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Indoor environmental quality index for conservation environments: the importance of including particulate matter

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ABSTRACT: It is commonly known that the conservation state of works of arts exhibited inside museums is strongly influenced by the indoor environmental quality (IEQ). Heritage institutions traditionally record and evaluate their IEQ by monitoring temperature, relative humidity, and -more rarely- light. However, smart use of technology enables monitoring other parameters that give a more complete insight in environmental ‘air aggressiveness’. One of this parameters is particulate matter (PM) and especially its concentration, size distribution and chemical composition. In this work, we present a selection of data sets which were obtained in a measuring campaign performed in the War Heritage Institute in Brussels, Belgium. A continuous monitoring of PM concentration with a light scattering based particle counter was performed. In addition the daily mass concentration and size distribution of airborne PM was monitored by means of Harvard impactors. The chemical composition of sampled PM was inferred from the results of XRF and IC analysis. The insights from these datasets are combined with the results of traditional environmental monitoring (temperature, relative humidity and light intensity), and assessed against the recommended guidelines for conservation environments. By using an integrated approach based on the calculation of an IEQ-index, we present a straightforward methodology to evaluate and visualize the IEQ including also continuous PM monitoring. It is clear from the results of this study how including PM in IEQ analysis allows to identify potential risks for museum collections that remain invisible when only traditional parameters are considered.

KEYWORDS: Indoor Environmental Quality, Airborne Particulate Matter, Cultural Heritage, Conservation
1. INTRODUCTION

In order to protect and conserve the relics of human history, it is important to address the environmental factors that may cause damage to museum collections and cultural heritage in general. Consequently, continuous indoor environmental quality (IEQ) measurements are a prerequisite to evaluate best practices in an exhibition or storage environment.

Currently, the continuous evaluation of conservation environments is usually based on physical parameters only, such as temperature, relative humidity and the intensity of visible (Vis) and ultraviolet (UV) light [1-9]. It is generally accepted that these parameters pose the largest threat towards hygroscopic and light sensitive objects, influencing also the conservation of general collections [10]. Several commercial systems are available on the market to monitor these physical parameters. However, deterioration is also influenced by gaseous pollutants and particulate matter (PM) [11-16]. A large number of studies taking into account the average levels of these pollutants in conservation environments has been published through the years [17-28]. However, only in few cases a continuous monitoring was performed [29-32]. This can be explained for gaseous pollutants by the lack of suitable sensors for a continuous and sensitive monitoring. The commercially available sensors for common pollutants such as O₃, NO₂, SO₂, H₂S, formaldehyde and acetic acid [33-42] are usually created for industrial applications and present limit of detections higher than the recommended levels in conservation environments [43]. On the other hand, PM can rather easily be continuously monitored using airborne particle counters based on light scattering principles. Both precise and accurate sensors created for clean rooms monitoring and inexpensive sensors for home
or office applications exist [44-46]. However, these systems are not yet employed to their full potential in cultural heritage.

In this article, a system is presented that enables the continuous monitoring of temperature (°C), relative humidity (%), illuminance of visible light (lux), UV light intensity (mW/m²) and PM concentration (number of particles/m³) in a simultaneous and synchronous way. Extended measuring campaigns were performed at two locations in the former Royal Museum of the Armed Forces and of Military History, or shorter Royal Military Museum, in Brussels, Belgium. The Royal Military Museum is integrated in the War Heritage Institute (WHI) since May 1st 2017. [47] Since the risk associated with the presence of airborne PM in conservation environments strongly depends on the concentration and aerodynamic dimension of the particles [27, 48], but also on their chemical composition [13-14, 49-51], the continuous monitoring is supplemented by an in depth chemical analysis of PM. The total mass concentration and chemical composition of fine (PM₁, PM₂.₅) and coarse particles (PM₁₀) is analyzed. This qualitative and quantitative analysis of PM allows to register sudden changes of the indoor environmental quality which are invisible with physical parameters. The available guidelines for conservation environments are then used to calculate an IEQ-index [52] by combining the information of temperature, relative humidity and light exposure with that of PM. No univocal guideline has been found in literature prescribing an optimal PM concentration for conservation environments expressed in number of particles per unit volume. A mass per unit volume threshold is therefore considered and converted into number of particles/m³ values on the basis of site-specific empirical correlations. Taking into account the extreme variety of objects exposed in the museum, the ASHRAE maximum limit for general collection of 10 µg/m³ for PM₂.₅ is applied [1].
This article discusses in detail how and why continuous PM data should be included in indoor environmental quality studies. The versatile methodology discussed can be applied to the monitoring of environmental quality in any type of indoor conservation environment. When the main concern is directed towards objects or materials with more specific needs, potentially dangerous conditions can be promptly recognized by simply changing the threshold values applied in the calculation of the IEQ-index.

2. MATERIALS AND METHODS

2.1. Sampling locations and campaigns. The Royal Military Museum is located in the Parc du Cinquantenaire area in the city center of Brussels in Belgium (50°50'29.4"N 4°23'31.6"E). The building dates from the end of the 19th century. The collections of the Royal Military Museum, now integral part of the War Heritage Institute (WHI), illustrate ten centuries of military history ranging from medieval times up until this day. The more than 125,000 objects include amongst others uniforms and headgear, edged weapons and firearms but also paintings and sculptures, medals and music instruments. The museum building even houses airplanes, armored vehicles and artillery. The collections consist of an extremely wide range of materials. The museum occupies five large exhibition galleries covering approximately 40,000 m².

First, measurements were performed in one location from April 11 to April 24 in 2016, followed by a period in a second place from April 25 to May 9 in 2016. The first location is the central storage facility situated underground (further indicated as ‘storage’), the second location is the Historic...
Gallery, situated at the ground level of the Army museum and further indicated in the text as ‘gallery’ (Figure 1).

**Figure 1. (2-column fitting image)** Images of the sampling locations: the ‘Historic Gallery’ and the central alley of the storage facility with indication of the sampling unit (white box).

The collection stored in the storage is a mixed collection, including paintings, textiles, leather, metal, stone and ceramic objects. The measuring equipment was located in the central alley of the storage. This space is equipped with a HVAC-system with two different types of filters (TL7U600 class F7 and TM9U600 class F9, AL-KO KOBER SE, Germany) to control environmental conditions and PM levels. The storage is located above a highway tunnel and indirectly connected to it through a shared emergency exit. This connection could negatively affect the IEQ in this environment, potentially causing high levels of gaseous pollutants and particulate matter.

The gallery was inaugurated in 1923 and is dedicated to the Belgian army between 1831 and 1914. It houses a collection consisting of hundreds of oil paintings on canvas, uniforms, flags, weapons and musical instruments. The gallery has a large roof with a skylight that enables direct sunlight entering...
(Figure 1), causing a severe temperature increase on sunny days. In winter period the gallery is heated but no cooling nor humidity control are installed. This strongly influences the thermo-hygrometric conditions for the collection. The gallery is not equipped with an air filtering system, which also might affect the PM levels. The objects are exhibited on the walls and in oaken display cases originating from the early 20th century. This type of display cases has a high air exchange rate, allowing infiltration of dust [53-54].

The very different environmental conditions in the two locations do not allow to obtain an overview of the conservation conditions in the whole museum, but represent an ideal context for testing the methodology discussed in this work. The “controlled” environment in the storage and the “uncontrolled” environment in the gallery, in fact, ideally represent the two extremes of the possible range of conditions that can be found in this conservation environment.

2.2. Monitoring of environmental parameters (temperature, relative humidity, Vis and UV light, CO₂). The monitoring of environmental parameters was performed with a frequency of 15 minutes during the sampling campaigns. For simplicity, and to underline the difference between these monitoring techniques and other PM sampling methods used, this “semi-continuous” monitoring will be referred to as “continuous” throughout the rest of this article. Well-calibrated, commercial off-the-shelf sensors were connected to a multi-purpose data logger (DataTaker DT85, Thermo Fischer scientific, Australia) [55]. Data was available online using a 4G network. Temperature, relative humidity and carbon dioxide (CO₂) were measured with a GMW90 sensor (Vaisala, Finland) [56], while the intensity of visible and UV light were measured with the sensors SKL310 [57] and SKU421 [58] (Skye Instruments, UK). For the light sensors, the orientation and distance from the light source
have a substantial impact on the intensity. The sensors were placed in a vertical position to simulate vertically stored objects (e.g., paintings). To monitor the real light exposure, the sensors should be placed next to the object of interest. The monitoring unit and its sensors were placed on a table at a height of 1.10 m. This corresponds with the average height of the lower edge of exhibited paintings. [59]

2.3. Sampling and analysis of particulate matter. PM was monitored with a frequency of 15 minutes by means of a Lighthouse Handheld 3016 IAQ continuous air particle counter (Lighthouse Worldwide Solutions, USA) [60]. This device is designed to respect the ISO 21501-4 directives in terms of accuracy and precision for the measurement of size and concentration of particles suspended in air. Particles in the range of 0.3 µm – 10 µm are resolved into 6 particle size channels. Black carbon was continuously monitored using a Portable Aethalometer® Model AE42 (Magee Scientific Corporation, USA) [61]. The concentration is deduced from the absorption of an 880 nm light bundle, black carbon being the only aerosol component that significantly absorbs at this wavelength.

The monitoring campaign was supplemented by an in depth analysis of PM mass. Different fractions of particulate matter (PM₁, PM₂.₅, PM₁₀) were sampled with 3 different Harvard-type impactors (MS&T area sampler, Air Diagnostics and Engineering Inc., USA). [62] PM₂.₅ and PM₁₀ were collected on mixed cellulose ester filters (SKC MCE 0.45 µm 225-1914, SKC Limited, UK), PM₁ on Teflon membrane filters (Pall 7227 TK15-G3M 37MM, Pallflex® Air Monitoring Filters, PALL Life Sciences, USA). All the fractions of PM were sampled for 24 hours, for the period of two consecutive weeks during each sampling campaign. The pumping units connected to Harvard impactors worked with the flow rate of 10 L/min for PM₂.₅ and PM₁₀ and with the flow rate of 23 L/min for PM₁. Changes in the average flowrate were monitored by means of a rotameter model P.
equipped with a 044-14-N tube (Aalborg Instruments & Controls, USA). Outliers based on the flowrate were identified with the interquartile range method and excluded from further considerations. The filters were weighed before and after sampling to obtain the mass of collected matter and calculate PM concentrations in µg per unit volume of sampled air. The gravimetric analysis was performed on an analytical microbalance MT5 (Mettler, USA) with capacity: 5.1 g, readability: 0.001 mg, repeatability: 0.0008 mg and linearity: 0.004 mg.

The elemental composition of the different fractions of PM was determined using a energy-dispersive X-ray fluorescence spectrometer (EDXRF-Epsilon 5, PANalytical, The Netherlands), equipped with a Gd X-ray tube (600W, 100 kV and 24 mA). Three different sets of settings were used to cover the entire range of elements of interest. Na, Mg, Al, Si, P, S, Cl, K and Ca concentrations were determined by using a Ti secondary target with an operating voltage, current and measuring time of respectively 25 kV, 24 mA and 500 s. Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn and Ba concentrations were determined using a Ge secondary target (75 kV; 8 mA; 1500 s). Finally a Mo secondary target (100 kV; 6 mA; 1500 s) was used to determine the As, Se, Br, Sr, Zr and Pb concentrations.

Then, the water-soluble fraction was ultrasonically leached in 8 mL of Milli-Q water allowing a quantitative analysis of positive and negative ions by means of ion chromatography (Metrohm 883 basic IC plus, Switzerland). Metrosep C 6 -150/4.0 column was used for the analysis of cations, Metrosep Asupp5 – 250/4.0 for the analysis of anions.

On the basis of x-ray fluorescence (XRF) and ion chromatography (IC) results the relative amounts of soil dust, sea salts, ammonium salts and mineral salts (soluble fraction of soil dust) in the different PM fractions was estimated. The method used is described in detail by Anaf et al. [28].
2.4. Data evaluation.

2.4.1. Univariate data analysis. Eventual anomalies and sudden perturbations of single parameters are thoroughly investigated in order to identify and interpret situations of potential risk for the museum objects. To deepen the understanding on the causes of anomalous behaviors the data obtained in the different indoor locations are assessed against the outdoor values for the corresponding period.

2.4.2. IEQ-index calculation.

The IEQ assessment was performed using an IEQ-index that combines the measurement data of temperature, relative humidity, visible light, UV, and PM$_{2.5}$ into a simpler magnitude representing the degree of air aggressiveness for collections [52]. This index was calculated based on thresholds already applied and well recognized by the cultural heritage conservation community: the ASHRAE standards for ‘Museums, Libraries, and Archives’ for temperature, relative humidity, and PM$_{2.5}$, [1] and the Thomson standards for visible light and UV [63].

The ASHRAE standards propose a set of five control classes depending on the potential risk for a collection under specific thermo-hygrometric conditions (Table 1). These classes vary from situations with the lowest possible risk for most objects (class AA) to a considerably high risk of damage for most of the artefacts (class D). The specific IEQ-index for temperature and relative humidity is defined by associating each of the aforementioned classes to a numerical value in each data point.

The index range from 100 to 0, as it covers the situations from the lowest to the highest risk of degradation or damage. Consequently, class AA corresponds to the maximum IEQ-index=100, A to IEQ-index=80, B to IEQ-index=60, C to IEQ-index=40, and D to IEQ-index=20. Below class D (relative humidity larger than 75%) the IEQ-index will reach the minimum value of 0.
Table 1: Overview of the ASHRAE standards for temperature and relative humidity [1] and their correspondence to the IEQ-index.

<table>
<thead>
<tr>
<th>ASHRAE Class</th>
<th>Temperature</th>
<th>Relative Humidity</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Class AA</td>
<td>15-25°C</td>
<td>50%</td>
<td>No risk of mechanical damage to most artefacts and paintings.</td>
</tr>
<tr>
<td>IEQ-index=100</td>
<td>Short fluctuations: ±2°C. Seasonal adjustments: ±5°C</td>
<td>Short fluctuations: ±5%. No seasonal adjustments.</td>
<td></td>
</tr>
<tr>
<td>Class A</td>
<td>15-25°C</td>
<td>40-60%</td>
<td>Small risk of mechanical damage to high vulnerability artefacts; no mechanical risk to most artefacts, paintings, photographs, and books.</td>
</tr>
<tr>
<td>IEQ-index=80</td>
<td>Short fluctuations: ±2°C. Seasonal adjustments: +5°C -10°C</td>
<td>Short fluctuations: ±10%. Seasonal adjustments: No variation.</td>
<td></td>
</tr>
<tr>
<td>Class B</td>
<td>15-25°C</td>
<td>40-60%</td>
<td>Moderate risk of mechanical damage to high-vulnerability artefacts; tiny risk to most paintings and photographs and no risk to most artefacts and books.</td>
</tr>
<tr>
<td>IEQ-index=60</td>
<td>Short fluctuations: ±5°C. Seasonal adjustments: ±10°C but &lt;30°C</td>
<td>Short fluctuations: ±10%. Seasonal adjustments: ±10%</td>
<td></td>
</tr>
<tr>
<td>Class C</td>
<td>&lt;30°C</td>
<td>25-75%</td>
<td>High risk of mechanical damage to high vulnerability artefacts; moderate risk to most paintings, photographs, and tiny risk to most artefacts and books.</td>
</tr>
<tr>
<td>IEQ-index=40</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Class D</td>
<td>≤75%</td>
<td></td>
<td>High risk of sudden or cumulative mechanical damage to most artefacts and paintings due to low humidity fracture, but high humidity delamination and deformations. Mold growth and rapid corrosion avoided.</td>
</tr>
<tr>
<td>IEQ-index=20</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The index for light and PM is evaluated based on the maximum allowed thresholds available in literature for general collections (not particularly sensible objects). The thresholds applied for light
are 200 lux for visible light illuminance and 75 µW/lm (=15 mW/m²) for UV [63]. For particulate matter the guideline applied is 10 µg/m³ of PM$_{2.5}$ [1]. The behavior of the IEQ-index for light and PM differs from the one of temperature and relative humidity due to the specific characteristics of these thresholds. Now the values equal or below the thresholds are considered acceptable (IEQ-index=100) and those above would imply IEQ-index=0; without any gradual transition between the two categories.

The general IEQ-index in each data-point is set by the lowest value of the parameter-specific indexes in that point. For example, a situation where it was measured a temperature of 20°C, with a short time fluctuation of ±2°C and seasonal adjustment of ±5°C, a relative humidity of 50%, short fluctuations of ±5% relative humidity, no seasonal adjustment, and UV of 16 mW/m² would translate in IEQ-index(temperature, relative humidity)=100, IEQ-index(UV)=0, and consequently IEQ-index=0.

In order to facilitate the visualization, the evolution of the IEQ-index through time is represented by a colour map with a fixed scale of colours. The scale varies from red to blue conforming the index varies from 0 to 100. The IEQ-index calculation and graphical representation was performed using MATLAB R2017a (The MathWorks, Inc., 2017).

3. RESULTS AND DISCUSSION

First, a traditional interpretation of the environmental data is presented. The thermo-hygrometric data and light values recorded in the storage are compared with the gallery. Subsequently, an extensive range of PM monitoring techniques are evaluated. Finally, in order to investigate the effect
of the inclusion of particulate matter in indoor environmental quality monitoring, the IEQ-index is calculated for traditional environmental parameters both including and not including PM.

3.1. Environmental parameters (temperature, relative humidity, Vis and UV light, CO₂). The collected data streams are visualized using graphs (Figure 2). The information in the graphs is enhanced by adding a coloured zone that denote the acceptable range as defined by ASHRAE guidelines [1] for temperature and relative humidity and by Thomson [63] for Vis and UV light intensity. ASHRAE classes take into account also short time fluctuations, in this case only the absolute limit values for temperature and relative humidity for classes As, A and B (from no risk to moderate risk only for high-vulnerability artefacts) are represented in the graph.

Figure 2a shows the temperature trend at the two selected locations. The temperature in the storage is very stable due to the controlled environment, with a minor daily difference of ±0.3 °C. The gallery experiences more extreme daily temperature differences of ±3.7 °C. Figure 2b shows the relative humidity trends in both environments. The relative humidity in the storage is more stable compared to the gallery. Moreover, the average relative humidity is 47%, close to the 50% average recommended by ASHRAE [1]. The relative humidity in the gallery on the other hand drops below 40% with an average of 30%, close to the lowest limit (25%) of ASHRAE class C (high risk for sensible objects and moderate to tiny risk for all other hygroscopic artefacts) [1]. The presence of daily fluctuations of ±8% also increases the risk of damage towards high vulnerability artefacts [1]. The stabilizing effect of the HVAC in the storage is clearly visible in the temperature and relative humidity gradient in contrast to the uncontrolled environment in the gallery.

Figure 2c and d show the light evolution of Vis and UV respectively. In the storage, visible light is originating from artificial lights with an intensity of 450 lux when switched on, zero when switched
This light source emits 14.8 mW/m$^2$ UV on average. The artificial light level, significantly higher than the maximum suggested threshold value of 200 lux, leads to the exposure of the collection to potentially endangering conditions on a daily basis. UV levels are extremely close to the threshold. Even though these values are not strictly exceeding the recommended limit in this case, it is anyway important to remember that it is advised to keep UV radiation as low as possible [63]. A further risk for the stored objects comes from the fact that lights in the storage remain switched on for long periods also during the night. This depends on security reasons connected to an increased terror threat after the 2016 Brussels bombings, unfortunately causing the unnecessary exposure of the objects to potentially dangerous levels of light. However, by simply changing the type of lamps installed, this danger can be easily averted. In the gallery high values for illuminance and UV are reached during day time, with peaks above the suggested thresholds on a daily basis. An increase in the maximum Vis light is observed after 3/05, from an average of 514 lux to an average of 1039 lux. Temperature also presents a growing trend in the same days, from an average maximum of 21 ºC (before 3/05) to an average maximum of 28 ºC (after 3/05). The higher temperatures observed when more light is entering the building from the windows underlines the central role played by direct sunlight on the indoor environmental quality.

To get an idea about human activity in the indoor environment, the CO$_2$ level was monitored (Figure 2e). In the storage, the CO$_2$ average level rises during weekdays but significantly decreases in the weekends (highlighted days in Figure 2). On the contrary, in the gallery the CO$_2$ level variation is more irregular and no clear distinctions are present between weekdays and weekends. This depends on the presence of visitors in the museum also in the weekends, but also on the variation of outdoor levels of the gas. As an example, on May 1$^{st}$ the level of CO$_2$ is extremely low compared to the
previous days in the gallery, while on May 2\textsuperscript{nd} an absolute maximum is observed. On both days the
museum was closed to visitors, but on May 1\textsuperscript{st} the outdoor level of CO\textsubscript{2} is around the average for the
period of analysis, while on May 2\textsuperscript{nd} one of the highest values for whole 2016 was registered in the
surroundings of the museum (data from IRCELINE, monitoring stations 41WOL1 and 41002) [64]. The rate at which the CO\textsubscript{2} level decreases when people are not present in the environment is linked
to the air exchange rate between indoor and outdoor [65]. The drop in concentration observed every
night in the gallery appears to be much faster than the slow decrease registered in the storage, leading
to the conclusion that the air exchange rate in the former is higher than in the latter. To our knowledge
a threshold for CO\textsubscript{2} levels in generic collections is still not defined, the concentrations observed in
both environments remain anyway well below the threshold for humans of 1000 ppm. [1]
Figure 2 (1.5-column fitting image). Physical parameters continuous monitoring in storage and gallery: a) temperature, b) relative humidity, c) Vis light illuminance and d) UV light intensity, e) CO$_2$ concentration. Grey areas represent the suggested range for general collections.
According to the guidelines for the traditionally monitored parameters, the storage is on average better suited for preserving general collections compared to the gallery, strongly influenced by the outdoor environment. In general the variations observed are mainly connected to day/night cycles, without allowing to clearly identify specific events.

3.2. **Average amounts of airborne particulate matter.** The deposition of particulate matter on cultural heritage materials has been proven to represent a concrete risk for their conservation [11-14]. PM transport and deposition mechanisms are strictly linked to concentration and aerodynamic dimension of the particles, practically deciding the fate of the suspended matter. [27,48] The threats associated with the presence of airborne PM in conservation environments therefore strongly depend on these factors [14].

Daily measurements of particulate matter over a period of two weeks were performed in storage and gallery via traditional Harvard impactors. Figure 3 shows the average concentrations of PM₁, PM₂.₅, and PM₁₀ for both locations. The average PM values in the storage are lower compared to the gallery. As it is clear from Figure 3, the PM concentration in the gallery reaches the outdoor levels of the corresponding PM fraction, confirming the close interaction with the outdoor environment. Also the average particle size distribution in the gallery matches the one outside, with similar percentages of PM₁₀-2.₅ (31%, 39%), PM₂.₅-₁ (36%, 31%) and PM₁ (33%, 30%) in the total PM₁₀ mass. The storage room is more protected from the outdoor environment, filtering particles and especially the coarse fraction. On average, in fact, PM₁₀-2.₅ represents only 7% of the total PM₁₀ mass in the storage, while PM₂.₅-₁ and PM₁ respectively represent 56% and 37%. The HVAC system installed in the storage is responsible both for the general lower PM content and for the different size distribution observed in this room. A very similar situation to the one registered in this HVAC controlled environment is
observed by Ligocki et al. [17] in three different museums where filtering HVAC systems are installed. Also in this case very similar concentrations of PM$_{2.5}$ and PM$_{10}$ are observed, showing the strong influence of air filtering systems in terms of coarse fraction reduction.

Figure 3 (1.5-column fitting image). Daily average of a) PM$_1$, b) PM$_{2.5}$ and c)PM$_{10}$ measurements in storage and gallery. The grey area represents the ASHRAE suggested maximum concentration for PM$_{2.5}$ in general collections. [1] N/A= data not available (flow rate outliers).

Indoor to outdoor ratios (I/O) of mass concentrations of PM$_1$, PM$_{2.5}$ and PM$_{10}$ were calculated for both locations (Figure 4). In the storage, the values observed are significantly lower than unity, except for some higher I/O values for fine PM at specific days. On the other hand, in the gallery the values
for I/O ratios are closer to unity, indicating a high natural ventilation rate. This behavior appears very similar to the one observed by Ligocki et al. [17], with close to unity I/O ratios in two historical buildings not equipped with PM filtration systems (Sepulveda House and Southwest Museum, Los Angeles) and significantly lower values in HVAC-equipped museums (J. Paul Getty Museum, Malibu; Norton Simon Museum, Pasadena; Huntington Library, San Marino). This difference between storage and gallery underlines the close contact between the latter and the outdoor environment, causing the variability in PM values to depend mainly on factors such as wind speed, wind direction and road traffic outside the museum. On the contrary, given the higher average level of protection from the outdoor environment observed in the storage, the presence of peaks can be attributed to indoor human activities. In detail, some extreme sudden variations from a quite constant trend are present on April 18, 19 and 22. Indoor, the temperature and relative humidity are mildly disturbed on those days and the CO₂ levels are slightly higher compared to the other days, evidence that suggests the presence of people in the storage. During this week the floor of the storage was treated with a sealant and a number of objects were moved. These activities might have caused the increase in fine PM, exposing the collection to a potential threat undetectable by monitoring only conventional parameters. The monitoring of PM therefore constitutes an important tool for the assessment of the real risk level the stored objects are exposed to.
3.3. Average chemical composition of particulate matter. The potential risk associated with airborne particulate strongly depends also on the chemical composition of the deposited material. [12-14, 27, 49-51] In particular, the presence of airborne soil dust and black carbon has been linked to the soiling of works of art. The deposition of these particles on the surface of museum objects can lead to the building up of dark deposits, which are difficult, expensive and often even impossible to remove safely. [12,14,27] Other types of inorganic particles, such as ammonium salts, sea salts and Fe-rich particles, can also pose a chemical hazard to specific materials. [13,49-51] In this section the reconstruction of the abundances of different particle types in different PM fractions is introduced.

First, the abundances of soil dust, sea salts, ammonium salts and mineral salts (soluble fraction of soil dust) in the different locations are calculated. The daily average results of the analysis of black carbon are also included in the chemical reconstruction of PM mass. Since in general both primary
and secondary soot particles fall into the submicron size fraction [66-67], all the observed black carbon is considered to be part of PM$_1$. Black carbon was not analyzed in the outdoor environment, therefore IRCELINE values (monitoring station 41R001) [68] are used in order to allow a comparison. Considering the difference in position between museum and monitoring station these values are only an approximation of the real values.

The total inorganic and black carbon mass obtained from these results represents only a limited part of the total PM mass collected. This is due to the presence of organic dust and aerosol-associated water [17,28]. In the storage, in particular, the unexplained mass accounts for over 90% of the total mass on average, with higher values in the fine fraction than in the coarse (Table 2). The low air exchange rate and the HVAC filtration system account for the extremely high content of organic aerosols in this environment. [17] On the other hand, in the gallery the percentage of unknown mass is lower, with a total average close to the one observed for the outdoor environment. Also in this case the values are higher for the fine fraction than for the coarse fraction. When compared to the outdoor results, the amount of unknown mass in the gallery appears slightly higher in all fractions. This suggests the presence of indoor sources of organic dust. The chemical composition of this fraction in museum environments has been linked before to the shedding of particles by visitors and to maintenance operations, such as vacuum cleaning and floor waxing [17]. In order to have a complete vision of the indoor environmental quality in the museum, the nature of this organic fraction has to be further investigated both in the storage and in the gallery.

Table 2. Percentage of unexplained mass in different PM fractions in storage, gallery and outdoor.
<table>
<thead>
<tr>
<th></th>
<th>PM$_1$</th>
<th>PM$_{2.5}$</th>
<th>PM$_{10-2.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Storage</td>
<td>96.7±0.9%</td>
<td>99.8±0.1%</td>
<td>93.5±4.5%</td>
</tr>
<tr>
<td>Gallery</td>
<td>71±7%</td>
<td>81±9%</td>
<td>55±20%</td>
</tr>
<tr>
<td>Outdoor</td>
<td>50±11%</td>
<td>68±8%</td>
<td>44±14%</td>
</tr>
</tbody>
</table>

The results for the chemical reconstruction of the inorganic fraction of PM$_{10-2.5}$, PM$_{2.5-1}$ and PM$_1$ are presented in Figure 5-7. Soil dust and all other species considered are present in much lower concentrations in the storage than in the gallery, reflecting the trend observed for the total PM amount. The relatively high concentrations of inorganic PM and black carbon in the gallery compared to the storage confirms the important influence of the outdoor environment on the IEQ for this room. This close relation between the two environments is supported by the distribution of the single species in the different fractions. In particular, the fine fraction is mainly composed of ammonium salts and black carbon, with small amounts of sea salts in the PM$_{2.5-1}$ fraction in both environments. In the same way, the coarse fraction contains soil dust, sea salts and mineral salts mainly. Soil dust is often carried indoor from the outside through shoes, in particular when outdoor relative humidity is high, and resuspended by human activities [28,69]. A significant difference is observed only for the sea salts concentration, much lower in the gallery than outdoor. Black carbon concentration is also lower indoor, but the different location of museum and IRCELINE monitoring station could have influenced these results.

On the other hand, in the storage the very small portion of mass identified consists mainly of black carbon, ammonium salts and soil dust in the fine fraction and almost exclusively of soil dust in the coarse fraction. The relatively high content of soil dust is connected to the presence of people in the...
environment, while the other inorganic particles concentrations are efficiently reduced by the HVAC system installed [17]. The level of soil dust appears to be relatively high compared to the other species also in the fine fraction. The absolute concentration in PM$_1$ and PM$_{2.5-1}$ remains anyway low compared to the one observed for the coarse fraction, where soil dust is usually more abundant [66]. The chemical composition of PM observed in the storage confirms the similarity between this environment and the HVAC-equipped museums studied by Ligocki et al. [17] In both cases in fact extremely high percentages of organic matter and an inorganic fraction dominated by soil dust were observed.

Figure 5. (2-column fitting image, colour) Daily composition of the airborne inorganic fraction of PM$_1$. N/A= data not available (flow rate outliers).
Figure 6. (2-column fitting image, colour) Daily composition of the airborne inorganic fraction of PM$_{2.5}$. N/A = data not available (flow rate outliers).
In order to further explore the origin of the inorganic aerosols observed inside the museum, Indoor/Outdoor (I/O) PM ratios are calculated. In Figure 8-9, the average daily indoor to outdoor ratios of soil dust, sea salts, ammonium salts and mineral salts in PM$_1$, PM$_{2.5}$ and PM$_{10}$ are presented. As expected, in the well-insulated and well-controlled storage the ratios are significantly lower than 1 on average. This value is exceeded only in precise events and only for certain species in the fine fraction. In particular an ammonium concentration slightly higher than the outdoor level on 18/04 (I/O=1.2) is observed, together with an important soil dust enrichment on 18 and 19/04 (I/O=13.5 and 12.8 respectively). This peak of material corresponds to the days in which a new floor sealant was

**Figure 7. (2-column fitting image, colour)** Daily composition of the airborne inorganic fraction of PM$_{10-2.5}$. N/A = data not available (flow rate outliers).
applied in the storage, probably causing the resuspension of important amounts of particulate matter.
The increase registered in the soil dust ratios is more drastic than the one for ammonium salts on these
days. The very low concentration of soil dust in outdoor PM$_1$ accounts for the relative difference
observed for the two species. It is not clear if the event was caused directly by the sealing intervention
or if the fine particulate suspension was caused by the movement of the stored objects. What is certain
is that this perturbation of the preexistent equilibrium condition represents a potential risk for the
stored objects, emphasizing the importance of including PM in the monitoring of indoor
environmental quality.

In the gallery the situation appears different, with ratios generally closer to 1 for all the fractions.
In particular, the ratios for the fine fraction tend to be higher than the ratios for PM$_{10}$. This evidence
is connected to the fact that PM$_1$ usually infiltrates more efficiently than the other fractions through
the building shell. [27] In detail, soil dust presents ratios on average slightly lower than 1, but
significantly higher than 1 on 26/04 for PM$_{10}$ and PM$_{2.5}$ and on 7 and 8/05 for PM$_1$. Sea salts seem to
enter less efficiently or to be less resuspended in the exhibition environment than soil dust, presenting
lower I/O ratios especially for PM$_{10}$. The introduction of soil dust through the shoes of visitors
accounts for this difference in behavior [28]. A single higher ratio can be noticed on 7/05 only for
PM$_1$. Ammonium salts present a similar behavior to the one observed for soil dust. The ratios in the
gallery are in general smaller than 1 while one single enrichment is present on 26/04 for PM$_1$. Mineral
salts are characterized by generally higher and extremely variable ratios; on average lower than 1 for
PM$_1$ and PM$_{10}$ and slightly higher for PM$_{2.5}$. Particularly high mineral salt I/O ratios in the gallery
are present on 26/04 for PM$_{2.5}$ and PM$_{10}$ and on 7/05 for PM$_1$. Generally speaking this situation
confirms the dominant outdoor influence on the environmental quality of the gallery, with variations
associated only to single discrete events of which the causes are difficult to identify. Knowing the type and dimension of the inorganic particles suspended in the gallery during these events can be an important tool for defining the best conservation strategies for the objects.

**Figure 8. (1.5-column fitting image)** Indoor/outdoor (I/O) ratios for a) soil dust and b) sea salts in PM$_{10}$, PM$_{2.5}$ and PM$_{1}$. N/A=data not available (flow rate outliers).
Figure 9. (1.5-column fitting image) Indoor/outdoor (I/O) ratios for c) ammonium salts and d) mineral salts in PM$_{10}$, PM$_{2.5}$ and PM$_{1}$. N/A=data not available (flow rate outliers).

Indoor/Outdoor ratios were calculated also for black carbon (Figure 10). The observed values remain approximately stable in both locations throughout the whole sampling period, with an average
ratio of 0.04±0.01 in the storage and 0.31±0.08 in the gallery. The low average ratios confirm the absence of indoor sources. As a consequence, the closer contact of the gallery with the outside accounts for the 10 times higher ratios registered in this environment compared to the storage. Since elemental carbon enters the atmosphere mainly through incomplete combustion of fossil fuels and biomass [67], the black carbon observed inside the museum is mostly produced by outdoor traffic. From this point of view it is interesting to notice how the levels of elemental carbon in the storage are relatively low, even though this environment is indirectly connected to a highway tunnel. The efficiency of the storage insulation in preventing the contamination even from submicron sized outdoor particles is therefore confirmed. The role played by the filtration system in this case is probably secondary to the one played by the underground location of the storage. The black carbon I/O ratios appear in fact from 2 to 10 times lower than the ones observed in HVAC-equipped museums by Ligocki et al., a difference that cannot be explained by an higher filtration efficiency given the state-of-the art quality of the systems installed in these museums and the lower efficiency of filters in removing submicron particles. [17]
3.4. Continuous monitoring of PM$_{2.5}$. The indoor concentration of PM$_{2.5}$ in terms of number of particles per unit volume was continuously determined by means of Lighthouse Handheld 3016-IAQ particle counter. Continuous measurements allow to obtain much more detailed information about short-time PM variations than daily average measurements. This makes it easier to identify sudden risks for museum collections as well as to reconstruct the sources of PM enrichments. An example is given in Figure 11, where the continuous data registered with the particle counter are compared to their daily average. Due to a malfunctioning during the measurements the data from 21/04 23:15 to 26/04 10:15 are missing.
Figure 11. (2-column fitting image) Comparison between continuous monitoring and daily averages of PM$_{2.5}$ with Lighthouse Handheld 3016-IAQ particle counter. N/A=data not available

Before being able to implement the IEQ-index with continuous PM data, it is necessary to convert the PM$_{2.5}$ threshold value for general collections ($\mu$g/m$^3$) [1] into an average number of particles/m$^3$. For this reason, the daily average of Lighthouse results is compared with the mass data from Harvard impactors. Different results were obtained for the two different environments, with higher correlation in the storage ($r=0.84$) than in the gallery ($r=0.57$). In Figure 12 the centered daily average concentrations (Average=0, Standard deviation=1) for storage and gallery are presented. Because of the different correlation obtained, the standardization of the data was performed separately for the two sets. The better correlation observed for the storage is clear from the figure. This behavior is likely caused by the uncontrolled environmental conditions observed in the latter location, closer to...
an outdoor environment than to an indoor one. It means that defining an IEQ index in this specific
environment simply does not make sense. The response of light scattering based particle counters is
a function not only of particle size, but also of particle refractive index and shape. In an outdoor-like
environment particulate matter can present widely differing morphologies and chemical
compositions, potentially causing changes in both optical properties and density [70]. These factors
lead to the poor correlation between particle counts and mass measurements observed in the gallery.
For this reason, the calculation of a PM threshold expressed in counts/m$^3$ in this environment is not
feasible without introducing a substantial uncertainty. This makes it impossible to implement the
IEQ-index with continuous PM data in a simple and accurate way. On the contrary, in the storage the
good insulation from the outdoor environment and the HVAC system installed grant a significant
stability both from a physical and chemical point of view. These conditions produce the good
correlation observed in this environment, allowing to convert the PM$_{2.5}$ maximum threshold for
general collections (10 µg/m$^3$) [1] into a site-specific counts/m$^3$ PM limit value. In this specific case
this mass threshold was chosen, but any threshold value can be selected according to the specific
needs of the collection considered.
Figure 12 (2-column fitting image). Comparison between the standardized daily concentrations of PM$_{2.5}$ obtained by Lighthouse particle counter and Harvard impactor. The data from the two locations are standardized separately.

The result of the threshold conversion for the storage is $901043 \pm 171674$ counts/m$^3$. Given the significant standard error of regression to prevent the underestimation of possible risks for the collection the greatest lower bound of the range of calculated values is considered. In other words, the difference between the average counts/m$^3$ calculated threshold and the standard error of regression is determined. The resulting value of 729369 counts/m$^3$ is then used as final PM$_{2.5}$ threshold. By applying this approximated limit value to the continuous PM$_{2.5}$ data collected with Lighthouse, it is possible to include PM concentrations in the calculation of the IEQ-index. In this way, a continuous estimate of the risk associated to indoor PM levels can be taken into account when monitoring the environmental quality in an indoor location. The use of continuous PM data for the calculation of the IEQ allows to identify short time events and potentially dangerous situations for the collection, deepening the understanding of the conservation conditions. By using only the daily averages these events may remain hidden, in particular in a relatively clean environment such as the storage.

Further studies will be necessary to better understand the factors influencing the correlation between particle counters response and mass measurements, and consequently the range of applicability of this methodology. The subsequent step will be to try to simplify and reduce the cost of this approach by substituting the expensive Lighthouse device with a low-cost laser particle counter. Different devices suggested for home or office applications are commercially available, but
their effective precision and accuracy are still a matter of debate [45-46] and will have to be preventively tested.

3.5. IEQ-index. To visualize the evolution in time of the indoor environmental quality, the IEQ-index method [52] is applied to the continuous data of the traditional environmental parameters and PM. In Figure 13 the environmental data are presented in terms of IEQ analysis with colour coding. This intuitive representation allows a fast evaluation of the indoor environmental quality compared to the selected threshold values. The first two bars in Figure 13a represent general IEQ indexes for the collection. In order to underline the effect of including PM analysis in the study of indoor environmental quality, these general indexes are calculated both including and not including particulate matter. In these bars, a specific time instant corresponds to the lowest IEQ value in that instant for the parameter-specific IEQ components. Therefore, red areas indicate that one or more parameters are potentially posing a threat to the objects. The separate bars per parameter (Figure 13b,c,d,e) allow to identify which parameter should be improved. Since the implementation of the index with continuous PM data was not possible in the gallery, an average IEQ-index is calculated by including the daily measurement of PM mass with Harvard impactors in this location. The inclusion of average values is not ideal and leads to a loss of short-term information, but it is sufficient to clearly visualize the strong influence of the inclusion of PM on indoor environmental quality evaluation.

Generally speaking, the IEQ analysis highlights the presence of different conservation conditions in the two environments. In the gallery, in particular, the important outdoor influence causes the IEQ to assume very low values for all the considered parameters. This situation exposes the whole collection to a high risk of deterioration and damage. A better insulation of the building and the
installation of a HVAC system capable of controlling temperature, relative humidity and PM infiltration rate would significantly improve the conservation conditions. Unfortunately, the dimension of the gallery makes this type of intervention expensive and complicated. The risk associated with temperature and illumination can be anyway partially contained by limiting the access of direct sunlight (e.g. by covering with reflecting materials the southern facing side of the roof).

On the other hand, in the storage low IEQ values are limited to light intensity and single PM events. Light sensitive objects should therefore not be stored in this room without accurately protecting them by covering and/or without preventively changing the light source to a lower intensity LED light.

The inclusion of PM$_{2.5}$ in the general IEQ index leads to different results for storage and gallery. In the storage, in fact, the inclusion of PM data does not cause visible changes. However, when considering the single IEQ bar for PM, the presence of potentially dangerous peaks of particulate can be observed on the days in which a floor sealant was applied in the room. Therefore, this data representation allows the museum personnel to easily recognize potential threats and to understand their possible causes. The detailed analysis of PM supporting the continuous monitoring an IEQ-index calculation allows to estimate more accurately the real risk on the basis of the type of particles observed. In this case a specific analysis of the organic fraction of PM will be also necessary. The information obtained should anyway lead to the development of safety measures to prevent high PM levels when maintenance operations are performed or to preventively protect the objects if this is not possible.

On the contrary, in the gallery the inclusion of discrete PM$_{2.5}$ data in the general IEQ index leads to a significant change in the results. The collection appears in fact to be under a constant threat when also PM is considered, even in periods in which only short events are taking place according to
traditional environmental parameters. The introduction of this additional information underlines the importance of including PM data in IEQ studies. Without considering this parameter the collection would be considered safe, even in moments when PM levels could potentially endanger it. Nevertheless, the impossibility of implementing continuous PM measurements in the IEQ for this location strongly limits the amount of information obtainable with this method. The development of a methodology that can be applied even on the monitoring of uncontrolled environments is therefore of capital importance for the future.

**Figure 13 (2-column fitting image, colour).** Representation of the IEQ-index [52] in storage (11/04/2016-21/04/2016) and gallery (27/04/2016-7/05/2016) using colour codes (red=higher risk for the collection, blue=lower risk): a) general indexes calculated combining all the considered parameters, PM included and excluded; b) temperature (T) and relative humidity (RH) combined, c)
Vis light illuminance, d) intensity of UV radiation, e) PM$_{2.5}$ Lighthouse data (storage) and Harvard impactors data (gallery).

4. CONCLUSIONS

The present study showed how particulate matter is present in conservation environments in concentrations that can significantly and suddenly vary with time. The presence of sudden events could not be identified by temperature, relative humidity and light monitoring, suggesting the presence of many more risks for museum collections that remain invisible when only traditional parameters are considered. For this reason it is important to include a continuous monitoring of PM concentration in the evaluation of indoor environmental quality. The methodology discussed in this work allows to implement PM concentrations in the IEQ-index calculation, easily recognizing potentially dangerous conditions for the collections. After recognizing in which periods the collection was at risk, one can further analyse the graphs and understand which parameters need to be improved. When it comes to PM, the chemical characterization of the particulate helps to deeper understand the real risk for the objects and the reasons behind sudden increases in concentration.

The implementation of PM in the IEQ calculation was not possible in uncontrolled environments, therefore the proposed method still needs to be perfected. However, reliable results were obtained when stable environmental conditions and a good insulation from the outdoor environment were granted (ideal conditions for a conservation environment).

All things considered, these results show why and how IEQ calculation methods should be discussed and implemented, not only in terms of PM levels. This, in fact, is not the only potentially
endangering parameter excluded from traditional continuous IEQ monitoring. Also the inclusion of continuous data for gaseous pollutants, now hindered by the sensitivity of commercially available sensors, could allow to identify further threats to indoor cultural heritage.

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REFERENCES


(38) Sulfur Dioxide Meter Model Z-1300 technical datasheet on Environmental Sensor Co. Website:


(39) Hydrogen Sulfide Meter Model Z-900 technical datasheet on Environmental Sensor Co. Website:


(40) Formaldehyde Meter Model Z-300 technical datasheet on Environmental Sensor Co. Website:


(41) Acetic Acid Gas Sensor Cell specifications on PureAire monitoring systems, Inc. website:


(42) Portable and Fixed Monitor Gas Sensor Specifications on Aeroqual™ website:


Kauneliene, V.; Wierzbicka, A.; Martuzevicius, D. Characterization of indoor aerosol temporal
variations for the real-time management of indoor air quality, Atmos. Environ. 2015, 118, 107-117.

Harrison, D.; Lin, C.; Wu, H. Personal exposure monitoring of PM\(_{2.5}\) in indoor and outdoor

Manikonda, A.; Zíková, N.; Hopke, P. K.; Ferro, A. R. Laboratory assessment of low-cost PM

War Heritage Institute website: www.warheritage.be

Riley, W. J.; McKone, T. E.; Lai, A. C. K.; Nazaroff, W. W. Indoor Particulate Matter of
36 (2), 200–207.

Anaf, W., Janssens, K., De Wael, K. Formation of metallic mercury during photodegradation/
photodarkening of α-HgS: electrochemical evidence. Angewandte Chemie International Edition
2013, 52(48), 12568-12571.

Anaf, W., Trashin, S., Schalm, O., Van Dorp, D., Janssens, K., De Wael, K. Electrochemical

Li, S. and Hihara, L. H. In situ Raman Spectroscopic Study of NaCl Particle-Induced Marine


(55) DataTaker DT85 specifications on DataTaker website: http://www.datataker.com/DT85.php

(56) GMW90 carbon dioxide, temperature and humidity sensor specifications on Vaisala website: http://www.vaisala.com/en/products/carbondioxide/Pages/GMW90.aspx


(62) Specifications for MS&T Area Sampler Harvard-type impactor on Air Diagnostics and Engineering Inc. website: https://www.airdiagnostics.com/indoor_samp_equip.html


(64) IRCELINE: carbon dioxide levels (ppm) from 11/04/2016 to 9/05/2017, monitoring stations 41WOL1 and 41002; http://viewer.irceline.be/?ts=6636__irceline,6612__irceline&span=2016-04-11T00:00:00+02:00/2016-05-09T00:00:02:00.


(68) IRCELINE: black carbon concentration (µg/m³) from 11/04/2016 to 8/05/2017, monitoring stations 41001; http://viewer.irceline.be/?ts=6569__irceline&span=2016-04-11T01:29:42+02:00/2016-05-08T02:24:42+02:00.