

CHARACTERISTICS OF AIRBORNE PARTICLES INSIDE SOUTHERN CALIFORNIA MUSEUMS

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Abstract—The concentrations and chemical composition of suspended particulate matter were measured in both the fine and total size modes inside and outside five southern California museums over summer and winter periods. The seasonally averaged indoor/outdoor ratios for particulate matter mass concentrations ranged from 0.16 to 0.96 for fine particles and from 0.06 to 0.53 for coarse particles, with the lower values observed for buildings with sophisticated ventilation systems which include filters for particulate matter removal. Museums with deliberate particle filtration systems showed indoor fine particle concentrations generally averaging less than $10 \mu\text{g m}^{-3}$. One museum with no environmental control system showed indoor fine particle concentrations averaging nearly $60 \mu\text{g m}^{-3}$ in winter and coarse particle concentrations in the $30\text{--}40 \mu\text{g m}^{-3}$ range. Analyses of indoor vs outdoor concentrations of major chemical species indicated that indoor sources of organic matter may exist at all sites, but that none of the other measured species appear to have major indoor sources at the museums studied. Significant fractions of the dark-colored fine elemental (black) carbon and soil dust particles present in outdoor air are able to penetrate to the indoor atmosphere of the museums studied, and may constitute a soiling hazard to works of art displayed in museums.

Key word index: Museum, indoor air quality, particulate matter, soiling.

INTRODUCTION

The extent to which particulate matter in outdoor, polluted atmospheres infiltrates into buildings remains largely unknown despite several studies over the last 20 years (Yocom *et al.*, 1971; Alzona *et al.*, 1979; Spengler *et al.*, 1981). While indoor air quality is of concern from a health-effect perspective, it is also relevant in terms of material damage. There is growing concern among museum curators that works of art displayed in urban museums are subject to damage from exposure to indoor air pollution (Thomson, 1978; Baer and Banks, 1985; NRC, 1986). Potential damage could take the form of chemical degradation due to exposure to reactive gases, or soiling due to deposition of airborne particles, particularly those containing elemental or "black" carbon. Studies have shown that the sulfur dioxide, nitrogen oxides and ozone found in outdoor air can penetrate the ventilation systems of museums and rare book libraries, with consequent risk of damage to the collections (Thomson, 1965; Hughes and Myers, 1983; Shaver *et al.*, 1983; Hackney, 1984; Davies *et al.*, 1984; Druzik *et al.*, 1989). For that reason, many such facilities have

begun to integrate activated carbon or other chemical sorbents into their heating, ventilation and air conditioning (HVAC) systems.

In the absence of particle filtration systems, concentrations of suspended particles indoors can be as high or higher than those outdoors (Biscontin *et al.*, 1980; Dockery and Spengler, 1981). Filters for particle removal have been incorporated into most museum HVAC systems for many years. But surprisingly little attention has been paid to determining the effect that these particle removal systems have on the concentration, chemical composition and size distribution of airborne particles within museums. Those studies which have been conducted on aerosol composition and removal within museums generally have been designed to resolve specific problems encountered in a particular building (e.g. generation and deposition of cement dust inside a freshly poured concrete structure; Toishi and Kenjo, 1967, 1975).

The present study was initiated in order to determine the extent to which museum ventilation systems modify the concentration and composition of airborne particulate matter entering from outdoors. In addition, information is sought that will identify particulate chemical species for which there may be indoor sources. Additional facets of the present study, which are reported elsewhere, include measurements of the accumulation rates and deposition velocities of particles to vertical and horizontal surfaces inside museums (Ligocki *et al.*, 1990) and modeling studies of

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indoor particle concentrations and deposition to surfaces (Nazaroff and Cass, 1989a, b; Nazaroff *et al.*, 1990a, b). Taken as a whole, this information provides data and methods for assessing the effectiveness of building ventilation systems in protecting artifacts from damage due to the intrusion of outdoor particulate air pollution. The present paper defines baseline conditions for particulate air quality at the museum sites studied, discusses the composition of the indoor and outdoor aerosol at the various sites, determines indoor/outdoor concentration ratios of the individual chemical species, and presents evidence for indoor sources of particles inside museums.

EXPERIMENTAL METHODS

Sample collection

Five sites were chosen for indoor and outdoor airborne particle sampling: (1) the J. Paul Getty Museum in Malibu, CA; (2) the Norton Simon Museum in Pasadena, CA; (3) the Virginia Steele Scott Gallery located at the Huntington Library in San Marino, CA; (4) the Southwest Museum in Los Angeles, CA; (5) the Sepulveda House, a historic house located in El Pueblo de Los Angeles State Historic Park in downtown Los Angeles. The first three sites are modern museum structures equipped with sophisticated HVAC systems which include filters for particle removal. The Southwest Museum is a turn-of-the-century poured concrete structure which has central heating and zonal air conditioning in the hall in which sampling was conducted, but no supplementary particle filtration system. The Sepulveda House is a partially restored 19th-century brick building which has no mechanical ventilation system whatsoever. Building characteristics, ventilation system parameters and particle filtration devices at each museum are described in Table 1.

At all sites, with the exception of the Sepulveda House, the samplers were located in galleries where artworks or artifacts were displayed. At the Sepulveda House, the samplers were

located on a stairwell landing which was open to the central corridor of the first floor. The most valuable artifacts on display at the Sepulveda House during 1987–1988 were located in a small room which is isolated from the rest of the building by a glass partition, thereby effectively placing them inside a sealed display case.

Ambient 24-h samples of both fine ($d_p < 2.1 \mu\text{m}$) and total airborne particulate matter were collected indoors and outdoors at each site. A schematic of the samplers used is presented in Fig. 1. Air drawn through the fine particle sampler first passed through a cyclone separator (John and Reischl, 1980) for removal of coarse particles ($d_p > 2.1 \mu\text{m}$). The air flow was then divided among several parallel 47-mm diameter filter assemblies, each containing a filter substrate compatible with a particular chemical analysis procedure. In the total particle sampler, the same filter substrates were employed in open-face filter holders. A Teflon-coated metal sheet was positioned above the open-face filter holders in order to prevent the sedimentation of large particles onto the filters.

Two Teflon membrane filters (Teflo, Gelman, Ann Arbor, MI; filters A and B in Fig. 1) were used in each fine and total particle sampler. One Teflon filter of each pair (filter A) was used to collect particle samples for analysis of anions by ion chromatography (IC), ammonium ion by colorimetry, and water-soluble sodium and magnesium by atomic absorption spectroscopy (AA). The other Teflon filter (filter B) was used for gravimetric determination of aerosol mass concentration and for the measurement of the concentrations of 32 elements by X-ray fluorescence (XRF). Quartz fiber filters (Tissuquartz 2500 QAO, Pallflex, Putnam, CT; filter C in Fig. 1), which had been baked at 750°C overnight in order to reduce their carbon blank, were used to collect particles for analysis of organic and elemental carbon. The additional filter D in Fig. 1 contained a polycarbonate Nuclepore filter for the analysis of individual particle morphology and elemental composition by scanning electron microscopy (Ligocki *et al.*, 1990). The Teflon filters were stored in sterilized disposable plastic petri dishes with tight-fitting lids. The quartz fiber filters were stored inside the same type of petri dishes which had been lined with pre-baked aluminum foil liners. All samples were retrieved promptly after sample collection. The samples were placed in petri dishes and sealed with Teflon tape, re-

Table 1. Building characteristics of the study sites

Site	Use*	Ventilation system†	Outdoor air exchange rate (h^{-1})‡	Indoor air recirculation rate (h^{-1})§
Sepulveda House	1	I	1.6–3.6	0
Southwest Museum (California Room)	2	II	0.3	nd¶
Norton Simon Museum	3	III	0.4–0.7	5.4
Scott Gallery	3	III	0.3–1.0**	8.2
Getty Museum (Gallery 121)	3	III	1.2–1.3	7.1

* (1) Historical museum, (2) archeological museum, (3) art museum.

† (I) No mechanical ventilation system; (II) partial forced ventilation system, no additional particle filtration; (III) modern heating, ventilation and air conditioning (HVAC) system with particle filtration.

‡ Air changes per hour due to outdoor make-up air plus infiltration. Range of measurements obtained from perfluorocarbon tracer tests from the summer and winter sampling periods (Salmon *et al.*, 1990), and sulfur hexafluoride tracer-decay tests for a 24-h period in the spring of 1988 (Nazaroff *et al.*, 1990a).

§ Air changes per hour provided by recirculation of indoor air, from Nazaroff *et al.* (1990a) and Salmon *et al.* (1990).

|| Summer and winter seasons.

¶ Not determined.

** Summer and spring seasons.

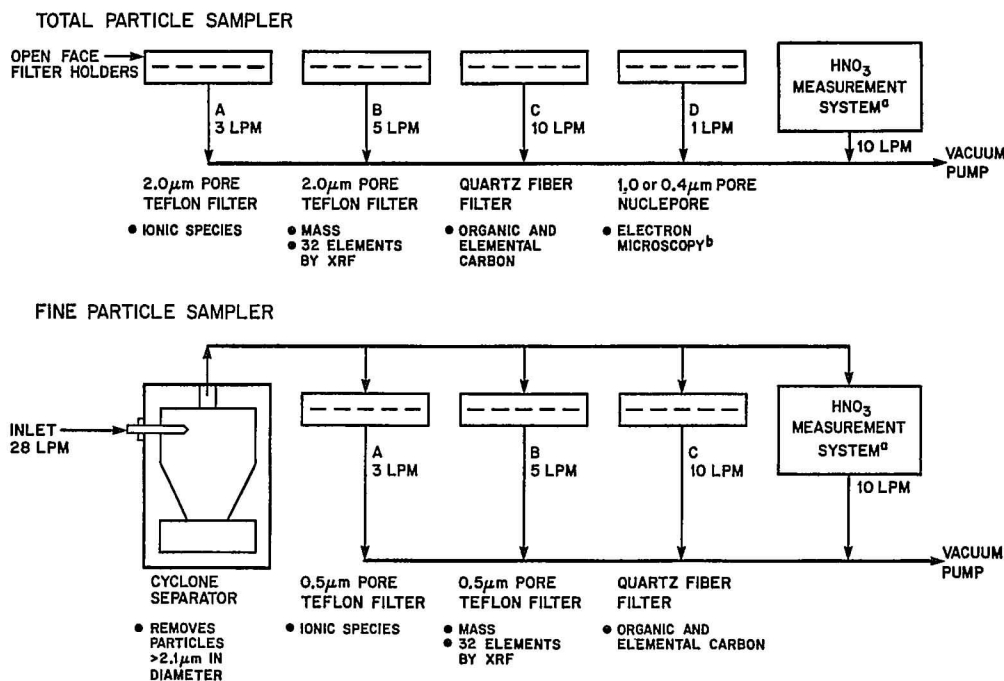


Fig. 1. Ambient total and fine particle samplers.

refrigerated immediately and then frozen in order to minimize sample contamination and losses of volatile material.

Ambient sampling was conducted during a summer period (2 July–31 August 1987) and a winter period (23 November 1987–30 January 1988). During each of these periods, ambient samples were collected every 6th day, resulting in a total of 11 sample days during the summer period, and 12 sample days during the winter period. Flow rates were checked with rotameters (Matheson, Secaucus, NJ) before and after each sampling day. The rotameters were calibrated in the laboratory and exhibited a precision of $\pm 3\%$. Field blank filters equivalent to 10% of the samples taken were acquired for each seasonal sampling period.

Analytical methods

The Teflon filters from sampler channel A were analysed for ionic material. Each filter was extracted in distilled deionized water by agitation in a sealed container on a mechanical shaker table for 3–4 h in a refrigerated room. Extracts were analysed for chloride, bromide, nitrate and sulfate ions by IC on a Dionex model 2020i ion chromatograph (Dionex Corp., Sunnyvale, CA); for ammonium ions by an indophenol colorimetric procedure (Bolleter *et al.*, 1961) on an Alpkem Model RFA-300 Rapid Flow Analyzer; and for sodium and magnesium ions by flame atomic absorption on a Varian Techtron Model AA-6 atomic absorption spectrophotometer. Replicate analyses were performed on approximately 10% of the filter extracts.

Instrument detection limits (IDL) for IC analysis, using a 50 μl sample loop, were 0.1, 0.2, 0.2 and 0.2 μg per filter for chloride, bromide, nitrate and sulfate, respectively, with an analytical precision of 1%. Bromide was not found in the samples in concentrations above the IDL. The IDL for ammonium was 0.4 μg per filter, with an analytical precision of 2%, while the IDLs for sodium and magnesium on the AA

were 0.1 and 0.2 μg per filter, respectively, with an analytical precision of 5%.

The Teflon filters from sampler channel B were equilibrated for 24–48 h at a temperature of 22 °C prior to weighing on a Mettler Model M-5S-A Balance (Mettler Instruments, Hightstown, NJ). The precision of the mass determination, based upon replicate weighings of samples and calibration filters, was $< 2 \mu\text{g}$ per filter. The variability in the instrument from day to day and from the initial to the final weighing periods (approx. 3 months) were 6 and 9 μg per filter, respectively. After mass determination, these filters were analysed for 32 elements* by XRF (Dzubay, 1977) at NEA Inc. The IDLs for the XRF technique varied from 0.01 to 0.9 μg per filter, depending upon the element, with an analytical precision of 5%. A subset of 10 of the open-face total particle filters were also analysed for aluminum by neutron activation analysis (NAA) in order to provide a calibration of the XRF particle size correction factor as it applied to the open-face total particle samples.

Analysis of elemental and organic carbon concentrations was performed at Sunset Laboratory by the thermal evolution and combustion technique of Johnson *et al.* (1981) as modified by Cary (1987). The IDLs for elemental and organic carbon were 0.9 μg per filter, with an analytical precision of 3.5%.

In all species mass determinations, mean field blank values were subtracted from the raw species concentrations. The variance of the field blank values, along with the measurement variances determined from replicate analyses and the uncertainties in the air volume sampled, were included in the error propagation procedure. The quantification limit for each species was taken to be twice the quantity obtained by propagating the IDL with the blank variance. Coarse particle mode concentrations were determined by subtracting the fine particle concentrations from the total particle concentrations. In a few instances, the measured fine particle concentration of a species was greater than the measured total particle concentration. In such a case, after ensuring that this was not due to a major sampling or analysis error, the fine and total concentrations were both set equal to the arithmetic

*Ag, Al, As, Ba, Br, Ca, Cd, Cl, Cr, Fe, Ga, Hg, In, K, Mn, Mo, Ni, P, Pb, Pd, Pb, S, Sb, Se, Si, Sn, Sr, Ti, V, Y, Zn and Zr.

tic mean of the two measured values, and the difference between the measured value and this mean value was added to the error propagation. The coarse mode concentration was taken as zero in these cases.

In the calculation of the indoor/outdoor ratios, in order to avoid division by values near zero, it was necessary to remove a few values from the calculation for which the outdoor concentration was below the quantification limit defined above. Samples for which the indoor concentration of a species was below that quantification limit were more frequent, but were considered to provide useful information as they generally demonstrated that the indoor/outdoor ratio was very low. In the latter case, the nominal values of the indoor/outdoor ratios were retained during the averaging calculations.

RESULTS AND DISCUSSION

Indoor and outdoor particle mass concentrations

The time series of indoor and outdoor fine and coarse aerosol concentrations for both the summer and winter periods are shown in Figs 2 and 3, respectively. The highest outdoor concentrations for both fine and coarse particles were observed at the Sepulveda House site in downtown Los Angeles (mean concentrations for fine and coarse particles, 30 and 86 $\mu\text{g m}^{-3}$ in the summer; 62 and 70 $\mu\text{g m}^{-3}$ in the winter; highest fine concentration 154 $\mu\text{g m}^{-3}$ on 11 December 1987; highest coarse concentration 132 $\mu\text{g m}^{-3}$ on 22 January 1988). The lowest outdoor concentrations were found at the Getty Museum in Malibu, which is located generally upwind of the city of Los Angeles, near the coast (mean concentrations for fine and coarse particles, 15 and 34 $\mu\text{g m}^{-3}$ in the summer; 21 and 27 $\mu\text{g m}^{-3}$ in the winter; highest fine concentration 49 $\mu\text{g m}^{-3}$ on 11 December 1987; highest coarse concentration 52 $\mu\text{g m}^{-3}$ on 19 August 1987).

Figure 2 shows that indoor fine particle concentrations at the Sepulveda House were nearly identical to those outdoors. This is not surprising in view of the fact that the building has no air filtration system, and that during the hours when the building is open to the public (10 a.m. to 3 p.m. daily, except Sunday), two large doors on the lower floor are kept open, leading to large outdoor air exchange rates. In addition, large gaps around the exterior doors, combined with ceiling vents, lead to relatively large outdoor air exchange rates even when the building is closed. However, as shown in Fig. 3, coarse particle concentrations indoors at the Sepulveda House are substantially lower than those outdoors even under these rapid ventilation conditions. This reduction in coarse particle concentration represents particle depletion by gravitational settling onto indoor surfaces, combined with possible removal during air passage through narrow cracks in the building shell.

The Southwest Museum has no central air conditioning system, and relies on a system of high-powered fans to circulate air through the main section of the building. The California Room, in which the ambient samplers were located, has a local air condi-

tioning system which draws air from an interior hallway that is connected to the main unconditioned space in the building. Consequently, the outdoor air exchange rate for this room is lower than that for the main portion of the building. In the summer months, indoor fine particle concentrations at the Southwest Museum closely approach those outdoors, as was the case at Sepulveda House, while during the winter, there is less correlation between indoor and outdoor fine aerosol concentrations than was observed at Sepulveda House.

In contrast to the situation at the Sepulveda House and Southwest Museum, indoor particle concentrations at the three facilities employing sophisticated HVAC systems with particle filtration are substantially below those measured outdoors. The lowest indoor particle concentrations were found at the Norton Simon Museum. At this site, the outdoor air entering the ventilation system passes through a fibrous mat filter and an activated carbon filter, and is then blended with return air that has also passed through a fibrous mat filter. The supply-air blower at the Norton Simon Museum has been shut off to improve thermal control, resulting in a low outdoor air exchange rate and a high internal air recirculation rate (Table 1). The high recirculation rate results in repeated passes of the indoor air through the particle filters before the air exits the building. The benefits of such a high ratio of recirculated air to outdoor air are demonstrated by the observed average fine particle concentration reduction of 82% achieved at the Norton Simon Museum, in spite of the fact that the single-pass filtration efficiency of the filters used at the Norton Simon Museum ranges from only 5% for 0.3 μm diameter particles to 95% for 2 μm diameter particles, with a weighted-average single-pass filtration efficiency for the fine fraction of about 35% (Nazaroff *et al.*, 1990a). As shown in Fig. 3, the concentrations of coarse particles inside the Norton Simon Museum were extremely low.

Between the summer and winter study periods, an activated carbon system was added to the particle filtration system already in place at the Scott Gallery. At that time, the outdoor air intake dampers to the mechanical ventilation system were closed, again in order to improve thermal control. Thus, during the winter study, the building was operated with a higher ratio of recirculated air to outdoor air than during the summer. That change in operations is clearly reflected in the decrease in indoor fine particle concentrations during the winter period.

At the Getty Museum, the indoor particle concentrations are low in an absolute sense, but the percentage reduction of the indoor concentrations relative to those outdoors is not as impressive as that achieved at the Norton Simon Museum. The Getty Museum employs efficient bag filters for particle removal. However, the ventilation system that serves the main (antiquities) floor only operates between the hours of 8 a.m. and 6 p.m. In addition, when the museum is

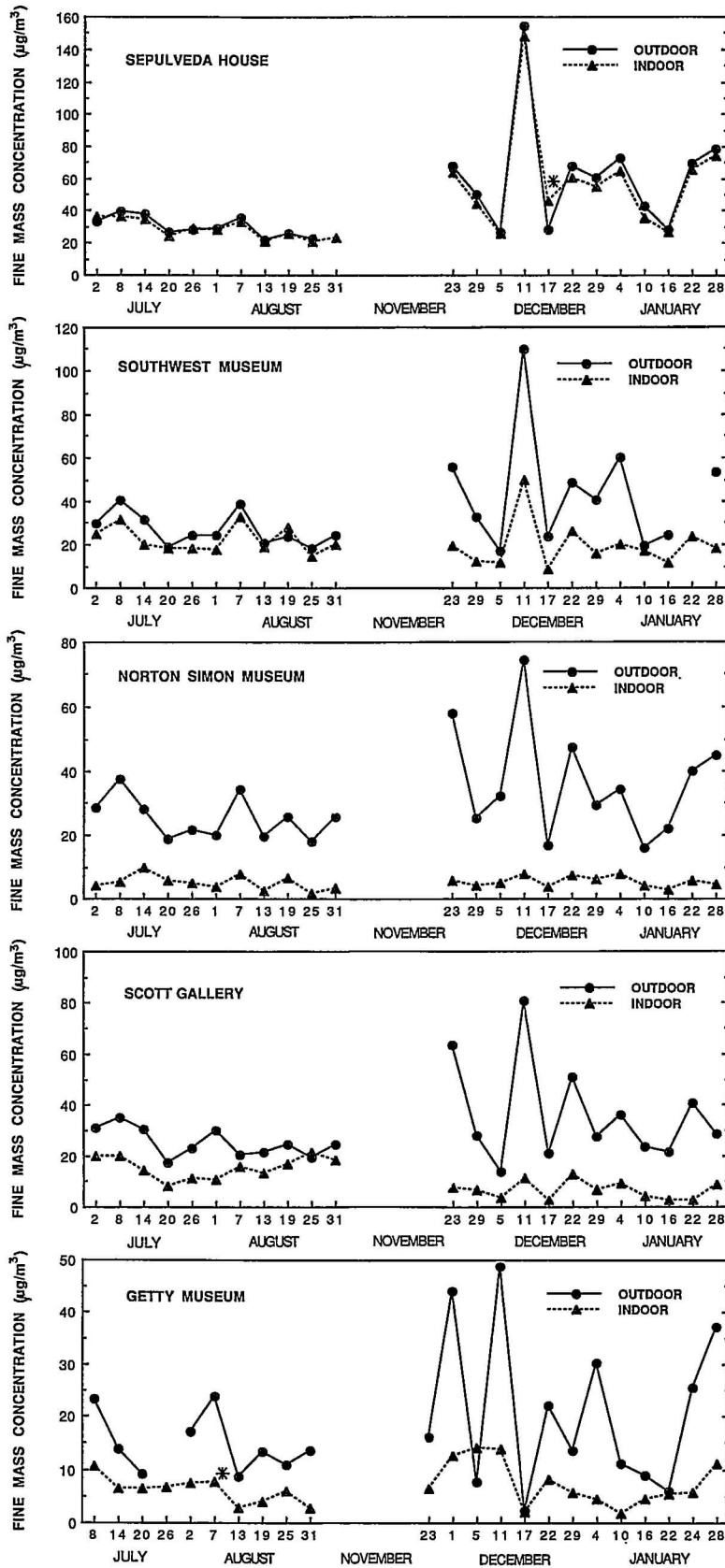


Fig. 2. Time series of ambient fine particle mass concentrations indoors and outdoors at the five museums investigated in southern California.

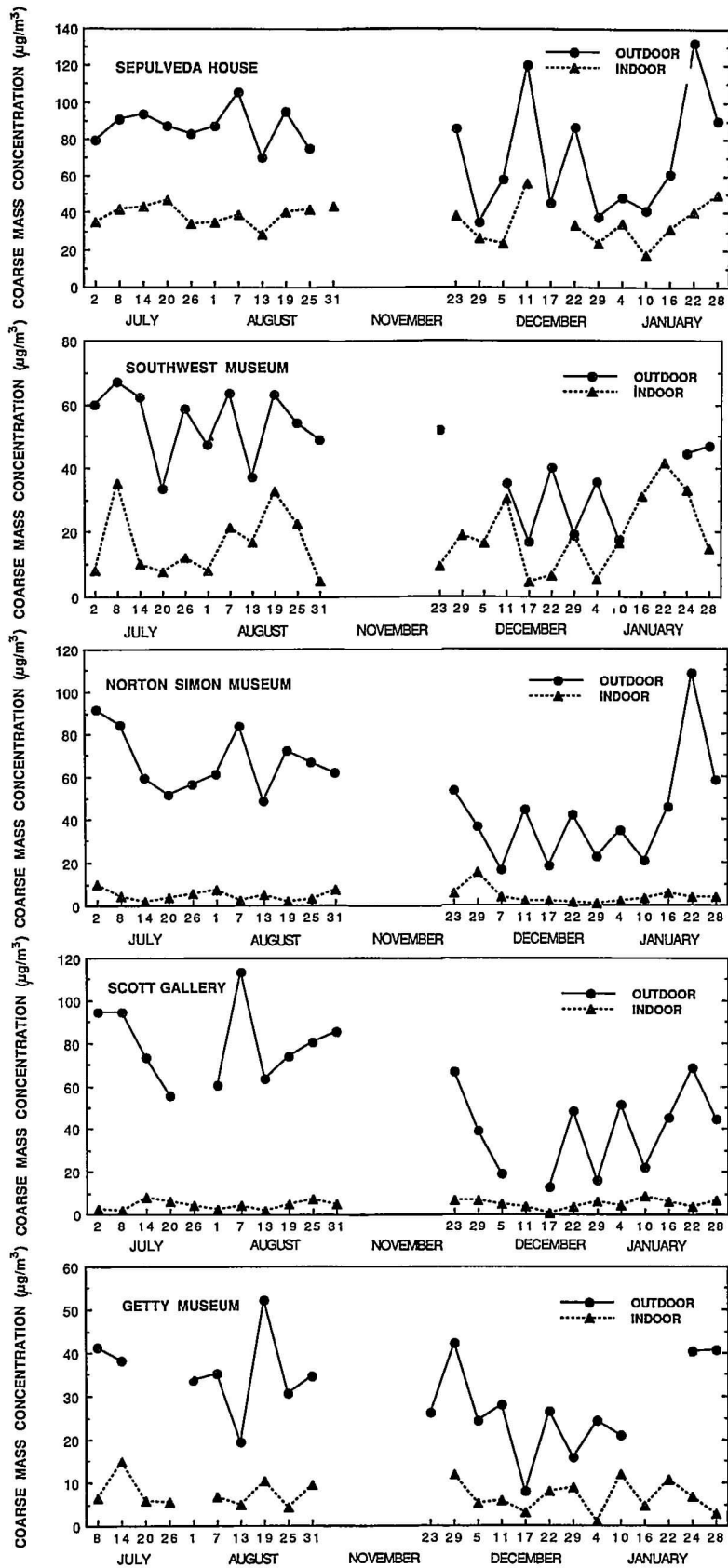


Fig. 3. Time series of ambient coarse particle mass concentrations indoors and outdoors at the five museums investigated in southern California.

open to the public, gallery doors on that floor are generally left open, allowing some unfiltered outdoor air to enter directly into the galleries despite attempts to pressurize the building such that the air flow is directed outward.

Summer and winter seasonal average indoor/outdoor concentration ratios for fine and coarse mass at all sites are presented in Fig. 4. The average indoor/outdoor ratios for particle mass ranged from 0.06 for coarse mass at both the Norton Simon Museum and the Scott Gallery in the summertime to 0.96 for fine mass at the Sepulveda House in the summertime. In general, the indoor/outdoor ratios were larger for fine than for coarse particles, reflecting both the decreased filtration efficiency for fine particles relative to coarse particles and the relatively large gravitational settling velocity of coarse particles. Between the summer and winter experiments, changes were made in the ventilation system at the Scott Gallery. The outdoor make-up air supplied to the building HVAC system was eliminated, while the number of passes of recirculated air through the particle filters was increased. The effect of these changes is clearly shown in Fig. 4. The indoor/outdoor coarse mass ratio increased from 0.06 in the summer to 0.16 in the winter. This can be attributed to the fact that elimination of the outdoor make-up air supply to the mechanical ventilation system forces the exhaust from the building to be balanced by an increase in the amount of air that leaked, unfiltered, into the building through the doors, rather than entering through the ventilation system filters. Coarse particles are removed with reasonable efficiency by even one pass through most HVAC filters yielding the characteristically low indoor/outdoor ratios for coarse particles at most sites having conventional HVAC systems. But if the first filtration pass for coarse particles is eliminated in favor of untreated infiltration air, then coarse particle concentrations can be expected to increase. In contrast, the single-pass fine particle filtration effi-

ency of most HVAC system filters is low enough (*ca* 35% removal efficiency) that several passes of recirculated air through the filters is needed to achieve low indoor fine particle concentrations. While additional fine aerosol undoubtedly also entered the building with the added infiltration air, the concurrent increase in the number of passes of recirculated air through the HVAC system filters at the Scott Gallery more than compensated for the increase in infiltration. As a result, the indoor/outdoor fine particle mass concentration ratio dropped from 0.63 during the summer study to 0.19 during the winter study.

Aerosol chemical composition

Mass balances were constructed on the chemical composition of the indoor and outdoor aerosol samples. Measured organic carbon concentrations were multiplied by a factor of 1.4 in order to account for the estimated contribution of oxygen and hydrogen to the total mass of organic matter (Grosjean and Friedlander, 1975; Benner *et al.*, 1984). Measured elemental concentrations were converted into concentrations of their stable oxides. Soil dust concentrations then were estimated by summing the oxides of aluminum and silicon, plus a fraction of the oxides of iron, calcium, potassium, titanium, manganese, sulfur and phosphorus. The proportion of these latter species which was included in the soil dust category was based on a composite soil dust profile developed for the Los Angeles area by Cooper *et al.* (1988). Finally, the total of these measured soil components was multiplied by a factor of 1.17 (Cooper *et al.*, 1988) to account for the fraction of the soil dust which is composed of the following mineral species which were not measured in Cooper *et al.*'s study: sodium and magnesium oxides, carbonate and water of hydration.

Material balances on the chemical composition of the outdoor and indoor aerosol samples in the fine mode are shown in Fig. 5. In the fine mode, from 90 to 96% of the mass concentration measured at each site in the summer study was accounted for on the basis of the measured chemical species, while 83–92% was accounted for in the winter study. The fine mass balances on the outdoor aerosol indicate a chemical composition similar to that found previously in the Los Angeles Basin (Gray *et al.*, 1986). The outdoor fine particulate levels and composition at the Sepulveda House, Southwest Museum, Simon Museum and Scott Gallery show similarities which reflect the fact that all are located within a 15 × 5 km area in the central Los Angeles Basin. Organic matter constitutes the largest component of the outdoor fine particle mass at these sites, comprising about 40% in both the summer and winter periods. Sulfate is the second largest component of the fine mass in the summertime (20%), while nitrate accounts for 20% of the fine mass in the wintertime. The outdoor fine particle composition at the coastal Getty Museum site differs somewhat from that seen at the sites in the central Los Angeles Basin. Sulfates comprise about 30% of the

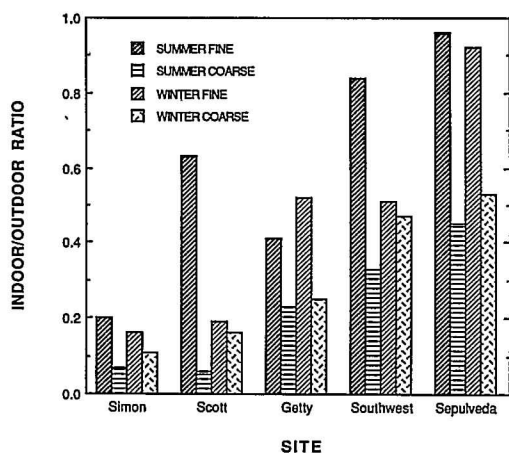


Fig. 4. Seasonal mean indoor/outdoor concentration ratios for fine and coarse particle mass.

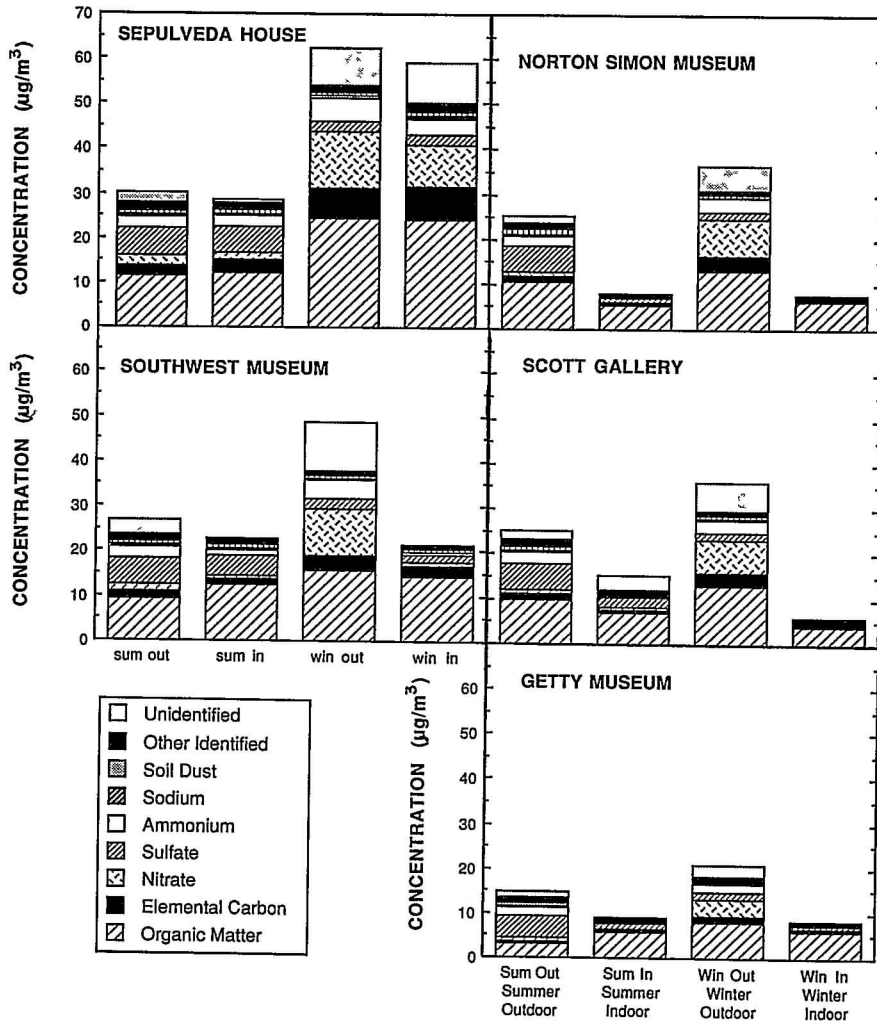


Fig. 5. Seasonal mean chemical mass balances for outdoor and indoor fine particles.

fine particle mass at this site in the summertime, with organic matter comprising only 20%. In the wintertime, a chemical composition similar to the other four sites is observed at the Getty Museum, although lower in absolute concentration.

At every site, the fine particle mode indoors is dominated by organic material, comprising about 40% of the fine mass at the Sepulveda House, up to 80% or more at the Getty and Norton Simon Museums. The measured species accounted for 82–105% of the gravimetrically measured fine mass in the indoor samples in the summertime at the sites with indoor fine mass concentrations $> 10 \mu\text{g m}^{-3}$. At the Getty and Norton Simon Museums, measured species consistently accounted for more than the gravimetrically measured mass in the fine mode. At these sites the measured organic material alone often represented a larger mass concentration than the measured mass. In many of these cases, the measured mass was near its detection limit, leading to a relatively large percentage uncertainty in the mass determinations. The high

organic matter levels are due, in part, to a known positive artifact in the organic carbon determination due to the adsorption of organic vapor onto the quartz filters during sampling (Cadle *et al.*, 1983; McDow, 1986). While this artifact generally amounts to 15–20% of the measured organic carbon concentrations, this percentage increases for lightly loaded samples (McDow, 1986; Rau, 1989). Because organic matter represents such a large fraction of the total mass indoors at the Getty and Norton Simon Museums, the effect of this artifact is magnified. If a positive artifact of 25% is assumed for organic matter and subtracted from the measured concentrations, the sum of measured species at these two sites would balance the measured mass within the error bars on both determinations.

Indoor concentrations of elemental carbon are of particular concern from a soiling perspective. The indoor concentrations of elemental carbon were uniformly higher in the wintertime than in the summertime. The indoor seasonal averages for fine

elemental carbon ranged from $0.14 \mu\text{g m}^{-3}$ at the Getty Museum in the summertime, to $7.4 \mu\text{g m}^{-3}$ at Sepulveda House in the wintertime. At the Norton Simon Museum and the Scott Gallery, the average indoor elemental carbon concentrations during the winter period were $0.7 \mu\text{g m}^{-3}$. This concentration is not large compared to the concentrations observed at the Sepulveda House, nevertheless elemental carbon represented the second largest component of the indoor fine aerosol in the winter samples at those two sites.

Material balances constructed from the chemical analysis of the total particle filter samples were in reasonable agreement with gravimetrically measured total aerosol mass. The per cent of total mass accounted for by chemical analysis ranged from 78 to 105% in the summertime and from 88 to 115% in the wintertime. In order to study the coarse mode aerosol under the conditions of this experiment, coarse mode concentrations and chemical composition were obtained by subtracting measured fine particle characteristics from measured total aerosol characteristics.

In the coarse mode, 62–75% of the measured mass outdoors was assigned to identified chemical species in this manner, as shown in Fig. 6. Because coarse mode data are obtained by difference, the uncertainties in the material balance are higher than for the fine aerosol data. However, the relatively larger amount of unidentified material still merits some discussion.

Soil dust is the largest component of the coarse particle samples shown in Fig. 6. In one study conducted in Denver, soil dust accounted for 80% of the coarse mass in particles $<30 \mu\text{m}$ in diameter (Counness *et al.*, 1980). In the Los Angeles area data of Fig. 6, soil dust accounted for only about 40% of coarse particle mass. In the present study, soil dust concentrations were measured by XRF, a technique which is not well suited for the analysis of large particles. X-ray signals produced by an element deep within a large particle may be re-absorbed before reaching the surface of the particle and beyond to the detector. The depth to which an element may be detected varies with the atomic number of the element, and is smallest for the elements Al and Si, which are the two largest

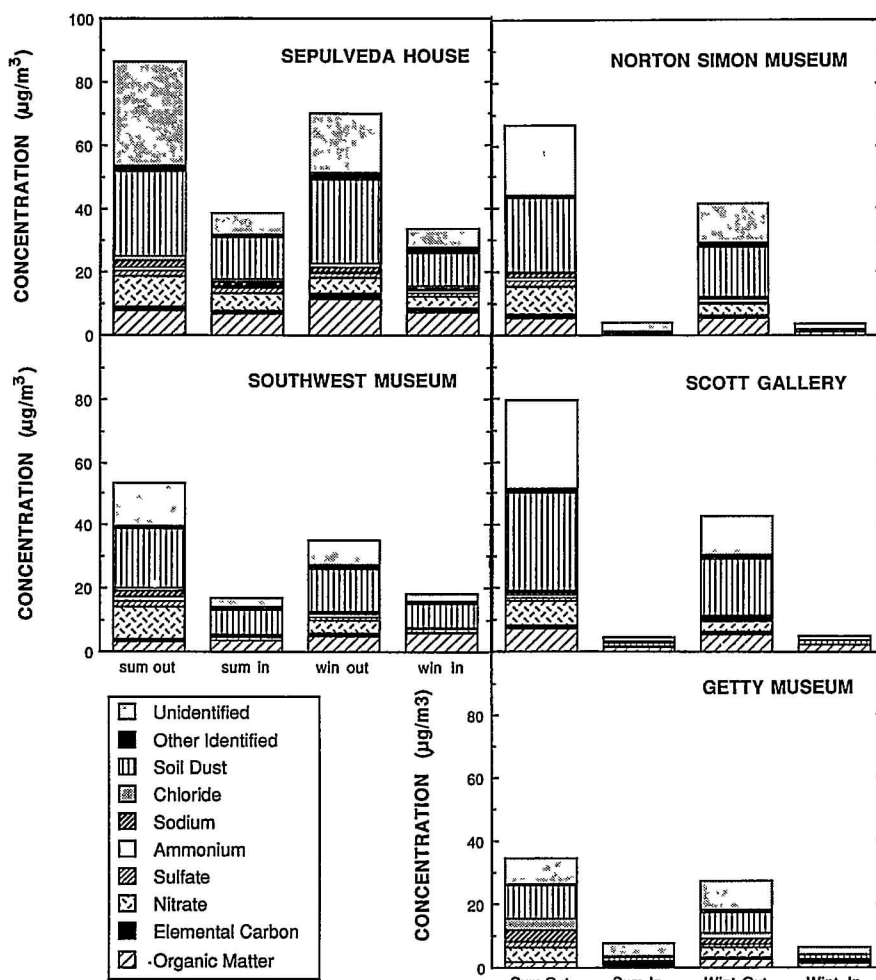


Fig. 6. Seasonal mean chemical mass balances for outdoor and indoor coarse particles.

components of soil dust. A calibration factor is applied to the concentrations of these elements in samples containing large particles. In this study, the calibration factor was obtained by comparison of the aluminum concentrations measured by XRF to those measured by neutron activation analysis. Because only a subset of the samples were analysed by neutron activation, and because Al was the only element on which this correction factor was based, the soil dust concentrations reported here are considered to be uncertain.

Another contributor to the unidentified coarse aerosol mass includes the concentrations of species which were not measured, such as aerosol liquid water. Liquid water has been estimated to comprise as little as 5% of aerosol mass at humidities below 70% (Countess *et al.*, 1980; Witz *et al.*, 1987), up to 40–50% at 70% relative humidity (Ho *et al.*, 1974; Hanel, 1976). In the present study, the material balances for the fine fraction suggest a liquid water content of approximately 10%. The unidentified coarse particle mass is therefore thought to consist of a combination of soil dust and liquid water.

At the Getty Museum, the coarse particle mode outdoors contains nearly equal amounts of soil dust and sea-salt (Na + Cl) mass, reflecting its coastal location. At the inland sites, the sea-salt concentration is greatly reduced, and organic material and nitrates are second in concentration behind the soil dust. The coarse mode composition shows little variation from summer to winter, with absolute concentrations somewhat lower in the wintertime than in the summertime.

The coarse particle mode indoors was very small at the three sites with engineered HVAC systems and was dominated by soil dust and organic material. Coarse mode concentrations were virtually undetectable at the Norton Simon Museum, where coarse mass concentrations appeared to be $\leq 4 \mu\text{g m}^{-3}$. At the other four sites, the measured species accounted for 46–74% of the measured indoor coarse particle mass concentration in the summertime, and 64–84% in the wintertime. The percentage of coarse mass accounted for by measured chemical species indoors at the Sepulveda House was 81% for both the summer and winter periods, compared to 62 and 73% obtained outdoors at the same site. This is further evidence that it was the largest particles, which would be depleted in the indoor air, which were responsible for the lack of agreement.

Indoor/outdoor ratios of individual chemical species

Summer and winter seasonal averages of the indoor/outdoor ratios for the major chemical species are presented in Fig. 7 for both the fine and coarse modes. At most sites, there were large species-to-species variations in the indoor/outdoor ratio. The largest indoor/outdoor ratios are observed for organic matter; small ratios often are observed for nitrate and ammonium ions. The indoor/outdoor ratios for the

coarse mode fraction of all species were lower than those for the fine fraction. Indoor/outdoor ratios for several species were immeasurably low in the coarse mode.

At the Sepulveda House, the seasonal averages of the indoor/outdoor ratios of all species were very close to 1.0 for fine particles, as would be expected if the high indoor fine particle concentrations were explained directly by intrusion of outdoor air with little or no fine particle removal. The lowest indoor/outdoor ratios were observed for ammonium and nitrate ions, with seasonal average values of around 0.7. The seasonal averages of the indoor/outdoor ratio for individual components of the coarse particle fraction ranged from 0.4 to 1.0 at the Sepulveda House. At the Southwest Museum, the seasonally averaged indoor/outdoor ratios for fine particles ranged from 0.3 for nitrate ion to 1.4 for organic matter during the summer period, and from 0.1 for nitrate ion to 1.3 for soil dust during the winter period. By comparison with the Sepulveda House, the species-to-species variability of the indoor/outdoor ratio was much greater at the Southwest Museum.

At the Norton Simon Museum, the fine particle indoor/outdoor ratios ranged from <0.03 for nitrate ion to 0.51 for organic matter. Indoor/outdoor ratios for coarse particles were immeasurably low for most species at the Norton Simon Museum. At the Scott Gallery the summer average fine particle indoor/outdoor ratios ranged from 0.2 for sodium and ammonium ions, up to 0.7 for organic matter. During the winter period, the indoor/outdoor ratios for most fine particle species were lower: 0.3 for organic matter and <0.02 for ammonium ion. The indoor/outdoor ratio for fine soil dust, however, was higher in the winter than in the summer. As observed at the Norton Simon Museum, indoor/outdoor ratios for coarse particles were immeasurably low for most species at the Scott Gallery, confirming the effectiveness of filtration of coarse particles at both sites.

At the Getty Museum, the indoor/outdoor ratio for organic material in the summer fine particle samples was 2.1. In absolute terms this translates into $2.7 \mu\text{g m}^{-3}$ higher concentration indoors than outdoors. The indoor/outdoor ratios of all other species were in the range 0.2–0.6 in the summer fine particle samples. The indoor/outdoor ratio for organic material in the wintertime at the Getty Museum was 1.0. While the indoor concentrations of organic material were similar in both seasons, the outdoor concentration was much larger in the wintertime. This will be discussed further in the section on indoor sources. The indoor/outdoor ratios for the coarse mode fraction at the Getty Museum ranged from 0.06 for sodium, to 0.57 for coarse organic material. Interestingly, the indoor/outdoor ratio for chloride was larger than that for sodium, which is noteworthy because these two elements are generally considered to be present in the same sea-salt particles.

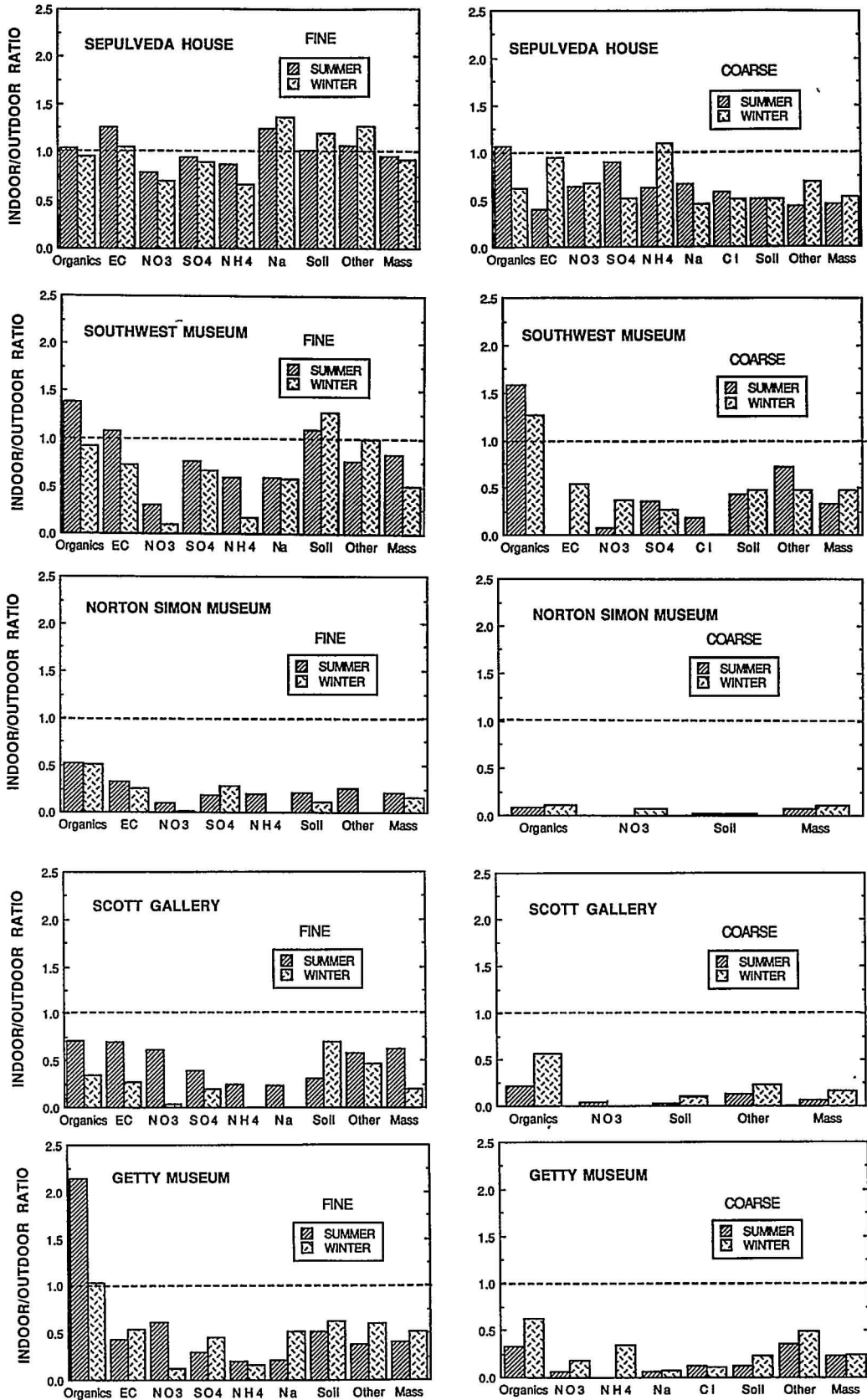


Fig. 7. Seasonal mean indoor/outdoor concentration ratios for individual chemical species.

Detection of indoor sources

The observation that fine particulate organic matter concentrations inside the museums studied are sometimes higher than those outdoors suggests that there may be indoor sources of organic particles. The possible existence of indoor sources of organic matter inside these museums is interesting because the usual activities associated with indoor organic particle generation, such as smoking and operation of indoor combustion devices, are prohibited inside museums. From observations of the operations at these facilities, possible indoor sources include particles shed by the visiting public as well as maintenance operations, such as floor waxing and vacuum cleaning.

The purpose of this section is to use the relationship between indoor and outdoor fine particle component concentrations to detect the presence or absence of a significant rate of indoor emissions of airborne particles. The basis for this exploration is a simplified, steady-state material balance model (Alzona *et al.*, 1979; Nazaroff and Cass, 1991). Elsewhere, a more detailed model of indoor aerosol dynamics is developed (Nazaroff and Cass, 1989b) and applied to short-term data collected at three of the museum sites (Nazaroff *et al.*, 1990a).

This simplified model yields the following relationship between the indoor and outdoor concentrations of the suspended fine particle mass of a particular component, k :

$$C_{in}(k) = A(k) + B(k) C_{out}(k). \quad (1)$$

In this formulation, $A(k)$ may be thought of as the contribution of indoor emissions to indoor concentrations. The dimensionless parameter, $B(k)$, represents the indoor-outdoor ratio that would prevail in the absence of indoor emissions. In terms of ventilation rates, filtration efficiencies and deposition rates onto indoor surfaces, $A(k)$ and $B(k)$ may be expressed as (Nazaroff and Cass, 1991):

$$A(k) = \frac{S(k)}{\eta_{ii}(k) f_{ii} + f_{oi} + f_{ox} + \sum_j A_j v_d(k)} \quad (2)$$

$$B(k) = \frac{f_{oi} + (1 - \eta_{ox}(k)) f_{ox}}{\eta_{ii}(k) f_{ii} + f_{oi} + f_{ox} + \sum_j A_j v_d(k)} \quad (3)$$

In these expressions, f_{oi} , f_{ox} and f_{ii} are the rates of air flow associated with infiltration, introduction of outdoor air through the mechanical ventilation system, and recirculation, respectively; $\eta_{ox}(k)$ and $\eta_{ii}(k)$ are the effective single-pass removal efficiencies for component k by filters in the supply and recirculating ventilation line, respectively; $S(k)$ is the emission rate of component k into the indoor volume; A_j is the area of the j th surface; and $v_d(k)$ is the effective deposition velocity of component k onto the j th surface.

A linear regression model based on Equation (1) was structured in which the time series of indoor fine particle component concentrations was regressed onto the corresponding outdoor concentrations. Best-

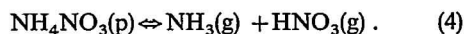
fit values of the parameters $A(k)$ and $B(k)$ were determined for each component and for each site. Summer and winter data were combined in most cases to maximize the number of observations used in the analysis. At the Scott Gallery, the summer and winter data were treated separately, because of the previously mentioned change that was made in the ventilation system between the summer and the winter study periods. Days for which the indoor and outdoor samples were not obtained concurrently were discarded.

The results from this analysis are presented in Table 2. The contribution of indoor emissions to indoor concentrations of fine organic particles range from 2 to $7 \mu\text{g m}^{-3}$, while the values of $A(k)$ for the other components are $\leq 1 \mu\text{g m}^{-3}$ in all cases. Figure 5 shows that the indoor concentration of organic matter in fine-mode particles ranges from 3 to $24 \mu\text{g m}^{-3}$. Thus, indoor emissions account for a significant portion of the total indoor concentration of fine organic particle mass.

The parameters $B(k)$ at a given site would be expected to be the same for species associated with similarly sized particles. The results presented in Table 2 show that values of $B(k)$ are generally constant at a given site for organic matter, elemental carbon, sulfates and total fine particle mass. However, at some sites, $B(k)$ values are higher for elemental carbon than those for organic matter or sulfate, consistent with the observation that elemental carbon is associated with very fine particles that are removed with lower efficiency by the HVAC systems than is the case for the latter two species (Quimette, 1981; Hansen and Novakov, 1989).

The $B(k)$ parameters for ammonium and nitrate ions, however, are significantly lower than for the other species in nearly all cases. In the case of ammonium ion, the summertime values are similar to those for the other species, but in the wintertime, the values are greatly reduced. This is significant because ion balances on the aerosol samples show that ammonium ion must be primarily associated with sulfate in the summertime, while in the wintertime it is primarily associated with nitrate. Thus, the indoor-outdoor concentration ratio for wintertime ammonium nitrate appears to be significantly smaller than that of the other fine species in those buildings that possess a heating system (i.e. all, except for the Sepulveda House).

Particulate ammonium nitrate in the atmosphere can exist in equilibrium with gaseous ammonia and nitric acid (Stelson *et al.*, 1979; Hildemann *et al.*, 1984):



When air which is in equilibrium at outdoor ambient conditions enters a building, a shift in this equilibrium could occur due to temperature change (e.g. heating during the winter season) or by removal of the gaseous species both by the ventilation system and by the

Table 2. Parameters $A(k)$, $B(k)$ and correlation coefficients (R) from the linear regression of indoor fine* particle concentrations on outdoor fine particle concentrations for major chemical species (see Equations 1-3)

		J. Paul Getty Museum	Norton Simon Museum	Sepulveda House	Southwest Museum	Scott Gallery	
						Summer	Winter
Organic matter	$A\ddagger$	4.96 ± 0.32	4.18 ± 0.49	1.99 ± 0.54	7.37 ± 3.14	4.05 ± 3.08	2.31 ± 0.50
	B	0.172 ± 0.042	0.151 ± 0.039	0.867 ± 0.026	0.473 ± 0.227	0.291 ± 0.304	0.139 ± 0.038
	R	0.682	0.656	0.992	0.405	0.303	0.775
Elemental carbon	$A\ddagger$	0.09 ± 0.05	0.13 ± 0.05	0.27 ± 0.26	0.65 ± 0.27	0.41 ± 0.11	0.26 ± 0.25
	B	0.369 ± 0.042	0.212 ± 0.019	1.031 ± 0.046	0.493 ± 0.103	0.298 ± 0.090	0.162 ± 0.088
	R	0.892	0.928	0.981	0.716	0.739	0.523
Sulfate	$A\ddagger$	0.23 ± 0.07	0.14 ± 0.05	-0.32 ± 0.16	-0.13 ± 0.28	0.88 ± 0.37	0.00 ± 0.05
	B	0.250 ± 0.018	0.154 ± 0.012	1.014 ± 0.032	0.742 ± 0.058	0.220 ± 0.059	0.197 ± 0.029
	R	0.952	0.941	0.990	0.939	0.781	0.915
Ammonium (summer)	$A\ddagger$	-0.09 ± 0.20	0.14 ± 0.08	-0.01 ± 0.25	0.00 ± 0.29	0.29 ± 0.14	
	B	0.257 ± 0.096	0.113 ± 0.030	0.886 ± 0.085	0.579 ± 0.104	0.084 ± 0.053	
	R	0.738	0.797	0.965	0.880	0.464	
Ammonium (winter)	$A\ddagger$	0.05 ± 0.05	-0.01 ± 0.03	-0.32 ± 0.21	-0.12 ± 0.17		0.00 ± 0.02
	B	0.072 ± 0.022	0.018 ± 0.008	0.778 ± 0.032	0.180 ± 0.033		0.014 ± 0.006
	R	0.706	0.592	0.992	0.857		0.591
Nitrate	$A\ddagger$	0.23 ± 0.05		-0.14 ± 0.20	0.02 ± 0.20	0.50 ± 0.31	0.05 ± 0.15
	B	0.012 ± 0.012	NA \ddagger	0.778 ± 0.016	0.124 ± 0.021	0.062 ± 0.272	0.026 ± 0.020
	R	0.228		0.996	0.785	0.077	0.397
Soil dust	$A\ddagger$	0.07 ± 0.03	0.04 ± 0.03	0.15 ± 0.05	0.01 ± 0.22	0.06 ± 0.19	0.08 ± 0.24
	B	0.271 ± 0.056	0.071 ± 0.042	0.875 ± 0.053	1.079 ± 0.335	0.230 ± 0.203	0.517 ± 0.373
	R	0.745	0.355	0.967	0.574	0.354	0.420
Mass	$A\ddagger$	3.24 ± 1.17	3.01 ± 0.95	-0.36 ± 0.94	11.45 ± 5.00	9.49 ± 6.21	1.84 ± 2.11
	B	0.189 ± 0.052	0.071 ± 0.028	0.943 ± 0.016	0.673 ± 0.151	0.234 ± 0.242	0.130 ± 0.060
	R	0.639	0.499	0.997	0.498	0.307	0.583

*Fine particles $<2.1 \mu\text{m}$ in diameter.

†Units are $\mu\text{g m}^{-3}$.

‡Not available, due to indoor concentrations below the detection limit.

interior surfaces of the buildings. In addition, ammonium nitrate collected on Teflon filters will volatilize and dissociate if clean air is drawn through the filters (Appel *et al.*, 1980). Because nitric acid concentrations inside many of the museums studied were extremely low (Salmon *et al.*, 1990), it is possible that some dissociation of this type occurred either in the air inside the buildings or during the indoor sampling. The dynamics of aerosol ammonium nitrate inside buildings may be complex and deserves more detailed study than was possible in this investigation.

CONCLUSIONS

The concentration and chemical composition of airborne particulate matter inside and outside five museums was measured. Indoor/outdoor ratios for fine particles ranged from 0.16 at the Norton Simon Museum in the winter to 0.96 at the Sepulveda House in the summer, while indoor/outdoor ratios for coarse particles were lower, ranging from 0.06 at the Norton Simon Museum and the Scott Gallery in summer, to 0.53 at the Sepulveda House in the winter. At the Sepulveda House, which lacked any air filtration system, indoor fine particle concentrations were vir-

tually as high as those outdoors, indicating that urban museums that lack an environmental control system can face a soiling hazard nearly as great as that outdoors. At the sites where the ventilation system included conventional HVAC system particle filters, the indoor airborne particles were mostly fine ($<2.1 \mu\text{m}$ in diameter) and consisted largely of organic matter. At two sites, indoor concentrations of organic matter exceeded outdoor concentrations by an average of about $3 \mu\text{g m}^{-3}$. Linear regression analysis of indoor vs outdoor concentrations of major species indicated that indoor sources of organic matter may exist at all sites, but that none of the other measured species appear to have major indoor sources at these museum facilities. Indoor/outdoor relationships for elemental (black) carbon particles and soil dust particles in museums are of particular interest because those species are light absorbing and can produce visible soiling deposits on works of art. The indoor-outdoor concentration ratio for fine elemental carbon and fine soil dust particles was roughly 0.2-0.5 for the facilities with particle filtration systems. For buildings that lack a conventional particle filtration system, fine indoor elemental carbon and soil dust particle concentrations were approximately the same as the concentrations outdoors, and coarse particle

concentrations in such facilities were approximately half as high as those outdoors. This implies that museums should be particularly watchful for the occurrence of indoor soiling problems caused by elemental carbon and soil dust particles.

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