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Potential effects of particulate matter from combustion during services on human health and on works of art in medieval churches in Cyprus

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Particulate matter in medieval churches of Cyprus.

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Indoor and outdoor particulate matter ($PM_{0.3-10}$) number concentrations were established in two medieval churches in Cyprus. In both churches incense was burnt occasionally during Mass. The highest indoor $PM_{0.5-1}$ concentrations compared with outdoors (10.7 times higher) were observed in the church that burning of candles indoors was allowed. Peak indoor black carbon concentration was 6.8 μg m^{-3} in the instances that incense was burning and 13.4 μg m^{-3} in the instances that the candles were burning (outdoor levels ranged between 0.6 and 1.3 μg m^{-3}). From the water soluble inorganic components determined in PM_{10}, calcium prevailed in all samples indoors or outdoors, whilst high potassium concentration indoors were a clear marker of combustion. Indoor sources of PM were clearly identified and their emission strengths were estimated via modeling of the results. Indoor estimated $PM_{0.3-10}$ mass concentrations exceeded air quality standards for human health protection and for the preservation of works of art.

1. Introduction

Medieval churches that host valuable works of art (frescoes, icons) function as museums but also as places of worship. The medieval churches of Cyprus are visited by 100 persons daily on an annual average basis. The burning of incense in a thurible during Mass services is common practice. Also, in many of them, burning of candles is allowed (see Pictures S1–S7 in supplementary data). An obvious particulate matter (PM) source in churches is the aforementioned combustion processes as described by Huynh et al. (1991). Also, Thatcher and Layton (1995) and Abt et al. (2000) demonstrated that PM can originate from visitor debris and movement, cleaning activities and infiltration from outdoors.

The effects of PM on human health, as demonstrated by Pope et al. (2004), or on the degree of degradation of the aesthetic value of the works of art, depends on particle size, number concentrations and chemical composition as Nazaroff et al. (1990) have shown. The same authors demonstrate that in the case of works of art, their effect depends also on the near surface airflow conditions. Combustion particles are also a major health concern for priests and elderly people, who stay for many hours in churches as Kennedy (2007) and Ho et al. (2005) reported.

The burning of incense and candles produces particulate matter and soot, among other air pollutants. Quantitative and qualitative data on their emissions have been obtained in test chamber studies (Pagels et al., 2009; Afshari et al., 2005; Lee and Wang, 2004; Jetter et al., 2002; Fine et al., 1999; Cheng et al., 1995). Particulate emissions from incense and candles vary, depending on their specific type and chemical composition, as well as on the burning conditions. Fine et al. (1999) have examined fine PM emissions from the burning of yellow paraffin wax candles from a Greek church in a test chamber, as well as beeswax candles from Chile. They found that burning candles emit fine particles, mainly of carbonaceous composition, at varying rates, depending on combustion conditions. The authors mentioned that under real world conditions the size distribution of emitted particles may differ to those found in the test chamber, due to the presence of ambient aerosol and different airflow patterns around the candle. Afshari et al. (2005) reported that the burning of pure wax candles emitted the highest concentration of ultra-fine particles in the few seconds after they were lit. The concentrations of particles larger than 0.3 μm were very low, but showed a sharp increase after 85 min, at the point when the candles were extinguished.

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In most of the publications that present PM in churches, the PM measurements were integrated measurements (24-h or more), conducted with filter packs as reported by Salmon et al. (2004). A recent study on detailed PM time-series conducted by Weber (2006) particle number and mass concentrations as well as particle dynamics in a Gothic Roman Catholic church were estimated, during the Christmas and New Year's Eve Mass, thus under real operating conditions. They reported that indoor PM_{10} mass concentrations, exhibited an 8.1-fold increase due to the burning of incense, in comparison to outdoor data taken from an adjacent atmospheric pollution station, whilst candle burning exhibited a little effect on indoor PM_{10} mass concentrations.

The Orthodox medieval churches are buildings with different design, structure and construction material characteristics than the Catholic churches. The candles used in the churches of Cyprus are usually made from beeswax. The only work concerning particulate matter measurements indoors and outdoors in Orthodox medieval churches was conducted by Salmon et al. (2004) in the World Heritage church of Asinou, in Cyprus. They found that particle deposition onto interior vertical surfaces could degrade the optical qualities of the frescoes in few years. In Asinou, the PM measurements were carried out using filter packs, with long sampling periods (24 h) that could not have captured the variability in indoor concentrations, particularly as they could not have recorded particle-generating activities occurring in time increments on the order of minutes to several hours.

The present study is set out to examine the variability of number concentration of indoor particles for 8 size bins ($PM_{0.3-0.5}$, $PM_{0.5-1}$, PM_{1-2}, PM_{2-3}, PM_{3-5}, PM_{5-10}, PM_{10-25} and $PM_{>25}$) and black carbon (BC), on a 15 min time resolution as well as the variability in particle chemical composition on a 12 h time resolution. The aim is to relate quantitatively all the above variability with indoor activities. In this way, indoor particulate matter and black carbon source strengths and emission rates in the church environment due to the burning of incense, candles, visitor presence and cleaning activities were estimated via their input in a numerical mass balance model. Based on the firm understanding of the above quantitative results, obtained in a real environment, one could then formulate mitigation strategies to protect human health and works of art from atmospheric particles in churches of similar use and architectural characteristics.

2. Methods

The churches were monitored for a minimum of 6 consecutive days in March 2004. A detailed description of the two Cypriot Churches Under Study (hereafter are referred to as *CCUS*), their floor plan and the location of on line sampling instruments has been presented elsewhere by Loupa et al. (2006) (see also Figs. S8–S9 in supplementary data). Shortly, both are stone-built and have a large number of single pane windows. The church of St. Paraskevi is located in the small village of Yeroskipou of Paphos. The cathedral of St. John is situated in the centre of the city of Nicosia. The data from St. Paraskevi, in Yeroskipou, Paphos are hereafter referred to as *PaMa* (Paphos March). The data from St. John's, in Nicosia hereafter are referred to as *NiMa* (Nicosia March). The air exchange rates (AER) of the CCUS were calculated by utilizing CO_2 data as reported by Loupa et al. (2006).

A Particle Measuring System LASAIR Model 5295 (PMS, Particle Measuring Systems, UK, Ltd) continuously measured particle number concentration and size distribution indoors and outdoors. Particle counting is divided in this instrument in eight channels, which correspond to the following size ranges, according to optical diameters: 0.3–0.5 μm, 0.5–1.0 μm, 1.0–2.0 μm, 2.0–3.0 μm, 3.0–5.0 μm, 5.0–10.0 μm, 10.0–25.0 μm, and >25.0 μm. Indoor and outdoor air was, alternately, drawn through a laboratory made stainless steel manifold that was connected to the instrument, similar to the manifold reported by Abt et al. (2000). The electrically actuated ball valves allowed air to be alternately sampled for 15 min from the indoor and the outdoor environments, hence allowing particle concentration to be recorded 15 times (one recording per minute) for each sampling interval. PM data were subsequently corrected for particle losses in the sampling manifold based on results from laboratory tests, according to the regression equation (Standard Error in parentheses): % particle loss = 2.82(0.03) × (particle diameter in μm) + 2.87(0.44).

Data during some important Mass services are missing, because the operation of the noisy instruments was not allowed.

Conversion of number to mass concentration was based on data obtained from the LASAIR instrument. This was accomplished by assuming spherical particle shapes, calculating the geometrical mean diameter of each size range, assuming a particle density of 1.4 g cm^{-3} for $PM_{0.3-1}$ and 2.4 g cm^{-3} for PM_{1-10}, and multiplying the mass of a single particle with a diameter equal to the geometric mean of a size range by the number of particles in that size range. The above density values were obtained from several values of densities reported in the literature (Nazaroff et al., 1990; Cheng et al., 1995; Ferro et al., 2004; Pagels et al., 2009).

A commercial instrument (PSAP; Particle Soot Absorption Photometer, Radiance Research, wavelength at 565 nm; Radiance Research; Seattle, USA) was used to measure in quasi-real time the mass concentration of black carbon (BC) in aerosol. The two mass-normalized absorption cross sections used in this study are 10 m^2 g^{-1}, as recommended by the manufacturers, and 7.5 m^2 g^{-1} in the case of freshly-generated particles, during combustion as demonstrated by Bond and Bergstrom (2006). A number of BC concentrations were also obtained outdoors.

Indoor and outdoor air was sampled with filter packs, for ca. 12-h periods to obtain PM_{10} chemical composition. Indoor and outdoor samplers were placed at a height of ca. 1.5 m from the ground. The sampling equipment consisted of a pair of laboratory made 90 mm diameter Dichotomous Stack Filter Units (DSFUs) that contained two filters in series. The first Teflon filter was analyzed for major anions (NO_3^-, PO_4^{-3}, SO_4^{-2}, Cl^-, $HCOO^-$, CH_3COO^-) and cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) of the water soluble fraction, by Ion Chromatography (DX500, DIONEX Inc., USA). A detailed description of the analytical methodology can be found in Karageorgos and Rapsomanikis (2007). The second nylon filter was analyzed for gaseous acids but the results were already presented in Loupa et al. (2007).

3. Results

3.1. Particulate matter diurnal profiles

In Fig. 1 the diurnal profile of PM number concentrations for two representative days in CCUS is presented. The corresponding time series of indoor BC and CO_2, NO_{in} and NO_{out}, O_{3in} and O_{3out}, data that were recorded simultaneously with PM are also presented Loupa et al. (2006).

In the case of NiMa (Fig. 1a, b), indoor PM number concentrations largely exceeded their corresponding outdoor concentrations in all size ranges. Between 0:00 and 7:00 h, the $PM_{0.3-2}$ concentrations were higher than outdoors, decaying slowly from their previous elevated concentrations from the evening Mass held on the previous afternoon. The opening of the church further decreased the fine particles by dilution with the fresh incoming air. The concentrations of larger particles had decayed much faster during the night and when the church opened, they started to increase due to the human movement. Between 11:00 and 11:40 h PM concentrations were increased in all size ranges (see also Fig. S10 and Table S1) because a funeral service (FS) took place, with the 30 people present holding lit candles. Incense was also burnt for a few minutes during this funeral service. Following the FS, the visitors that entered and lit candles induced a new increase to all sizes of particles, but in this case the higher increase was observed in the coarse particles. In the case of visitors which lit candles, the candles were lit near the main entrance and not at the centre of the church as it happened during the funeral service. Afterwards, candles were extinguished and the church was closed to the public. At that point, indoor $PM_{0.3-10}$ concentrations exhibited their maximum values, whilst the BC, CO_2, NO concentrations started to decrease. A similar behavior has also been observed by Afshari et al. (2005) and by Fan and Zhang (2001). Two hours later the church was opened again and a variable number of visitors (groups of 10–50 persons) entered and left the church within 10 min periods. The church was closed again for half an hour. When visitors were present without lighting candles, indoor fine and coarse PM concentrations appeared to decrease compared with the preceding times, but still remained higher than the corresponding outdoor concentrations. Simultaneously, indoor O_3 peaked, thus pointing out the absence of indoor combustion and enhanced ventilation rate because CO_2 did not increase. At 17:30 an evening Mass was started with few people present, the particle concentration counter had to be switched off, but an increase in