Exposure of Churchgoers to Airborne Particles

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Particle mass and number measurements in a church indicate significant increases of indoor particle concentrations during the burning of incense. Generally, varying concentration regimes can be attributed to different "modes of indoor activity" and emission sources. While periods of candle burning are negligible concerning particle concentrations, increases by a factor of 6.9 and 9.1 during incense burning were observed for PM₁₀ and PM₁, respectively. At maximum, indoor PM₁₀ shows an 8.1-fold increase in comparison to outdoor measurements. The increase of particles $<2~\mu m$ is significantly enhanced in comparison to larger particles. Due to a particle decay rate of 0.9 h⁻¹ post-service concentrations are elevated for a time span of $\sim\!24~h$ above indoor background concentrations.

1. Introduction

Research focusing on indoor particle concentrations and their potential impact on human health has increased largely during recent years. The studies cover a wide range of experimental sites and particle size distributions from coarse and fine to ultrafine particle fractions (1-3). In this process indoor aerosol mass and number concentrations were shown to reach significant concentration levels which temporarily are larger compared to simultaneous outdoor concentrations (4, 5). The indoor concentrations thereby depend on both the level of indoor activity, e.g., cooking, smoking, or cleaning, as well as the outdoor distribution of particles (6-9).

Particles are known to be significant contributors to health effects associated with respiratory, pulmonary, and cardio-vascular diseases (10-14). Regarding the fact that people in Central Europe spend 80-90% of their time indoors (e.g., 8) an increased knowledge of the temporal dynamics of indoor particles is important to assess the exposure to particle mass and number concentrations.

Recently, measurements of PM $_{10}$ during simulated candle and incense burning in a church showed that emissions of candles and incense are responsible for a 3-fold increase of PM $_{10}$ mass concentrations resulting in absolute concentrations of 658 μ g m $^{-3}$ (15). The authors concluded that regular exposure of churchgoers to high concentration levels might increase the risk of pulmonary diseases. However, they did not quantify the order of magnitude of either incense or candle emissions responsible for the increase of particle mass concentrations.

Several studies were published concentrating on the situation of air pollutants/particles and indoor meteorology in churches, however, most of them focused on the climatic situations and potential effects on cultural heritage (e.g., 16,

17). Besides the work of de Kok et al. (15) little is known about the effects of incense burning on indoor air quality in churches, although evidence of health effects from burning of incense was indicated in toxicological and epidemiological research (18, 19). Earlier measurements were conducted in Asian households or in the laboratory but due to significant differences in room sizes and mixing volumes those results are not directly comparable (20–22). Laboratory measurements thereby indicated large PM_{10} and $PM_{2.5}$ emission rates for incense but distinct variability with the chemical composition of different types of incense used (21).

The motivation of the present study was to estimate particle number and mass concentrations as well as particle dynamics in a church during the Christmas and New Year's Eve church services. During three mass services incense was burned, while ten "conventional" mass services without the burning of incense were held. The time period investigated was of special interest in the context of human exposure to particles due to the length of church services during Christmas (up to ~ 2.5 h) and the large number of churchgoers.

2. Materials and Methods

2.1 Study Site and Instrumentation. Measurements were performed from December 24, 2004 to January 5, 2005 within the Gothic Roman Catholic church of St. Engelbert, Mülheim/Ruhr, Germany (6° 53′ E, 51° 26′ N). The building has a capacity for roughly 450 visitors. Its interior outline is cross-shaped with dimensions of 48 m \times 20 m \times 16 m (length \times width \times height) resulting in a total interior volume of approximately 15,300 m³.

Particles were measured with two optical particle counters (OPC, Grimm Aerosol Germany, model 1.107) placed at 1.7 and 6.2 m above ground level (agl). The instruments were situated in the western corner of the church (not shown here). This was the most suitable measurement location inside the church to prevent measurement disturbances and influences by church visitors before or after service times. The OPC measured particles in the size range 0.3 μ m < particle diameter (D_n) < 32 μ m. However, in this study we focus on particles with aerodynamic diameters <10 μ m. The OPC measures particles by light scattering. Thereby the signal of a single particle passing a laser beam is counted by a recipient diode. The pulse height of the signal is detected by a multichannel classifier and measured as particle size distribution. The signal is then converted into the particle mass fractions PM₁₀, PM_{2.5}, and PM₁.

The OPC at 1.7 m agl measured mass and number concentration, while the second device of same construction installed at 6.2 m agl measured mass concentrations only. Due to incorrect software algorithms implemented within the second OPC the internal calculation of mass concentrations led to imprecise results. Therefore, absolute concentrations of both measurement heights cannot be compared directly, but relative comparison of the temporal dynamics of particle mass concentrations is possible. Data of both OPCs were sampled every 6 s and stored as 1 min averages.

To analyze the temperature stratification within the church interior a vertical air temperature profile (Thermistors, Thies Clima, Germany) was installed. Besides temperature measurements at 0.2, 1, 2, 4, 6, 7, and 8 m agl, relative humidity was measured at 7 m agl. The meteorological data were sampled every second and stored to a data logger (Combilog 1020, Th. Friedrichs, Schenefeld, Germany) as 1 min averages.

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To address differences in indoor and outdoor PM_{10} concentrations online data of an air quality measurement station (Mülheim-Styrum) of the North Rhine-Westphalia State Environment Agency (LUA NRW) were available. PM_{10} mass concentrations were measured by the tapered element oscillation microbalance method (TEOM) and processed as half-hourly values smoothed by an 8 h running mean. The station is situated at a distance of around 2.3 km to the northwest of the church and can be classified as characterizing the urban background concentration. Unfortunately, only PM_{10} was monitored at the station; fine particle mass concentrations were not available.

Particulate emissions from incense vary depending on the specific type and mixture of incense. In the present church a conventional incense (lat. Olibanum) from the resin of Boswellia carterii was used. The number of candles during service times did not vary notably between D and $D_{\rm INC}.$ During high mass at Christmas roughly 80 candles were burning in comparison to 20 candles during conventional church services.

During service times the building was warmed by means of hot-air blowers which are based at the church floor near to the walls. The heaters were running from approximately 30 min before to 30 min after church service times. The hotair input had a temperature of 16 °C and a flow velocity of around 1.2 m s⁻¹ at the exit and roughly 0.8 m s⁻¹ at 2 m agl (measured by a hand-held hot-bulb probe, Testoterm, model 490, Lenzkirch, Germany). The time periods during which heaters were turned on and off were documented throughout the study period.

2.2 Data Handling. For subsequent data analysis the 1-min particle data were labeled according to the different "modes of indoor activity" (MIA) within the church. During our study period we can define four modes with different sources of particle emission: periods before service times without any church visitors and potential particle sources (background, labeled B hereafter); periods during church service (D) where candles are a particle emission source; during church service with incense burning ($D_{\rm INC}$) when candles and incense are particle sources; and, since the hours after service with incense burning are assumed a priori to be characterized by a specific concentration-level (time-dependent decay of particles), the data during these periods was categorized as a sole class (P). The data were labeled P until measured concentrations decreased to background levels.

For the total of 14 measurement days the number of data assigned to each of the classes was 86% for B, 4% for D, 2% for $D_{\rm INC}$, and 8% for P. The emission of particles due to the hot-air blowers was not directly addressed in this study; however, their effects on indoor particle concentrations will briefly be discussed in a later part of this paper.

2.3 Particle Decay Rates. The time period needed for indoor particles concentrations to reduce to background levels after emission periods can be expressed by a decay rate constant, k. A time-dependent decay rate can be calculated from the point in time when indoor aerosol concentrations are significantly elevated above normal levels. The model is described in detail elsewhere (e.g., 4, 7) but will be briefly summarized here. The time rate of change of indoor aerosol concentration can be calculated as

$$\frac{dC_{\rm i}}{dt} = -kC_{\rm i} + \alpha C_0 F_{\rm p} \tag{1}$$

where C_i (N cm⁻³) is the indoor aerosol concentration, k is the decay rate constant (h⁻¹), α is the air exchange rate (h⁻¹), C_0 is the outdoor aerosol concentration (N cm⁻³), and F_p is the dimensionless penetration factor.

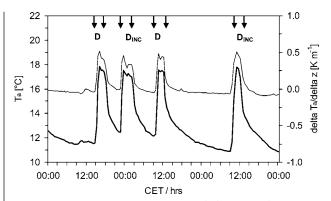


FIGURE 1. Time series of air temperature (T_a) at 2 m agl (thick solid line) and the vertical gradient of air temperature ($\Delta T_a/\Delta z$, thin solid line) calculated from air temperatures at 0.2 and 8 m agl for the time period December 24, 2004 to December 27, 2004. Start and end of church-service (D) during this time period are indicated by the black arrows.

The first term on the right-hand side, the particle decay rate, incorporates particles losses due to exfiltration (air exchange with outdoor air) and deposition:

$$k = \alpha + k_{\rm d} \tag{2}$$

where k_d is the particle deposition rate (h⁻¹).

The second term represents a source term due to infiltration of outdoor aerosol. If this term is assumed to be constant on short time scales (e.g., hours) and $kC_i \gg \alpha C_0 P$ then eq 1 reduces to

$$\frac{dCi}{dt} = -(\alpha + k_{\rm d})C_{\rm i} \tag{3}$$

This equation can be solved by integration

$$\ln\left(\frac{C_{\rm it}}{C_{\rm io}}\right) = -(\alpha + k_{\rm d})t\tag{4}$$

where $C_{\rm it}$ and $C_{\rm i0}$ are the indoor particle concentrations measured at time t and $t_{\rm 0}$. The left-hand side of eq. 4 can now be regressed against time from the moment when particle concentrations start to decay. The slope of the regression line equals the particle decay rate which incorporates decay due to exfiltration and deposition of particles.

3. Results

3.1 Indoor Meteorology. Indoor meteorology and thermal stratification are important factors when addressing air quality issues since they influence dispersion and accumulation processes during stable thermal stratification or particle dynamics (e.g., particle growth by condensation). The temperature regime inside the church is notably influenced by heating periods during service times (D, see Figure 1 and Figure 2a and b). During nonservice times the temperature distribution is quite homogeneous and barely influenced by insolation and outside temperatures. During heating periods which last approximately from 30 min before to 30 min after service times the air temperature increases from a mean temperature of 12.5 °C at 2 m agl ($\sigma_{T_a2m} = 2.4$ K) by about 3.7 K to a mean temperature of 16.2 °C during service (Figure 1). The vertical distribution of air temperature therefore changes significantly due to warm air input by heating indicated by a positive temperature lapse rate of about ΔT_a $\Delta z = 0.5 \text{ K m}^{-1}$ as evaluated from the air temperature measurements at 0.2 and 8 m agl (Figure 2a and b). The time periods during heating are therefore characterized by stable stratification of indoor air. However, with the heaters turned

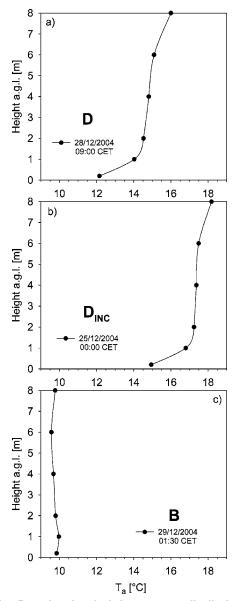


FIGURE 2. Examples of vertical air temperature distribution inside the church during different MIA (see text for details).

off after church service the temperatures start to decrease rapidly until isothermal stratification ($\Delta T_a/\Delta z \approx 0 \,\mathrm{K}\,\mathrm{m}^{-1}$) has developed through the entire air column just 2-2.5 h after the heating process ended. During nonservice times (B) the vertical temperature profile is generally distributed isothermally to slightly instable with increasing height above ground. However, the vertical temperature differences are very small with a negative lapse rate of only $\Delta T_a/\Delta z = -0.04 \text{ K m}^{-1}$ (Figure 2c). The mean relative humidity is 67% ($\sigma = 4\%$ relative humidity) during B and drops to levels of, on average, 55% ($\sigma = 6\%$ relative humidity) during D (not shown here). Considering the Kelvin effect, the indoor humidity levels are too low to affect particle dynamics by condensation and condensational growth (e.g., 16). As a result, particle dynamics in the size ranges as measured in this study are not influenced by indoor humidity fluctuations.

3.2 Particle Mass and Number Concentrations. The time series of particle mass concentrations for the entire study period (Figure 3) demonstrates a variable concentration regime inside the church which is clearly associated with the different MIA. Maximum concentrations occur during high masses at Christmas and New Year's Eve (burning of incense) which are elevated by factors of 12.6, 13.6, and 13.4 in

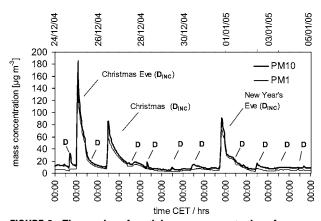


FIGURE 3. Time series of particle mass concentrations for mass fractions PM_{10} and PM_1 given for the study period from December 24, 2004 to January 5, 2005 (based on 30 min averages). PM2.5 data is not shown for reasons of clarity of the plot.

TABLE 1. Statistical Values for the Entire Study Period and Different MIA from December 24, 2004 to January 5, 2005 (Based on 1 min Averages)

		PM ₁₀ μg m ⁻³	PM _{2.5} μg m ⁻³	PM ₁ μg m ⁻³
entire period	average	17.5	15.5	12.4
	std. deviation	20.4	19.7	17.2
	median	10.8	9.2	6.0
В	maximum	219.5	210.5	166.3
	95 percentile	59.8	57.1	49.1
	average	10.8	9.3	7.2
	std. deviation	5.0	4.8	4.7
	median	9.2	7.8	5.6
	maximum	35.8	30.9	27.9
	95 percentile	14.2	11.8	9.2
D	average	19.4	15.7	12.9
	std. deviation	12.4	11.4	10.8
	median	14.4	11.3	7.2
	maximum	68.7	63.8	56.5
	95 percentile	51.1	49.6	44.5
D _{INC}	average	85.3	78.8	64.7
	std. deviation	55.6	53.4	43.6
	median	63.5	58.6	50.8
	maximum	219.5	210.5	166.3
	95 percentile	183.2	175.2	140.8

comparison to the average during the entire study period for PM₁₀, PM_{2.5}, and PM₁, respectively. The 95th percentile calculated for the entire study period is elevated by factors of 3.5–3.9 above the mean for all fractions. This demonstrates that periods of incense burning, incorporating only 2% of the study period, are characterized by significantly higher concentrations in comparison to the remaining measurement period. The maxima during D_{INC} are elevated by factors of 20.3, 22.6, and 23.1 above the average background B for PM₁₀, PM_{2.5}, and PM₁, respectively (Table 1). In terms of the churchgoers' exposure to particles we plotted the indoor/ outdoor ratio (I/O) in terms of PM₁₀ (Figure 4). Based on the PM₁₀ data measured indoors and outdoors (cf. Section 2.1) the three periods during D_{INC} are characterized by I/O ratios of 8.1, 4.8, and 4.2, respectively. For a time period of around 12 h after the start of D_{INC} the PM₁₀ concentrations are still elevated above outside values (see Section 3.3 for a discussion on particle decay rates). During the remaining study period the I/O is generally smaller than unity. The average I/O during B is 0.52 (\pm 0.27) rising to only D = 0.54 (\pm 0.27) during "conventional" liturgical service. The influence of candle burning on particle emissions inside the church is therefore negligible for PM₁₀. The I/O ratio of 1.2 on January 2, 2005 should not be considered here since is due to a very low outside value (indoor PM₁₀ shows normal background concentration of 14 μ g m⁻³).

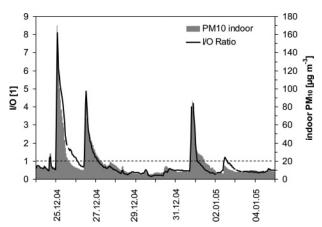


FIGURE 4. Indoor/outdoor ratio of PM_{10} concentrations measured in the church and at an air quality station at a distance of 2.3 km to the northwest of the church. The indoor concentration of PM_{10} is also given (both plots are based on hourly averages). The horizontal dashed line indicates $I/O\,=\,1.$

Overall, the indoor distribution of particle data classified into different MIA shows a very similar picture for the fractions $PM_{10},\ PM_{2.5},\ and\ PM_1$ (Figure 5a-c). The class D_{INC} is characterized by the highest concentrations while "conventional" service times (D) are only slightly elevated above the background concentrations (B) by factors of 1.4-1.6 (Figure 5d, Table 1). The PM_{10} concentration increases by a factor of 6.9 during D_{INC} in comparison to D, while the increases for $PM_{2.5}$ and PM_1 are even higher with factors of 7.5 and 9.1, respectively.

When comparing background concentrations B with D_{INC} a higher rise in fine mode particles (PM₁) in comparison to PM_{2.5} and PM₁₀ for the ratios D_{INC}/B and maximum D_{INC}/B can be observed (Figure 5d). On the other side, the ratio D/B slightly decreases to the finer modes. This behavior is due to the different sources of particle emission and the dynamics of number concentrations. During incense and candle burning a higher emission of particles in small size ranges can be observed in general. However, particle number concentrations are significantly larger during D_{INC} in comparison to D. For example, particle numbers during incense burning are on average 11,000 cm⁻³ for 0.3 μ m particles, while during service time without incense burning values of around 1,900 cm⁻³ in this size range were observed (not shown here). At diameters >1 μ m number concentrations were decreasing to $\, < 100 \, \, \text{cm}^{-3} \, \, \text{during D}_{\text{INC}}$ and $\, < 10 \, \, \text{cm}^{-3} \,$ during D, respectively. The ratio of D_{INC}/B is particularly elevated in the size range between $0.3 < D_p < 2 \mu m$, increasing by a factor of 10 to 15 on average compared to B (Figure 6). The absolute number concentration shows some variability due to its dependence on the actual mass of incense burned during the three D_{INC} services studied here. This is indicated by the difference in average and median ratio. However, the tendency for a significant increase in that size range is evident. The smaller size particles show only little enlargement during D with factors of around 1.7 but considerably increase to larger ratios at size ranges $D_p > 2 \mu m$. This increase at $D_p >$ $2 \mu m$ is observable during both D and D_{INC} and might be attributed to resuspension of deposited particles by the motion of churchgoers.

The effects of the different number dynamics are therefore associated with different emission characteristics (incense, candles, resuspension) where incense is responsible for a high number of particles primarily at particle sizes between 0.3 < $D_{\rm p}$ < 1 μm .

3.3 Temporal Dynamics and Decay Rate of Particles. In this section results on the temporal dynamics of indoor

particles are presented. The time series in Figure 3 already indicated that with the beginning of incense burning particle concentrations started to increase immediately. In Section 2.1 it was remarked that the second OPC installed at a height of 6.2 m agl failed in calculating correct absolute mass concentrations due to a imprecise internal software algorithm. Nevertheless the concentration time series can be used to evaluate the temporal particle dynamics between the two different measurement levels. Correlation coefficients of 0.98 and 0.99 ($r^2 \approx 0.98$), respectively, for the three particle fractions indicated that vertical mixing is not restricted by the air temperature stratification. Particles are well mixed within the whole air column inside the church, supported by warm air updrafts as long as the heaters are turned on (cf. Section 2.2).

After the end of D_{INC}, however, particle concentrations slowly start to decrease but are elevated above background concentrations for a time span of ~24 h (cf. Figure 4). With the decay rate which does not account for the specific process of particle removal as was described in Section 2.3 the temporal evolution of particle decay can be calculated (Figure 7). The decay for the particle bulk is $k = 0.9 \text{ h}^{-1}$ on average although a dependency on particle size can be observed which is consistent with findings by others (e.g., 4). The decay rate is larger for particles < 0.5 μ m and increases again at diameters $>1 \mu m$ where gravitational settling comes into account. While the decay rate varies between 0.85 and 0.90 h^{-1} in dependence on size range in this study, it is comparable to other estimates in the size range $< 1 \mu m$. However, a distinct increase of k to larger particle sizes, as reported by others (e.g., 7), was not observed in the present data.

4 Discussion of Results

Significant concentrations of indoor particles in the fine and coarse mode were measured inside a church. In comparison to the work of de Kok et al. (15) who found a 3-fold increase of PM $_{10}$ during candle and incense burning, we found a 1.6-fold increase of PM $_{10}$ by candle emissions (ratio D/B, cf. Figure 5d) but significant increases of 6.9 on average and 20.3 in maximum (ratio $D_{\rm INC}/B$) during simultaneous candle and incense burning. In absolute numbers PM $_{10}$ mass concentrations increased by $5.4\,\mu{\rm g}$ m $^{-3}$ during church service (when referring to the mean background concentration of 10.8 $\mu{\rm g}$ m $^{-3}$ PM $_{10}$ during B, cf. Figure 5a) and by $69\,\mu{\rm g}$ m $^{-3}$ on average during incense burning $D_{\rm INC}$ and up to 214 $\mu{\rm g}$ m $^{-3}$ during peak concentrations.

Although particle emissions by incense burning are variable in dependence on the mass of incense burned, it is the dominant source of particle emissions in a church with a significant increase especially in the fine size ranges $^{<}2\,\mu\mathrm{m}$ (cf. Figures 5d and 6).

Significant particle numbers by candle emissions are reported for size ranges <100 nm which might be important in terms of effects on human health (23). With present OPC technique it is not possible to measure ultrafine particles, therefore this would be a relevant question for future research. However, the absolute emission rate peaking at 50 nm as reported in ref 23 was rather small so that mass concentrations in the present church would have not been altered significantly by measurements in the ultrafine size range.

In Section 2.2 it was stated that potential emissions of hot-air blowers were not addressed by the measurement design, however, the time period before church service (heaters were turned on 30 min before church service) was not characterized by increases in particle mass or number concentrations and therefore they are negligible in this study.

When referring to potential impacts on human health the periods during important high mass times (e.g., Christmas, New Years's Eve) when incense is burned are of interest. The

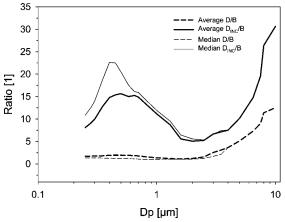


FIGURE 6. Ratios between the different modes of indoor activities vs particle diameter D_p as measured by the OPC at 1.7 m agl during the study from December 24, 2004 to January 5, 2005.

high masses can last for about 2.5 h and are characterized by indoor particle concentrations that are significantly elevated above outdoor PM₁₀ levels (I/O ratio between 4.2 and 8.1) and by high particle number increases in the size range $< 2 \mu m$. The decay of particles at a constant rate of 0.9 h-1 is comparable to findings of other measurements although a larger increase in decay rate at particle sizes >1 μm were observed mostly, although scatter of reference data is large at this size range due to different approaches (modeling, measurements) and varying room sizes and particle composition (4, 7). Here a significantly larger mixing volume (church interior ~15,300 m³) has to be considered in comparison to normal living room sizes in the other studies. The slow decay of particles is believed to be triggered mainly by indoor meteorology and the large mixing volume. Particles are well mixed by buoyancy effects during service times and

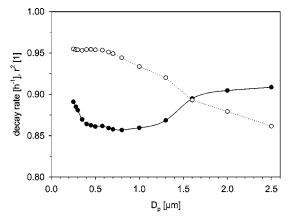


FIGURE 7. Particle decay rates (solid line) as evaluated by the method described in Section 2.3 (see text for details). The dotted line indicates the coefficient of determination (r^2) for the regression fit in each particle size class (cf. eq 4).

after the heaters are turned off a neutral to slightly instable thermal stratification develops which allows for further mixing of indoor air.

Candle burning shows only little effect on particle concentrations, while the burning of incense was identified to be the significant source of particle emissions in a church. It can be responsible for an absolute PM₁₀ increase of 68 μ g m⁻³ on average and 214 μ g m⁻³ during peak concentration situations. The distinct elevation of particle number concentrations by a factor of around 20 in the size range <1 μ m might be especially critical from the perspective of human health since recent epidemiological and toxicological research stresses the relevance of particle numbers in the fine and ultrafine range (*10*, *13*). The findings so far also stimulate future research on the influences of candle and incense burning on particle emissions within the ultrafine size ranges.

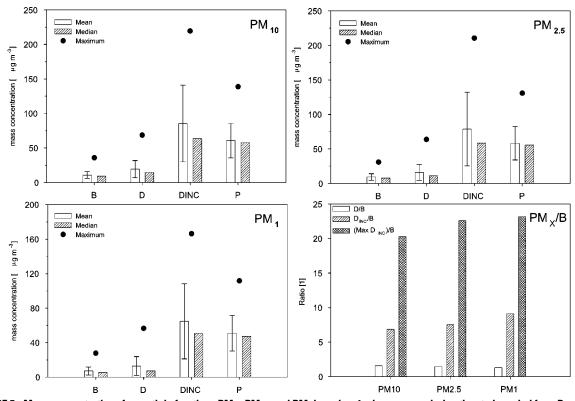


FIGURE 5. Mass concentrations for particle fractions PM_{10} , $PM_{2.5}$, and PM_1 based on 1 min averages during the study period from December 24, 2004 to January 5, 2005 as grouped according to the scheme described in Section 2.2. (Data significant on the p=0.01 level). Also the ratios of specific classes normalized by the background concentration (PM_X/B) are plotted for the particle fractions PM_{10} , $PM_{2.5}$, PM_1 .

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