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Article

Nanoparticle behaviour in an urban street canyon at different heights and implications on indoor respiratory doses

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Abstract: The amount of outdoor particles that indoor environments receive depends on the particle infiltration factors (F_{in}), peculiar of each environment, and on the outdoor aerosol concentrations and size distributions. The respiratory doses received, while residing indoor, will change accordingly. This study aims to ascertain to what extent such doses are affected by the vertical distance from the traffic sources. Particle number size distributions have been simultaneously measured at street level and at about 20 m height in a street canyon in downtown Rome. The same F_{in} have been adopted to estimate indoor aerosol concentrations, due to the infiltration of outdoor particles and then the relevant daily respiratory doses. Aerosol concentrations at ground floor were more than double than at 20 m height and richer in ultrafine particles. Thus, although aerosol infiltration efficiency was on average higher at 20 m height than at ground floor, particles more abundantly infiltrated at ground level. On a daily basis, this involved a 2.5-fold higher dose at ground level than at 20 m height. At both levels, such doses were greater than those estimated over the period of activity of some indoor aerosol sources, therefore they represent an important contribution to the total daily dose.

Keywords: Ultrafine particles; aerosol; urban street canyon; outdoor pollution; indoor air quality; respiratory doses; MPPD

1. Introduction

Outdoor and indoor air quality is a major determinant for human health. Indeed, World Health Organization estimated that air pollution kills seven million people worldwide every year and nine out ten individuals are exposed to high concentrations of airborne pollutants [1]. Particulate matter (PM) is one of the most relevant air pollutants, linked to the pathogenesis of several human diseases involving numerous systems and apparatuses. In particular, many studies highlighted that some specific PM fractions (particles diameter ≤ 10 μm and ≤ 2.5 μm, defined respectively as PM_{10} and $PM_{2.5}$) were associated to cardiovascular, respiratory and neurodegenerative diseases [2-4]. In addition, in 2015 PM in outdoor air was classified as group 1 carcinogen to humans by the International Agency for Research on Cancer (IARC) [5]. Over the last years the researches focused the attention on smaller particles, specifically on ultrafine particles (UFPs, that is particles with sizes ≤100 nm); indeed, particles > 2.5 μm are removed quickly (few hours) from the atmosphere due to dry and wet deposition, while particles < 1 μm persist for longer times and easily contaminate both outdoor and indoor air [6-8] and/or are transported over long ranges. Besides, UFPs have high number concentration, very high surface area and surface area reactivity, and small size compared to the dimensions of cellular structures. These characteristics allow them a great ability of both absorb

of organic molecules and penetrate into cellular targets [9,10]. Recent studies reported that UFPs are more toxic in comparison with larger particles and induce adverse effects on respiratory and cardiovascular systems via intracellular oxidative stress activation and inflammation response [11]. Besides, UFPs can pass directly in brain tissue through olfactory bulb and increase the risk of developing neurodegenerative diseases, in particular Alzheimer's disease [12]. Also, adverse outcomes on children's health associated with UFPs exposure have been recently reported, especially among children with respiratory diseases [13]. Such adverse effects on human health are of particular concern, considering that UFPs are released in high concentrations from many anthropogenic sources both outdoor and in many occupational and domestic scenarios [14]. Vehicular traffic is the main source of UFPs in urban area [15-18]. In indoor settings, UFPs are released by devices and appliances commonly used (conventional and electronic cigarettes, electric appliances, etc) or activities usually practiced (cooking) [19-24]. In the last years more attention was given to indoor air quality, because great part of general population spends most of the time (> 90%) in enclosed environments [25] and indoor air may be of worse quality than outdoor, since indoor air pollutants are the sum of those penetrated from outdoor and those directly produced indoor [26,27]. On this point, several studies [28-30] have addressed the issue of vertical profiles of particle concentrations in urban areas and have highlighted their relevance in terms of indoor air quality. However, to date, studies describing how the different vertical aerosol concentrations and size distributions affect the aerosol doses received by the respiratory system of individuals residing in the same building but at different heights are lacking. Thus, the aim of the present study was to ascertain to what extent such doses are affected by the vertical distance from the street level.

2. Experiments

2.1. Aerosol measurements

Atmospheric particle number-size distributions were simultaneously measured by means of two TSI Fast Mobility Particle Sizer (model 3091, FMPS, Shoreview, MN, USA) located at the ground level and at 20 m height in a building in downtown Rome during winter season. The site (41°53'46''N, 12°29'46'E) was characterized by high density of vehicular traffic, it was in a narrow double lane street (street width, W, of about 8 m), with high buildings on both sides (average height, H, of about 25 m). Such street can be considered a street canyon, as the aspect ratio H/W is about 3:1. FMPS counts and classifies particles, according to their electrical mobility, in 32 size channels, in the range 5.6-560 nm, with 1 s time resolution. It operates at a high flow rate (10 L min^{-1}) to minimize diffusion losses of UFPs and at ambient pressure, to prevent evaporation of volatile and semi-volatile particles [31]. The performance of the FMPS was investigated by Jeong et al. [32] by comparison with a Scanning Mobility Particle Sizer (SMPS). The authors evaluated that the SMPS number concentration, in the size range from 6 nm to 100 nm, is about 34% lower than the FMPS measurements, due to the diffusion losses of particles in the SMPS. The diffusion loss corrected SMPS number concentration is on average about 15% higher than the FMPS data. One-day outdoor aerosol measurements were selected to calculate the indoor aerosol concentration inside the building at the ground floor and at the 5th floor (20 m height) utilizing the size-resolved average infiltration factors (F_{in}) measured by Bennett and Koutrakis [33], using time-dependent aerosol concentrations (0.02-4 μm size range) and air-exchange rate (in the range of about 0.5 -1.5 h^{-1}) measurements carried out in nine homes with sampling duration from 6 to 12 consecutive days. The F_{in} relative to the FMPS size class were estimated by interpolation with a cubic spline [34] (Fig. S1 of supplemental material). For aerodynamic diameters (0.01-0.02 μm) outside the Bennett and Koutrakis [33] measurement range the F_{in} value of 0.02 μm particles was adopted.

Throughout the aerosol measurements atmospheric pressure, temperature, relative humidity, wind speed and wind direction were continuously measured with 5 min averaging time (Fig. S2 of supplemental material).

2.2. Aerosol dosimetry

To estimate aerosol doses due to the infiltration of outdoor aerosol in indoor environments, 22 hours daily indoor residence was supposed. Three time periods (t_r) were assumed according to the different work physical activities: 7 hours sleeping (from 11pm to 6 am), three hours sitting awake (6-7 am and 9-11 pm), 12 hours light work (8am-4 pm and 5pm-9pm). The particle regional deposition fractions per ($F^R(d_{ai})$) as a function of aerodynamic diameter (d_{ai}) have been estimated using the Multiple-Path Particle model Dosimetry (MPPD v3.01, ARA 2015, ARA, Arlington, VA, USA) [33]. The 60th percentile human stochastic lung was considered along with the following settings: (i) a uniformly expanding flow, (ii) an upright body orientation, and (iii) nasal breathing with a 0.5 inspiratory fraction and no pause fraction. Moreover, the following parameters were used for a Caucasian adult male under light work physical activity, based on the ICRP report [36]: (i) a functional residual capacity (FRC) of 3300 mL, (ii) an upper respiratory tract (URT) volume equal to 50 mL, (iii) a breathing frequency of 12 min^{-1} for sleeping and sitting awake activity and 20 min^{-1} for light work activity, and (iv) an air volume inhaled during a single breath (tidal volume, V_t) of 0.625 L, 0.750L and 1.25 L, respectively for sleeping, sitting awake and light work activities.

Since FMPS measures aerosol size number distribution as a function of the electrical mobility diameter (d), d values have been transformed to aerodynamic diameter (d_a) according to equation (1) [37].

$$d_a = d\sqrt{\chi \times \frac{\rho \times C_c(d_m)}{C_c(d_a)}}, \quad (1)$$

where C_c is the Cunningham slip factor for a given diameter, ϱ is the particle density (1.5 g cm^{-3} density was assumed) and χ is the particle dynamic shape factor. χ as a function of aerodynamic diameter was estimated by interpolating with a cubic spline the data reported by Hu et al. [38] in the range from 0.1 to 1.8 μm in Beijing. For da outside below this range the relevant lower bound χ value was adopted (1.13 for da <0.1 μm).

Aerosol number doses $D^R(d_{ai},t)$ deposited in the head (H), tracheobronchial (TB) and alveolar (Al) regions , as functions of time (t) and of the aerodynamic diameter (d_{ai}) were estimated according to eq. 2:

$$D^R(d_{ai},t) = F_{in} \times C(d_{ai},t) \times F^R(d_{ai}) \times V_t \quad \text{R= H, TB, Al,} \quad (2)$$

where $C(d_{ai},t)$ is the average aerosol concentration over a single inspiratory act as a function of time and of aerodynamic diameter.

Total regional aerosol doses have been estimated according to eq. 3, where the summation is carried out over the FMPS size classes:

$$D^R_{Tot}(t) = \sum_i D^R(d_{ai},t) \quad \text{R= H, TB, Al,} \quad (3)$$

Total aerosol doses deposited into the respiratory system has been calculated according to eq. 4:

$$D^{Tot}_{Tot}(t) = D^H_{Tot}(t) + D^{TB}_{Tot}(t) + D^{Al}_{Tot}(t), \quad (4)$$

Cumulative regional and total doses over a given residence time (t_r) have been calculated according equations 5 and 6:

$$D^R_{Tot}(t_r) = \sum_{t=0}^{t_r} D^R_{Tot}(t) \quad \text{R= H, TB, Al,} \quad (5)$$

$$D^{Tot}_{Tot}(t_r) = \sum_{t=0}^{t_r} D^{Tot}_{Tot}(t), \quad (6)$$

The cumulative regional ($D^R_{Tot}(t_{day})$) and total daily dose ($D^{Tot}_{Tot}(t_{day})$) has been calculated as the sum of the $D^R_{Tot}(t_r)$ and $D^{Tot}_{Tot}(t_r)$ doses relative to the tree time period (t_r) considered (7 h sleeping, 3 h sitting awake, 12 h light work, t_{day}=22 h).

The cumulative total dose size distribution over a given residence time ($D^{Tot}(d_{ai},t_r)$) has been calculated according to eq. 7:

$$D^{Tot}(d_{ai},t_r) = \sum_{t=0}^{t_r} D^H(d_{ai},t) + D^{TB}(d_{ai},t) + D^{Al}(d_{ai},t), \quad (7)$$

3. Results and discussion

3.1. Aerosol measurements

Outdoor aerosol total number concentrations (C_{Tot}) over the 24 hours considered were on average 2.3-fold higher at ground floor than at 20 m heights with spike concentration ratios exceeding 20, up to about 40. Both sets of data shared the same temporal modulation with two broad peaks centered at 8 am and 7 pm, determined by the daily traffic flow variation and by the planetary boundary layer (PBL) mixing height that was highest in the central part of the day during period of high solar radiation (Figure 1) [39-41].

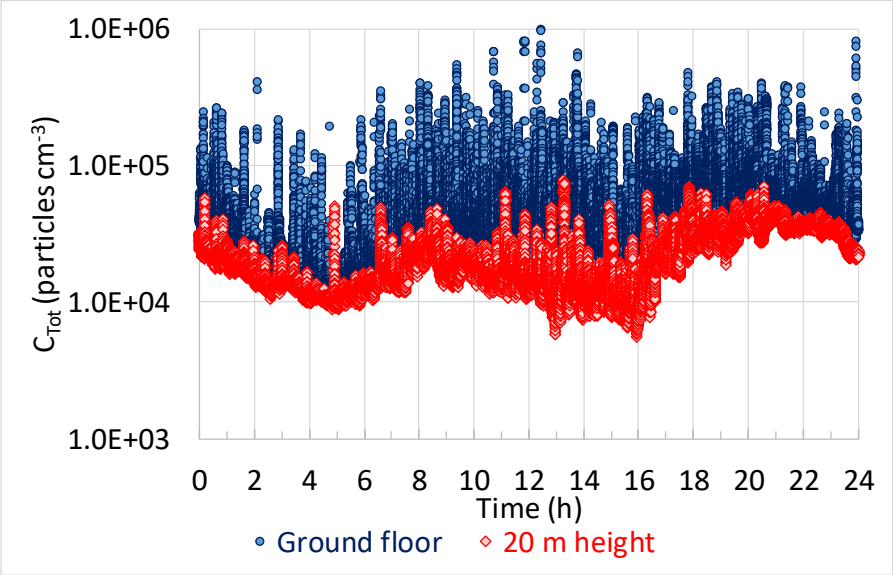

Figure 1. Total (5.6-560 nm electrical mobility diameter) aerosol concentrations measured with 1 s time resolution at ground floor and at 20 m height in a street canyon in downtown Rome.

Spike concentration were more intense and frequent at ground level than at 20 m heights due to the proximity of the vehicular exhausts. The contribution of UFPs at ground level was slightly higher than at 20 m height, on average respectively 88% and 84%, although at ground level it occasionally dropped below the relevant value at 20 m height. Such occurrence was probably due to the turbulence at ground level generated by the vehicular traffic, with consequent dust re-suspension and removal of smaller particles by impaction.

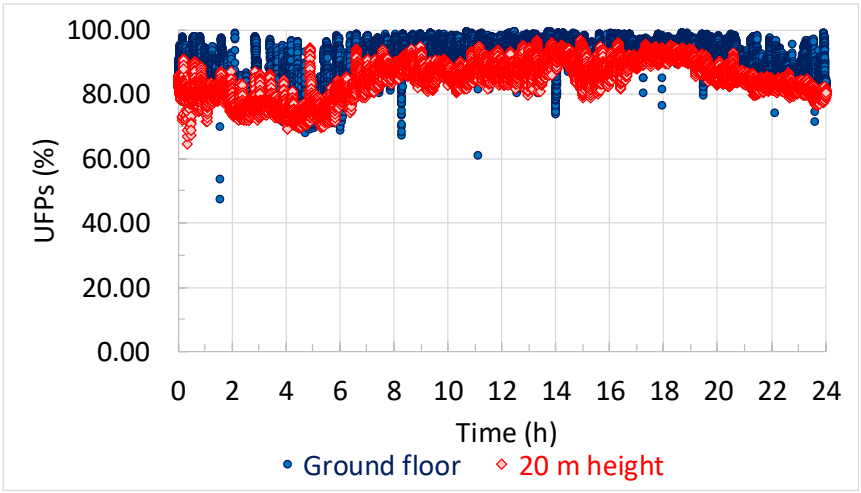

Figure 2. Temporal trend of UFP % contribution at ground floor and at 20 m height in a street canyon in downtown Rome.

Small particles (< 0.1 μm) deposition is efficient due to their Brownian diffusion, whereas interception, impaction and gravitational settling are important deposition mechanism for larger particles (> 2 μm) [42]. Minimum values of deposition velocities occur for particles in the range 0.1-2 μm, because neither Brownian diffusion nor impaction or interception are effective mechanisms [42]. Consequently, nucleation particles are less persistent than larger-sized particles. Therefore, they are predominant during traffic hours, when freshly formed from vehicular exhausts and decrease significantly at night, when traffic is less intense and particles in the accumulation mode predominate. Therefore, Geometric Mean Diameter (GMD), both at ground level and at 20 m height (Figure 3), follows a temporal trend a characterized by low values during the day and increases during nocturnal hours.

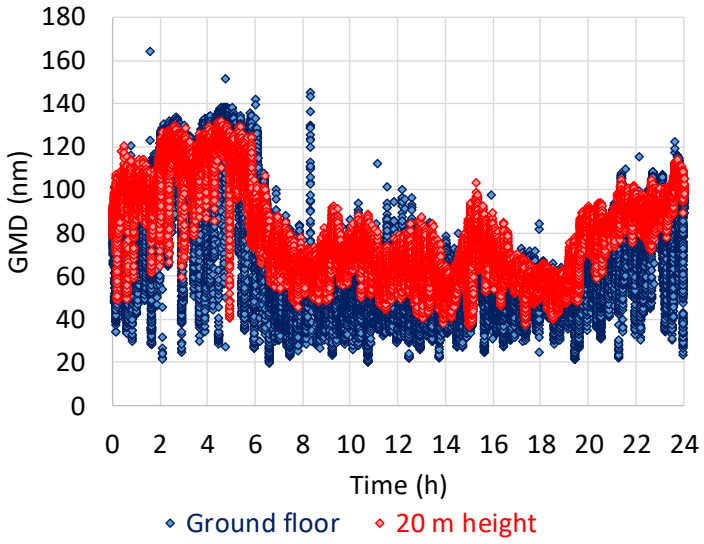

Figure 3. Temporal trend of the Geometric Mean aerodynamic Diameter (GMD) at ground level and at 20 m height in a street canyon in downtown Rome.

For these reasons, and due to the higher distance from the traffic exhaust, as a general trend GMD is higher at 20 m height than at ground level, as shown in Figure 4, where the ratio of GMD at the two levels is plotted a s function of time. This feature involves some consequences in terms of aerosol capability of infiltrating indoor environments.

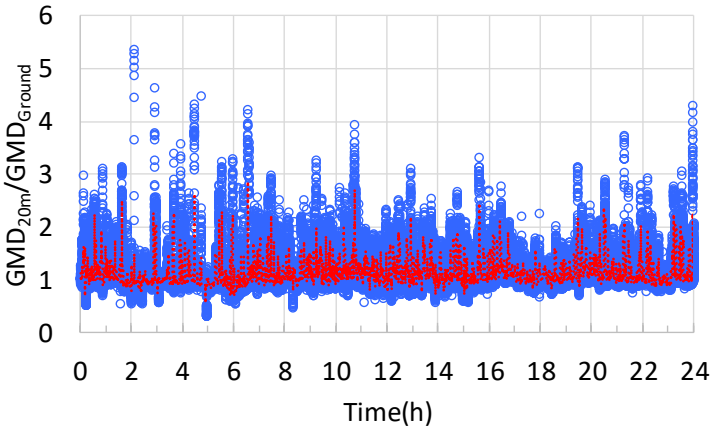

Figure 4. Ratio of geometric mean aerodynamic diameter (GMD) at 20 m height to GMD at ground level in a street canyon in downtown Rome.

To discuss this point, the infiltration factors pertaining to each GMD value have been calculated at both levels (Figure 5). Such dataset, being characterized by highly frequent spike values, has been despiked by calculating the relevant 1h mobile averages (F_{in}(GMD)). The smoothed dataset so obtained allows to assess a general daily trend of the average infiltration factor (Figure 5). At both levels the F_{in}(GMD) is higher during nocturnal hours than during daytime, in the same periods when a maximum is observed in the GMD temporal trends (Figure 3). Moreover, due to the higher GMD values, the aerosol infiltration efficiency at 20 m height is, on average, slightly higher than at ground level.

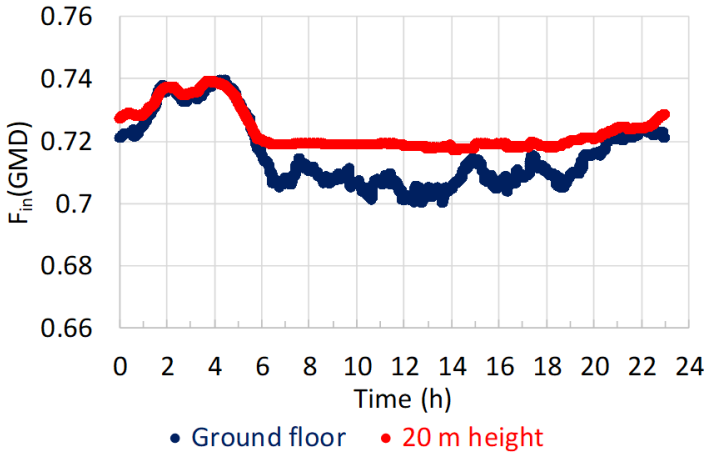

Figure 5. 1h-moving average of the infiltration factors (F_{in}) calculated at the GMD values at ground level and at 20 m height.

This occurs because the infiltration efficiency for small particles (< 0.1 μm) due to their efficient Brownian deposition is lower than for accumulation mode particles.

3.2. Aerosol dosimetry

Figure 6 describes the cumulative regional ($D_{Tot}^{R}(t_r)$) and total dose ($D_{Tot}^{Tot}(t_r)$) estimated for indoor environments at ground floor and at 20 m height, in the absence of indoor aerosol emission sources. They represent the contribution due to the indoor infiltration of outdoor aerosol. These doses are about 2.6, 1.7 and 1.9-fold higher at ground floor than at 20 m height, respectively for light work, sitting awake and sleeping physical activities. Such differences are determined by the higher outdoor aerosol concentrations at the street level (Figure 1) that abundantly outweigh the higher average infiltration efficiency of the aerosol at 20 m height (Figure 5). The highest $D_{Tot}^{Tot}(t_r)$ doses have been estimated during the period of light work activity (1.05×10^{11} particles at 20 m height and 2.80×10^{11} particles at ground floor) and the lowest for sitting awake activity (1.20×10^{10} particles at 20 m height and 2.02×10^{10} particles at ground floor) because of both the longer time period and of the higher tidal volume and breathing frequency for light work activities.

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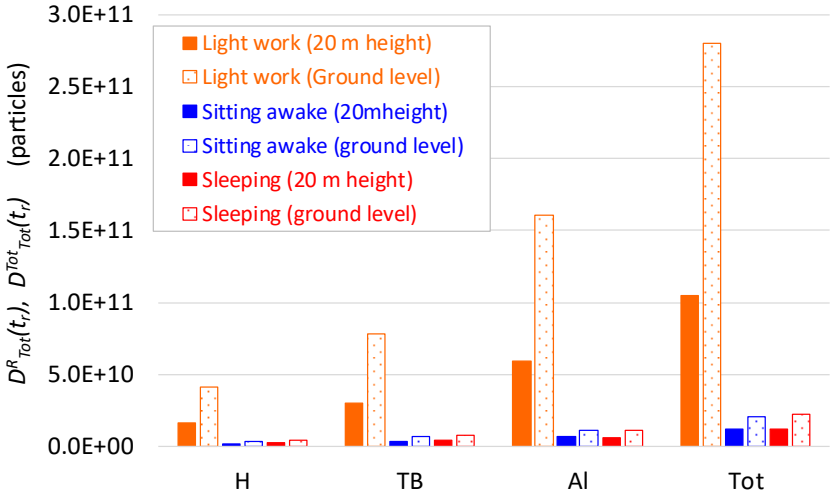
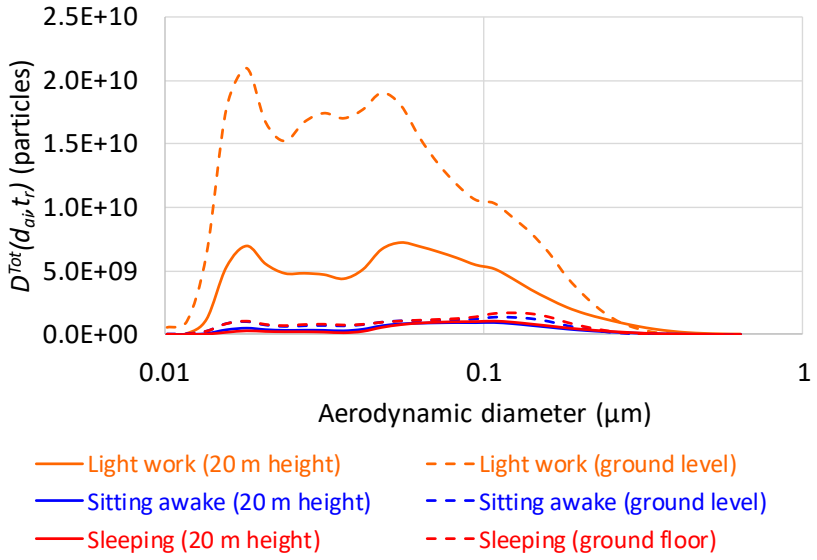


200

201 **Figure 6.** Cumulative regional ($D^R_{Tot}(t_r)$) and total ($D^{Tot}_{Tot}(t_r)$) aerosol doses over a residence time (t_r) of 7 h,
202 3 h and 12 h, respectively for sleeping, sitting awake and light work activities, at ground level and at 20 m
203 height.

204 The different work activities also affect the particle dose distribution within the respiratory
205 system. The fraction of particles deposited in the H and TB regions decreases passing from sleeping
206 (respectively 17% and 33%) to light work activity (respectively 15% and 28%); conversely, the Al
207 fraction increases (from 50% to 57%).

208 Figure 7 shows the size distributions of $\boldsymbol{D^R_{Tot}(t_r)}$ doses at ground level and at 20 m height. For
209 light work activity, at both levels, three modes of about the same importance were present at 0.018,
210 0.031 and 0.048 μm and a fourth one, less intense, at about 0.107 μm. The first two modes occurred
211 also for the sitting awake and sleeping activities, but the more intense modes were at higher particle
212 diameters (0.072 and 0.120 μm), reflecting the more abundant presence of accumulation mode
213 particles during nocturnal hours.



214

215 **Figure 7.** Cumulative total dose size distribution ($D^{Tot}(d_{ai}, t_r)$) at ground level and at 20 m height, over a
216 residence time (t_r) of 7 h, 3 h and 12 h, respectively for sleeping, sitting awake and light work activities.

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Figure 8 describes the cumulative regional ($D^{R}_{Tot}(t_{day})$) and total ($D^{Tot}_{Tot}(t_{day})$) daily doses estimated for one day residence in an indoor environment at ground floor and at 20 m height. 3.23×10^{11} and 1.29×10^{11} particles are respectively deposited into the respiratory system at ground floor and at 20 m height. About 16 %, 29 % and 55 % of such doses are respectively deposited in the H, TB and Al regions.

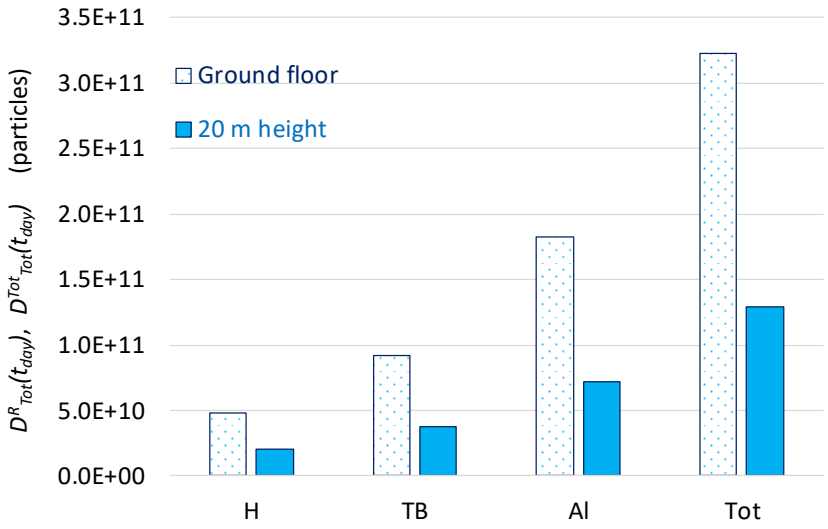

Figure 8. Cumulative regional ($D^{R}_{Tot}(t_{day})$) and total daily dose ($D^{Tot}_{Tot}(t_{day})$) at ground level and at 20 m height.

$\boldsymbol{D^{Tot}_{Tot}(t_{day})}$ at both levels are compared in Figure 9a with the total particle doses deposited into the respiratory system after 1 h exposure time (t_{exp}), at the same site and in the same day, at outdoor aerosol during the traffic peak hour and after 1h exposure to atmospheric aerosol in Mount Terminillo, a central Italy 2217 m high mount region ($D_c{}^{Tot}(t_{exp})$) [43]. In the same figure the comparison is carried out with the main combustion and non-combustion sources encountered in indoor environments [21-24], in this case t_{exp} represents the period of source operation (5, 1.5, 6, 5, 6, 6, 4, 4 and 5 min) respectively for hairdryer, electric drill, vacuum cleaner, hot flat iron, mosquito coil, incense cone, tobacco cigarette, e-cigarette and meat grilling.

Due to the longer exposure time (22 h), at both levels $D^{Tot}_{Tot}(t_{day})$ are higher than the doses relative to all the other operations, with the exception of the meat grilling without exhaust ventilation. In that case, a single 5 min exposure is associated to a dose that is 1.9 and 4.7 fold higher than the cumulative total daily doses at ground floor and 20 m height (22 h exposure). To account for the different time scales, the doses of Figure 9a have been referred to unit exposure time ($\dot{D}^{Tot}_{Tot}(t_{day})$, $\dot{D}^{Tot}_{c}(t_{exp})$) and plotted in Figure 9b. On this basis, with the exception of the relatively low aerosol emitting first generation e-cigarette [21,23], all the doses due to indoor aerosol sources are higher than the doses due to the infiltration of outdoor particles.

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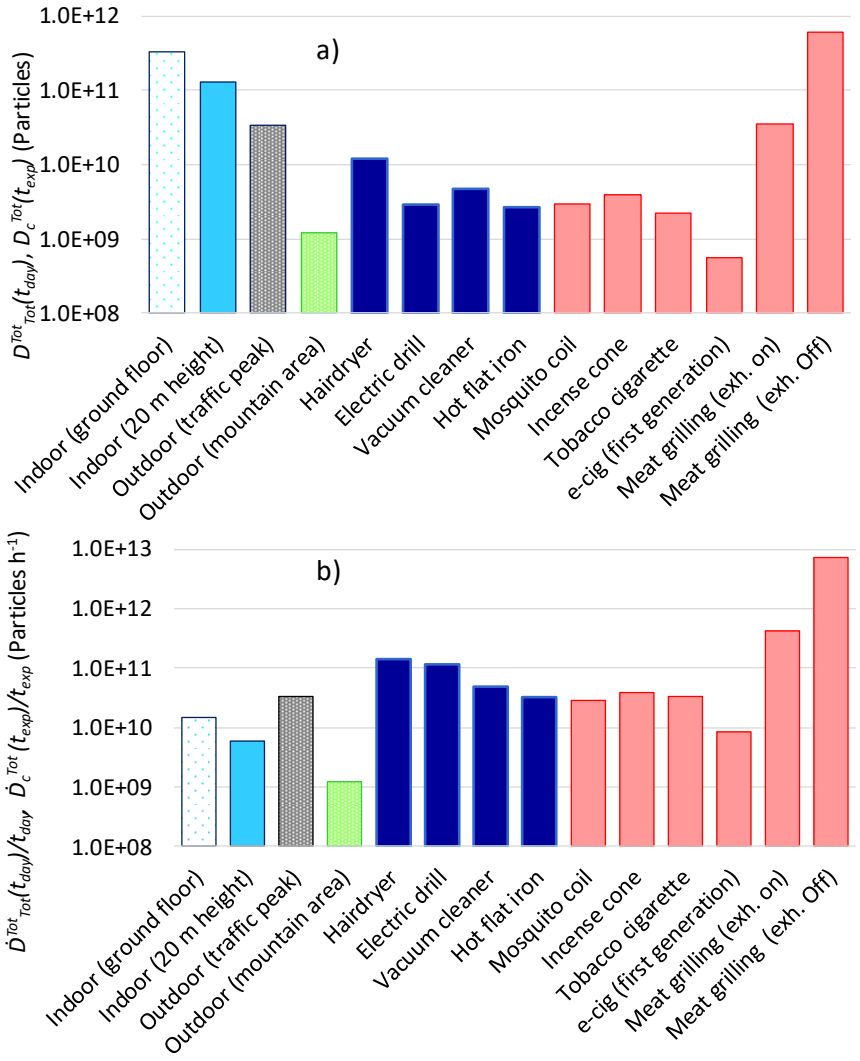

Figure 9. (**a**) $D^{Tot}_{Tot}(t_{day})$ at ground level and at 20 m height compared with the total particle doses deposited into the respiratory system ($D_c^{Tot}(t_{exp})$) after exposure to different indoor and outdoor aerosol sources for given exposute times (t_{exp}); (**b**) The same comparison is made for the relevant doses per unit exposure time ($\dot{D}^{Tot}_{Tot}(t_{day})$, $\dot{D}^{Tot}_{c}(t_{exp})$).

Moreover, the dose after 1h outdoor exposure in the Terminillo mountain area emphasizes the influence of the outdoor pollution on indoor air quality. Such dose is 12 and 5-fold lower than the 1 h doses relative respectively to the ground floor and to the 20 m height indoor environments in an urban street canyon.

4. Conclusions

Particle number size distributions have been simultaneously measured at street level and at about 20 m height, with 1 s time resolution, at a street canyon in downtown Rome. At both heights, the total particle number concentrations shared a temporal trend that on hourly time scale was determined by the daily traffic flow variations and by the PBL modulation. On a few second time scale, the two trends where characterized by spike concentrations, due to freshly emitted vehicular exhausts. Due to the closer proximity to traffic, such spikes were more frequent and more intense at ground level then at 20 m heights. This circumstance made the road level aerosol concentrations on average richer in UFPs, particularly in nucleation mode particles. Hence, the aerosol infiltration efficiency was on average slightly higher at 20 m heights than at ground level. On the other hand, for

the same reason, the road level aerosol concentration was on average more than double than at 20 m heights. As a result of that, the indoor aerosol concentration due to the penetration of outdoor particles was higher at ground floor than at 20 m height. With an estimated daily indoor permanence of 22 hours, the daily dose deposited into the respiratory system was 3.23×10^{11} and 1.29×10^{11} particles respectively at ground level and at 20 m height. Such doses are greater than those estimated over the period of activity of some common combustion and non-combustion sources in indoor environment, therefore they represent an important contribution to the total aerosol daily dose.

Supplementary Materials: The following are available online, Figure S1: Infiltration factors (F_{in}) estimated by interpolation of the average F_{in} measured by Bennett and Koutrakis [32], Figure S2: Atmospheric pressure, Temperature, Relative humidity, wind speed and wind direction throughout the aerosol measurements (averaging time 5 min).

Author Contributions: conceptualization, M.M. and P.A.; methodology, M.M. and P.A.; software, M.M.; validation, M.M., C.P., M.V. and P.A.; formal analysis, M.M.; writing—original draft preparation, M.M. and C.P.; writing—review and editing, M.V. and P.A.

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Conflicts of Interest: The authors declare no conflict of interest.

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