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Evaluating the infiltration of outdoor black carbon into residential indoor environments in Stockholm, Sweden

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Abstract

Combustion generated particulate matter (PM) have been more closely associated with adverse health effects, than PM from other sources. Black Carbon (BC) is a constituent in combustion generated PM from carbon containing fuels. More robust relationships have been found linking BC to estimated health effects, including cardiovascular and respiratory disease, than to $PM_{2,5}$ and PM_{10} which are the current subjects of measurement utilized in epidemiological contexts. Human exposure to traffic-related PM, including BC, is regularly assessed by the monitoring of ambient outdoor concentrations. This might not be representative for the real situation since people spend most of their time indoors. For improved estimations of human exposure, a better understanding of the mechanisms behind the infiltration of BC (generated outdoors) into residential buildings is needed. The most important parameters influencing particle infiltration are the air exchange rate (a) which is the rate in h^{-1} at which the indoor air is exchanged, the penetration factor (P) which is the fraction of the outdoor concentration following the inflowing air, and the deposition rate of particles indoors (k) which is the decay rate of particles due to deposition onto indoor surfaces. A particle infiltration factor (F_{inf}), defined as the amount of particles that enters the indoor environment and remains suspended there, can be quantified using parameters a, P and k in the equation $F_{inf} = (a \cdot P) / (a + k)$. This study aims to contribute to the knowledge of how these parameters are related to each other with the intent to improve future assessment of human exposure to BC. Time-resolved indoor and outdoor concentrations of BC and size distributed particles, ranging from 0,3 to 10 µm were monitored in three naturally ventilated, occupied apartments in Stockholm, Sweden. a and k values were experimentally determined by a decay rate test using candle generated particles, including BC, and carbon dioxide (CO_2). F_{inf} factors were estimated from measured indoor and outdoor concentrations, and then P values were calculated for BC and the size distributed particles. Indoor concentrations for BC are measured to 0,59, 0,75 and 0,46 µg m^{-3}, with indoor sources, and 0,58, 0,66 and 0,38 µg m^{-3} without indoor sources. Outdoor concentrations are measured to 0,77, 0,77 and 0,61 µg m^{-3}, a values are estimated to 0,35, 0,24 and 0,34 h^{-1} and BC specific k values are 0,56 (± 0,11 (s.d.), 0,34 (± 0,01) and 0,23 (± 0,07) h^{-1} for Artillerigatan, Grevgatan and Karlavägen, respectively. F_{inf} factors for BC, estimated to 0,55 (± 0,02 (95% C.I.)), 0,64 (± 0,02) and 0,41 (± 0,05) at Artillerigatan, Grevgatan and Karlavägen, respectively, suggests that results in epidemiological studies based on outdoor exposure to traffic-related BC might be misleading by approximately a factor of 2. The P values calculated to 1,41, 1,53 and 0,67 (theoretically expected to be in the range of 0 to 1) is assumedly due to the source specific composition of BC and that k values of candle generated BC are not sufficiently representative for traffic generated BC. There are many uncertainties involved when dealing with the complex environment that an occupied apartment is, and further developing of applicable methods are important for assessing human exposure to BC. The experimentally estimated values in this study are discussed in the light of corresponding results from previous studies. Size distributed estimations of variables k and F_{inf} reflected their size dependence. According to the results of this study, k for BC indicated a BC size distribution peaking at a particle size below 0,3 µm, which is in line with the ~150 nm estimated from literature.

Contents

Nomenclature	
C_{BC}	Concentration of black carbon
C_x	Concentration of particles with an equivalent particle diameter of x µm
C_{in}	Indoor concentration
C_{out}	Outdoor concentration
I/O ratio	indoor/outdoor ratio (C_{in}/C_{out})
F_{inf}	infiltration factor
a	air exchange rate (h^{-1})
k	deposition rate (h^{-1})
P	penetration factor (dimensionless)
When nothing else is stated, particle sizes refers to the equivalent particle diameter given in µm, corresponding to 10^{-6} m (SI units), and mass concentrations are given in µg m^{-3}.	

Introduction

Background

Particulate matter

Particulate matter (PM) in the atmosphere make up the suspended phase which is dissolved in a gas, a two-component system called an aerosol. PM can be a complex mixture of innumerable compounds with different physical and chemical properties that determines their fate in the atmosphere. Particles are generally categorized in respect to their size, which is the most important characteristic influencing a particles behavior (Hinds, 1999), commonly defined by the equivalent particle diameter using the denomination PM_x, where the x stands for the highest equivalent particle diameter, in µm, represented in that category. As an example, the concentration of $PM_{2,5}$ is the concentration of all particles with an equivalent particle diameter less than 2,5 µm. There is a lot of evidence that PM is responsible for adverse health effects and this is especially true for PM_{10} (particles with an equivalent particle diameter less than 10 µm), which are inhalable (Meng, et al., 2013). 1 µm is the upper limit of the submicron range, which is an important dividing line since submicron sized particles, can be deposited in the deep lung through inhalation (LaRosa, et al., 2002).

PM can be generated indirectly in the atmosphere by gaseous precursors interacting with e.g. sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds (VOCs) (Hänninen & Knol, 2011). There are both anthropogenic and natural direct sources of PM (Miljöförvaltningen, 2011). Major anthropogenic sources of PM are the exhaust emissions by combustion engines, residential burning of wood and coal, the use of heavy oil or coal for power supply and field burning of agricultural wastes. Natural sources include forest and vegetation fires, dust and pollen (Janssen, et al., 2012). There are a lot of studies done on component-specific associations with adverse health effects linked to PM, but in epidemiological contexts, it is still customary to relate health effects with total mass concentrations of PM_{10} and $PM_{2,5}$ (Hänninen & Asikainen, 2013).

Black Carbon

When carbon containing fuels, e.g. fossil fuels and biomass, are not completely combusted, a PM mixture is produced containing different proportions of black carbon, primary organic matter and inorganic salts (Laborde, et al., 2013). These substances have a large variety of chemical and physical

properties and accordingly, environmental behavior. Black carbon (BC) as a constituent in this mixture functions as an indicator of combustion sources (Janssen, et al., 2012, Ning, et al., 2013). BC is the main component in the historically used term “soot” and is operationally defined as carbon that absorbs light (LaRosa, et al., 2002), a property that is utilized in monitoring BC by light absorbance measurements. The major constituent in atmospheric BC, is elemental carbon (EC) formed by the combustion of natural carbon. It is present in two forms with differing chemical-, physical- and optical light-absorbing properties; brown “char-EC” with partly preserved graphite-like structure, and black “soot-EC” where the original structure of natural carbon is not preserved (Janssen, et al., 2012). BC is considered to be chemically stable but is subject for a number of physical alteration processes once emitted from its source, referred to as aging. Aging involves the processes in which primary “fresh” hydrophobic BC interacts with other aerosol particles and is transformed to become more hygroscopic e.g. through oxidation processes at the surface of BC particles. The size of BC is generally increased by aging, irrespective of the original size (Park, et al., 2011). Primary BC particulate matter, emitted during the combustion of diesel fuel is initially in the form of small (~ 25 nm) spherules and then immediately agglomerates into chainlike structures hence with larger equivalent particle diameters. In a study performed in a typical urban site of Hong Kong, the mass size distribution of BC peaked at an equivalent diameter of 100-150 nm when measured in aerosols associated with diesel exhaust and at somewhat larger equivalent diameters (~ 200 nm) in roadside and ambient measurements, indicating an ageing process of BC in the atmosphere once emitted (Ning, et al., 2013). In a study made in Paris, where traffic emissions are a dominant source of BC, the mass equivalent diameter was reported to peak at an equivalent diameter of 150 nm (Laborde, et al., 2013).

Health effects of BC

Fatal diseases caused by indoor and outdoor air pollution are ischaemic heart disease (causing insufficient blood supply), stroke, chronic obstructive pulmonary disease (COPD), lung cancer and acute lower respiratory infections in children. In March this year (2014) the world health organization (WHO) released data stating that exposure to air pollution, both indoors and outdoors, was responsible for “one in eight of total global deaths” in the year of 2012, and that the link between exposure to air pollution and the development of diseases caused by it, such as cardiovascular diseases and cancer in addition to respiratory, is stronger than previously estimated (Jasarevic, et al., 2014). Diesel PM emissions are linked to a number of adverse health effects and are classified as a Category 1 carcinogen by WHO (Ning, et al., 2013).

In a publication from the World Health Organization in 2012 evidences were considered being sufficient to state that short-term variations in BC concentrations can be associated with short-term variations in public health. Both short- and long-term studies show much higher estimated health effects caused by BC compared with $PM_{2,5}$ and PM_{10} when expressed in the same mass concentration unit ($\mu g/m^3$). Adverse effects that are associated with daily variations in BC concentrations are mortality, all-cause and cardiovascular, and cardiopulmonary hospital admissions. In the same publication it is also considered sufficiently evident, that long-term average exposure to BC is associated with all-cause and cardiopulmonary mortality (Janssen, et al., 2012).

Although BC is generally considered to be chemically inert (Ning, et al., 2013), recent toxicological studies suggests that EC, which is the major constituent in BC (LaRosa, et al., 2002), act as a universal carrier for toxic compounds. Gases, suspended particles and co-released combustion derived substances, all including toxic compounds, adhere to the active surface areas of BC during regional and/or long-range transport (Janssen, et al., 2012). During the combustion of diesel and gasoline, BC is coated with co-released organic matter including carcinogenic nitrous compounds and polycyclic hydrocarbons (PAH). The size of most BC particles emitted by diesel engines enable them to penetrate deep into the lungs of a human inhaling them, where co-penetrated compounds subsequently can cause cancer when deposited (LaRosa, et al., 2002).

BC in Stockholm, Sweden

In Stockholm, the capital of Sweden, the major source of BC in the more urban parts is fuel combustion associated with traffic, especially by diesel driven vehicles which emit more compared with gasoline driven vehicles. Heavy-duty vehicles, like trucks and buses, emit more BC per distance compared with light-duty vehicles, like cars and light vans. Other sources of BC in Stockholm are shipping, power plants and residential wood burning. During winter, wood burning are sometimes the major source of BC. In Stockholm, the street level and the urban background concentrations of BC are recorded at stationary monitoring stations situated at Hornsgatan and Torkel Knutssonsgatan, respectively. The concentration of BC typically decreases with height and the concentrations measured at the station on Hornsgatan is usually three times the concentration on Torkel Knutssonsgatan, which is reflected by the 2013 annual mean concentration of 2,5 µg BC m^{-3} and 0,78 µg BC m^{-3}, respectively (SLB-analys, 2014).

Human exposure in an urban environment

Human exposure to BC in urban areas occurs when people breathe in traffic-related fumes (AethLabs, 2014). A large number of previously conducted studies have shown a significant relationship between the risk of adverse health effects and the exposure to traffic-related PM, from which the exposure to BC can be estimated. To date, epidemiological risk analyses are based on modelled or measured outdoor concentrations of PM. On average, people spend more than 85 % of their time indoors (Diapouli, et al., 2013, Pagels, et al., 2009, Lunden, et al., 2008, Polidor, et al., 2007), therefore indoor exposure needs to be included in the assessment of human exposure.

Indoor concentrations

Indoor particle concentrations, C_{in}, are a result of all the generating- and removing processes of BC in that indoor environment (Nazaroff, 2004). Black Carbon is present in indoor environments both due to infiltration of BC being emitted outdoors and from indoor sources generating BC (LaRosa, et al., 2002). Health benefits can be won from reducing the indoor exposure to BC, and this can be done by optimizing ventilation and filtration as well as improving the identification of the indoor sources of BC (Hänninen & Asikainen, 2013). In this study, alterations in indoor particle concentrations are assumed to depend solely on particle movement in space, e.g. by advective flow or through diffusion. Processes like coagulation, chemical reactions and thermodynamically induced phase changes are not considered. C_{in} can be expressed by the equation $C_{in} = F_{inf} * C_{out} + C_{ś,}$ where F_{inf} is the infiltration factor of the outdoor concentration, C_{out}, and $C_{ś}$ is the indoor particle concentration resulting from indoor sources (Chen & Zhao, 2011). Important indoor sources of BC are cooking and candle burning (LaRosa, et al., 2002). The magnitude and characteristics of particles produced from cooking activities, that ad up to C_{in}, varies with the type of cooking, including preparation approaches and cookware being used, as well as building characteristics, such as air flow patterns and construction factors. The particle concentration increase is also dependent on the space between the particle source and the measurement location. In a previous study, the maximum value of the particle number concentration observed when cooking on an electrical stove was $1{,}1*10^6$ cm^{-3}, compared to a corresponding value for a gas stove which was observed to be $5{,}6*10^6$ cm^{-3} (Hussein, et al., 2006).

Different modes of candle burning generate large differences in particle size distribution of the candle emissions. The different modes are steady burn, which emissions consists primarily of ultrafine particles of phosphates and alkali nitrates, sooting burn, which emissions contain larger particles consisting mainly of EC, and smouldering upon extinction, which primarily consists of organic matter. Sooting burn, with an average “mobility diameter” of 270 nm was shown to generate the largest emissions in a study by (Pagels, et al., 2009).

The indoor exposure to particles of outdoor origin is dependent on the infiltration processes of these particles along with the infiltrating air. The ratio between the indoor- and the outdoor concentrations (I/O ratio) of a particle pollutant, is quantified by calculating the fraction C_{in}/C_{out}.

Even in the absence of indoor sources of BC, C_{in} is not equal to C_{out} due to particle loss during infiltration processes and the different kinds of interactions occurring indoors compared with outdoors (Liu & Nazaroff, 2001). The most important parameters describing the infiltration, referred to as the controlling parameters in an indoor aerosol model by (Hussein & Kulmala, 2007), are the air exchange rate between indoors and outdoors, a (h^{-1}), the penetration factor, P (dimensionless), and the deposition rate, k (h^{-1}). In naturally ventilated buildings, such as the ones in this study, k and P are even more important than in mechanically ventilated buildings in influencing the size dependent C_{in} of particles (Chao, et al., 2003). The rate of change in C_{in}, assuming the absence of indoor sources, is generally expressed as a function of time by the equation $\Delta C_{in}/\Delta t = a*P*C_{out} - KC_{in}$, where K is the total decay rate constant, a + k (h^{-1}) (Vette, et al., 2001).

Ventilation

In order to predict the concentration of PM, including soot, in an indoor environment, the air flow between the outside and the inside of the building shell needs to be estimated. Ventilation of an apartment is referred to the process where outflowing indoor air is replaced with inflowing outdoor air, and thereby the indoor air is exchanged. The ventilation of an apartment can be quantified by the air exchange rate (h^{-1}), which is defined as the total inflow rate of outdoor air (commonly expressed in $m^3\ h^{-1}$) divided by the internal volume that make up the indoor environment of the apartment (expressed in m^3). The three main ways by which an apartment is usually ventilated are mechanical ventilation, natural ventilation and infiltration of air by leakage flow (Nazaroff, 2004). Even if significant infiltration of particles of outdoor origin can occur in mechanically ventilated buildings (Chen & Zhao, 2011), mechanical ventilation was not considered in this work.

Natural ventilation occurs when outdoor air flow through an open window or door with wind and temperature as the driving forces (Orme, et al., 1998). The indoor air having a higher temperature compared with the outdoor air gives rise to a pressure gradient across the building shell with the indoor air having a lower pressure compared with the outdoor air. This pressure gradient generates a “chimney effect”, which natural ventilation is based on and it causes the inflow of outdoor air into the suppressed indoor environment. The indoor air rises, as it is heated, and higher up in the building the pressure of the indoor air is higher compared with the outdoor air, causing an outflow of the indoor air (Hussein, et al., 2006). The chimney effect is also the principle behind the air leakage through the building envelope which is the operating air exchange process when windows and doors are closed. Since there is always a leakage through the building envelope (Liu & Nazaroff, 2001), the air exchange is carried out by a combination of leakage flow and natural ventilation in the absence of mechanical ventilation. In this study, the air exchange rates generated by only the leakage flow were estimated.

Infiltration factor

Particles of outdoor origin are infiltrated indoors along with the air leaking from outdoors through the building envelope (Chen & Zhao, 2011). The infiltration factor (F_{inf}) is a unitless term quantifying the fraction of the outdoor concentration of a pollutant that has entered an indoor environment and remained suspended there (Bennett & Koutrakis, 2006). It can be derived from the site specific values of a, P and k using the equation $F_{inf} = ((a*P)/(a+k))$, assuming steady-state. When windows and doors are closed in a naturally ventilated house, this is the primary pathway for particles to enter the indoor environment why the infiltration processes are important to understand in order to make good exposure estimations. F_{inf} can be calculated when indoor sources are absent. According to the equation $C_{in} = F_{inf} * C_{out} + C_{ś}$, C_{in}, $C_{in} = F_{inf} * C_{out}$ when there are no indoor sources and F_{inf} then equals the variable with which C_{out} is multiplied to calculate C_{in}. The slope generated by plotting the values of C_{in}, that are measured when indoor sources are negligible, against temporarily paired values of C_{out}, correspond to F_{inf}. The effect of indoor sources can be reduced by taking measurements at night or in an uninhabited building (Chen & Zhao, 2011).

Penetration factor

The fraction of C_{out} passing through the building shell along with the inflowing air is determined by a site specific penetration factor (P) (Chen & Zhao, 2011). When air enters the apartment through natural ventilation while opening a window or door, P is assumed to be 1, since there is not expected to be any particle attenuation due to the large openings through which the air is passed (Nazaroff, 2004, Hussein, et al., 2006). When outdoor air is entering the indoor environment by infiltration through cracks and wall cavities, the size distributed concentrations of PM, and thus the total PM concentration and its size fractions are altered. This is a result of loss processes occurring when the air is passing through the building shell. Particle losses are a consequence of three particle deposition mechanisms occurring in the building cracks (Liu & Nazaroff, 2001). These are Brownian diffusion, caused by net particle displacement (relative to its wandering motion) which results in the adhesion of particles to the building material due to van der Waals forces, impaction which is the collision of particles with other surfaces (Hinds, 1999) and gravitational settling (Liu & Nazaroff, 2001). Through the equation $F_{inf} = ((a*P)/(a+k))$, P is related to the ratio of a to K ((a)/(a + k)) assuming steady state, and P can thus be estimated when the site specific values of F_{inf}, a and k are known (Chao, et al., 2003).

Deposition

Particle deposition is here defined as total loss rate of particles in the indoor environment being monitored. There are many mechanisms that contribute to this deposition, and gravitational settling is the most important mechanism of deposition for coarse particles (> 2 µm (Nazaroff, 2004)), generating an increasing deposition rate with particle mass. When airborne particles, predominantly submicron particles, are sufficiently close to a solid surface, typically less than 1 cm (Nazaroff, 2004), to which they can adhere, they are deposited onto that surface. As a consequence, the air concentrations of those particles are decreased close to the surface, approaching zero very close to the surface, causing a lateral concentration gradient. According to Fick's first law of diffusion this will be counteracted by the equilibrating of particles at higher concentrations further away from the surface, resulting in a continuous decrease of the particle concentration in the room and acts as a sink of the airborne particles irrespective of initial source. In the calculations in this work, the surfaces in question are the ones present in the physical room in which the concentration measurements take place. They are walls, windows, roofs, floors and other objects with surfaces to which particles can adhere, like miscellaneous furniture and fabrics (Nomura, et al., 1997). The combined set of surface textures in a residence influence the deposition velocity of suspended particles. Higher deposition rates are obtained on rough and heterogenic surfaces which in existing residences are commonly represented by carpets and rough wallpaper (Abadie, et al., 2001). Calculating the diffusive deposition can be rather complicated when taking into account all the mechanisms responsible for the phenomena (Nomura, et al., 1997). In this work, the focus lies on defining deposition rates for the different particle sizes and residences that are the most probable real average deposition rate values.

Monitoring Black Carbon Mass Concentrations

In order to assess the real-time indoor and outdoor air concentrations of optically-absorbing BC, an Aethalometer can be used. The Aethalometer principle is based on measuring the change in rate of absorption by measuring the gradual increase in light attenuation caused by optically-absorbing particles deposited on a filter. An air sample with a known flow rate passes through the filter and suspended particles are continuously deposited on a sampling spot with a known area at a T60 Teflon-coated borosilicate glass fiber filter. Light at a wavelength of 880 nm produced by a LED light source is transmitted through the sampling spot as well as through a blank reference spot on the same filter. The incremental change in light absorption caused by the continuous increase in light absorbing particles on the sampling spot is measured by a photo diode detector. The light

attenuation is then converted to real-time BC mass, giving the BC concentration in ng m^{-3} when divided by the volume of air being sampled, which is decided by the flow rate (AethLabs, 2014).

Aim of this study

Improving the understanding of the mechanisms acting behind diseases caused by air pollution, together with the progress in the field of monitoring air pollution and exposure patterns, the assessment of health risks due to air pollution is continuously developed. With better understanding more requisite guidelines can be forwarded to policy makers hopefully resulting in public health gains (Jasarevic, et al., 2014). To date, all epidemiological studies relating BC exposure and most of the studies relating PM exposure to the risks of adverse health effects are based on measured and/or modeled outdoor concentrations of BC and PM, respectively. People spend most of their time indoors leading to biased estimations of the relationships between health effects and BC exposure when solely based on outdoor exposure (Johannesson, et al., 2007). To effectively reduce adverse health effects caused by both short- and long-term exposure to BC, more knowledge about the relationships between indoor and outdoor BC concentrations is needed. Identifying the major sources of indoor BC enables a more robust evaluation of which abatement measures, or regulating actions is predicted to give the largest health gains (LaRosa, et al., 2002). Neither in Sweden nor in the EU is BC an object for regulation, but the increasing evidences of the association between BC and morbidity and mortality have raised the awareness of BC in the last years (SLB-analys, 2014). These health effects are more distinctly associated with BC variations than with the variations of PM, which is a motive for measuring the BC mass concentration and use it as an indicator of combustion derived toxic compounds (such as traffic generated particles) rather than measuring the mass concentration of PM (Janssen, et al., 2012).

This work aims to contribute to a better understanding of the mechanisms of indoor infiltration of urban BC, as a more potent indicator of health disrupting agents than $PM_{2,5}$. By real-time monitoring of the indoor/outdoor concentration relationship of BC and the estimation of indoor parameters a, k and P in three different occupied apartments, in Stockholm Sweden, the intention was to assess the behavior of BC in common naturally ventilated indoor environments and thus being able to predict modeled future indoor concentrations of BC, when the outdoor BC concentration is known.

Method and materials

The monitoring took place in 3 different occupied apartments in Östermalm, which is one of the more residentially oriented city districts in the city of Stockholm, Sweden. Information about the apartments is compiled in Table 1.

Table 1, apartment characteristics

	Artillerigatan	*Grevgatan*	*Karlavägen*
area	130 m^2	34 m^2	150 m^2
ceiling height	3,33 m	3,1 m	2,6 m
volume	432,9 m^3	105,4 m^3	390 m^3
number of floors from street level	1	1	3
number of rooms	5	1	4
number of valves	5	2	7
windows facing	street	courtyard	street

A picture taken on the outdoor surroundings along with a drawing of the apartment is presented in Figure 1, 2, and 3 for Artillerigatan, Grevgatan and Karlavägen.

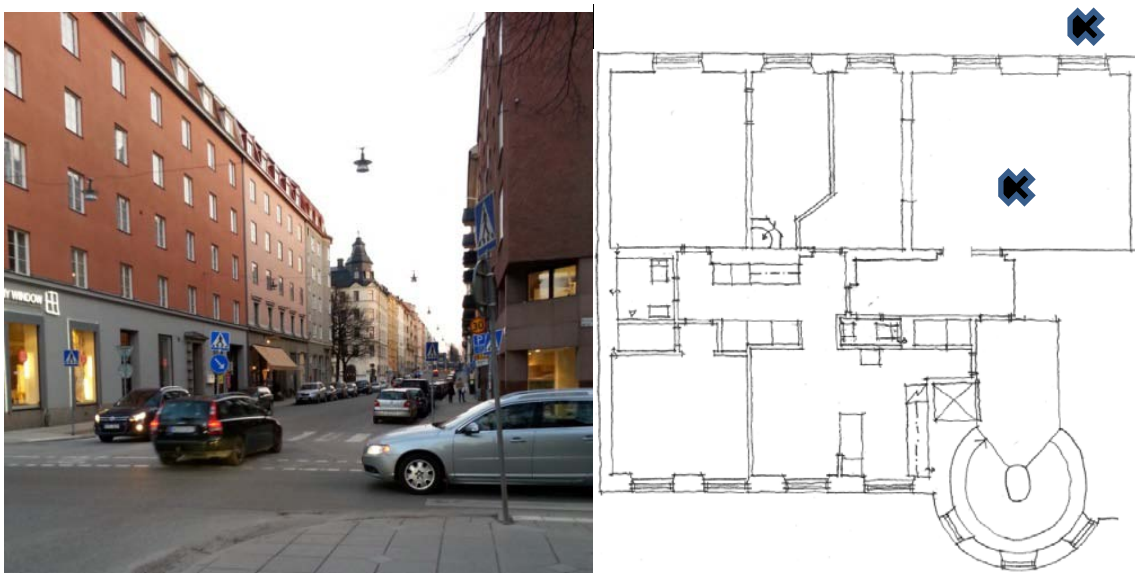

Figure 1 a, Artillerigatan street view (Ekebergh, 2014) b, drawing of the apartment on Artillerigatan with the measurement locations marked (Frondell, 2014)

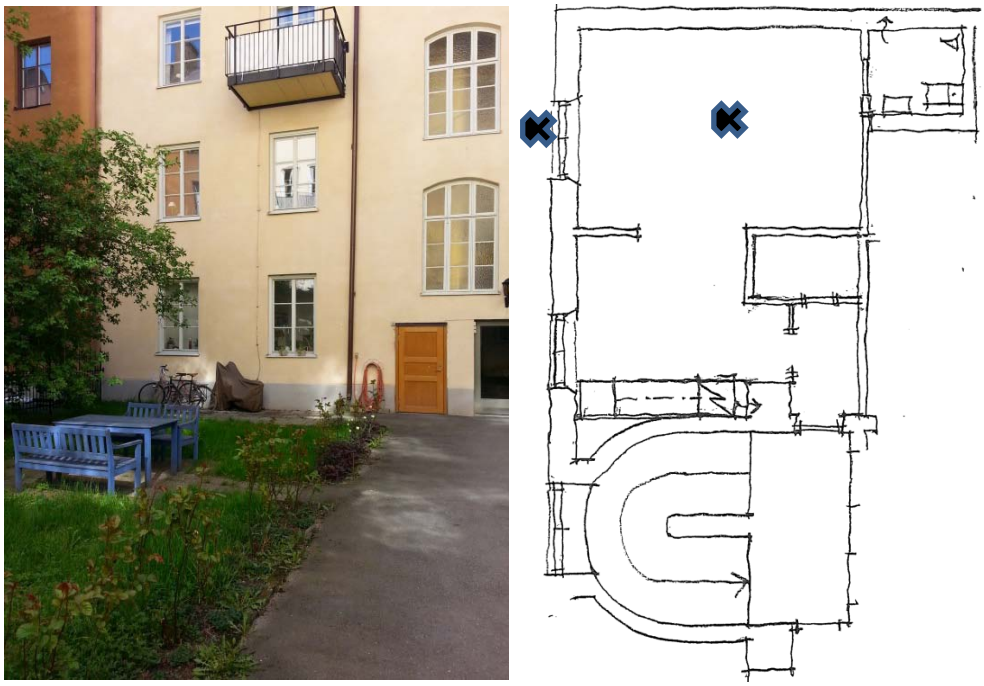

Figure 2 a, Grevgatan courtyard view (Ekebergh, 2014) b, drawing of the apartment on Grevgatan with the measurement locations marked (Frondell, 2014)

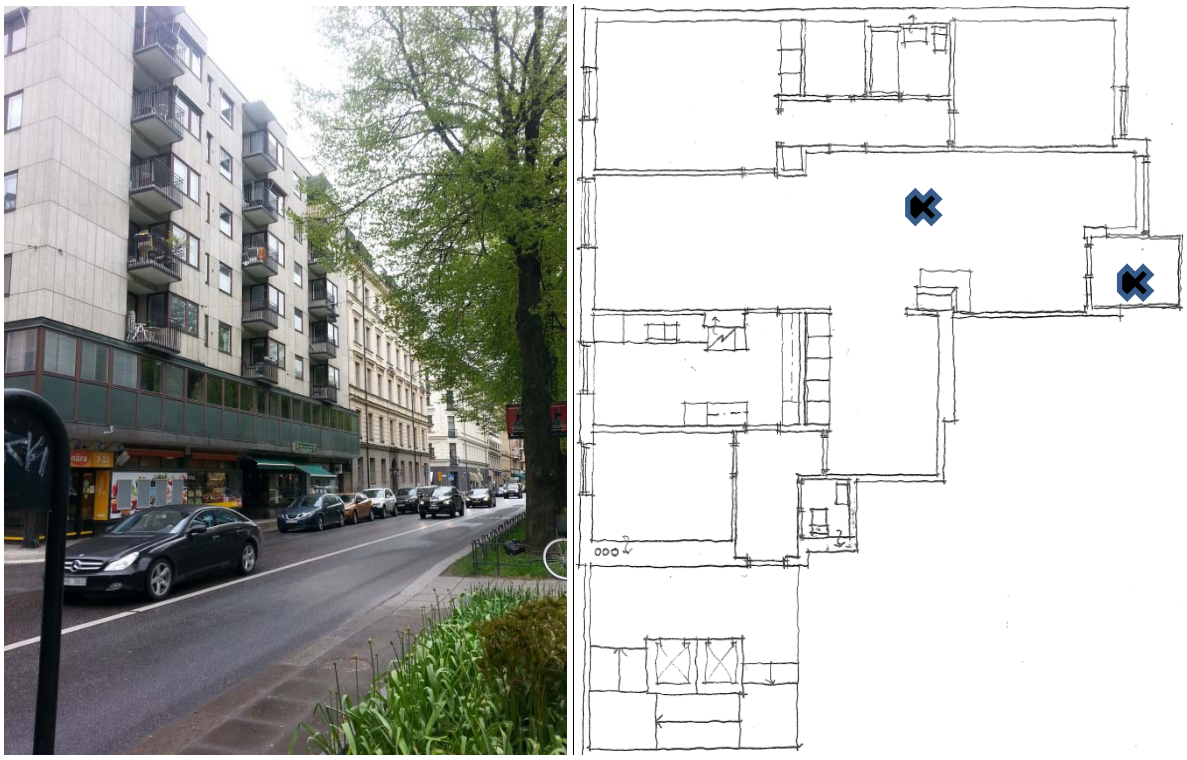

Figure 3 a, Karlavägen street view (Ekebergh, 2014) b, drawing of the apartment on Karlavägen with the measurement locations marked (Frondell, 2014)

Study design

The time-resolved indoor and outdoor concentrations of BC ($C_{BC,in}$ and $C_{BC,out}$) were measured in the living room of each apartment as well as right outside the windows of those living rooms (cross marked in the drawings in Figure 1 b, 2 b, and 3 b.

For a better understanding of the behavior of the measured BC, monitoring of size distributed particles in the size range of 0,3-10,0 µm were measured in parallel at several occasions when measuring the C_{in} and C_{out} of BC at Artillerigatan and Karlavägen. Also, the parameters a, k and P were estimated for size distributed particles in all three apartments. k and P are size dependent (Thatcher & Layton, 1995). The intent with size distributed measurements was to facilitate the assessment of indoor BC.

Since the air exchange rates varies with meteorological conditions like wind, temperature and air pressure the data of the parameters temperature, air pressure, relative humidity, wind speed and wind direction for the time periods of the measurements were obtained from measurements in central Stockholm and used to assess any erratic appearance in the C_{out} data series as well as to assess if there was any detectable correlation between meteorological conditions and the I/O ratios of the particles measured.

Measurement instruments and procedure

Black Carbon

BC concentrations were measured using 4 portable Aethalometers (Model AE51, Magee Scientific) with a measurement resolution of 0,1 * 10^{-2} µg BC m^{-3} and a measurement precision of ± 0,1 µg m^{-3} with the average time base of one minute and a flow rate of 150 ml min^{-1}.

For all measurements at apartments Artillerigatan and Grevgatan and for the indoor measurements at Karlavägen, the aethalometers were supplied with electricity through appurtenant

USB to AC-USB wall adapters connected to a microAeth® USB Port. For the outdoor measurements on the balcony at Karlavägen, the internal rechargeable lithium-ion battery with a total run time of minimum 24 hours (with 5 minute averages at a flow rate of 100 ml h^{-1}) was utilized. The two aethalometers used at Karlavägen were interchanged between each measurement allowing the batteries to recharge while measuring indoors. For the most part $C_{BC,in}$ and $C_{BC,out}$ were measured with 5 minute averages at a flow rate of 100 ml h^{-1}. The decay of C_{BC} in the air exchange rate- and particle deposition experiments was measured with 30 seconds averages at a flow rate of 150 ml min^{-1}. The filters were exchanged when it was assumed to be needed based on instructions in the operating manual from the AethLabs website. The data produced was transferred to a Windows®-based PC with the appropriate microAethCOM PC software installed (AethLabs, 2014).

Figure 4, Aethalometer measuring $C_{BC,out}$ at Artillerigatan with red ring marking the inlet of the hose sticking out through the potrhole.

For the time resolved monitoring of $C_{BC,in}$ and $C_{BC,out}$, two aethalometers were used pairwise for each apartment. At the apartment on Artillerigatan, $C_{BC,out}$ was measured with an aethalometer placed indoors in the window, with an attached sampling hose that was sticking out through a porthole beneath the window, which thus was not entirely closed but in a mode reflecting normal conditions. The inlet of the hose was sheltered from precipitation and placed with a distance of approximately 0,2 m to the building wall.

At the apartment on Grevgatan, $C_{BC,out}$ was measured in the same manner as on Artillerigatan, except the porthole here was placed above the window and not available, so the sampling hose had to stick out through the open window which then was closed provisionally as tight as possible using foam rubber, plastic and sticky tape.

Figure 5, Hoses for outdoor monitoring sticking out through the provisionally closed window at Grevgatan.

The apartment on Karlavägen had a balcony on which the aethalometer measuring $C_{BC,out}$ was placed in a protecting plastic box with a hole where the sampling hose was sticking out.

In all three apartments, $C_{BC,in}$ was measured with an aethalometer placed indoors in the middle of the living room at a height representing the breathing zone of the residents while sitting down on the living room couch. All indoor measurements were also done with a sampling hose attached to the aethalometer for consistency.

Inter-comparison measurements were carried out before initiating a measurement with new filters by placing the aethalometers in parallel and letting them measure the same air under equal conditions for a period of time assumed to yield a sufficient amount of data points. For the most part, this was also done before exchanging the used filters. The attempt was to give a fairly even coverage of the order of concentrations being measured in the monitoring measurements. In the startup of a measurement, there is some amount of time required for the instrument to be warmed up and stabilized and therefore, not all data produced in the control measurements were used to represent the consistency between the two aethalometers, but rather a sufficient amount of data points to conclude weather the intercept is significantly different from zero and the slope is significantly different from 1 (at a 95% confidence level) for the linear regressions produced by the temporally paired data points plotted against each other. If the

measurement differences were significant, the functions of these linear regressions were used to convert the subsequent $C_{BC,in}$ and $C_{BC,out}$ data series, measured with the associated filters.

Size distributed particles

Indoor and outdoor $C_{0,3}$, $C_{0,5}$, $C_{1,0}$, $C_{2,5}$, $C_{5,0}$ and $C_{10,0}$ were simultaneously measured with a minute time resolution using an airborne particle counter designed and built by Lighthouse (model HANDHELD 3016 IAQ) having these particle diameters as standard channel sizes. The mass concentration reported by the instrument assumes an approximate particle density of 2,500 g ml^{-1} which is equal to 2,5· 10^3 g dm^{-3} and 2,5· 10^{12} µg m^{-3} (Lighthouse Worldwide Solutions, 2006-2014).

Size distributed C_{in} and C_{out} were measured alongside the measurements of BC at several occasions in the apartments at Artillerigatan and Grevgatan. The particle counter measuring C_{out} (L2) were installed in the same manner as the aethalometer measuring $C_{BC,out}$, in the window with a hose attached to its inlet sticking out assembled to the aethalometer hose. The particle counter measuring C_{in} (L1) was placed in order to measure the same air as the aethalometer measuring $C_{BC,in}$.

At five occasions during the measurement period, inter-comparison measurements were carried out with the instruments L1 and L2, by placing them in parallel and letting them measure the same air under equal conditions for a period of time assumed to yield a sufficient amount of data points. A regression function was received for each inter-comparison measurement relating the data points measured by L2 to the temporarily paired data points measured by L1. If the slope and/or the intercept were significantly different from 1 and/or 0, respectively, the function received was used to convert the data recorded by L1. The measurement data were downloaded using the lighthouse data transfer software LMS XChange v1.6 (Lighthouse Worldwide Solutions, 2006-2014).

CO_2

In the air exchange rate- and deposition rate analysis, the real-time concentrations of CO_2 were measured every tenth second using an indoor air quality monitor (model TSI 8551 Q-TRAK™ Monitor). The instrument had a measurement size range of 0-5000 ppm CO_2 with the resolution of 1 ppm and an accuracy of ±50 ppm at 25°C. The measurement data were downloaded to a computer and operated using the TRAKPRO™ Data Analysis Software (TSI incorporated, 2006).

Experimentally estimated parameters

Air exchange and particle deposition rate

The three apartments were ventilated by a combination of natural ventilation and infiltration by leakage flow. For simplification, the ventilation and deposition mechanisms were assumed to be constant in each apartment, giving constant apartment specific values of a and k that were applied to all calculations in the data analysis independently of the time of day that the data was recorded. Furthermore, the internal air was assumed to be uniform in terms of temperature and pressure and well mixed in terms of particle concentrations.

The air exchange and deposition rates (a and k) were measured at the same sites that the measurements of C_{in} took place by measuring of the decay in soot particles and carbon dioxide (CO_2). A sufficient amount of candles were lit to increase the concentrations of soot particles and CO_2 above background levels as a fan mixed the indoor air. CO_2 was used as a tracer gas since it was not expected to be subject for deposition but only be decreased by the influence of the air exchange rate. When the concentrations were considered being high enough, the candles were extinguished and then the decrease in particle- and CO_2-concentration were monitored against time. The apartment was left unoccupied for at least 1 hour immediately after extinguishing the candles with

the intention to remove all indoor sources for a more accurate definition of the ventilation- and deposition mechanisms.

The ventilation, a (h^{-1}), can be calculated from the decrease in CO_2 by the equation $a = (1/(t-t_0)*\ln(C/C_0)$ (Vette, et al., 2001) where C is the concentration at the end of the sampling interval, t (in hours), and C_0 is the concentration at the beginning, at t_0. By plotting the $\ln(C/C_0)$ for all t measured a linear regression is generated from which the slope is equal to the air exchange rate. This was done for a few CO_2 decay experiments in each apartment and then apartment specific mean values of a were estimated.

Decay rates were defined for the BC and size distributed particles generated from the candles in the same way as for the CO_2. When particle concentrations are let to decline from levels that are significantly elevated above background levels, the change in concentration over time, dC_{in}/dt, is equal to the summed air exchange and deposition rate, $-(a+k)*C_{in}$, If $\ln(C/C_0)$ for BC and the size distributed particles are plotted against time, thus, the slope of that linear regression would equal the total decay rate of a and k. In order to estimate the specific k values the apartment associated a value was subtracted from the total decay rate for each particle species (Vette, et al., 2001, Chao, et al., 2003).

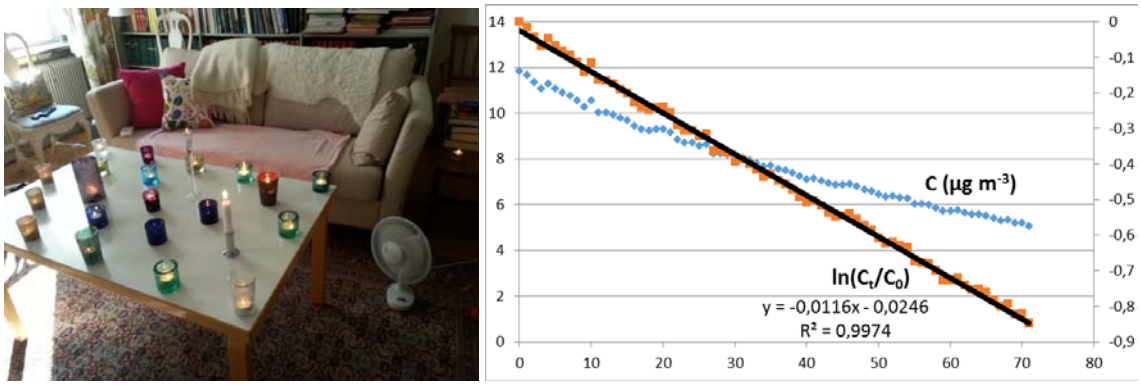

Figure 6 a, air exchange- and deposition rate experiment using candles and a fan for air mixing. B, decay in particle concentration (µg m^{-3}) and $\ln(C_t/C_0)$ plotted against time (min).

Indoor sources

A logbook was kept over indoor activities during the measurements of C_{in} and C_{out} in order to keep track of indoor sources. Cooking activities taking place during the measurement periods included frying or oven-baking with an electrical stove, heating in a microwave oven and using a waffle iron. The types of food prepared were not noted. Aerosol particles generated from cooking activities have been reported to have a life-time in the kitchen varying between 4-6 h (Hussein, et al., 2006) whereof cooking activities in this study were considered influencing the indoor particle concentrations for a time period of 6 h after the cooking had terminated.

Indoor/Outdoor ratio and infiltration factor

C_{in} and C_{out} data that was recorded in the absence of indoor sources was put together as a certain data set. This data was used to calculate the I/O ratio and F_{inf} for each apartment for BC and for all particle sizes measured. The I/O ratios were calculated as average values of the C_{in}/C_{out}. Values of F_{inf} were estimated from the linear approach in which the C_{in} equals the $F_{inf} * C_{out} + C_s$ by plotting all the C_{in} data against the temporarily paired C_{out} data and estimate F_{inf} as the slope of the regression (Chen & Zhao, 2011).

Penetration factor

Apartment specific penetration factors were calculated from the estimated values of a, k and F_{inf} for BC and all particle sizes using the equation $F_{inf} = (a*P)/(a+k)$ found in (Bennett & Koutrakis, 2006).

Correlation with meteorological conditions

Correlation factors for the outdoor BC concentration and the infiltration factor (C_{in}/C_{out}), respectively, were calculated for the meteorological parameters temperature, air pressure, relative humidity, wind speed, at the three apartments. The meteorological parameters were obtained from measurements in central Stockholm. The data used for calculating the correlation of C_{in}/C_{out} with different parameters were the one adjusted for indoor sources. The correlation factors calculated are average values of correlation factors calculated for each measurement series separately for easier detection of how the change in e.g. the relative humidity affect the $C_{BC,out}$.

Data analysis

All calculations and data analysis were made using the Microsoft office program Microsoft Excel 2010®. The $C_{BC,in}$ and $C_{BC,out}$ data along with the data received from the inter-comparison measurements were processed prior to analysis applying the Local polynomial regression algorithm found on the aethlabs.com website with 5 smoothing number of points.

After converting the data, all concentrations were recalculated to 15 minute average values in order to reduce sudden changes in the time series that is difficult to interpret.

Results

Time-resolved BC data

Summary statistics of C_{in} and C_{out} are presented in Table 2. In order to compensate for the differing distribution of time of day that the monitoring went on in the different apartments, mean values for every fourth of an hour were calculated for C_{in} and C_{out} throughout the whole day, from which the concentration mean, median and percentiles were calculated. The minimum and maximum values reported in Table 2 are derived from the entire dataset of 15 min means of the real-time measurement data points.

Table 2, Results of the measured C_{in} and C_{out} at the apartments at Artillerigatan, Grevgatan and Karlavägen.

Artillerigatan	Cin	Cout	Grevgatan	Cin	Cout	Karlavägen	Cin	Cout
n (data points)	96	96	**n (data points)**	96	96	**n (data points)**	96	96
Mean	0,59	0,77	**Mean**	0,75	0,77	**Mean**	0,46	0,61
Median	0,61	0,78	**Median**	0,68	0,76	**Median**	0,40	0,59
St. deviation	0,16	0,21	**St. deviation**	0,35	0,22	**St. deviation**	0,17	0,15
C.I.	0,03	0,04	**C.I.**	0,07	0,04	**C.I.**	0,03	0,03
5th percentile	0,36	0,47	**5th percentile**	0,38	0,41	**5th percentile**	0,33	0,42
95th percentile	0,83	1,05	**95th percentile**	1,54	1,11	**95th percentile**	0,78	0,91
Minimum	0,04	0,02	**Minimum**	0,11	0,07	**Minimum**	0,05	0,06
Maximum	4,37	3,61	**Maximum**	11,03	5,70	**Maximum**	5,35	2,48

To put the measured $C_{BC,out}$ in context, they were compared with BC-data obtained from one urban background site, Torkel Knutssonsgatan, and one street level site with known high concentrations of BC, Hornsgatan, in central Stockholm. The data were obtained separately for Artillerigatan, Grevgatan and Karlavägen at the respective time periods that the measurement had proceeded in each apartment. The summary statistics derived from the entire dataset of 15 min means of the real-time measurement data points are presented in Table 3, 4, and 5.

Table 3, summary statistics of fourth of an hour means of $C_{BC,out}$ measured at Artillerigatan and the corresponding $C_{BC,out}$ at Hornsgatan and Torkel

	Hornsgatan (street level)	Torkel Knutsongatan (urban background)	Artillerigatan (measured)
Mean	1,64	0,80	0,77
Median	1,65	0,85	0,78
Minimum	0,56	0,50	0,44
Maximum	4,63	1,09	1,13
percentile			
5th	0,65	0,55	0,47
95th	3,04	1,02	1,05

Table 4, summary statistics of fourth of an hour means $C_{BC,out}$ measured at Grevgatan and the corresponding $C_{BC,out}$ at Hornsgatan and Torkel Knutssonsgatan.

	Hornsgatan (street level)	Torkel Knutsongatan (urban background)	Grevgatan (measured)
Mean	1,33	0,71	0,77
Median	1,32	0,71	0,76
Minimum	0,50	0,48	0,39
Maximum	2,84	1,04	1,40
percentile			
5th	0,59	0,52	0,40
95th	1,97	0,88	1,10

Table 5, summary statistics of fourth of an hour means $C_{BC,out}$ measured at Karlavägen and the corresponding $C_{BC,out}$ at Hornsgatan and Torkel Knutssonsgatan.

	Hornsgatan (street level)	Torkel Knutsongatan (urban background)	Karlavägen (measured)
Mean	1,32	0,40	0,61
Median	1,42	0,37	0,59
Minimum	0,51	0,23	0,36
Maximum	2,97	0,63	1,04
percentile			
5th	0,54	0,28	0,42
95th	2,14	0,57	0,91

Mean values were calculated on a 15 minute basis for the whole day in order to compare the diurnal $C_{BC,out}$ characteristics between the measured data and the one obtained from Hornsgatan and Torkel Knutssonsgatan. In Figure 7 these mean values are plotted against the time of day.

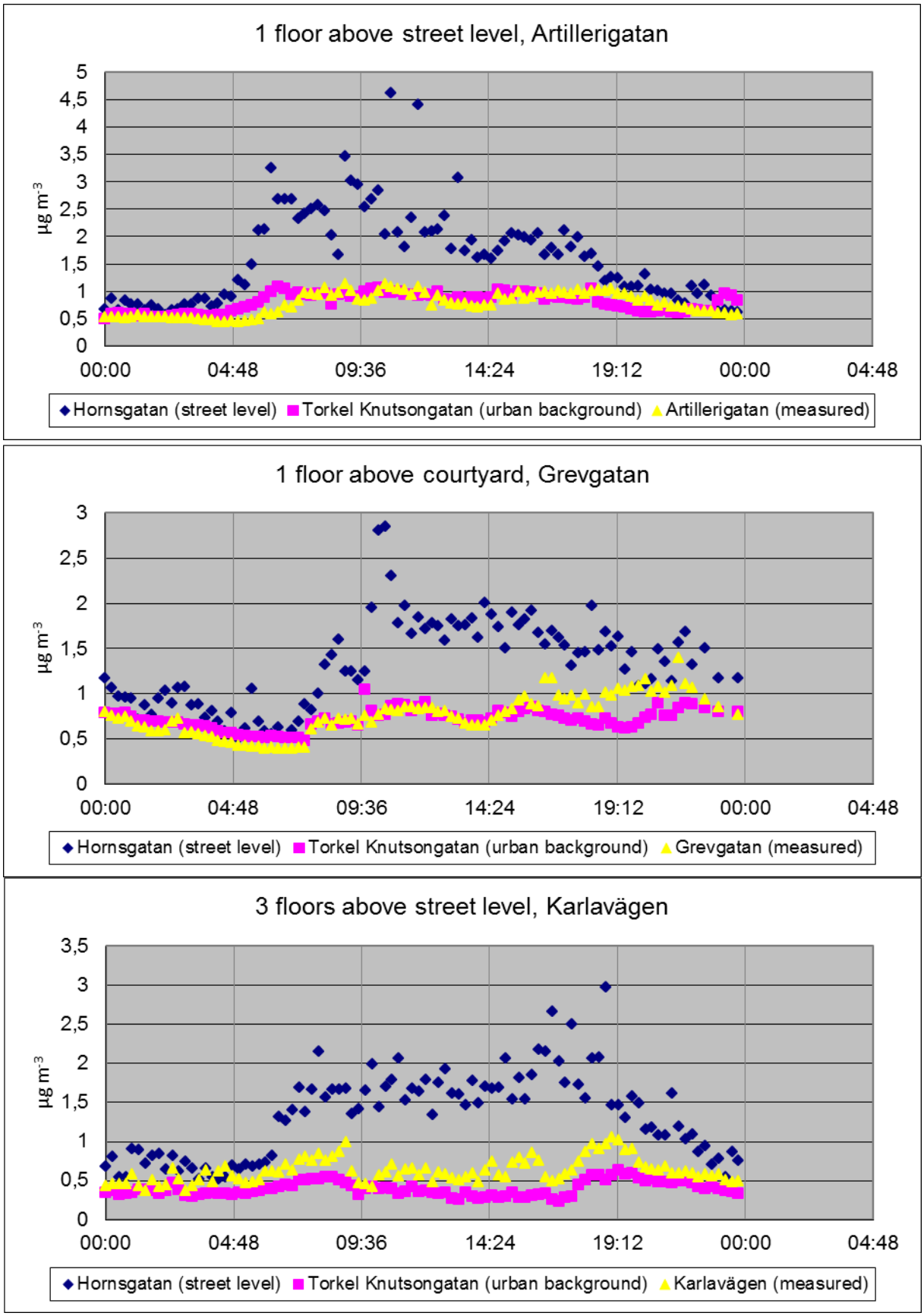

Figure 7, comparison of the measured $C_{BC,out}$-data at Artillerigatan, Grevgatan and Karlavägen, with C_{BC} measured at Hornsgatan and Torkel Knutssonsgatan in central Stockholm.

Meteorological data

Table 6, meteorological conditions during the specific measurement periods at Artillerigatan, Grevgatan and Karlavägen.

	Artillerigatan	Grevgatan	Karlavägen
	mean (± st. deviation)	mean (± st. deviation)	mean (± st. deviation)
temperature (°C)	1,80 (1,91)	2,33 (0,69)	4,55 (3,44)
air pressure (hPa)	1007,41 (9,92)	993,66 (5,48)	1012,25 (12,05)
relative humidity (%)	89,51 (13,31)	97,69 (3,47)	66,21 (20,40)
wind speed (m s^{-1})	3,66 (1,66)	4,50 (1,74)	4,39 (2,20)

Correlation of $C_{BC,in}/C_{BC,out}$ with meteorological parameters

Table 7, correlation coefficients (c.c.) between $C_{BC,in}/C_{BC,out}$ and meteorological data from Central Stockholm.

c.c. between $C_{BC,in}/C_{BC,out}$ and:	mean C.C. (± 95% C.I.) Artillerigatan	mean correlation factor ((± 95% C.I.) Grevgatan	mean correlation factor ((± 95% C.I.) Karlavägen
temperature (°C)	0,04 (0,22)	0,03 (0,69)	0,04 (0,278)
air pressure (hPa)	0,02 (0,23)	-0,31 (0,72)	-0,15 (0,27)
relative humidity (%)	-0,17 (0,14)	-0,04 (0,64)	-0,07 (0,26)
wind speed (m s^{-1})	0,21 (0,14)	0,03 (0,56)	0,16 (0,17)

No correlation could be detected between the $C_{BC,in}/C_{BC,out}$ ratios calculated for the apartments at Artillerigatan, Grevgatan and Karlavägen

Experimentally estimated parameters

The estimated values of a and (a + k) are presented in Table 8, along with the calculated k values for Artillerigatan, Grevgatan and Karlavägen, respectively.

Table 8, experimentally estimated values of a and k

Particle specie	particle decay rate (a+k) (h^{-1})	Air exchange rate (h^{-1})			Deposition rate (h^{-1})		
Artillerigatan		a	C.I.	st. deviation	k	C.I. *	st. deviation
BC	0,92	0,35	0,10	0,10	0,56	0,97*	0,11
0,3 µm	0,40	0,35	0,10	0,10	0,05	0,22*	0,02
0,5 µm	0,61	0,35	0,10	0,10	0,26	0,28*	0,03
1,0 µm	0,74	0,35	0,10	0,10	0,39	0,38*	0,04
2,5 µm	1,04	0,35	0,10	0,10	0,69	0,21*	0,02
5,0 µm	2,46	0,35	0,10	0,10	2,11	1,33*	0,15
10,0 µm	3,50	0,35	0,10	0,10	3,15	3,92*	0,44
Grevgatan							
BC	0,58	0,24	0,14	0,06	0,34	0,03	0,01
0,3 µm	0,27	0,24	0,14	0,06	0,03	0,22	0,09
0,5 µm	0,39	0,24	0,14	0,06	0,15	0,12	0,05
1,0 µm	0,58	0,24	0,14	0,06	0,34	0,07	0,03
2,5 µm	0,92	0,24	0,14	0,06	0,68	0,20	0,08
5,0 µm	2,75	0,24	0,14	0,06	2,51	0,56	0,23

10,0 µm	3,72	0,24	0,14	0,06	3,48	0,80	0,32
Karlavägen							
BC	0,56	0,34	0,04	0,02	0,23	0,12	0,07
0,3 µm	0,73	0,34	0,04	0,02	0,40	0,20	0,18
0,5 µm	0,90	0,34	0,04	0,02	0,57	0,25	0,16
1,0 µm	1,13	0,34	0,04	0,02	0,80	0,39	0,25
2,5 µm	1,38	0,34	0,04	0,02	1,04	0,58	0,37
5,0 µm	2,09	0,34	0,04	0,02	1,75	1,12	0,71
10,0 µm	3,79	0,34	0,04	0,02	3,45	1,96	1,23

* The deposition rates on Artillerigatan are only based on two measurements, why the confidence intervals for these values are not really representative.

Air exchange rates

In Figure 8, the calculated mean values of the air exchange rates for apartments Artillerigatan, Grevgatan and Karlavägen are presented in h^{-1} together with a 95 % confidence interval.

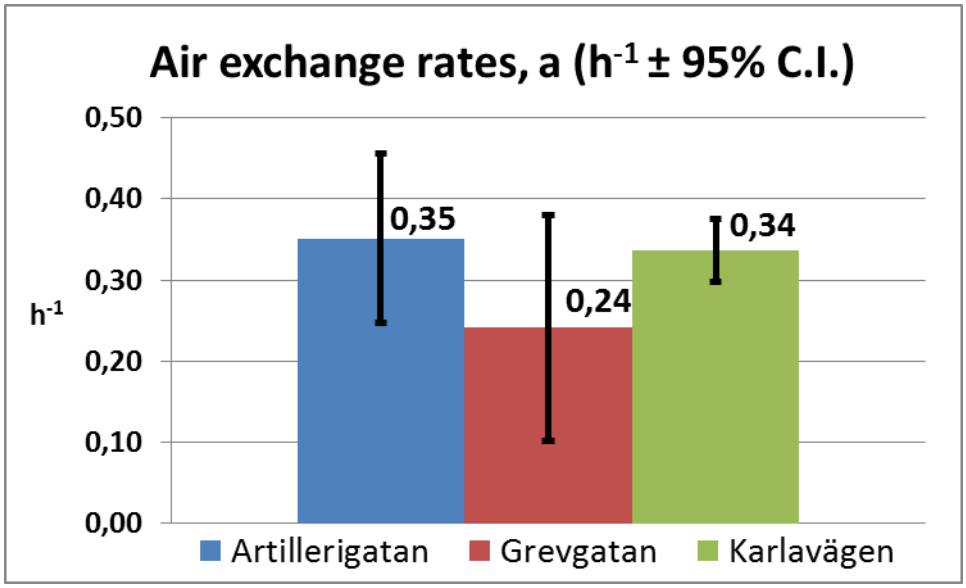

Figure 8, Air exchange rates for the apartments at Artillerigatan, Grevgatan and Karlavägen

A Kruskal Wallis test was performed to see if the air exchange rate mean values were significantly different from one another in the different apartments (Townend, 2002). The critical value of H was determined to 5,61 for a significance level of 95% and 7,5 for a significance level of 99%. The calculated value of H became 3,10, why it was not rejected that the air exchange rates from the different apartments all had the same mean value.

Particle deposition rates

The deposition rates in h^{-1} for BC and the particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen are presented.

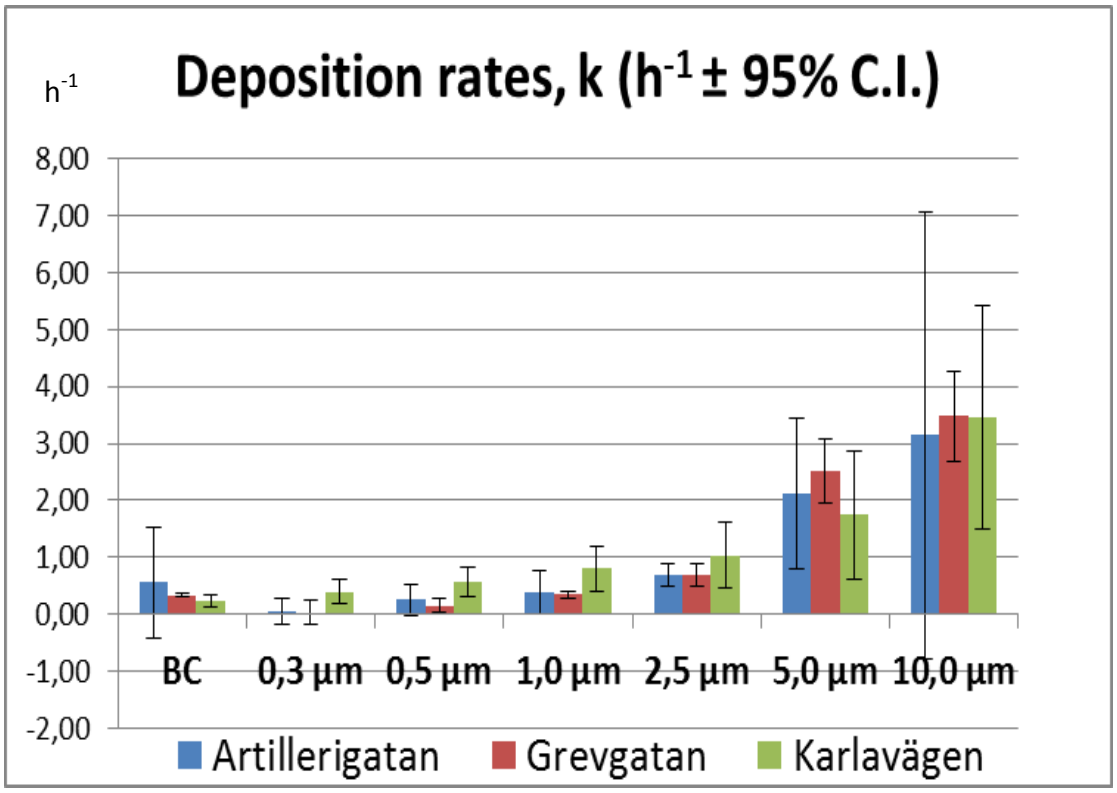

Figure 9, deposition rates for BC and the different particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen.

In Figure 10, the deposition rates, in h^{-1}, converted to deposition velocities, in cm s^{-1}, using the estimated area to volume ratio of the apartments are presented.

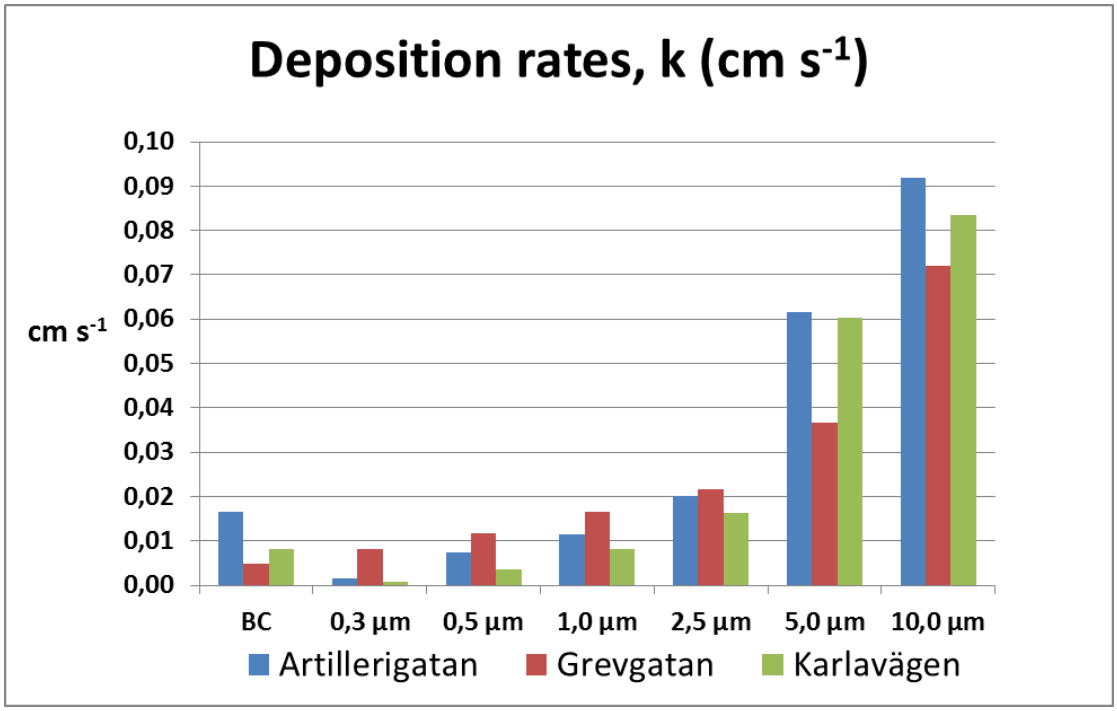

Figure 10, deposition rates (cm s^{-1}) for BC and the different particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen.

I/O ratio

The I/O ratios are presented in Table 9, for BC and all particle sizes measured in the apartments. Indoor sources were considered negligible in the data used for the calculations of the I/O ratios.

Table 9, I/O ratiosfor BC and the particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen.

I/O ratio	Artillerigatan mean C_{in}/C_{out} (± s.d.)	Grevgatan mean C_{in}/C_{out} (± s.d.)	Karlavägen mean C_{in}/C_{out} (± s.d.)
BC	0,81 (0,46)	0,83 (0,35)	0,78 (0,57)
0,3 µm	0,69 (0,15)	0,66 (0,22)	
0,5 µm	0,40 (0,08)	0,75 (0,39)	
1,0 µm	0,63 (0,27)	1,50 (1,68)	
2,5 µm	0,81 (0,65)	1,68 (2,22)	
5,0 µm	1,00 (2,34)	1,71 (3,54)	
10 µm	24,21 (77,01)	18,19 (106,14)	

In Figure 11, the I/O ratios for BC and particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen are presented with the I/O ratios for 10 µm in a separate graph due to its much higher values compared with the other particle sizes.

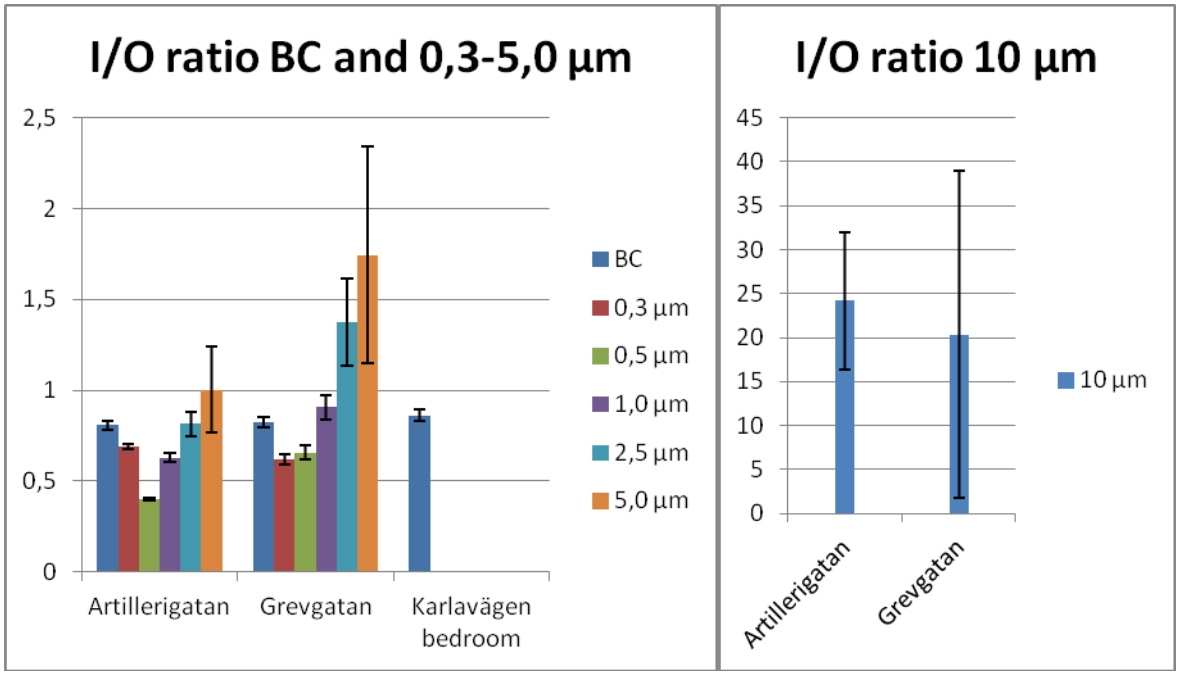

Figure 11, I/O ratios for BC and the particle sizes measured in the apartments Artillerigatan, Grevgatan and Karlavägen

The I/O ratios were also calculated for night data (recorded between 2 and 6 a.m.) to see if the results would be different. The night values of the I/O ratios are presented in Table 10.

Table 10, I/O ratios for BC and the particle sizes measured in the apartments Artillerigatan, Grevgatan and Karlavägen at night. n.d. means no data and refers to the shortage of data available for calculating that I/O ratio.

I/O ratio at night (2-6 a.m.)	Artillerigatan mean C_{in}/C_{out} (± s.d.)	Grevgatan mean C_{in}/C_{out} (± s.d.)	Karlavägen mean C_{in}/C_{out} (± s.d.)
BC	0,73 (0,24)	1,02 (0,16)	0,76 (0,32)
0,3 µm	0,79 (0,15)	0,96 (0,27)	
0,5 µm	0,41 (0,08)	1,02 (0,35)	
1,0 µm	0,45 (0,11)	1,23 (0,72)	
2,5 µm	0,29 (0,16)	2,51 (2,48)	
5,0 µm	n.d.	5,31 (6,92)	
10 µm	n.d.	28,91 (18,09)	

Infiltration factors

The estimated F_{inf} factors, defined as the slope of the C_{in}/C_{out} linear regression functions, for BC and the particle sizes measured in the apartments are presented in Table 11**.** The individual graphs that are associated with the calculated F_{inf} factors for BC and size distributed particles are presented in appendix a.

Table 11, Infiltration factors

	Artillerigatan		Grevgatan		Karlavägen	
	F_{inf} (± 95% C.I.)	R^2	F_{inf} (± 95% C.I.)	R^2	F_{inf} (± 95% C.I.)	R^2
BC	0,55 (0,02)	0,73	0,64 (0,02)	0,83	0,41 (0,05)	0,39
0,3 µm	0,70 (0,08)	0,46	0,46 (0,03)	0,80		
0,5 µm	0,33 (0,02)	0,81	0,39 (0,02)	0,87		
1 µm	0,16 (0,05)	0,12	0,43 (0,05)	0,60		
2,5 µm	0,54 (0,13)	0,16	0,53 (0,29)	0,15		
5 µm	0,10 (0,13)	0,01	0,50 (0,40)	0,05		
10 µm	-2,25 (1,05)	0,08	6,21 (18,05)	0,02		

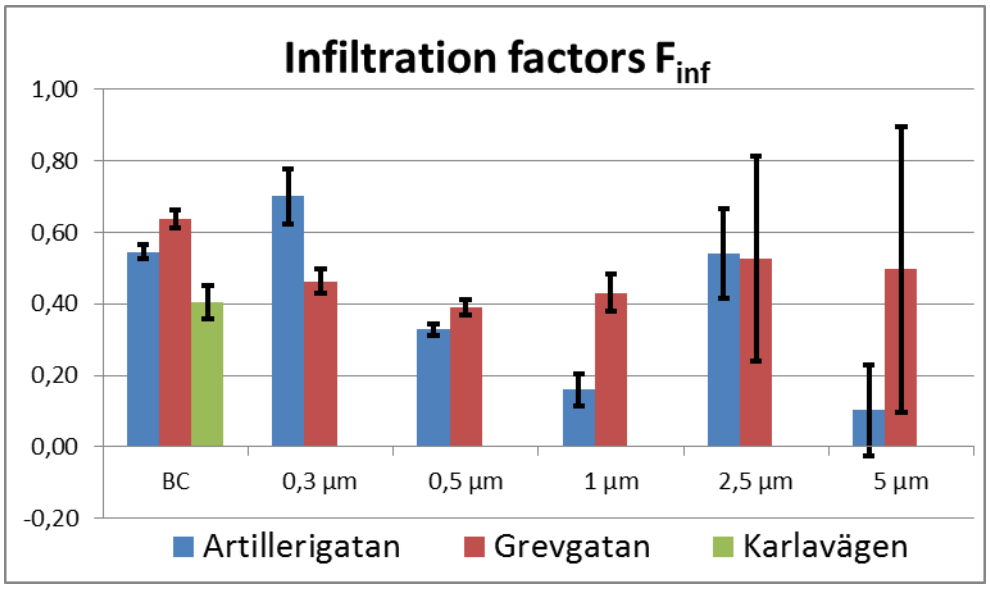

Figure 12, Infiltration factors for BC and the particles measured in apartments Artillerigatan, Grevgatan and Karlavägen

The F_{inf} factors calculated from data only recorded at night are presented in Table 12 and Figure 13.

Table 12, Infiltration factors calculated from night data

	Artillerigatan		Grevgatan		Karlavägen	
	F_{inf} (± 95% C.I.)	R^2	F_{inf} (± 95% C.I.)	R^2	F_{inf} (± 95% C.I.)	R^2
BC	0,72 (0,05)	0,80	0,81 (0,16)	0,69	0,55 (0,06)	0,75
0,3 µm	1,09 (0,21)	0,61	0,17 (0,05)	0,80		
0,5 µm	0,32 (0,05)	0,73	0,17 (0,05)	0,80		
1 µm	0,07 (0,04)	0,16	-0,07 (0,15)	0,06		
2,5 µm	-0,03 (0,02)	0,10	-0,51 (0,49)	0,25		
5 µm	-0,03 (0,02)	0,17	-1,79 (1,68)	0,26		
10 µm	-0,30 (0,51)	0,02	-0,44 (11,83)	0,00		

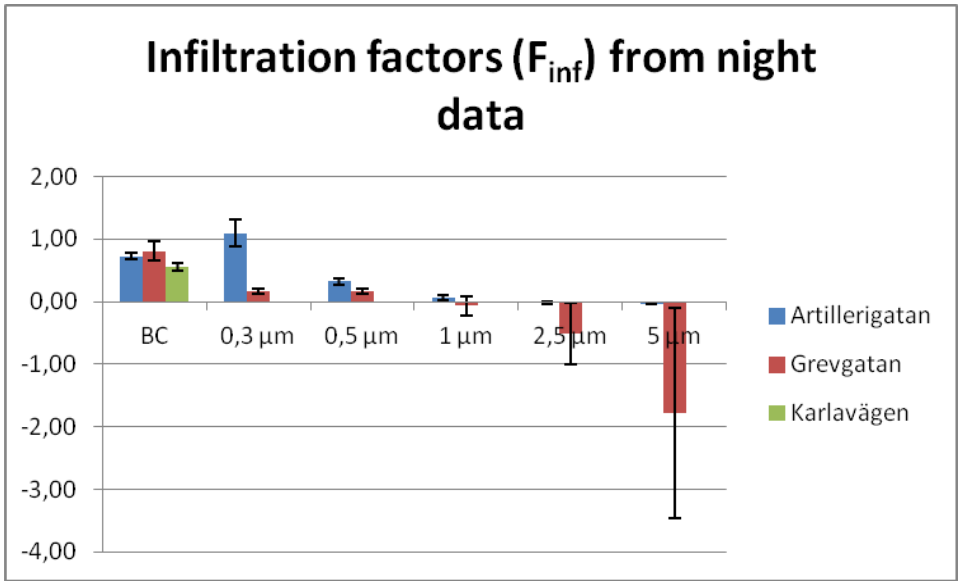

Figure 13, Infiltration factors calculated from night data

Penetration factors

P calculated from the estimated F_{inf} for BC and the particle sizes measured in the apartment at Artillerigatan, Grevgatan and Karlavägen are presented in Figure 14.

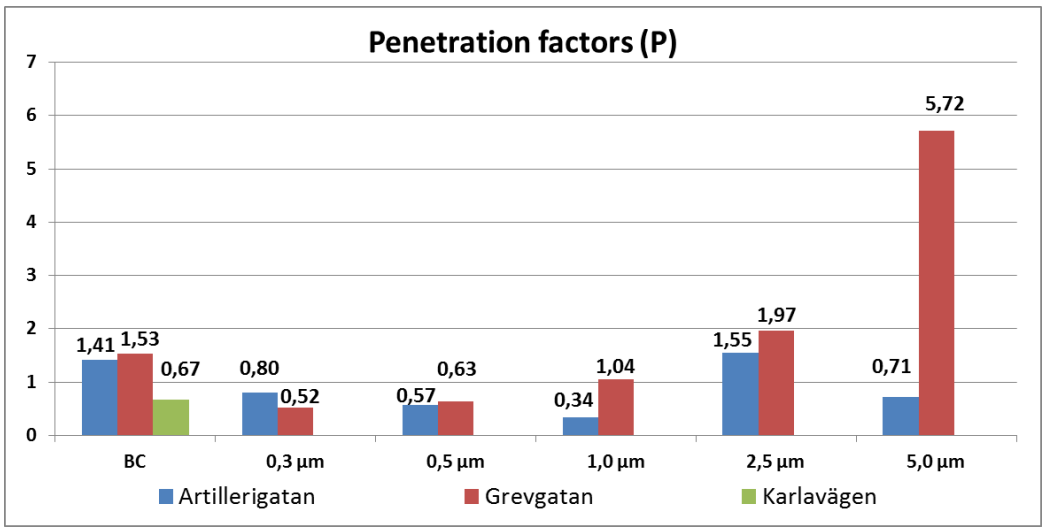

Figure 14, calculated penetration factors for BC and particle sizes measured in the apartments at Artillerigatan, Grevgatan and Karlavägen.

Discussion

In this study, the apartments were all treated as single wellmixed zones with the measured indoor concentration thought to represent the entire apartment. This enables the use of uncomplicated mathematical procedures for the data processing and the measurement data can easily be compared with corresponding data from another apartment. The simplicity of the study design makes it applicable in occupied "everyday life" apartments. The downside to using this method is the shortage of detail in the resulting data. In real life the dynamic behaviour of aerosol particles indoors is described with greater validity using a multiple-compartment size resolved model accounting for spatial differences in the particle concentrations of each particle size (Hussein & Kulmala, 2007). Nevertheless, since the analysis of the conducted measurement data are based on the temporal variations in concentrations of BC and size distributed particles, the location for the measurement is considered representative in the sense of fluctuations of indoor concentrations in comparison with simultaneous outdoor concentrations.

Black Carbon

The C_{in} values of 0,59 (± 0,16), 0,75 (± 0,35), and 0,46 (± 0,17) µg m^{-3} at Artillerigatan, Grevgatan and Karlavägen, respectively, are in the expected magnitude. In the literature, C_{in} measured in the presence of indoor sources have been reported to be 0,462 (±3,197) (LaRosa, et al., 2002), and 1,57 ± 1,0 µg m^{-3} reported by (Lunden, et al., 2008). The C_{out} values of 0,77 (± 0,21), 0,77 (± 0,22), and 0,61 (± 0,15) µg m^{-3} at Artillerigatan, Grevgatan and Karlavägen, respectively, are higher than the corresponding C_{in} values which is expected in an urban environment that Stockholm is. According to the summary statistics, presented in Table 2, where indoor sources have not been neglected, the C_{in} values are close to C_{out} values due to a few, relatively high concentrations recorded during cooking activities. The peak C_{in}, presented in appendix a, at Artillerigatan of 4,37 µg m^{-3} is attributable to the use of a waffle iron and the peak C_{in} of 11,03 µg m^{-3} and 5,35 µg m^{-3} at Grevgatan and Karlavägen, respectively, are both attributable to the use of a frying pan.

The highest $C_{BC,in}$ mean and 95th percentile value is measured at Grevgatan. The increase in particle concentration caused by cooking activities is decreased with the amount of space between the particle source and the measurement location (Hussein, et al., 2006), why this is reasonable since cooking activities take place in the same room as the measurements in the apartment at Grevgatan, compared with Artillerigatan and Karlavägen, where all cooking activities take place in a separate kitchen.

The highest $C_{BC,out}$ mean and $maximum_{BC,out}$ value is measured at Grevgatan even though the outdoor concentrations there were measured facing a courtyard. All the measurements done at Grevgatan were performed in February while in the other apartments; the monitoring proceeded until April 1st. This could be the explanation due to more residential wood burning associated with lower temperatures (Table 6) in Stockholm, which by this time of the measurement period could also be smelled in the outdoor air. The BC generated by wood burning can be seen in the time-resolved graphs in Figure 7, where the BC concentrations don't decrease after 7 p.m. like they do for the other apartments. A similar evening peak for wintertime data thought to be due to wood burning was found by (LaRosa, et al., 2002).

Interpreting the measured outdoor concentrations with the obtained ones from Hornsgatan and Torkel Knutssonsgatan, presented in Figure 7, it can be seen that the BC levels at Hornsgatan is approximately twice as high during the daytime than on Artillerigatan, Grevgatan and Karlavägen respectively. According to these data, the magnitude of diurnal mean values of $C_{BC,out}$ measured at

Artillerigatan seems to be in line with the $C_{BC,out}$-data obtained from Torkel Knutssonsgatan which is thought of as the urban background to heavily polluted Hornsgatan. $C_{BC,out}$ is lower at Grevgatan than at both Hornsgatan and Torkel Knutssonsgatan between midnight and morning, possibly reflecting its facing towards a courtyard instead of the street. $C_{BC,out}$ measured at Karlavägen is higher than at Torkel Knutssonsgatan throughout the entire day. Contradictive, the mean value and 95th percentile of the $C_{BC,out}$ is higher for Artillerigatan than for Karlavägen, which explanation then probably has to do with the monitoring taking place at differing time periods on the two streets. This underlines the importance of assessing the actual real-time concentration of the outdoor air that is penetrating the indoor environment, in order to make good estimations of the indoor concentrations.

Meteorological conditions

(LaRosa et. al 2002) performed regression analysis on outdoor concentrations of BC and meteorological parameters including temperature, relative humidity, and wind speed. They found that wind speed was the strongest independent variable and that an increase of one s.d. in wind speed generated a decrease in 0,3 s.d. in the log-normally distributed BC concentration. Outdoor relative humidity was found to be the second strongest variable with one increase in the s.d. increasing the logarithmic BC concentration data with 0,27 s.d. In this study, however, no real correlation can be seen between the fluctuation in BC concentration and meteorological conditions, presented in Figure 7. In urban areas, wind velocity (and direction) can be spatially highly variable (Olsson-Jonsson, 2011), which could be the explanation of the non-correlation since the meteorological data is obtained from another site of Stockholm city.

Experimentally estimated parameters

Air exchange rate

The apartment specific a-values, presented in Figure 8, has values of 0,35 h^{-1}, 0,24 h^{-1} and 0,34 h^{-1} for the apartments at Artillerigatan, Grevgatan and Karlavägen, respectively. These values were not significantly different from each other according to a Kruskal Wallis test and there is an overlap in their 95 % confidence intervals calculated by Microsoft excel®. Despite this the apartment specific a-value was used in all calculations associated with that apartment. The air exchange utilized for the calculations in a study on naturally ventilated residences by (Chao, et al., 2003) was reported to 1,28 ± 0,54 h^{-1}. In a review from 2011 the median air exchange rate was 0,71 h^{-1} based on a study of about 500 residences (Chen & Zhao, 2011). The estimated a-values in this study seem low in comparison with this litterature. However, they are in the same order of magnitude as the deposition rates which is in line with previous findings (Chao, et al., 2003).

Deposition rate

The k-values, presented in Figure 9, for the size distributed particles were in the range of 0,03 h^{-1} for 0,3 µm at Grevgatan, and 3,48 h^{-1} for 10 µm at Grevgatan. This agrees fairly well with the deposition rates in Figure 15 published by (Hussein et. al, 2006).

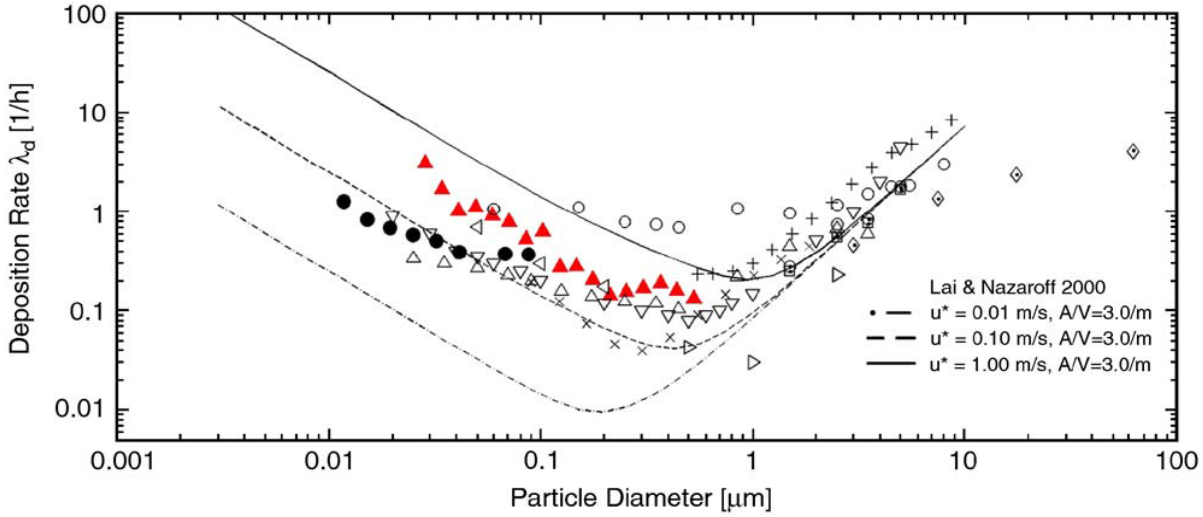

Figure 15, Particle deposition rates (h^{-1}) as a function of particle diameter (µm) copied by (Hussein, et al., 2006).

The minimum k was at 0,3 µm and increased with larger sized particles, in accordance with the size-dependence of k, and within the particle size range of 0,2-0,5 µm reported as the interval for k minimums (Chao, et al., 2003). The higher rates found for larger particle sizes was presumably due to the gravitational settling forces, which are increased with particle mass. In the literature, a deposition rate increase is also to be seen for particle sizes below the k minimum due to diffusion, and thus the minimum k is found for particle sizes that are little affected by diffusion due to being too large as well as gravitational settling due to being too light (Chao, et al., 2003). k-values for a certain particle size varies largely among different studies due to its site specificity. Air exchange rates and the apartment specific area to volume ratio are influencing factors (Nazaroff, 2004) as well as the variety in differing material on the surfaces to which the particles deposit. No effort was put in to assess the real surfaces of the apartments of this study, but the surface areas were calculated roughly by the equation (2*apartment area) + (4*square root of the apartment area*ceiling height)) corresponding to the floor, ceiling and walls. The volume to area ratios were then used to convert the estimated k values with the unit h^{-1} to k values with the unit cm s^{-1} (presented in Figure 10). Judging by the shape of the column heights in relation to particle size for k-values are expressed in both h^{-1} and cm s^{-1}, the size of BC does not seem to agree with any other size above 0,3 µm, why the candle generated BC is assumed to be below 0,3 µm. The k-values for BC at Artillerigatan and Karlavägen are approximately ten times higher than the k-value for 0,3 µm. According to the data in Figure 15 this corresponds to a particle size of 1,0 µm or larger, which is probably unlikely when it comes to traffic-related BC. At Karlavägen, this could be the case as well and the explanation for BC having a lower k than 0,3 µm particles might be due to a minimum at a particle size below 0,3 µm. Since the apartments are structurally differentiated from each other, this is assumedly the explanation for the variation in k being observed among the apartments for the same particle sizes. In a previous study on six residences in Hong Kong, the variation in the calculated k-values was about 50-92% of their calculated mean value, reasoned to be an effect of the differing structure of the residences surfaces in terms of materials and roughness (Chao et. al, 2003).

I/O ratio

The I/O ratio, presented in Figure 11, for BC are 0,81 (± 0,46), 0,83 (± 0,35) and 0,78 (± 0,57) at Artillerigatan, Grevgatan and Karlavägen, respectively, meaning that the mean C_{in}/C_{out} fraction of BC in the three living rooms were below unity as would be expected when indoor sources of BC are absent. The lowest I/O ratio was found for the particle size of 0,5 µm at Artillerigatan (0,40 (± 0,08)) and 0,3 µm at Grevgatan (0,66 (± 0,22)). In a study characterizing particle sizes in a family home with natural ventilation in Espoo, Finland, I/O ratios were estimated to 0,50 ± 0,49 for particle sizes < 100 nm and 0,70 ± 1,36 for particle sizes between 100 nm and 400 nm when measured in a ground floor living room (Hussein et al. 2005), which is in line with the calculated I/O ratio for 0,3 µm in this study. In another study on BC infiltration performed in Barcelona, Spain, minimum I/O ratios were assessed to be 0,7 for BC and 0,73 for PM_1 (Viana, et al., 2011).

Since the apartments were occupied during the whole measurement period, most of the data was recorded when indoor sources were not completely absent. Even if the data recorded during cooking activities, and one candle burning that took place on Artillerigatan, were removed prior to calculations, the fact that someone is at home should make it impossible to state that indoor sources are negligible. However, since the I/O ratios are calculated from a large amount of C_{in} and C_{out} data, spread out over the time of day, the I/O ratios are supposed to be representative for the purpose of this study. Since Grevgatan is a studio apartment, all activities carried out by the residents occurs in the room where the measurements take place. The residents´ bed is placed close to the station for the C_{in} measurements, why the night data probably should not be considered free from indoor sources. When moving blankets and walking by the instruments, deposited particles are resuspended into the air close to the instruments inlets, which in the recorded data is detected as higher particle concentrations in the indoor environment. This affects the C_{in}-measurements and thereby also the I/O ratios. This is probably is the cause for I/O ratios above one for BC measured at night at Grevgatan. At Artillerigatan and Karlavägen, the indoor activities and thereby the indoor sources are more spreadout due to more spaceous apartments and the measurement station in the living room is left for longer periods without the influence of indoor sources. The high I/O ratios of 1,00 (± 2,34) and 24,21 (±77,01) for 5,0 and 10,0 µm, respectively, on Artillerigatan is however best explained by the presence of indoor sources. The I/O ratio is dependent on many variables (Chen & Zhao, 2011), and the influencing factors behind the calculated I/O ratios in this study cannot really be assessed to a more satisfying degree.

The I/O ratio differs with indoor sources of particles and ventilation system, wind characteristics outdoors and the configuration of the cracks and cavities through which the air penetrates (Chen & Zhao, 2011).

Infiltration factor

In this study, only using the data recorded at night was not sufficient to receive values of the I/O ratios or F_{inf} factors that could be used in the further calculations. In the calculations of the penetration factors, "all-day" C_{in}- and C_{out}-data were used where indoor sources were considered negligible.

The particles in the inflowing airstream will penetrate the indoor environment to different degrees depending on their tendency to be affected by the respective deposition mechanisms. Brownian diffusion is an important mechanism for ultrafine particles (< 0,1 µm), and impaction and gravitational settling are more important mechanisms for coarse particles (> 2,0 µm) (Nazaroff, 2004).

F_{inf} factors for BC was estimated to be 0,55 (± 0,02), 0,64 (± 0,02) and 0,41 (± 0,05) for Artillerigatan, Grevgatan and Karlavägen, respectively. Infiltration have been shown to be higher for BC than $PM_{2,5}$ with respective values of 0.84 versus 0.48 in Los Angeles homes and 0,61 versus 0,41 in a home in Clovis, California (Sarnat, et al., 2006, Lunden, et al., 2008). This can be related to the F_{inf} factors for $PM_{2,5}$ estimated here at 0,54 and 0,53 for Artillerigatan and Grevgatan, respectively, which are smaller than the F_{inf} factors for BC but more similar than in the found literature.

Part of the explanation for the differences in F_{inf} factors between the apartments can be the differing distances from traffic, giving rise to BC particles of different sizes due to the aging processes in the outdoor environment.

The F_{inf} factors for 10 µm large particles were -0,24, and 6,21 for Artillerigatan and Grevgatan, respectively, and judging by the looks of the linear regression functions behind these data, presented in appendix a, they were considered invalid and no penetration factor was calculated for the particle size of 10 µm. A F_{inf} factor is expected to be within the range of 0 and 1 in the absence of indoor sources. The remaining F_{inf} data, presented in Table 11, seemed to be reasonable comparing with values of calculated dynamic infiltration factors reported by (Bennett & Koutrakis, 2006) for particle sizes ranging from 0,3 4,0 µm presented in Table 13, and was used to calculate penetration factors.

Table 13, infiltration factors from litterature

Particle size (µm)	0,3	0,4	0,7	1	2	3	4
Mean across homes	0.74	0.68	0.53	0.48	0.37	0.32	0.32

The peak in F_{inf} seen for particles with a particle size of 0,3 µm (0,79 (± 0,15) and 0,96 (± 0,27) at Artillerigatan and Grevgatan, respectively) compared with larger particle sizes (0,5 and 1,0 µm) is in accordance with the deposition rate minimum. F_{inf} quantifies the particles that remain suspended and a relatively low deposition rate would be expected to promote this suspension. Except the difficulty to address all the processes occurring in the building shell when particles pass through it, the method for estimating the F_{inf} is easily applied, and it has previously been used by many researchers (Chen & Zhao, 2011). By taking the slope as the estimated F_{inf} is thought to compensate for the indoor sources not being entirely absent which was suitable because of the already discussed occupation of the apartments. The F_{inf} values were used in the calculations of the P values using the equation $P = (F_{inf} *((a + k)/(a))$ which assumes steady state and no indoor sources (Chen & Zhao, 2011). This approach was thought to allow a calculation of P assuming steady state, but still allowing for temporal variations in the way that the F_{inf} factors were estimated.

Interpreting the R^2 values of the C_{in}/C_{out} linear regressions, presented in appendix a, the indoor concentration correlated best with the outdoor concentration for particles with the size of 0,5 µm. BC has the next best correlation between C_{in} and C_{out} and third best are the correlation for particles with the size of 0,3 µm. The higher correlation for BC, R^2 = 0,73 and 0,83, than for $PM_{2,5}$, R^2 = 0,16 and 0,15, at Artillerigatan and Grevgatan, respectively, suggests that C_{in} of BC is more dependent on outdoor concentrations and less influenced by indoor sources than is C_{in} of $PM_{2,5}$ (Gotschi, et al., 2002).

Pentration factor

The calculated P values for the size distributed particles was in the range of 0,34 to 5,72. The penetration factor is thought to represent the fraction of outdoor particles that passes through the building envelope along with the inflowing air (Chao, et al., 2003), and thus the variables used when calculating P needs to be estimated in the absence of other factors affecting the indoor concentration of the considered particle specie. The P values for 2,5 µm at Artillerigatan and 2,5 and 5,0 µm at Karlavägen was above 1 which is theoretically not possible. The mechanism behind this could be resuspension of deposited particles due to ordinary indoor activities such as walking and folding of clothes (Nazaroff, 2004) acting as an indoor sources for particles of these sizes. (Vette, et al., 2001) also concluded that resuspension is an important indoors source of aerosols with a particle size of >1,0 µm. Since the apartments were occupied throughout the measurement period the

indoor sources for particles larger than 1,0 µm was not possible to disregard, which is reflected in the linear regression diagrams produced when estimating the F_{inf} factors, from which P is calculated. The P values calculated in this study for sizes ranging from 0,3 – 1,0 µm was in the range of 0,34 < 1,04 which could be reasonable considering the amount of uncertainties involved in the estimations of the variables that P is dependent on. In Figure 16, penetration factors obtained from a number of previous studies on real buildings by (Chen & Zhao, 2011) lies in the range of 0,6 < 1,0 for the particle size range of 0,05 < 2 µm.

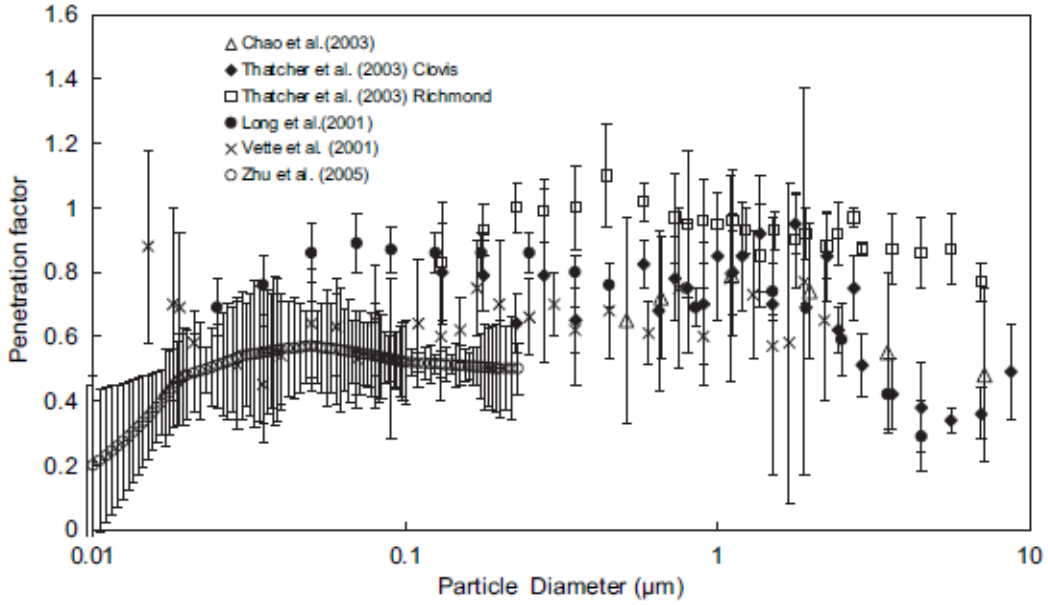

Figure 16, penetration factors obtained from experiments in real buildings, copied from (Chen & Zhao, 2011).

The P minimum was found for particles with a size of 1,0 µm (0,34) in the apartment at Artillerigatan and at 0,3 µm (0,52) at the apartment at Grevgatan. The explanation for this could lie in the study design. At Artillerigatan, the small opening resulting from the hose sticking out through the porthole beneath the window in the living room, was a source for a small amount of inflowing air, and did assumedly not give rise to any filtration of the particles passing through it. The fact that P for BC is 1,41 and 1,53 at Artillerigatan and Grevgatan, respectively, might be due to the approach of estimating the deposition rate of BC. BC generated from candles differs from traffic-related BC that enters an apartment, and the deposition rate of BC is probably over-estimated since the BC measured in the decay experiments is "freshly" generated and in a smaller size range than the one originating from outdoors. The over-estimated k is thus not comparable with the estimated F_{inf} for BC generated outdoors, why P is over-estimated for BC when calculated through the equation $P = (F_{inf}*(a + k)/(a))$.

Conclusion

The simple study design of this project enables the avoidance of complex calculations that would be needed to take into account all characteristics and properties of the specific study objects involved.

The C_{in} values at Artillerigatan, Grevgatan and Karlavägen, respectively, are in the expected magnitude and higher when indoor sources are present than when they are considered negligible.

The C_{out} values are higher than the corresponding C_{in} values which is expected in an urban environment that Stockholm is.

Cooking activity related peaks can easily be detected in the C_{in}-data from Artillerigatan, Grevgatan and Karlavägen, respectively. Also, the effect of residential wood burning on the $C_{BC,out}$-data from Grevgatan can be seen on the evening time mean values in Figure 7, emphasizing the importance of taking other combustion sources into account when analyzing traffic related BC in this manner.

The air exchange and deposition rates were easily estimated measuring the decay in candle generated CO_2 and particles, and the size dependence shown in the deposition rates suggests that this method is applicable, with the condition that the ventilation rate does not vary significantly with time, and that a proper substitute for traffic related BC is used when estimating the decay rate for BC.

Since outdoor BC sources have shown to account for 16-31% of the total BC indoors (LaRosa, et al., 2002), the quality of the estimated F_{inf}- and penetration factors is dependent on the assessment of indoor sources in terms of chemical and physical characterization of the BC that are generated from them. The results conducted in this study are relevant for occupied apartments and affected by the fact that indoor sources cannot be entirely neglected. Size distribution assessment of indoor and outdoor BC could have broadened the analysis of the origin of indoor BC. Despite the difficulty in removing indoor sources completely, I/O ratios and infiltration factors estimated from the C_{in} and C_{out} data for BC are in line with the literature. These parameters were estimated in a more straightforward way than the experimentally estimated parameters a and k, which complexity probably is the reason for the resulting, questionable, P values in this study. Candles might not be a suitable substitute for generating BC with relevant characteristics for the assessment of traffic related BC.

The volume of literature on indoor concentrations of traffic-related BC was found to be somewhat scarce and a lot of work needs to be done in the attempt to better assess human exposure to BC through the analysis of indoor concentrations. Besides future risk assessment based on indoor exposure of traffic-related BC, it would be useful to perform health risk analysis on BC generated indoors, e.g. on candle burning, in order to make relevant estimations on health effects from the exposure of BC generated both indoors and outdoors simultaneously, as this is the real scenario in residential indoor environments.

The higher correlation between the indoor and outdoor concentration of BC than of $PM_{2,5}$ at Artillerigatan and Grevgatan, respectively, and thus the larger influence of outdoor concentrations for indoor exposure for BC than $PM_{2,5}$ emphasizes the advantage of monitoring BC when assessing the infiltration of traffic-related PM in future epidemiological studies.

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