Artikel i Husbyggaren
No 6, år 2007

Luftens partiklar påverkar sjuklighet och dödlighet

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INNEMILJÖ Luften vi andas innehåller partiklar. De bildas exempelvis vid slitage, uppvärmning eller kemiska reaktioner. Riktigt små partiklar är extra reaktiva. Detta ställer krav på var man bygger, på byggmaterialet och på ventilationen.

Luftens partiklar påverkar sjuklighet och dödlighet

Av MATS BOHGARD, professor, *Lunds tekniska högskola/Lunds universitet*

ALL LUFT INNEHÅLLER svävande partiklar. Ibland – särskilt på senvintern då solen ligger lågt – brukar man i bostäder kunna se massor av dammpartiklar som seglar runt i luften när solstrålarna kommer in genom fönstret och reflekteras mot dem.

Partiklarna finns där också när vi inte ser dem. Men vi ser bara en bråkdel av alla partiklar som finns i luften. De flesta är för små för att vi ska kunna se hur ljuset sprids mot dem. Vid höga koncentrationer kan vi då i stället se ett blåaktigt dis om partiklarna är riktigt små.

Inomhusluftens partiklar

Luften i våra inomhusmiljöer innehåller normalt mellan tusen och några hundratusen partiklar per milliliter. Partikelstorleken sträcker sig från en tusendels mikrometer till hundra mikrometer (en miljondels till en tiondels millimeter).

Det är ett brett storleksintervall. Om vi tänker oss att vi förstorade upp partiklarna så den minsta partikeln blev som ett knappnålshuvud, skulle den största vara ungefär lika stor som Globen i Stockholm. De allra minsta partiklarna, de under en tiondels mikrometer (100 nanometer), kallas ibland nanopartiklar eller ultrafina partiklar. Den kemiska sammansättningen och formen kan variera mycket.

Partiklarna kan i princip uppkomma på tre olika sätt: genom sönderdelning av material och uppvirvling, genom att gasmolekyler slår ihop sig till större kluster (till exempel när en ånga kyls ner i luften) och genom kemiska reaktioner i luften.

Slitage från däck och vägbana

Partiklar mellan 1 och 100 mikrometer uppstår vanligen när material finfördelas. I naturen kan de bildas genom jorderosion och havsspray. Från biltrafiken tillförs slitagepartiklar från däck och vägbana till luften. I hemmiljön finns till exempel partiklar från hud och textilier och från uppvirvling från dammiga ytor. Vid nötning (till exempel skor mot golv) kan det också bildas partiklar i dessa storlekar.

Partiklar mindre än 1 mikrometer uppstår vanligen genom kondensationsprocesser och kemiska reaktioner. Naturliga källor är skogsbränder och vulkanutbrott. Förbränningsanläggningar för energiproduktion och bilmotorer är exempel på källor till småpartiklar som människan har skapat. Matlagning, rökning och tända stearinljus ger små partiklar i hemmiljön. Laserskrivare och kopiatorer kan också ge upphov till små partiklar.

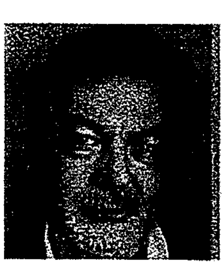
I inomhusmiljöer kan partiklar också uppstå genom att organiska ångor, till exempel från rengöringsmedel, reagerar med ozon från uteluften varvid mycket små partiklar bildas.

Dör i förtid

De senaste åren har allt större uppmärksamhet riktats mot hälsoeffekter av luftens partiklar. Det finns flera orsaker till detta. Det får nu anses fastlagt att det finns starka samband mellan exponering för små partiklar och dödlighet/sjuklighet utifrån epidemiologiska studier baserade på stora populationer.

Enligt de senaste uppskattningarna dör varje år hundratusentals människor i förtid i Europa (cirka fem tusen i Sverige) på grund av luftföroreningspartiklar. Då är inte tobaksrökningens effekter medräknade.

De luftburna partiklarna kan påverka hälsan på olika sätt. Framför allt är det sjukdomar i andningsvägarna och hjärtkärlsjukdomar som brukar förknippas med partiklarna.



FÖRFATTAREN

Mats Bohgard är professor vid avdelningen för ergonomi och aerosolteknologi vid Lunds tekniska högskola/Lunds universitet. Han har lång erfarenhet av forskning om luftburna partiklar och deltar för närvarande i flera tvärvetenskapliga forskningsprojekt.

Andra orsaker till den ökande uppmärksamheten, särskilt i inomhusmiljöer, är den ökade förekomsten av allergier, astma och överkänslighet, samt sjukahusproblematiken.

Mäter mest utomhus

De mätningar som ligger till grund för uppskattningarna om total dödlighet/sjuklighet är huvudsakligen gjorda i utomhusluften. Det görs ständigt omfattande mätningar av utomhusluftens partiklar. Luftens partiklar utanför husväggarna har vi relativ god kunskap om.

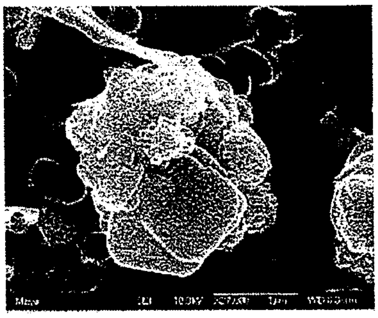
Men normalt är vi inomhus mer än 85 procent av vår tid (hemma, på arbetet, i skolan, på daghem, i bil, på buss etcetera). Det har inte gjorts lika många studier av inomhusluftens partiklar. Vi har därför ganska dåliga kunskaper hur det kan se ut i olika slags inomhusmiljöer. Och vi har inga säkra kunskaper om eventuella hälsoeffekter av de flesta partiklar som brukar finnas inomhus.

Från de studier som har gjorts på olika håll i världen kan man sluta sig till att i folktomma lokaler domineras partiklarna av sådana som kommer med uteluften via ventilation eller otätheter i byggnaden. Koncentrationen och typen av dessa partiklar beror på var byggnaden är belägen (närheten till trafikerade vägar, järnvägar, förbränningskällor) och hur den är utformad med tanke på ventilation, filter, täthet och var friskluftsintaget är beläget med mera.

I folktomma lokaler utan starka partikelkällor, är normalt partikelhalten lägre inomhus än utomhus. Men det blir ofta tvärtom vid olika slags aktiviteter. En stor del av partiklarna kommer också då från uteluften, men städning, textilhantering, matlagning, tända stearinljus, tobaksrökning och att människor och husdjur rör sig i miljön kan ge mycket stora bidrag.

I princip kan alla material som vi använder till byggnader, inventarier och konsumentprodukter hamna i finfördelad form i luften om vi hanterar dem. Det innebär att partikelsammansättningen kan bli mycket komplex. Sammansättningen av partiklar ändras också med tiden på grund av nya vanor, nya konsumentprodukter och ny hemutrustning.

Bilden visar en liten partikel som har bildats av ett brinnande stearinljus. Partikeln har samlats in genom att den laddats elektriskt och deponerats på en platta varefter den har fotograferats med hjälp av ett elektronmikroskop. Partikeln är ett aggregat som har bildats genom att partiklar med några tiotals nanometers diameter har slagits ihop. Foto: Joakim Pagels och Lisa Karsson



Bilden visar en partikel som frigjorts till luften från textilier som har tvättats med tvättmedel innehållande svårlösliga zeoliter vilka har fastnat i textilen. Partikeln har sedan blivit luftburen när textilen har hanterats, varefter den har infångats på ett filter och fotograferats med ett elektronmikroskop. Vid hantering av textilier har det uppmätts relativt höga halter av luftburna partiklar. Partikelns diameter är 2–3 mikrometer – vilket gör att den kan tränga långt ner i andningsvägarna vid inandning.
Foto: Anders Gudmundsson och Jakob B. Wagner

Tvättmedelsrest frigörs till luft

Vi har ännu inga metoder för att säkert bedöma om inomhusluftens kvalitet är godtagbar. Det här är ett ökande problem, särskilt för människor som är känsliga för luftföroreningar.

På senare tid har partiklar som frigörs från golvs ytbeläggning, från tvättme-

Fortsättning s. 22 ▸

➤ delsrester i textilier och från rengöringsmedel i sprayform uppmärksammats. Vi har idag otillräcklig kunskap om inomhuspartiklarnas sammansättning och hur de påverkar vår hälsa.

Det verkar rimligt att anta att sådant som livsstil, inventarier, konsumentprodukter och partikelalstrande apparater har betydelse, men också planlösning och hur ventilationen är utformad, verkar ha betydelse även för exponering för de partiklar som har sitt upphov i den inre miljön.

Små partiklar når lungblåsor

Vi vet att partikelstorleken har betydelse för effekterna på människan när vi andas in partiklar. Deras storlek har betydelse för hur de transporteras och hur de deponeras i andningsvägarna. Generellt kan man säga att partiklar som är större än fem till tio mikrometer hamnar i näsa, svalg och i andra delar av de övre luftvägarna. Mindre partiklar har förmåga att ta sig ner djupt i lungorna – ända ner till lungblåsorna. Riktigt små partiklar kan både hamna högt upp och långt ner i andningsvägarna.

Den kemiska sammansättningen har betydelse för hälsofarligheten. Men det faktum att partiklarna är små kan i sig utgöra en hälsofara, eftersom de kan tränga ner i andningsvägarna. Dessutom blir material mer reaktiva ju mer de finfördelas. Det beror på att ett stort antal partiklar tillsammans har större yta än en stor, även om de innehåller samma mängd material. Det faktum att små partiklar har kraftigt krökt yta, innebär också i sig att små partiklar kan reagera lättare med vävnader.

Kvartsdamm, asbestfibrer, mjöldamm och trädamm är sedan länge kända för att ge allvarliga yrkessjukdomar trots att den kemiska sammansättningen i sig inte innebär att ursprungsmaterialet är giftigt.

Söker kunskap

Det pågår en hel del arbete på många håll i världen för att öka förståelsen av de luftburna partiklarna i olika slags inomhusmiljöer och vilka effekter de kan ha på människor. Detta görs också vid olika forskningsinstitutioner i Sverige.

I vårt laboratorium vid Lunds universitet pågår just nu fyra olika projekt som handlar om inomhusmiljöns partiklar. I ett av dessa görs detaljerade mätningar i olika inomhusmiljöer. I ett annat utvecklas mätmetodik för enkla snabba mätningar i byggnader. I ett tredje, som görs tillsammans med institutionen för yrkes- och miljömedicin vid Lunds universitet och kardiologisk expertis från Karolinska institutet, exponeras försökspersoner för vanliga inomhuspartiklar i välkontrollerade kammarstudier. Effekter på andningsvägar och hjärt-kärlsystemet mäts. I det fjärde projektet deltar vi i en studie som leds från arbets- och miljömedicin vid Uppsala universitet. I studien genomförs enkäter och mätningar och medicinska undersökningar av boende i svenska bostäder. Forskningen stöds av Formas och SBUF.

Dessa studier, tillsammans med många andra som görs i inomhusmiljöer, bör medföra att vi om några år har betydligt bättre kunskaper om hälsoeffekterna än vi har idag.

De slutsatser som vi kan dra idag om hur man kan begränsa exponeringen är att hålla avstånd mellan byggnader och trafikerade vägar och förbränningskällor, att tänka på var friskluftsintaget är placerat, att använda effektiva partikelfilter i ventilationssystem och vädringsfönster. att undvika otätheter i byggnaden och att tänka på vilka material man använder – särskilt när det gäller sådant som utsätts för friktion och nötning.

När det gäller partiklar som bildas inomhus verkar det ha mycket att göra med konsumentprodukter, aktiviteter och livsstil. Men även här har byggnadens utformning betydelse när det gäller ventilationens utformning och hur byggnaden planeras med avseende på olika slags partikelalstrande aktiviteter. □

Fotnot:
För den som är intresserad av att läsa mer rekommenderas en rapport som gjorts av arbets- och miljömedicin vid Uppsala universitet på uppdrag av Socialstyrelsen: *Partiklar i inomhusmiljön – en litteraturgenomgång* (2006-123-1). Den kan laddas ner från Socialstyrelsens hemsida, www.socialstyrelsen.se.

Abstract, accepterat som bidrag till
Indoor Air Conference 2008, Köpenhamn

A Low Cost Aerosol Monitor for Screening Measurements in Indoor Environments.

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A Low Cost Nanoparticle Monitor for Screening Measurements in Indoor Environments

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Keywords: Aerosol instrument, Ultrafine particle

1 Introduction

This paper is focused on the improvement of a common ionization type smoke detector for monitoring nanoparticle concentrations in indoor environments at a low cost and a low level of maintenance.

2 Method

In the detector air ions are generated by a radioactive source. The ions make an electrical field between two electrodes inhomogenous and a third electrode senses the potential in a point in the field. When airborne particles enter the electrical field, the ions attach to the particles and this changes the potential of the sensing electrode with dV according to:

$$dV = -dV_{max}\cdot(1-e^{-\alpha\cdot\beta\cdot N}) \qquad (1)$$

(Litton et al. 2004) where dV_{max} is the maximum shift, α depends on the dimensions of the sensor, β is the ion attachment coefficient which is linear towards the particle diameter (D_p). N is the particle concentration. For low concentrations equation 1 can be approximated to:

$$dV = k\cdot N\cdot D_p \qquad (2)$$

where k represents the sensor response. The product of N and D_p is the particle diameter concentration.

The output of the sensor is also affected by the pressure, temperature and humidity as these parameters affect the electrical properties of air. With a similar monitor Edwards et al. (2006) used a numerical approach to correct for the air properties. In contrast, this monitor measures the difference between filtered and untreated air and thereby only the response from the particles affect the output of the sensor.

3 Results and discussion

The sensor was calibrated with a NaCl aerosol and the size distribution was measured with a particle sizer. The sensor gave a response (k) of 4 mV per mm/cm^3. The detection limit was 1.5 mm/cm^3 which corresponds to a number concen-tration of 15 000 #/cm^3 100 nm particles.

The sensor was tested on aerosols of candle smoke and welding fumes and the signal response versus particle length concentration and the results are shown in Figure 1.

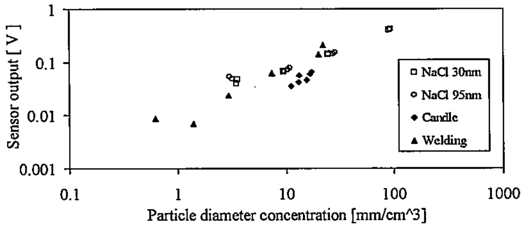

Fig 1. Particle diameter concentration of calibration, candle and welding aerosols versus sensor output.

The results suggest that this sensor can be used for detection of temporarily raised particle levels that occur in indoor environments.

Acknowledgement:

This work has been supported by the Development Fund of the Swedish Construction Industry (SBUF) and the Swedish Research Council FORMAS.

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Luftburna partiklar i ny kontorsmiljö

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ISSN 1650-9773
Publikation 31, år 2007
LUTMDN/TMAT-3028-SE
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Sammanfattning

Luftburna partiklar har mätts i ett nybyggt kontor i Malmö under en treveckorsperiod i juni 2006.

Med en APS (Aerodynamic Particle Sizer) mättes halten av stora partiklar (0.5 – 20 µm). Dessa mätningar visade:

- under arbetstiden är det en förhöjd partikelkoncentration till en nivå på ca 2 µg/m^3,
- efter arbetstid sjunker partikelhalten ner till en bakgrundsnivå kring 0.3 µg/m^3,
- aktiviteter i kontoret bidrar med ca 1.7 µg/m^3 till inomhusluften,
- medelvärdet av masskoncentrationen över hela perioden var 1.0 µg/m^3.

Fina partiklar (0.01- 1 µm) mättes med ett SMPS-system (Scanning Mobility Particle Sizer) och resulterade i:

- inget tydligt regelbundet mönster eller koppling till någon partikelkälla kunde observeras,
- medelvärdet över hela perioden var för masskoncentrationen 3.5 µg/m^3 och för antalskoncentrationen 830 partiklar/cm^3.

I medel var totala masskoncentrationen (0.01-20 µm) ca 4.5 µg/m^3 och i medel under arbetstid 5.5 µg/m^3. Motsvarande PM10 koncentrationer är knappt 10% lägre. Partikelhalterna av både fina partiklar och stora partiklar får anses vara låga. Masskoncentrationen i kontoret var jämfört med utomhusluften i Malmö för fina partiklar (< 1 µm) uppskattningsvis 50% lägre och för stora partiklar (1- 10 µm) 90% lägre.

Inledning

Under tre veckor, 2 juni till 26 juni 2006, mättes halten luftburna partiklar i ett kontor beläget i ny byggnad i Malmö hamn, Hjälmarekajen, kvarteret Scylla. Förutom vanligt kontorsarbete förekom arkivering och hantering av pappersritningar samt ett specialventilerat rum för utskärning av t ex frigolit till modeller.

Mätningarna påbörjades en dag (fredag 2 juni) innan inflyttning av inredning och möbler (lördag 3 juni till 6 juni). Första arbetsdagen med all inredning, möblemang och annan utrustning på plats var tisdagen 7 juni. Därefter pågick mätningarna i ytterligare knappt 3 veckor.

Mätutrustning

De luftburna partiklarna mättes med två olika partikelinstrument.

- APS (Aerodynamic Particle Sizer, model 3321, TSI Inc) för mätning av stora partiklar (0.5 – 20 µm, aerodynamisk diameter)
- SMPS (Scanning Mobility Particle Sizer, TSI Inc.) för fina partiklar (0.01 – 0.7 µm).

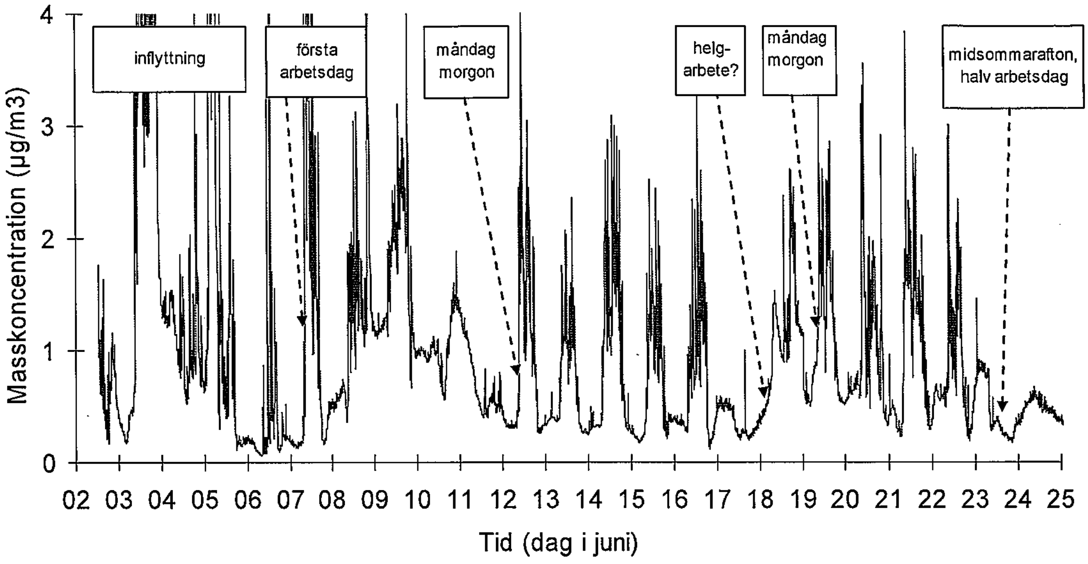
Mätutrustningen var placerad i en öppen kontorsmiljö och på ett samma ställe under hela mätperioden.

Resultat

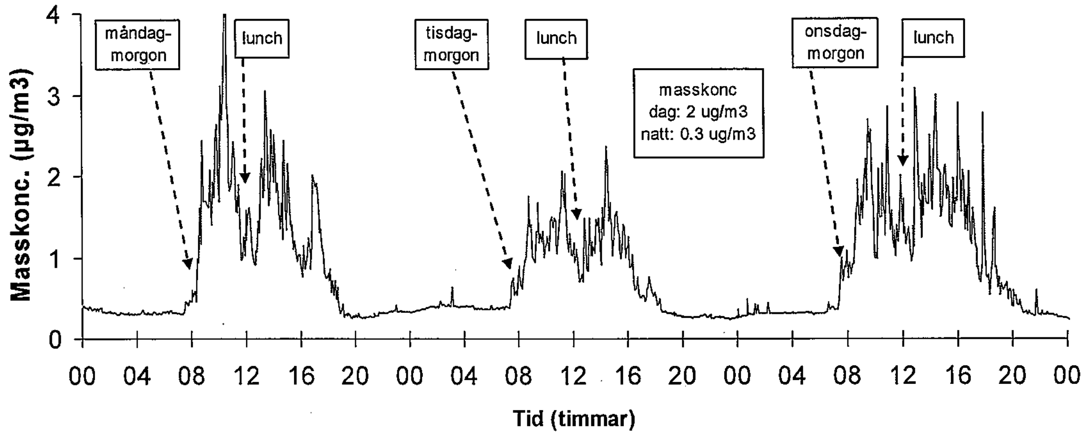
Figur 1 visar hur masskoncentrationen (APS-mätningar, 0.5 – 20 µm) varierar med tiden för stora partiklar > 0.5 µm över hela mätperioden. I figur 2 visa mer detaljerat hur partikelhalten varierar över några dygn. Sammanfattningsvis kan noteras:

- under arbetstiden är det en förhöjd partikelkoncentration till en nivå på ca 2 µg/m^3,
- något lägre koncentration under lunchen
- efter arbetstid sjunker partikelhalten ner till en bakgrundsnivå kring 0.3 µg/m^3.

Partikelmätningarna visar att partikelkällorna i kontoret bidrar med ca 1.7 µg/m3 till inomhusluften.



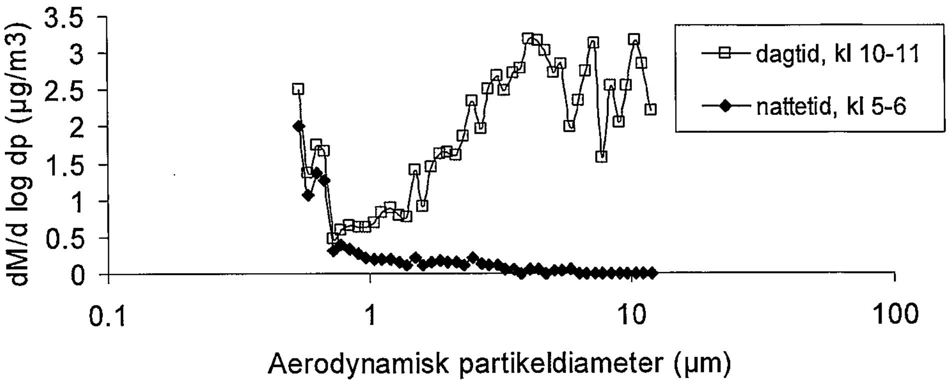
Figur 1. Masskoncentrationens (µg/m³) (APS-mätningar) variation med tiden i inomhusluften för stora partiklar >0.5 µm.



Figur 2. Masskoncentrationens (µg/m³) (APS-mätningar) variation under tre dygn i inomhusluften för stora partiklar >0.5 µm.

Figur 3 visar partikelstorleksfördelningen under arbetstid och under natt. Figuren visar att:

- under dagtid bildas stora partiklar > 1-2 µm
- förekomsten av partiklar 0.5 – 0.7 µm är lika under dag och natt



Figur 3. Storleksfördelningen för partikelmassan (APS-mätningar) under dagtid och nattetid i kontoret. Under dagtid sker aktiviteter som bildar luftburna partiklar > 1 µm.

Mätningarna av mindre partiklar med SMPS (0.01 – 0.7 µm) visar ingen tydlig dygnsrytm som för de stora partiklarna. Detta kan tolkas som att de anställdas aktiviteter skapar stora partiklar medan halten fina partiklar beror mer på bidrag från utomhusluften och att det finns inga starka inomhuskällor av fina partiklar. I medel är antalskoncentrationen av fina partiklar ca 800 partiklar/cm3 och masskoncentrationen ca 3.5 µg/m3.

Den totala masskoncentrationen av luftburna partiklar i kontoret är alltså ungefär: SMPS (0.01 – 0.7 µm) + APS (0.7 – 20 µm):

Medel under hela mätperioden = 3.5 + 1.0 = 4. 5 µg/m3
Medel under arbetstid = 3.5 + 2.0 = 5.5 µg/m3

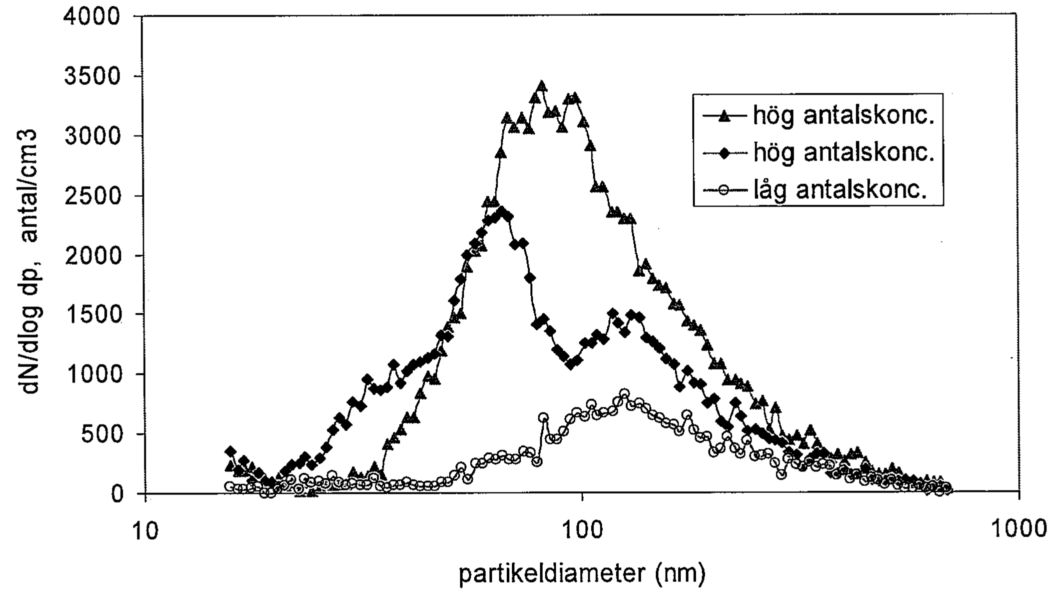
Diskussion

För att relatera partikelmätningar av luftburna partiklar till effekt på människan brukar man mäta halten av partiklar med aerodynamisk partikeldiameter < 10 µm, vilket benämn PM10.

Masskoncentrationen för utomhusluften på taknivå i Malmö ligger normalt runt 16 µg/m^3 mätt som dygnsmedelvärde för PM10 (Henric Nilsson, Malmö miljöförvaltning, 2006). Halten av PM1 (partiklar < 1 µm) i Malmö är normalt ca 50% av PM10 (Henric Nilsson, 2006). Det innebär att halten av partiklar mellan 1 och 10 µm är uppskattningsvis ca 8 µg/m^3 i utomhusluften. APS-mätningarna visar att halten av partiklar mellan 1 och 10 µm i kontorets inomhusluft är 0.7 µg/m^3. Detta visar att utomhusluftens stora partiklar (1 -10 µm) effektivt reduceras från ca 8 till en nivå på 0.7 µg/m^3 i inomhusluften, dvs en faktor 10. Partikelhalterna av stora partiklar (> 1 µm) i kontoret får betraktas som låga.

Vi kan också jämföra PM1 halten för utomluften på ca 8 µg/m^3 med uppmätta halter av medelvärdet för SMPS-mätningar av inomhusluften, som var ca 3.5 µg/m^3. SMPS mäter mellan 0.01 och 0.7 µm och bidraget mellan 0.7 och 1 µm är uppskattningsvis ytterligare 20% dvs 0.7 µg/m^3. PM1-halten inomhusluften är alltså ca 4.2 µg/m^3 (3.5+0.7), vilket kan jämföras med utomluftens PM1-koncentration på 8 µg/m^3. Inomhusluftens PM1-halter är halverade jämfört med utomluftens PM1 halter.

Att reduceringen av PM1-partiklar är mycket mindre än reduceringen av stora partiklar (>1 µm) beror på att ventilationsfilter har sin sämsta effektivitet för partiklar kring 0.3 µm. Det betyder att stora delar av utomluftens PM1-partiklarna kan ta sig genom filter in till inomhusluften. Figur 4 visar hur partikelstorleksfördelningen i inomhusluften ser ut vid olika tillfällen: vid låg koncentration ca 500 partiklar/cm^3 och vid högre koncentrationer 1500 respektive 1000 partiklar/cm^3. Vid höga halter inomhus finns alltså ett bidrag av fina partiklar mellan 0.05 och 0.5 µm.



Figur 4. Storleksfördelningen för partikelantalet (SMPS-mätningar) vid olika koncentrationsnivåer för inomhusluften i kontoret.

Methodology for identifying particle sources in indoor environments

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Received 10th January 2007, Accepted 12th June 2007
First published as an Advance Article on the web 29th June 2007
DOI: 10.1039/b708731b

Growing concern about airborne particles in indoor environments requires fast source identification in order to apply remedial actions. A methodology for identifying sources emitting particles larger than 0.5 μm was designed and applied. It includes: (1) visual inspection of interior surfaces in order to identify deposited particles and inspection of potential sources (equipment, materials, activities *etc.*) of airborne particles. (2) Technical measurements of airborne particles at different positions in a building with simultaneous logging of activities. (3) Isolating potential activities/particle sources in a test chamber, initially free from particles, for controlled characterizations of the particles generated. The methodology was applied in a study of three houses in southern Sweden. The results show that source identification is facilitated by knowledge of concentration variations between different rooms, real-time measurements together with activity reports and information on particle characteristics that are comparable with results from laboratory simulations. In the houses to which the methodology was applied, major particle emissions from textile handling were identified, which were likely due to detergent zeolite residues.

Introduction

Exposure to airborne particles in indoor environments is considered to be an important factor for various health effects and for how inhabitants perceive the environment. For some types of aerosols present in indoor environments there is conclusive evidence[1] (environmental tobacco smoke, radon progenies, asbestos, allergens, *etc.*), but there have been few conclusive reports on health effects of particles in general indoor environments.[2]

Numerous epidemiological studies have shown that elevated outdoor particle concentrations measured as PM_{10} and $PM_{2.5}$ are associated with increased prevalence of respiratory and cardiovascular morbidity and mortality.[3–6] As most people seem to spend more than 85% of their time indoors,[7] it is reasonable to assume that the major exposure to particles also occurs in these environments. Although there are few conclusive studies on the relationships between indoor exposure and health effects, there are reasons to believe that indoor exposure affects health.

Indoor exposure consists of a mixture of ambient primary particles or secondary created particles of outdoor origin (infiltration of outdoor air pollutants followed by chemical transformation/condensation) and particles from indoor activities and processes. Typical sources for fine particles are thermal and combustion activities/processes such as smoking, incense, cooking and kerosene heaters,[8] whereas typical sources for coarse particles are activities causing disintegration of materials and resuspension.[9,10] When problems related to indoor sources are perceived or suspected, the first aim of inhabitants and technical practitioners is not to find relationships between exposure and effects. It is rather to identify the source and eliminate the problem—if possible.

The indoor environment is complex. There are many potential particle emission sources. Concentrations and composition can change quickly and the variation is huge between buildings at various activities. Furthermore, in a given indoor environment (dwelling, office, school, *etc.*), new consumables and equipment as well as changes in lifestyle and routines can cause changes in concentration levels and dust composition over time. In a study by Wallace[11] of indoors environments, the fraction of unknown sources was estimated to be about 25%. To deal with this complexity, there is a need for fast and practical procedures and methods to identify the emission sources. As soon as a source or some sources are identified, methods more suited for the examination of effects can be applied to explore potential health consequences and/or measures can be taken to eliminate or reduce the emissions/exposure.

Airborne particulate matter can be characterized with respect to various features (number, area, volume/mass concentrations and size distributions, physical, chemical and biological properties and shape and morphology) with various measurement techniques. In a comprehensive review[2] on associations between exposure to particles in buildings and health effects, it was concluded that exposure measurements, reported from published scientific studies on indoor air are performed in very diverse ways and that they are very often far from optimal. In many studies, the measurement techniques used were not related to the objectives of the studies. The measurement methods seemed often to be chosen in an arbitrary way or estimated from proxies. There is a need for better measurement methodology designs which give information of aerosol features which are relevant for different

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objectives. This work is a contribution to the efforts to develop study designs which permit performance of efficient identification of particle sources in the indoor environment.

The aim of this study was to develop and apply a methodology for fast source determination of particles larger than 0.5 μm, in cases where inhabitants experienced unusually high dust levels or else when airborne dust is believed to cause annoyance or to affect health. The methodology was applied to home environments where the residents complained about high levels of airborne and deposited dust.

Methods

The method for identifying sources consists of three steps: (1) visual inspection of interior surfaces in an attempt to find deposited particles and examination of potential sources (such as equipment, materials, activities, *etc.*) of airborne particles. (2) Technical measurements of airborne particles in different positions of a building with simultaneous logging of activities. (3) Experimental characterisation of particles generated from isolated activities/particle sources in a laboratory test chamber, initially free from particles.

The three steps were applied in a study of three neighbouring houses in a small town in southern Sweden. Two of the houses had reported dust problems: House A (no dust problems, four persons, 130 m^2), House B (dust problems, two persons, 132 m^2) and House C (dust problems, four persons, 135 m^2). The three houses were built in 2001 and all were one-storey brick buildings on concrete ground plates.

The first step, the visual inspection, was made by the building contractor, by the inhabitants and by researchers from the University.

In the second step, particle concentrations were measured in four rooms in every house. The inhabitants were asked to keep all windows and outer doors closed. The technical instruments used were: an aerodynamic particle sizer, APS (Model 3321, TSI Inc., USA) and three optical particle counters, OPC (Remote 5010, Lighthouse Worldwide Solutions, USA). The APS provides a detailed size distribution of the aerodynamic equivalent particle diameter in the range 0.5–20 μm. The OPC classifies the detected particles into two size ranges: small particles, 0.5 to 5 μm, and large particles, ≥ 5 μm. As we needed to measure in different rooms in the houses when the inhabitants were present, disturbing noise from the instruments had to be avoided. Therefore, OPCs without internal pumps were used. Instead, a central pump placed in the laundry room of each house was connected to the OPCs with thin plastic tubing. The noisy APS was located in the laundry along with the central pump, see Fig. 1. The instruments measured continuously for three days and nights in each house while the families recorded different activities such as cooking, washing, cleaning and sleeping times in a logbook.

The regional background particle concentration was not measured but obtained from continuous measurements made by The Swedish National Environmental Protection agency using a TEOM (tapered element oscillating microbalance) situated in the country side about 70 kilometres north-west of the houses. The source region of the air was traced using back-trajectories calculated with the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's HYSPLIT model.

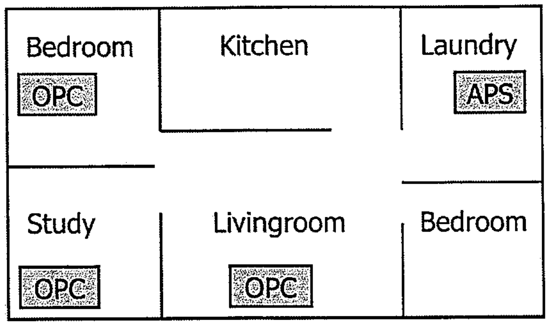

Fig. 1 Schematic diagram of the layout of the houses and placement of the three OPCs and the APS.

The third step, simulation of activities/sources in the laboratory, was carried out in an airtight test chamber (volume 22 m^3, walls of stainless steel, temperature 20 °C, RH 25–40%, ventilated with 24 m^3 h^{-1} particle free air to a background concentration below 50 cm^{-3}, in the size range 10–500 nm and below 1.0 cm^{-3} in the range 0.5–20 μm). From the conclusions made in step 1 and 2, textiles washed with zeolite detergents were considered to be a likely particle emission source (see Results section). Therefore, the particle emissions from textiles washed with a detergent containing zeolites was compared to those from textiles washed with a detergent containing phosphates. The textiles were washed with a household washing machine (AEG Lavamat W1229). On each occasion, the washing machine was operated with a standard washing program. The textiles were then handled with standardized motions in the test chamber while the particle emissions were studied. The person treating the textiles was wearing clean-room clothing to minimize contamination. First, the textiles washed with traditional detergent containing phosphate were studied and then the textiles washed with detergent containing zeolites. Measurements were done on-line with an APS and a scanning mobility particle sizer (SMPS,[12] design: Lund University). The APS was placed on the floor of the chamber at a distance of about one metre from the source, while the SMPS sampled outside the chamber from a tube through the wall. Particles were also sampled on NuclePore® filters (track-etched membranes of polycarbonate, pore size 0.4 μm, 10 L min^{-1} during 15 min) for analysis with scanning electron microscopy (SEM, JEOL JSM-6700F, 20 kV) and X-ray energy dispersive spectrometry (XEDS).

Results

Step 1. Visual inspection

In the visual inspection a number of frequently occurring potential indoor particle sources were investigated. For instance, all houses were furnished with normal household equipment such as electrical stoves and washing machines and they were equipped with central vacuum cleaner systems. House A had a drying chamber in the laundry; Houses B and C had condenser tumble driers. The inhabitants in House A used detergent powder containing phosphate, while the inhabitants in Houses B and C used detergent powder containing zeolites (30% zeolites).

Obvious unnecessary particle sources were minimized. No open fires or candles or the like were used during the measurement period. Inhabitants in House A reported daily cigarette smoking (1–3 cigarettes) in the kitchen close to the cooker hood ventilator.

The houses were ventilated using mechanical exhaust air systems with cooker hood ventilators in the kitchen and exhaust ventilators in the laundry and bathroom. Supply air ventilators were placed in all the other rooms including the bedroom, study and living-room. Thus, there was a substantial mixing of the air in the houses. The supply air was filtered through an electret filter (Fresh AB, Sweden). The houses were heated with water radiators. The outdoor daytime temperature during the measurement period ranged from 15 to 22 °C and night time from 10 to 15 °C.

No major particle sources were identified nearby the houses. They were situated in a residential district and the local environment consisted of newly planted gardens (2–3 years old). There were no industrial activities or major busy streets in the vicinity of the houses.

For the two houses with dust problems, deposited particles appeared to be evenly distributed over the interior surfaces in all rooms. They could be visually observed both on vertical and horizontal surfaces. Using a simple microscope (magnification 30×), it was observed on location that a vast majority of the particles were less than about 20 µm in diameter. Only single occurrences of larger fibres and large soil particles could be detected. Microscope and element analysis conducted by the building contractor showed that the deposited dust did not arise from the insulating material of the house.

Step 2. Field particle measurements

Fig. 2, 3 and 4 show the results from the three houses using the three OPC instruments, situated in the bedroom, living-room/kitchen and study and from the APS in the laundry. The OPC measurements are presented as the number concentration for particles between 0.5 and 5 µm and the APS measurements are presented as calculated particle mass distributions between 0.5 and 20 µm, assuming a density of 1000 kg m^{-3}.

Table 1 shows the mean values of three days and nights (daytime 06:00–22:00). The particle concentrations measured in the living-room, study and bedroom using the OPCs were very similar and are therefore given as a mean value for all three rooms. The values from the OPCs and the APS can not be compared quantitatively as the instruments measure different size-related properties of the particles. The OPCs determine the particle size from the total scattered light of a particle and the APS determines the aerodynamic diameter using the particle inertia.

The only significant difference between the houses, in mean concentration of particles, was the APS mass concentrations in

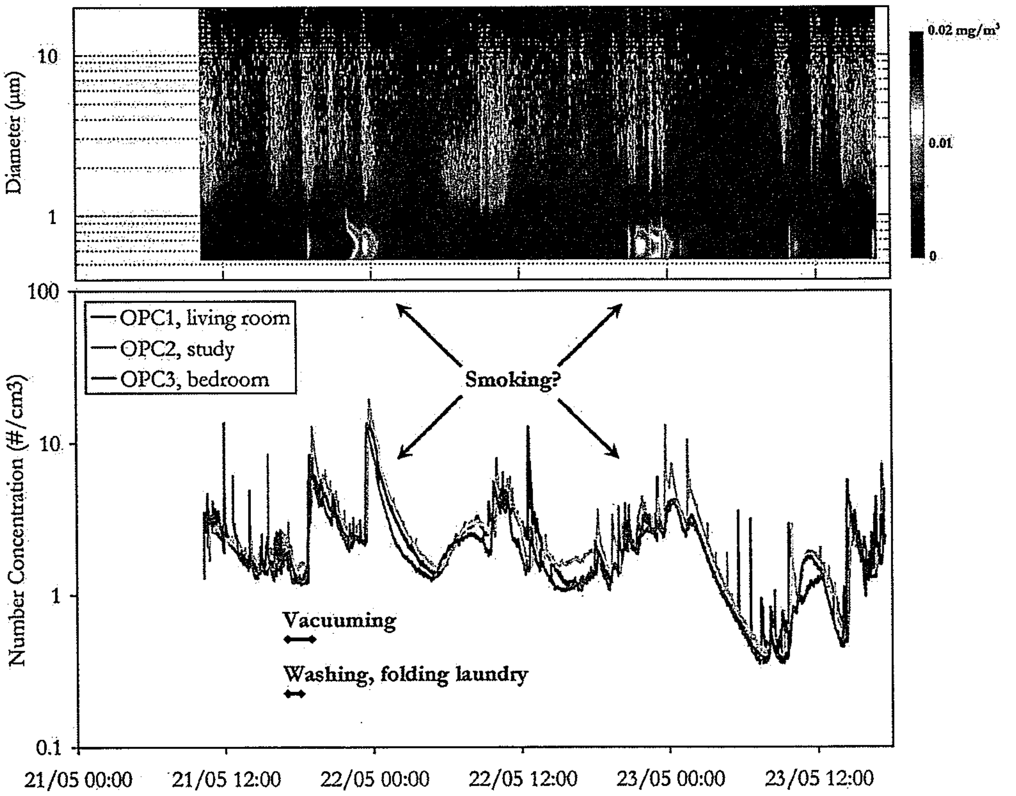

Fig. 2 House A. The upper figure shows the particle mass distribution in the laundry measured by an APS and the lower figure shows the particle number concentration in three different rooms measured by OPCs (0.5–5 µm) during three days and nights. Different activities and sleeping times logged by the members in the family are inserted in the diagram.

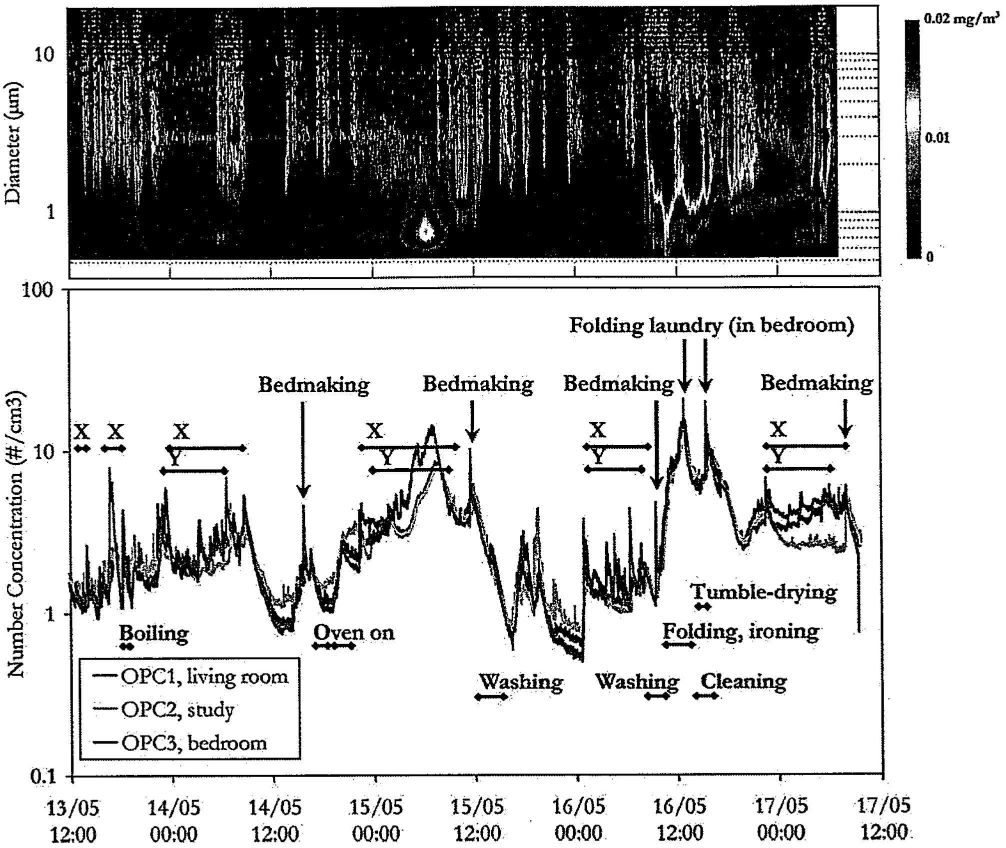

Fig. 3 House B. The upper figure shows the particle mass distribution in the laundry measured by an APS and the lower figure shows the particle number concentration in three different rooms measured by OPCs (0.5–5 μm) during three days and nights. Different activities and sleeping times (*X* and *Y*) logged by the members in the family are inserted in the diagram.

the laundry, which were two times higher in Houses B and C (the houses with dust problems) than in House A. The average number concentration measured in the living-room, study and bedroom (using the OPCs) and in the laundry (using the APS), did not show any significant differences between the houses. Because of this, mean values of the indoor concentrations were difficult to compare without continuous monitoring of the outdoor concentration, which would complicate the study. The indoor mean mass concentrations measured by the APS were in the same range as the regional background of PM_{10}. The regional background concentration was around 7 μg m^{-3} for $PM_{2.5}$ and 13 μg m^{-3} for PM_{10} during the measurement period. The air mass was traced back in direction towards the north-west of Scandinavia. This region has been associated with clean air.[13] The houses were situated in the north-western part of the city which, considering the wind direction, indicates a low city contribution to PM.

The most important information was obtained by comparison of the fluctuations in real time with activities noted by the inhabitants. All the activities related to the handling of textiles were followed by high particle emissions, for instance, when the inhabitants made the beds, went to sleep, got up, or worked with the laundry. Sometimes mass concentrations above 1 mg m^{-3} were reached for short time periods. There were clear differences between day and night. During the daytime, the concentration of particles was more fluctuating and the mass concentration was higher, probably due to increased activity in the houses.

Emission episodes were more noticeable in Houses B and C and were often registered within a short time interval by all instruments in the house, even for particles larger than 5 μm. This indicates a difference in sources between the houses and show that the air is mixing rapidly. The houses have forced mechanical ventilation giving a net flow of air from rooms with supply air ventilators to rooms with exhaust air ventilators. As a consequence there are high correlations between the concentration of small particles measured with the OPCs in the three rooms (living-room, study and bedroom). The correlation coefficient in Houses B and C is about 0.88 for small particles (0.5–5 μm) and about 0.60 for large particles ($\geq$ 5 μm), while the correlation is only 0.20 for large particles in House A (0.85 for small particles). The reason is probably that House A has fewer emission episodes containing large particles.

Fig. 5 shows the OPC particle measurements during a typical emission episode, in this case occurring in House B, 11:30 pm on the 15th of May. The elevated concentration of particles in the bedroom during bed making was followed by an increased concentration in the other rooms in the house with a delay time of about 10 to 15 minutes—typically

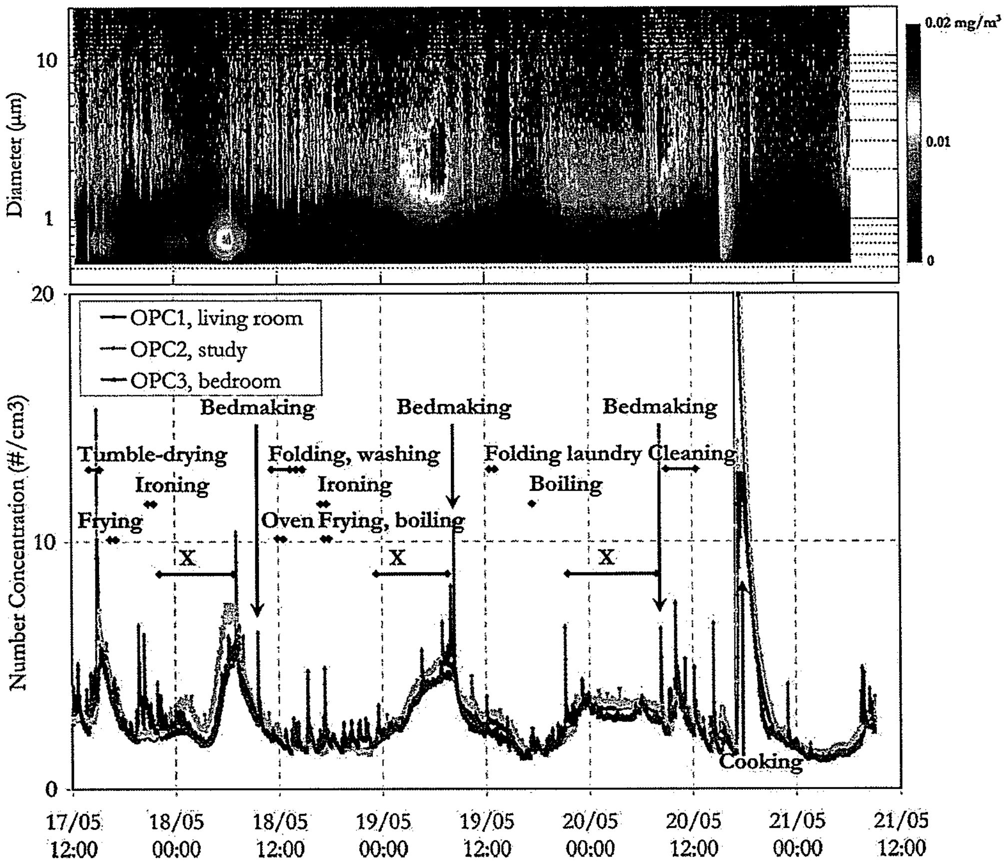

Fig. 4 House C. The upper figure shows the particle mass distribution in the laundry measured by an APS and the lower figure shows the particle number concentration in three different rooms measured by OPCs (0.5–5 µm) during three days and nights. Different activities and sleeping times (*X*) logged by the members in the family are inserted in the diagram.

the time also for other episodes. Equal episodes were observed repeatedly for both small (0.5–5 µm) and large (≥ 5 µm) particles.

Emission episodes were used to estimate the particle rate loss coefficient in the houses. Data from APS measurements in the laundry were analysed as this instrument gives a high particle size resolution. Calculations were made for several episodes in all three houses with approximately the same result. Fig. 6 shows the particle rate loss coefficient as a function of the aerodynamic particle diameter in House B after folding laundry on the afternoon, 16th of May. The shape of the curve and the determined values are equivalent to those from other indoor studies as reviewed by Lai.[14] Assuming a negligible deposition for the particles with the lowest particle rate loss coefficient (about 0.7 µm in diameter), the specific ventilation rate for the studied houses becomes 0.5 to 1.5.

In summary, the following important observations were made from the measurements in the houses:• Particle concentration varies largely over time. During daytime the concentration of particles was more fluctuating and the mass concentration was higher, likely due to increased activity in the houses.

• The APS mass concentrations in the laundry of Houses B and C (the houses with dust problems) were two times higher than in House A.

Table 1 Mean values of three days and nights for the houses studied. The OPC values are the mean value of the concentrations measured in the living-room, study and bedroom. The OPC determines the number concentration in two classes while the APS determines the aerodynamic particle size distribution in the size range 0.5 to 20 µm, fractionated in 52 intervals. Assuming a particle density of 1000 kg m^{-3}, the mass distribution can be estimated

	Mean OPC (living-room, study, bedroom)				APS (laundry)			
	Number conc. 0.5–5 µm/# cm^{-3}		Number conc. ≥ 5 µm/# cm^{-3}		Number conc./# cm^{-3}		Mass conc./µg m^{-3}	
	Day	Night	Day	Night	Day	Night	Day	Night
House A	2.53	2.90	0.16	0.09	9.26	8.67	5.4	3.4
House B	3.15	2.91	0.15	0.08	8.22	7.85	13.2	7.0
House C	2.85	2.68	0.13	0.05	8.20	9.20	10.1	5.0

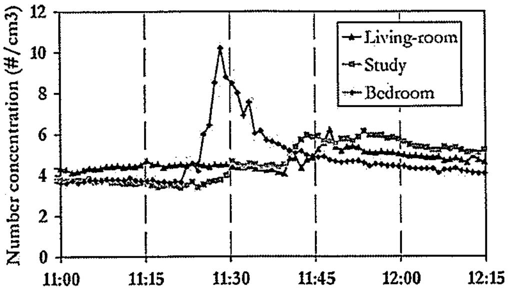

Fig. 5 An example of how particles generated in one room (in this case the bedroom) increase the concentration and is followed by an increased concentration in the other rooms in the house. This emission episode is taken from House B at 11:30 pm, 15 May. The data is obtained from the OPCs and for small particles, 0.5–5 μm.

- Handling of textiles generates particles, for instance, when the inhabitants make the beds, go to sleep, get up, or handle textiles in the laundry.
- The mass concentration reached high values, above 1 mg m^{-3} for short time intervals.
- Emission episodes were more frequent in Houses B and C than in House A.
- Some emission episodes can be seen in the whole house (typically within 10–15 min), indicating that the mixing is rather rapid. The reason is the forced mechanical ventilation in the houses.
- It was not possible to explain all the peaks by observed activities.

Step 3. Laboratory particle measurements

Textile handling was studied further in the laboratory, a conclusion from the visual inspection was that only the houses with dust problems used zeolite detergents, and the measurements showed a correlation between activities related to textiles and exceptionally high number/mass concentrations.

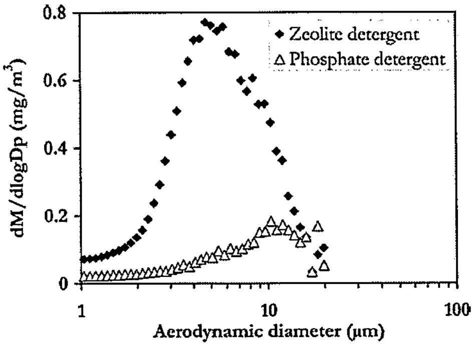

Fig. 7 Particle mass concentrations in the test chamber measured after handling of textiles washed with detergents containing zeolites and phosphates, respectively. The particle size distribution was measured using an APS.

The mass concentrations measured in the test chamber when handling textiles washed with phosphate and zeolite detergent, respectively, are shown in Fig. 7. The textiles washed with the zeolite detergent emitted about 5 times more particles by mass than the others (0.5 mg m^{-3}, respectively 0.1 mg m^{-3}). By number, most of the particles were in the size range 0.1–1 μm. The number concentration was about twice as high for the textiles washed with detergent containing zeolites.

Typical SEM images of the particles emitted from the textiles washed with the two different detergents are shown in Fig. 8. The particle loading was much higher on the filter from the experiment with textiles washed with zeolite detergent. As can be seen on the XEDS graph, these particles typically had high peaks for aluminium (Al) and silicon (Si) and intermediate peaks for sodium (Na) and calcium (Ca) (Fig. 8A). Particles were also found on the filter sampled during handling of the textile washed with phosphate detergent, but these were scarcer (Fig. 8B). These particles often seemed to contain phosphate (P) and calcium (Ca). The two filters had a similar concentration of textile fibres.

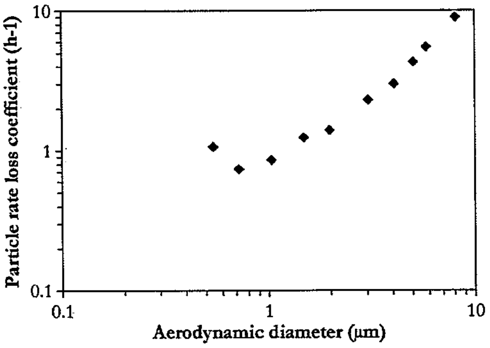

Fig. 6 The particle rate loss coefficient calculated from APS measurements of an emission episode in House B after folding laundry on the afternoon of 16th of May.

Discussion

It cannot be definitely concluded that the perceived dust problems were related to the particles measured with the instruments. However, there are several reasons to assume that the dust originates from textiles containing detergent zeolite residues. Using the described methodology, a number of indications that strengthened this hypothesis were found:

Step 1. Visual inspection

- The households with dust problems used zeolite detergents; the household without problems did not.
- The dust was spread on all surfaces throughout the house, which implies that it arose from relatively small particles.
- No major outdoor particle source was observed.

Step 2. Field particle measurements

- The mean mass concentration in the laundry was twice as high in Houses B and C (dust problems) compared to House A.
- Activities involving textiles in Houses B and C, such as tumble-drying and bed making, give high particle concentrations.
- Emission episodes were more noticeable in Houses B and C.
- The APS particle measurements in the laundry shows high mass concentrations in the 1 to 5 μm size range in Houses B and C when textiles were handled, and about the same sizes

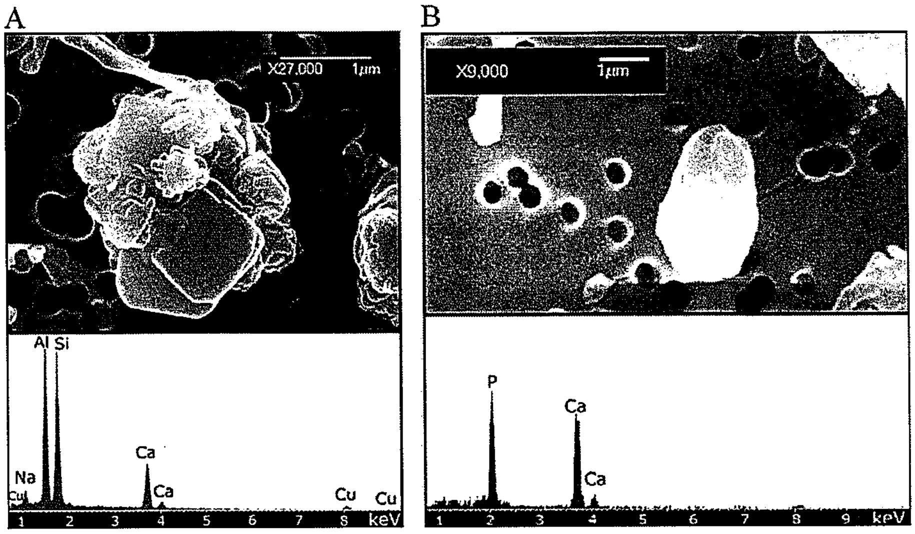

Fig. 8 SEM images of particles sampled during the chamber experiment. (A) A typical particle emitted from textiles washed with zeolite detergent and (B) a typical particle from textiles washed with phosphate detergent.

are reported for zeolite particles in detergents using a Coulter Counter.

Step 3. Laboratory particle measurements

- The study in the test chamber clearly demonstrates the large amount of particles suspended from textiles washed by a detergent containing zeolites. The emissions from these textiles were two times higher by number and about five times higher by mass compared to textiles washed with phosphate detergents.
- The SEM images show clear differences between the two filters. The sample from the textile washed with phosphate detergent mostly contains fibres. The sample from the textile washed with zeolite detergent is loaded with angular particles.
- XEDS analysis of the particles from the phosphate and zeolite textile, respectively, yielded a high amount of P and Ca on the former, and Na, Al, Si and Ca on the latter. The zeolites in detergents consist of synthetic sodium aluminium silicates.

A few short comments might be needed about the laboratory measurements. For the XEDS analysis the particles from both detergents had clear peaks for calcium. The purpose of zeolite and phosphate, respectively, is to soften the water by scavenging calcium and other metal ions. Adsorption of this material on the particles could thus be expected. The size distributions measured in the laboratory and in the houses were difficult to compare. The differences could be due to the brand of the washing detergent, type of textile, particle losses, ventilation, and last but not least that other sources were also present in the houses.

Studies have shown that zeolites remain in clothes after washing. Matthies and Ippen[15] have reported values in the range 0.1–3.5% of zeolites per kg textile after washing. The amount was dependent on the type of washing machine used, composition of the detergent and type of textile. Later studies[16] found residues in textiles of 5%. The Swedish Consumer Agency[17] has reported that modern washing machines have an insufficient rinse in order to decrease water consumption, which results in increased amounts of detergent residues in the washed textiles. Zeolites have increasingly been replacing phosphates as a softening agent over the last two decades and about one million tons of zeolites are used worldwide every year in powerful laundry detergents.[18]

Despite four commercially available instruments, none were placed outside the house and therefore it could be argued that a large local outdoor source increased the indoor levels. However, for these houses we assumed that the outdoor contribution was best given by a measurement close to a ventilator in a room with little other activity. All doors and windows were closed during the study, all ventilators were supplied with filters and the houses were well insulated. The outdoor air would not penetrate into the houses unaffected. Therefore, it was assumed that the studied peaks of the measured indoor concentrations, with values many times higher than the mean concentration, were caused by indoor particle sources. Furthermore, to determine the time dependent outdoor contribution to the indoor concentration it is not enough just to measure the outdoor concentrations. It also requires advanced measurement of the air exchange rate in the different rooms and modelling of the deposition rate and penetration efficiency.[19] It is not a simple task to draw conclusions about the outdoor contribution to the indoor air.

There were further reasons not to do any outdoor measurements in this particular case. First, the visual inspection could not identify any outdoor source, which could possibly explain the high amounts of dust found on both horizontal and vertical indoor surfaces. Second, of three neighbouring houses only two had dust problems—outdoor emissions would affect also the reference house (House A). Third, the dust had been a problem during more than a year and it would most likely have been noted if there was an activity going on close to the houses with such high emissions. Fourth, Wallace and Howard-Reed[20] found in their study of the indoor particle concentration in a townhouse that the concentration was 2 to 20 times higher than average, when an indoor source was operating. The measurements in Step 2 showed that the particle concentration sometimes reached values that were 10–100 times higher than normally measured outside. Even close to a busy road in one of the larger cities in Sweden, PM_{10} seldom is above 50 $\mu g\ m^{-3}$ at the time of year for the study. In two of the three houses the mass concentration could be several mg m^{-3} for periods of some minutes, *i.e.* one to two orders of magnitude higher. During other circumstances a local outdoor measurement could be considered.

It might be possbile, with some modifications, to apply the methodology to indoor environments other than households. In buildings with more people passing by, such as schools, offices or industries, it becomes a formidable task to log relevant activities. Instead of log books, video documentation could be used. High measured peaks could be followed by inspection of the video in search for explaining activities. If a fixed schedule is followed, that could also be a valuable source of information. Some sources, as for example electric equipment, could be monitored with simple sensors registering when they are turned on. In large or badly ventilated rooms the mixing of the air has to be taken into account when placing the instruments.

Several problems have to be solved to extend the methodology for identification of sources to particles below 0.5 μm in diameter. Only the basic procedure of visual inspection, field measurements and laboratory experiments could be helpful. Especially the field study needs to be thought over. To perform parallel measurements with high time resolution is difficult because low-cost optical particle counters are not able to detect particles much smaller than the wavelength of the light. The instruments needed, electrometers or condensation particle counters, are at present more expensive. Maybe alternative techniques, *e.g.* using some chemical characteristic, could be used to track the source. Moreover, the laboratory experiments are not as straightforward. If the particles originate from gas-phase chemistry it could be difficult to isolate a single source in the laboratory. For example, the particles might be formed by nucleation of organic vapours in the presence of ozone emitted from a copier. Often it is also of interest to quantify the sources and not only identify them. This requires a more extensive statistical analysis and is beyond the scope of this work. The best approach to handle these problems is a question for continued methodological discussion and future research.

Conclusion

With the suggested methodology it was possible to identify one probable major indoor particle source—textile handling. To the best of the authors' knowledge, this has not been considered before. Valuable in the source examination was, among other things, knowledge about differences in concentration between different rooms, real-time measurements compared to activity reports from the inhabitants, information on particle size distribution and simulation of activities in the laboratory. The methodology will be used in future investigations of buildings where dust problems are reported.

The lowest adverse effect level (LOAEL) has been determined to be 1 mg m^{-3} for the most common detergent zeolites.[16] Thus, the dust concentration in the two houses with zeolite detergents is close to the margin-of-safety (MOS = 100). Considering that a major part of the European population uses such detergents, a potential health risk is implied.

Acknowledgements

This work was partly financed by The Building and its Indoor Environment, a research school at Lund University (financed by the Knowledge Foundation), the Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS) and the Development Fund of the Swedish Construction Industry (SBUF). We thankfully acknowledge Dr Jakob B. Wagner and Mrs Gunnel Karlsson at the Department of Chemistry, Division of Polymer and Materials Chemistry, Lund University, for the SEM analysis.

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Abstract, accepterat som bidrag till
European Aerosol Conference 2008, Thessalonikia

Sources of fine particles in a residential apartment

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Keywords: indoor particles, indoor air quality

There is an increasing interest in fine particles (particles smaller than 2.5 μm in diameter) found indoors, as it has become apparent that indoor sources periodically can generate particles that in numbers and mass concentrations frequently exceed ambient levels (He et al 2004, Abt et al., 2000). Considering also the fact that we tend to spend 65 % of our time in homes (Leech et al., 2002) information on particle characteristics found in residential homes is of great importance for health effects assessments. The aim of this study was to investigate characteristics of particles found in a residential apartment, indentify sources of the particles and estimate their strengths.

Measurements were performed for ten consecutive days in February 2007 in a residential apartment in Malmö city (Sweden). A log book of occupants' activities during the measurement period was created for the identification of particle sources. Measurements were recorded continuously with the following instruments: an aerodynamic particle sizer TSI APS 3321, a scanning mobility particle sizer (SMPS), (consisting of differential mobility analyzer (TSI DMA) and condensation particle counter (TSI CPC 3010)), and two DustTracks (model 8520 TSI Inc.) The two DustTracks were used for indirect mass concentration measurements of particles smaller than 2.5μm ($PM_{2.5}$) to ~0.1 μm, they enabled simultaneous measurement of indoor and outdoor mass concentration. $PM_{2.5}$ was also collected on quartz filters for organic and elemental carbon analysis.

Identified sources of fine particles consisted of different types of cooking (frying sausages, onions, vegetables, chicken, omelette, pizza in the oven), burning candles, burning incenses, peeling oranges and tangerines, plugging in air freshener. Only a few of these activities led to a significantly increased mass concentration, namely: frying vegetables, frying chicken, cooking and heating pizza in the oven. The highest number peak contribution (Fig. 1) was observed at party dinner, with vigorous cooking followed by 7 candles burning. Typically a sharp increase in particle number concentration at the beginning of each indoor activity was observed. When the activity was ceased slow exponential decrease followed. The decrease to background (no source) level in the apartment took up to 12.5 hours, therefore elevated concentrations of particles were observed for long period of time. Average particle number concentration during an active source period was about 13 times higher than during no source period (15300 and 1200 particles cm^{-3}, respectively). Active source period was characterised by smaller average number geometric mean diameter of 82 nm in comparison to 95 nm during no source time. Estimated total source strengths for cooking events range from 5.5×10^{11} to 1.5×10^{12} particles min^{-1}. The highest contribution is observed from ultrafine particles (<100 nm), their source strengths range from 5.1×10^{11} to 1.1×10^{12} particles min^{-1}, which accounts on average for 84 % of total generated submicron particles. Peeling tangerines produced 3.5×10^{10} particles min^{-1}, where ultrafine particles accounted for 90%.

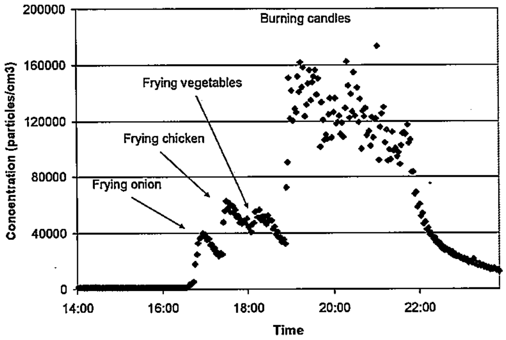

Figure 1. Particle number concentration during party

Mass and number concentrations of particles in residential apartment can vary to a great extent and these variations depend on numerous factors. To state conclusively typical levels of particles in residential homes more data is needed as this study is limited to just one location at a specific time. Although the results suggest that in order to decrease personal exposure to particles from indoor sources, efforts should be made to enhance ventilation at least in locations where indoor particles are generated frequently, namely in the kitchen.

This work has been supported by the Swedish Research Council FORMAS and the Development Fund of the Swedish Construction Industry (SBUF)

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Abstract, accepterat som bidrag till
Indoor Air Conference 2008, Köpenhamn

Fine Particles in Various Swedish Indoor Environments

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Keywords: fine particles, indoor environments

1. Introduction

The aim of this study was to gain information about fine particles (particles with diameter smaller than 2.5 µm, $PM_{2.5}$) loads and their characteristics in various Swedish indoor non-industrial environments, namely: in two schools, fast food restaurant, supermarket, residential apartment close to a busy road situated in a city and residential house in a remote area. Assessed particles' characteristics are: number and mass concentrations, size distributions and organic and elemental carbon content.

2. Materials and methods

Measurements were performed for seven consecutive days in each location during winter time 2006 and 2007. Log books of activities during the measurement period were created for the identification of particles' sources. Measurements were recorded continuously with the following instruments: time-of-flight instrument, differential particle sizer, condensation particle counter and two photometers. Additionally alternate measurements of indoor and outdoor particles were carried out. The two photometers enabled simultaneous indirect mass concentration measurement of indoor and outdoor particles. Particles were also collected on quartz filters for organic and elemental carbon analysis.

3. Results

Median approximation of $PM_{2.5}$ mass concentration (photometer readings) varied between the sites from 6 to 25 $\mu g/m^3$ (with min and max values of 2 and 403 $\mu g/m^3$, respectively. Median particle number concentration varied between 600 and 2000 particles/cm^3 (min 100 and max 173000 particles/cm^3). Indoor sources that generated particles were identified as cooking, burning candles, burning incenses, using air fresheners, cleaning products (due to terpenes - ozone reactions) (Wierzbicka et. al., 2008). Values which are representative for personal exposure while indoors to both indoor generated and infiltrated from outdoors particles were recalculated using only the time when occupants were inside the apartment and house. Thus occupancy time average particle number concentration in the apartment and house accounted for 10200 ± 24600 and 2300 ± 2700 particles/cm^3, respectively and average $PM_{2.5}$ mass concentration in the apartment was 36 ± 51 $\mu g/m^3$. When the indoor sources were active in the apartment, number concentrations were about 13 times higher in comparison to average background concentration with no active source (1200 particles/cm^3).

4 Conclusions

Mass and number concentrations of particles in different indoor environments vary to a great extent and these variations depend on numerous factors. To state conclusively typical levels of particles in various indoor environments more data is needed as this study is limited to just a few locations at specific time. Although the results suggest that in order to decrease personal exposure to particles from indoor sources efforts should be made to enhance ventilation at least in locations where indoor particles are generated frequently, namely in the kitchen.

This work has been supported by the Swedish Research Council FORMAS and the Development Fund of the Swedish Construction Industry (SBUF).

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Preliminärt manus till vetenskaplig artikel. Kommer att ingå i Aneta Wierzbicka doktorsavhandling, som ska förvaras den 25 april 2008. Tryckning av avhandling 13 mars.

Characteristics of Airborne Particles in Various Swedish Indoor Environments

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PRELIMINARY VERSION

Characteristics of Airborne Particles in Various Swedish Indoor Environments

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Abstract

The aim of this study was to gain information about airborne particle in various Swedish indoor environments, namely in: two schools, a supermarket, a restaurant, an apartment and a house. Assessed particle characteristics were: number and mass concentrations, size distributions and organic and elemental carbon content. Continuous measurements for 7 consecutive days in each location were conducted during winter time 2006/7 using a time-of-flight instrument (APS), a scanning mobility particle sizer (SMPS) consisting of a differential mobility analyser and a condensation particle counter, and two photometers (DustTrak).

Median mass concentration (DustTrak readings) varied among the sites between 6 and 25 µg m^{-3}. In the apartment the short-term peak mass concentrations were observed up to 400 µg m^{-3} (about 20 times higher than the ambient concentrations), and were due to cooking and candle burning. Median values of submicron PNC for all studied locations were similar, between 600 and 2000 particles cm^{-3}. An indoor source of particles in the supermarket was observed, possibly due to local emissions of terpenes (from washing powders, cleaning products and air fresheners), which in presence of ozone from outdoors form particles due to gas-to-particle conversions.

During occupancy time (occupants inside) average submicrometer particle number concentration (PNC) in the apartment and house were 10200 ± 24600 and 2300 ± 2700 particles cm^{-3}, respectively and average PM2.5 mass concentration in the apartment was 36 ± 51µg m^{-3}. These occupancy time averages represent personal exposure values while indoors due to both indoor generated and infiltrated from outdoors particles. Duration of elevated PNC (due to indoor activities) ranged from 5.5 to 12.5 hours. Average PNC during active source period was about 13 times higher than during no source period (15300 ± 29600 and 1200 ± 300 particles cm^{-3}, respectively). Contribution of ultrafine particles (< 100 nm) to total submicron PNC accounted for 75 % and 50 % during active and no source period, respectively. About 94 % of total submicron PNC consisted of particles smaller than 300 nm, for both active and no source period. Estimated total source strengths for cooking events range from 5.5×10^{11} to 1.5×10^{12} particles min^{-1}.

Introduction

In numerous epidemiological studies ambient fine particulate matter PM2.5 (particles with diameter smaller than 2.5 µm) has been associated with cardiovascular and respiratory morbidity and mortality (Pope III, 2000; Pope C.A. III et al., 1995; Dockery et al 1993). Recently focus has been put on exposure to particulate matter in indoor environments, as it is known that we spend majority of the time indoors (up to 90 % Leech et al., 2002; Brasche and Bischof, 2005; Klepeis et al., 2001). Poor correlations were found between ambient PM2.5 and personal exposure, as not only ambient PM sources contribute to personal exposure but also indoor, work related, invehicle sources and personal activities (Wilson and Brauer, 2006; Wallace et al., 2006; Meng et al., 2005; Wallace and Williams, 2005; Burke et al., 2001). Particles emitted from different sources have different physical characteristics (e.g. number, size distribution, surface area) and chemical composition. Therefore, it might be that particles generated indoors have different health effects than those of ambient origin.

Indoor particles consist of ambient particles that infiltrated indoors, particles emitted indoors and particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Meng et al. 2005, Uhde and Salthammer, 2007, Morawska and Salthammer, 2003). More information is available on outdoor particles characteristics i.e. concentration, size distributions, emission factors for specific sources etc, while data on indoor particle characteristics remains limited.

Measured values in indoor environments are influenced by many factors such as building characteristics (type of ventilation, tightness of the building envelope, airing practices), geographical location (outdoor particle concentrations, climate), type of interiors (carpeted floor, amount of textile decorations, etc.), type of cooking appliances, human activities and hobby, cleaning practices, other practices (burning candles, incenses etc.) which all vary to a great extent.

Indoor sources of fine particles were identified as tobacco smoking, cooking (frying, grilling, baking, boiling water, making toasts etc.), use of gas and electric stoves, gas-powered clothes dryers, candle and incense burning, use of photocopiers, laser printers, use of cleaning products and air fresheners containing terpenes (which in presence of ozone form secondary aerosols) (Ogulei et al. 2006; Wallace 2006; Danekamp et al., 2001; He et al., 2005). Indoor sources of coarse particles were identified to come from sweeping, hovering, dusting, people movement, resuspension from clothes, carpets, washing powder residues (zeolite) (Ogulei et al., 2006; Abt et al., 2000; Gudmundsson 2007).

There are many studies based on integrated particle mass concentration measurements (filter measurements) but studies investigating continuously changes in particle mass and number are limited. Scattered information on particle number concentrations and emission factors for indoor sources can be found in several publications but the available data is far away from being concise and complete (Ogulei et al. 2006; Hussain et al., 2005; Morawska et al., 2003; He et al., 2005; Wallace 2006; Wallace 2005; Wallace et al., 2004; Danekamp et al., 2001).

Detail characterisation of particles found indoors is of interest for epidemiologic and toxicological studies and to guide development of control strategies as well as raise general awareness.

The aims of this study were: 1) To gain information about airborne particle loads in various Swedish indoor environments, 2) To assessed indoor particle characteristics by means of: number and mass concentrations, size distributions and organic and elemental carbon content, 3) To identify indoor sources of particles and estimate their source strength.

Materials and methods

Sampling sites

The sampling sites consisted of: two schools A and B, a supermarket, a restaurant, an apartment and a house, all situated in southern Sweden. Particle measurements were performed for at least seven consecutive days in each location during winter time (February 2006 and October - March 2007). In Table 1 sampling duration, type of ventilation and measured air exchange rates are summarised for all sampling sites. Activities were recognized using information from log books created by occupants of the apartment and house and further clarified within a week after the measurements, with a support of preliminary analysis. In case of the schools, supermarket and restaurant, time schedules and discussions with employees were used to identify various activities.

School A is an elementary school situated in residential area on the outskirts of town (Lund, Sweden). It is one-level building, built in 1970s with 4 regular classrooms, 3 study rooms, and a play room. Total volume of the building was ~ 900 m^3, with a floor area of ~ 300 m^2. In the mechanical ventilation system electrically charged synthetic filters of F7 class (EN 779: 2002) are used. Ventilation works during weekdays between 5:00 and 18:00. The measurements were performed in a play room with some textiles (upholstered sofa, few cushions, small carpet) and numerous open shelves with accessories. Additionally many activities took place there, such as games, puzzles, handcraft etc., and all of them were connected with considerable movement and children' activity.

School B is a secondary school situated within the same area as school A. It is a two- level building, built in 1997. There are 8 classrooms. Total volume was ~ 3000 m^3, with a floor area of ~ 1000 m^2. Measurements were performed in a classroom where regular classes took place with lower activities and movement intensity in comparison to school A. Additionally smaller amounts of open shelves and nearly no textiles (only curtains), were in the classroom in comparison to site in school A.

Supermarket is situated on the outskirts of town (Lund, Sweden), close to a busy street within the shopping area. Total volume of the supermarket was ~ 23400 m^3, with a floor area of ~ 3600 m^2. Mechanical ventilation supplies heated and filtered air, the system is equipped with glass-fibre filters of F7 class (EN 779: 2002). Ventilation on weekdays works between 4:30 and 22:00 and during weekends between 5:00 and 20:30. Measurements were performed within the area with washing powders, cleaning products and air fresheners.

Restaurant was a fast food restaurant, situated in the centre of town (Lund, Sweden) Total volume of the restaurant was ~ 2400? m^3 with a floor area of ~ 600? m^2. Mechanical ventilation system is equipped with glass -fibre filters of F7 class (EN 779: 2002). The ventilation works 24 hours per day, seven days a week. Measurements were performed in a dining area, with ... sitting places.

Apartment was situated on the third floor of the residential building, close to a busy street in an urban centre in Malmö (Sweden). The volume of the apartment was ~ 281 m^3, with a floor area of 108 m^2. The apartment's layout is given in Figure 1, where the placement of the enclosure with instruments is marked. Occupants had modified the original layout of the apartment, they took down the wall between kitchen and living room (marked with dashed line in Fig 1). They had also eliminated one of the ventilation outlets in one of the bedrooms, thus most probably the air exchange rate (AER) was reduced. Most of the time the windows were kept closed due to low outdoor temp (daily mean temperature between 0.5 and 3.5 ^0C). Two non-smoking, employed adults lived in the apartment during the measurements. On one occasion dinner party was organised with 6 invited guests. An electric stove was used for cooking. Even though an extraction hood above the oven was installed occupants did not use it. Hot water and heat was supplied from district heating.

House was situated in a remote area within rural surroundings. It was a three-level (basement, first and second floor) stand-alone villa. The volume of the house was ~ 780 m^3, with a floor area of ~ 320 m^2. The measurements were performed on the second floor close to the staircase. Two non-smoking adults lived in the house. Due to low outdoor temperatures windows were also kept closed (daily mean temperatures between -5 to 1 ^{0}C). Electric stove was used for cooking. A boiler fired with wood logs was used for heat and hot water supply in the house.

Instruments and analysis used

Due to safety precautions and aim to minimize the disturbance in occupants' daily activities, instruments for particle measurements indoors were placed in specifically designed enclosure. The enclosure was insulated to reduce the noise from instruments and pumps and had an inbuilt system of fans to lower the temperature. Placement of the enclosure with the instruments at different sampling locations was dependant on site-specific limitations, occupants' preferences and the aim to cause the least possible disturbance in daily activities.

All measurements were recorded continuously for the entire duration of sampling. Instruments placed in the enclosure are illustrated in Figure 2 and consisted of:

APS - aerodynamic particle sizer (APS 3320, TSI Inc., USA) was used for the determination of number concentration and size distribution of particles in the 0.7 – 20 μm size range.

DMA + CPC - a differential mobility analyzer (Long Column DMA, TSI Inc., USA) and condensation particle counter (CPC 3010, TSI Inc., USA). These two instruments working together form a scanning mobility particle sizer (SMPS 3934, TSI Inc., USA), which was used for measuring number concentrations and size distribution of particles between 15 – 700 nm. Particles within this size range are referred as submicrometer particles in this article. The SMPS hardware was controlled by a custom-made computer program, which enables measurement on both up and down scan and includes corrections for the smearing of the CPC signal (Collins et al. 2002) and diffusional losses in DMA (ref to Bengts paper?). A scan time of 180 s was used, aerosol flow was 0.3 l min^{-1}, sheath air 3 l min^{-1}. Corrections were made for particle losses in the inlet tubing and in the DMA. A valve was incorporated into the sampling system, which after every second SMPS scan switched the sampling air between indoor and outdoor, or indoor and supplied ventilation air. Time lag of 13 s, after switching the valve, was incorporated. Placement of the enclosure at different sampling sites dictated possibility of these alternate measurements. A list of these configurations is given in Table 1.

DustTrak - a photometer (DustTrak 8520, TSI Inc., USA) was used for indirect mass concentration measurements of particles smaller than 2.5 μm (PM2.5) down to 0.1 μm (see Corrections for DustTrak). Additionally in locations, where it was technically feasible, second photometer (the same model) was placed on the roof of the building for simultaneous outdoor concentrations monitoring (see Table 1). In the apartment simultaneous DustTrak measurements were additionally supported by TEOM's PM 2.5 mass concentration readings from the monitoring station situated about 2 km away from the apartment (within Malmö city).

OC EC line - a set-up for PM 2.5 collection for organic (OC) and elemental (EC) carbon analysis consisted of pre-collection cyclone with a cut off diameter of about 2.5 μm, set of quartz and Teflon filters, and a pump. Detail description of the set up was published elsewhere (Wierzbicka et al, 2004). In short, method with an additional parallel line, where Teflon filter followed by quartz backup filter, was used for the estimation of the positive artefact (amount of gas phase organic vapours adsorbed onto quartz filter) (Turpin et al., 2000). All samples were collected for 24 h, with an exception of a sample from the house which was collected for 72 hours. A thermal-optical method, using a carbon analyser system developed by Sunset Laboratory Inc., was used for OC and EC analysis (Johnson et al., 1981; Turpin et al., 2000; Viidanoja et al., 2002).

Air exchange rate (AER) was measured at each sampling site by tracer decay method using sulphur hexafluoride (SF_6) as the tracer gas. Measurements of AER were not performed at the same time as the particle measurements due to some technical problems. Outdoor temperature during AER measurements was slightly higher (daily mean between 12 and 15 ^{o}C) in comparison to actual particle measurements at sites with natural ventilation (-5 to 5 ^{o}C), therefore the AER could be slightly underestimated, as the temp gradient influences the natural ventilation. An Innova 1312 photoacoustic multi-gas monitor together with Innova 1303 multipoint sampler and doser were used for measuring SF_6 concentration levels. One sampling inlet was placed exactly were the particle monitoring enclosure was situated, and 2 to 4 inlets (depending on sampling site) in different parts of the sampling site. SF_6 was dosed up to level of around 10 ppm, a fan was used to help mixing for several minutes, after switching it off the distribution across the sampling site was checked by monitoring the level of SF_6 in few locations. When even distribution of the tracer gas was achieved the decay process was monitored only in the previous enclosure location. The instrument's detection limit for SF_6 was 0.009 ppm. During the AER measurements in the apartment and house conditions which were representative for the duration of the particle sampling period were kept, i.e. windows closed, while doors between the rooms were opened. In schools frequent doors and windows opening occurred during the duration of particle measuring, determined AER are for the low ventilation conditions with the windows and doors closed. In the supermarket and fast food restaurant it was not possible to control the ventilation conditions (i.e. doors opening), as the measurements were performed during normal opening hours of the sites, therefore average value from a few measurements is given.

Corrections for DustTrak

DustTrack operates on the principle of light scattering, its response is highly depended on aerosol material (refractive index) and on the size distribution of the sampled aerosol. The instrument was factory calibrated with Arizona dust particles and it was reported previously that for fine aerosols commonly encountered in indoor environments the instrument's response might be significantly higher than the true PM2.5 mass concentration (Ramachandran et al., 2000; Morawska et al., 2003). Therefore correction factors are needed. Correction factors used in previously published studies were derived from comparisons based on ambient particles (Morawska et al. 2003; Kingham et al., 2006). There were two studies conducted indoors but with no active indoor sources (Heal et al., 2000; Yanosky et al., 2002). We have conducted laboratory comparison of DustTrack and TEOM mass readings, using typical indoor sources such as candles, incenses and frying onions, as the loadings and variations in lower range of submicrometer particles (close to 100 nm) indoors are greater than those encountered outdoors in the studied Scandinavian locations. *Whereas DustTrak's lower sensitivity in this size range was reported previously (Morawska et al., 2003).* SMPS and APS measurements were performed simultaneously to obtain information on number concentration and size distribution of generated particles. It was observed that with the increase of number geometric mean diameter (GMD) of generated particles from 130 to 300 nm the DustTrak's over-reading factor ($C_{PM2.5\ DustTrak}/C_{PM2.5\ TEOM}$) decreased from 6 to 1.8. During events when concentrations were dominated by ultrafine particles, which are outside the DustTrak's detection range, two different scenarios were observed: a) when the particle number concentration was about 10^5 particles/cm^3 with number GMD of 51 nm over-reading factor was 1.3 (only tail of the number size distribution was within the DustTrak detection range; b) when the particle number concentration was about 10^6 particles/cm^3 with number GMD of 37 nm, values given by DustTrak underestimated TEOM values ($C_{PM2.5\ DustTrak}/C_{PM2.5\ TEOM} \approx 0.2$).

During our indoor measurements ultrafine particles number concentrations, while the indoor sources were active, were about 10^4 particles/cm^3 (with one occasion reaching values 10^5) therefore underestimation of PM2.5 mass concentration by DustTrak seems not to have an influence. Derived average factor for indoor environments used for correction of PM2.5 mass concentration measured by DustTrak equals 0.49 ($C_{PM2.5\ TEOM}/\ C_{PM2.5\ DustTrak}$). Obtained value is slightly higher in comparison to the one in regression equations estimated on the basis of ambient particles by Morawska (2003) PM2.5(TEOM) = 0.394 PM 2.5 (DustTrack) + 4.450.

Additionally ambient DustTrak's readings were assessed by placing the instrument on the roof of the background monitoring station in Vavihill (Southern Sweden), and compared with the TEOM PM2.5 values. Derived correction factor equals 0.29, and was used for all DustTrak measurements performed outside.

Additionally the difference in readings by the two used DustTraks was assessed and accounted for ± 2 μg/m^3. Precision for the instrument given in manufacturer's specifications accounts for ± 1.5 μg/m^3 (1-hour average).

Nevertheless all presented DustTrak readings even after correction remain only approximation of the PM2.5 mass concentration as the instrument was not calibrated for all the indoor-specific aerosols studied, and use of correction factor can give only approximate values. DustTrak seems to be useful to detect changes in mass concentration of particles with diameters within its detection range, although its readings for heterogeneous aerosols even after applying correction factors should never be treated as absolute mass concentrations values.

Source strength estimations

The mass balance differential equation describing the particle concentration indoors (Koutrakis et al., 1992) used by other researchers for source strength calculations (He et al., 2004; Wallace et al, 2004) was employed:

$$dC_{in}/dt = PaC_{out} - (a + k)C_{in} + S/V \qquad (1)$$

where C_{in} and C_{out} are indoor and outdoor particle number concentration (particles cm^3), respectively, P is a penetration factor (h^{-1}), a is AER (h^{-1}), k is a deposition rate (h^{-1}), S is source strength (particles h^{-1}), V is a volume of the apartment (m^3).

Simplified solution for S to the equation (1) given by Wallace et al. (2004) was used:

$$S = V(a + k)\Delta C_{in}/\{1\text{-}\exp(-(a+k)t\} \qquad (2)$$

where following assumptions where made: 1) equation (1) refers to a particular aerosol size, where P, k and S may all be functions of particle size, 2) coagulation and condensation processes were neglected, 3) constant outdoor concentration during the active indoor source episode and subsequent particle decay, 4) constant values for P, a, k and S during the active indoor source time, 5) apartment is a single well mixed zone with instantaneous mixing, 6) the conditions, at the beginning of the active indoor source event, have held long enough before that to reach equilibrium.

Criteria for selecting an active indoor source episode for source strength calculations, adopted from Wallace et al. (2004) were: 1) a sharp increase in particle number concentration above average background (no active source) concentration level, 2) no other reported activity at the time and a smooth decay (indicating no other significant active indoor source), 3) a return to initial (before the episode) concentration level indicating no change in outdoor concentration.
These criteria limited active source episodes in the apartment, suitable for the source strength calculations to 8. For ΔC_{in} calculation, initial background concentration (15 min average preceding first observed concentration increase) and peak concentration for the given active source episode were used. This determined the maximum concentration change and allowed estimation of average source strength for this period on the basis of equation (2).

Decay rates where determined for given particle size intervals from equation that describes the decay of particles indoors:

$$C_{(t)} = e^{-(a+k)t} C_{(t-1)} \qquad (3)$$

where $C_{(t)}$ and $C_{(t-1)}$ are the indoor concentrations for given particle size interval at times t and t-1. Decay rates are determined by taking the natural logarithm of both sides of the equation (3) and regressing over time. The negative slope of the regression is a + k, i.e. it is a total decay rate due to exfiltration and deposition. The decay curves lasted between 5.5 to 12.5 hours, on average about 100 data points were used for the decay rate calculations.

Results and discussion

Indoor/outdoor DustTrak measurements

Simultaneous measurements of indoor and outdoor approximation of PM2.5 mass concentration were carried out with the use of two DustTraks in three sampling locations (see Table 1). One DustTrak was placed indoors within the monitoring enclosure and the other on the building's roof.
Simulataneos DustTrak measurements in the apartment were additionally supported by TEOM's PM 2.5 mass concentration readings from the monitoring station situated about 2 km away from the apartment (within Malmö city). The obtained results are shown in Figure 3. Peak mass concentration contributions from indoor sources are clearly visible, reaching values up to 20 times higher in comparison to observed outdoor concentration. These short term peaks were identified, with the help of occupant's logbooks of activities, as cooking and candle burning. Similar findings were reported previously: Abt et al. (2000) reported cooking among activities contributing to short-term peak indoor volume concentrations, which were 2 - 33 times higher than background and outdoor concentrations; Wallace et al. (2004) reported that levels of PM2.5

were increased during cooking by factor of 3; He et al. (2004) reported increased PM2.5 over the background level by 89 and 32 times during grilling and frying, respectively; Morawska et al. (2003) reported increased PM2.5 during cooking activities up to 100 times higher than the background levels.

In school B, with natural ventilation, PM2.5 mass concentration measured by DustTrak inside showed always lower concentrations than the DustTrak placed outside on the roof of the building (not shown here). During weekdays clear pattern inside with increased PM2.5 mass concentrations to about 10 μg/m^3 during hours when pupils were at school could be observed, outdoor concentration were about 15 μg/m^3 at the time. Strong influence of outdoor concentration on indoor measured values was observed, when the outdoor concentrations increased to about 30 μg/m^3 increased inside concentrations were observed of about 15 μg/m^3. This could be explained by enhanced infiltration, probably due to increased AER as doors and windows were frequently opened. During weekend influence of outdoor concentration on indoor measured values was decreased, possibly due to decreased AER (all windows and doors closed).

In fast food restaurant DustTrak inside by mistake measured PM1 mass concentration, which was very stable throughout sampling duration with median value of 8 μg/m^3. Influence of outdoor concentrations was not noticed. This indicates high removal efficiency of used filters in the ventilation system.

DustTrak readings (comparison of sampling sites)

In Table 2 DustTrak readings, which give approximation of PM 2.5 mass concentrations, are summarised for the studied indoor environments. Median values were calculated for opening hours in the supermarket and restaurant, teaching hours in the schools and entire duration of measurements in the apartment and house. Highest median value was observed in the school A (25 μg/m^3), the lowest in the supermarket (6 μg/m^3).

There is a significant difference between median values for the two studied schools A and B (25 μg/m^3 and 8 μg/m^3, respectively). As a strong influence of ambient PM2.5 on indoor values was observed and discussed earlier, outdoor and indoor values were compared. Different outdoor PM2.5 mass concentrations during measurements in schools A and B was observed (median for corresponding days and hours 16 and 9 μg/m^3, respectively). During measurements in school A simultaneous measurements were not performed, instead mass concentration was obtained from monitoring background station (TEOM). After subtracting median outdoor concentration from median indoor concentration the difference between schools A and B was still about 9 μg/m^3. This difference possibly could be explained by differences in available dust reservoir and pupils' activities influencing the resuspension of particles. School A is an elementary school where younger children attend, the sampling site was a play room with more textiles (sofa, few cushions, small carpet) and numerous open shelves with accessories additionally many activities took place there, such as games, puzzles, handcraft etc., and all of them were connected with considerable movement and children' activity. School B is a secondary school with older pupils, with much smaller amounts of open shelves and nearly no textiles (only curtains), in the studied classroom regular classes took place with lower activities and movement intensity in comparison to school A. Therefore the higher PM2.5 mass concentration in school A possibly could be attributed to larger amounts of dust available for resuspension and higher movement intensity and activity, which in turn resulted in more resuspended particles. The following conclusion can be supported by observed differences in mass size distribution calculated on the basis of APS measurements assuming 1000 kg m^{-3} density presented in Figure 4. An hour average mass size distribution is plotted for the school A and B during the times when pupils were in the classroom and when there was no one. When pupils are not in the classrooms higher mass concentration is observed for supermicron particles in school A which would indicate dustier location (introduced air movement due to mechanical ventilation could contribute to particle resuspension in this case). Increase in supermicron particles mass concentration is observed in both schools during the time when pupils are in the classrooms. Similar results were reported by Ward et al. (2004) see Table 3 for details, where difference in measured PM2.5 was seen between elementary and middle school, authors attributed it to sampler position and dustier site in elementary school. Sampler in elementary school was placed in gymnasium, which definitely is a place with increased pupils' movement and activity, which could have been a contributing factor to observed higher concentrations. Obtained values for both schools in this study are comparable to other reported measurements (summarised in Table 3) which are in range from 6 to 37 μg/m^3 with min and max from 3 to 54 μg m^{-3}. When comparing reported PM 2.5 mass concentrations (given in Table 3) one has to bear in mind that frequently different instrumentation and analytical methods were used, therefore given values should be interpreted with caution.

Measured 18 μg/m^3 PM2.5 median value in the apartment is within the range of values reported in other publications i.e. between 8 and 30 μg/m^3 (see Table 3 for references.) Given values in Table 3 comprise only results for non-smoking residences, even if many of the studies were conducted for both smoking and non-smoking ones. Smoking residences were excluded as tobacco smoking is a known major source fine particles, which was reported to increase PM2.5 mass concentration by 58 – 130 % (Stranger et al, 2007, Breysse et al., 2005) therefore care was taken to eliminate this source in comparisons. The very high maximum concentration of PM 2.5 in the apartment (403 μg/m^3) was due to cooking and candle burning during party dinner, and this value can be compared to median peak concentration values due to cooking reported by He et al. (2004) where even higher concentrations were reported i.e. 745, 735, 718 μg/m^3 median peak concentration due to frying, cooking pizza and grilling, respectively.

In the restaurant PM_1 was measured, authors did not find any reference value for PM_1 concentrations measured in restaurants. $PM_{2.5}$ values given by Travers et al., 2004 for 20 bars and restaurants after the smoking prohibition implemented in 2003 in USA are higher (24 μg/m^3). Many reference values were found for Chinese style of cooking but values reported there during cooking activities were much higher 312 and 1167 μg/m^3 in See and Balasubramanian (2006) and Lee et al. (2001), respectively, it could be due to not adequate ventilation system (natural ventilation in the first study, no information on AER in the latter), different type of cooking (oil heated to high temperatures), gas appliances and it was not clear if there was a smoking restriction. As it was mentioned before, low mass concentration in the studied restaurant could be attributed to effective ventilation system working 24 hours per day.

Authors did not find any reference value for PM2.5 mass concentration measured in the supermarket.

Submicron particle number concentrations in indoor/outdoor and indoor/supplied ventilation air

In mechanically ventilated school A submicron particles inside, measured by SMPS system 15 - 700 nm, mirrored the outdoor number concentration and size distribution (not shown here. In naturally ventilated school B a time lag was observed, although submicron particle number concentration (PNC) showed the same pattern (i.e. indoor fluctuations followed outdoor ones). This could be explained due to lack of indoor sources of submicron particles in schools (no cooking, candle burning, tobacco smoking strictly prohibited etc.). In school B due to low AER once indoor submicrometer concentration increased, the time required for the concentration to decrease was longer in comparison to the observed outdoor concentrations decrease. This could explain the slightly higher indoor submicrometer PNC in school B (median 1500, min 700, max 3500 particles/cm^3) in comparison to outdoor values (median 1200, min 500, max 3600). In school A median submicrometer PNC was the same indoors and outdoors 1600 particles/cm^3, ranging indoors from 800 to 3100 particles/cm^3, and outdoor from 600 to 3200 particles/cm^3. Given values are restricted to teaching hours 8:00 to 16:00 only, as these are the most representative for pupils' exposure. Values obtained in this study are lower in comparison to the reported values by Fromme et al. (2007), where median PNC during teaching hours measured in 36 classrooms in Munich area was 5700 particles/cm^3 (min 2600, max 12100 particles/cm^3). As we could see clear dependence of indoor submicrometer particles on outdoor concentrations, the difference can be attributed to substantially different submicrometer particle loads in ambient air in these two locations i.e. Munich city with 1.3 million inhabitants and much higher traffic in comparison to a small town in southern Sweden with 80000 inhabitants.

Inside the supermarket during opening hours median submicron PNC was 2000 particles/cm^3, ranging from 900 to 8000 particles/cm^3. In the supplied, heated ventilation air submicron PNC was much lower with a median value of 700 particles/cm^3, ranging from 140 to 3400 particles/cm^3. Indoor particle sources' contributions with peak values of about 7500 particles/cm^3 with median GMD of 44 nm (during opening hours) were observed, GMD of supplied ventilation air was 60 nm. Observed concentrations are not high, but considering high AER (~10 h^{-1}) and supply of air with very low particle loads these contributions seem to be significant. There are three possible sources of these particles. Most likely explanation of the observed nucleation mode particles could be due to formation from organic vapours as the enclosure with the instruments was placed intentionally within the area with washing powders, cleaning products and air fresheners containing terpenes, known to form particles by gas to particle conversions in the presence of ozone (Wainman et al. 2000, Sarwar and Corsi, 2007, Leungsakul et al., 2005). Weschler (2003) reported that even in a high latitude country such as Denmark (which approximates southern Sweden conditions) during winter time with limited sunlight, outdoor ozone levels (28 - 30 ppb) were sufficient, when coupled with adequate outdoor-to-indoor transport and an indoor terpene source, to contribute significantly to the growth of indoor particles. Ozone levels measured at the monitoring station about 3 km from the supermarket were between 23 to 30 ppb, the high AER (due to mechanical ventilation) provided efficient outdoor-to-indoor transport therefore ozone values are comparable to those Weschler's study. Terpenes concentrations were not measured during the study but it is possible that their concentrations in this specific location was higher than normally encountered levels indoors, Wainman et al. (2000) suggests that peak limonene concentrations at homes may easily exceed 100 ppb through the use of lemon-scented furniture polish, Weschler and Shields quoted indoor emission rate of 5 ppb/h of d-limonene from a cleaning product, considering that in the supermarket we have thousands of products the emissions might be considerable. Weschler (2003) points out that even at very high

AER, reactions between ozone and some terpenes (such as α-terpinene) are fast enough to be significant. Published articles, where increased particle concentrations due to ozone/terpenes reactions were studied at such high AER (~10 h^{-1}) are limited (Li et al., 2002; Weschler and Shields, 2000). In Li et al. study ozone and d-limonene concentrations were high, 100 – 125 ppb and 160 – 240 ppb, respectively. Weschler and Shields used similar amounts of ozone (up to 30 ppb) but used 5 ppb/h emission rate of d-limonene at AER of 12 h^{-1} that resulted in no detectable particle number increase, we believe that emission rates of mixture of terpenes are significantly higher than 5ppb/h in this specific location in the supermarket.

In Figure 5 total submicron PNC and GMD inside the supermarket and in the supplied ventilation air are plotted. Differences were observed between the time when the ventilation was operating and when it was switched off, therefore discussion is split into the two scenarios.
Ventilation on. During the hours when the ventilation was operating (ventilation was switched off between 22:00 and 4:30) PNC inside the supermarket is clearly much higher than in the supplied, heated ventilation air, which indicated an indoor source of particles. The median GMD inside the supermarket (40 nm) is about 20 nm smaller than in the supplied ventilation air (60 nm), therefore nucleation seems to be likely process explaining presence of these particles. PNC inside the supermarket fluctuates with clear peak contributions (up to 7500 particles/cm^3). The fluctuations could be due to: not complete mixing, varying AER due to door opening and draughts, changing source strengths of terpenes (elevated when shelves stocking took place due to movement and opening packaging), or influence from other sources of fine particles.
Ventilation off. When the ventilation was switched off (between 22:00 and 4:30) the removal due to high AER and supply of heated air with low particle loads were eliminated. It might have resulted in local accumulation of terpenes and temperature decrease. Thus observed sudden increase in TNC with GMD about 28 nm, could be explained by nucleation via gas-to-particle conversions from terpenes in the presence of ozone. After reaching the peak (within a half to an hour after ventilation was switched off) a decrease in the PNC was seen, possibly ozone levels were decreasing by then. At the same time the GMD increase was observed, see example in Figure 6 where GMD increased from 25 to 53 nm during the time when the ventilation was switched off. This GMD increase could be due to organic vapour condensation *and some coagulation*, which is in line with Weschler and Shields (2000) findings that reduced AER results in more time available for gas phase chemistry and particle growth. But the dominant process for the overall PNC decrease was due to diffusional wall/surface deposition, which is en efficient removal process for the particles smaller than 100 nm. Large available surface areas (shelves) support this explanation, as the deposition is also dependant on surface area.

Second possible contributor to observed nucleation mode particles could be from freshly formed particles from the exhaust emissions brought by draughts from the delivery section. The monitoring enclosure was situated about 10 m from the main doors from storage area, with nearby port doors to delivery bay. Although observed PNC increase when the ventilation is switched off and all the doors closed, does not support this explanation. Some influence during the day time from this source can not be excluded.
Third possible source of daytime contribution of ultrafine particles into the supermarket indoor air could be from electric ovens. In the supermarket two electric ovens for pastries, bread and rolls baking are used, although considering high AER (~ 10 h^{-1}) and the distance between ovens

and monitoring enclosure (opposite sides of the supermarket with floor area of 2000 m^2), influence of the baking does not seem to be significant.

In the fast food restaurant PNC found inside mirrored the concentrations measured in the supplied ventilation air with median value of 600 particles cm^{-3}, ranging from 100 to 9000 particles cm^{-3}. This low concentrations could be explained by efficient ventilation system (working 24 h per day with AER ~ 9 h^{-1}) in the dinning section where our enclosure was placed, and adequate separate extraction systems in the kitchen where grilling and frying were taking place.

Particle number concentrations (comparison of sampling sites)

Table 4 summarises number concentration of submicron particles (SMPS measurements, size range 15 - 700 nm) for studied indoor environments. Median and average values were calculated for opening hours in the supermarket and restaurant, teaching hours in the schools and entire duration of measurements in the apartment and house.

Median values for all studied locations are similar, with the lowest value of 600 particles cm^{-3} observed in the restaurant and the highest in the supermarket 2000 particles cm^{-3}. In case of the restaurant two very efficient ventilation systems i.e. in the dining and kitchen area can explain so low particle concentration. In the supermarket ventilation system is also very efficient (AER ~10 h^{-1}) but influence of a strong indoor source is observed, in our opinion it is due to local emissions of terpenes, which in presence of ozone form particles due to gas-to-particle conversions (for details see section Submicron particle number concentrations in indoor/outdoor and indoor/supplied ventilation air).

Studies reporting number concentration of submicron particles in similar environments are limited. Values reported by Ogulei et al. (2006) for a non-smoking townhouse in Boston as four months averages (in particles cm^{-3}: average 8100, standard deviation 17250, min 180 and max 308000) are similar in comparison to average concentration in the studied apartment (in particles cm^{-3}: average 7500, standard deviation 19000, min 700 and max 173000) but higher in comparison to the house. Most of found references are given as averages for no source and active source period (Hussain et al., 2005; Morawska et al., 2003) or as peak concentration values for specific sources or activities (He et al., 2005; Wallace 2006; Wallace 2005; Wallace et al., 2004; Danekamp et al., 2001), therefore not directly comparable with median or average values for the entire measurement period given in Table 4. Detail characterisation and division into non-source and active source period will be discussed later.

Values measured in schools in this study are lower in comparison to the reported values by Fromme et al. (2007), for detail description see section: Submicron particle number concentrations in indoor/outdoor and indoor/supplied ventilation air.

See and Balasubramanian (2006) reported 773000 particles cm^{-3} in a chineese food stall in Singapore (measured with ELPI, size range 8 – 2500 nm) with AER between 0 and 1.1 h^{-1}, Levy et al. (2002) reported 140000 particles cm^{-3} in a seating area of a mall's food court in Boston (measured with TSI P-Trak, range 20 – 1000 nm) no information on AER was provided. These values are much higher in comparison to measured concentrations in this study in a fast food restaurant. As mentioned before effective ventilation system and possibly lower intensity of cooking (considering amount of potential customers i.e. higher cooking intensity in Boston's mall and Singapore's food stall than in a small town's fast food restaurant) could contribute to it.

No reference values were found for the supermarket. Measured values in Boston's mall (Levy et al., 2002) are much higher with average PNC of 13000 particles cm^{-3} and again not directly comparable due to different type of sampling site (central corridor of urban mall near entrance (10 m from street, where influence of outdoor air with 25000 particles cm^{-3}, could be significant) in comparison to section with washing powders, cleaning products and air fresheners in well ventilated supermarket).

Apartment – active source periods, source strengths

For the personal exposure assessment it is important to know the average concentration during the time occupants actually are in the residences, as this will vary from the total average for given residence. Results presented here are calculated only for the time when occupants were present in the apartment (occupancy time), which accounted for about 70 % of the total sampling time. Out of the time occupants spend in the apartment 64 % was during the active source period. Active source period was defined as time when elevated particle number concentration associated with indoor activity was observed until the concentration went down to average background (no source) level. Duration of elevated particle concentration (due to indoor activities) ranged from 5.5 to 12.5 hours. During active source period particles smaller than 100 nm contributed to 75% of total submicron particles number concentration, while during no source period their contribution accounted for 50%. About 94% of total submicron PNC consisted of particles smaller than 300 nm, for both active and no source period.

In Table 5 submicrometer PNC is given only for the time when occupants were present in the apartment (occupancy time), with the division for total, active and no source activity period. Average PNC during active sources is about 13 times higher than during no source period (15300 and 1200 particles cm^{-3}, respectively). Average values for no source period observed by Hussein et al. (2005) in a family house in Finland were 2300 particles cm^{-3} and are comparable with values observed in our study 1200 particles cm^{-3}, outdoor concentration given by Hussain et al. (2005) was 5600 particles cm^{-3}, which is comparable to annual average concentrations reported for Malmö 7000 particles cm^{-3} (Dahlberg, 2007). Morawska et al., (2003) measured average concentrations for 48h in 15 houses in Brisbane, reported value for activity period is comparable to this study (18200 ± 3900 and 15300 ± 29600 particles cm^{-3}, respectively) but observed variations are higher in this study. Given values by Morawska et al. (2003) for non-activity period, 12400 particles cm^{-3}, are higher in comparison to this study (1200 particles cm^{-3}), but reported average outdoor concentrations in Brisbane was 10900 particles cm^{-3}, thus considerably higher than in Malmö. *It looks like Scandinavian houses with tight building envelope and reduced air exchange rates have a protective effect i.e. infiltration of ambient particles in submicrometer range to indoors is reduced.*

Active source period was characterised by smaller average number GMD 82 nm in comparison to 95 nm during no source time. What is worth noting average PNC during the total sampling period when the occupants were in the apartment is higher than the average PNC during entire measurement time (10200 and 7500 particles cm^{-3}, respectively) which is not surprising as occupants' presence and activities dictate occurrence of major indoor particle sources.

Measured continuous particle concentration data together with the log of occupants' activities allowed identification of indoor activities leading to increased particle levels. Identified activities that increased significantly particle number concentration consisted of cooking (frying sausages, onions, vegetables, chicken, omelette, pizza in the oven), burning candles, burning incenses, peeling oranges and tangerines, plugging in air freshener. Only a few of these activities led to a significantly increased mass concentration, namely: frying vegetables, frying chicken, cooking and heating pizza in the oven. In Figure 7 total particle number and mass concentration is plotted as an example of one day. APS and SMPS mass concentration refers to particle mass concentration which was calculated on the basis of SMPS (15 - 700 nm) and APS (0.8 – 20 μm) number size concentrations assuming unit density. Typically a sharp increase in particle number concentration at the beginning of each indoor activity is observed. When the activity is ceased slow decrease follows, the decrease to background (no source) level in the apartment took up to 12.5 hours, therefore elevated concentrations of particles were observed for long period of time. Similar patterns, i.e. sharp increases due to indoor activities with slower decrease, were observed in earlier studies (Morawska et al., 2003; Wallace, 2006; Afshari et al., 2005). In Figure 8 number size distribution at peak concentrations from different activities in the apartment are presented. The highest peak contribution was observed at party dinner, with vigorous cooking followed by 7 candles burning. Although observed peak concentrations depend on air exchange rate, apartment volume and deposition rates thus source strengths independent of theses variables are better suited for comparisons and are presented below.

Estimated source strengths for some activities in the apartment are summarised in Table 6, together with peak concentration and decay rates for given size intervals. Activities listed in Table 6 met the selection criteria, described in section Source strengths estimation. Estimated total source strengths for cooking events range from 5.5×10^{11} to 1.5×10^{12} particles min^{-1}, lowest for frying omelette (no oil, Teflon frying pan used) and highest for cooking. The highest contribution is observed from ultrafine particles, their source strengths range from 5.1×10^{11} to 1.1×10^{12} particles min^{-1}, which accounts on average for 84 % of total generated submicron particles. Peeling tangerines produced 3.5×10^{10} particles min^{-1}, and ultrafines accounted for 90%. Source strengths estimated by He et al. (2004) in 15 houses in Brisbane for cooking pizza, frying, cooking and grilling range from 1.7×10^{11} to 5.7×10^{11} particles min^{-1}, thus are slightly lower but comparable with results obtained in this study. Values reported by Wallace et al. (2004) 3.0×10^{12} particles min^{-1} from 44 cooking episodes and agree with our upper limit of obtained source strengths. Wallace (2006) published source strengths per cooking event for 381 cooking events (from about 3 years long study in a town house in Reston, Virginia), assuming average active cooking time lasting 20 min, recalculated source strengths for tea and toast, tortillas and broiled fish were 2.5×10^{11}, 2.0×10^{12} and 1.4×10^{12} particles min^{-1}, and again are comparable to obtained values in this study. Afshari et al. (2005) in a full-scale chamber study reported source strength of 8.3×10^{11} particles min^{-1} for frying meat, which again is within the range of values obtained here. Evident limitation of our study is the calculation of source strengths based on singular events in one apartment only.

OC/EC analysis

Average particulate OC and EC concentration and their contribution to PM.2.5 mass for studied indoor environments is presented in Table 7. Given concentrations were corrected for positive

sampling artefacts accomplished by subtraction from OC on the quartz filter, the gas phase OC adsorbed onto quartz backup filter (placed in a parallel line after Teflon filter). Turpin et al. (2000) has shown that this is a good method of estimating the adsorption of the gas phase on a quartz filter. The gas phase OC measured on the quartz backup filter is also given in Table 7. Particulate organic matter (OM), an estimate of the average organic molecular weight per carbon weight was obtained by multiplying OC by 1.4 (Polidori et al., 2006; Turpin and Lim, 2001)
Polidori et al. (2006) in a part of RIOPA study, where OC and EC were measured in 173 homes in US, reported that average indoor OM concentration was 9.8 μg m^{-3} and accounted for 48 – 61% of PM2.5 mass, results from the apartment in this study for OM (6 μg m^{-3}) are slightly lower but comparable, but their contribution to PM2.5 is much lower (18%). This observed lower contribution to PM2.5 mass could be due to combination of the three following factors: 1) In this study only three OC/EC measurements were done in one apartment, thus differences are expected. 2) Observed difference in OC contribution to PM2.5 may indicate not accurate estimation of mass concentration on the basis of DustTrak readings, 3) Particulate OC could be underestimated as filters for OC/EC collection were placed in the enclosure where temperature was about 35 – 40 ^{0}C, thus some particulate OC might have been lost due to evaporation. Measured much higher concentrations of gas phase OC, accounting for 40 to 100% of particulate OC could support this explanation.
Sawant et al. (2004) reported for non smoking residences and classrooms OM of 13.5 and 5.0 μg m^{-3}, respectively (OM=1.9OC). When recalculating our values with the same factor, obtained concentrations are slightly lower, for apartment and school 8.5 and 3.7 respectively. Reported by Sawant et al. (2004) EC concentrations 2 and 1 μg C m^{-3} for non-smoking residences and classrooms, are higher in comparison to 0.7 and 0.4 μg C m^{-3} in this study, respectively.
Ward at al. (2007) reported in the middle school average OC and EC of 8.8 and 0.9 μg C m^{-3}, respectively. These values are higher in comparison to what was measured in the school B (1.9 and 0.4 μg C m^{-3} for OC and EC respectively), but Ward et al. study was done in area with very high contribution of residential wood combustion to observed PM2.5, thus higher OC and EC values could be expected. *Though sum of measured OC and EC concentrations for PM 2.5 in middle school in Ward et al. (2007) study was higher than measured PM 2.5 mass concentration, possibly due to sampling artefacts.*

Conclusions

Airborne particle were measured in various Swedish indoor environments. Assessed particle characteristics were: number and mass concentrations, size distributions and organic and elemental carbon content. Median mass concentration (DustTrak readings) varied among the sites between 6 and 25 μg m^{-3}, measured for school and supermarket, respectively. In the apartment the short term peak mass concentrations were observed up to 400 μg m^{-3} (about 20 times higher than the ambient concentrations), and were due to cooking and candle burning. Median values of submicron PNC for all studied locations were similar, between 600 and 2000 particles cm^{-3}. Both sites have an efficient ventilation system (AER ~10 h^{-1}) but influence of a strong indoor source in the supermarket was observed, possibly due to local emissions of terpenes (in section with washing powders, cleaning products and air fresheners), which in presence of ozone form particles due to gas-to-particle conversions.

During occupancy time (occupants inside) average submicron PNC in the apartment and house was 10200 ± 24600 and 2300 ± 2700 particles cm^{-3}, respectively and average PM2.5 mass concentration in the apartment was 36 ± 51μg m^{-3} (not measured in the house). These occupancy time averages represent personal exposure values while indoors due to both indoor generated and penetrated from outdoor particles. Duration of elevated PNC (due to indoor activities) ranged from 5.5 to 12.5 hours. Average PNC during active source period is about 13 times higher than during no source period (15300 ± 29600 and 1200 ± 300 particles cm^{-3}, respectively). During active source period particles smaller than 100 nm contributed to 75% of total submicron particles number concentration, while during no source period their contribution accounted for 50%. About 94% of total submicron PNC consisted of particles smaller than 300 nm, for both active and no source period. Estimated source strengths for cooking events range from 5.5×10^{11} to 1.5×10^{12} particles min^{-1}.

The importance of adequate ventilation system, capable of removing/diluting and not allowing the accumulation of both particles and not studied here gaseous emissions generated by indoor sources, becomes apparent when comparing results from apartment and fast food restaurant. Even though amount of cooking in the restaurant is incomparably higher than in the apartment, due to efficient ventilation there are no observed impacts of these sources on particle loads in the restaurant's dinning area, while in the apartment peak concentrations of submicron particles can reach 170000 particles cm^{-3}. It is not possible to regulate activities in private homes to reduce particle exposure, but much can be done to enhance ventilation in locations where strong indoor sources of particles occur, namely kitchen. Raising general public awareness should be one of the priorities.

Acknowledgments

This work has been supported by the Swedish Research Council FORMAS and the Development Fund of the Swedish Construction Industry (SBUF).

References

To be added ....

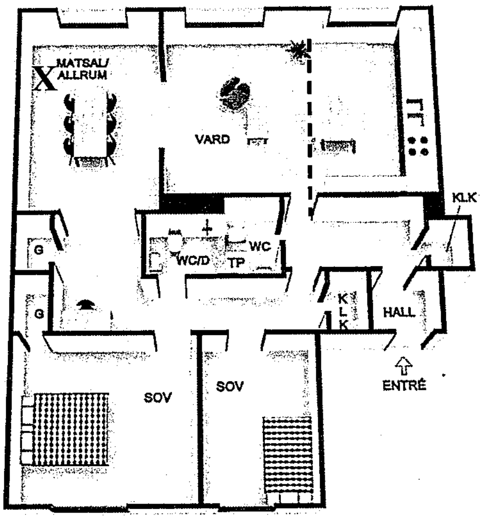

Figure 1. Apartment's layout, X indicates placement of the enclosure with instruments

Table 1. List of sampling sites, measurements duration, type of ventilation, air exchange rates and simultaneous/alternate measurements of indoor and outdoor air.

Sampling location	**Measurements**		**Type of ventilation**	**AER**	**Simultaneous/alternate[d] measurements of**	
	Date	**Number of days**		**(h^{-1})**	**indoor and outdoor air**	**indoor and supplied ventilation air**
School A	Oct 2006	10	mechanical	2.4[a]	SMPS	
School B	Mar 2007	11	natural	0.4[a]	SMPS + 2 DustTraks	
Supermarket	Jan 2007	7	mechanical	9.7[b]	2 DustTraks	SMPS
Restaurant	Dec 2006	7	mechanical	8.5[b]		SMPS
Apartment	Feb 2007	10	natural	0.3[c]	2 DustTraks	
House	Feb 2007	8	natural	0.12[c]		

[a]) measured at low ventilation conditions i.e. windows and doors closed
[b]) measured during normal opening hours, doors opening not controlled
[c]) measured at low ventilation conditions i.e. windows closed, doors between rooms opened
[d]) simultaneous measurements by two DustTraks, alternate measurements by SMPS system

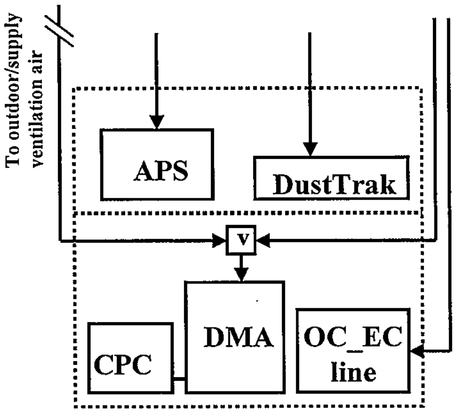

Figure 2. Enclosure with the instruments

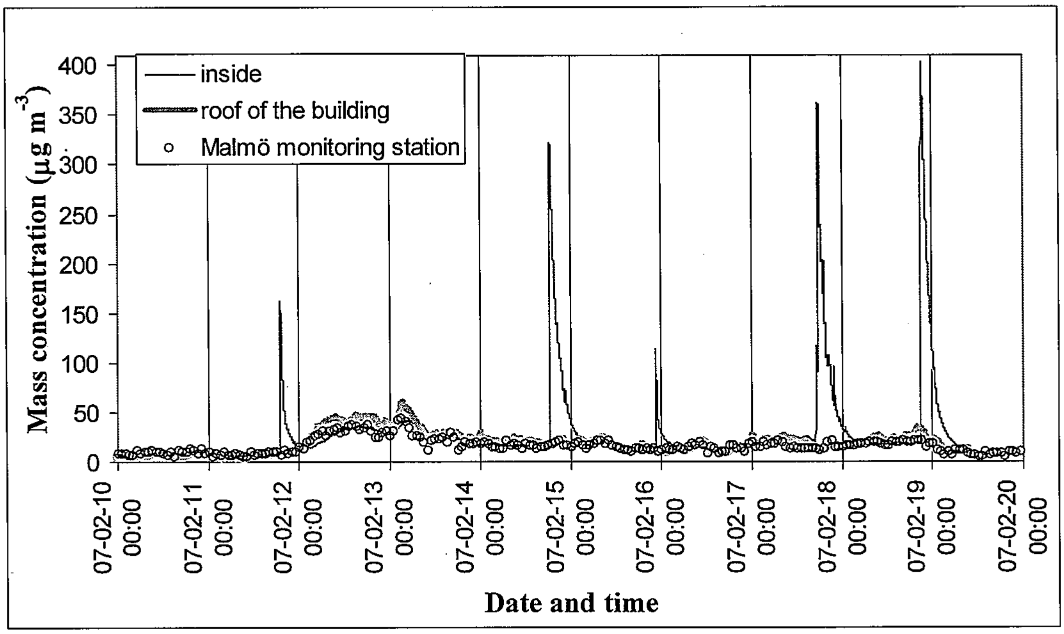

Figure 3. Comparison of DustTraks' readings indoors in the apartment and on the roof of the building with mass concentration measured by TEOM at the monitoring station in Malmö.

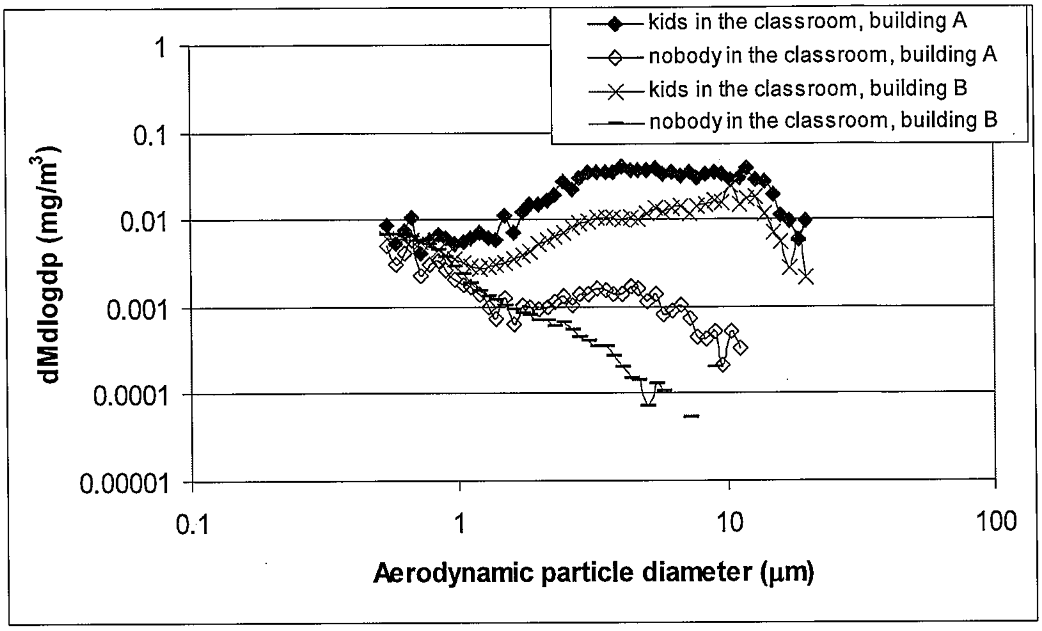

Figure 4 One hour average mass size distribution in school A and B when pupils were in the classroom and when there was no one. Calculated on the basis of APS measurements assuming unit density

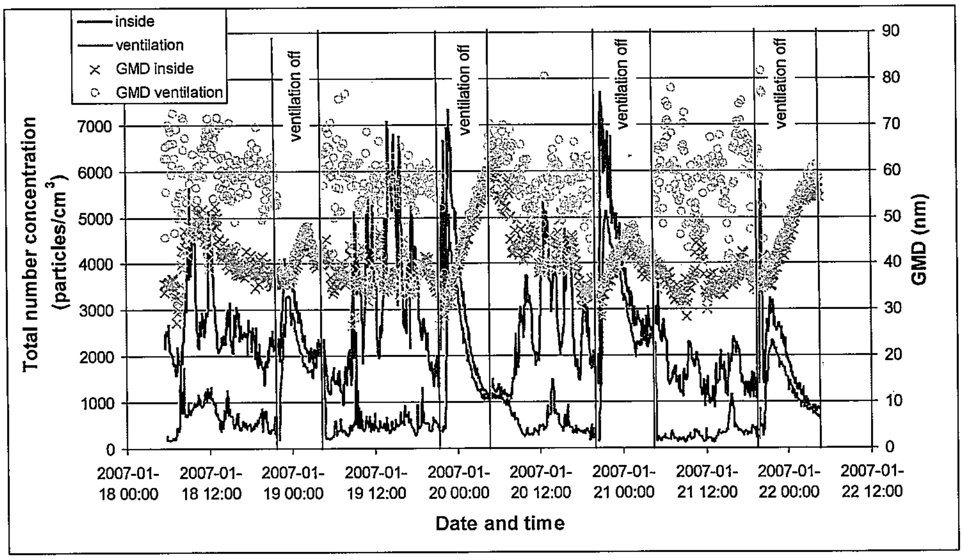

Fig 5 TNC and GMD inside and in the supplied ventilation air in the supermarket

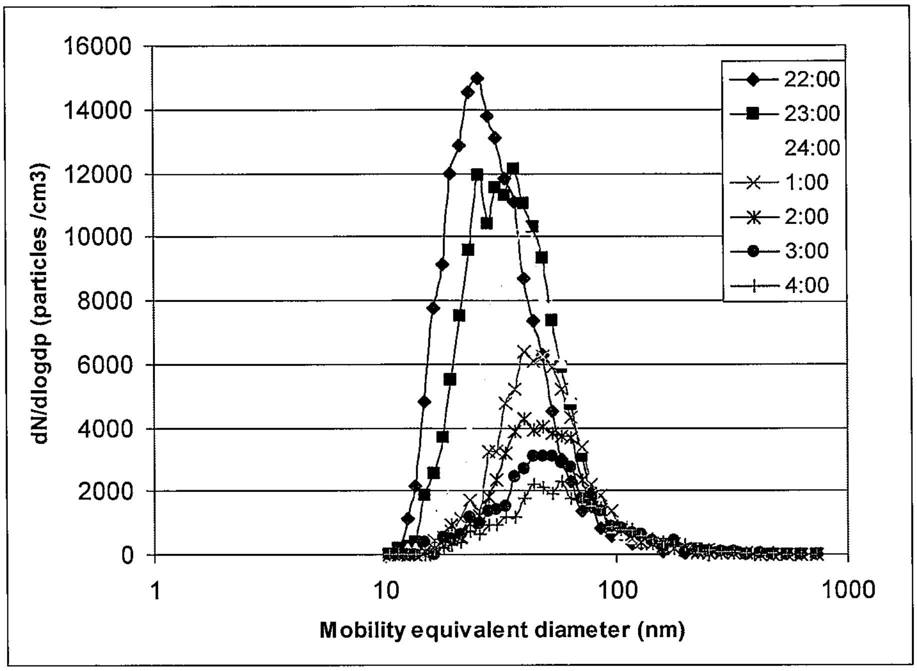

Figure 6 GMD increase in the supermarket when the ventilation was switched off.

Table 2. Approximation of PM 2.5 mass concentration (DustTrak measurements) for studied indoor environments

	Approximation of $PM_{2.5}$ mass concentration ($\mu g\ m^{-3}$)				
	Supermarket	**Restaurant (PM_1)**	**School A**	**School B**	**Apartment**
Median	6	8	25	8	18
Minimum	5	2	8	2	5
Maximum	20	14	43	47	403

Table 3. Reported PM2.5 mass concentration for non-smoking residences, schools and restaurants

PM2.5 mass concentration (µg m^{-3})							
	median	min	max		median	min	max
Residences				Morawska et al. 2003 (Brisbane, 15 houses, activity period 24h)	16 ± 8[b]	8	37
Wigzell et al., 2000[a] (UK, 10 homes, 48h in living rooms)	9	6	19	Morawska et al. 2003 (Brisbane, 15 houses non-activity period 24h)	11 ± 3[b]	8	18
Molnar et al., 2005 (homes with wood burning)	10.3	7	18	**Schools**			
Sawant et al., 2004[a] (California, 20 homes, 24h)	29	19	54	Sawant et al., 2004 (California, clasrooms' values only, 24h)	18	17	25
Molnar et al., 2007 (Stockholm, 20 homes, 10 classrooms, 10 preschools)	8	3	19	Fromme et al., 2007 (Munich, 64 schools, teaching hours)	20 - 37	4	73
Koistinen et al., 2004 (Helsinki, 102 residences)	9			Ward et al., 2007 (Libby, Montana, elementary school, 24h)	37	14	54
Breysse et al. 2005[a] (Baltimore, 100 houses)	26 ± 15[b]			Ward et al., 2007 (Libby, Montana, middle school, 24h)	6	5	11
Stranger et al., 2007 (Antwerp, Belgium, 15 residences, 24h)	30[c]	11	75	Janssen et al., 2001 (Netherlands, 24 schools, teaching hours)	23	8	53
Long et al., 2000 (Boston, 9 houses, 12h)	12 ± 10[b]	2	86	**Restaurants**			
Abt et al. 2000 (Boston, 4 homes)	14 ± 15[b]	3	128	Travers et al, 2004 (New York, 20 bars and restaurants after smoking ban)	24	6	83

[a]) values for non-smoking residences only, the whole study comprised both non- and smoking residences
[b]) mean concentration ± standard deviation
[c]) mean concentration

Table 4. Total submicron PNC (15 - 700 nm, SMPS measurements) for studied indoor environments

	Partcle number concentration (particles cm^{-3})					
	Supermarket	**Restaurant**	**School A**	**School B**	**Apartment**	**House**
Median	2000	600	1600	1500	1700	1400
Average	2300	700	1700	1600	7500	2300
STD[a]	1200	500	500	700	19000	2500
Minimum	800	100	800	700	700	250
Maximum	10000	9100	3200	3500	173000	35300

[a]) STD – standard deviation

Table 5. Total submicron PNC (15 - 700 nm, SMPS measurements) during the time occupants were in the apartment

Occupancy time	**Average**	**STD**[a]	**Median**	**Min**	**Max**	**Average number GMD**[b]	**Average GSD**[c]
	particles cm^{-3}					**nm**	
Total	10200	24600	1800	660	173000	87	2.0
Active sources	15300	29600	4000	950	173000	82	1.9
No sources	1200	330	1200	660	2300	95	2.1

[a]) standard deviation
[b]) geometric mean diameter
[c]) geometric standard deviation

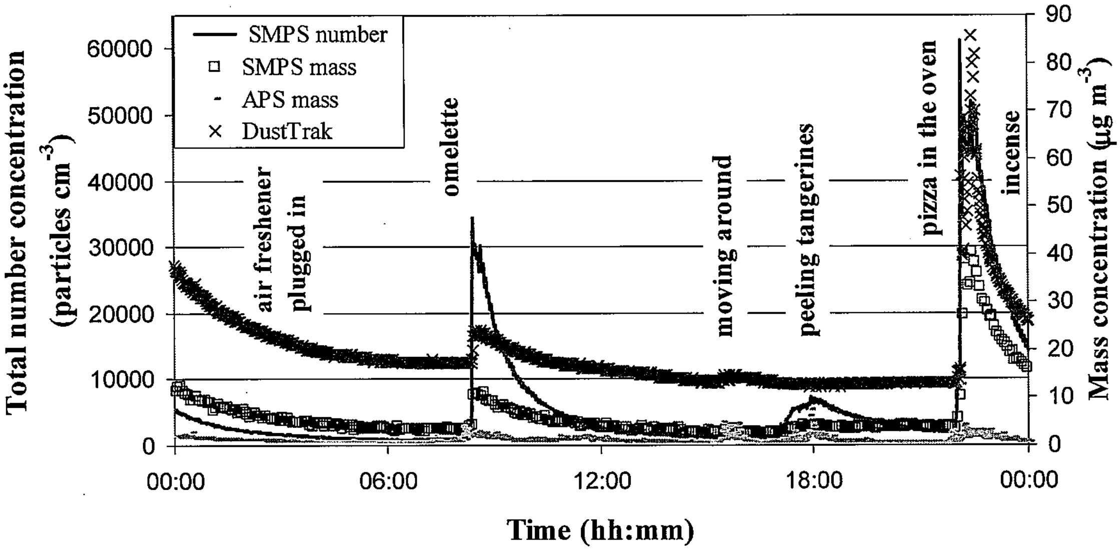

Figure 7. Total number and mass concentration in the apartment - example of one day. SMPS (15 -700 nm) and APS (0.8 – 20 µm) mass concentration was calculated from number size concentrations assuming density of 1 kg/m^3.

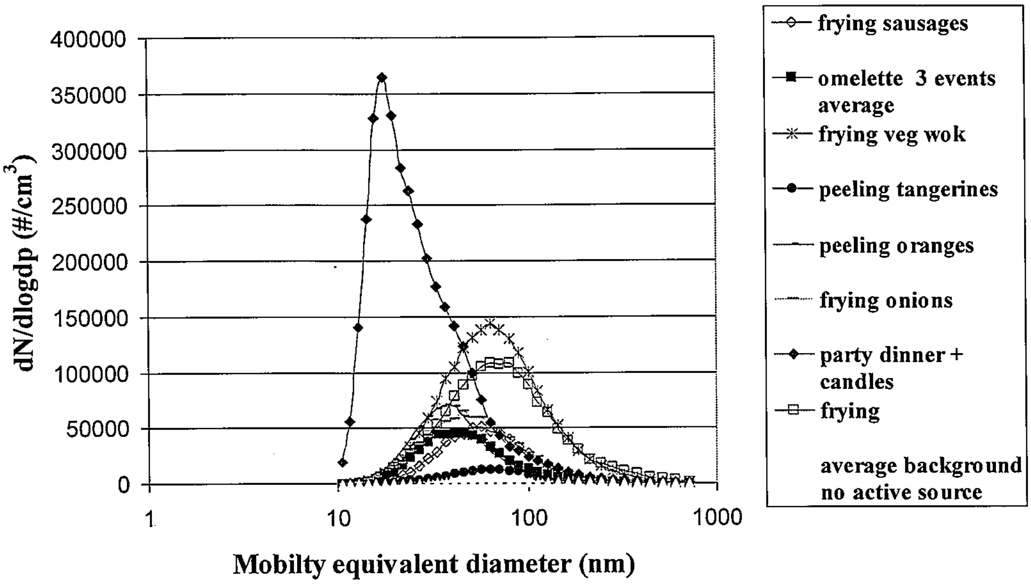

Figure 8. Number size distribution at peak concentrations from different activities in the apartment

Table 6. Source strengths estimations for some activities in the apartment.

Activity	Size interval	Decay rate[a] (a+k)	Peak			Time[d]	Source strength
			Concentration	Number GMD[b]	GSD[c]		
	(nm)	(h^{-1})	(particles cm^{-3})	(nm)		(min)	(particles min^{-1})
Cooking							
	15 - 50	1.07	20330				4.26E+11
	50 - 100	0.76	35171				7.06E+11
	100 - 300	0.53	17574				3.39E+11
	300 - 700	0.48	2445				4.69E+10
		total	**75519**	**69**	**1.9**	**15**	**1.52E+12**
Frying sausages							
	15 - 50	1.16	11040				2.86E+11
	50 - 100	0.66	13435				3.27E+11
	100 - 300	0.36	3916				8.05E+10
	300 - 700	0.11	300				4.03E+09
		total	**28691**	**58**	**1.8**	**12**	**6.97E+11**
Veg wok							
	15 - 50	1.17	21370				2.61E+11
	50 - 100	0.82	49477				5.59E+11
	100 - 300	0.52	21368				2.23E+11
	300 - 700	0.45	2135				2.16E+10
		total	**94350**	**64**	**1.9**	**30**	**1.06E+12**
Peeling 1 kg of tangerines							
	15 - 50	0.56	2220				1.15E+10
	50 - 100	0.38	3923				1.95E+10
	100 - 300	0.18	1070				3.44E+09
	300 - 700	0.11	66				6.55E+07
		total	**7279**	**61**	**1.8**	**59**	**3.45E+10**
Frying onions							
	15 - 50	1.85	18185				3.04E+11
	50 - 100	0.78	15980				2.18E+11
	100 - 300	0.65	5143				6.35E+10
	300 - 700	0.76	584				7.03E+09
		total	**39891**	**52**	**1.8**	**23**	**5.92E+11**
Frying omelette (3 events average)							
	15-50	0.89	14369				3.10E+11
	STD[e]	(0.17)	(5757)				(1.53E+11)
	50-100	0.59	9561				2.04E+11
	STD[e]	(0.13)	(5612)				(1.45E+11)
	100-300	0.27	2091				3.43E+10
	STD[e]	(0.15)	(1338)				(2.84E+11)
	300-700	0.11	153				1.38E+09
	STD[e]	(0.06)	(88)				(1.32E+09)
		total	**26174**	**45**	**1.8**	**15**	**5.49E+11**
	STD[e]		**(12221)**				**(3.21E+11)**

[a]) measured AER at low ventilation conditions equals 0.3 h^{-1}

[b]) geometric mean diameter

[c]) geometric standard deviation

[d]) time from initial background concentration (one measurement point preceding first observed concentration increase) until peak concentration was reached

[e]) standard deviation

Table 7. Average particulate organic and elemental carbon, and their contribution to PM2.5 mass for studied indoor environments. In brackets standard deviation is given. Given concentrations were corrected for positive sampling artefacts by subtraction from OC on the quartz filter, the gas phase OC adsorbed onto quartz backup filter (placed in a parallel line after Teflon filter).

	OC		**Gas phase OC**[a]		**EC**		**OM**[b]	**Contribution to $PM_{2.5}$ mass**					
								OC		**OM**		**EC**	
	$\mu g\ m^{-3}$		$\mu g\ m^{-3}$		$\mu g\ m^{-3}$		$\mu g\ m^{-3}$	%		%		%	
Supermarket	n.d.[c]	n.d.[c]	8.91	(2.83)	0.22	(0.13)	-	-	-	-	-	3	(2)
Restaurant	0.99	(0.90)	2.96	(2.61)	0.13	(0.14)	1.38	17[d]	(6)	24[d]	(8)	2[d]	(1)
School B	1.93	(1.91)	4.15	(1.26)	0.44	(0.42)	2.71	26	(25)	36	(35)	5	(3)
Apartment	4.48	(5.69)	7.52	(0.81)	0.67	(0.17)	6.27	13	(11)	18	(15)	3	(2)
House[e]	1.17	-	1.60	-	0.22	-	1.56	51[f]		73[f]		10[f]	

[a]) gas phase organics detected on the quartz backup filter placed after Teflon filter (positive artefact)

[b]) OM=1.4×OC, particulate organic matter

[c]) n.d. not detected, method's detection limit approximately 2 $\mu g/m^3$

[d]) contribution to PM_1

[e]) only 1 measurement (72h), no standard deviation

[f]) PM2.5 estimated on the basis of SMPS and APS number size concentrations assuming density 1 kg/m^3