Physical properties, chemical composition, sources, spatial distribution and sinks of indoor aerosol particles in a university lecture hall

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Abstract

PM$_{10}$ mass, particle number ($N$) and CO$_2$ concentrations, particle number size distributions and meteorological parameters were determined with high time resolution, and daily aerosol samples were collected in the PM$_{10}$, PM$_{2.5}$ and PM$_{2.0}$ size fractions for chemical analysis in the middle of a university lecture hall for one week. Median concentrations for the PM$_{10}$ mass and $N$ of 15.3 mg m$^{-3}$ and 3.7 $\times$ 10$^3$ cm$^{-3}$, respectively were derived. The data are substantially smaller than the related outdoor levels or typical values for residences. There were considerable concentration differences for workdays, weekends and various lectures. Main sources of PM$_{10}$ mass include the usage of chalk sticks for writing, wiping the blackboard, ordinary movements and actions of students and cleaning. High PM$_{10}$ mass concentration levels up to 100 mg m$^{-3}$ were realised for short time intervals after wiping the blackboard. The mass concentrations decreased rapidly after the emission source ceased to be active. Two classes of coarse particles were identified. General indoor dust particles exhibited a residence time of approximately 35 min, while the residence time for the chalk dust particles was approximately 20 min as lower estimates. Emission source rate for wiping the blackboard was estimated to be between 8 and 14 mg min$^{-1}$. This represents a substantial emission rate but the source is active only up to 1 min. Suspension of the chalk (made mainly of gypsum) dust particles was confirmed by enrichment of Ca and S in the hall with respect to ambient urban aerosol. Contribution of ambient aerosol via the heating, ventilation and air conditioning (HVAC) facility was considerable for time intervals when the indoor sources of PM$_{10}$ mass were not intensive. The HVAC facility introduces, however, the major amount of aerosol particles from the outdoors as far as their number concentration is regarded. Mean contribution of ultrafine particles to the total particle number was (69 $\pm$ 7)%$, which is smaller than for the related outdoor urban environment. This can indicate aged ultrafine aerosol. The major amounts of CO$_2$ arrive from the corridors through open doors by infiltration. Spatial distribution of the PM$_{10}$ mass concentration within the hall was derived by CFD modelling, and spatial inhomogeneities were obtained.

1. Introduction and objectives

It is estimated that people in industrialised countries spend more than approximately 80% of their time indoors and in closed environments.
spaces (Jantunen et al., 1999; Schweizer et al., 2007). During this period, they are exposed to a broad range of pollutants of indoor and outdoor origin. Indoor emission sources and formation processes include outgassing from building materials, furniture, carpets and textiles, heating and cooking, smoking, chemicals and residues from cleaning and personal care, plants, pesticides, pets, mould, specific indoor physical activities such as cleaning or vacuum cleaning, some metabolic processes (Morawska and Salthammer, 2003), and indoor air chemistry (Fan et al., 2003). Outdoor pollutants can penetrate through open doors and windows, ventilation systems and the building envelope. Outdoor ambient air is often of better quality than indoor air, so one way of ensuring indoor air quality (IAQ) is by replacing interior air with outside fresh air. The lack of adequate ventilation is a frequent cause of poor IAQ. In indoor spaces occupied by people, CO₂ concentration is often used as a surrogate indicator of the adequacy of air ventilation relative to occupant density and metabolic activity (Seppälä et al., 1999). To eliminate most health complaints and discomfort feeling, indoor CO₂ should be less than approximately 600 ppm above the outdoor levels (ASHRAE, 2004).

Various research studies dealt with indoor aerosol. Their results and conclusions were summarised by account type or review publications by Wallace (1996, 2004), Lai (2002), Morawska and Salthammer (2003), and Holmes and Morawska (2006). It was shown that aerosol particles are a common class of indoor pollutants, and that they (particularly those of biological origin) represent serious health risks from air pollution indoors. Sources and properties of indoor aerosol exhibit large variability in time and space, as far as both the variability within a room (among its different compartments), and the changes from room to room of similar type (among different microenvironments) are concerned. It is, therefore, desirable to perform research studies in various closed spaces belonging to the same, well-defined type of indoor microenvironment to acquire a statistically significant knowledge which enables to identify both common tendencies and specific features of the indoor aerosol. Larger variations in the IAQ for mechanically ventilated halls are expected since the design and operating parameters can differ substantially (Zhong et al., 2010). Multiplicity of the factors affecting the physical and chemical characteristics of indoor aerosol particles also causes a large variation in their properties, and, therefore, it seems to be more useful to understand the nature and relative importance of the influential factors and their implications.

Educational institutions including university lecture halls are an important type of indoor environments (Janssen et al., 1997; Lee and Chang, 2000; Patterson and Eatough, 2000; Lee et al., 2002; Braniš et al., 2009, 2011; Guo et al., 2010; Szoboszlai et al., 2011). They are dedicated rooms equipped with facilities for teaching, usually equipped with ventilation and humidity control, and accommodate a larger number of young people periodically for a considerable time interval in one occasion. They should provide an acceptable feeling of comfort so the students and lecturer can focus constant attention on lectures. Considering all these, an intensive aerosol research program was conducted in a university lecture hall to follow up and complement the existing international studies for this specific indoor microenvironment. Our major objectives were to determine time-resolved aerosol mass and number concentrations, CO₂ concentration, and size-resolved chemical composition of aerosol particles, to evaluate them together with micrometeorological variables in order to understand the formation, behaviour and sinks of aerosol particles, and the effect of various indoor activities on the aerosol properties. A computational fluid dynamics (CFD) model was developed to estimate the spatial distribution of the experimental and derived data within the hall.

### 2. Applied methods

#### 2.1. Measurement location and its environment

The measurement and sample collection campaign was organised in the Rudolf Ortvay Lecture Hall of the Faculty of Science, Eötvös University (Budapest, 11th district, 1 Pázmány promenade) from Thursday, 8 April to Thursday, 15 April 2010 for one week in an education term-time. The building is situated next to the city centre in the downwind direction on the bank of the river Danube in a larger open area. Wind channels that often form above the river result in good overall ventilation of the whole location (Salma et al., 2011). The building was put into operation in 1998. Smoking inside the university buildings is not allowed. The lecture hall has a floor space of 126 m² and a capacity of 120 persons in eight bench rows. It is located on the ground and first floors, and has no windows. The total volume of the hall is 485 m³. The lecture hall has mechanical ventilation. Two air supply ducts are located below the ascending floor of the hall in service rooms which have connected air-spaces to the hall by eight rows of ventilation slots in each stair riser. Inflow velocities were measured, and the total ventilation flow-rate was 0.91 m³ s⁻¹⁻¹. Five rows of ventilation outlet ducts are situated in the ceiling. Four lecture halls located next to each other have a joint heating, ventilation and air conditioning (HVAC) facility. The outgoing air flows from the halls are joined and partially returned back to the hall. The recycled flow is mixed with fresh make-up air flow coming from an open outer area. The mixed flow passes through dry media bag filters (grade EU4 with an average arrestance of η > 90%) into the halls. The air turnover rate is approximately 6.8 h⁻¹. On workdays, the HVAC system is automatically switched on at 6:00 local time, and switched off at 21:00. The system can also be turned off and on manually from the lecture hall. There is no direct sunlight entering the hall. Ordinary blackboards and dustless white chalk sticks are used as writing and drawing implements. The chalk sticks utilised (Omega, Israel) are prepared from mineral gypsum (mainly CaSO₄ × 2H₂O) as the major component and mineral calcite (mainly CaCO₃). They also contain several percents of carbamyl cellulose, polyvinyl alcohol and starch as binder glue and thickener, and 1–2% of ZnO as additive to enhance the dustless property. The density of the sticks is 1.83 g cm⁻³. Occupancy of the hall by students during the lectures was counted by overlooking persons, and it was recorded into a logbook together with other relevant supporting information. There were no plants in the hall during the campaign. There are two tight doors for passage on the lower level and two tight doors for passage on the upper level of the hall located in opposite directions. The doors were usually closed during the lectures. They lead to corridors with glass roof.

The measuring sensors and sampling devices were set up on two metal poles located approximately in the middle of the hall in the bench area in heights from 1.5 to 2 m. The four surrounding seats were closed off by band bars for students. The controlling and data acquisition systems and the pumps were placed in the service rooms below the hall.

During the first 2.5 days of the campaign, there were anticyclonic weather conditions in the area of Budapest with warm (daily maximum temperatures up to 20–21 °C) and dry weather. A cold front passed the city in the afternoon of 10 April, and it was followed by a cyclonic system. The weather became colder (with daily maximum temperatures up to 11–15 °C) and there was 2–10 mm rain each day till the end of the campaign. Mean PM₁₀ mass, NOx, O₃ and SO₂ concentrations over the campaign — calculated from the measured data of the closest municipal air quality measuring station (OLM, 2012) in a distance of about 1.6 km from the building — were 22, 69, 37 and 5.4 µg m⁻³, respectively.
2.2. Measuring instruments and sample collection devices

Particulate mass concentration for the PM$_{10}$ size fraction was measured on-line by a tapered-element oscillating microbalance equipped with a PM$_{10}$ inlet (TEOM, model 1400a, Rupprecht and Patashnick, USA). It was operated with the filter heated to 40 °C to prevent moistening. Dependence of the standard deviation of the baseline on the integration time was investigated in a laboratory experiment earlier (Salma et al., 2007), and a time resolution of 1 min was adopted on the basis of its conclusions. Number size distribution of aerosol particles was measured by a flow-switching type differential mobility particle sizer (DMPS) with aerosol flows of 2 and 0.3 L min$^{-1}$ for an electrical mobility diameter range from 6 to 1000 nm in 30 channels (Salma et al., 2011). There was no upper size cut-off inlet deployed. Diameters of particles in their dry state were pre-equilibrated before weighing at 20 °C, and were measured on-line by a tapered-element oscillating microbalance using a box model as:

$$\frac{dc_{in}}{dt} = \frac{1}{V} \frac{dq_{in}}{dt} = \frac{1}{V} (P + F_{in} - R - F_{out}), \quad (1)$$

where $q_{in}$ is the amount of the material in the air of the hall (box), $V$ is the total volume of the hall, $P$ is the sum of the production rates for all sources of the material in the hall, $R$ is the sum of the removal rates of the material due to all sinks in the hall, and $F_{in}$ and $F_{out}$ are the material flows into and out of the hall, respectively. It is commonly assumed that the material is uniformly mixed within the hall. The validity of this assumption is linked to the comparison of the extent of the concentration difference to the actual concentration level, and it only holds with limitations. The material flows for the halls with a HVAC facility can be expressed as (Abt et al., 2000):

$$F_{in} = c_{out} p Q_{inf} + (1 - \eta) (c_{in} Q_{ina} + c_{in} Q_{ret}). \quad (2)$$

$$F_{out} = c_{in} Q_{out} + c_{in} Q_{exh} + c_{in} Q_{ret}. \quad (3)$$

where $c_{out}$ is the concentration in the outdoor air, $p$ is the penetration factor, $Q_{ina}, Q_{ret}, Q_{exh}$ and $Q_{out}$ are the volumetric airflow rate from the outdoors to the HVAC system (make-up flow rate), from the HVAC system back to the outdoors (return flow rate), from the indoors to the outdoors by the HVAC system (exhaust flow rate), the natural volumetric airflow rate from the outdoors to the indoors (infiltration flow rate), and from the indoors to the outdoors (exfiltration flow rate), respectively, and $\eta$ is the efficiency of the HVAC filter. Gravitational sedimentation and diffusional deposition were considered as the major sinks for the particles that mainly contribute to the PM$_{10}$ mass concentration. Both surface deposition processes can be described by first order kinetics as $R = k q_{in}$, where $R$ is the rate for the two processes and $k$ is their joint rate coefficient (Hinds, 1999; He et al., 2005). If the source and sink terms can be considered as time-independent over the time of interest, then the solution of Eq. (1) utilising Eqs. (2) and (3) yields an analytical expression of (Koutrakis et al., 1992):

$$c_{in} = (c_0 - c_w) \exp[-(\alpha + \eta \beta + k)t] + c_w = (c_0 - c_w) \exp[-(\alpha + k)t] + c_w, \quad (4)$$

$$c_0 \text{ is the concentration at } t = 0, \quad c_w \text{ is the steady-state concentration, } \eta \text{ is the air exchange rate, and } \beta = \frac{Q_{exh}}{Q_{inf}} \text{ is the return (recirculated) air exchange rate. Since } \alpha, \beta \text{ and } \eta \text{ are characteristics of the HVAC facility, they are denoted jointly as } a = \alpha + \eta \beta. \text{ On the basis of Eq. (4), the}$$

Al, Si, P, S, Cl, K, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, Br, Sr, Ba and Pb in the in-vacuum PIXE chamber at the Laboratory of Ion Beam Applications using a proton beam with an energy of 2 MeV and with a current of 40 nA (Borbély-Kiss et al., 1985). The quartz filters were analysed by a real-time RT-OC/EC analyser (Sunset Laboratory, USA) in its off-line mode to determine the concentrations of organic carbon (OC) and elemental carbon (EC). A temperature program close to the NIOSH protocol was selected. The field blank filters were analysed, and the data obtained from them were used for blank corrections. The measured DMPS data were utilized for calculating concentrations of particles in the diameter ranges from 6 to 1000 nm and from 6 to 100 nm. The former size fraction represents the total number of aerosol particles ($N$), while the latter fraction represents ultrafine particles ($N_{Uf}$, Salma et al., 2011). Particle number size distributions were also derived from the DMPS data by inversion, and were fitted by lognormal functions to obtain modal concentrations, number median mobility diameters (NMMMDs) and geometric standard deviations for the modes identified.

Change of the concentration $c_{in}$ of a material of interest in the hall as function of time $t$ can be expressed from the material balance using a box model as:
residence time of (coarse) particles \( t = 1/(a + k) \) can be derived from a decay curve analysis of the measured mass concentration data. The residence time estimated in this way is related to further uncertainties if stratification in the concentration is realised within the hall.

Eqs. (1)–(3) and the calculated residence time can be further utilised to estimate the emission rate \( P \) for the material of interest. It was assumed that 1) the infiltration and exfiltration flow rates are negligible in comparison with the sum of the exhalation and return flow rates when the doors of the hall are closed, 2) the make-up, return and exhaust flow rates, and the filtration efficiency are constant. The differential of \( c_{\text{in}} \) in time was replaced by its finite difference over a time interval \( \Delta t \) for which the concentration increase is considered to be linear, and, furthermore, the instant value of \( c_{\text{in}} \) was replaced by its mean value for the same time interval \( \Delta t \). By adopting these sensible simplifications and approximations, the mean emission source rate can be estimated as:

\[
P = V \frac{\Delta c_{\text{in}}}{\Delta t} + (a + k) \bar{c}_{\text{in}} = V \frac{\Delta c_{\text{in}} + \bar{c}_{\text{in}}}{\Delta t}.
\]

(5)

It was also assumed implicitly that the material under investigation has a conservative character in the hall, thus its transformation processes can be neglected. For aerosol particles such processes generally involve nucleation, condensation and evaporation. These can be indeed ignored for coarse particles and for overall conditions normally encountered in ordinary, non-industrial indoor environments (Hinds, 1999).

A transient three-dimensional CFD model was developed with the purpose of qualitative analysis of flow phenomena as well as the transport of particles emitted from the dominant source. It was utilized to extend the interpretation of the measured data for a single point to the larger space, and model validation was not among the investigation aims. The model — set up in the ANSYS-FLUENT simulation system (version 14.0) — was built of finite volume mesh consisting of mostly hexahedral elements, and it had an average spatial resolution of 0.1 m. The flow field in the hall was treated jointly with that of the service rooms. Identical air extraction intensity was assumed through the five ventilation outlet ducts. Unsteady Reynolds-averaged Navier—Stokes model was applied with a temporal resolution of 1 s. Physical dimensions and structure of the indoor spaces, HVAC system, and the measured \( T_{\text{air}}, T_{\text{surf}} \) and ultrasonic sensor WS data, together with the occupancy records in the logbook were utilised as input parameters for the model. Human heat sources were only taken into account because the power of the heat sources was more aged with respect to general ambient urban aerosol. This is also related to the location of the building (see Sect. 2.1).

### Table 1

<table>
<thead>
<tr>
<th>Variable [unit]</th>
<th>Type</th>
<th>All days</th>
<th>Workdays</th>
<th>Weekends</th>
<th>Opening hours</th>
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<tr>
<td>PM_{10} [\mu g m^{-3}]</td>
<td>Minimum</td>
<td>2.6</td>
<td>3.4</td>
<td>2.6</td>
<td>7.1</td>
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<td></td>
<td>Median</td>
<td>15.3</td>
<td>17.7</td>
<td>11.1</td>
<td>24</td>
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<tr>
<td></td>
<td>Maximum</td>
<td>101</td>
<td>101</td>
<td>30</td>
<td>101</td>
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<tr>
<td>( N \times 10^{-3} ) [cm^{-3}]</td>
<td>Minimum</td>
<td>1.36</td>
<td>1.36</td>
<td>1.76</td>
<td>1.59</td>
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<tr>
<td></td>
<td>Median</td>
<td>3.7</td>
<td>4.0</td>
<td>3.0</td>
<td>5.8</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>10.6</td>
<td>10.6</td>
<td>8.3</td>
<td>10.6</td>
</tr>
<tr>
<td>( N_{\text{ultra}} \times 10^{-3} ) [cm^{-3}]</td>
<td>Minimum</td>
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<td>0.81</td>
<td>1.00</td>
<td>1.09</td>
</tr>
<tr>
<td></td>
<td>Median</td>
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<td>3.0</td>
<td>2.0</td>
<td>4.2</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>8.3</td>
<td>8.3</td>
<td>6.3</td>
<td>8.3</td>
</tr>
<tr>
<td>CO_{2} [ppm]</td>
<td>Minimum</td>
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<td>390</td>
<td>381</td>
<td>401</td>
</tr>
<tr>
<td></td>
<td>Median</td>
<td>402</td>
<td>415</td>
<td>392</td>
<td>589</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>1405</td>
<td>1405</td>
<td>530</td>
<td>1405</td>
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<tr>
<td>( T_{\text{air}} ) [\degree C]</td>
<td>Minimum</td>
<td>22</td>
<td>22</td>
<td>23</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>Median</td>
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<td>26</td>
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<tr>
<td></td>
<td>Maximum</td>
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<td>RH [%]</td>
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<td>17.6</td>
<td>15.6</td>
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</tr>
<tr>
<td></td>
<td>Median</td>
<td>28</td>
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<tr>
<td></td>
<td>Maximum</td>
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<td>48</td>
<td>28</td>
<td>48</td>
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</table>

ambient \( PM_{10} \) concentration of 22 \( \mu g m^{-3} \). The median concentration is also smaller two or three times than for naturally ventilated domestic residences, restaurants and malls, and is comparable to air-conditioned offices (Lee et al., 2002; He et al., 2004). Median concentration for the workdays was larger than for the weekend by approximately 60%. The difference is caused by internal emission sources, which are more intense on workdays than on weekends.

Median particle number concentration \( N \) in the hall of \( 3.7 \times 10^{3} \) cm\(^{-3} \) is much smaller than the yearly median \( N \) of 11.8 \times 10^{3} \) cm\(^{-3} \) for the same outdoor location (Salma et al., 2011). The median indoor concentration corresponds to the near-city background location in Budapest with a median \( N \) of 3.6 \times 10^{3} \) cm\(^{-3} \). Median \( N \) concentration for the workdays was larger than for the weekend by 50%. This implies that there are production processes for aerosol particles active within the hall and building, or that the ambient \( N \) concentration levels are generally higher for workdays than for weekends. The median concentration is also smaller than for ordinary residential environments with natural ventilation exhibiting average values of approximately \((1–2) \times 10^{3} \) cm\(^{-3} \) (Morawska et al., 2001; Wallace and Howard-Reed, 2002; Franck et al., 2003; He et al., 2004). The maximum measured data was smaller than ordinary concentrations near some specific indoor sources (such as cooking, vacuum cleaning and smoking) by up to 3—4 orders of magnitude (Morawska et al., 2003). There are few studies conducted in mechanically ventilated offices and spaces (Fisk et al., 2000; Koponen et al., 2001; Wang et al., 2010). The present median particle number concentration is smaller than the typical value for office buildings. Mean contribution and standard deviation of ultrafine particles to total particle numbers were \((69 \pm 7\%) \). The mean value was practically constant for the different time intervals considered, and it is substantially smaller than for various outdoor urban environments in Budapest, i.e., \((77 \pm 9\%) \) for a near-city background, \((79 \pm 6\%) \) for the city centre, \((85 \pm 3\%) \) for a street canyon, and \((85 \pm 2\%) \) for a road tunnel. A possible reason for this small ultrafine contribution could be that the smallest particles were lost in the HVAC facility more that the larger particles. Another contribution is that their emission and formation processes are likely located in a larger distance from the receptor site, and that the indoor aerosol was more aged with respect to general ambient urban aerosol. This is also related to the location of the building (see Sect. 2.1).

Median CO\(_{2}\) concentration of 402 ppm was much smaller than the recommended limit of 992 ppm for indoor spaces (i.e., 600 ppm
above the ambient median concentration level of 392 ppm for the weekend). The minimum concentrations for the different time intervals considered agreed within the experimental uncertainty interval of \(\pm 5\%\). Median CO\(_2\) concentration for the workdays was larger than for the weekend by approximately 28\%. The difference is related to internal emission sources, mainly respiration of humans, which are larger on workdays when the building is more occupied by students than on weekends.

Average meteorological parameters basically reflected the outdoor weather situations that were modified by the HVAC facility and occupants as heat source. The RH data were rather low in general. Till 11 April, the RH was small because of the dry weather, and after this date, the weather was wet but colder, which caused that the RH of the incoming air flow remained small.

### 3.2. Temporal variations

Time variations of the PM\(_{10}\) mass, \(N\) and CO\(_2\) concentrations, \(T_{\text{air}}\) and RH over the time interval studied are shown in Fig. 1. The time variation of ultrafine particles closely followed the total particle number (with a correlation coefficient of 0.985), and, therefore, it is not displayed. In a few occasions on workdays and for short time intervals, the PM\(_{10}\) mass concentration was larger than the EU 24-h health limit. All measured indoor \(N\) data were smaller — usually much smaller — than the yearly median ambient \(N\) concentration of \(11.8 \times 10^4\) cm\(^{-3}\) \(\pm 7\) (Salma et al., 2011). Measured concentrations of CO\(_2\) were usually below the recommended value. On Tuesday, 13 April, the indoor CO\(_2\) concentration was above the limit in three occasions for some ten minutes each. Air temperature on workdays with active HVAC system varied according to its daily pattern for the ambient air which shows a maximum typically in the early afternoon. Its level was usually rather constant during nights when the HVAC system was switched off. Variability of the PM\(_{10}\) mass concentration was the largest; it changed by a factor of about 40, while the variability of the \(N\) and CO\(_2\) concentrations were much smaller. They only changed by factors of approximately 8 and 4, respectively. Correlation coefficients between PM\(_{10}\) mass and CO\(_2\), between PM\(_{10}\) mass and \(N\), and between CO\(_2\) and \(N\) for the whole interval were 0.376, 0.573 and 0.043, respectively. These imply that the major sources of particulate mass, particle number and CO\(_2\) in the hall are not directly connected to each other. The air temperature changed together with the CO\(_2\) concentration during the daytime intervals when the HVAC system was switched off, e.g., on Tuesday, 13 April. This means that their major sources were identical. This is unambiguously the metabolism of humans. The largest concentration levels were realised when the HVAC facility was switched off manually during the opening hours, and when the indoor sources were active and frequent (see Sect. 3.3). During the weekend and nights on workdays, changes in the concentrations were only modest (see also Table 1). Concentration of the CO\(_2\) had returned to a constant (ambient) value of approximately 390 ppm every evening before the HVAC system was switched off. The aerosol concentration levels for the weekend and nights on the workdays were, however, influenced by their actual values at the time when the HVAC system was turned off, and, therefore, the mean levels for these time intervals were not identical.

### 3.3. Major sources and sinks

On workdays, sudden changes on the PM\(_{10}\) mass and CO\(_2\) concentration curves occurred usually in the form of peaks, while changes for the \(N\) concentration resemble more a fluctuation. Detailed variations of the PM\(_{10}\) mass, \(N\) and CO\(_2\) concentrations from 7:00 to 18:00 (during which the hall was opened) on Monday, 12 April are shown in Fig. 2 as example. The HVAC system was

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**Fig. 1.** Time variations of PM\(_{10}\) mass concentration, total particle number concentration (\(N\)), and CO\(_2\) concentration, air temperature (\(T_{\text{air}}\)) and relative humidity (RH) over the whole campaign. The EU 24-h health limit for PM\(_{10}\) mass and advised concentration limit for CO\(_2\) are indicated by horizontal lines in the corresponding panels. Time periods when the HVAC system was in operation are indicated by shaded areas.
active during this interval. The lecture hall was cleaned by one person for 5–10 min, and the blackboard was washed by sponge at 7:10. These activities resulted in a substantial increase in the PM10 mass concentration and influenced little the N and CO2 concentrations. There were five lectures that day labelled as L1 through L5 in Fig. 2. It is seen that CO2 concentration peaks occurred before and after the lectures, when the students arrived to or departed from the hall. The peaks were assigned to intensive infiltration from the corridors through the opened doors. This is confirmed with the large CO2 peak at 16:05 which was generated by a door that was reopened in the beginning of the lecture, and was closed only after some minutes during the lecture. The extent of the infiltration (the area of the band of peaks) is related to the number of students moving in and out, thus to the frequency of door opening, its duration and to the CO2 concentration in the corridors. The very small number of students who arrived at the hall around 10:00 and the large time span of more than 45 min to the next lecture explain why there is no increase in the CO2 concentration before the lecture L2. The two expected CO2 concentration peaks between the lectures L4 and L5 were merged into a single peak due to the shorter time break of 15 min between these two lectures instead of regular breaks that last for 30 min. No chalk was utilised at all during the lectures L2, L3 and L5, while it was intensively applied during the lecture L4. Wiping of the blackboard by dry sponge at 14:15, 14:43, 14:58, 15:08, 15:21 and 15:32 was noted by overlooking persons, and this activity coincides with the concentration peaks of PM10 mass occurring with a mean delay time of 2.8 min. It is worth recognising that the total time interval — for which the actual concentration was above the EU 24-h health limit value — added up to approximately half of the L4 lecture time. At the same time, the baseline levels for the PM10 mass concentration during the lectures were similar to each other. It also suggests that the filtration efficiency of the HVAC facility for the aerosol particles which have large contribution to the PM10 mass (coarse particles) is satisfactory. The situation was different for the day (i.e., 13 April, see Fig. 1) when the HVAC system was mostly switched off manually. Concentration of CO2 exhibited an (exponential) increase during the lectures most likely due to the occupants’ respiration, and the PM10 mass concentration levels changed irregularly but were higher than for the lectures when the ventilation was switched on.

Particle number concentrations could not be connected to the activities discussed above. Particle numbers are dominated by
nucleation-, Aitken- and accumulation-mode particles (see the \(N_{10}/N\) ratio in Sect. 3.1), and they are basically non-resuspendable by physical activities mentioned above under ordinary indoor conditions (Thatcher and Layton, 1995; Fisk et al., 2000). These particles are generated by high temperature direct emissions or atmospheric nucleation. There was no new mode (i.e., the nucleation mode) appearing below 10 nm in the contour plots (displaying the joint variation of the particle number concentration and particle diameter in time), which means that no new particle formation and growth event happened in the hall. Similarly, no intense and persistent high-temperature emission source was identified and localised on the contour plots. The individual particle number size distributions invariably contained the Aitken and accumulation modes. They could be resolved by fitting. The accumulation mode was larger than the Aitken mode which is typical for distant emission sources. Moreover, the mean NMMDs for the Aitken and accumulation modes were approximately 45 and 130 nm, respectively. The values are larger than the ambient data of 26 and 93 nm, respectively at the same urban location (Salma et al., 2011) indicating again aged aerosol. This all implies that most particles — with respect to their number concentration — were transported from the outdoor air by the HVAC facility, and, therefore, their indoor concentrations are mainly influenced by the outdoor levels. There are several emission sources and formation mechanisms of aerosol particles active in the ambient air in Budapest, and their intensities are rather complex in time and space (Salma et al., 2011). This caused the time dependency that recalls fluctuations.

3.4. Residence times and emission source rate

Residence time of the coarse particles was estimated from the time evolution of its concentration after huge and isolated peaks (e.g., at 7:10, 14:21 and 15:41 in Fig. 2) after the major indoor source ceased to operate. The decay curve analysis after cleaning (i.e., after the peak at 7:10 in Fig. 2) resulted in a residence time of 34 min, while the decay curves belonging to the other peaks consisted of two components with mean residence times of 17 min and 32 min, respectively. These two residence times can most likely be associated with two classes of coarse particles present in the lecture hall. The longer time is related with general indoor dust particles, while the shorter value can be likely linked to suspended chalk particles. It was implicitly assumed in the procedure that the effect of all the other ordinary sources can be neglected or regarded to be constant over time, which means that no new particle formation and growth event happened in the hall. Similarly, no intense and persistent high-temperature emission source was identified and localised on the contour plots. The individual particle number size distributions invariably contained the Aitken and accumulation modes. They could be resolved by fitting. The accumulation mode was larger than the Aitken mode which is typical for distant emission sources. Moreover, the mean NMMDs for the Aitken and accumulation modes were approximately 45 and 130 nm, respectively. The values are larger than the ambient data of 26 and 93 nm, respectively at the same urban location (Salma et al., 2011) indicating again aged aerosol. This all implies that most particles — with respect to their number concentration — were transported from the outdoor air by the HVAC facility, and, therefore, their indoor concentrations are mainly influenced by the outdoor levels. There are several emission sources and formation mechanisms of aerosol particles active in the ambient air in Budapest, and their intensities are rather complex in time and space (Salma et al., 2011). This caused the time dependency that recalls fluctuations.

Emission source rates between 8 and 14 mg min\(^{-1}\) were derived with a mean and standard deviation of \((11.1 \pm 2.6)\) mg min\(^{-1}\). The actual value clearly depends on the coverage of the blackboard area by chalk ripples and the intensity of wiping. The mean value is comparable to but larger than for other indoor activities studied so far, e.g., for cooking \((1.7 \pm 0.6)\) mg min\(^{-1}\), frying \((2.7 \pm 2.2)\) mg min\(^{-1}\), grilling \((2.8 \pm 17.8)\) mg min\(^{-1}\) or smoking \((0.99 \pm 0.81)\) mg min\(^{-1}\), all unspecified data from He et al., 2004) obtained for the PM\(_{2.5}\) mass. The difference between the present rate and the other values listed is reasonable because dust particles are generated by mechanical disintegration, and, thus, they are larger than those produced by high temperature emission sources, and, therefore, their contribution to the mass concentration is substantially larger. Wiping the whole blackboard caused a linearly increasing concentration for only 1 min. As a result, the concentration levels reached in the lecture hall are usually much smaller than for the other indoor activities mentioned above, which can be active for several hours (Morawska and Salthammer, 2003).

3.5. Spatial distributions

Three dimensional geometrical model of the lecture hall as the starting point of the CFD analysis, and spatial distribution of air temperature and air flow in three longitudinal cross section planes as contour plots are shown in Fig. 3. Two of the planes (numbered as 1 and 3) are in a distance of 3 m from the side walls, and the remaining plane (numbered as 2) is in the middle of the hall. The contour plots of the temperature show that a stable vertical stratification developed with a difference of approximately 3 °C. Human heat sources can be clearly identified as rising air plumes. Under the benches, lower temperatures are observed because of the fresh, colder incoming air from the service rooms, which is not mixed with the warmer indoor air. As far as the velocity distribution is concerned, relatively large (approximately 0.2 m s\(^{-1}\)) differences can be only seen in the plumes of persons and under the benches. Typical air velocity front level is approximately 0.13 m s\(^{-1}\). The flow structure is rather complex. Natural convections caused by the internal heat sources are important elements of the flow field. Most horizontal components of the flow field converge from the blackboard toward the audience. There are differences in both the flow velocity and direction in the space above the benches. The first two and the last bench rows are poorly ventilated, the third and fourth rows seem to be over-ventilated, while the other rows receive ordinary ventilation. The air flows from the hall to the service room through the first several ventilation slots, while it flows from the service room to the hall via the other slots. The most intensive outflow is realised through the fourth ventilation slot. Advection and turbulence are the dominant factors of the transport process for PM\(_{10}\) aerosol particles. Turbulence — characterised by turbulent kinetic energy and turbulent dissipation rate — exhibited the largest intensity under the benches, near the ventilation inlets and above the heat sources.

Spatial distribution of PM\(_{10}\) mass concentration in 1 min steps after wiping the blackboard was also calculated. Distribution for 5 min is shown in Fig. 4 in form of an iso-concentration surface as example. It also indicates that the distribution of the particles is heterogeneous within the hall. A plume of particles is created along the central longitudinal band of the hall, while its spreading in the lateral directions is limited. The plume rises above the bench area near the first several rows, and reaches the students’ level at the back benches. As noted in Sect. 3.3, the measured concentration peaks lagged behind the times of the blackboard wiping. The typical calculated time needed for the coarse particles to arrive from the blackboard to the central part of the hall (at the measuring device) was approximately 3 min, while the plume reached the full length of
The hall in ca. 5 min. The modelled time development of the PM$_{10}$ aerosol mass concentration at the receptor point agreed with the measured time behaviour within a relative uncertainty of 15%. The students receive unequal exposure to coarse particles for short times after wiping the blackboard, and sitting at the outermost seats seems favourable from this point of view. The lecturers who use chalk sticks intensively for writing and wipe the blackboard dryly are exposed to higher concentration levels than the students.

Fig. 3. Isometric geometrical model of the lecture hall without the ceiling and side walls together with the distributions of air temperature and air flow both in three longitudinal cross section planes. The temperature is shown as a contour plot, while the flow is displayed as a contour plot for the flow velocity magnitude and as arrows for the flow directions.
The mean PM10 mass of 32 measured analytical data of Co, Ni and Br were very close to their iso-concentration surface 5 min after wiping the blackboard. Ambient aerosol in central Budapest (adopted from Salma and the indoor aerosol to the crustal EF of the same constituent for the average crustal rock composition (Mason and Moore, 1982) with Si averaging the TEOM data over the aerosol sample collection PM10 matter (PM) mass in the PM10 for elemental carbon (EC), organic carbon (OC), chemical elements and particulate mean atmospheric concentrations, crustal enrichment factors (EFs) and indoor EFs Table 2. Modelled three-dimensional spatial distribution of PM10 aerosol particle as an iso-concentration surface 5 min after wiping the blackboard.

3.6. Chemical composition and derived properties

Mean atmospheric concentrations of chemical constituents for the PM10-2.0 and PM2.0 size fractions are shown in Table 2. The measured analytical data of Co, Ni and Br were very close to their detection limits, and, therefore, these elements were not included. The mean PM10 mass of 32 μg m⁻³ — calculated as the sum of the PM10-2.0 and PM2.0 masses from the Nuclepore filters — agrees reasonably well with the mean PM10 mass of 28 μg m⁻³ derived by averaging the TEOM data over the aerosol sample collection intervals in particular if the known desorptive sampling artefact of the TEOM is considered.

Crustal enrichment factors (EFs) were calculated relative to the average crustal rock composition (Mason and Moore, 1982) with Si as the reference element. Ratio of the crustal EF of a constituent for the indoor aerosol to the crustal EF of the same constituent for the ambient aerosol in central Budapest (adopted from Salma and Maenhaut, 2006) expresses an indoor enrichment relative to the mean urban aerosol composition, and is called indoor EF. Crustal and indoor EFs are also shown in Table 2. The constituents have an indoor EF below 10¹ (usually below approximately 3), which suggests that they mainly originate from the ambient urban air via the HVAC system. Indoor EFs of PM10-2.0-fraction Ca and Sr were between 7 and 9 which indicate that their substantial indoor emission sources, while elevated indoor EFs of S, PM2.0-fraction K and PM10-2.0 mass imply their non-negligible indoor sources. The indoor EFs of the other constituents in the PM2.0 size fraction are close to unity suggesting that their major amounts arrive from the outdoor air. Blackboard chalk contains large amounts of Ca and S, and trace amounts of Sr. Its usage can explain the increased indoor abundances of the PM10-2.0-fraction Ca and S, and possibly Sr. Crustal EFs for S, Cl, Cu, Zn, Pb, PM2.0-fraction Ba and mass were large (usually above 40), which indicates their non-crustal, likely anthropogenic (urban) source types.

Concentration ratio of the PM2.0-fraction mass to the PM10-2.0-fraction mass averaged over all Nuclepore samples was 1.07. This value is larger than the mean mass ratio for the outdoor central urban locations in Budapest (Salma and Maenhaut, 2006) by a factor of 1.58. Most ratios for elements were also larger than their corresponding outdoor values by factors between 2 and 3. This indicates that the abundances of the fine constituents are larger for the indoor particles than for the ambient particles. The smallest factors were derived for S (1.35) and Ca (1.49). These two data were the only values that were also smaller than the factor for the particulate mass. It all implies again that there were indoor emission sources for coarse S and Ca in the hall. Usage of chalk sticks was assigned to them.

4. Conclusions

Mean mass and particle number concentrations of particulate matter were lower in the lecture hall with the HVAC system switched on than the corresponding general ambient concentration levels. Measured concentrations of CO2 were usually also smaller than the recommended health limit. The HVAC system is effective to maintain low CO2 concentration levels if the doors are closed. Large amounts of CO2 arrive from the corridors through opened doors by infiltration. Large concentrations of PM10 mass were realised for short time intervals. Their peak values were substantially larger than the 24-h health limit. The chalk usage and particularly wiping the blackboard were identified as the major emission sources. Durations of the concentration peaks add up to time intervals that can represent a considerable part of a lecture. There were spatial differences in the PM10 mass concentrations within the hall during the times when elevated PM10 levels occurred, and the lecturers are subjected to larger exposures than the students. More detailed spatial distributions can be derived by measurements at several points in the hall in parallel, which could also facilitate the comparison of the experimental data and modelled distributions. Resuspension of general indoor dust particles within the hall by mechanical and thermal turbulences caused by students are also important. Residence time of suspended chalk particles is approximately 20 min. Most aerosol constituents have smaller indoor concentrations than their outdoor counterparts. Most of them are transported from the ambient air via the HVAC system. At the same time, Ca and S showed larger indoor EFs than the other constituents. This is explained by the chalk usage for writing and drawing. The crustal enrichment factors and PM2.0/PM10-2.0 concentration ratios revealed that mechanical disintegration is the major source for many aerosol constituents. Concentration levels of the PM10 mass and CO2 for particular lectures are not influenced by the IAQ history in the hall if the HVAC system is in operation.
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