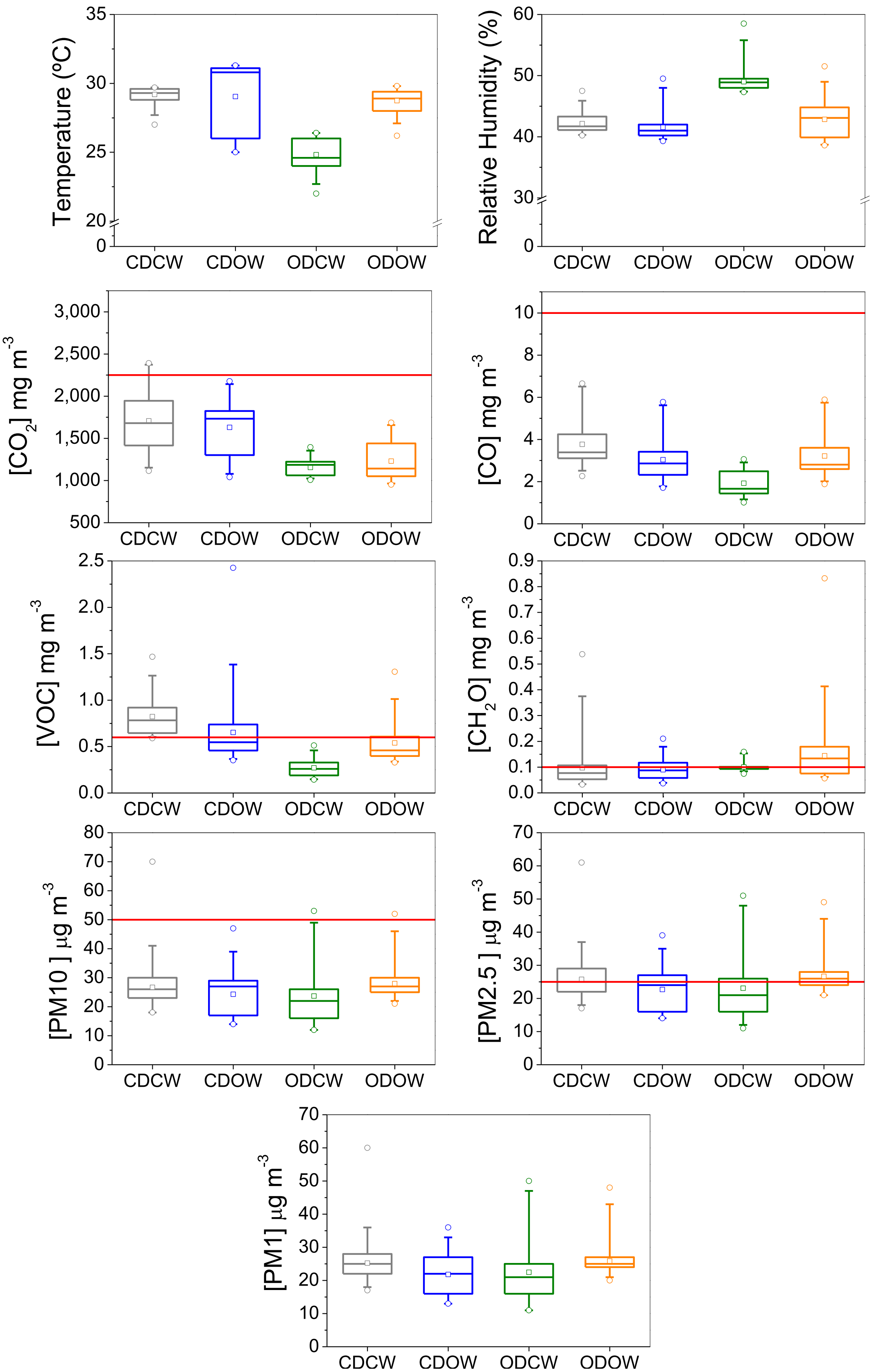

Fig. 3. Levels of indoor pollutants and comfort parameters during sleep under 4 different ventilation conditions (CDCW – closed door and closed window, CDOW – closed door and open window, ODCW – open door and closed window, ODOW – open door and open window). Red line represents the limit value defined by the Portuguese legislation (Ordinance no. 353-A/2013, 2013).

Table 3
Air change rates for each studied ventilation setting; n is the number of used CO_2 events for ACH calculation, n_{bu} refers to the build-up phase and n_{ss} refers to the steady state phase.

ACH (h^{-1})	Ventilation Setting			
	CDCW	CDOW	ODCW	ODOW
Mean ± SD	0.67 ± 0.28	2.20 ± 0.86	3.63 ± 0.75	4.85 ± 0.57
[Min-Max]	[0.46–1.31]	[1.27–4.05]	[2.65–4.81]	[3.95–5.69]
n (n_{bu} + n_{ss})	8 (8 + 0)	8 (8 + 0)	8 (1 + 7)	8 (1 + 7)

and 1390 mg m^{-3}), respectively.

Carbon dioxide is produced exclusively by the occupant's breathing and, therefore, may indicate how well ventilated the bedroom is. Usually, high CO_2 concentrations are associated with low ventilation rates, which promote the indoor accumulation of pollutants, maximising the exposure levels of the occupants (Canha et al., 2016).

Fig. 4 shows the temporal variability of CO_2 levels. As expected, due to the lowest ventilation rates, CDCW promoted a continuous increase of CO_2 levels, even exceeding the Portuguese limit value over the last hour of the sleeping period. The ventilation settings with open window showed higher standard deviations due to typical meteorological variability of outdoor air, such as wind speed and direction.

Only the mean CO_2 level provided by ventilation setting CDCW was slightly higher than the mean value of 1500 mg m^{-3} (835 ppm) described by Strøm-Tejsen et al. (2016). This is the threshold below which the occupants showed significantly improved sleep quality, along with perceived air quality, next-day reported sleepiness and ability to concentrate, when compared to occupants exposed to mean CO_2 levels of 4311 mg m^{-3} (2395 ppm).

3.4. Carbon monoxide

Regardless of the ventilation conditions, CO levels were always below the Portuguese limit value of 10 mg m^{-3}, as shown in Fig. 3 and Fig. S1 (Supplementary information section). The lowest CO mean level was found in ventilation setting ODCW with 1.6 ± 0.3 mg m^{-3} (median of 1.5 mg m^{-3} and ranging between 1.0 and 2.6 mg m^{-3}). The highest CO mean value was obtained under ventilation setting CDCW with 3.8 ± 1.0 mg m^{-3} (median of 3.4 mg m^{-3} and ranging between 2.3 and 6.7 mg m^{-3}). Both ventilation conditions involving open window promoted similar CO levels, with CDOW presenting a mean value of 3.0 ± 1.0 mg m^{-3} (median of 2.9 mg m^{-3} and ranging between 1.7 and 5.8 mg m^{-3}) and ODOW leading to a mean value of 3.5 ± 1.2 mg m^{-3} (median of 3.2 mg m^{-3} and ranging between 1.9 and 5.9 mg m^{-3}).

Carbon monoxide is a toxic by-product of incomplete combustion. It can be generated indoors by combustion processes (e.g. gas cooking appliances) (Mullen et al., 2016) or outdoors due to traffic exhaust emissions (Ramos et al., 2016), or from other human activities, such as smoking (Konstantopoulou et al., 2014). In the present study, probable indoor sources of CO may be the appliances used in the kitchen, such as stove or water heater (both gas fuelled). Carbon monoxide may also have outdoor sources, such as the restaurant located on the ground floor of the building, where, besides the exhaust of cooking appliances, clients only smoke outdoors which may promote CO infiltration through the window and exposure to second hand smoke (SHS). Moreover, the building was located in a street of moderate traffic, which can contribute to CO infiltration into the apartment.

Mullen et al. measured CO levels in 316 kitchens of homes in California and found mean hourly CO levels of 6.4 ppm (7.9 mg m^{-3}) (Mullen et al., 2016), which, as expected, are above those observed in the present study, since typically dilution occurs until CO may infiltrate into the bedroom. However, as stated above, indoor produced CO may penetrate into the bedroom, along with CO from outdoor sources.

Zhong et al. monitored the indoor CO levels in a naturally ventilated and unoccupied bedroom, obtaining mean values of 2.97 ± 0.43 mg m^{-3} (ranging from 2.24 to 4.11 mg m^{-3}) and 2.00 ± 0.19 mg m^{-3} (ranging from 1.71 to 2.52 mg m^{-3}) during weekdays and weekend days, respectively (Zhong et al., 2013). The indoor/outdoor (I/O) ratios were for both cases near 1 (0.990 and 0.985, respectively), which is expected if no indoor CO sources are present, as it was the case. The higher mean CO level during weekdays shows the impact of outdoor local sources, such as traffic

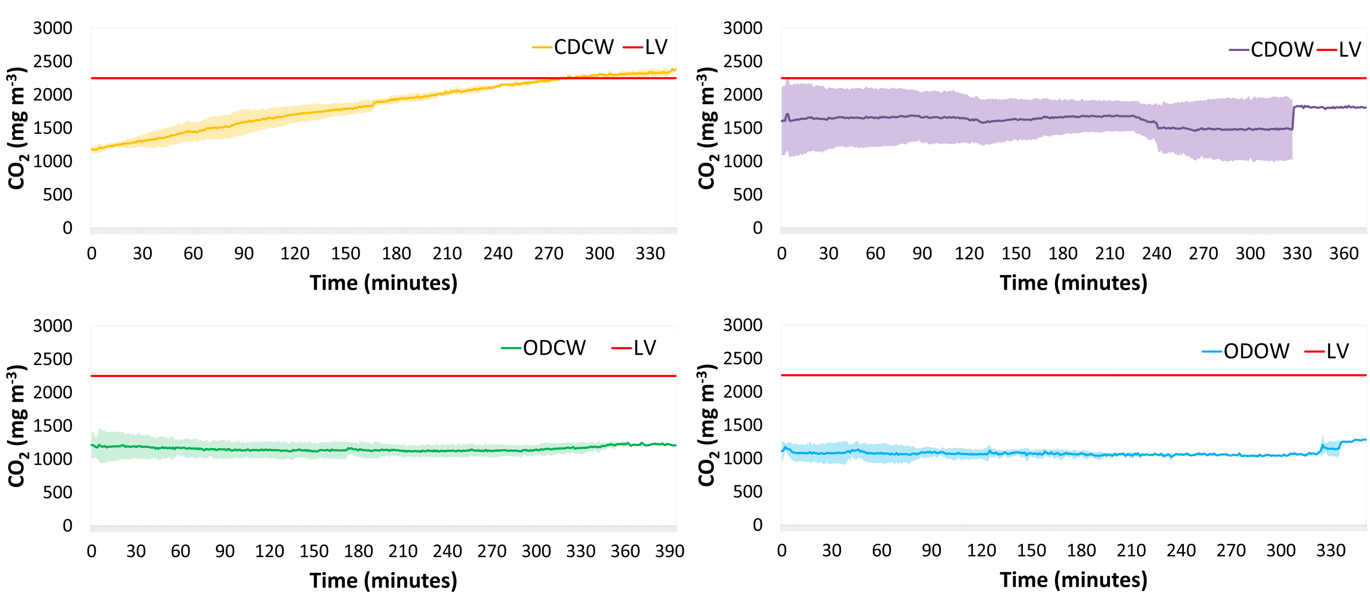

Fig. 4. Temporal variability of CO_2 (line – mean; light coloured area – standard deviation) during the sleeping period for the four studied ventilation settings: (top, left) CDCW, (top, right) CDOW, (bottom, left) ODCW and (bottom, right) ODOW. Red line represents the CO_2 limit value of 2250 mg m^{-3} defined by the Portuguese legislation (Ordinance no. 353-A/2013, 2013).

and smoking outdoors (Zhong et al., 2013). The CO levels found in the present study are similar to the ones described by Zhong et al.

The variability of the present results (high standard deviation in some cases and moments) are likely influenced by meteorological conditions, such as wind velocity and direction, as well as by the intensity and frequency of specific indoor and outdoor sources.

3.5. Volatile organic compounds

The temporal variability of VOC concentrations during sleep under the four ventilation settings is presented in Fig. S2 (Supplementary information section). Global statistics can be found in Fig. 3.

The ventilation setting ODCW promoted the lowest values with a mean value of 0.23 ± 0.08 mg m^{-3} (median of 0.20 mg m^{-3} and ranging between 0.14 and 0.51 mg m^{-3}), while the ventilation setting CDCW conducted to the highest levels with a mean value of 0.82 ± 0.20 mg m^{-3} (median of 0.78 mg m^{-3} and ranging between 0.59 and 1.47 mg m^{-3}).

Both ventilation conditions with open window led to similar VOC levels. Regarding ventilation settings CDOW and ODOW, mean levels of 0.65 ± 0.34 mg m^{-3} (median of 0.55 mg m^{-3} and ranging between 0.35 and 2.43 mg m^{-3}) and 0.58 ± 0.21 mg m^{-3} (median of 0.46 mg m^{-3} and ranging between 0.33 and 0.13 mg m^{-3}) were found, respectively.

Ventilation settings with closed door (CDCW and CDOW) presented mean VOC levels above the limit value of 0.6 mg m^{-3} established by the Portuguese legislation (Ordinance no. 353-A/2013, 2013). Thus, opening the bedroom door seems to help keep the VOC levels below the protection threshold. Common indoor sources of VOCs include household products and building materials, such as paint and varnishes, and cleaning or consumer products (Vicente et al., 2017). Environmental tobacco smoke (ETS) is also a major source of several VOCs. Infiltration from polluted outdoor air is an additional source that may contribute to indoor levels of traffic-generated VOCs.

Almeida-Silva et al. carried out a monitoring campaign in bedrooms of 10 Portuguese elderly care centres (ECCs). The researchers also registered mean VOC levels always above the limit value of 0.1 mg m^{-3}. A mean value of 4.1 ± 1.5 mg m^{-3} was measured in the bedroom with the highest concentrations (Almeida-Silva et al., 2014). Compared to other spaces, such as kitchens or living rooms, bedrooms have been shown to register generally higher VOC levels when comparing (Ohura et al., 2009).

It is important to highlight that the real time monitor of VOCs used in this study was based on a Photo-Ionisation Detection (PID) sensor, while the reference method established by the Portuguese legislation (Ordinance no. 353-A/2013, 2013) is based on FID. Taking into account the differences between both approaches (PID uses isobutylene as reference compound, while FID uses toluene), the results obtained by the PID sensor in this study may be slightly different from the ones that would be obtained using a FID approach (for example, PID does not respond to methane). However, despite the low selectivity of a PID sensor, its use in the specific studied micro-environment has several advantages, such as portability, continuous monitoring and noiseless, which showed to be important for supplying information regarding the exposure of the bedroom's occupant to VOCs.

3.6. Formaldehyde

Fig. S3 (Supplementary information section) presents the temporal variability of CH_2O during the sleeping period for the four studied ventilation settings. Fig. 3 supplies the overall statistics. The ventilation setting that promoted the lowest CH_2O mean level was CDOW with a mean value of 0.090 ± 0.034 mg m^{-3} (median of 0.087 mg m^{-3} and ranging between 0.037 and 0.210 mg m^{-3}), while the ventilation setting ODOW led to the highest mean CH_2O levels with a mean value of 0.205 ± 0.082 mg m^{-3} (median of 0.178 mg m^{-3} and ranging between 0.122 and 0.832 mg m^{-3}).

Regarding ventilation settings CDCW and ODCW, mean levels of 0.097 ± 0.068 mg m^{-3} (median of 0.078 mg m^{-3} and ranging between 0.032 and 0.538 mg m^{-3}) and 0.095 ± 0.006 mg m^{-3} (median of 0.094 mg m^{-3} and ranging between 0.074 and 0.109 mg m^{-3}) were registered, respectively. Only ventilation setting ODOW promoted mean levels above the limit value of 0.1 mg m^{-3} established for formaldehyde by the Portuguese legislation (Ordinance no. 353-A/2013, 2013).

Formaldehyde levels found in the present study are generally higher than those described in the literature. Mullen et al. monitored 340 bedrooms in Californian houses and found a mean value of 0.017 mg m^{-3} (with a 90th percentile of 0.030 mg m^{-3}) (Mullen et al., 2016). Dallongeville et al. obtained a mean level of 0.031 mg m^{-3} (with a 90th percentile of 0.046 mg m^{-3} and ranging from 0.007 to 0.113 mg m^{-3}) in 147 children's bedrooms of French dwellings (Dallongeville et al., 2015). Similar values were reported by Rovira et al. who registered a mean level of 0.027 mg m^{-3} (ranging from 0.011 to 0.048 mg m^{-3}) in bedrooms of 10 houses of Catalonia (Spain) (Rovira et al., 2016).

Almeida-Silva et al. measured CH_2O in bedrooms of 10 ECCs in Lisbon metropolitan area and found mean concentrations ranging from 0.03 to 0.15 mg m^{-3}. Only one of the ECC presented values above the threshold of 0.1 mg m^{-3} (Almeida-Silva et al., 2014).

Formaldehyde is a common indoor air pollutant due to its general use in the production of resins and binders (used on wood-products, such as furniture and plywood), plastics, coatings and paints, flooring materials and others building materials, along with consumer products (Nielsen et al., 2010). Other sources include combustion processes such as smoking, heating or candle burning, among others (World Health Organization (WHO), 2010). Higher emissions rates of formaldehyde are known to be promoted by higher temperature and relative humidity (Haghighat and Bellis, 1998), as confirmed by Uchiyama et al. (2015), who studied 602 Japanese houses, registering mean and maximum CH_2O concentrations of 0.013 and 0.058 mg m^{-3}, respectively, in winter (mean temperature of 17 °C and mean relative humidity of 48%). Mean and maximum CH_2O concentrations of 0.034 and 0.220 mg m^{-3}, respectively, were recorded in summer (mean temperature of 28 °C and mean relative humidity of 63%). Therefore, the high concentrations of formaldehyde registered in the present study may be due to high indoor temperatures, since the monitoring was carried out in summer.

It is noteworthy to consider that the use of the real-time monitoring device Formaldemeter htV-M in the present study may present some limitations, such as a lower specificity for lower concentrations (<0.1 ppm) (Hirst et al., 2011) along with the interference of aldehydes and alcohols (such as acetaldehyde, methanol and ethanol), and phenol in the electrochemical sensor. Therefore, CH_2O concentrations reported in the present study may be overestimated, especially at lower real concentrations. However, the use of such monitoring technique allows to provide insights regarding the concentration's temporal variability during sleep.

3.7. Particulate matter concentrations

The ventilation setting with higher mean PM levels was ODOW, regarding all the studied PM fractions. According to the Portuguese legislation (Ordinance no. 353-A/2013, 2013), mean PM10 levels should not exceed the protection limit of 50 μg m^{-3}, which was verified for all the ventilation settings.

ODOW promoted the highest mean PM10 level with 27.9 ± 4.6 μg m^{-3} (median of 27.0 μg m^{-3} and ranging from 21.0 to 52.0 μg m^{-3}), followed by CDCW with 26.7 ± 5.6 μg m^{-3} (median of 26.0 μg m^{-3} and ranging from 18.0 to 70.0 μg m^{-3}) and then by CDOW with 24.3 ± 6.6 μg m^{-3} (median of 27.0 μg m^{-3} and ranging from 14.0 to 47.0 μg m^{-3}), with ODCW showing the lower mean levels, namely 18.5 ± 4.7 μg m^{-3} (median of 21.0 μg m^{-3} and ranging from 12.0 to 31.0 μg m^{-3}). These results are higher than those reported by Almeida-Silva et al. (2014) who studied the PM10 levels in bedrooms of Portuguese elderly care centres and registered a mean PM10 concentration of 11 μg m^{-3} (total of 4 bedrooms studied, with 2 occupants each). However, Du et al. measured total suspended particles (TSP) in 115 bedrooms (Michigan, USA). TSP concentrations of 28 ± 23 μg m^{-3} were obtained, peaking at 96 μg m^{-3} (Du et al., 2011), which are similar to the PM10 values of the present study.

The protection limit established by the Portuguese legislation (Ordinance no. 353-A/2013, 2013) for PM2.5 is 25 μg m^{-3}. Only two ventilation settings promoted mean PM2.5 levels below this value, namely CDOW (mean of 22.7 ± 5.8 μg m^{-3}, median of 24.0 μg m^{-3} and ranging from 14.0 to 39.0 μg m^{-3}) and ODCW (mean of 17.9 ± 4.5 μg m^{-3}, median of 20.0 μg m^{-3} and ranging from 11.0 to 30.0 μg m^{-3}). Higher PM2.5 levels were found for CDCW (mean of 25.8 ± 5.0 μg m^{-3}, median of 25.0 μg m^{-3} and ranging from 17.0 to 61.0 μg m^{-3}) and ODOW (mean of 26.3 ± 4.3 μg m^{-3}, median of 26.0 μg m^{-3} and ranging from 21.0 to 49.0 μg m^{-3}), slightly above the established limit value.

In Denmark, Sørensen et al. evaluated PM2.5 levels in bedrooms of central areas of Copenhagen during warm and cold seasons and found median PM2.5 concentrations of 9.5 μg m^{-3} (25th Percentile − 6.5 μg m^{-3}; 75th Percentile − 15.5 μg m^{-3}) and 13.4 μg m^{-3} (25th Percentile − 9.4 μg m^{-3}; 75th Percentile − 20.6 μg m^{-3}), respectively (Sørensen et al., 2005). Later, Raaschou-Nielsen et al. registered PM2.5 levels in the bedrooms of 389 Danish infants and found mean concentrations of 19 μg m^{-3} (5th Percentile − 5.7 μg m^{-3}; 95th Percentile − 58 μg m^{-3}) (Raaschou-Nielsen et al., 2011). In Manchester (UK), Gee et al. found mean 5-day concentrations of 19.0 ± 12.4 μg m^{-3} (ranging from 4.7 to 65.1 μg m^{-3}) in children's bedrooms of 69 homes (Gee et al., 2002).

The 1997 revisions to the particulate matter National Ambient Air Quality Standards (NAAQS) in the USA included changes in the reporting method of measured concentrations. Pollutant concentration data compiled in the Environmental Protection Agency (EPA) Aerometric Information Retrieval System (AIRS) prior to the 1997 revisions had been required to be reported in units corrected to standard temperature and pressure (25 °C, 760 mm Hg). This requirement was removed so that, in the new regulations, the PM data will be reported at local temperature and pressure. PM concentrations expressed in terms of local pressure may be a few percent lower than those reported at standard pressure with the largest decreases occurring in high elevation areas. Recently, Chithra and Nagendra (2014) evaluated the impact of outdoor meteorology on indoor PM10, PM2.5, and PM1 concentrations in a naturally ventilated classroom. Maximum PM concentrations were observed under stagnant atmospheric conditions. The highest concentrations of coarse and fine particles in the classroom were associated with strong and low winds, respectively. Among the other meteorological variables, temperature, relative humidity and precipitation showed moderate dependence with indoor PM concentrations. However, solar radiation and atmospheric pressure showed poor correlation with indoor PM. Chan (2002) also reported a weak correlation demonstrating that pressure seems to have very little effect on the I/O PM ratio. However, taking into account that atmospheric pressure is not always reported, comparisons between different works should be done with some precaution.

PM1 levels accounted for, on average, between 96 and 98% of PM2.5 levels, indicating that most of the respirable fraction is mainly composed of submicrometre-sized particles. The lowest mean levels were found for ODCW (17.3 ± 4.5 μg m^{-3}) followed by CDOW, CDCW and ODOW with 21.8 ± 5.6 μg m^{-3}, 25.2 ± 4.8 and 25.5 ± 4.1 μg m^{-3}, respectively.

Table 4 supplies the ratios between particles of different sizes. The fraction of PM1 accounted for 90% or more of PM10 during the sleeping period in all the studied cases. Higher fine-to-coarse particles ratios are expected during sleep since, because coarse particles have higher deposition velocities than the smaller ones (Vicente et al., 2017) and due to the absence or less active emitting sources (Canha et al., 2014). A higher contribution of the coarse fraction to PM10 was found under ventilation conditions with open window. Both ventilation settings (CDOW and ODOW) showed PM2.5/PM10 ratios of 0.94, while for ventilation settings with closed window (CDCW and ODCW), a PM2.5/PM10 ratio of 0.97 was obtained. This suggests outdoor infiltration of coarse particles to the bedroom from outdoors. On the other hand, by opening a window we are allowing air currents to move through the room, causing additional resuspension of coarse particles.

Overall, levels of fine particles found in all ventilation settings were relatively high with mean values nearby the PM2.5 Portuguese limit value of 25 μg m^{-3}.

Indoor PM levels are influenced by emission of specific indoor PM sources (such as, candles or wood burning), activities of the occupants (smoking, vacuuming, cooking involving frying and grilling, movement and cleaning) and outdoor infiltration (Nasir and Colbeck, 2013). However, during sleeping periods, most of the specific indoor PM sources or activities that contribute for PM levels are absent and it can be observed a trend of PM deposition if ventilation is low, such as under CDCW, as shown by the example of Fig. 5-A.

When ventilation is higher, with outdoor infiltration, as verified for ODOW, PM levels remain roughly constant over time, which indicates a continuous exposure to similar PM levels (Fig. 5-B).

In order to evaluate the contribution of the occupant's movements to the PM levels, a test was conducted in a bedroom with the ventilation setting CDCW (similar bedroom, one occupant), where an alarm clock was set during the sleeping period and the occupant was requested to move on the bed by lifting the bedclothes one time. Fig. 5-C shows the PM levels during this test. It is possible to clearly identify the movements of the occupant (red arrows) due to the sudden increase of PM levels (mainly coarse fraction). The movement of the occupant promoted an increase of the PM levels ranging from 22% to 90%, averaging 56%. These results agree with the findings by Spilak et al. (2014), who studied the impact of bedding arrangements, pillows and blankets on particle resuspension during sleep. It was shown that resuspension induced by movements during sleep is comparable to resuspension induced by other human indoor activities, such as walking (Spilak et al., 2014).

3.8. Final considerations

Mean levels above the limit values established by the

Table 4
Ratios between particles of different sizes.

Ventilation Setting	PM1/PM10	PM1/PM2.5	PM2.5/PM10	*n*
CDCW	0.95 ± 0.04	0.98 ± 0.02	0.97 ± 0.03	1072
CDOW	0.90 ± 0.06	0.96 ± 0.03	0.94 ± 0.05	729
ODCW	0.93 ± 0.03	0.96 ± 0.03	0.97 ± 0.02	755
ODOW	0.92 ± 0.02	0.97 ± 0.02	0.94 ± 0.02	946

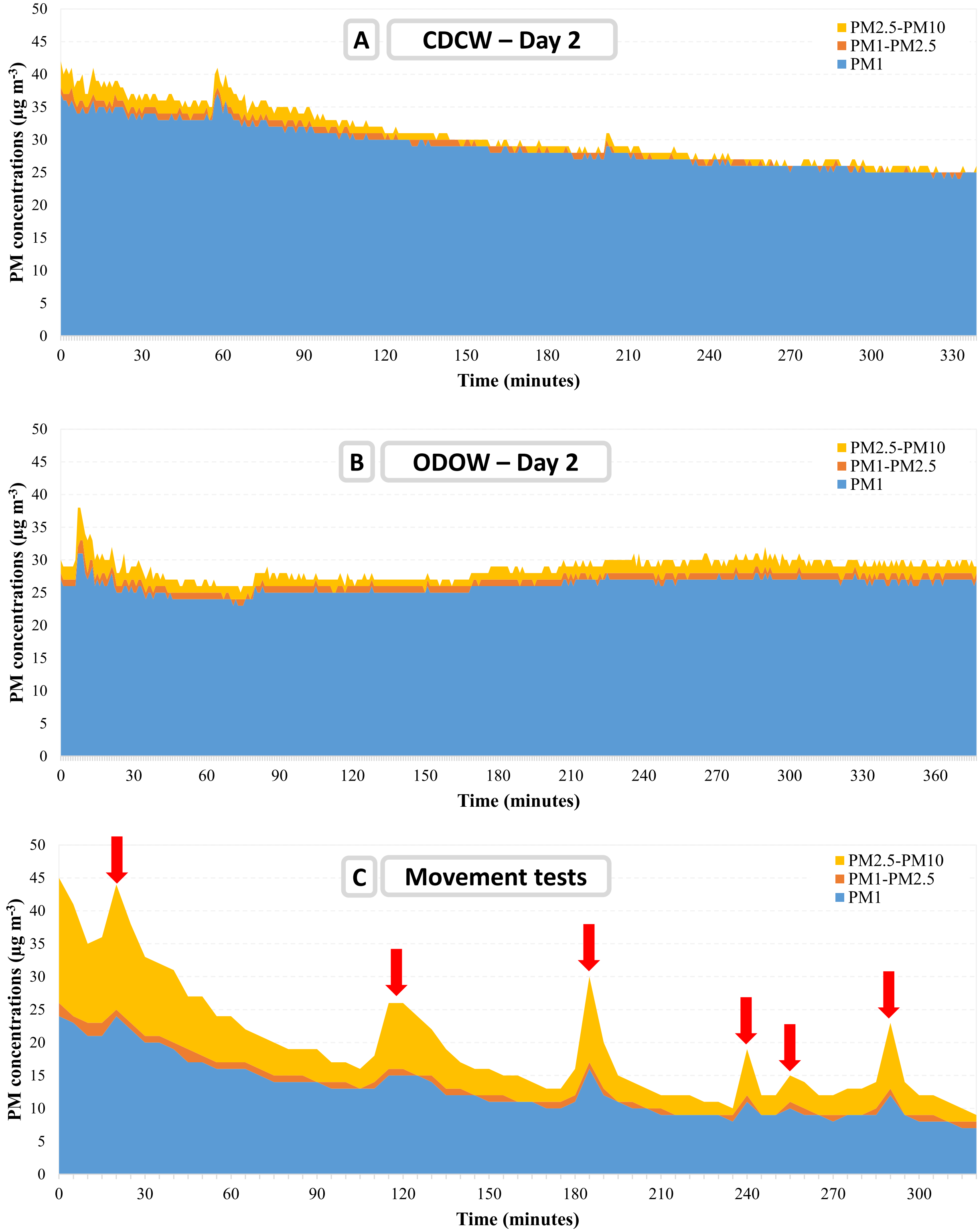

Fig. 5. Temporal variation of PM during sleeping: (A) Ventilation setting CDCW – day 2; (B) Ventilation setting ODOW – day 2 and (C) Movement tests during sleep with ventilation setting CDCW (red arrows indicates scheduled movements).

Portuguese legislation were found for VOCs (for settings CDCW and CDOW), formaldehyde (for settings CDCW and CDOW) and PM2.5 (for settings CDCW and ODOW). Several factors may influence exposure during sleep: 1) emissions from indoor sources in the bedroom or from the rest of the house that penetrate into the bedroom; 2) emissions from outdoor sources that infiltrate into the sleeping environment; 3) ventilation of the bedroom; 4) atmospheric conditions (temperature, relative humidity, wind direction, and other atmospheric conditions) that may contribute to enhanced outdoor levels, and exert an influence on the strength and patterns of infiltration from outdoors5) the variability of emission rates of indoor pollutants (such as formaldehyde or VOCs).

Almeida-Silva et al. found that the exposure of elderly in bedrooms of ECCs contributed, on average, to 70% of the total daily CO_2 exposure (Almeida-Silva et al., 2014). Regarding CO and VOCs, the same researchers concluded that the bedroom represented, at least, 60% and 40%, respectively, of the daily exposure in 9 ECCs amongst the 10 studied (Almeida-Silva et al., 2014). Particulate matter in the bedroom (PM1, PM2.5 and PM10) accounted for, at least, 40% of the overall daily exposure in 80% of the studied ECCs.

Taking into account the great contribution of the sleeping environments to the daily exposure, along with, in some cases, relatively high concentrations of some specific pollutants, as the present study showed, these microenvironments should be carefully considered and studied when evaluating human exposure to contaminants.

The main aim of the present study was to perform a comprehensive characterisation of indoor air during sleep, focusing on different ventilation settings, in order to supply an understanding of the occupant's exposure. However, the study design presents some limitations, such as: 1) lack of monitoring outside the building in order to understand the outdoor contribution to the bedroom's environment; 2) the study was only performed in one bedroom and should be, in a next phase, replicate in a higher number of dormitories to find out the representativeness of the present results; 3) simultaneous characterisation of IAQ in other areas of the house, such as living room and kitchen, to understand its impact on pollutant levels and comfort parameters in the bedroom; 4) evaluation of the sleep's quality and try to figure out the impact of bedroom's IAQ on it. Despite these considerations, the present study supplies a first overview of the IAQ during sleep and the impact of the ventilation on it, showing that these specific micro-environments should not be neglected.

4. Conclusions

Sleeping environments are usually characterised by low air change rates and, as this preliminary study showed, some indoor pollutants, such as VOCs, formaldehyde and PM2.5, may reach concentrations above the established guidelines. Improving natural ventilation, by opening doors and windows, may increase air change rates but, simultaneously, the infiltration of specific pollutants from sources other than the ones in the bedroom, such as from outdoors or from other spaces of the house (e.g. kitchen).

Further research on the indoor air quality during sleep should be conducted in more and different bedrooms in order to understand the exposure of the occupants. Concomitantly, the evaluation of sleep's quality should be done to realise the impact of IAQ on it and, specifically, which are the main pollutants contributing to the degradation of sleep's quality and, therefore, to the occupant's overall well-being.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.05.004.

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