Concept of high quality simultaneous measurements of the indoor and outdoor aerosol to determine the exposure to fine and ultrafine particles in private homes

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Summary Simultaneous measurements of indoor and outdoor aerosol particles are crucial for determining the relationship of indoor to outdoor particle concentrations (Indoor/Outdoor ratio; I/O ratio) as well as the indoor particle exposure. This article describes an improved concept to determine the exposure to indoor particles, including the indoor and outdoor measurements of particle number size distribution (PNSD) of fine and ultrafine particles, and coarse mode particle mass concentrations. Special mobility particle size spectrometers (Mobility Particle Size Spectrometer; MPSS), designed and built at TROPOS, have been employed to determine the indoor and outdoor PNSD (0.01 to 0.8μm). The MPSS should be calibrated against a reference MPSS at a calibration facility (e.g. the World Calibration Center for Aerosol Physics; WCCAP) every second month. This article describes the set up and quality assurance of the indoor and outdoor measurement system, as well as the logistic of the measurement. Examples of yearlong measurements at Leipzig are shown as illustrative results. Ultrafine aerosol particles are especially produced due to cooking activities or candle burning. The median I/O shows that the indoor aerosol is increasingly decoupled from outdoor during nighttime. During daytime, the influence of the outdoor aerosol is more significant and the I/O ratio is close to 1.0.

Konzept für qualitativ hochwertige simultane Messungen des Innenraum- und Außenaerosols zur Bestimmung der Exposition von feinen und ultrafeinen Partikel in privaten Wohnungen

Zusammenfassung Simultane Messungen von Aerosolpartikeln im Innenraum und Außenbereich sind entscheidend für das Verhältnis von Innenraum- zu Außenbereich-Partikelkonzentrationen (I/O-Verhältnis) sowie der Partikelbelastung im Innenraum. Dieser Artikel beschreibt ein verbessertes Konzept zur Bestimmung der Exposition von Innenraumpartikeln, einschließlich der Messungen der Partikelgrößenverteilung (Particle Number Size Distribution; PNSD) von feinen und ultrafeinen Aerosolpartikeln im Innenraum und Außenbereich sowie der Partikelmassenkonzentrationen im groben Partikelgrößenbereich. Spezielle Partikelgrößenspektrometer (Mobility Particle Size Spectrometer; MPSS), von TROPOS entwickelt und gebaut, wurden eingesetzt um die PNSD im Innen- und Außenbereich (0,01 bis 0,8 μm) zu bestimmen. Die MPSS sollten jeden zweiten Monat in einer Kalibrierungseinrichtung (z. B. dem World Calibration Center für Aerosol Physics; WCCAP) gegen ein Referenz-MPSS kalibriert werden. Dieser Artikel beschreibt den Aufbau und die Qualitätssicherung der Innen- und Außenmesssysteme sowie die Logistik der Messung. Als anschauliche Ergebnisse werden Beispiele von einjährigen Messungen in Leipzig gezeigt. Ultrafeine Aerosolpartikel werden insbesondere aufgrund von Kochaktivitäten oder brennenden Kerzen erzeugt. Der Median des I/O-Verhältnisses zeigt, dass das Innenraumaerosol nachts zunehmend vom Außenbereich entkoppelt ist. Tagsüber ist der Einfluss des Außenaerosols signifikanter und das I/O-Verhältnis liegt nahe bei 1,0.

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1 Introduction

Ambient aerosol particles have drawn special attention by many studies because of the health effects (e.g. asthma, cardiovascular diseases) caused by the inhalation [1-4]. Other studies have indicated that health effects of fine (particle diameter <1 μm) and ultrafine (particle diameter <0.1 μm) aerosol particles could potentially be more harmful than those of $PM_{2.5}$ (particle diameter <2.5 μm) [5-7].
Additionally, there is a growing concern about the exposure to indoor particles, since people spend most of their time indoors, in developed countries around 60% [8-10]. Indoors, people are exposed to a mixture of particles from outdoor and generated by human indoor activities [11; 12]. Many studies reported that the outdoor particle number size distribution (PNSD) is modified due to ventilation process [13; 14]. Furthermore, indoor particle losses are strongly particle size-dependent [15; 16] and highest for ultrafine and course particles. To understand the indoor particles exposure, it is therefore important to study indoor particle emissions due to residential activities as well as the influence of the outdoor aerosol on the indoor particle number size distribution (PNSD).
The most commonly used methods of indoor particles exposure measurements are: outdoor measurements to predict indoor concentrations, the use of a direct personal monitor, or simultaneously indoor and outdoor measurements [17]. For the first two methods there are missing the indoor source or outdoor source information, respectively. The method of simultaneous measurement of indoor and outdoor PM_{10} and $PM_{2.5}$ has been well studied [14; 17 to 20]. Until now, either studies lacked measurements of the PNSD of fine and ultrafine particles, or their measurements were statistically not robust, because of an insufficient number of probed homes. One could therefore only retrieve an incomplete overview, which was probably not representative.
This article describes an improved concept to determine indoor and outdoor PNSD of fine and ultrafine particles with high quality, as well as measurement of course mode particle mass concentrations. The whole concept is designed to perform measurement in multiple private homes over a period of more than one year in order to obtain statistically more representative data. This concept includes a) the design of instrumentation to make investigations around the year possible, b) the quality assurance of the instruments and data, and c) the logistic of the measurement.

Figure 1. Indoor system, custom-built by TROPOS.

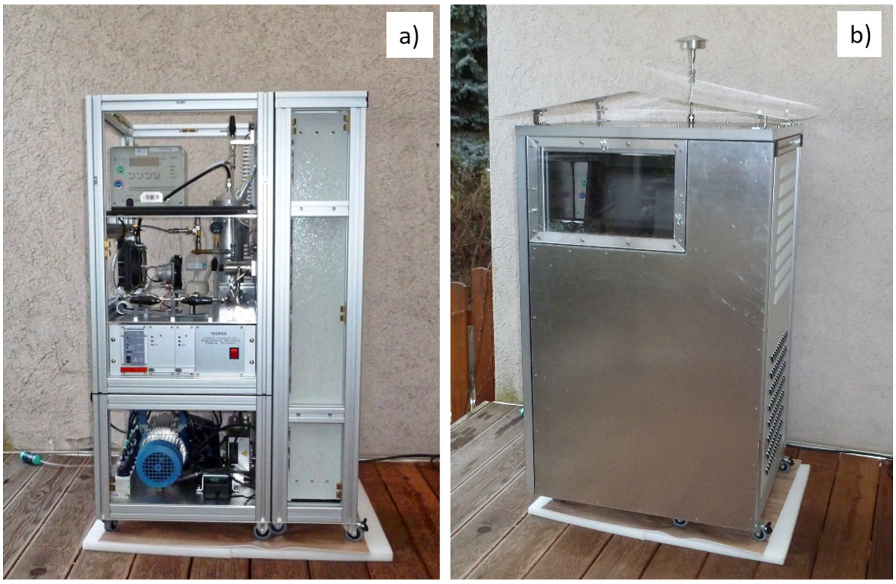


Figure 2. a) Outdoor system with outer panels and roof dismantled. The air-conditioner occupies the right hand segment of the rack. b) Complete assembly of the outdoor system as employed in the field.

2 Instrumentation

2.1 Design considerations

To provide high-quality indoor and outdoor particle number size distributions across the range 0.01 to 0.8 µm, special mobility particle size spectrometers (MPSS) were designed and constructed by TROPOS (**Figures 1** and **2**). Measuring air quality in occupied indoor environments requires enhanced standards for the technical equipment with regard to avoiding hazards and nuisances. One aspect is the avoidance of radioactive material or organic solvents that are usually employed in standard MPSS instruments. In addition, it is advocated to minimize the noise emerging from the equipment. Other aspects include portability – in order to be able to move the instrumentation easily from one dwelling to another, and operational flexibility. Outdoor equipment, in contrast, needs to be sheltered from extreme temperatures, rain and excessive humidity. Preliminary considerations of the planned indoor/outdoor measurements led to the conclusion that these standards could be best achieved with a pair of newly designed and custom-built instruments.

2.2 Indoor mobility particle size spectrometer

The indoor measurement system has been designed to provide a minimum of nuisance to the dwelling's occupants with respect to noise as well as hazardous odours and radiation. Regular MPSS instruments are typically equipped with a Condensation Particle Counter (CPC) operated with butanol as the working fluid and a bipolar diffusion charger containing radioactive nuclides. To meet higher safety standards, the indoor system described here is equipped with a water-based CPC (TSI Inc., GP-WCPC Model 3787). The bipolar diffusion charger is custom-designed and contains a Ni^{63} nuclide. The decay activity is 95 MBq, which is within the free legal limit of 100 MBq, thus minimizing its impact on the indoor environment. The entire indoor MPSS system is pictured in Figure 1. Its housing has been made soundproof to reduce noise of the CPC and other small pumps. In addition, the original pump of the water CPC was replaced by a low-noise pump, because the vibrations of the original pump turned out to be too noisy for applications in private homes. Moreover, the ventilation of the water-based CPC was augmented to prevent overheating. The entire measurement rack is ventilated to keep the system temperature close to the room temperature.

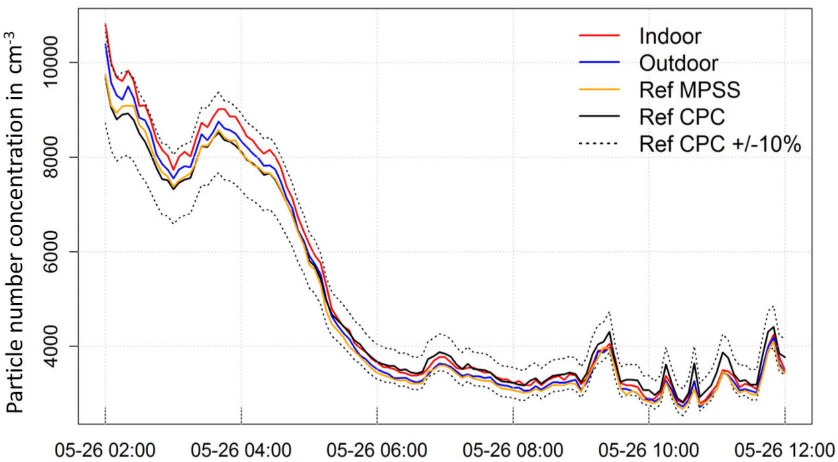


Figure 3. Example of the intercomparison results from May 26, 2017. The time series of the PNCs (10 to 800 nm) of the indoor, outdoor, reference MPSS and reference CPC are compared. The dashed lines represent the +/-10% target uncertainty range around the reference CPC.

2.3 Outdoor mobility particle size spectrometer

To resist temperature changes in the outdoor environment, the outdoor instrument was thermally insulated, and incorporated into a waterproof shelter. The MPSS rack (Figure 2a) includes a special air-conditioner for cooling and heating (Rittal CS 9776500, 650 W for cooling, and 400 W for heating). Due to the availability of both, air-conditioning and heating, the inside temperature can be kept between 22 and 26°C for ambient temperatures between -10 and 35°C. Although the outdoor system is designed to operate outside (balcony, terrace, garden), the potential nuisance by butanol exhaust vapors from the CPC needs to be controlled. An active carbon filter is therefore used at the exhaust tube of the outdoor system, which is regularly changed to avoid saturation.

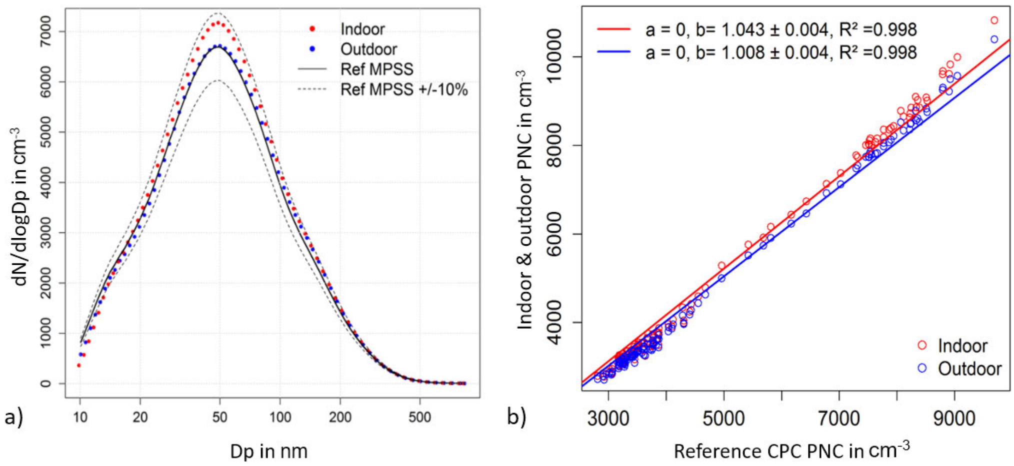


Figure 4. Example of the intercomparison results from May 26, 2017. a) The mean PNSD of the indoor, outdoor, and reference MPSS. The dashes lines represent the +/-10% target uncertainty around the reference MPSS. b) Linear regression between the particle number concentrations of the indoor and outdoor MPSS, and reference CPC. Legend at top left shows the curve fit results, coefficient values ± one standard deviation.

2.4 Additional measurements

Both MPSS are complemented by optical particle size spectrometers (Grimm, Model 1.108) that provide PM_{10}, $PM_{2.5}$ and PM_1 particle mass concentration. Measurements of equivalent black carbon mass concentration (microAeth Model AE51) and CO_2 concentration (CO_2 sensor, GMP252 Vaisala) were added for the indoor instrument. CO_2 concentrations measurements are useful to document ventilation events such as opening doors or windows. (CO_2 generated by the occupants will be diluted with ambient air.) Furthermore, equivalent black carbon mass concentrations are helpful to determine the impact of the outdoor aerosol and to identify indoor sources that are related to combustion or heating processes such as candle burning or baking. For easier transport and installation, both instruments were designed in a modular fashion, also having their weight optimized. Steady and lockable wheels were installed for easier handling.

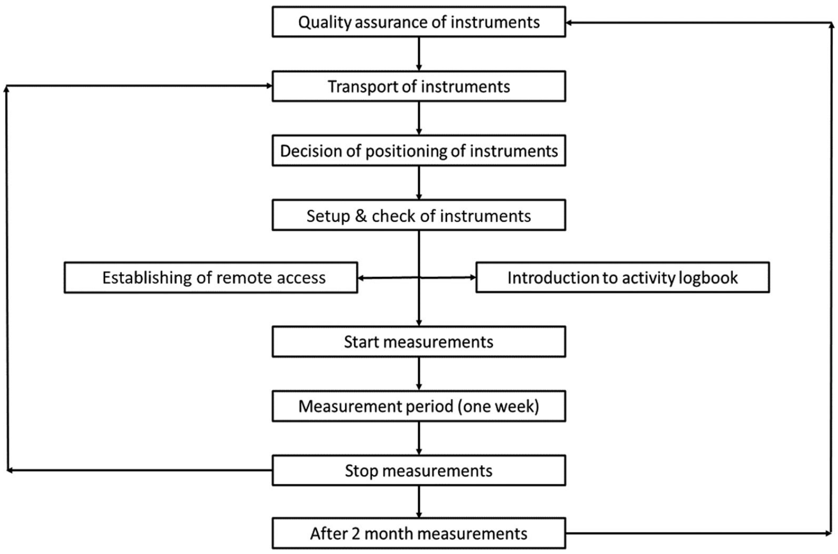


Figure 5. Flowchart of indoor and outdoor simultaneous measurement at private homes.

3 Quality assurance

To maintain a high reliability of the measurement data the indoor and outdoor MPSS are calibrated against a reference MPSS at the World Calibration Center for Aerosol Physics (WCCAP) every second month. During these intercomparisons, the indoor, the outdoor, and the WCCAP reference MPSS measure the PNSD of the ambient aerosol simultaneously. Furthermore, the CPCs are calibrated against a Faraday Cup Aerosol Electrometer (FCAE).

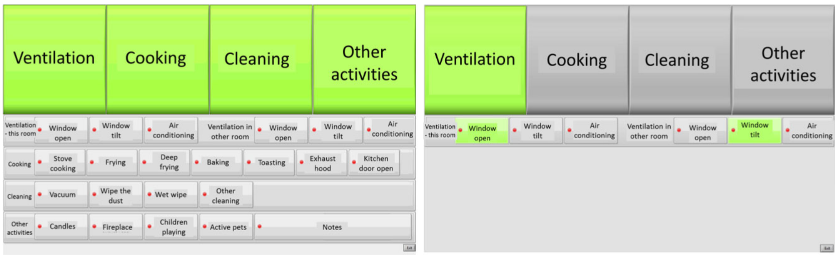
Particle number concentration (PNC) and mean particle number size distribution (PNSD) of indoor and outdoor MPSS should be within a ±10% target uncertainty compared to the reference MPSS and reference CPC as shown in **Figure 3** and **Figure 4a**. The PNC of indoor and outdoor MPSS should have a close linear relationship with the reference CPC as shown in **Figure 4b**. The calibration procedures and target uncertainties are described in more detail in *Wiedensohler* et al. [21].

Figure 4a shows the mean PNSD of the indoor, the outdoor and the reference MPSS. The PNSD of the indoor and outdoor MPSS are within the target uncertainty of +/10% over a wide range against the reference MPSS. Only below 20 nm in particle diameter, the deviation become somewhat greater. This increased uncertainty of MPSS is a general feature, which was already described in a former study [22]. The PNSD have been calculated using the inversion routine [23] and corrected for internal losses using the method of equivalent length [22] as well as for size-dependent counting efficiency of the butanol and water CPCs.

4 Planning and logistics of indoor/outdoor measurements

The instruments described in this article are, at the time of writing, being used for a cross-sectional study across 40 homes in Germany. The typical logistics of such field measurements consist of the transportation of the instruments between the dwellings under study, the training of study participants to use a digital activity logbook, and to establish Internet access to both instruments for the purpose of remote maintenance. The measurement cycle is illustrated in **Figure 5**.

Each MPSS instrument is running under control of a custom-made data acquisition program (LabVIEW Version



a) b)

Figure 6. Interface of digital activity logbook. a) All options of activities selected. b) Ventilation and the subcategories "window open" and "window tilt" selected.

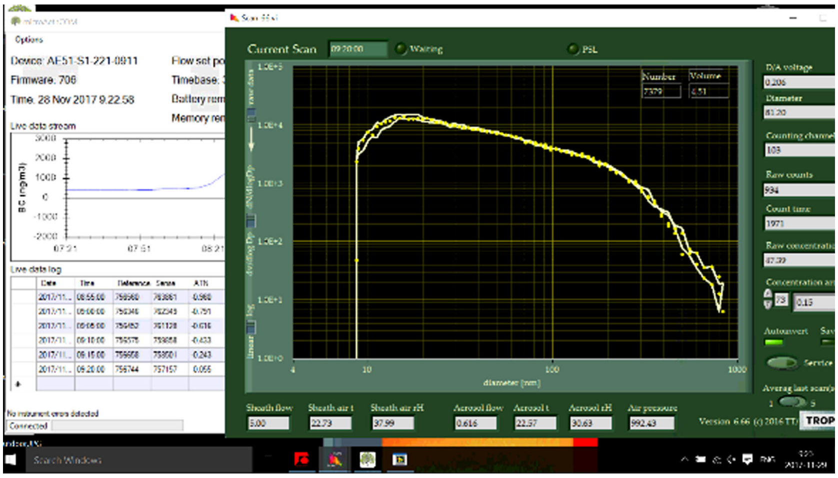


Figure 7. Example of remote control observation. Indoor system PC interface.

6.66, National Instruments). The program shows the instrument's status parameters (e.g. temperature, relative humidity, error code) and time series of preliminary measurement results. After installation of the instruments, a zero check (total filter connect inlet) and a flow check for the whole system ensure that the system has no leaks and all the air pumps are working correctly. When the PNSD plot of the MPSS program shows outliers, the high voltage checking and calibration of DMA is also necessary.

To be able to interpret the indoor measurement and to minimize the effort of the people, a digital active logbook was developed as shown in **Figure 6a**. Specific activities can easily be selected by clicking as illustrated in **Figure 6b**.

In order to obtain more statistically significant measurement data, measurements should be continuous and last days long. At the same time, during the measurement in the household, the operation staffs should be absent to avoid an impact on residence's normal life. It is however necessary to be able to remotely check and control measurements. Therefore, a remote control software (e.g. TeamViewer) is installed on every PC. Through an Internet connection, we could easily obtain access to the PC (**Figure 7**) at any time, and operate troubleshooting if necessary.

5 Illustrative results

5.1 Time series of indoor CO_2 and black carbon concentration

"Open window" is one of the most common indoor activity, and it has a non-negligible influence on the contribution of indoor sources to indoor air. For the example in **Figure 8**, the indoor particle number concentration shows one sharp peak, reaching the outdoors concentration. At the same time, a drop in the CO_2 concentration indicates an open window.

Figure 9 shows an example of toasting. During this event of approximately 15 minutes, the total particle concentration increases by more than two orders of magnitude and is far higher than the outdoor concentration. At the same time, the equivalent black carbon mass concentration increases by a factor of eight.

5.2 Time series of the indoor and outdoor relationship

Figure 10 shows an example of one private home in Leipzig. One can clearly see the nighttime period from 00:00 to 08:00 with no activities and the active period from 08:00 to

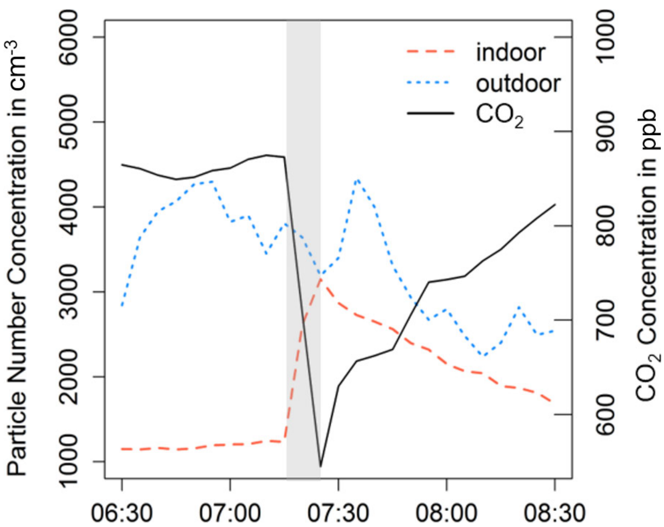


Figure 8. Example of time series of indoor and outdoor particle number concentrations, and indoor CO_2 concentration, the shaded area marks the time of open windows.

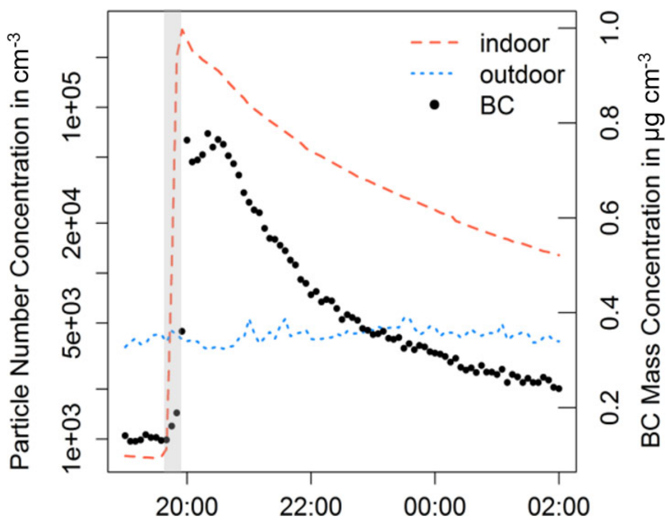


Figure 9. Example of time series of indoor particle number concentration, and indoor black carbon concentration, the shaded area indicates the time when a toaster was used.

24:00. Figure 10a shows the time series of the median and 25-75 percentile of PNCs measured indoors and outdoors. From the median of the indoor and outdoor PNCs, one can see that the outdoor PNC is generally higher than the indoor one. During nighttime, both indoor and outdoor PNCs decreased constantly due to less traffic and no indoor emissions. On the other hand, the indoor PNC and I/O ratio show peaks during breakfast, lunch and dinnertime. Ultrafine aerosol particles are produced, especially due to cooking activities or candle burning.

The influence of the outdoor aerosol on the indoor PNC or PNSD is usually described by the ratio between indoor and outdoor (I/O ratio) of the PNC or PNSD. In this example, the I/O ratio was calculated by dividing the indoor PNC by the outdoor PNC (10 to 800 nm) for each 5 min measurement. Figure 10b shows the time series of the median and 25 to 75 percentile of I/O ratio. The median I/O ratio is in the range of 0.5 to 2.4. During nighttime, the indoor aerosol is increasingly decoupled from outdoor, while during daytime the influence of the outdoor aerosol is more significant (I/O ratio close to 1.0). The 75 percentile of I/O ratio is however in the range of 0.7 to 8.5, showing the presence of indoor particle sources, mainly to activities related to combustion or heating processes.

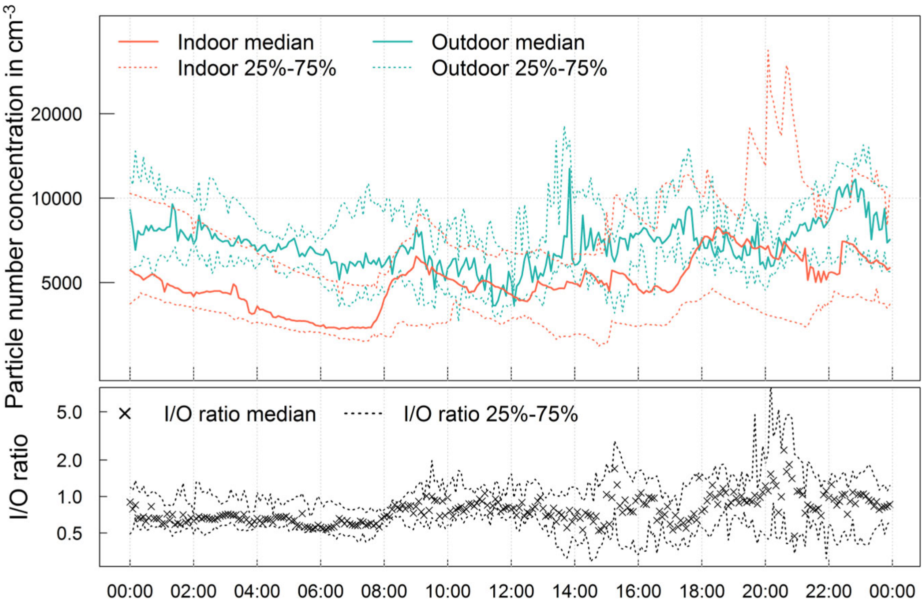


Figure 10. Time series of (a) the indoor and outdoor median PNC, and (b) I/O (indoor to outdoor) ratio measured at a private home over a period of one week.

5.3 Indoor source

Figure 11 shows an example of the indoor PNSD before, during and after toasting. The black dots represents PNSD before heating, while the red dotd show the PNSD directly after toasting. It can be seen that the majority of the emitted particle are in the range around 20 nm particle diameter. In the following, the particles grow by coagulation or are lost by diffusion to surfaces in the living room. The peak of PNSD consequently moves from approximately 20 to 50 nm, while the PNC drops by more than a factor of ten.

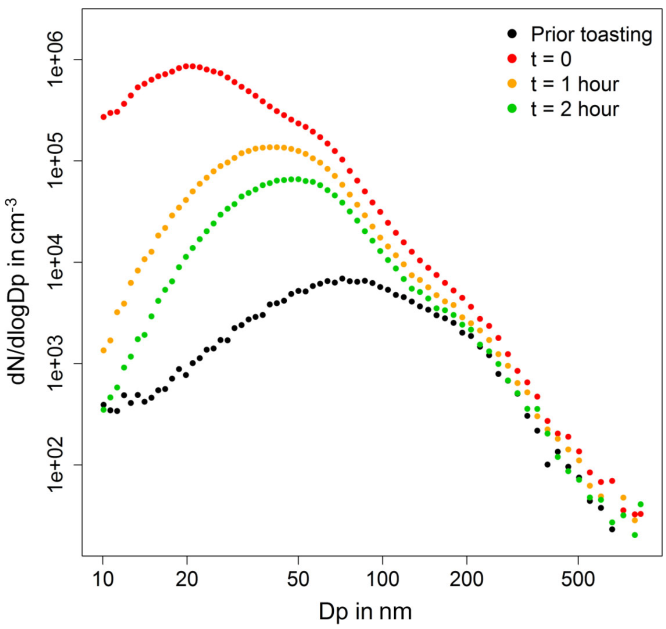


Figure 11. Example of one toasting event, PNSD of prior (black dots), during (red dots) and after toasting (orange and green dots).

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References

[1] *Atkinson, R. W.; Anderson, H. R.; Sunyer, J.; Ayres, J.; Baccini, M;. Vonk, J. M.; Boumghar, A.; Forastiere, F.; Forsberg, B.; Touloumi, G.; Schwartz J.; Katsouyanni, K.*: Acute effects of particulate air pollution on respiratory admissions: results from APHEA 2 project. Air Pollution and Health: a European Approach. Am. J. Respir. Crit. Care Med. 164 (2001) No. 10, pp. 1860-1866.

[2] Human respiratory tract model for radiological protection. A report of a Task Group of the International Commission on Radiological Protection. Ann. ICRP 24 (1994) No. 1-3, pp.1-482.

[3] *Kreyling, W. G.; Semmler, M.; Erbe, F.; Mayer, P.; Takenaka, S.; Schulz,H.; Oberdörster, G.; Ziesenis, A.*: Translocation of ultrafine insoluble iridium particles from lung epithelium to ex-

trapulmonary organs is size dependent but very low. J. Toxicol. Environ. Health, Part A, 65 (2002) No. 20, pp. 1513-1530.
[4] *Arhami, M.; Minguillón, M.C.; Polidori, A.; Schauer, J. J.; Delfino R. J.; Sioutas, C.*: Organic compound characterization and source apportionment of indoor and outdoor quasi-ultrafine particulate matter in retirement homes of the Los Angeles Basin. Indoor Air 20 (2010) No. 1, pp. 17-30.
[5] *Franck, U.; Odeh, S.; Wiedensohler, A.; Wehner, B.; Herbarth, O.*: The effect of particle size on cardiovascular disorders –The smaller the worse. Sci. Total Environ. 409 (2011) No. 20, pp. 4217-4221.
[6] *Oberdörster, G.*: Pulmonary effects of inhaled ultrafine particles. Int. Arch. Occup. Environ. Health 74 (2000) No. 1, pp. 1-8.
[7] Air Quality Guidelines: Global Update 2005: Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur Dioxide. Edited by: World Health Organization, 2006.
[8] *Brasche, S.; Bischof, W.*: Daily time spent indoors in German homes – Baseline data for the assessment of indoor exposure of German occupants. Int. J. Hyg. Environ. Health 208 (2005) No. 4, pp. 247-253.
[9] *Odeh, I.; Hussein, T.*: Activity pattern of urban adult students in an Eastern Mediterranean Society. Int. J. Environ. Res. Public Health 13 (2016) No. 10, pp. 960.
[10] *Franck, U.; Tuch, T.; Manjarrez, M.; Wiedensohler, A.; Herbarth, O.*: Indoor and outdoor submicrometer particles: exposure and epidemiologic relevance („the 3 indoor Ls“). Environ. Toxicol. 21 (2006) No. 6, pp. 606-613.
[11] *Uhde, E.; Salthammer, T.*: Impact of reaction products from building materials and furnishings on indoor air quality – a review of recent advances in indoor chemistry. Atmos. Environ. 41 (2007) No. 15, pp. 3111-3128.
[12] *Meng, Q. Y.; Turpin, B. J.; Korn, L.; Weisel, C. P.; Morandi, M.; Colome, S.; Zhang, J.; Stock, T.; Spektor D.; Winer, A.*: Influence of ambient (outdoor) sources on residential indoor and personal PM2.5 concentrations: analyses of RIOPA data. J. Expo. Anal. Environ. Epidemiol. 15 (2005) No. 1, pp. 17-28.
[13] *Hussein, T.; Hämeri, K.; Heikkinen M. S. A.; Kulmala, M.*: Indoor and outdoor particle size characterization at a family house in Espoo, Finland. Atmos. Environ. 39 (2005) No. 20, pp. 3697-3709.
[14] *Morawska, L.; He, C.; Loveday, J.; Parappukkaran S.*: The relationship between indoor and outdoor airborne particles in the residential environment. Atmos. Environ. 35 (2001) No. 20, pp. 3463-3473.
[15] *Hussein, T.; Dada, L.; Juwhari H.; Faouri, D.*: Characterization, fate, and re-suspension of aerosol particles (0.3-10 µm): The effects of occupancy and carpet use. Aerosol Air Qual. Res. 15 (2015), pp. 2367-2377.
[16] *Hussein,T.; Kubincová, L.; Džumbová, L.; Hruška, A.; Dohányosová, P.; Hemerka J.; Smolík, J.*: Deposition of aerosol particles on rough surfaces inside a test chamber. Building and Environment 44 (2009) No. 10, pp. 2056-2063.
[17] *Morawska, L.; Afshari, A.; Bae, G. N.; Buonanno, G.; Chao, C. Y.; Hanninen, O.; Hofmann, W.; Isaxon, C.; Jayaratne, E. R.; Pasanen, P.; Salthammer, T.; Waring, M.; Wierzbicka, A.*: Indoor aerosols: from personal exposure to risk assessment. Indoor Air 23 (2013) No. 6, pp. 462-487.
[18] *Chen, C.; Zhao, B.*: Review of relationship between indoor and outdoor particles: I/O ratio, infiltration factor and penetration factor. Atmos. Environ. 45 (2011) No. 2, pp. 275-288.
[19] *Geller, M. D.; Chang, M.; Sioutas, C.; Ostro B. D.; Lipsett, M. J.*: Indoor/outdoor relationship and chemical composition of fine and coarse particles in the southern California deserts. Atmos. Environ. 36 (2002) No. 6, pp. 1099-1110.
[20] *Hassanvand, M. S.; Naddafi, K.; Faridi, S.; Arhami, M.; Nabizadeh, R.; Sowlat, M. H.; Pourpak, Z.; Rastkari, N.; Momeniha, F.; Kashani, H.; Gholampour, A.; Nazmara, S.; Alimohammadi, M.; Goudarzi G.; Yunesian, M.*: Indoor/outdoor relationships of PM10, PM2.5, and PM1 mass concentrations and their water-soluble ions in a retirement home and a school dormitory. Atmos. Environ. 82 (2014), pp. 375-382.
[21] *Wiedensohler, A.; Wiesner, A.; Weinhold, K.; Birmili, W.; Hermann, M.; Merkel, M.; Müller, T.; Pfeifer, S.; Schmidt, A.; Tuch,T.; Velarde, F.; Quincey, P.; Seeger, S.; Nowak, A.*: Mobility particle size spectrometers: Calibration procedures and measurement uncertainties. Aerosol Sci. Technol. 52 (2018) No. 2, pp. 146-164.
[22] *Wiedensohler, A.; Birmili, W.; Nowak, A.; Sonntag, A.; Weinhold, K.; Merkel, M.; Wehner, B.; Tuch, T.; Pfeifer, S.; Fiebig, M.; Fjäraa, E. A. A. M.; Sellegri, K.; Venzac, H.; Villani, P.; Laj, P.; Aalto, P.; Ogren, J. A.; Swietlicki, E.; Roldin, P.; Williams, P.; Quincey, P.; Hüglin, C.; Fierz-Schmidhauser, R.; Gysel, M.; Weingartner, E.; Riccobono, F.; Santos, S.; Grüning, C.; Faloon, K.; Beddows, D.; Harrison, R.; Monahan, C.; Jennings, S. G.; O'Dowd, C. D.; Marioni,A.; Horn, H.-G.; Keck, L.; Jiang,J.; Scheckman, J.; McMurry, P. H.; Deng, Z.; Zhao, C. S.; Moerman, M.; Henzing, B.; de Leeuw, G.; Löschau, G.; Bastian, S.*: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions. Atmos. Meas. Tech. 5 (2012), pp. 657-685.
[23] *Pfeifer, S.; Birmili, W.; Schladitz, A.; Müller, T.; Nowak, A.; Wiedensohler, A.*: A fast and easy-to-implement inversion algorithm for mobility particle size spectrometers considering particle number size distribution information outside of the detection range. Atmos. Meas. Tech. 7 (2014) No. 1, pp. 95-105.