Asinou Church, Cyprus World Heritage Site Aerosol Pollutant Study

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ABSTRACT: The World Heritage church of Asinou, in Cyprus, is filled with frescoes dating from the 12th century AD. During April 1997 - January 1998, an air quality monitoring program measured indoor and outdoor particulate matter samples each season. Particle deposition samples also were collected. Average coarse particle concentrations outside the church were 28.1 μg m^{-3} while outdoor fine particle concentrations averaged 16.4 μg m^{-3}. Indoor average concentrations were 17.3 μg m^{-3} and 16.9 μg m^{-3} for coarse and fine particles, respectively. Particle deposition onto interior vertical surfaces over a nine-month period caused color changes that would be perceptible by a human observer (maximum color change ΔE=1.87). At these rates, the optical qualities of the frescoes will be clearly degraded over a period of five years. The installation of a mechanical air filtration system could reduce particle concentrations inside the church to less than 3% of uncontrolled levels.

1 INTRODUCTION

The famous Byzantine Church of Panagia Phorbiotissa of Asinou is part of UNESCO's World Cultural Heritage List. Located three miles to the south of the village of Nikitari in the Troodos Mountain range, it is home to perhaps the finest examples of Byzantine mural painting on the island of Cyprus. Asinou comes from the Greek form Asine, which is an ancient city founded by immigrants from the Greek city Argolis in the 11th century BC. Once belonging to the monastery of Phorbia, the church dates from the early 12th Century AD and the murals inside range from the 12th Century through the 17th Century.

The church is a rectangular building [See Figure 1] with vaults and high arched recesses on the sides. The church is covered with a steep-pitched roof and flat tiles. Fortunately, two-thirds of the original church decoration from the 1100's survive today. Through these murals we are able to determine that the church was probably originally constructed as a family chapel for Nicephoros Magistros, who later died here in 1115 AD (Stylianou & Stylianou, 1997).

A parking lot is located on the south side of the building. The church is a popular tourist destination and receives perhaps as many as four buses with tourists daily during the summer season. In addition, the church is sometimes used for weddings and baptisms, which further increases the number of visitors and the number of buses and cars that are parked in front of the church.

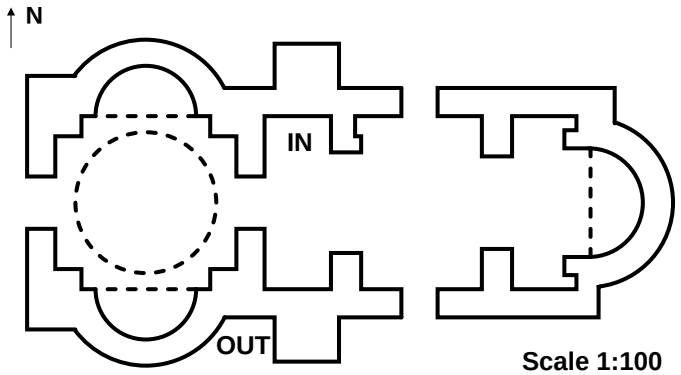

Figure 1. Church of Asinou floorplan with sampler locations.

Church art and museum collections and artifacts in general are threatened by air pollution, and particles soil artifacts in interiors as well as building facades. In addition, depending on the chemistry, the airborne grime can significantly hasten the deterioration of many materials. Measurements of outdoor airborne pollutant concentrations in the Cyprus atmosphere are scarce, and corresponding indoor levels are essentially unknown. These airborne pollutants can be important agents of decay of materials typically displayed indoors (e.g. textiles, pigments, paper) (Grossi & Brimblecombe, 2002; Christoforou et al. 1994; Nazaroff & Cass, 1991). Information about airborne pollutants will serve as a basis for determining appropriate protection methods for sensi-

tive cultural properties displayed indoors. These protection methods might include: (re)design of ventilation systems, increased cleaning schedules, display cases for sensitive materials, or recommendations for controlled storage conditions for especially sensitive materials.

Of special interest is the damage caused to frescoes and icons by black soot. Elemental or "black" carbon is generally associated with particles less than a micrometer in diameter. Once lodged in the pores in surfaces, these very fine black particles cannot be removed easily. This problem is caused in part by the intrusion of outdoor pollutants, such as diesel car exhaust, into the church interior, and possibly by combustion sources inside the church, such as candle burning. One of the goals of this study was to estimate the relative importance of the various sources that contribute to this problem. Larger particles, generally a few micrometers in diameter or larger often consist of soil dust, which is usually brown in color. Much other airborne particulate matter consists of light-colored ionic material, or organic matter, which is often yellow in color. It is possible to measure the mass flux of particles to surfaces, and to measure the darkening of surfaces by optical methods.

This study documents over four seasons the concentrations and chemical makeup of airborne and deposited particles inside the Byzantine church in Asinou, where all frescoes and icons are openly displayed. The study relates outdoor ambient concentrations to indoor particle deposition, both by comparing parallel concentration measurements indoors and out and by measuring particle deposition fluxes directly. These data can be used to support the design of systems that will control indoor air pollutant levels in order to protect this and other similar important collections.

2 AIRBORNE PARTICLE CONCENTRATIONS

2.1 *Experimental Methods*

Filter-based air pollutant samplers were used to measure airborne particle concentration and chemical composition both indoors and outdoors at the Asinou Church. The indoor sampler was located in the church as shown in Figure 1. The outdoor sampler was placed on the south side of the building. The sampling system used during this experiment has been described previously in the literature (Salmon et al. 1994; Ligocki et al. 1993) and is only briefly summarized here.

The sampling system is illustrated schematically in Figure 2. The ambient samplers measure airborne particle concentrations and chemical composition in two size ranges: (1) fine particles ($d_p < 2.1$ μm), and (2) total particles (no size discrimination). Coarse particle concentrations (here defined as those particles with $d_p > 2.1$ μm) are calculated by subtracting the fine particle concentrations from the total particle concentrations. Discrimination between coarse and fine particles is important because particle size has a considerable effect on particle deposition rates. Coarse particles settle onto horizontal surfaces under the influence of gravity while fine particles are capable of depositing onto vertical surfaces by diffusion and thermophoresis.

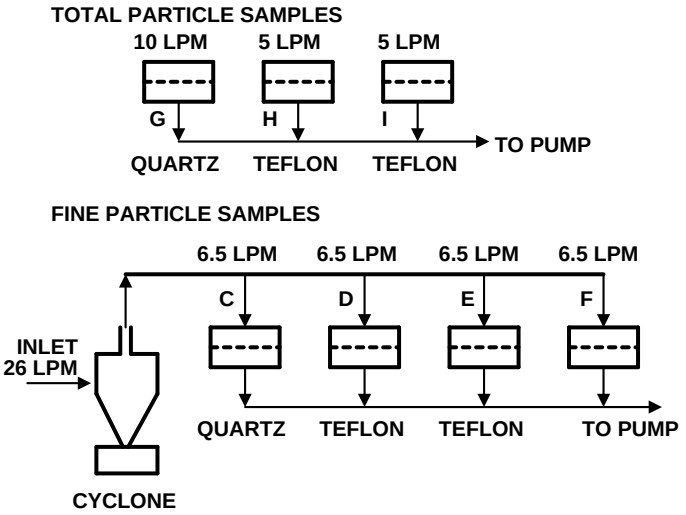

Figure 2. Schematic diagram of the particle sampler.

In the fine particle sampling system shown on the lower part of Figure 2, ambient air passes under a stainless steel rain cap and is pulled at a nominal flow rate of 26 lpm through an acid-washed Pyrex glass inlet line (1.27 cm ID, 100 cm long) that is protected from the sun. The air then passes through a Teflon-coated AIHL-design cyclone separator which, when operated at flow rate of 26 lpm, removes coarse particles with diameters larger than 2.1 μm (John & Reischl, 1980). The air stream containing only fine particles then enters a sampling manifold where it is split into several portions and ducted using Teflon tubing to separate filter holder assemblies. The flow rate through each filter holder is controlled by a critical orifice. Two of these sampling lines (D and E; Figure 2) collect fine particles on Teflon filters while sampling lines C and F collect fine particles on quartz fiber filters for subsequent chemical analyses.

The filter substrates used to collect particulate matter are chosen to be compatible with particular chemical analyses. Pretreated quartz fiber filters (47 mm diameter, Pallflex, 2500 QAO, baked at 550^o C for at least 8 hours in order to reduce their carbon blank levels) are used to collect samples destined for determination of atmospheric carbonaceous aerosol concentration. Polytetrafluoroethylene (PTFE) filters (47 mm diameter, Gelman Teflo, 1 μm pore size) are employed to obtain atmospheric particulate matter samples for mass concentration determination and for subsequent measurement of a wide variety of major and trace elements and water soluble anions and

cations. The combination of measurements made on the quartz fiber and Teflon filters allows a nearly complete material balance on the chemical composition of the fine particles to be obtained.

A similar set of filters are depicted in the upper part of Figure 2 comprising the total suspended particulate (TSP) samples which measure particles with no size discrimination. Filters are held in open-faced filter holders in order to collect all airborne particles. The filters are analyzed in the same manner as the corresponding fine particle filters to yield information for the total particles.

Airborne particle samples were collected over 24-hour periods once every six days for five consecutive weeks during four time periods encompassing a full year's seasonal cycle. Sampling periods took place beginning in April, 1997 and were repeated in July, 1997, October, 1997, and January, 1998. This frequency is more than adequate to obtain representative average values and to encounter representative peak 24-hour periods. Samples were collected from midnight to midnight to associate the samples with a particular day. Filters were installed the day prior to, and removed the day after sample collection.

Flow rates were measured before and after sampling with a rotameter which has been factory calibrated with an accuracy of 1 percent full scale. The flow rate checks are done to ensure that filter holders are not leaking and to determine that filter clogging did not occur. As an additional precaution to help ensure the integrity of the samples after collection, the PTFE, and quartz filters were stored in self-sealing petri dishes, further sealed with Teflon tape, refrigerated until returned from the field on the day of sample retrieval, and then frozen until sample analysis.

2.2 *Results*

The mass concentrations of fine, coarse and total airborne particles inside and outdoors at the Asinou Church are shown in time series in Figures 3 and 4. The highest measured concentrations occurred on 23 April, 1997 when 24-hour average outdoor total particle concentrations reached 93.5 μg m^{-3} and indoor total particle concentrations were 57 μg m^{-3}. The highest outdoor fine and coarse particle concentrations were also reached on 23 April with values of 28.4 and 65.0 μg m^{-3}, respectively.

Indoor fine particle concentrations reached a peak of 29.3 μg m^{-3} on 1 October, 1997 just slightly higher than the 29.2 μg m^{-3} measured on 23 April. Indoor coarse particle concentrations peaked at 35.0 μg m^{-3} on 27 July, 1997. Indoor/outdoor fine particle ratios averaged 1.05 for the year with indoor concentrations typically very close to and sometimes higher than those outdoors. The indoor/outdoor ratio for total particles averaged 0.81 over the year.

Days with indoor particle concentrations higher than those outdoors suggest that a significant indoor source of airborne particles exists at Asinou Church. This may be explained in part by the visit of tourists at the church. At times, there were more than 25 people inside the small church, several times per day. Moreover, wax candles inside the church were sometimes used, and two oil candles were lit on a constant basis.

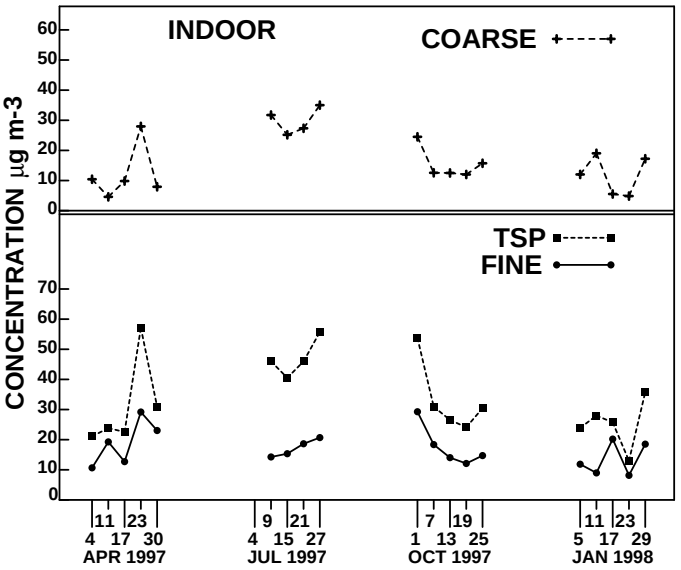

Figure 3. Mass concentrations of indoor airborne particles.

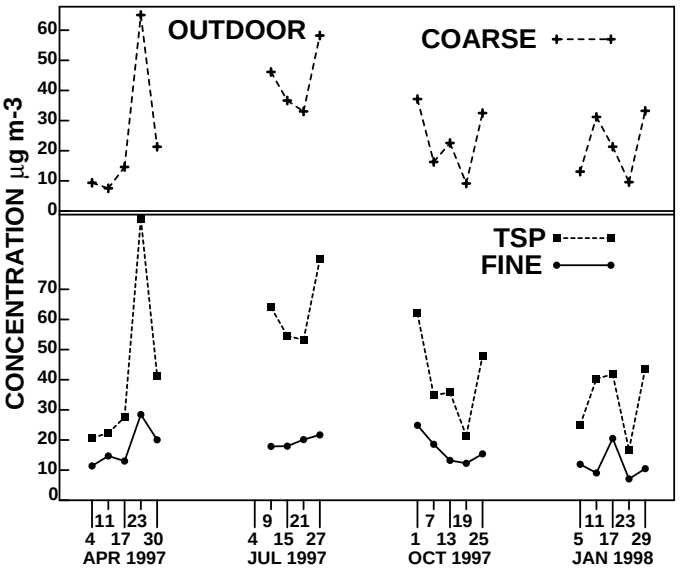

Figure 4. Mass concentrations of outdoor airborne particles.

Annual average concentrations were estimated by first averaging the data within each season and then averaging the four seasonal periods: April-1997, July-1997, October-1997, and January-1998. Annual average coarse particle concentrations were found to be 28.1 μg m^{-3} outside Asinou Church while outdoor fine particle concentrations averaged 16.4 μg m^{-3} for the year. Annual average indoor coarse and fine particle concentrations were similar, measuring 17.3 and 16.9 μg m^{-3}, respectively. Note that indoor annual average concentrations of fine particles are slightly higher than outdoor average.

By comparison, we found average coarse particle concentrations outside the World Cultural Heritage Wieliczka Salt Mine in Poland to be 30.4 μg m^{-3} while annual average outdoor fine particle concentrations there were 53.3 μg m^{-3} (Salmon et al. 1996). Coarse particle concentrations in the air outside museums in downtown Los Angeles, California, USA, average 78 μg m^{-3}, and fine particle concentrations in Los Angeles average about 25 to 44 μg m^{-3} (Salmon et al. 2004; Christoforou et al. 2000; Ligocki et al. 1993). Los Angeles is widely recognized as experiencing high airborne fine particle loadings; fine particle concentrations at Asinou are about a factor of two lower.

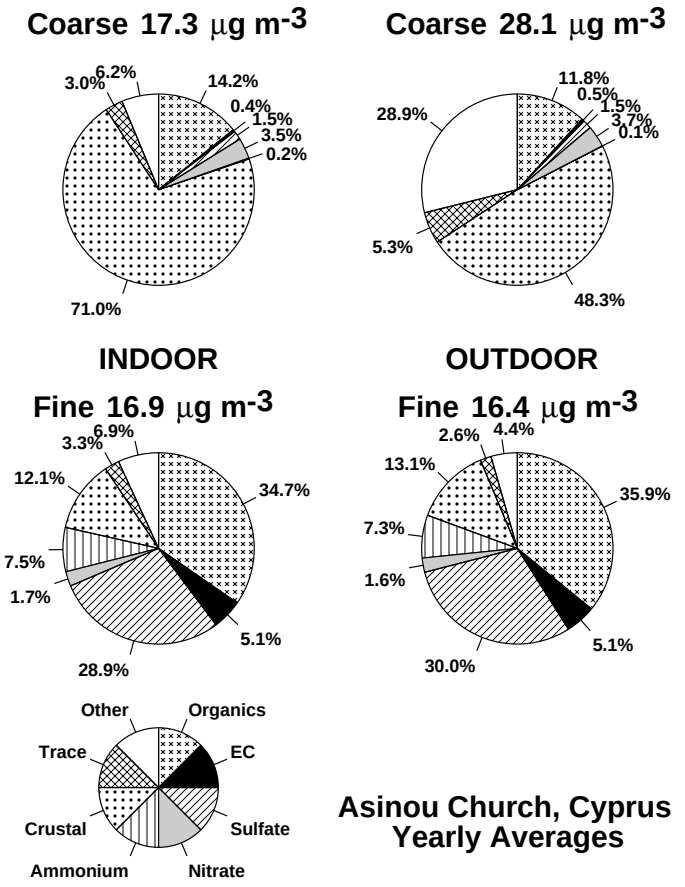

Figure 5. Yearly average aerosol chemical composition.

Figure 5 depicts the annual average chemical composition of the fine and coarse particles both indoors and outdoors. Organic carbon compounds are the most abundant contributor to the fine particle concentrations, with noticeable amounts of sulfate, and crustal material (mineral dust). Particulate carbon black (EC) is also high at approximately 5%. Among coarse particles the largest fraction is from mineral dust, with a sizable organic carbon contribution. Indoor particles show almost identical composition as the outdoor particles indicating that there is no protection for the frescoes and icons on the interior.

Coarse soil dust particles settle out of the atmosphere fairly rapidly by gravitational sedimentation. In the absence of air filtration, indoor airborne coarse mineral dust concentrations lower than those outdoors indicate particle deposition onto horizontal surfaces within the building studied. Fine particle mineral dust concentrations in excess of those outdoors suggest indoor sources, possibly due to visitors to the church, sweeping or other indoor activities.

The seasonal variation in the chemical composition of the airborne particles is illustrated in Figures 6 and 7 for fine and coarse particles, respectively. In Figure 6 it is seen that particle concentrations and composition outdoors are very similar to those indoors during all seasons. Both indoor and outdoor levels of elemental (black) carbon particles during April, 1997 are 3-5 times more than during the other months studied averaging nearly 2 μg m^{-3}, with maximum concentrations as high as 3.2 μg m^{-3} measured indoors.

By comparison, indoor elemental carbon concentrations inside museums in a very polluted region like Krakow Poland averaged between 2.9 and 3.3 μg m^{-3} (Salmon et al. 1995). To put this in perspective, outdoor elemental carbon particle concentrations averaged about 4.3 μg m^{-3} at downtown Los Angeles during 1986 (Solomon et al. 1989), indicating that there is nearly half as much soot inside the Asinou Church air as in the most heavily trafficked city in the United States. Fine black elemental carbon particles do not settle out of the atmosphere easily under the influence of gravity but instead can deposit on surfaces by convective diffusion.

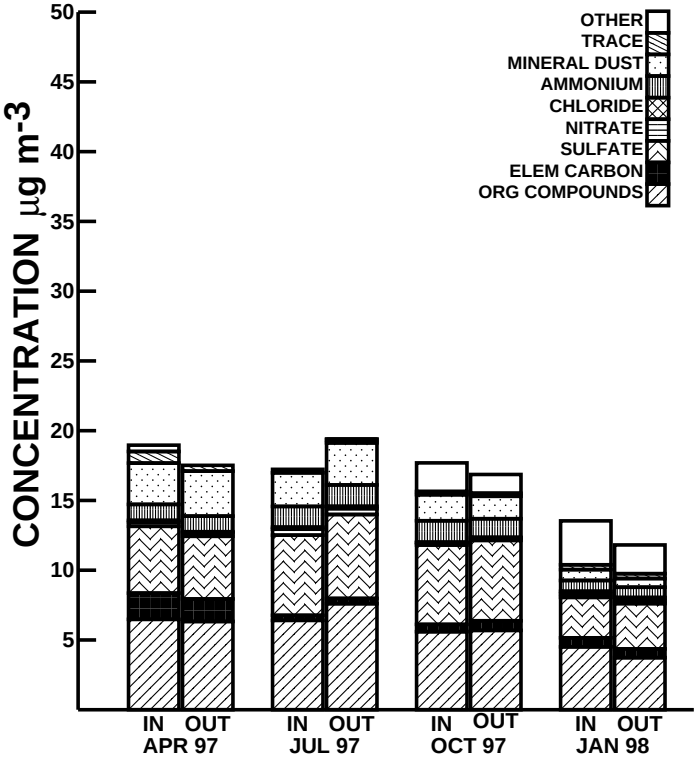

Figure 6. Seasonal chemical composition of fine particles.

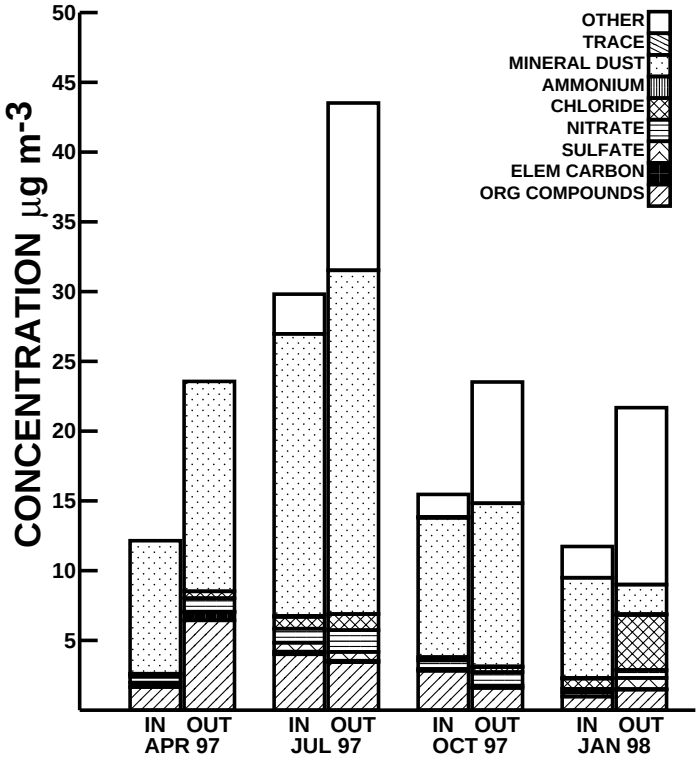

Figure 7. Seasonal chemical composition of coarse particles.

3 PARTICLE DEPOSITION ONTO SURFACES

3.1 *Experimental Methods*

Deposition plates were used to measure the flux of particles depositing to surfaces within the Asinou Church. Horizontally and vertically oriented deposition plates were deployed at the beginning of May, 1997 and left in place for 9 months. The vertically-oriented plates were mounted directly on the church walls at two indoor locations and one outdoor location. The outdoor plates were protected from the rain by an overhanging portion of the building. Horizontally-oriented deposition plates were placed indoors near the ambient particle sampler (no horizontal plates were used outdoors).

Quartz fiber filter material (Pallflex) mounted in aluminum holders with beveled edges was used as a dry deposition surface. These quartz fiber surfaces were analyzed to determine organic carbon and elemental carbon particle deposition rates.

A number of "white" deposition surfaces were used for optical analysis of the soiling of surfaces. These included the quartz filter materials mentioned above, and Arches hot-pressed water color paper surfaces. Color changes were used to determine the amount of soiling of each sample over the course of the 9-month exposure period. A Diano MatchScan II spectrophotometer was used to measure the diffuse reflectance spectra of the deposition surfaces. Reflectance spectra of all samples were taken prior to placement and the color of each sample was remeasured at the same location on each sample after 9 months in the field.

3.2 *Results*

Coarse particles by virtue of their large size have negligible diffusivity in air and hence do not deposit readily to vertical surfaces. Instead, particulate matter deposits on vertical surfaces are due to the flux of smaller diameter fine particles.

The deposits collected on vertical surfaces indoors over 9 months were not large. Therefore, the color change on all of the vertically oriented deposition substrates is very small, as seen in Figure 8. The initial color of the samples and the color after exposure were calculated using CIE's (Commission International de l'Eclairage) L*,a*,b* color scale (Billmeyer & Saltzman, 1981) for the CIE 2^o standard observer using illuminant C. The darkness of the sample is given by the L* coordinate, while its coloration is described by the a* (red-green) and b* (yellow-blue) coordinates. The overall color change of a sample, ΔE, is given by the formula in Equation 1.

$$\Delta E = \sqrt{(\Delta L*)^2 + (\Delta a*)^2 + (\Delta b*)^2} \quad (1)$$

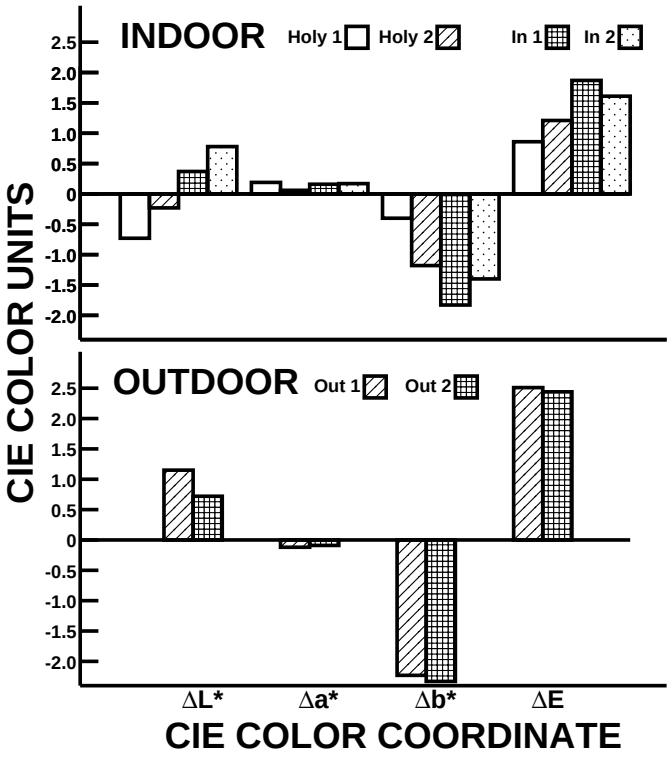

Figure 8. Color change observed on vertical substrates.

Figure 8 shows the color change observed on each of the vertically-oriented white deposition substrates over a 9 month period. Changes on indoor vertical surfaces over this time period are large enough to be considered perceptible by a human observer with a maximum $\Delta E = 1.87$ due predominantly to a shift in the yellow-blue coordinate at that location.

Typically, a ΔE value of less than 0.5 represents an imperceptible difference to a human observer, and a

ΔE value of 1 is generally still considered to be a good color match. Therefore, the minimum color change $\Delta E = 0.86$ on one of the samples placed in the Holy is at the threshold of what would be noticeable.

Outdoors the measured color change in 9 months was up to $\Delta E = 2.51$ again mostly due to a shift toward blue in yellow-blue coordinate. While the color change of indoor vertically oriented white surfaces is just over the threshold of human perception for a nine month period, exposure over a five year period to the same conditions would be expected to produce $\Delta E > 6$ which would be clearly noticeable.

Table 1 shows the dry deposition flux of carbon species to both vertically and horizontally-oriented deposition substrates. Dry deposition to horizontal surfaces generally is dominated by gravitational sedimentation of large (coarse) airborne particles. Carbonate carbon which is found in the coarse size range, was therefore only found on the horizontally-oriented deposition plate. Vertically-oriented indoor deposition plates experienced a higher carbon loading than those placed outdoors. However, this could be related to placement of the plates nearer to sources of pollutants (e.g. candles). The outdoor plates were placed such that they were protected from rain, which could have reduced their exposure to particles as well.

Table 1. Carbon accumulation on quartz deposition substrates over 9 months (μg cm^{-2}).

	Indoor	Outdoor
Vertical plates:		
Organic	14.09 ± 0.91	4.38 ± 0.43
Elemental	0.07 ± 0.20	0 ± 0.20
Horizontal Plates:		
Organic	104.15 ± 6.02	N/a
Elemental	1.81 ± 0.29	N/a
Carbonate	12.22 ± 1.20	N/a

Placed: 8 May, 1997
Collected: 22 Feb, 1998

4 PROTECTION FROM DAMAGE DUE TO SOILING BY AIRBORNE PARTICLES

From the field experimental study just described, it is seen that fine particle concentrations in the atmosphere inside Asinou Church are as high as those outdoors. These concentrations are high enough that frescoes and icons inside the church exposed directly to indoor air that cannot easily be cleaned eventually will become blackened.

Deposition of black particles onto vertical surfaces occurs more slowly than onto upward facing horizontal surfaces. The situation faced by vertically-oriented objects is possibly more dangerous precisely because the effects are more subtle and because the surfaces may be more difficult to clean.

Optical measurements of the test surfaces placed indoors during the present study show that undisturbed vertical white surfaces will be darkened to the extent of a ΔE value of 1-2 within one calendar year. Taking a ΔE value of about 2 as the point where a color change would begin to be noticed, it is reasonable to project that undisturbed white vertical surfaces indoors in Asinou church will become visibly soiled in less than 2 years time.

Perception that a colored surface has darkened would probably require a longer exposure (Bellan et al. 2000). These changes are just slow enough that they may not be noticed by personnel who view the interior day-after-day, but over a period of two decades, the accumulated change would be profound: darkening of an object by a ΔE value of 12 over a two decade period would be a very serious change in appearance. If the object involved is placed at risk during cleaning, then serious consideration should be given to preventative measures such that the frequency of cleaning can be decreased.

Control measures for reducing the accumulation of particles onto indoor surfaces include reducing the infiltration of untreated outdoor air, filtration of particles from the air within the building and encasement of objects within display cases or framing pictures behind glass. Reduction of the outdoor airborne particle concentrations, of course, provides a preferred solution, but such pollution abatement programs are generally not under the control of authorities at the local level.

The situation faced by Asinou Church is typified by conditions where an historical building lacks any mechanical ventilation system. Particulate matter control in this situation has been studied by Nazaroff and Cass (1991) for the case of an historical residence used as a museum in downtown Los Angeles, CA. Also, Christoforou et al. (1999) have studied control methods for the abatement of the problem of deposition of atmospheric particles onto interior surfaces at the Yungang Grottoes.

One of the problems inherent in retrofitting a cultural monument with a mechanical ventilation system is that it may not be possible to do so without significantly altering the appearance of the building. However, such an air conditioning system is oftentimes the only way to ameliorate the air pollution problems faced by churches and other ancient structures.

In the case of fine black carbonaceous particle control, the preferred solution is installation of a mechanical air filtration system if the system can be constructed in a way that is compatible with the historic structure. The reason for this is that outdoor fine particles will infiltrate most buildings that are not placed under positive pressure by a deliberate mechanical ventilation system.

A conventional mechanical air conditioning filter like the one illustrated in Figure 9 could be installed at the Asinou church. The system does not necessarily

have to be elaborate mechanically, as only particulate air filtration is considered here; central heating, cooling and humidity control is not necessarily required for particle filtration to be provided (although temperature and relative humidity control may be desirable for other reasons).

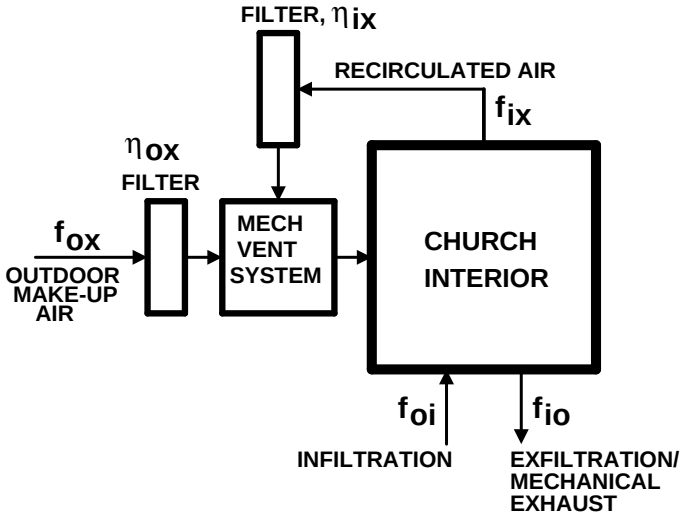

Figure 9. Schematic of mechanical filtration system.

Filters are placed in both the outdoor make-up air supply (η_{ox}) and in the return air supply to the air handlers (η_{ix}). Based on work done by Nazaroff and Cass (1991) at a Los Angeles, CA, USA, museum, some indicative particle control efficiencies may be obtained. Here it is assumed that widely available high efficiency filters are used (87% collection efficiency for fine particles; 95% collection efficiency for coarse particles) and that outdoor air exchange rates are kept fairly low: about 0.68 air changes per hour (0.5 hr^{-1} due to filtered make-up air and 0.18 hr^{-1} due to unfiltered infiltration air).

Under the conditions described above, both indoor fine black carbon particle and coarse soil dust concentrations would fall to approximately 3% of the uncontrolled case. Changes in particle deposition rates and soiling rates are similar (but not identical) to the changes in indoor airborne particle concentrations just discussed.

As an alternative to centralized air filtration systems, smaller units can be built to clean the air in single rooms that contain particularly sensitive objects. If at all possible, these smaller systems should be designed to filter both the make-up air flow (f_{ox}) and recirculated air flow (f_{ix}). Stand-alone units within a single room that filter only air from within the room are less effective than a system that also filters the make-up air supply and that can be used to pressurize the room so that leakage occurs to the outdoors.

Absent a mechanical air filtration system, the remaining alternatives for reduction of soiling by airborne particles involve careful use of physical barriers such as display cases and framing behind glass. Since fine black particles are capable of infiltrating through small cracks such as are found around doors and windows, a really effective display case must be fairly tightly sealed. Before sealing objects in a tight case, it is important to be sure that the object will not be damaged by the resulting temperature and humidity conditions within the case and that harmful gases (e.g. acetic acid) do not accumulate inside the case due to the wood used in the case or due to slow decomposition of the objects.

The church at Asinou has frescoes covering the entire surface of the interior walls. In essence, only the floor of the church requires no special protection. It is possible to encase all surfaces inside the church, in effect turning the interior into a giant walk-in display case. This approach was used in a California museum to prevent soiling of an entire room filled with historic furniture covered with sensitive textiles. A glass alcove was built just inside the entrance door to the room. The public can walk into this glass enclosure and look around the room, but the glass enclosure is sealed to the entrance door of the room, thereby turning the room into a giant walk-in display case.

5 CONCLUSIONS

During April 1997 - January 1998, an air quality monitoring program conducted at the Asinou church simultaneously measured indoor and outdoor ambient particulate matter. In addition, particle deposition samples were collected.

Fine particulate matter, which is more likely to deposit onto vertical surfaces and therefore cause soiling, was found to be nearly the same indoors and outdoors, averaging 16.9 μg m^{-3} and 16.4 μg m^{-3} respectively. Chemical characterization of the fine particulates has shown that there is basically no difference between the particles at the two locations. This means that there is essentially no protection to the frescoes inside the church from outdoor particles.

Particle deposition onto the interior vertical surfaces of the church was found to be causing color changes that are barely perceptible by a human observer (average color change $\Delta E \approx$ 1-2). Even though these rates are small, if they continue unabated, would cause serious change in the optical qualities of the frescoes.

The installation of mechanical air filtration systems could reduce particle concentrations inside the church to less than 3% of uncontrolled levels and thus significantly lengthen the time required for perceptible soiling to occur. Other methods of control, such as the encasing of the frescoes into tightly sealed display cases, could further protect the church.

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