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

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 O_3, NO_2, SO_2, H_2S, 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

60 or office applications exist [44-46]. However, these systems are not yet employed to their full
61 potential in cultural heritage.

62 In this article, a system is presented that enables the continuous monitoring of temperature (ºC),
63 relative humidity (%), illuminance of visible light (lux), UV light intensity (mW/m^2) and PM
64 concentration (number of particles/m^3) in a simultaneous and synchronous way. Extended measuring
65 campaigns were performed at two locations in the former Royal Museum of the Armed Forces and
66 of Military History, or shorter Royal Military Museum, in Brussels, Belgium. The Royal Military
67 Museum is integrated in the War Heritage Institute (WHI) since May 1st 2017. [47] Since the risk
68 associated with the presence of airborne PM in conservation environments strongly depends on the
69 concentration and aerodynamic dimension of the particles [27, 48], but also on their chemical
70 composition [13-14, 49-51], the continuous monitoring is supplemented by an in depth chemical
71 analysis of PM. The total mass concentration and chemical composition of fine (PM_1, $PM_{2.5}$) and
72 coarse particles (PM_{10}) is analyzed. This qualitative and quantitative analysis of PM allows to register
73 sudden changes of the indoor environmental quality which are invisible with physical parameters.
74 The available guidelines for conservation environments are then used to calculate an IEQ-index [52]
75 by combining the information of temperature, relative humidity and light exposure with that of PM.
76 No univocal guideline has been found in literature prescribing an optimal PM concentration for
77 conservation environments expressed in number of particles per unit volume. A mass per unit volume
78 threshold is therefore considered and converted into number of particles/m^3 values on the basis of
79 site-specific empirical correlations. Taking into account the extreme variety of objects exposed in the
80 museum, the ASHRAE maximum limit for general collection of 10 $\mu g/m^3$ for $PM_{2.5}$ is applied [1].

81 This article discusses in detail how and why continuous PM data should be included in indoor
82 environmental quality studies. The versatile methodology discussed can be applied to the monitoring
83 of environmental quality in any type of indoor conservation environment. When the main concern is
84 directed towards objects or materials with more specific needs, potentially dangerous conditions can
85 be promptly recognized by simply changing the threshold values applied in the calculation of the
86 IEQ-index.

87

88 **2. MATERIALS AND METHODS**

89 **2.1. Sampling locations and campaigns.** The Royal Military Museum is located in the Parc du
90 Cinquantenaire area in the city center of Brussels in Belgium (50°50'29.4"N 4°23'31.6"E). The
91 building dates from the end of the 19th century. The collections of the Royal Military Museum, now
92 integral part of the War Heritage Institute (WHI), illustrate ten centuries of military history ranging
93 from medieval times up until this day. The more than 125,000 objects include amongst others
94 uniforms and headgear, edged weapons and firearms but also paintings and sculptures, medals and
95 music instruments. The museum building even houses airplanes, armored vehicles and artillery. The
96 collections consist of an extremely wide range of materials. The museum occupies five large
97 exhibition galleries covering approximately 40,000 m^2.

98 First, measurements were performed in one location from April 11 to April 24 in 2016, followed
99 by a period in a second place from April 25 to May 9 in 2016. The first location is the central storage
100 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, CO_2). 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 (CO_2) 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 (PM_1, $PM_{2.5}$, PM_{10}) were sampled with 3 different Harvard-type impactors (MS&T area sampler, Air Diagnostics and Engineering Inc., USA). [62] $PM_{2.5}$ and PM_{10} were collected on mixed cellulose ester filters (SKC MCE 0.45 µm 225-1914, SKC Limited, UK), PM_1 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 $PM_{2.5}$ and PM_{10} and with the flow rate of 23 L/min for PM_1. 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].

182 **2.4. Data evaluation.**

183 *2.4.1. Univariate data analysis.* Eventual anomalies and sudden perturbations of single parameters
184 are thoroughly investigated in order to identify and interpret situations of potential risk for the
185 museum objects. To deepen the understanding on the causes of anomalous behaviors the data obtained
186 in the different indoor locations are assessed against the outdoor values for the corresponding period.

187 *2.4.2. IEQ-index calculation.*

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

194 The ASHRAE standards propose a set of five control classes depending on the potential risk for a
195 collection under specific thermo-hygrometric conditions (Table 1). These classes vary from situations
196 with the lowest possible risk for most objects (class AA) to a considerably high risk of damage for
197 most of the artefacts (class D). The specific IEQ-index for temperature and relative humidity is
198 defined by associating each of the aforementioned classes to a numerical value in each data point.
199 The index range from 100 to 0, as it covers the situations from the lowest to the highest risk of
200 degradation or damage. Consequently, class AA corresponds to the maximum IEQ-index=100, A to
201 IEQ-index=80, B to IEQ-index=60, C to IEQ-index=40, and D to IEQ-index=20. Below class D
202 (relative humidity larger than 75%) the IEQ-index will reach the minimum value of 0.

203

11

204 **Table 1:** Overview of the ASHRAE standards for temperature and relive humidity [1] and their

205 correspondence to the IEQ-index.

ASHRAE Class		Temperature	Relative Humidity	Remarks
Class AA IEQ-index=100		15-25°C Short fluctuations: ±2°C. Seasonal adjustments: ±5°C	50% Short fluctuations: ± 5%. No seasonal adjustments.	No risk of mechanical damage to most artefacts and paintings.
Class A IEQ-index=80	As	15-25°C Short fluctuations: ±2°C. Seasonal adjustments: +5°C -10°C	40-60% Short fluctuations: ± 5%. Seasonal adjustments: ±10%	Small risk of mechanical damage to high vulnerability artefacts; no mechanical risk to most artefacts, paintings, photographs, and books.
	A		Short fluctuations: ± 10%. Seasonal adjustments: No variation.	
Class B IEQ-index=60		15-25°C Short fluctuations: ±5°C. Seasonal adjustments: ±10°C but <30°C	40-60% Short fluctuations: ± 10%. Seasonal adjustments: ±10%	Moderate risk of mechanical damage to high-vulnerability artefacts; tiny risk to most paintings and photographs and no risk to most artefacts and books.
Class C IEQ-index=40		<30°C	25-75%	High risk of mechanical damage to high vulnerability artefacts; moderate risk to most paintings, photographs, and tiny risk to most artefacts and books.
Class D IEQ-index=20			≤75%	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.

206

207 The index for light and PM is evaluated based on the maximum allowed thresholds available in

208 literature for general collections (not particularly sensible objects). The thresholds applied for light

209 are 200 lux for visible light illuminance and 75 μW/lm (=15 mW/m^2) for UV [63]. For particulate
210 matter the guideline applied is 10 μg/m^3 of $PM_{2.5}$ [1]. The behavior of the IEQ-index for light and
211 PM differs from the one of temperature and relative humidity due to the specific characteristics of
212 these thresholds. Now the values equal or below the thresholds are considered acceptable (IEQ-
213 index=100) and those above would imply IEQ-index=0; without any gradual transition between the
214 two categories.

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

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

225 **3. RESULTS AND DISCUSSION**

226 First, a traditional interpretation of the environmental data is presented. The thermo-hygrometric
227 data and light values recorded in the storage are compared with the gallery. Subsequently, an
228 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, CO_2). 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.

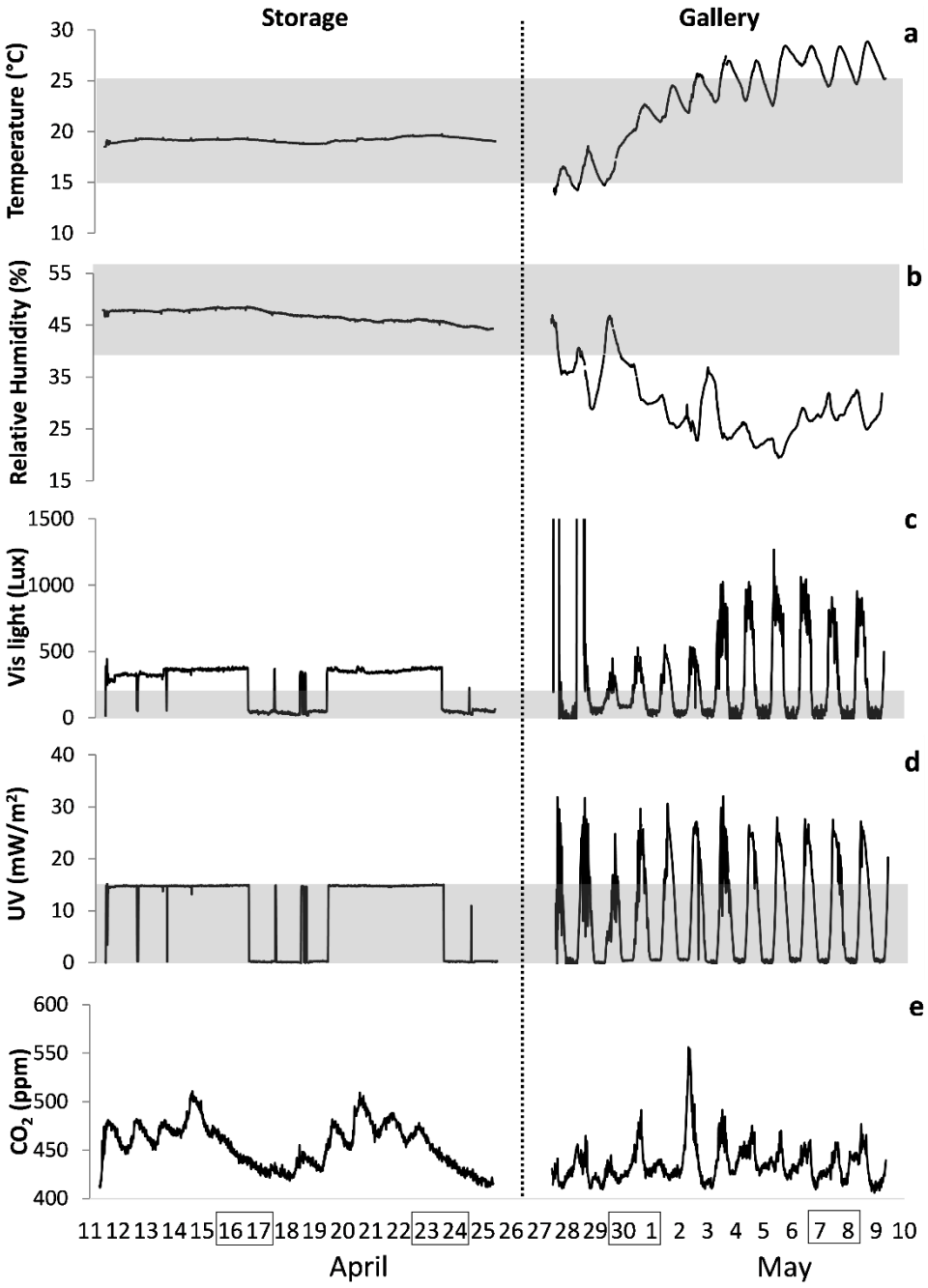
Figure 2a shows the temperature trend at the two selected locations. The temperature in the storage is very stable due to the controlled environment, with a minor daily difference of ±0.3 °C. The gallery experiences more extreme daily temperature differences of ±3.7 °C. Figure 2b shows the relative humidity 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

off. 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 1st the level of CO_2 is extremely low compared to the

previous days in the gallery, while on May 2nd an absolute maximum is observed. On both days the museum was closed to visitors, but on May 1st the outdoor level of CO_2 is around the average for the period of analysis, while on May 2nd 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_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_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]



283

284 **Figure 2 (1.5-column fitting image).** Physical parameters continuous monitoring in storage and
285 gallery: a) temperature, b) relative humidity, c) Vis light illuminance and d) UV light intensity, e)
286 CO_2 concentration. Grey areas represent the suggested range for general collections.

287 According to the guidelines for the traditionally monitored parameters, the storage is on average
288 better suited for preserving general collections compared to the gallery, strongly influenced by the
289 outdoor environment. In general the variations observed are mainly connected to day/night cycles,
290 without allowing to clearly identify specific events.

291 **3.2. Average amounts of airborne particulate matter.** The deposition of particulate matter on
292 cultural heritage materials has been proven to represent a concrete risk for their conservation [11-14].
293 PM transport and deposition mechanisms are strictly linked to concentration and aerodynamic
294 dimension of the particles, practically deciding the fate of the suspended matter. [27,48] The threats
295 associated with the presence of airborne PM in conservation environments therefore strongly depend
296 on these factors [14].

297 Daily measurements of particulate matter over a period of two weeks were performed in storage
298 and gallery via traditional Harvard impactors. Figure 3 shows the average concentrations of PM_1,
299 $PM_{2.5}$, and PM_{10} for both locations. The average PM values in the storage are lower compared to the
300 gallery. As it is clear from Figure 3, the PM concentration in the gallery reaches the outdoor levels of
301 the corresponding PM fraction, confirming the close interaction with the outdoor environment. Also
302 the average particle size distribution in the gallery matches the one outside, with similar percentages
303 of $PM_{10-2.5}$ (31%, 39%), $PM_{2.5-1}$ (36%, 31%) and PM_1 (33%, 30%) in the total PM_{10} mass. The storage
304 room is more protected from the outdoor environment, filtering particles and especially the coarse
305 fraction. On average, in fact, $PM_{10-2.5}$ represents only 7% of the total PM_{10} mass in the storage, while
306 $PM_{2.5-1}$ and PM_1 respectively represent 56% and 37%. The HVAC system installed in the storage is
307 responsible both for the general lower PM content and for the different size distribution observed in
308 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.

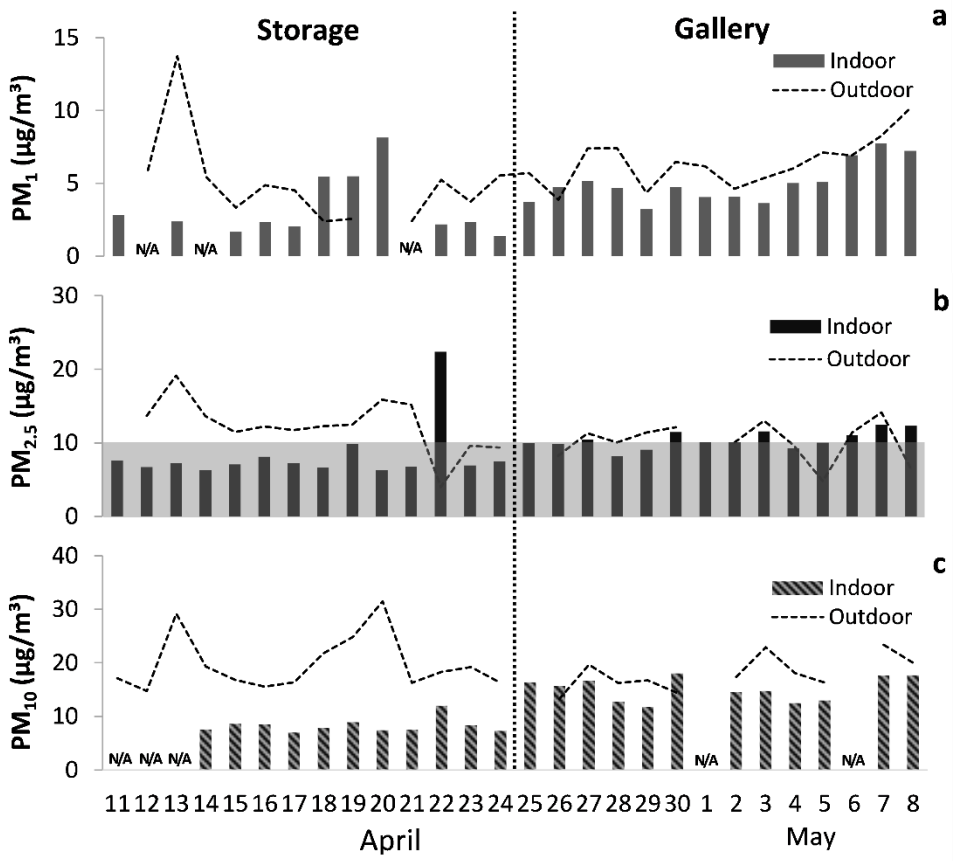


Figure 3 (1.5-column fitting image). Daily average of a) PM_1, b) $PM_{2.5}$ and c)PM_{10} measurements in storage and gallery. The grey area represents the ASHRAE suggested maximum concentration for $PM_{2.5}$ in general collections. [1] N/A= data not available (flow rate outliers).

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 CO_2 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.

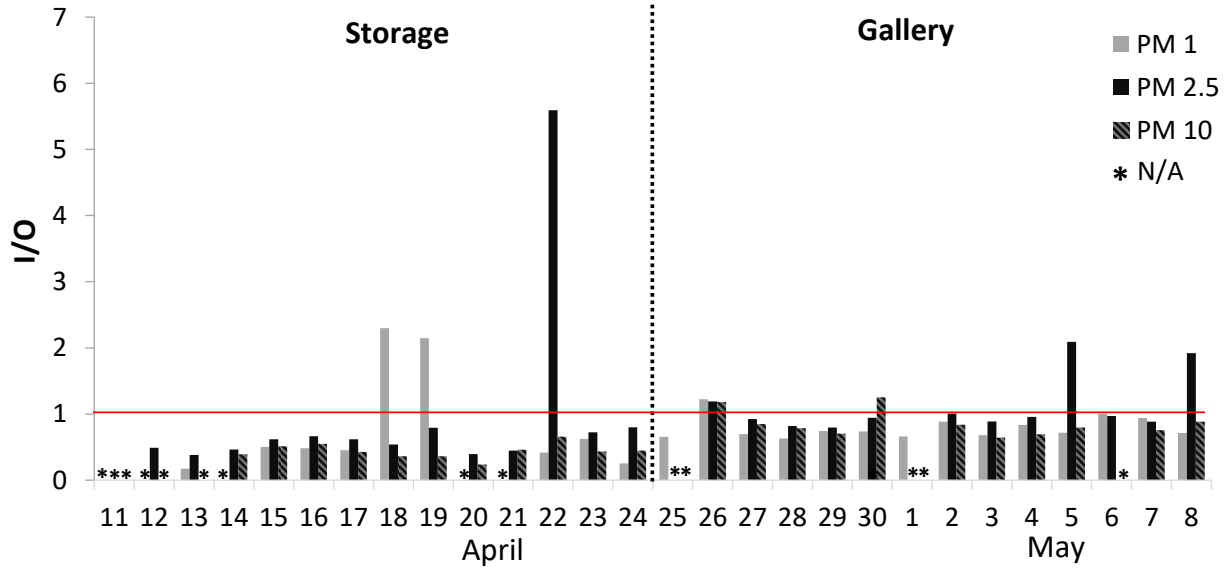


Figure 4 (2-column fitting image). Indoor/outdoor (I/O) ratios for PM_1, $PM_{2.5}$ and PM_{10} concentration in storage and gallery. N/A=data not available (flow rate outliers).

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.

	PM_1	$PM_{2.5-1}$	$PM_{10-2.5}$
Storage	96.7±0.9%	99.8±0.1%	93.5±4.5%
Gallery	71±7%	81±9%	55±20%
Outdoor	50±11%	68±8%	44±14%

371

372 The results for the chemical reconstruction of the inorganic fraction of $PM_{10-2.5}$, $PM_{2.5-1}$ and PM_1
373 are presented in Figure 5-7. Soil dust and all other species considered are present in much lower
374 concentrations in the storage than in the gallery, reflecting the trend observed for the total PM amount.
375 The relatively high concentrations of inorganic PM and black carbon in the gallery compared to the
376 storage confirms the important influence of the outdoor environment on the IEQ for this room. This
377 close relation between the two environments is supported by the distribution of the single species in
378 the different fractions. In particular, the fine fraction is mainly composed of ammonium salts and
379 black carbon, with small amounts of sea salts in the $PM_{2.5-1}$ fraction in both environments. In the same
380 way, the coarse fraction contains soil dust, sea salts and mineral salts mainly. Soil dust is often carried
381 indoor from the outside through shoes, in particular when outdoor relative humidity is high, and
382 resuspended by human activities [28,69]. A significant difference is observed only for the sea salts
383 concentration, much lower in the gallery than outdoor. Black carbon concentration is also lower
384 indoor, but the different location of museum and IRCELINE monitoring station could have influenced
385 these results.

386 On the other hand, in the storage the very small portion of mass identified consists mainly of black
387 carbon, ammonium salts and soil dust in the fine fraction and almost exclusively of soil dust in the
388 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.

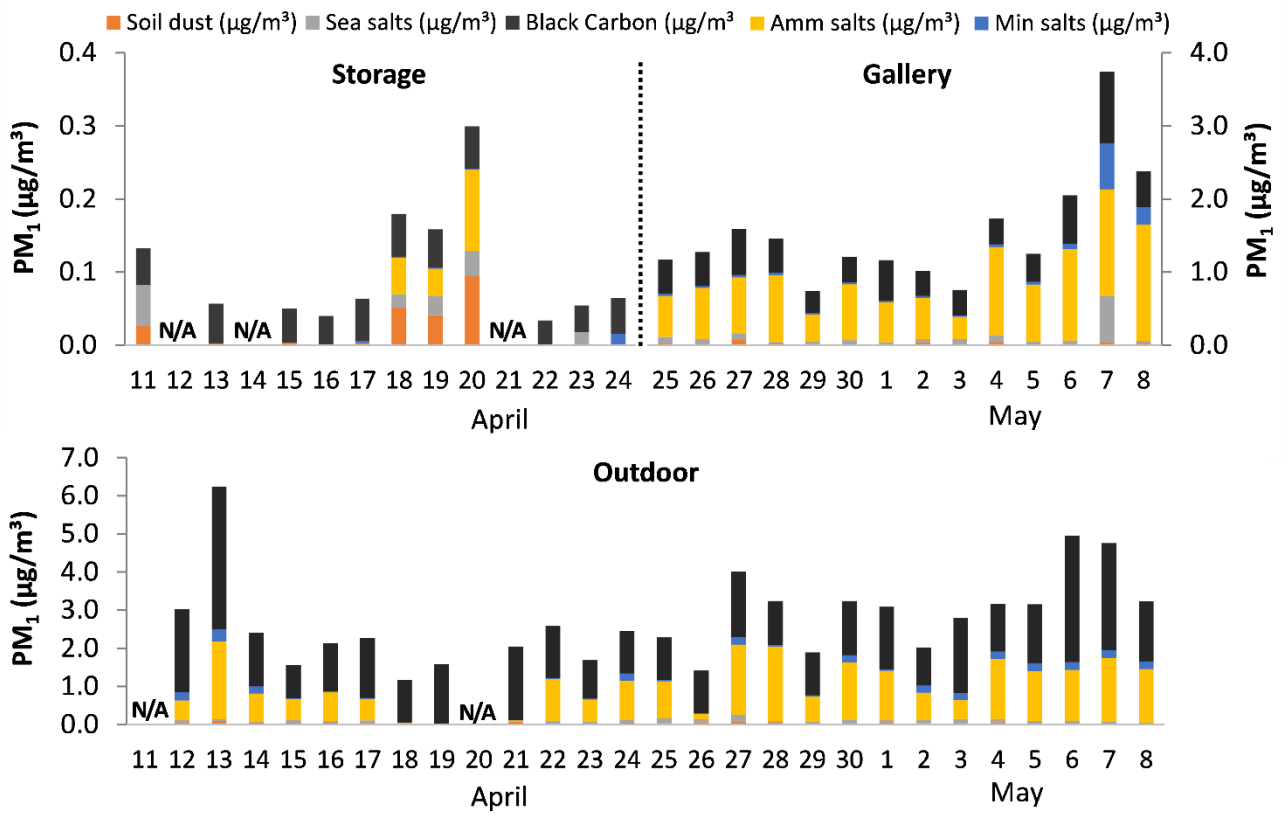
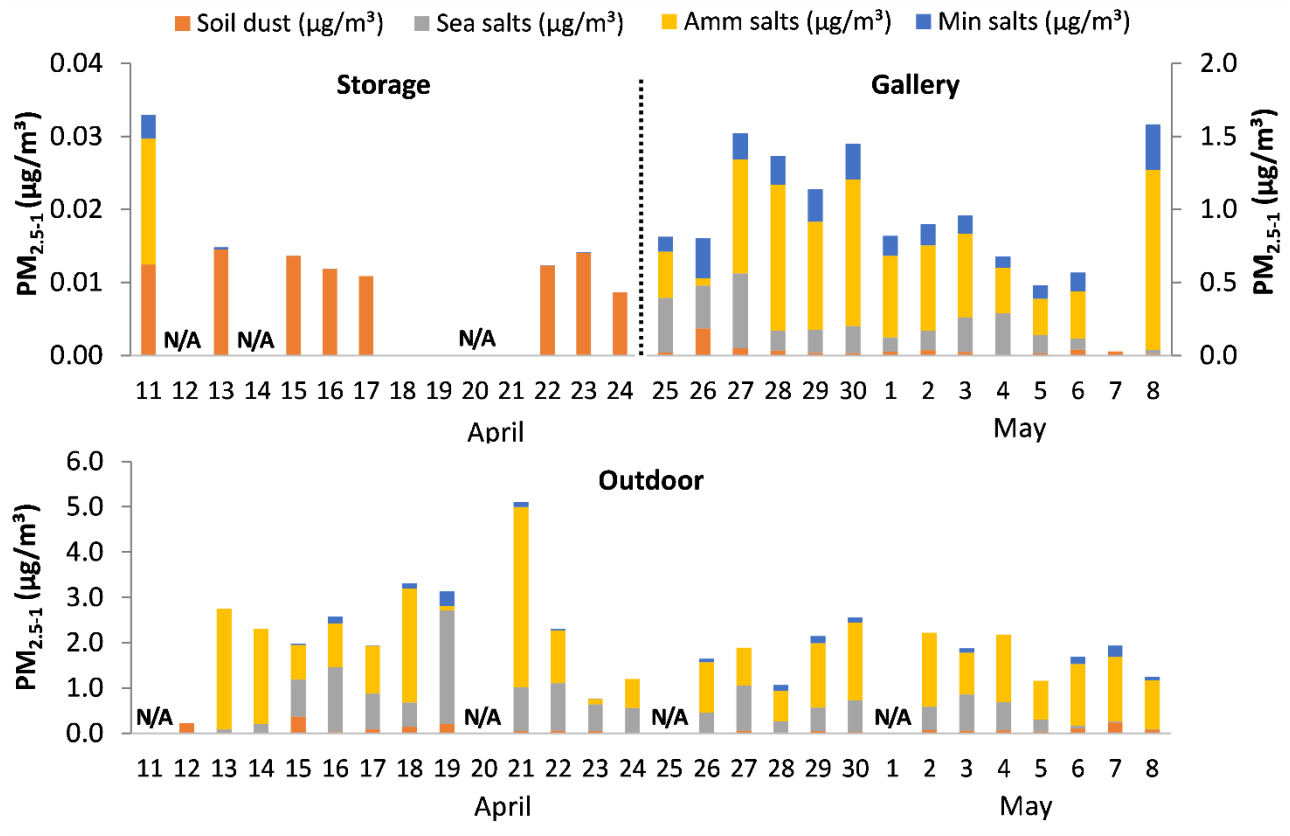


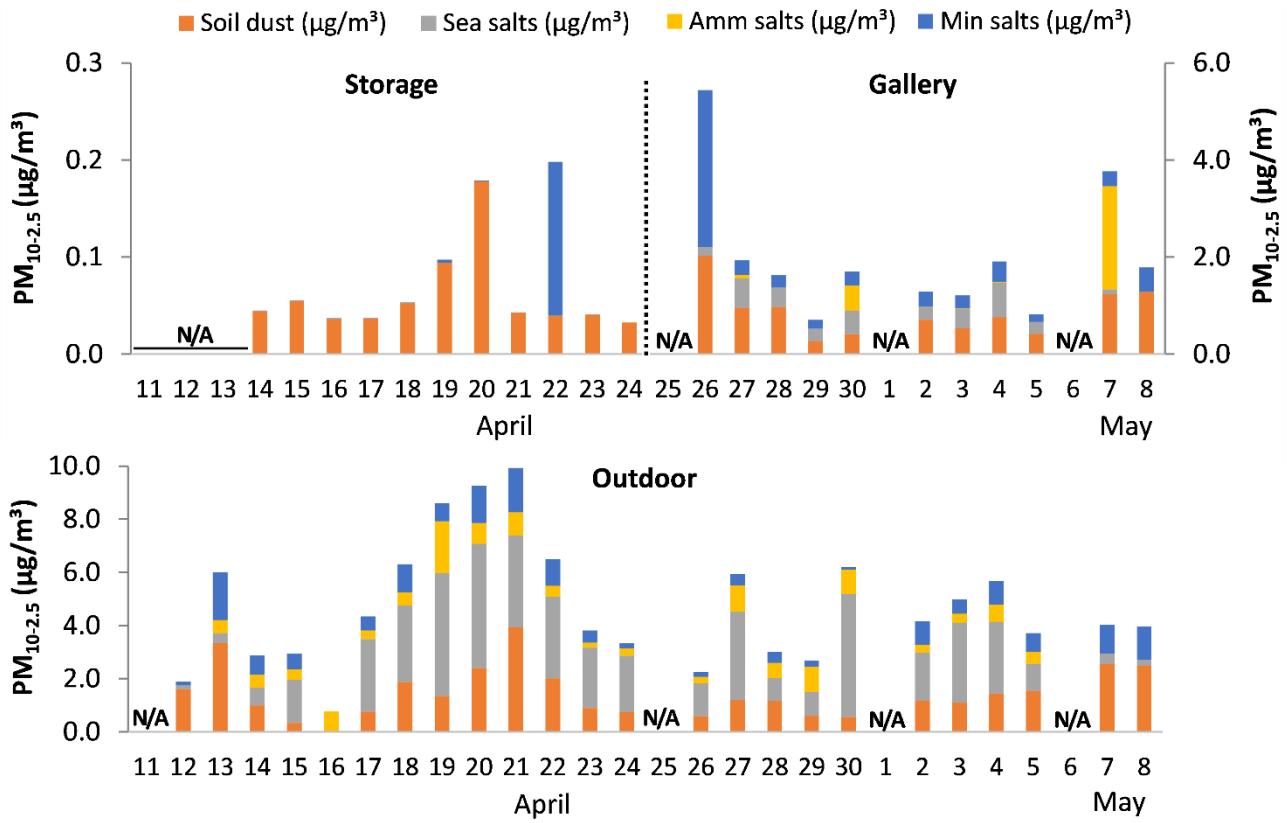
Figure 5. (2-column fitting image, colour) Daily composition of the airborne inorganic fraction of PM_1. N/A= data not available (flow rate outliers).



400

401 **Figure 6. (2-column fitting image, colour)** Daily composition of the airborne inorganic fraction of

402 $PM_{2.5-1}$. N/A= data not available (flow rate outliers).



403

404 **Figure 7. (2-column fitting image, colour)** Daily composition of the airborne inorganic fraction of

405 $PM_{10-2.5}$. N/A= data not available (flow rate outliers).

406 In order to further explore the origin of the inorganic aerosols observed inside the museum,

407 Indoor/Outdoor (I/O) PM ratios are calculated. In Figure 8-9, the average daily indoor to outdoor

408 ratios of soil dust, sea salts, ammonium salts and mineral salts in PM_1, $PM_{2.5}$ and PM_{10} are presented.

409 As expected, in the well-insulated and well-controlled storage the ratios are significantly lower than

410 1 on average. This value is exceeded only in precise events and only for certain species in the fine

411 fraction. In particular an ammonium concentration slightly higher than the outdoor level on 18/04

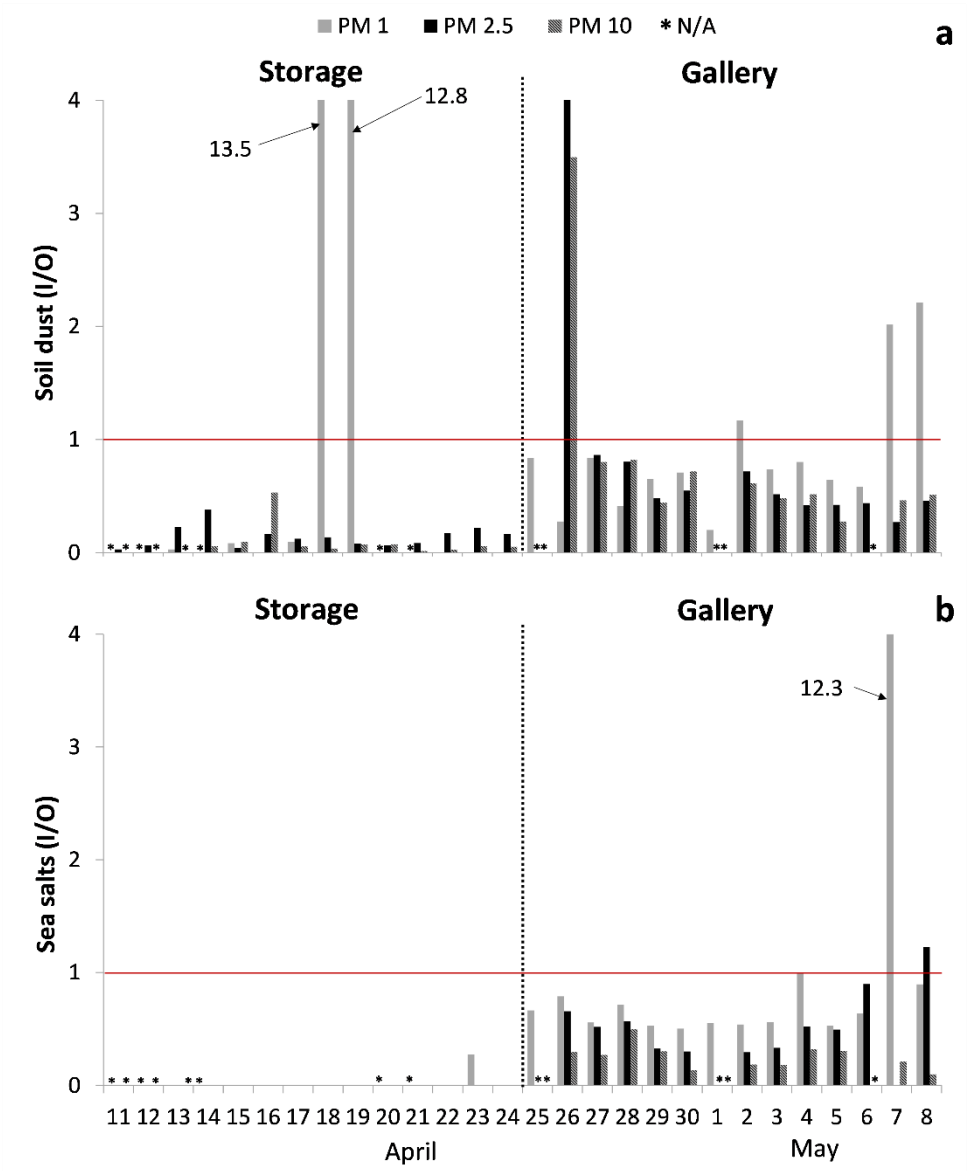
412 (I/O=1.2) is observed, together with an important soil dust enrichment on 18 and 19/04 (I/O=13.5 and

413 12.8 respectively). This peak of material corresponds to the days in which a new floor sealant was

414 applied in the storage, probably causing the resuspension of important amounts of particulate matter.
415 The increase registered in the soil dust ratios is more drastic than the one for ammonium salts on these
416 days. The very low concentration of soil dust in outdoor PM_1 accounts for the relative difference
417 observed for the two species. It is not clear if the event was caused directly by the sealing intervention
418 or if the fine particulate suspension was caused by the movement of the stored objects. What is certain
419 is that this perturbation of the preexistent equilibrium condition represents a potential risk for the
420 stored objects, emphasizing the importance of including PM in the monitoring of indoor
421 environmental quality.

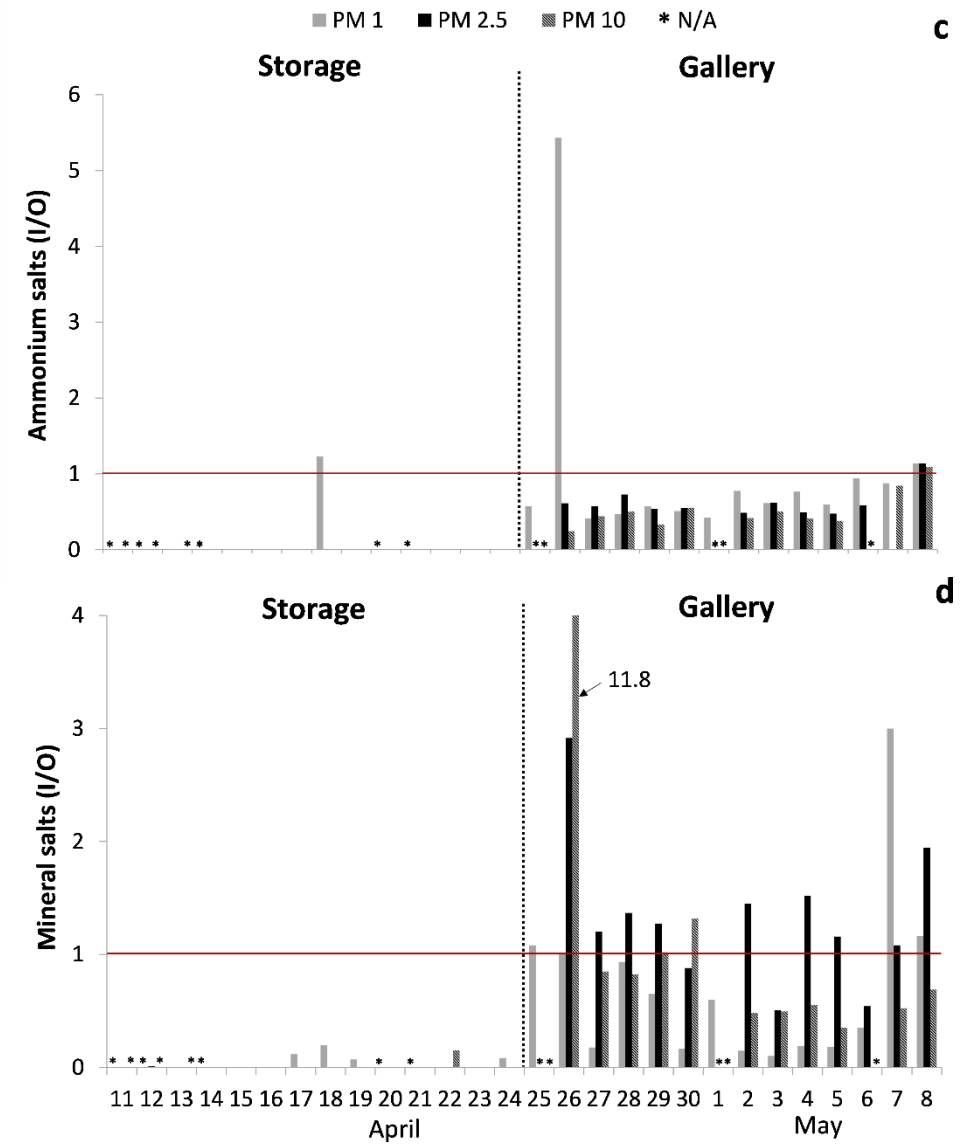
422 In the gallery the situation appears different, with ratios generally closer to 1 for all the fractions.
423 In particular, the ratios for the fine fraction tend to be higher than the ratios for PM_{10}. This evidence
424 is connected to the fact that PM_1 usually infiltrates more efficiently than the other fractions through
425 the building shell. [27] In detail, soil dust presents ratios on average slightly lower than 1, but
426 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
427 enter less efficiently or to be less resuspended in the exhibition environment than soil dust, presenting
428 lower I/O ratios especially for PM_{10}. The introduction of soil dust through the shoes of visitors
429 accounts for this difference in behavior [28]. A single higher ratio can be noticed on 7/05 only for
430 PM_1. Ammonium salts present a similar behavior to the one observed for soil dust. The ratios in the
431 gallery are in general smaller than 1 while one single enrichment is present on 26/04 for PM_1. Mineral
432 salts are characterized by generally higher and extremely variable ratios; on average lower than 1 for
433 PM_1 and PM_{10} and slightly higher for $PM_{2.5}$. Particularly high mineral salt I/O ratios in the gallery
434 are present on 26/04 for $PM_{2.5}$ and PM_{10} and on 7/05 for PM_1. Generally speaking this situation
435 confirms the dominant outdoor influence on the environmental quality of the gallery, with variations

436 associated only to single discrete events of which the causes are difficult to identify. Knowing the
437 type and dimension of the inorganic particles suspended in the gallery during these events can be an
438 important tool for defining the best conservation strategies for the objects.



439

440 **Figure 8. (1.5-column fitting image)** Indoor/outdoor (I/O) ratios for a) soil dust and b) sea salts in
441 PM_{10}, $PM_{2.5}$ and PM_1. N/A=data not available (flow rate outliers).



442

443 **Figure 9. (1.5-column fitting image)** Indoor/outdoor (I/O) ratios for c) ammonium salts and d)

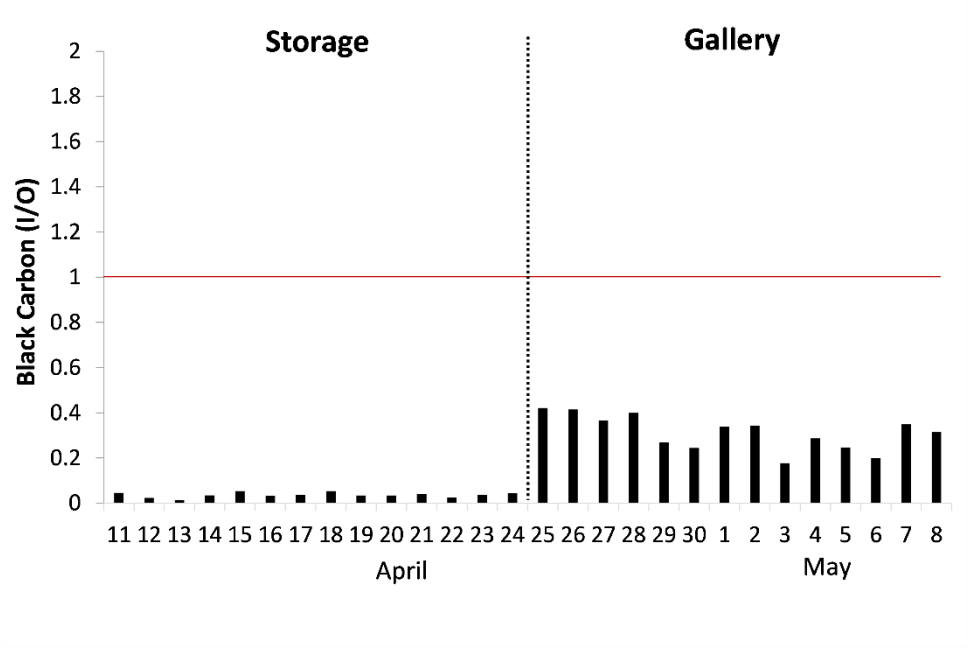
444 mineral salts in PM_{10}, $PM_{2.5}$ and PM_1. N/A=data not available (flow rate outliers).

445

446 Indoor/Outdoor ratios were calculated also for black carbon (Figure 10). The observed values

447 remain approximately stable in both locations throughout the whole sampling period, with an average

448 ratio of 0.04±0.01 in the storage and 0.31±0.08 in the gallery. The low average ratios confirm the
449 absence of indoor sources. As a consequence, the closer contact of the gallery with the outside
450 accounts for the 10 times higher ratios registered in this environment compared to the storage. Since
451 elemental carbon enters the atmosphere mainly through incomplete combustion of fossil fuels and
452 biomass [67], the black carbon observed inside the museum is mostly produced by outdoor traffic.
453 From this point of view it is interesting to notice how the levels of elemental carbon in the storage
454 are relatively low, even though this environment is indirectly connected to a highway tunnel. The
455 efficiency of the storage insulation in preventing the contamination even from submicron sized
456 outdoor particles is therefore confirmed. The role played by the filtration system in this case is
457 probably secondary to the one played by the underground location of the storage. The black carbon
458 I/O ratios appear in fact from 2 to 10 times lower than the ones observed in HVAC-equipped museums
459 by Ligocki et al., a difference that cannot be explained by an higher filtration efficiency given the
460 state-of-the art quality of the systems installed in these museums and the lower efficiency of filters in
461 removing submicron particles. [17]

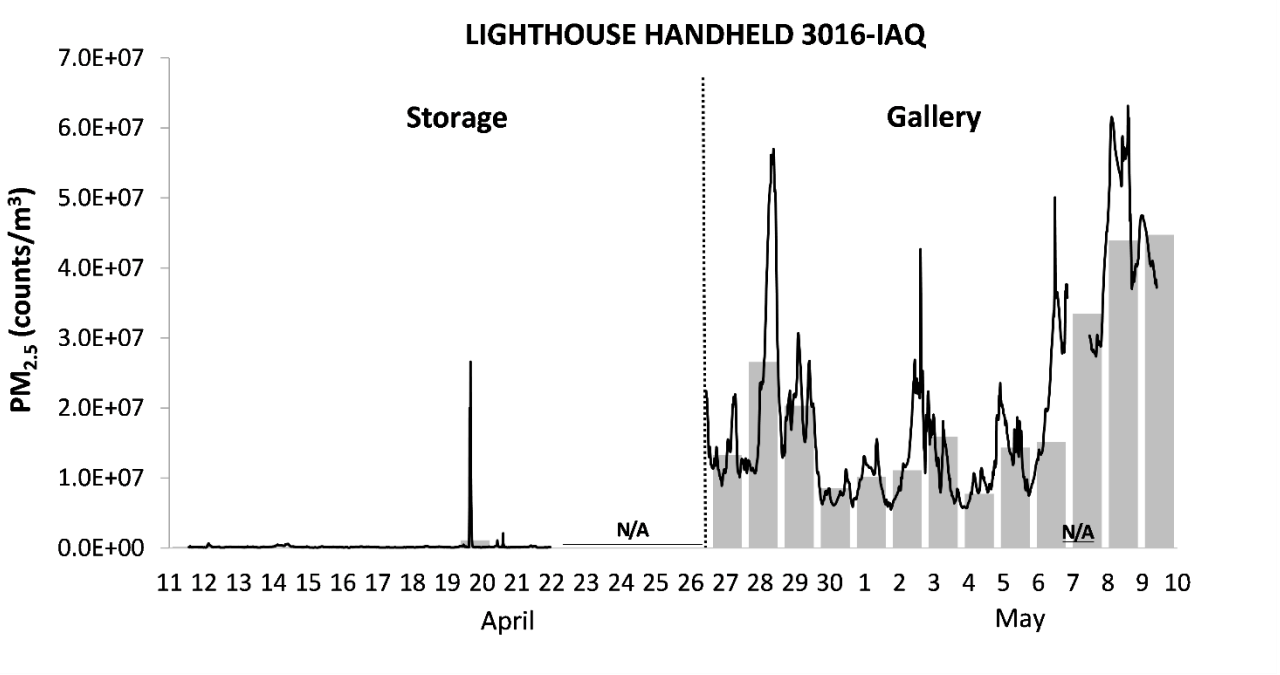


462

463 **Figure 10. (1.5-column fitting image)** Indoor/outdoor (I/O) daily average ratios for black carbon
464 in storage and gallery.

465

466 **3.4. Continuous monitoring of $PM_{2.5}$.** The indoor concentration of $PM_{2.5}$ in terms of number of
467 particles per unit volume was continuously determined by means of Lighthouse Handheld 3016-IAQ
468 particle counter. Continuous measurements allow to obtain much more detailed information about
469 short-time PM variations than daily average measurements. This makes it easier to identify sudden
470 risks for museum collections as well as to reconstruct the sources of PM enrichments. An example is
471 given in Figure 11, where the continuous data registered with the particle counter are compared to
472 their daily average. Due to a malfunctioning during the measurements the data from 21/04 23:15 to
473 26/04 10:15 are missing.



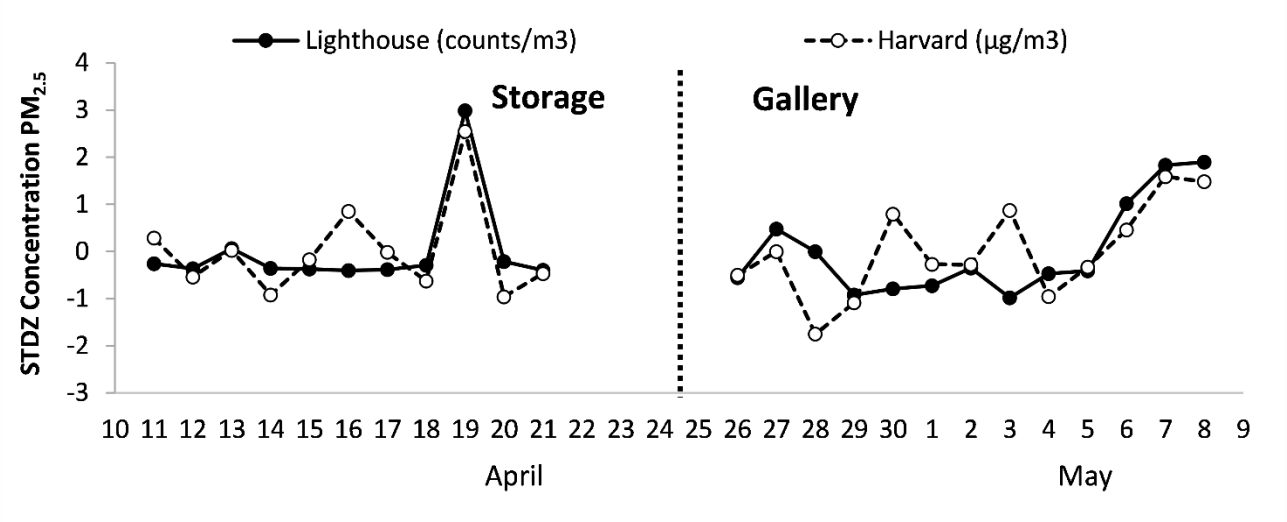
474

475 **Figure 11. (2-column fitting image)** Comparison between continuous monitoring and daily
476 averages of $PM_{2.5}$ with Lighthouse Handheld 3016-IAQ particle counter. N/A=data not available

477

478 Before being able to implement the IEQ-index with continuous PM data, it is necessary to convert
479 the $PM_{2.5}$ threshold value for general collections ($\mu g/m^3$) [1] into an average number of particles/m^3.
480 For this reason, the daily average of Lighthouse results is compared with the mass data from Harvard
481 impactors. Different results were obtained for the two different environments, with higher correlation
482 in the storage (r=0.84) than in the gallery (r=0.57). In Figure 12 the centered daily average
483 concentrations (Average=0, Standard deviation=1) for storage and gallery are presented. Because of
484 the different correlation obtained, the standardization of the data was performed separately for the
485 two sets. The better correlation observed for the storage is clear from the figure. This behavior is
486 likely caused by the uncontrolled environmental conditions observed in the latter location, closer to

487 an outdoor environment than to an indoor one. It means that defining an IEQ index in this specific
488 environment simply does not make sense. The response of light scattering based particle counters is
489 a function not only of particle size, but also of particle refractive index and shape. In an outdoor-like
490 environment particulate matter can present widely differing morphologies and chemical
491 compositions, potentially causing changes in both optical properties and density [70]. These factors
492 lead to the poor correlation between particle counts and mass measurements observed in the gallery.
493 For this reason, the calculation of a PM threshold expressed in counts/m^3 in this environment is not
494 feasible without introducing a substantial uncertainty. This makes it impossible to implement the
495 IEQ-index with continuous PM data in a simple and accurate way. On the contrary, in the storage the
496 good insulation from the outdoor environment and the HVAC system installed grant a significant
497 stability both from a physical and chemical point of view. These conditions produce the good
498 correlation observed in this environment, allowing to convert the $PM_{2.5}$ maximum threshold for
499 general collections (10 µg/m^3) [1] into a site-specific counts/m^3 PM limit value. In this specific case
500 this mass threshold was chosen, but any threshold value can be selected according to the specific
501 needs of the collection considered.



502

503 **Figure 12 (2-column fitting image).** Comparison between the standardized daily concentrations of
504 $PM_{2.5}$ obtained by Lighthouse particle counter and Harvard impactor. The data from the two locations
505 are standardized separately.

506

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

519 Further studies will be necessary to better understand the factors influencing the correlation
520 between particle counters response and mass measurements, and consequently the range of
521 applicability of this methodology. The subsequent step will be to try to simplify and reduce the cost
522 of this approach by substituting the expensive Lighthouse device with a low-cost laser particle
523 counter. Different devices suggested for home or office applications are commercially available, but

524 their effective precision and accuracy are still a matter of debate [45-46] and will have to be
525 preventively tested.

526 **3.5. IEQ-index.** To visualize the evolution in time of the indoor environmental quality, the IEQ-index
527 method [52] is applied to the continuous data of the traditional environmental parameters and PM. In
528 Figure 13 the environmental data are presented in terms of IEQ analysis with colour coding. This
529 intuitive representation allows a fast evaluation of the indoor environmental quality compared to the
530 selected threshold values. The first two bars in Figure 13a represent general IEQ indexes for the
531 collection. In order to underline the effect of including PM analysis in the study of indoor
532 environmental quality, these general indexes are calculated both including and not including
533 particulate matter. In these bars, a specific time instant corresponds to the lowest IEQ value in that
534 instant for the parameter-specific IEQ components. Therefore, red areas indicate that one or more
535 parameters are potentially posing a threat to the objects. The separate bars per parameter (Figure
536 13b,c,d,e) allow to identify which parameter should be improved. Since the implementation of the
537 index with continuous PM data was not possible in the gallery, an average IEQ-index is calculated
538 by including the daily measurement of PM mass with Harvard impactors in this location. The
539 inclusion of average values is not ideal and leads to a loss of short-term information, but it is sufficient
540 to clearly visualize the strong influence of the inclusion of PM on indoor environmental quality
541 evaluation.

542 Generally speaking, the IEQ analysis highlights the presence of different conservation conditions
543 in the two environments. In the gallery, in particular, the important outdoor influence causes the IEQ
544 to assume very low values for all the considered parameters. This situation exposes the whole
545 collection to a high risk of deterioration and damage. A better insulation of the building and the

546 installation of a HVAC system capable of controlling temperature, relative humidity and PM
547 infiltration rate would significantly improve the conservation conditions. Unfortunately, the
548 dimension of the gallery makes this type of intervention expensive and complicated. The risk
549 associated with temperature and illumination can be anyway partially contained by limiting the access
550 of direct sunlight (e.g. by covering with reflecting materials the southern facing side of the roof).

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

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

565 On the contrary, in the gallery the inclusion of discrete $PM_{2.5}$ data in the general IEQ index leads
566 to a significant change in the results. The collection appears in fact to be under a constant threat when
567 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.

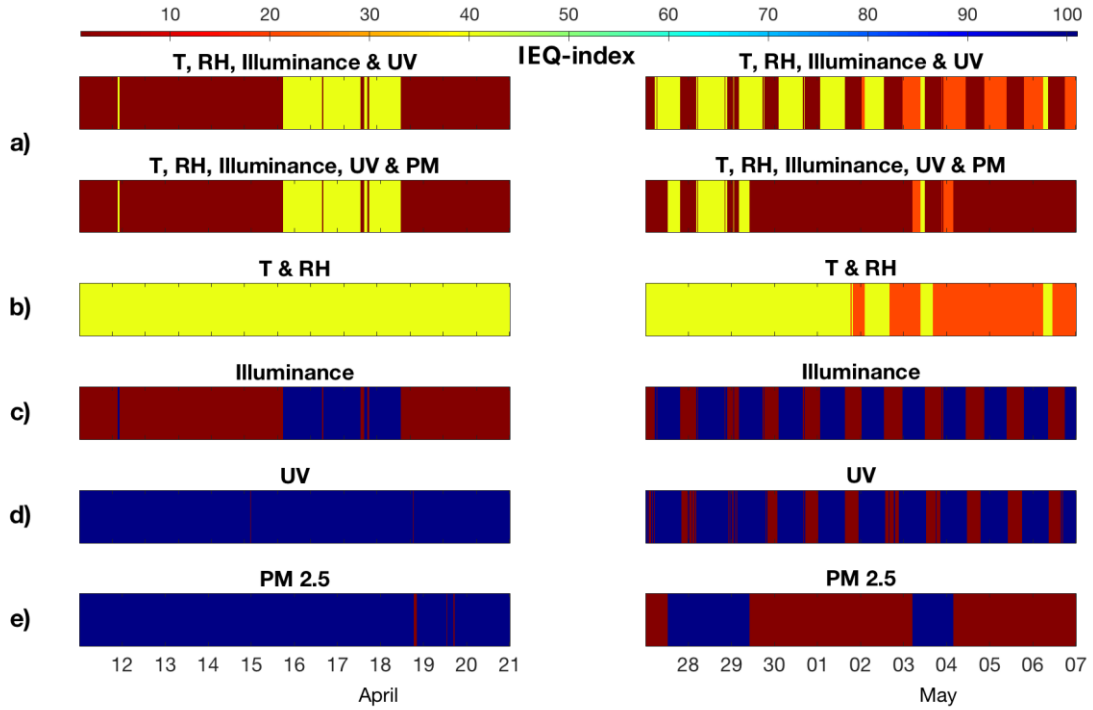


Figure 13 (2-column fitting image, colour). Representation of the IEQ-index [52] in storage (11/04/2016-21/04/2016) and gallery (27/04/2016-7/05/2016) using colour codes (red=higher risk for the collection, blue=lower risk): a) general indexes calculated combining all the considered parameters, PM included and excluded; b) temperature (T) and relative humidity (RH) combined, c)

580 Vis light illuminance, d) intensity of UV radiation, e) $PM_{2.5}$ Lighthouse data (storage) and Harvard
581 impactors data (gallery).

582

583 **4. CONCLUSIONS**

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

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

599 All things considered, these results show why and how IEQ calculation methods should be
600 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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