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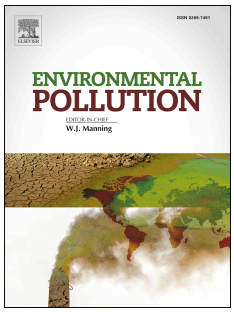

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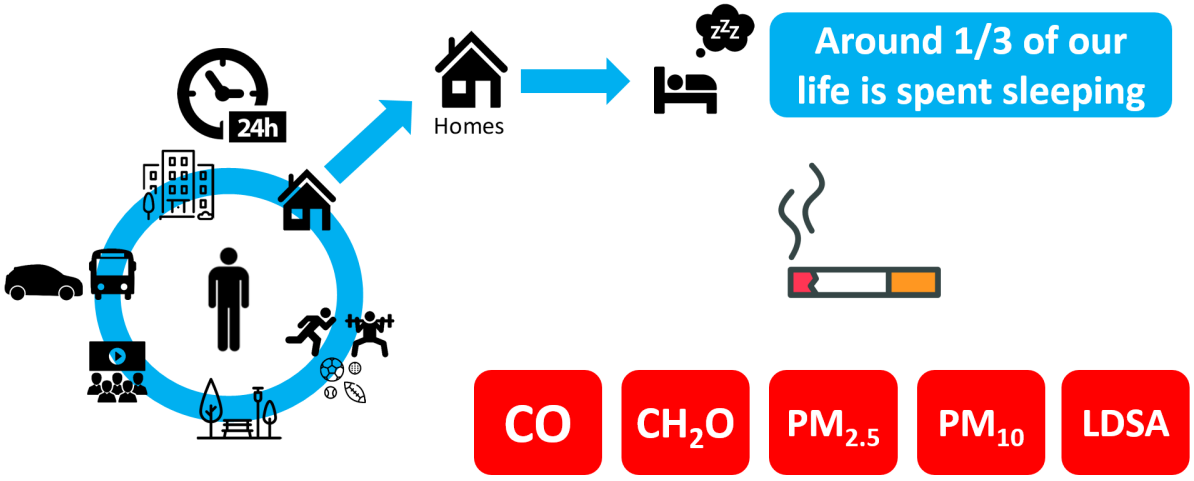

Comparison of indoor air quality during sleep in smokers and non-smokers' bedrooms: a preliminary study

Nuno Canha[1, 2, *], Joana Lage[1], Joana Teixeira Coutinho[1], Célia Alves[2], Susana Marta Almeida[1]

[1] Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, Km 139.7, 2695-066 Bobadela LRS, Portugal
[2] CESAM - Centre for Environmental and Marine Studies, Department of Environment and Planning, University of Aveiro, 3810-193 Aveiro, Portugal

[*]*Corresponding email: nunocanha@ctn.tecnico.ulisboa.pt*

Abstract

People spend one third of their life sleeping, but the bedroom, as a specific micro-environment, is often neglected when assessing human exposure to air pollutants. However, exposure during sleep may be significant in the long-term to the integrated individual exposure. This study aimed to assess the exposure during sleep, focusing on a multi-pollutant approach (comfort parameters, carbon dioxide – CO_2, carbon monoxide – CO, formaldehyde (CH_2O), total volatile organic compounds (VOCs), particulate matter – $PM_{2.5}$ and PM_{10} – and ultrafine particles, particle number concentrations – PNC - and lung deposited surface area - LDSA). For that, the air quality during sleep (in real conditions) was monitored using real-time devices in 12 bedrooms of urban (Lisbon and Vila Franca de Xira) and rural (Ponte de Sor) areas of Portugal for one night. Volunteers were smokers and non-smokers. Considering the Portuguese legislation for indoor air quality (IAQ), 67% of the bedrooms registered CO_2 levels above the limit value, while CH_2O, VOC, PM_{10} and $PM_{2.5}$ thresholds were exceeded in 30, 100, 36, and 45% of cases, respectively. Regarding ultrafine parameters, LDSA and PNC ranged from 7.3 to 95.2 $\mu m^2/cm^3$ and from 0.6 to 4.8 x $10^3/cm^3$, respectively. Even with no smoking indoors, smokers' bedrooms were found to have significant higher levels of CO, CH_2O, $PM_{2.5}$, PM_{10} and LDSA than non-smokers' bedrooms, showing the effect of thirdhand smoke, exhalation of pollutants after smoking and infiltration on the degradation of the air quality in the bedroom. A recent new model of real-time monitor was also used for a wide set of IAQ parameters. Its performance to measure $PM_{2.5}$ and CO_2 was assessed, showing its applicability in real conditions. Although often neglected, these micro-environments should be considered in the integrated individual exposure to air pollutants and further studied.

Main findings of the work: Several pollutants (CO_2, PM, VOCs and CH_2O) exceeded the guidelines during sleep; smokers are exposed to higher levels of CO, CH_2O, PM, and LDSA than non-smokers while sleeping.

KEYWORDS
Indoor air quality; sleep; smoking; exposure; ultrafine particles

1. Introduction

Sleep plays a key-role in human welfare since it promotes body recovery from daily physical and psychological fatigue (Krueger et al., 2016), enables productivity of people (Catarino et al., 2014; Reis et al., 2016) and their athletic performance (Thun et al., 2015). Multiple factors can affect sleep, such as health and emotional states, bedding conditions or environmental factors (Thun et al., 2015), especially temperature (Okamoto-Mizuno and Mizuno, 2012) and noise levels (Halperin, 2014).

Despite sleep has a vital role in daily welfare of people, the impact of the quality of the rest environment has been scarcely studied (Lan and Lian, 2016). Both research issues (sleep and indoor air quality - IAQ) have been addressed in the worldwide scientific literature separately but never fully exploited together. Thus, the impact of indoor air on sleep and all its implications is a task yet to be achieved.

The rest environment should be considered a micro-environment of particular interest due to the following reasons:

1) importance of essential body functions during a sleep period of quality to the human being's welfare, health and daily productivity;

2) exposure to pollutants during sleep may have a great contribution to the daily personal exposure and, moreover, have a greater contribution to long-term exposure, since humans spend about one third of their lives sleeping;

3) low ventilation conditions usually found (Bekö et al., 2010; Canha et al., 2017) may potentiate the accumulation of pollutants, increasing exposure levels.

The environmental characterisation during sleep will enable understanding the factors that may contribute to the degradation of sleep quality and will allow to devise mitigation measures to improve conditions during sleep. Few studies regarding this topic are found in the literature and the few available are focused only on some specific pollutant/parameter. For instance, lower levels of carbon dioxide (CO_2) during sleep were found to significantly improve sleep quality and perceived freshness of the bedroom air by the occupants, together with the performance on the next day (Strøm-Tejsen et al., 2016). Reduction of 74% on $PM_{2.5}$ concentrations in households with indoor fuel pollution were found to improve significantly

children's sleep and respiratory related symptoms, such as difficulty falling asleep, sore throat and morning headache (Accinelli et al., 2014).

One of the challenges of assessing IAQ in a multi-pollutant approach during sleep is the use of standard methodologies since their volume and noise (pumps for air sampling) may interfere with the occupant's sleep (Canha et al., 2014). This issue is especially important for particulate matter (PM).

In 2017, a preliminary multi-pollutant monitoring study in one bedroom evaluated the impact of different ventilation conditions on IAQ while sleeping (Canha et al., 2017). This study revealed that the concentrations of some indoor pollutants, such as formaldehyde (CH_2O), total volatile organic compounds (VOCs) and $PM_{2.5}$, could exceed the established guidelines. The improvement of natural ventilation in sleep environments can be implemented by opening windows or doors to promote the increase of air change rates, which in turn can increase the infiltration of pollutants to the bedroom, such as from outdoors or from other spaces of the house (e.g., kitchen) (Canha et al., 2018).

Smoking is known as an important source of multiple pollutants, both in the gaseous and particulate phases, in indoor environments, which promotes the degradation of air quality (Holcomb, 1993; Kaunelienė et al., 2018; Mueller et al., 2011). However, the impact of indirect smoking (smoking outside the home) on the IAQ during sleep has not been assessed previously. On the other hand, the human exhaled breath, especially of smokers, has been described as a long-neglected pollutant source of several VOCs, nitrogen oxide, carbon monoxide, among others, which may also affect IAQ (Filipiak et al., 2012; Sun and Yang, 2013; Zhang et al., 2013).Therefore, the aim of the present study was to understand the exposure of individuals while sleeping, using a multi-pollutant approach, and to evaluate the difference in exposure between smokers and non-smokers. For that, a strategy was developed using a set of portable monitoring instruments, including a new model, whose performance was assessed. Among the several pollutants studied, a special focus was given to particulate matter and ultrafine particles.

2. Materials/Methods

2.1. Study site and individuals' characterisation

The IAQ during the sleeping period of the occupants was monitored in twelve bedrooms in rural and urban areas of Portugal. The urban areas were in the municipalities of Lisbon and Vila Franca de Xira, while the rural area was located in the municipality of Ponte de Sor.

The occupants of the studied bedrooms were aged between 24 and 53, with six males and six females, 7 non-smokers and 5 smokers. None of the smokers smoked inside the household, but rather outside the building (e.g., balcony). The households ranged from apartment-type (9 cases) in different floors (varying from ground to fifth floor) to detached house-type (3 cases). All bedrooms had natural ventilation, no indoor plants and only one door (to a corridor) and one window. More details about the volunteers and their bedrooms are shown in Table S1 (in Supplementary Information Section). No cleaning procedures were performed during the day prior the night of the monitoring in any studied bedroom. Each bedroom only had one volunteer sleeping during the IAQ monitoring programme. No specific criterion was followed to choose bedrooms, except the availability of volunteers, since the aim was to provide an overview of IAQ during sleep. It was only requested to the volunteers to sleep in similar conditions as they usually sleep, in particular regarding ventilation conditions.

2.2. Indoor air quality monitoring

IAQ assessment was conducted using four different real-time monitoring devices for the selected parameters: i) Graywolf (IQ-610 probe, WolfSense Solutions, USA) for temperature (T), relative humidity (RH), CO_2, carbon monoxide (CO) and total VOCs; ii) Formaldemeter (htV-M, PPM Technology, UK) for formaldehyde (CH_2O); iii) DustTrak DRX monitor (8533 model, TSI, USA) for particulate matter of aerodynamic diameter of 2.5 µm and 10 µm - $PM_{2.5}$ and PM_{10}, respectively; and iv) Pegasor AQTM Indoor Air Quality (Coorstek Amazing Solutions) for T, RH, $PM_{2.5}$, particle number concentration (PNC) and lung deposit surface area (LDSA). Monitoring devices i) to iii) are commonly used in IAQ studies (Canha et al., 2017) and more details about their specifications can be found in the supplementary section (7.1 Indoor air quality monitoring – additional information). Device iv) is a recently launched model in the market that relies on the diffusion charging operating principle for assessing $PM_{2.5}$ (measuring range: 0.001 to 200 $mg.m^{-3}$, resolution of ± 0.1% of reading of 0.001 $mg.m^{-3}$), with a built-in suction pump operating at a flow rate of 3 L/min. Furthermore, this device also allows PNC and LDSA monitoring, along with CO_2, T and RH. The particle size range measured is from 10 nm to 2.5 µm.

All devices were calibrated according to the manufacturers' specifications and the sampling frequency was set to 60 seconds. The monitoring devices were placed at the centre of the bedroom, at approximately one meter from the bed and at about 80 cm from the floor, since this height corresponds reasonably to the breathing level of a person lying in bed. The monitoring period in each bedroom occurred during only one night, usually between 23:00 and 08:00. Depending on the individuals, the sleep period ranged from a minimum of 4h30m to a maximum of 8h45m. The monitoring programme took place from 29 October to 10 November of 2016. For the environmental characterisation of the sleep period, all parameters were reported in relation to their mean values.

Air changes per hour (ACHs, h^{-1}) were calculated for the monitored period using a computerised tool that relies on the build-up phase of the CO_2 curve. This method has already been fully described elsewhere (Hänninen, 2013), along with several examples of its application (Canha et al., 2017, 2016; Hänninen et al., 2017). Table S2 (in Supplementary Information Section) provides the ACHs for each studied bedroom, which ranged from 0.39 ± 0.03 h^{-1} (bedroom 2) to 3.24 ± 0.70 h^{-1} (bedroom 5). These values agree with the ones previously described for different ventilation settings in bedrooms (Canha et al., 2017).

2.3. Statistical analysis

Analysis of data was performed by applying statistics with a significance level of 0.050. To assess the normality of data, the Shapiro-Wilk test was used since all datasets had a number of cases below 30. The results of normality tests for all datasets are available in the supplementary information section (see Table S3 in Supplementary Information). When data was parametric, statistical difference between two independent samples (e.g. smoker vs non-smoker) was evaluated using the *t-test*, while if data was non-parametric the Mann-Whitney test was applied (see Table S4 in Supplementary Information). All statistical analyses were performed by the XLSTAT 2014.1.09 software program.

3. Results and Discussion

3.1. Comparison between devices: Pegasor vs. Graywolf & DustTrak

The performance and comparability of the new model Pegasor was assessed for two parameters, CO_2 and $PM_{2.5}$, against two devices commonly used in IAQ studies, namely Graywolf and DustTrak, respectively. Figure 1 shows the relationships between CO_2 and $PM_{2.5}$ concentrations obtained with Pegasor and the two monitoring devices (Graywolf and DustTrak). For CO_2, all 12 studied cases were used and a very good correlation (R^2 = 0.99)

was found between both instruments. Regarding $PM_{2.5}$, it was not possible to assess concentrations for two (bedrooms 5 and 7) due to operational problems. Additionally, for comparison purposes between monitoring devices, one bedroom (bedroom 11) was excluded from the analysis, since the Pegasor monitor supplied a concentration 11 times higher than the one monitored by the DustTrak, which was taken as an outlier. A good correlation was found for $PM_{2.5}$ levels, with a R^2 value of 0.89, despite the fact that Pegasor provided $PM_{2.5}$ concentrations slightly higher than DustTrak in 67% of the cases.

3.2. Comfort parameters

The mean relative humidity in the 12 bedrooms during the sleep period varied from 43.7 ± 1.2 % to 61.6 ± 1.1%, with a median value of 57.8%. Mean temperatures ranged from 18.4 ± 0.1 ºC to 25.5 ± 0.18 ºC, with a median value of 22.8 ºC among the 12 bedrooms. Considering the international guideline ISO 7730:2005 (ISO 7730:2005, 2005) that establishes, for the colder period, ranges of temperature (20ºC – 24ºC) and relative humidity (30% - 70%) in indoor environments for the occupants' comfort, all bedrooms showed RH mean values within the comfort range. However, only 58% of the bedrooms (7 out of 12) presented temperatures within the comfort range (with one bedroom below the minimum of 20ºC and four bedrooms with temperatures above the maximum of 24ºC).

3.3. Carbon dioxide

Only 33% of the bedrooms (4 out of 12) showed mean CO_2 concentrations below the limit value of 1250 ppm stipulated by the Portuguese legislation for indoor environments (Figure 2). Overall, CO_2 mean concentrations ranged from 553 ± 24 ppm (bedroom 7) to 2671 ± 633 ppm (bedroom 8).

Figure S1 (Supplementary Information – section 7.4) depicts the temporal variability of CO_2 concentrations during the sleep period in bedrooms 7 and 8. A rather constant CO_2 concentration in bedroom 7 can be observed, while levels in bedroom 8 increased successively during the sleep period, reaching a maximum of 3589 ppm (ca. 2.5 times higher than the initial concentration of 1417 ppm). This pattern is due to the different ways of promoting natural ventilation by both occupants. As described in the "Materials/Methods" section, the volunteers were requested to sleep under the usual conditions. Individual of bedroom 7 slept with the door of the bedroom opened and window closed, promoting natural ventilation, while individual of bedroom 8 slept with both door and window closed, contributing to the accumulation of pollutants. Given that the occupants' breathing is the only

significant source of CO_2, these levels reflect different ventilation rates. The impact of opening a door and/or window, during the sleep period, on the pollutant concentrations has already been described in the literature (Canha et al., 2017). Moreover, mean CO_2 levels were significantly different between smokers and non-smokers: 2029 ± 429 ppm and 1123 ± 479 ppm, respectively (Graywolf data). The mean CO_2 levels for smokers were above the limit value (1250 ppm) established by the Portuguese legislation, while values for non-smokers' bedrooms were below the threshold.

Considering the reported threshold of 835 ppm as the value below which the sleep quality is significantly improved, along with perceived air quality, next-day reported sleepiness and ability to concentrate (Strøm-Tejsen et al., 2016), in the present study, only three bedrooms registered levels below this limit (bedrooms 6, 7 and 10).

3.4. Carbon monoxide

The measurement of exhaled CO level may provide an immediate, non-invasive method of assessing smoking status. In a study carried out by Deveci et al. (2004), the exhaled CO levels were measured in 322 subjects (243 healthy smokers, 55 healthy non-smokers, 24 passive smokers). The mean level was 17.13 ± 8.50 ppm for healthy smokers and 3.61 ± 2.15 ppm for healthy non-smokers, and 5.20 ± 3.38 ppm for passive smokers. There was a significant positive correlation between CO levels and daily cigarette consumption, and CO levels and duration of smoking in healthy smokers ($r = 0.550$, p-value < 0.001, $r = 0.265$, p-value < 0.001, respectively). Other studies also confirmed that the level of CO in exhaled air is higher in healthy smokers than in non-smokers (Cunnington and Hormbrey, 2002; Middleton and Morice, 2000; Zhang et al., 2013).

In the present study, CO levels were always below the limit value of 9 ppm (*Ordinance no. 353-A/2013*, 2013) in all bedrooms (Table 1). This was expected since CO is a toxic by-product of incomplete combustion and indoor sources in the bedroom are not supposed to exist. Nevertheless, CO can be generated indoors by combustion processes (e.g., cooking appliances, water heating systems or fireplaces (Canha et al., 2018; Mullen et al., 2016)), by other human activities, such as smoking (Konstantopoulou et al., 2014), but can also originate from outdoor air due to exhaust emissions from traffic (Ramos et al., 2016). Moreover, as described above, low levels of CO are released due to normal human metabolism and due to previous exposure to CO sources, such as smoking (Wu, L., Wang, 2005; Zhang et al., 2013).

Therefore, the detection of CO in the bedroom is likely due to infiltration from other rooms with active sources (e.g. kitchen), penetration of polluted outdoor air and exhaled breath.

Carbon monoxide mean values ranged from undetected to 4.21 ppm, averaging 0.79 ± 0.43 ppm (median of 0.49 ppm). Of the 12 studied bedrooms, only two presented CO levels above 1 ppm. Both bedrooms belonged to smokers. CO infiltration from outdoors will depend on the outdoor levels where the household is located. However, in the present study, no statistical difference between rural and urban dwellings was found (see Table S4, in Supplementary Information).

In a preliminary study on the influence of ventilation in a bedroom during the sleep period on air pollutant levels (Canha et al., 2017) at Setúbal (Portugal), the lowest mean value of CO (1.40 ± 0.26 ppm) was found for the ventilation condition ODCW (open door and closed window), while the highest mean value (3.32 ± 0.87 ppm) was measured with CDCW (closed door and closed window). The mean values of the present study are below those documented in the previous work (Canha et al., 2017) and also below the ones found in a naturally ventilated and unoccupied dormitory room evaluated during weekdays and weekends in Shanghai (Zhong et al., 2013), with mean CO levels of 2.97 ± 0.43 ppm and 2.00 ± 0.19 ppm, respectively.

Figure S2 (Supplementary Information – section 7.5) shows the CO levels in bedrooms of smokers (n = 5) and non-smokers (n = 7). CO levels in smokers' bedrooms were found to be significantly higher than the ones in non-smokers' bedrooms (p-value of 0.006). A mean CO value of 1.60 ± 1.52 ppm (ranging from 0.59 to 4.21 ppm) was registered in bedrooms of smokers during the sleep period, while CO levels approximately 8 times lower (mean value of 0.21 ± 0.22 ppm, ranging from undetectable to 0.50 ppm) were obtained for non-smokers.

Despite none of the volunteers smoked inside the household, some of them had smoked a cigarette one hour prior to their sleep period outside the household (on the balcony or outside the front door). Thus, smoke infiltration from outdoors or the presence of CO in exhaled air may be the reasons justifying the higher levels in the smokers' rooms. As already mentioned, previous studies focused on exhaled carbon monoxide from smokers and non-smokers, using specific devices, showed that smokers exhaled higher levels of carbon monoxide than non-smokers. A study in Poland documented that smokers in a small city (less than 100,000 inhabitants) had mean CO concentrations in their exhaled breath around five times higher than non-smokers (10.77 ± 8.02 ppm and 2.22 ± 1.43 ppm for smokers and non-smokers, respectively), while in a big city (more than 100,000 inhabitants) CO mean concentrations for smokers were about two times higher than for non-smokers (13.54 ± 8.36 ppm and 6.57 ±

8.36 ppm for smokers and non-smokers, respectively) (Maga et al., 2017). Similar results can also be found in studies conducted in China (11.5 ppm and 3.7 ppm for male smokers and non-smokers, respectively) (Zhang et al., 2013) and in Turkey (17.13 ± 8.50 ppm and 3.61 ± 2.15 ppm for smokers and non-smokers, respectively) (Deveci et al., 2004). Therefore, a plausible source of CO during the sleep period may be the air exhaled by smokers.

3.5. VOCs and formaldehyde

Levels of VOCs and formaldehyde monitored in the studied bedrooms are presented in Figure S3 (Supplementary Information – section 7.6). Due to operational problems of the monitoring devices, it was only possible to assess VOC levels in 11 bedrooms (except bedroom 1) and to assess CH_2O levels in 10 bedrooms (except bedrooms 10 and 12).

All monitored bedrooms presented VOC levels above the limit value of 262 ppbv established by the Portuguese legislation, with a mean VOC concentration of 1040 ± 130 ppbv (ranging from 830 to 1230 ppbv), which is around four times higher than the threshold. No statistical differences between VOC levels in bedrooms of smokers and non-smokers were found (smokers: 1070 ± 140 ppbv; non-smokers: 1010 ± 120 ppbv). These levels were all above the maximum VOC concentration of 641 ppbv registered in a preliminary study in a bedroom with only one occupant and restricted ventilation conditions, namely, closed window and door (Canha et al., 2017).

Regarding CH_2O, the limit value of 0.081 ppm established by the national guidelines was only exceeded in three bedrooms (out of 10) with a mean value of 0.060 ± 0.027 ppm (ranging from 0.037 to 0.116 ppm). CH_2O levels in bedrooms of smokers and non-smokers was statistically different, with bedrooms of smokers presenting CH_2O levels two times higher than bedrooms of non-smokers (smoker: 0.087 ± 0.022 ppm; non-smoker: 0.042 ± 0.010 ppm; p-value of 0.014). The mean CH_2O concentration of the present study was below the concentration of 0.073 ppm, which was the minimum recorded in a preliminary study in a bedroom with closed window and door (Canha et al., 2017).

3.6. Particles

3.6.1. Particulate matter (PM)

Figure 3 and Table S5 (Supplementary Information – section 7.7) show the concentrations of $PM_{2.5}$ and PM_{10} monitored during the sleep period in 10 different bedrooms (except bedrooms 5 and 7), using the DustTrak device.

The overall mean $PM_{2.5}$ concentration was 35.1 ± 32.4 µg.m^{-3}, which is above the threshold value stipulated by the Portuguese legislation (*Ordinance no. 353-A/2013*, 2013) of 25 µg.m^{-3} in indoor environments. However, it should be noted that the only bedrooms surpassing this limit value belongs to smokers (Figure 3) , with a mean value of 61.2 ± 24.4 µg.m^{-3}, while for non-smokers the value is around 7 times lower, i.e. 8.9 ± 7.0 µg.m^{-3} (Table S 4).

For PM_{10}, the overall mean value was 39.2 ± 33.8 µg.m^{-3}, not exceeding the national threshold of 50 µg.m^{-3} (*Ordinance no. 353-A/2013*, 2013). Once more, the concentrations found in smokers' bedrooms (67.5 ± 22.8 µg.m^{-3}) were approximately 6 times higher than those of non-smokers (11.0 ± 6.9 µg.m^{-3}). A higher fine mass fraction was observed in smokers' bedrooms compared to non-smokers, with $PM_{2.5}$ accounting for 89 ± 6% of PM_{10} versus 79 ± 19 %, respectively (Table S5).

A preliminary single-room study with a non-smoking occupant in an urban area was designed to evaluate different natural ventilation patterns (focusing on opening of windows and door) and their impact on IAQ. With this purpose $PM_{2.5}$ and PM_{10} were continuously monitored (Canha et al., 2017). The ventilation condition that led to higher PM concentrations was the one with open door and open window (PM_{10} = 27.9 ± 4.6 µg.m^{-3} and $PM_{2.5}$ = 26.3 ± 4.3 µg.m^{-3}), while open door and closed window gave rise to the lowest mean PM concentrations (PM_{10} = 18.5 ± 4.7 µg.m^{-3} and $PM_{2.5}$ = 17.9 ± 4.5 µg.m^{-3}). Therefore, outdoor infiltration may contribute to enhanced PM levels inside bedrooms, which may depend on the type of area where the house is located (urban versus rural, for instance). Although ventilation was not under consideration in the present work, the mean concentrations found in this preliminary study are higher than those reported here for non-smokers, but lower than for smokers. In addition, for non-smokers, the values were similar to the ones reported in a study performed in 4 bedrooms, with 2 occupants each, in Portuguese elderly care centres (Almeida-Silva et al., 2014b), with mean PM_{10} concentrations of 11 µg.m^{-3}.

The $PM_{2.5}$ concentrations provided by the present study are in the range of values reported for UK households of smokers and non-smokers (Semple et al., 2015). The smokers' homes presented a median concentration of 31 µg.m^{-3} (ranging from 10 to 111 µg.m^{-3}, n = 93), whereas this value decreased to 3 µg.m^{-3} in smoke-free homes (ranging from 2 to 6.5 µg.m^{-3}, n = 17). These values were monitored in the living room of the households for 24h, instead of in a bedroom during the sleep period, as it was done in the present study. However, the magnitude of values is similar in both studies, attesting the contribution of smoking to the degradation of IAQ.

3.6.2. Lung deposited surface area (LDSA)

Ultrafine particles are characterised by having a high surface area per mass (Reche et al., 2015). It has been reported that particle surface plays a significant role in determining the toxicological activity of these particles (Reche et al., 2015; Weichenthal, 2012). Lung Deposited Surface Area (LDSA) has been considered as a more relevant potential biological metric in terms of exposure and risk assessment (Levin et al., 2016) since it provides insights into the association between aerosol particle properties and health outcomes (Hama et al., 2017).

Figure 4 presents LDSA concentrations during the sleep period in 11 different bedrooms. Bedroom 11 was not assessed due to operational problems of the monitoring device. The mean LDSA concentration monitored in all studied bedrooms was 30.5 ± 28.3 $\mu m^2.cm^{-3}$ (ranging from 7.3 to 95.2 $\mu m^2.cm^{-3}$). In smokers' bedrooms, a mean LDSA concentration of 49.6 ± 31.7 $\mu m^2.cm^{-3}$ (ranging from 21.4 to 95.2 $\mu m^2.cm^{-3}$) was found, while for non-smokers lower values were obtained, in the range from 7.3 to 33.7 $\mu m^2.cm^{-3}$, averaging 19.5 ± 11.2 $\mu m^2.cm^{-3}$. Mean concentrations of LDSA in the bedrooms of smokers and non-smokers were found to be significantly different (p-value of 0.047).

Table S6 (Supplementary Information – section 7.7) provides an overview of LDSA concentrations in different types of outdoor and indoor environments documented in the literature. Mean outdoor LDSA concentrations ranged from 12 $\mu m^2.cm^{-3}$ (Helsinki, Finland (Kuuluvainen et al., 2016)) to 153 $\mu m^2.cm^{-3}$ (Los Angeles, USA (Ntziachristos et al., 2007)), while mean indoor LDSA levels varied from 10 $\mu m^2.cm^{-3}$ (in a bedroom with two occupants at an elderly care centre in Lisbon, Portugal (Almeida-Silva et al., 2014a)) to 150 $\mu m^2.cm^{-3}$ (at schools in Cassino, Italy (Buonanno et al., 2012)). The LDSA concentrations of the present study are fairly within this interval, ranging from a minimum value of 7.3 ± 1.0 $\mu m^2.cm^{-3}$ (bedroom 9) to a maximum of 95.2 ± 30.4 $\mu m^2.cm^{-3}$ (bedroom 12). However, the results of the present study are lower than those (42 to 140 $\mu m^2.cm^{-3}$) reported for the children's sleep period in Cassino, Italy (Buonanno et al., 2012). The main distinguishable factor between these studies is the fact that the research in Italy was focused on personal exposure, which means that the monitoring device was closer to the children's breathing area while sleeping, whereas this study aimed to assess the LDSA concentrations in the bedroom ambient air, positioning the monitoring device 1 m away from the bed. It has been reported that LDSA concentrations in the personal cloud of the individual are higher than in the surrounding environment (Cattaneo et al., 2010; Licina et al., 2017).

Cooking is a major source of ultrafine particles indoors, as reported by several studies (Geiss et al., 2016). Cooking different types of meals showed LDSA values ranging from 73 ± 7.4 $\mu m^2.m^{-3}$ (baseline) to 890 ± 38.3 $\mu m^2.m^{-3}$ (boiling fish) at an unventilated kitchen (Lisbon, Portugal) (Bordado et al., 2012). In a study conducted in a private house in Ispra (Italy), LDSA concentrations ranging from 19 to 134 $\mu m^2.m^{-3}$, averaging 61 $\mu m^2.m^{-3}$, were obtained in the living room when the woodstove was working (Geiss et al., 2016). Specific activities may also produce high concentrations of LDSA with peaks several orders of magnitude above the usual levels in indoor or outdoor environments, such as incense burning (peak of 872 $\mu m^2.m^{-3}$), candle burning (226 $\mu m^2.m^{-3}$), 3D-printer (72 $\mu m^2.m^{-3}$) and tobacco cigarette (1040 $\mu m^2.m^{-3}$) (Geiss et al., 2016). As shown before, the infiltration of pollutants from other rooms of the house, such as the kitchen, or from the outdoor, to the bedroom, can take place and may promote accumulation of contaminants in this specific micro-environment (Canha et al., 2017). This can explain the significantly high LDSA concentrations found in the present study in the smoker's bedrooms when compared to the non-smoker' bedrooms.

3.6.3. Particle number concentration

Figure 5 presents the particle number concentrations (PNC) during the sleep period in 11 different bedrooms. As described in section 3.6.2., bedroom 11 was not assessed. Mean PNC were found to be (1.7 ± 1.2) x $10^3.cm^{-3}$ in all studied bedrooms, ranging from 0.6 to 4.8 x $10^3.cm^{-3}$. Mean PNC were higher in smokers' bedrooms (mean value of (2.4 ± 1.7) x $10^3.cm^{-3}$, ranging from 1.0 to 4.8 x $10^3.cm^{-3}$) than in non-smokers' bedrooms (mean value of (1.2 ± 0.7) x $10^3.cm^{-3}$, ranging from 0.6 to 2.0 x $10^3.cm^{-3}$). However, mean PNC in smokers and non-smokers' bedrooms were not significantly different (p-value of 0.156).

In a study in 56 residences of non-smokers in Copenhagen (Denmark), a geometric mean of 5.1 x $10^3.cm^{-3}$ was found when the occupants were asleep (Bekö et al., 2013). However, those PNC values were monitored in the living rooms, instead of the bedrooms. Several studies have shown that PNC in households are mainly originated from candle burning and cooking activities (Bekö et al., 2013; Isaxon et al., 2015). No PNC values monitored during the sleep period can be found in the literature, to the best of our knowledge.

3.6.4. Association between $PM_{2.5}$ and LDSA/PNC

Figure 6 shows the correlations of $PM_{2.5}$ with LDSA and PNC. The LDSA concentrations presented an excellent correlation with $PM_{2.5}$ (R^2=0.95). This linear regression can be used to roughly estimate the LDSA concentration in sleep environments based on the $PM_{2.5}$ measurements. In a study carried out in outdoor environments in Helsinki (Finland), the slopes of the $PM_{2.5}$ vs. LDSA regression ranged from 1.8 (residential area – suburban) to 7.2 (traffic site – city centre), increasing with the influence of traffic (Kuuluvainen et al., 2016). PNC presented a good correlation with $PM_{2.5}$ (R^2=0.87) during the sleep period, which can be also used to roughly estimate PNC values in sleep environments from the $PM_{2.5}$ measurements.

3.7. Considerations

An increase number of scientific evidences in the last decades confirmed the negative health impacts of smoking and exposure to secondhand smoke (SHS) (Öberg et al., 2011; United States Department of Health and Human Services, 2014). In order to protect non-smoking population of SHS, several countries worldwide have implemented restrictions on smoking in public areas, establishing minimum distances from doorways where smokers could smoke, non-smoking buildings and smoking bans in specific sites, such in some university campi (DeCarlo et al., 2018). In recent years, a different human exposure route to smoking's products has been studied, namely thirdhand smoke (THS), which is the persistent residue generated from aged SHS that adheres to clothing, indoor dust and surfaces and reemits into the air (Northrup et al., 2016). In a simpler way, THS is the fraction of cigarette smoke that persists in indoor environments after smoking (Hang et al., 2017). It was already showed that early exposure to THS may have a negative health impact on mice, namely regarding their body mass and the development of immunity. More recently, it was also found that skin exposure to an important component of THS can exacerbate pathological features of asthma in mouse (Yu et al., 2018).

The present study showed that the fact of a person is a smoker will somehow constrain the air quality during the sleep period, with some influence of THS, probably due to reemission of SHS previously adsorved to surfaces, such as clothes, hair and skin, as described in previous researches (Bahl et al., 2014).

It is noteworthy to highlight the need to critically evaluate LDSA concentrations since the different available techniques may not be fully comparable (Levin et al., 2016). Furthermore, the impossibility of measuring particle size distributions renders difficult the evaluation of air quality and its effects (Todea et al., 2015). LDSA concentrations of the present study agree

with those described in the literature, which indicates that the instrument is not completely off the scale. However, to obtain firm conclusions on its applicability/reliability it would be necessary to take some "gold standard" reference instrument and run it in parallel.

A limitation of this study is the monitoring of only one night per individual and the small study group of only 12 individuals. Since this research can be classified as a preliminary evaluation, further studies should consider monitoring over several nights to assess the possible variation of pollutant concentrations, as well as a higher number of individuals to increase the population representativeness. Moreover, in future studies, specific VOCs, such as acetone, may have a particular interest to be monitored during sleep since it is a by-product of the human metabolism and is exhaled by breath (from 300 ppbv in healthy individuals to more than 1800 ppbv in diabetics (Righettoni et al., 2012).

Since the use of a portable device, based on a PID sensor, can withdraw selectivity to the measured VOCs as compared to the reference method (active sampling on Tenax TA® sorbent, thermal desorption and analysis by gas chromatography using Mass Spectrometer/Flame Ionisation Detector) (*Ordinance no. 353-A/2013*, 2013), the high values obtained in this study should be taken as indicative and as a warning for a more exhaustive monitoring in the future.

4. Conclusions

This study provided some insights into the IAQ that people are exposed to while sleeping, considering a multi-pollutant approach. IAQ monitoring during sleep is a challenge due to eventual interferences of instruments under operation with the sleep of individuals. However, the strategy adopted showed to be successful, allowing to characterise IAQ during sleep. The Pegasor AQ™ Indoor provided reliable results regarding particulate matter and carbon dioxide, with the advantage of gathering in one easy to use device several parameters for a wider characterisation of IAQ. Overall, this instrument allowed to assess temperature, relative humidity, CO_2, $PM_{2.5}$ and ultrafine particles (focusing on LDSA and PNC).

Taking into account the limit values for some IAQ parameters established by the national legislation, it was found that some non-smoking subjects are exposed to higher VOCs levels, while smokers are exposed to higher values of CO_2, CO, VOCs, CH_2O, $PM_{2.5}$ and PM_{10} during sleep. Taking into account the good correlations between $PM_{2.5}$ concentrations and measurements of either LDSA or PNC, it seems that there is a possibility of constructing predictive models to estimate the latter parameters. However, given the poor sample

representativeness, to confirm this hypothesis, additional measurements involving a thorough analysis of time-series comparisons with more sophisticated instruments would be required.

Despite no smoking was done indoors, the results suggest that smokers exhibit a significant higher exposure to CO, $PM_{2.5}$, CH_2O, PM_{10} and LDSA during sleep than non-smokers.

Further studies regarding exposure to air pollutants during sleep should be conducted involving a wider target group. The preliminary conclusions that people are usually exposed to higher levels of pollutants during sleep, which can greatly contribute to their daily exposure, should be corroborated by additional investigations. Moreover, considering these results, future studies should also focus on the impact of IAQ on the sleep quality of the occupants in order to assess which environmental factor may interfere with a good night of sleep.

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Figures of manuscript

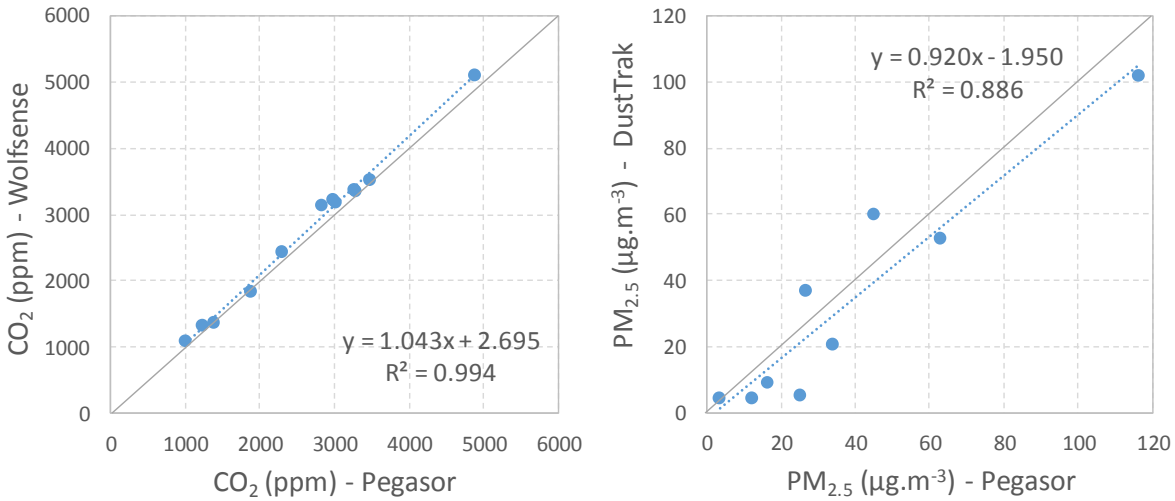


Figure 1. Comparison between devices: (left) CO_2 concentrations by Pegasor and Graywolf; and (right) $PM_{2.5}$ concentrations by Pegasor and DustTrak.

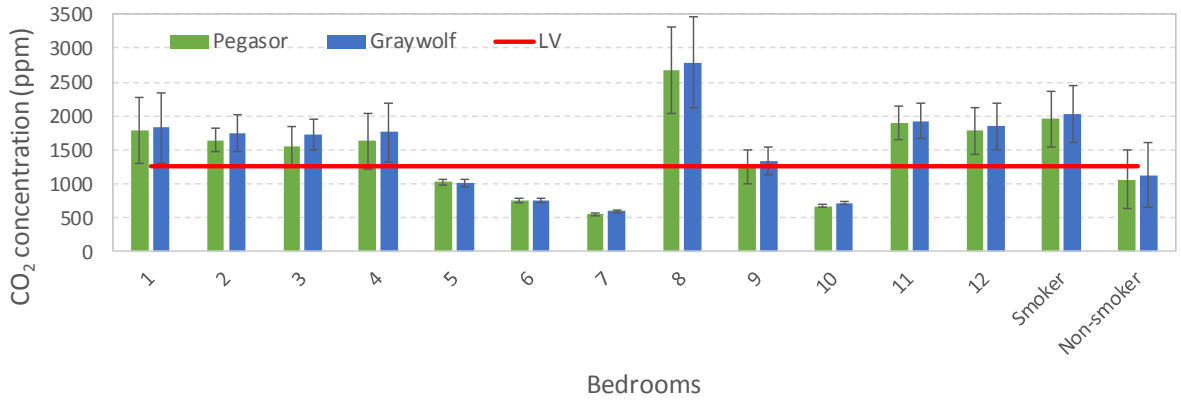
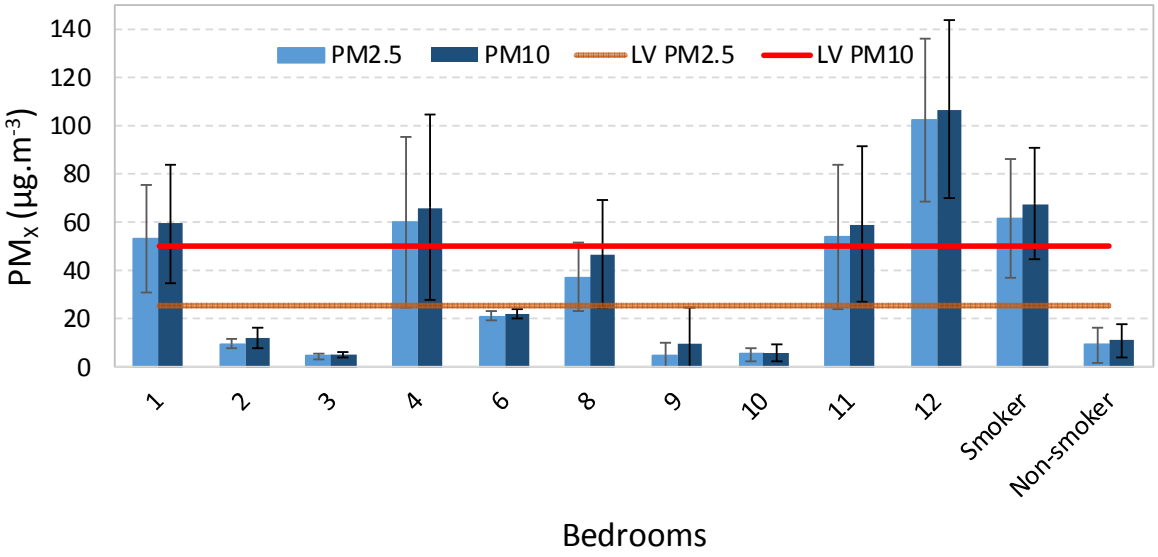


Figure 2. Carbon dioxide concentrations monitored during the sleep period in 12 different bedrooms, using two different monitoring devices: Pegasor and Graywolf. Red line represents the CO_2 limit value of 1250 ppm defined by the Portuguese legislation (*Ordinance no. 353-A/2013*, 2013).



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695 Figure 3. $PM_{2.5}$ and PM_{10} concentrations monitored during the sleep period in 10 different
696 bedrooms.
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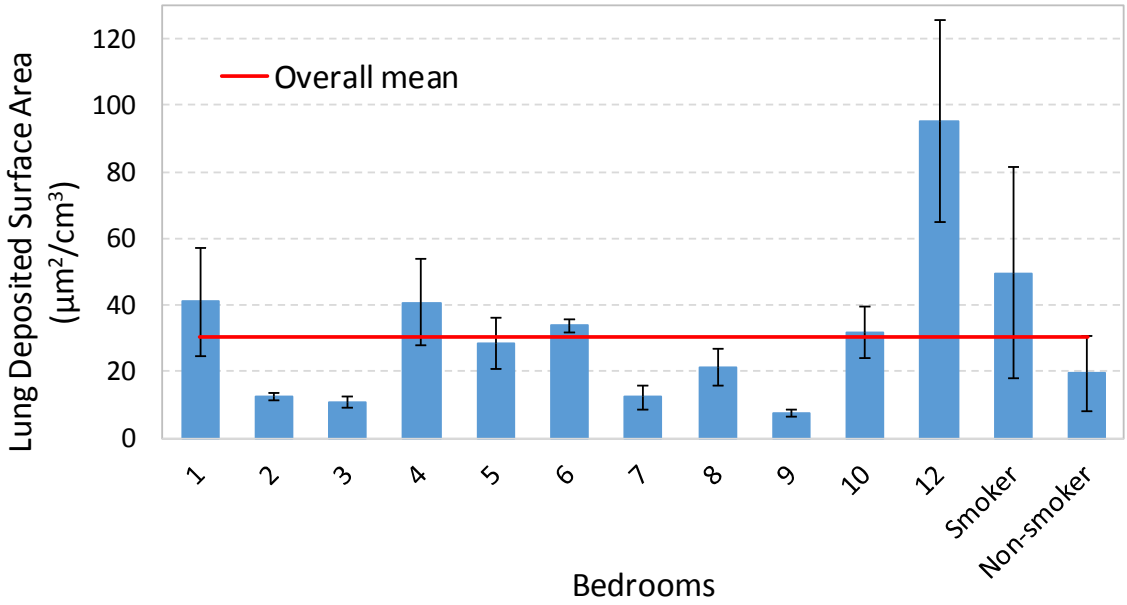

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700 Figure 4. Lung deposited surface area (LDSA) of particles monitored during the sleep period
701 in 11 different bedrooms. Red line is the mean value of the 11 bedrooms (30.5 $\mu m^2.cm^{-3}$).

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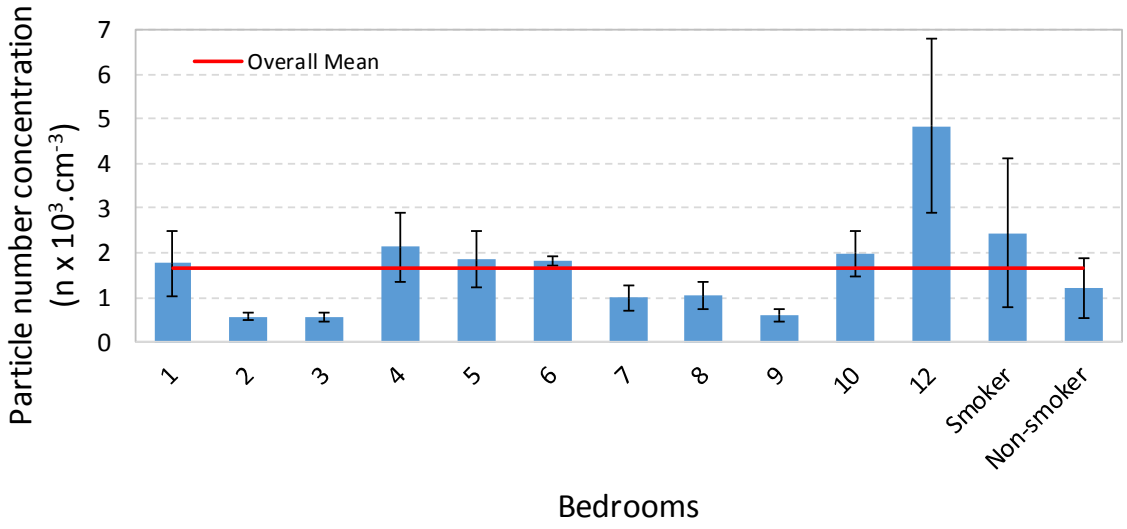

Figure 5. Particle number concentration during the sleep period in 11 different bedrooms. Red line is the mean value of the 11 bedrooms (1.7 x 10^3.cm^{-3}).

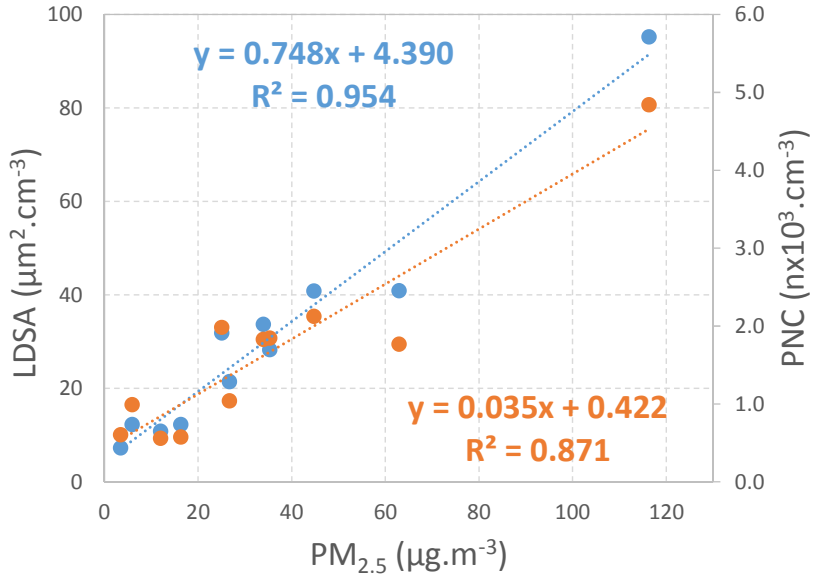

Figure 6. Correlation of $PM_{2.5}$ with and lung deposited surface area (blue) and particle number concentration (orange) during the sleep period.

716 **Tables of manuscript**
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718 Table 1. Levels of carbon monoxide monitored during the sleep period in 12 different
719 bedrooms, using Graywolf monitoring devices. LV stands for the CO limit value of 9 ppm
720 defined by the Portuguese legislation (*Ordinance no. 353-A/2013*, 2013).

	CO concentration (ppm)		
Individuals	**Mean ± SD**	**Min**	**Max**
1	4.21 ± 0.41	3.4	5.3
2	0.12 ± 0.11	0.0	0.3
3	0.34 ± 0.10	0.2	0.6
4	0.59 ± 0.16	0.3	0.8
5	0.47 ± 0.39	0.1	1.0
6	0.00 ± 0.00	0.0	0.0
7	0.00 ± 0.01	0.0	0.1
8	0.80 ± 0.09	0.6	0.9
9	0.50 ± 0.16	0.2	0.8
10	0.04 ± 0.05	0.0	0.2
11	1.71 ± 0.21	1.2	2.0
12	0.71 ± 0.07	0.3	0.9
LV	9		

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Impact of smoking on indoor air quality during sleep by Canha et al.

Highlights

- Multi-pollutant assessment of indoor air quality in 12 bedrooms during sleep.
- CO_2, PM_x, VOCs and CH_2O levels during sleep were found to be above guidelines.
- Comparative study of smokers and non-smokers' exposure in bedrooms while sleeping.
- Smokers are exposed to higher levels of CO, CH_2O, PMx, and LDSA than non-smokers.