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Tracheobronchial and alveolar dose of submicrometer particles for different population age groups in Italy

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Abstract – Exposure to ultrafine particles (diameter less than 100 nm) is an important topic in epidemiological and toxicological studies. This study used the average particle number size distribution data obtained from our measurement survey in major micro-environments, together with the people activity pattern data obtained from the Italian Human Activity Pattern Survey to estimate the tracheobronchial and alveolar dose of submicrometer particles for different population age groups in Italy. We developed a numerical methodology based on Monte Carlo method, in order to estimate the best combination from a probabilistic point of view. More than 10^6 different cases were analyzed according to a purpose built sub-routine and our results showed that the daily alveolar particle number and surface area deposited for all of the age groups considered was equal to 1.5×10^{11} particles and 2.5×10^{15} μm^2, respectively, varying slightly for males and females living in Northern or Southern Italy. In terms of tracheobronchial deposition, the corresponding values for daily particle number and surface area for all age groups was equal to 6.5×10^{10} particles and 9.9×10^{14} μm^2, respectively. Overall, the highest contributions were found to come from indoor cooking (female), working time (male) and transportation (i.e. traffic derived particles) (children).

Keywords: tracheobronchial and alveolar particle deposition, ultrafine particles, time activity pattern, Monte Carlo method.

1. Introduction

People are exposed to particulate pollution from a range of indoor and outdoor sources. Epidemiological data from air pollution studies associate particulate matter with negative effects on human health, including breathing difficulties, cardiovascular disease and a general increase in mortality and morbidity (Schwartz, 1991; Vedal, 1997; Cheng et al., 1999; Cheng, 2003; Kreyling et al., 2006). Currently, there is considerable research activity to investigate the possible associations between short term fluctuations in airborne particulate matter and morbidity or mortality (Brugge et al., 2007), as well as mortality from long term chronic exposure (Miller et al., 2007). A number of epidemiological studies associated the above effects with particle mass concentration, including $PM_{2.5}$ (Pope, 2000) and PM_{10} (Loomis, 2000), as well as ultrafine particle (UFP, particle diameter < 100 nm) number concentration (Hauser et al., 2001), surface area concentration (Giechaskiel et al., 2009) and overall exposure level (Siegmann and Siegmann, 1998). Most of the epidemiological investigations rely on measurements of particle concentration at some central location in the urban area of interest and then use these data to estimate individual exposure. This methodology can be easily applied for larger fractions of particulate matter (PM) but is generally not applicable to UFPs, which present a higher concentration decay with respect to distance to the source and, hence displaying a concentration gradient with less spatial homogeneity (Buonanno et al., 2011a). In general, exposure to particulate pollution can come from a range of sources, including inside buildings, in vehicles, and in the general urban environment. Human exposure was defined by Ott (1982) as 'the event when a person comes into contact with a pollutant of a certain concentration during a certain period of time'. Consequently, exposure implies that both the pollutant and the person must be present (Ashmore, 2009).

Despite a number of pollution abatement policies which have been recently proposed in Western countries (Sundqvist, 2002; Bramoulle, 2005; Dellink, 2006), basic information concerning the air quality monitoring data and time activity patterns are not adequate to permit policy-makers to develop an accurate exposure profile of, or to identify the major contributors to human exposure. This information is essential for policy-makers to identify the potential at risk groups and to develop appropriate risk reduction measures. In fact, though there is an increasing number of studies reporting pollutant concentrations in different micro-environments (Chau et al., 2002; Kaur et al., 2007; Hänninen et al., 2009), the integrated exposure or integrated dose of the people was not successfully estimated.

The main objectives of this study were: i) to provide quantitative information on the levels of UFPs (including particle number and surface area concentrations, as well as size distribution), in over 20 smoke-free micro-environments in Cassino, Central-Southern Italy (average concentrations were obtained from our measurement survey data); ii) to estimate the daily alveolar- and trachebronchial-deposited particle number and surface area concentrations for people of different age groups on the basis of time activity patterns reported in Bastone et al. (2003; 2006), Soggiu et al. (2005; 2010) and

ISFORT (2010), which represent the Italian Human Activity Pattern Survey; and iii) to determine the exposure profiles of people with respect to different commuting and behavior patterns, in particular by comparing the lifestyle of people living in the Northern and Central-Southern Italy. Hence, this work represents a population-based study: the results would be very different between individuals, especially when on and off road environments are considered.

2. Methodology

In order to estimate the daily tracheobronchial and alveolar-deposited fraction of airborne particles, the following aspects were considered: i) matching the locations in which each exposed person spends time with the average time spent in each location, ii) the particle number size distributions that are thought to exist in each location, iii) the selection of inhalation rates to be used as a function of the age of the exposed population and the corresponding specific activity levels, and iv) the fraction of inhaled particles deposited in the respiratory.

In the indirect exposure assessment approach (Ott, 1982; USEPA, 2004; Klepeis 2006), the daily number (N) or surface area alveolar-deposited (S) dose for each age group ($\delta_{A,x}$), where x can be N or S for particle number and surface area concentration, can be evaluated through the following equation:

$$\delta_{A,x} = \sum_{j=1}^{n} \left\{ IR_{activity} \left[\int_0^{\infty} \varphi_A \left(IR_{activity}, D \right) \cdot \frac{dx(D)}{dD} \cdot dD \right] T_j \right\} \quad (1)$$

where $IR_{activity}$ is the inhalation rate of each age group depending on the human activity, $\varphi_A(IR_{activity},D)$ is the fractional alveolar deposition depending on inhalation rate and particle diameter, $\frac{dx(D)}{dD}$ is the particle number size distribution for each microenvironment and T_j is the time spent for an activity in a defined location. The same methodology can be applied to evaluate the daily number or surface area tracheobronchial-deposited dose for each age group ($\delta_{TB,x}$) by considering the corresponding fractional tracheobronchial deposition $\varphi_{TB}(IR_{activity},D)$.

In this formulation, locations are termed micro-environments and they are assumed to have a range of particle number size distributions. Furthermore, we have considered that different activities can be performed in the same micro-environment. Therefore, equation (1) has to be applied to the different daily activities $j = 1 \ldots n$ carried out by each age group (male/female younger than 1 year, 1-5 years, 6-10 years, 11-18 years, 19-40 years, 41-65 years and older than 65 years). A stochastic algorithm based on the Monte Carlo method was proposed to solve equation (1) and the following paragraph provides a detailed description of the model and of the input parameters used.

2.1 *Inhalation rates*

Inhalation rates as a function of the different activities and age groups are reported in Table 1. These values, adapted from EPA (2009) and CARB (1993), vary between 0.3 $m^3 h^{-1}$ for children and young people who are at rest or sleeping, and 2 $m^3 h^{-1}$ for adults during sporting activities.

2.2 Fractional deposition in regions of the respiratory tract

Particle properties, dimensions of airways and breathing patterns are among the most important parameters required to evaluate the fraction of inhaled particles deposited in the respiratory system. The International Commission on Radiological Protection (ICRP) dosimetry model (ICRP, 1994) was applied by the authors to estimate alveolar and tracheobronchial deposition. To determine the amount of atmospheric particle number or surface area deposited per cubic centimeter of air inhaled, the number or surface area of the particles were multiplied by the deposited fraction (DF) (reported in ICRP, 1994) for the size of the midpoint of each $\Delta logD$ interval. In order to obtain the functional relationship between particle diameter and alveolar and tracheobronchial deposition, the author interpolated the data from ICRP (1994) as piecewise cubic Hermite interpolating polynomials (Kahaner et al., 1988) of $\varphi_A(IR_{activity}, D)$ versus $\log(D)$, for each activity selected. When evaluating the fractional deposition in different regions of the respiratory tract, only normal nose breathers were considered.

2.3 Particle number size distributions

According to equation (1), one fundamental piece of information necessary to estimate the dose is the particle number size distribution to which each group is exposed in the different micro-environments .

The maximum and minimum values of the particle number size distributions for the different micro-environments were obtained from our previous measurement survey data (Buonanno et al., 2009a; 2009b; 2010; 2011a; 2011b; 2011c), as reported in Table 2. Cassino is located in Central-Southern Italy (41°30’00” N -13°50’00” E), and can be considered a typical busy Italian middle town (resident population: 33 000 inhabitants; daily commuter workers and students: 20 000; surface area: 83 km^2). Particle size distributions used to evaluate the daily particle dose were fitted through lognormal distribution. In Table 2, log-normal distribution parameters (mode, standard deviation, total particle number) for each micro-environment are reported, as well as the maximum (N_{max}) and a minimum (N_{min}) total particle number concentration value. The particle number concentration values in Table 2 show higher concentrations during cooking activities (not only due to particle emission from cooking but also because of the natural ventilation conditions normally used in an Italian kitchen during cooking), in some working conditions and during transportation. As expected, the concentration level varied widely among different types of micro-environments and such concentrations are comparable to those reported in other studies (see Table 2). It is noteworthy to point out that we also present a range of particle number size distributions for each micro-environment, in order to help the individuals set-up a time budget plan for optimizing their lifestyles, and consequently reducing their total particle dose.

2.3 Daily time activity patterns

The starting point for the determination of daily time activity patterns is based on Bastone et al. (2003; 2006) and Soggiu et al. (2005;2010). The data reported in these studies can be considered the Italian Human Activity Pattern Survey for the Northern and Southern parts of Italy, respectively. These were carried out using specific questionnaires, which provided a valid method for acquiring information on individual variability, in order to perform the exposure evaluation. In fact, even though Cassino is located in the Central-Southern part of Italy, we also applied our analysis to Northern Italy, in order to evaluate the influence of lifestyle on particle dose. In Italy, a more Western style of living is typical in the North, rather than a more traditional Mediterranean style of living in the South. In terms of daily time activity patterns for transportation, data were obtained from ISFORT (2010). In particular, transportation data are reported up to the first semester of 2010 for different age groups, geographical areas, gender, working occupation and city dimensions.

Firstly, we generated a combination matrix ($\boldsymbol{C}_{i,j}$) by alternatively considering the presence (value 1) or absence (value 0) of some activities, such as: indoor or outdoor entertainment, indoor or outdoor sport, and having a sedentary or non- sedentary job. Therefore, 24 combinations were obtained (n_c) for a total number of 21 activities (n_a).

From an arithmetic point of view, a total time greater than 1440 min (24 h) was found by considering the total daily time spent by the age groups for each combination. Consequently, in order to normalize the data to 1440 min, the following procedure was applied:

$$T^*_{i,j} = \frac{T_i \cdot C_{i,j}}{\sum_{i=1}^{n_a} T_i \cdot C_{i,j}} \cdot 1440 \qquad \text{for } i = 1 \ldots n_a \text{ and } j = 1 \ldots n_c \tag{2}$$

and:

$$\begin{gathered} \overline{T}_i = \frac{\sum_{j=1}^{n_c} T^*_{i,j}}{n_c} \\ \sigma\left(\overline{T}_i\right) = \sqrt{\frac{1}{n_c - 1} \cdot \sum_{j=1}^{n_c} \left(\overline{T}_i - T^*_{i,j}\right)^2} \end{gathered} \qquad \textit{for } i = 1 \ldots n_a \textit{ and } j = 1 \ldots n_c \tag{3}$$

where $\boldsymbol{C}\left(n_a \times n_c\right)$ is the activity combination matrix, $\boldsymbol{T}\left(n_a \times 1\right)$ is the vector of the time activity patterns corresponding to the different activities (see Table 3), $T^*\left(n_a \times n_c\right)$ is the matrix of the normalized times where $n_a = 21$ is the number of activities and $n_c = 24$ is the number of combinations, $\overline{T}_i\left(n_a \times 1\right)$ and $\boldsymbol{\sigma}\left(\overline{T}_i\right)\left(n_a \times 1\right)$ are the normalized time vector and its corresponding standard deviation. The procedure described by equations (2-3) was applied for each gender and each age group considered in the present investigation.

2.4 Application of the Monte Carlo method in the evaluation of the tracheobronchial and alveolar dose

In order to apply equation (1), the authors performed a Monte Carlo simulation (Hammersley and Handscomb, 1964) to generate both particle size number distributions and daily time activity patterns. In particular:

- for each activity, a sample of 50 particle size number distributions was generated according to a random uniform distribution between the minimum and maximum values reported in Table 2 for the corresponding microenvironment;
- for each age group, a sample of 10^6 daily time activity pattern combinations was generated following a Gaussian distribution based on the parameters reported in equation (3). Among these combinations, a sub-sample was chosen with the constraint that the total daily time was equal to 24±0.5 hours. Then, each sample activity time combination belonging to the sub-sample was normalized to 1440 min;
- a random sample of 10^6 combinations of the generated particle size number distributions and daily time activity patterns was then obtained; and
- the daily alveolar $\delta_{A,x}$, or tracheobronchial, $\delta_{TB,x}$ number or surface area deposited dose for each age group was finally evaluated using equation (1) on the basis of a total number of combinations of 10^6.

3. Results and Analysis

3.1 Normalized daily time activity patterns

Figure 1 shows the normalized daily time activity patterns for Northern and Southern Italy, and the corresponding standard deviations for each age group and gender, by considering the 21 activities. The surveyed age groups mainly engage in different primary activities. For example, for the 1-5 years age group, sleeping and resting, (1) sedentary activities (2), eating (6) and entertainment (15-16) were the primary activities. For 6-10 and 11-18 years, the contribution of study time to their primary increases. In the case of females, a higher contribution of house cleaning (4) and cooking (5) was observed for Southern Italy than for Northern Italy (confirming the influence of a more traditional lifestyle). For adults (18–41 and 41-65 years old), working time makes an important contribution to the primary activity pattern. Our survey results showed that the total time spent undertaking indoor activities, by individuals from all age groups varied slightly, ranging from 1180 min (82%) for males in Southern Italy to 1224 (85%) for females in Northern Italy.

The average value for total time spent at home was about 1000 min (69%) and in general, Women spent more time indoors (generally at home) than men. This difference is mainly due to the different lifestyles and climate between Northern and Southern Italy. As a function of age group, the minimum values for time spent at home were found for the 19-40 years are group. The total time spent using transportation was reasonably constant and equal to about 60-73 min (4-5%). The obtained values were in good agreement with the data presented by previous research. For example, Chau et al. (2002) reported that individuals from Hong Kong (China) spent an average of 86% of their time indoors, 3–7% in enclosed transit and 3–7% outdoor, for all three of the age groups analyzed, (6–18 years, 18–60 years and >60 years).

Brasche and Bischof (2008) carried out an analysis of the time spent indoors at home, based on a study of about 12,000 persons living in 5,530 randomly selected apartments and houses in Germany. The mean time spent at home was equal to 942 min (65%) with higher values for women (996 min, 69%) than men (882 min, 61%). This difference is very pronounced in the age range 25–54 years. Generally, older persons and pre-school children are characterized by the longest periods at home.

The overall mean time spent at home is also in good accordance with results from American (940 min, 65%) and Canadian (950 min, 66%) human activity surveys carried out in the nineties, as reported by Leech et al. (2002). The comparison between the presented data and the current literature highlights an adopted Italian lifestyle that is typical of Western countries, with slight differences found between Northern and Southern Italy.

3.2 Alveolar and tracheobronchial particle number and surface area deposited dose

In Figure 2, the average daily alveolar and tracheobronchial particle number and surface area deposited are reported with the corresponding standard deviations, by considering age groups, gender and location (Northern and Southern Italy).

The daily alveolar particle number (and surface area) deposited for males from the all age groups ranged from $1.39 \times 10^{11} \pm 2.6 \times 10^{10}$ particles ($2.28 \times 10^{15} \pm 4.9 \times 10^{14}$ μm^2) in Northern Italy to $1.51 \times 10^{11} \pm 7.2 \times 10^{9}$ particles ($2.51 \times 10^{15} \pm 4.7 \times 10^{14}$ μm^2) in Southern Italy. The corresponding values for females ranged from $1.44 \times 10^{11} \pm 2.9 \times 10^{10}$ particles ($2.64 \times 10^{15} \pm 6.5 \times 10^{14}$ μm^2) in Northern Italy to $1.51 \times 10^{11} \pm 4.8 \times 10^{9}$ particles ($2.56 \times 10^{15} \pm 3.1 \times 10^{14}$ μm^2) in Southern Italy.

In terms of tracheobronchial deposition, the daily particle number (surface area) deposited for males from all age groups, ranged from $5.88 \times 10^{10} \pm 9.6 \times 10^{9}$ particles ($8.04 \times 10^{14} \pm 1.7 \times 10^{14}$ μm^2) in Northern Italy to $6.59 \times 10^{10} \pm 2.9 \times 10^{9}$ particles ($9.88 \times 10^{14} \pm 6.1 \times 10^{13}$ μm^2) in Southern Italy. The corresponding values for a female varies from $6.72 \times 10^{10} \pm 1.3 \times 10^{10}$ particles ($1.06 \times 10^{15} \pm 2.7 \times 10^{14}$ μm^2) in Northern Italy to $6.67 \times 10^{10} \pm 2.2 \times 10^{9}$ particles ($1.11 \times 10^{15} \pm 5.2 \times 10^{13}$ μm^2) in Southern Italy.

These values highlight a slightly higher dose for people living in Southern Italy (compared to Northern Italy), and for females (compared to males). However, these differences are lower than the corresponding standard deviations and, therefore, have to be considered as not significant. Another noteworthy point is the general increase in dose from the younger (1-18 years old) to older (19-65 years old) age groups, with a decrease in dose for people > 65 years old). The main reason for this is less to do with differences between the inhalation rates for the different age groups, and more to do with different level of particle exposure in the different micro-environments where people spend their time. In fact, when compared to adults (41-65 years old), the percentage difference in alveolar (tracheobronchial) number deposition for children (6-10 years old) and the elderly (> 65 years old) was equal to 74% (57%) and 17% (14%), respectively. By normalizing the data with respect to mean inhalation rate as a function of activities for all age groups (0.57 m^3 h^{-1}), these differences only decrease to 69% (50%) and 14% (11%), which confirms that the main factor contributing to increased exposures was related to the different micro-environments rather than physiology.

3.3 Contributions to the alveolar and tracheobronchial particle number and surface area deposited dose

In Figure 3, the time contribution for each activity, as well as the average daily alveolar and tracheobronchial particle number and surface area deposited for each activity are reported. In order to analyze these contributions in more depth, we introduced a new parameter (Wang et al., 2011) termed "dose intensity", defined as the ratio of the daily dose contribution compared to the daily time contribution.

The average contribution to alveolar deposited number and surface area for indoor micro-environments was equal to 69% ± 9.3% and 86% ± 5.5%, respectively. In terms of dose intensity, this ranged between 0.82 ± 0.10 and 1.26 ± 0.11. Overall, the maximum dose intensity values found for indoor environments were for children (1-5 years old), while the minimum values found were for teenagers (11-18 years old). In terms of daily total particle number deposition, these values show that the high contribution from indoor activities mainly depends on the time contribution these activites make to the overall daily activity profile, as opposed to a high dose intensity for indoor micro-environments. The corresponding values for tracheobronchial deposition in indoor micro-environments are comparable, in terms of both particle number and surface area (72% ± 6.2% and 84% ± 4.6%), as well as dose intensity (0.84 ± 0.07 and 1.17 ± 0.06).

Another important contributor to daily total particle deposition is transportation micro-environments. Even though the average contribution to alveolar deposited number and surface area for these micro-environments is low (4.7% ± 0.6% and 9.8% ± 5.2%), the dose intensities are significantly higher than those observed for indoor environments, equaling 3.8 ± 1.6 and 2.0 ± 0.9, respectively. This trend was also observed for tracheobronchial deposition, with the only difference being that the higher values were found for surface area deposition as opposed to number deposition dose intensity, being 3.1 ± 1.4 and 0.5 ± 0.2, respectively. In terms of particle number deposition, the highest dose intensity values were found for children (6-10 years old), indicating that it is very important to consider the exposure of children to ultrafine particles emitted by traffic.

Based on the obtained data, we also investigated the contributions of single human activities, which varied considerably for each age group. For 1-5 year olds, the following activities represent a contribution to alveolar particle number deposition greater than 10%: sleeping and resting (18%), cooking time (17%), eating time (21%) and transportation (12%). Similar values are found for tracheobronchial number deposition. In terms of alveolar and tracheobronchial surface area deposition, the predominant contributions came from cooking and eating time, which ranged from 61-67% when taken together. These values highlight the fact that, in addition to transportation, the particles generated from cooking activities also represent a significant source of exposure for children (1-5 years old), with a dose intensity higher than 10.

The major contributors to alveolar and tracheobronchial particle number deposition in children (6-10 years old) were sleeping and resting (13% and 15%, respectively), eating (22% and 18%, respectively) and transportation (19% and 20%, respectively). With regard to surface area, the predominant contribution was from eating time (>54%).

For the teenager group (11-18 years old), the main contributions to alveolar particle number deposition arose from cooking time (15%, with higher values for females), eating time (19%), non-sedentary work (outdoor) (15%) and transportation (11%). These values were quite different from those observed for tracheobronchial particle number deposition, which were: sleeping and resting (13%), eating time (17%), and transportation (20%). In terms of surface area deposition, this finding also confirms the importance of eating time, where high particle concentrations are likely to remain in the air following cooking activities

For the 19-40 and 41-65 year age groups the main contributors to alveolar particle number deposition were cooking time for females and eating time for males (including time spent in restaurants and pizzerias), with a mean value of 34%, as well as working activities for both groups (39%). For the elderly group (>65 years old), the main contributor was cooking and eating time, representing 45% and 31% for alveolar and tracheobronchial particle number deposition, 71% and 73% for alveolar and tracheobronchial surface area deposition, respectively.

Conclusions

In this work, activity pattern data were combined with micro-environmental data (human activities and particle number size distributions) using an indirect approach, in order to evaluate the doses of alveolar and tracheobronchial deposited particle number and surface area experienced by different age groups in Cassino (South Italy). The analysis of activity pattern data was also extended to Northern Italy, in order to compare exposure levels for the two different lifestyles. Our survey results showed that the total time spent undertaking indoor activities varied slightly for all age groups, ranging from 82% for a males in Southern Italy to 85% for a females in Northern Italy. This difference was mainly due to the different lifestyles and climates found in Northern and Southern Italy. Overall, women were found to spend more time indoors (generally at home) than men, however the total time spent on transportation was fairly constant at 4-5%. Overall, the comparison of these data to current scientific literature showed good agreement with both time spent indoors, as well as time spent in transportation micro-environments .One fundamental piece of information necessary to estimate dose is the particle number size distribution to which the different age groups were exposed in the different micro-environments. The maximum and minimum values of the particle number size distributions for the different micro-environments were obtained from our measurement survey data, with the highest particle number concentrations observed during cooking activities, under some working conditions and in transportation micro-environments. As expected, the concentration level varied widely among different types of micro-environments, with the daily alveolar particle number (surface area) deposited from the all age groups equal to 1.5 x 10^{11} particles (2.5 x 10^{15} μm^2), varying slightly for males and females living in Northern or Southern Italy. In terms of tracheobronchial deposition, the corresponding values were equal to 6.5 x 10^{10} particles (9.9 x 10^{14} μm^2). Overall, deposition increased as a function age group, with the maximum value observed for 41-65 years old. The elderly age group (> 65 years), on the other hand, displayed lower deposition rates with respect to adults. This trend mainly depended on the increased exposure of adults in the different micro-environments, as opposed to physiological differences, such as respiratory inhalation rate. Finally, the major contributions to exposure dose, which were estimated on the basis of "exposure intensity" were found to arise from indoor cooking and eating times (female), working time (male) and transportation (children).

References

Ashmore, M.R., Dimitroulopoulou, C., 2009. Personal exposure of children to air pollution, Atmospheric Environment 43, 128–141

Bastone, A., Soggiu, M.E., Vollono, C., Masciocchi, M., Rago, G., Sellitri, C., 2003, Assessment study on inhalation exposure to atmospheric pollution in Ferrara. First phase, Rapporti ISTISAN 03/19, Istituto Superiore di Sanità (Italy), in Italian.

Bastone, A., Soggiu, M.E., Vollono, C., Viviano, G., Masciocchi, M., Rago, G., Sellitri, C., Spagnolo, S., Spartera, M., 2006. Lifestyles and behaviour of Taranto, Massafra, Crispiano and Statte populations for inhalation exposure assessment to atmospheric pollution, Rapporti ISTISAN 06/36, Istituto Superiore di Sanità (Italy), in Italian.

Berghmans, P., Bleux, N., Int Panis, L., Mishra, V.K., Torfs, R., Van Poppel, M., 2009. Exposure assessment of a cyclist to PM_{10} and ultrafine particles. Science of the Total Environment 407(4), 1286-1298.

Bramoullé, Y, Olson, L.J., 2005. Allocation of pollution abatement under learning by doing, Journal of Public Economics, 89, 1935–1960.

Brasche, S., Bischof, W., 2005. Daily time spent indoors in German homes – Baseline data for the assessment of indoor exposure of German occupants, Int. J. Hyg. Environ.-Health, 208, 247–253.

Brugge, D., Durant, J.L., Rioux, C., 2007. Near-highway pollutants in motor vehicle exhaust: a review of epidemiologic evidence of cardiac and pulmonary health risks. Environmental Health 6, 23.

Buonanno G. Lall A., Stabile L., 2009a. Temporal size distribution and concentration of particles near a major highway, Atmospheric Environment, 43, 1100–1105

Buonanno G. Morawska L., Stabile L., 2009b. Particle emission factors during cooking activities, Atmospheric Environment, 43, 3235–3242

Buonanno G., Morawska L., Stabile L., Viola A., 2010. Exposure to particle number, surface area and PM concentrations in pizzerias, Atmospheric environment, 44, 3963-3969.

Buonanno, G., Fuoco, F.C., Stabile, L., 2011a, Influential parameters on particle exposure of pedestrians in urban microenvironments, Atmospheric Environment 45, 1434-1443.

Buonanno G., Graham J., Morawska L., Stabile L. 2011b. Volatility characterization of cooking-generated aerosol particles, in press on Aerosol Science & Technology.

Buonanno G., Morawska L., Stabile L., 2011c. Exposure to welding particles in automotive plants, in press on Environmental Science & Technology.

California Environmental Protection Agency. Measurement of breathing rate and volume in routinely performed daily activities. Sacramento (CA): California Air Resources Board; 1993.

Chau, C.K., Tu, E.Y., Chan, D.W.T., Burnett J., 2002. Estimating the total exposure to air pollutants for different population age groups in Hong Kong, Environment International 27, 617–630.

Cheng, Y.S., 2003, Aerosol Deposition in the Extrathoracic Region, Aerosol Science and Technology 37. 659–671.

Cheng, Y.S., Zhou, Y., Chen, B. T., 1999. Particle Deposition in a Cast of Human Oral Airways, Aerosol Science and Technology 31. 286-300.

Dellink, R., van Ierland, E., 2006. Pollution abatement in the Netherlands: A dynamic applied general equilibrium assessment Journal of Policy Modeling 28, 207–221

Dennekamp, M., Mehenni, O.H., Cherrie, J.W., Seaton, A., 2002. Exposure to Ultrafine Particles and $PM_{2.5}$ in Different Micro-environments. Annal of Occupational Hygiene 46(S1) 412–414.

Elihn, K., Berg, P., 2009. Ultrafine Particle Characteristics in Seven Industrial Plants, Ann. Occup. Hyg. 53(5), 475–484.

EPA, U.S. Environmental Protection Agency, 2009, Metabolically derived human ventilation rates: a revised approach based upon oxygen consumption rates, National Center for Environmental Assessment, Office of Research and Development, EPA/600/R-06/129F

Evans, D.E., Heitbrink, W.A., Slavin, T.J., and Peters,T.M., 2008. Ultrafine and Respirable particles inanautomotive grey iron foundry. Annals of Occupational Hygiene 52(1), 9-21.

Fromme, H., Twardella, D., Dietrich, S., Heitmann, D., Schierl, R., Liebl, B., Rüden, H., 2007. Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area. Atmospheric Environment 41, 854–866.

Giechaskiel, B., Alföldy, B., Drossinos, Y., 2009. A metric for health effects studies of diesel exhaust particles. Journal of Aerosol Science 40, 639-651.

Hammersley, J. M., Handscomb, D. C., 1964. Monte Carlo Methods, Chapman and Hall, London & New York.

Hänninen, O., Zauli-Sajani, S., De Maria, R., Lauriola. P., Jantunen, M., 2009. Integrated ambient and microenvironment model for estimation of PM10 exposures of children in annual and episode settings, Environmental Monitoring & Assessment 14, 419-429.

Hauser, R., Godleski, J.J., Hatch, V., Christiani, D.C., 2001. Ultrafine particles in human lung macrophages. Archives of Environmental Health 56, 150–156.

He, C., Morawska, L., Hitchins, J., Gilbert, D., 2004. Contribution from indoor sources to particle number and mass concentrations in residential houses, Atmospheric Environment, 38, 3405–3415.

International Commission on Radiological Protection (ICRP), 1994. Human Respiratory Tract Model for Radiological Protection: A Report of a Task Group of the International Commission on Radiological Protection. Elsevier Science Ltd., Oxford, U.K., pp. 1-482.

Istituto Superiore Formazione e Ricerca per i Trasporti (ISFORT), AUDIMOB – Osservatorio sui comportamenti degli italiani – La domanda di mobilità degli italiani, Giugno 2010, in Italian.

Kahaner, D., Moler, C., Nash, S., Numerical Methods and Software, Prentice Hall, 1988

Kaur, S., Nieuwenhuijsen, M.J., Colvile, R., 2007. Fine particulate matter and carbon monoxide exposure concentrations in urban street transport microenvironments. Atmospheric Environment 41, 4781-4810.

Klepeis, NE. 2006. Modeling Human Exposure to Air Pollution. In: Human Exposure Analysis, ed., Ott et al., CRC Press.

Kreyling, W.G., Semmler-Behnke M., Moller W., 2006. Health implications of nanoparticles. Journal of Nanoparticle Research 8, 543–562.

Leech, J.A., Nelson, W.C., Burnett, R.T., Aaron, S., Raizenne, M.E., 2002. It's about time: a comparison of Canadian and American time-activity patterns. J. Expo. Anal. Environ. Epidemiol. 12, 427–432.

Loomis, D., 2000. Sizing up air pollution research. Epidemiology 11, 2-4.

Mejìa, J.F., Wraith D., Mengersen K., Morawska L., 2007. Trends in size classified particle number concentration in subtropical Brisbane, Australia, based on a 5 year study, Atmospheric Environment 41, 1064–1079

Miller, K.A., Siscovick, D.S., Sheppard, L., Shepherd, K., Sullivan, J.H., Anderson, G.L., Kaufman, J.D., 2007. Long-term exposure to air pollution and incidence of cardiovascular events in women. The New England Journal of Medicine 365, 447-458.

Ott, W.R., 1982. Concepts of human exposure to air pollutants. Environmental International 7, 179–196.

Pope, C.A., 2000. What do epidemiologic findings tell us about health effects of environmental aerosols?. Journal of Aerosol Medicine 13, 335-354.

Schwartz, J., 1991, Air Pollution and daily mortality in Philadelphia. Presented at the 1991 Meeting of the American Lung Association, Anaheim, CA.

See, S.W., Balasubramanian, R., 2006. Risk assessment of exposure to indoor aerosols associated with Chinese cooking. Environ. Res. 102 (2), 197–204.

Shi J. P., Khan, A.A. Harrison, R.M., 1999. Measurements of ultrafine particle concentration and size distribution in the urban atmosphere, The Science of the Total Environment, 235, 51-64

Siegmann, K., Siegmann, H.C., 1998. Molecular Precursors of Soot and Quantification of the Associated Health Risk. Plenum Press, New York, pp. 143–160.

Soggiu, M.E., Bastone, A., Vollono, C., Masciocchi, M., Rago, G., Sellitri, C., 2005, Study of inhalation exposure assessment to atmospheric pollution in Ferrara. Second phase, Rapporti ISTISAN 05/09, Istituto Superiore di Sanità (Italy), in Italian.

Soggiu, M.E., Vollono, C., Bastone, A., 2010. Human exposure assessment to environmental contaminants: exposure scenarios. Rapporti ISTISAN 10/19, Istituto Superiore di Sanità (Italy), in Italian.

Sundqvist, G, Letell, M., Lidskog, R., 2002. Science and policy in air pollution abatement strategies, Environmental Science & Policy 5,147–156

USEPA. 2004. Air Quality Criteria for Particulate Matter (Final Report, Oct 2004) (No. EPA 600/P-99/002aF-bF). Washington, DC: Environmental Protection Agency.

Vedal, S., 1997. Ambient particles and health: lines that divide. Journal of the Air and Waste Management Association 47, 551–581.

Wallace, L., and Ott, W., 2011. Personal exposure to ultrafine particles. Journal of Exposure Science and Environmental Epidemiology 21, 20–30.

Wang, L., Morawska, L., Jayaratne, E.R., Mengersen, K., Heuff, D., 2011. Characteristics of airborne particles and the factors affecting them at bus stations, Atmospheric Environment, 45 (3), 611-620.

Zhu, Y., Hinds, W.C., Krudysz, M., Kuhn, T., Froines, J., Sioutas, C., 2005. Penetration of freeway ultrafine particles into indoor environments, Aerosol Science, 36, 303–322.

Table 1 – Inhalation rates (m^3 h^{-1}) as a function of activity and age group.

Activities	Age (years)					
	1-5	**6-10**	**11-18**	**19-40**	**41-65**	**>65**
Sleeping and Resting	0.31	0.31	0.36	0.36	0.41	0.41
Sedentary activities	0.42	0.42	0.45	0.45	0.49	0.49
Walking (not along a road)	0.58	0.58	0.60	0.60	0.63	0.63
House cleaning	0.62	0.62	0.64	0.64	0.67	0.67
Cooking	0.48	0.48	0.50	0.50	0.54	0.54
Eating	0.42	0.42	0.45	0.45	0.49	0.49
Entertainment indoor	0.91	0.91	0.94	0.94	1.01	1.01
Entertainment outdoor	0.91	0.91	0.94	0.94	1.01	1.01
Sport indoor	1.27	1.27	1.36	1.36	1.59	1.59
Sport outdoor	1.44	1.44	1.67	1.67	1.94	1.94
Sedentary job	0.42	0.42	0.45	0.45	0.49	0.49
Non sedentary job	0.91	0.91	0.94	0.94	1.01	1.01
School	0.42	0.42	0.45	0.45	0.49	0.49
Studying	0.42	0.42	0.45	0.45	0.49	0.49
Playing indoor	0.91	0.91	0.94	0.94	1.01	1.01
Playing outdoor	1.27	1.27	1.36	1.36	1.59	1.59
Transportation	0.58	0.58	0.60	0.60	0.63	0.63

Table 2 – Particle number size distributions, in terms of lognormal functions, applied in the different microenvironments

Microenvironment	**Mode nm**	**Standard deviation σ**	**N_{max} (N_{min}) particles cm^{-3}**	**Our measurement survey data**	**Other references (average particle number concentration, particles cm^{-3})**
Indoor evening & night	42	2.0	1.0×10^4 (7.0×10^3)	Buonanno et al., 2011a	Zhu et al., 2005 (9.0×10^3)
Indoor day	40	2.2	2.0×10^4 (1.0×10^4)	Buonanno et al., 2011a	Fromme et al., 2007 (1.2×10^4)
Outdoor day	40	2.1	2.5×10^4 (1.40×10^4)	Buonanno et al., 2011a	Shi et al., 1999 (2×10^4) Mejìa et al., 2007 (1×10^4)
Cooking	65	1.9	6.9×10^5 (1.0×10^5)	Buonanno et al., 2009b	See and Balasubramanian, 2006; (7.7×10^5) He et al., 2004 (2×10^5)
Eating	70-100	1.7-1.8	2.0×10^5 (7.0×10^4)	Buonanno et al., 2009b	Dennekamp et al., 2002 (6×10^4)
Working	47	1.8	1.5×10^5 (1.0×10^5)	Buonanno et al., 2011c	Elihn and Berg, 2009 (1.0×10^5)
Restaurants & pizzerias	50	1.8-2.0	1.2×10^5 (5.7×10^4)	Buonanno et al., 2010	Wallace et al., 2011 (1.7×10^5)
Pedestrian and bikes	*mode 1:* 11 *mode 2:* 40	σ_1: 1.6 σ_2: 1.6	9.0×10^4 (4.5×10^4)	Buonanno et al., 2011a	Berghmans et al., 2008; (4×10^4) Kaur et al., 2007 (8×10^4)
Car and buses	*mode 1:* 11 *mode 2:* 38	σ_1: 1.6 σ_2: 1.6	7.0×10^4 (4.0×10^4)	Buonanno et al., 2011a	Kaur et al., 2007; (6×10^4)

Table 3 - Daily time activity patterns (minutes) for Northern and Southern Italy (F - female, M – male)

Activity identifying number	**Activities (n_a)**	**Gender**	**Age (years)**						
			<1	**1-5**	**6-10**	**11-18**	**19-40**	**41-65**	**>65**
1	Sleeping & Resting	F/M (South) F/M (North)	703/660 -/-	584/611 615/622	537/526 550/569	511/518 509/514	469/460 466/452	443/442 454/451	435/434 466/470
2	Sedentary activities	F/M (South) F/M (North)	-/- -/-	127/141 152/165	149/144 166/170	132/140 184/179	128/131 171/194	171/155 199/215	267/236 320/302
3	Walking (not along a road)	F/M (South) F/M (North)	90/80 -/-	58/55 36/-	49/57 31/-	58/68 43/2	70/76 48/16	73/77 42/14	69/96 48/28
4	House cleaning	F/M (South) F/M (North)	-/- -/-	38/- -/-	29/- 2/-	66/29 7/29	141/45 73/45	166/73 108/73	126/82 105/82
5	Cooking	F/M (South) F/M (North)	-/- -/-	30/- -/-	19/- 1/1	39/100 4/1	78/42 51/17	87/61 82/18	79/62 82/28
6	Eating	F/M (South) F/M (North)	86/128 -/-	70/80 -/-	73/65 -/-	76/72 -/-	70/72 -/-	69/74 -/-	71/77 -/-
7	Hobbies indoor	F/M (South) F/M (North)	330/180 -/-	109/165 1/3	148/131 2/-	155/158 4/2	153/163 24/13	139/148 32/18	141/153 30/29
8	Hobbies outdoor	F/M (South) F/M (North)	171/45 -/-	189/230 3/3	184/264 10/15	198/242 28/33	194/210 16/21	176/183 24/31	172/187 21/56
9	Sport indoor	F/M (South) F/M (North)	-/- -/-	90/- 1/3	94/87 17/12	111/119 19/17	101/115 7/10	82/88 5/5	120/93 1/4
10	Sport outdoor	F/M (South) F/M (North)	-/- -/-	-/300 1/2	70/102 4/17	116/115 9/30	101/116 6/12	90/98 7/12	99/220 2/7
11	Sedentary job	F/M (South) F/M (North)	-/- -/-	-/- -/-	-/- -/-	-/- -/-	339/346 122/168	323/348 113/182	-/- 7/32
12	Non sedentary job	F/M (South) F/M (North)	-/- -/-	-/- -/-	-/- -/-	-/357 2/-	339/442 95/147	284/367 66/102	-/325 4/9
13	School	F/M (South) F/M (North)	-/- -/-	250/296 -/-	304/297 -/-	324/308 -/-	281/237 -/-	-/- -/-	-/- -/-
14	Studying	F/M (South) F/M (North)	-/- -/-	-/- 8/1	94/83 158/167	137/117 203/198	237/216 30/23	80/53 6/12	-/- 1/2
15	Entertainment indoor	F/M (South) F/M (North)	260/297 -/-	184/220 266/250	138/134 105/100	120/109 21/46	-/- 18/16	-/60 3/5	-/- 2/6
16	Entertainment outdoor	F/M (South) F/M (North)	276/- -/-	147/147 85/101	145/136 62/55	141/174 27/24	-/- 6/7	-/- 2/2	-/- 1/3
17	Walking or bicycle	F/M (South) F/M (North)	12/14 13/14	12/14 13/14	13/15 12/16	13/15 12/16	11/14 13/16	14/17 14/16	22/23 20/24
18	Motorbike	F/M (South) F/M (North)	-/- -/-	-/- -/-	-/- -/-	3/6 4/5	3/4 3/5	1/2 2/3	-/- -/-
19	Car	F/M (South) F/M (North)	43/45 44/47	43/46 44/46	43/45 43/46	41/44 42/45	44/47 45/48	42/45 44/45	26/27 27/29
20	Bus	F/M (South) F/M (North)	16/17 17/18	16/17 17/18	16/17 17/18	15/17 16/17	11/12 10/13	5/6 6/8	5/8 7/9
21	Restaurants and pizzerias	Evaluated as 49.3% of the total eating time for working people and one time a week for the other age groups older than 18 years.							

Figure Captions

Figure 1 - Normalized daily time activity patterns for Northern and Southern Italy and corresponding standard deviations for each age group and gender, by considering the 21 activities: Sleeping & Resting (1), Sedentary activities (2), Walking through different indoor microenvironment (3), House cleaning (4), Cooking (5), Eating (6), Hobbies indoor (7), Hobbies outdoor (8), Sport indoor (9), Sport outdoor (10), Sedentary job (11), Non sedentary job (12), School (13), Studying (14), Entertainment indoor (15), Entertainment outdoor (16), Walking or bicycle (17), Motorbike (18), Car (19), Bus (20), Restaurants and pizzerias (21).

Figure 2 - Average daily dose with the corresponding standard deviations as a function of age group, gender and location (Northern and Southern Italy). a) Alveolar particle number deposited, b) alveolar surface area deposited, c) tracheobronchial particle number deposited, b) tracheobronchial surface area deposited.

Figure 3 – Contribution of different activities to daily activity patterns, average daily alveolar and tracheobronchial particle number and surface area deposited for each age group investigated: Sleeping & Resting (1), Sedentary activities (2), Walking (not along a road) (3), House cleaning (4), Cooking (5), Eating (6), Hobbies indoor (7), Hobbies outdoor (8), Sport indoor (9), Sport outdoor (10), Sedentary job (11), Non sedentary job (12), School (13), Studying (14), Entertainment indoor (15), Entertainment outdoor (16), Walking or bicycle (17), Motorbike (18), Car (19), Bus (20), Restaurants and pizzerias (21).