ELSEVIER

Contents lists available at ScienceDirect

Building and Environment

journal homepage: www.elsevier.com/locate/buildenv





Climate-induced risk for the preservation of paper collections: Comparative study among three historic libraries in Italy

Elena Verticchio ^{a,*}, Francesca Frasca ^b, Chiara Bertolin ^c, Anna Maria Siani ^b

- ^a Department of Earth Sciences, Sapienza University of Rome, Rome, Italy
- ^b Department of Physics, Sapienza University of Rome, Rome, Italy
- ^c Norwegian University of Science and Technology, Trondheim, Norway

ARTICLE INFO

Keywords: Library collection Microclimate investigation Preventive conservation Historical building Paper deterioration Preservation risk assessment

ABSTRACT

The conservation of historic libraries can be referred towards both the ancient book collections and the buildings themselves. Heritage collections made of paper are threaten by climate-induced deterioration risks such as cellulose hydrolysis. Several studies have investigated the microclimate inside historic libraries but comparisons are difficult due to the lack of long-term microclimate observations and uniformity in the use of standards and risk assessment methods. For the first time, the long-term microclimate observations collected in three historic libraries in Italy were comparatively studied to outline differences and similarities of their microclimates in terms of paper preservation. A multidisciplinary approach was applied to assess the building performance (a) and the deterioration risks for the collections (b). As for a), a common feature of the libraries was the high thermal inertia and low indoor-outdoor air exchanges. As for b), the Time Weighted Expected Lifetime (TWEL) was defined to account for an average chemical risk on a seasonal and yearly basis. TWEL allowed to highlight the impact of the most adverse conditions on the overall chemical risk for acidic paper preservation (e.g., temperatures above 20 °C reached naturally in summer/artificially in winter). It resulted that the measured microclimate conditions in the libraries would lead to the loss of their acidic collections in less than 300 years. Demographic plots were finally used to inform about the risk resulting from the synergy between handling and microclimate as well as to explore the effectiveness of possible preservation measures such as the deacidification of 10% of the collections.

1. Introduction

Libraries are unvaluable witnesses of the human knowledge over the centuries [1]. The term library can refer either to a collection of books and other sources of recorded information or to the building where such a collection is conserved. In this sense, the conservation of historic libraries may be referred towards both the ancient collection of books and the buildings themselves (if with cultural value). The importance of public libraries has been acknowledged since classical antiquity (e.g., Pliny wrote on Julius Caesar: *ingenia hominum rem publicam fecit*, "He made men's talents a public possession"). During the Middle Ages and the Renaissance, libraries were assembled in monasteries and universities, that preserved them over many centuries. After the French Revolution, the abolition of monastic orders led to the expropriation and dispersion of several libraries [2]. Some of them were transformed into private collections, which afterwards became the core of today's national libraries. Nowadays, digital libraries have broadened the access to

library contents from any place [3], so users do not need any more to visit the library building nor to physically interact with the materials in order to consult their contents. Although the reduced handling undoubtedly favours book conservation [4], the preservation of historic libraries and their buildings still demands to be carefully managed. In fact, their materiality is an irreplaceable testimony of our past for the present and future generations.

Sections 1.1, 1.2 and 1.3 of the Introduction present the state-of-theart on the preservation of library collections, focusing on the damage risk assessment for paper objects, the international policy framework and the previous microclimate monitoring studies in historic libraries worldwide. Section 1.4 outlines the research aims of this work.

1.1. Damage risk assessment for paper collections

Library collections are made of a wide range of organic and inorganic materials, where paper is the most represented. All library materials undergo a natural and unavoidable ageing process, however the rate of

E-mail address: elena.verticchio@uniroma1.it (E. Verticchio).

https://doi.org/10.1016/j.buildenv.2021.108394

Received 11 June 2021; Received in revised form 9 September 2021; Accepted 23 September 2021

^{*} Corresponding author.

Abbreviations			Δz Vertical distance between probes (m) DP Paper degree of polymerisation (–)			
ANSI	American National Standards Institute	k	Rate of paper degradation (years ⁻¹)			
ASHRAE		MR	Mixing Ratio (g•kg ⁻¹)			
110111412	Conditioning Engineers	pН	Acidity of paper (–)			
BSI	British Standards Institution	RH	Relative Humidity (%)			
CEN	European Committee for Standardization	RH _{30d}	centred 30-day moving mean of relative humidity (%)			
CREA	Consiglio per la Ricerca in Agricoltura e l'Analisi	RH _{90d}	centred 90-day moving mean of relative humidity (%)			
	dell'Economia Agraria	T	Temperature (°C)			
HVAC	Heating, Ventilation, and Air Conditioning	T_{24h}	centred 24-h moving mean of temperature (°C)			
IFLA	International Federation of Library Associations and	T _{d.14} , T _d	Temperature at 14:00 ($T_{d,14}$) and 22:00 ($T_{d,22}$) of dth			
	Institutions	.,	calendar day (°C)			
ISO	International Organization for Standardization	T d,Mean,	$T_{d,max}$ Mean $(T_{d,mean})$ and maximum $(T_{d,max})$ temperature			
NISO	National Information Standards Organization		of dth calendar day (°C)			
PAS	Publicly Available Specification					
PD	Portable Document	Indexes				
TR	Technical Report	CI	Continuity Index (–)			
UNI	Italian National Unification Institute	CoI	Completeness Index (–)			
	1	EL	Expected Lifetime of historic paper (years)			
	rs and variables	eLM	equivalent Lifetime Multiplier (–)			
$\Gamma_{\rm d}$	Adiabatic gradient of vertical temperature for dry air (°C	LM	Lifetime Multiplier (–)			
4.) (D	m ⁻¹)	NDR	Normalized Diurnal Range (–)			
Δ MR	Absolute difference between outdoor and indoor mixing	PI	Preservation Index (-)			
ADII	ratio (g•kg ⁻¹)	RH ratio				
ΔRH_{24h}	Daily relative humidity fluctuation (%)	TWEL	Time Weighted Expected Lifetime (years)			
ΔT	Vertical temperature gradient (°C)	TWPI	Time Weighted Preservation Index (years)			
ΔT_{24h}	Daily temperature fluctuation (°C)					

decay is dependent on the properties of the constituent materials (e.g., acidity of paper and physical strength) and is greatly affected by the indoor climate [5]. Paper can be classified into three types based on their average acidity (pH) and degree of polymerisation (DP): acidic paper (pH = 5.2, DP = 826.3), rag paper (pH = 6.4, DP = 1481.2) and contemporary paper (pH = 7.6, DP = 1526.2) [6].

Cellulose hydrolysis represents the main concern for damage risk of paper collections, being the rate of chemical degradation mainly driven by temperature [7]. This deterioration mechanism has been extensively studied by using the dose-response functions, such as the Lifetime Multiplier (LM) for various materials such as varnishes and cellulose [7] and the Preservation Index (PI) for organic materials (e.g. paper, textiles, plastics, dyes, leather, fur, etc.) [8]. Strlič et al. [9] derived the isochrones for paper (i.e., curves of equal expected lifetime, EL), based on a damage function which relates the DP loss with the pH of paper and the indoor temperature and relative humidity at the reference conditions of dark storage (i.e., without considering natural and artificial light). In this way, the isochrones allow to assess the chemical deterioration risk for the different paper types by taking into account their different vulnerability to cellulose hydrolysis as a function of their pH and DP [9].

Handling is the main responsible for the accumulation of wear and tears of paper [4]. The time required for a library collection to become unfit for use by readers due to the combined effect of handling and cellulose hydrolysis can be estimated through a dose-response function [4] that depends on the percentages of paper types constituting the collection (i.e., its demography) [9].

Thermal stratification may occur in high-ceilinged libraries. Hence, if too high temperatures are experienced, the chemical decay of paper collections located at the upper levels is accelerated based on the vulnerability of the paper types. Moreover, high relative humidity levels might occur in poor ventilated areas in the proximity of cooler surfaces, favouring mould growth [10]. Finally, temperature and relative humidity fluctuations might accelerate paper degradation (e.g., tensile creep of paper), but consensus on their influence on the degradation has

not been reached yet among the researchers [5].

Pollutants do not generally represent a significant threat to the overall rate of chemical degradation, as their concentrations are small (e.g., acetic acid<250 $\mu g \cdot m^{-3}$; formic acid<35 $\mu g \cdot m^{-3}$; nitrogen dioxide<15 $\mu g \cdot m^{-3}$; ozone<25 $\mu g \cdot m^{-3}$; sulphur dioxide<3 $\mu g \cdot m^{-3}$ [11,12]) and their effect on historic paper preservation is limited [13]. Dust particles can affect paper conservation in terms of cellulose degree of polymerisation, thus increasing the vulnerability of paper to the other environmental parameters [14].

Fungal spores, which are ubiquitous, may become a biological risk when RH>65% for a sufficient period of time. This risk is frequently assessed using the Sedlbauer isopleths for spore germination and mould growth for biologically recyclable materials [15].

Digital platforms, such as HERIE [16] and Collection Demography app [17], are freely available for conservation professionals and decision makers to easily and effectively carry out the quantitative assessment of the climate-induced risks for paper collections based on the prevailing environmental conditions where the objects are displayed or stored.

1.2. Standards and guidelines on library conservation

This section describes the international policy framework on the preservation of cultural heritage including library and archive materials. Table 1 summarises standards and guidelines on the library conservation. Standards are published by national or international standards bodies (e.g., UNI, BSI, CEN, ISO), whereas guidelines are usually formulated by unions (e.g., ASHRAE, IFLA). The earlier standards specified temperature (T) and relative humidity (RH) ranges for various types of materials including paper, parchment and leather [18–22]. It is worth noticing that sometimes, for the same classes of materials, these standards provide conflicting recommendations about T and RH ranges, thus arising doubts about which indication has to be followed. The introduction of the concepts of "proofed fluctuations" [23] and "historical climate" [24] led to more flexible T and RH ranges with respect to the target values previously recommended as satisfactory to mitigate

Table 1
Italian and international policy framework on the preservation of cultural heritage including library and archive materials. Remarks' field provides the microclimate specifications indicated in each document. The withdrawn standards (highlighted in footnotes) are here reported to support the interpretation of Table 2.

REF.	YEAR	DOCUMENT	INSTITUTION	TYPE OF DOCUMENT	REMARKS [®]
[18]	1997	UNI 10586	Italian National Unification Institute (UNI)	Standard	T = 14 \div 20 °C, RH = 50 \div 60% (illustrative documents). T = 18 \div 23 °C, RH = 50 \div 65% (consultation and reading). Acclimatisation is advised if T and RH in consultation places differ more than \pm 4 °C and \pm 5% respectively from storage facilities conditions.
[19]	1999	UNI 10829	Italian National Unification Institute (UNI)	Standard	T = 13 \div 18 °C, RH = 50 \div 60% (archival document and books); T = 19 \div 24 °C, RH = 45 \div 55% and $\Delta T_{24h} = \pm 1.5$ °C, $\Delta RH_{24h} = \pm 6$ % (book bindings in leather and parchment).
[20]	2001	D. lgs. 112/98,art. 150, comma 6	Italian Ministry of Cultural Heritage (MIBAC)	Legislative Decree	T = $19 \div 24$ °C, RH = $50 \div 60\%$ (books and manuscripts). T < 21 °C, RH = $40 \div 55\%$ to avoid microbiological attacks on organic materials.
[21]	2001	ANSI/NISO Z39.79 ^b	American National Standards Institute (ANSI) – National Information Standards Organization (NISO)	Standard	T < 21 °C \pm 3 °C, RH = 35 \div 50% \pm 5% (library and archival materials).
[22]	2003	ISO 11799°	International Organization for Standardization (ISO)	Standard	$T=2\div18$ °C±1 °C, RH = 30 \div 45%±3% (optimum preservation) $T=14\div18$ °C±1 °C, RH = 35 \div 50%±3% (staffed stack areas, items in regular use)
[24]	2010	EN 15757	European Committee for Standardization (CEN)	Standard	T= no limits; $RH=$ within safe bands calculated from the historical climate.
[30]	2011	ASHRAE Handbook—HVAC Applications Chapter 23 ^d	American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE)	Guidelines	Class B is the reference for historic buildings (no risk to most books): T = 15 \div 25 °C, with seasonal adjustment T+10 °C (but not above 30 °C), and T down as low as necessary to maintain RH control, and RH = 50% \pm 10% (or historic annual average for permanent collections); short-term fluctuations T±5 °C, RH±10%.
[25]	2012	PD 5454 ^e	British Standards Institution (BSI)	Guidelines	T = 13 \div 20 °C, RH = 35 \div 60% (mixed collections); T = 5 \div 25 °C, RH = 25 \div 60% (paper records storage).
[26]	2012	PAS 198 ^f	British Standards Institution (BSI)	Publicly Available Specification	Tables on the relative risk of damage and deterioration due to T and RH [26] as a function of the sensitivity to hydrolysis (rag paper = low; wood pulp paper = medium) are reported as an informative appendix.
[27]	2015	ISO 11799	International Organization for Standardization (ISO)	Standard	Recommended T and RH ranges not explicitly specified.
[32]	2016	IFLA - Principles for the Care and Handling of Library Materials	International Federation of Library Associations and Institutions (IFLA)	Guidelines	$T<10\ ^{\circ}\text{C}$ to favour paper chemical stability and physical appearance; $RH=50\div65\% \text{ to minimise mechanical damage (while reducing the risk of biological attacks)}.$
[28]	2017	BS 4971	British Standards Institution (BSI)	Standard	T = 13 \div 23 °C, with annual average T < 18 °C; RH = 35 \div 60% (mixed archives).
[29]	2018	EN 16893	European Committee for Standardization (CEN)	Standard	Tables on the relative risk of damage and deterioration due to T and RH [26] as a function of the sensitivity to hydrolysis (rag paper = low; wood pulp paper = medium) are reported as an informative appendix.
[31]	2018	ISO/TR 19815	International Organization for Standardization (ISO)	Standard	Tables on the relative risk of damage and deterioration due to T and RH [26] as a function of the sensitivity to hydrolysis (rag paper = low; wood pulp paper = medium) are reported as an informative appendix.
[10]	2019	ASHRAE Handbook—HVAC Applications Chapter 24	American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE)	Guidelines	Class B is the reference for historic buildings (no risk to most books): $T \le 30$ °C, $RH = 30 \div 70\%$, seasonal adjustment from annual average $T+10$ °C, $T-20$ °C) and $RH\pm10\%$, short-term fluctuations $T\pm5$ °C, $RH\pm10\%$.

^a The paper types to which microclimatic specifications are suitable for are specified in this column, if mentioned in the document.

mechanical risks for organic hygroscopic materials. As a result of the above concepts, the standards have evolved along different paths: ISO and BSI updated their documents [22,25,26] with new specifications [27–29], while UNI did not change the original version of the norms [18, 19]. The ASHRAE handbook [30] introduced museum climate classes without prescriptive T and RH specifications, which have been extensively adopted for both environmental design and risk management in museums, galleries, archives and libraries. In the last years, EN 16893:2018 [29], ISO/TR 19815:2018 [31] and the updated ASHRAE guidelines [10] focused on sustainability and greater flexibility in the

climate control strategy based on risk management principles and existing literature on best practices in museums as well as the collections' response to the surrounding environment. Although EN 16893:2018 [29] gives useful recommendations for buildings intended for the storage and use of heritage collections (including reading rooms), no European standards exist by CEN/TC 346, i.e. the Technical Committee of the European Committee for Standardization dedicated to the Conservation of cultural property, which specifically deals with the conservation in libraries and archives.

^b Withdrawn in 2013.

 $^{^{\}mathrm{c}}$ Withdrawn and superseded by ISO 11799:2015 (that will be replaced by ISO/AWI 11799, currently under development).

^d The ASHRAE Handbook —HVAC Application of 2015 did not modify Chapter 23.

^e Withdrawn and superseded by EN 16893:2018 and BS 4971:2017.

f Withdrawn in 2018.

 Table 2

 Indoor microclimate monitoring studies in historic libraries over the last 20 years.

Ref.	Year	Library (foundation year)	Place	Monitoring period	HVAC	Damage risk assessment	Policy framework
[34]	2002	Guildhall library (1876) Bronte Parsonage Museum library	Leicester (UK) Haworth (UK)	12 months	Yes No		BS 5454
		(1778–79)	naworai (ore)		140		
[49]	2007	Audley End House (early 17th-	Saffron Walden	not mentio-ned	Yes	Time Weighted Preservation Index	BS 5454
		century)	(UK)			[8].	
		Brodsworth Hall (1861)	Doncaster (UK)				
		Eltham Palace (1933)	London (UK)				
		Iveagh Bequest (1754)	London (UK)				
		Walmer Castle (1539)	Walmer (UK)				
		Dover Castle (1912)	Dover (UK)				
[11]	2011	National Library (1726)	Prague (Czech Republic)	9 months (on monthly basis)	No		
[38]	2014	Malatestiana (1454)	Cesena (Italy)	3 months	No		UNI 10829; MIBAC D. lgs 112/98.
[39]	2016	Palatina (1761)	Parma (Italy)	2 months (spot)	No		UNI 10829; MIBAC D. lgs 112/98;
							ASHRAE guidelines
[12,	2016	Classense (1513)	Ravenna (Italy)	15 days summer/15	No	Wear and tear dose-response function	UNI 10586;
50]				days winter		[4]; paper isochrones [9].	UNI 10829
[48]	2017	Tire Necip Pasa (1827)	Izmir (Turkey)	1 year	No	Lifetime Multiplier [7]; Sedlbauer	ASHRAE guidelines
		• • • •		•		isopleth [15].	· ·
[47]	2018	Fine Arts Library (1905)	La Plata	4 months	Yes	•	UNI 10829
		Physics Library (1897)	(Argentina)		Both		
		Historical Archive of the Presidency			Yes		
		of the UNLP (1884)					
		Historical Archive of the Natural			Yes		
		Sciences Museum (1877–1884)					
[36]	2018	Baroque Library of the University	Coimbra	6 months	No	equivalent Lifetime Multiplier;	UNI 10829
		(1716)	(Portugal)			Sedlbauer isopleth [15].	
[51]	2018	National Library (1677)	Warsaw (Poland)	13 months	Yes	•	
[37]	2020	Library of the National Observatory (1842)	Athens (Greece)	1 month	Yes		ISO 11799:2003; UNI 10586

1.3. Microclimate monitoring in historic libraries

In Italy, almost half of the historic libraries was founded before the XIX century [33]. Historic libraries are frequently conserved in massive buildings characterised by high thermal inertia [12,34-37]. Most of these buildings have natural ventilation and unconditioned indoor climate (i.e., without any HVAC systems) [11,12,36,38,39]. Windows, usually small-sized, were often placed on two sides of the main reading hall [12,35,39] to maintain relatively constant daylight levels for visual comfort. The orientation of the building was chosen to protect from dampness and to take advantage of natural daylighting. To cite but a few, Vitruvius advised that libraries should have an eastern exposure to the morning light useful to dispel dampness [40] and, many centuries after, the Italian Renaissance architect Leon Battista Alberti wrote that private libraries should have western exposure to benefit the reading at twilight [41]. Recently, it has been highlighted that hygroscopic collections act as buffers on air relative humidity fluctuations due to the exchanges of moisture with the surrounding air [42]. The RH buffering, already visible at smaller scale (e.g. in sealed boxes containing paper [43]), can be evaluated at large scale (e.g. in libraries and historical buildings) through microclimate analysis and hygrothermal simulation

Table 2 provides a synthetic overview of studies on microclimate monitoring conducted in historic libraries over the last 20 years. A significant variability can be observed in the length of the monitoring period as well as in the policy framework adopted for microclimate analysis and, more in general, in the methods followed for damage risk assessment. The T and RH ranges recommended by standards and guidelines in Table 1 have been frequently used as threshold values to evaluate the quality of the environmental conditions for the conservation of library materials [24,26,28,29,34]. Sedlbauer spore germination isopleths and fungal growth curves have been employed in Refs. [36,48]

to evaluate the biological risk for collections. Synthetic indexes, such as the equivalent Lifetime Multiplier (eLM) and the Time Weighted Preservation Index (TWPI), have been used in Refs. [36,49] to account for average values of LM and PI over the monitored period. The damage function for historic paper [9] was used in Ref. [50] for the estimation of the collection expected lifetimes in various conservation scenarios.

1.4. Research aims

Several studies have separately evaluated the microclimate inside historic libraries; however, few of them were based on long-term microclimate observations and there is lack of uniformity in the use of standardised microclimate specifications and risk assessment methods (Table 2). Taking advantage of the availability of annual time series of data collected by the authors, the microclimates of three historic libraries located in different sites of Italy (Milan, Udine and Rome) were analysed. Besides, the collected hygrothermal conditions were compared with respect to those recommended by both Italian and European regulations (Table 1) and used as an input in dose-response functions for paper. Moreover, the comparison allowed us to investigate to what extent the differences in the external climate, building features and library management can affect the climate-induced risk for the preservation of paper collections. The comparison can provide useful insight on the impact on paper collections of conditioned and unconditioned indoor climates to inform preservation strategies within historic buildings. Section 2 deals with the description of the three historic libraries, the microclimate monitoring campaigns and the methods for characterising the indoor climate and assessing the conservation risks. Section 3 is devoted to the presentation and discussion of the results. Finally, section 4 outlines the main conclusions of the work and the future perspectives in the research on library preservation.

2. Materials and methods

2.1. Case studies: the three historic libraries in Italy

The historic libraries under study (Fig. 1), hereafter named Ca' Granda (Milan), Delfiniana (Udine) and Collegio Romano (Rome), are located in different regions of Italy. Milan (Köppen-Geiger climate class Cfa, i.e. temperate climate, fully humid with hot summer [52]) and Udine (Köppen-Geiger climate class Cfb, i.e. temperate climate, fully humid with warm summer [52]) are in northern Italy, whereas Rome is in the central part of the peninsula (Köppen-Geiger climate class Csa, i.e. temperate climate, with dry and hot summer [52]).

2.1.1. Ca' Granda Library (Milan)

The library is hosted in the Ca' Granda Ospedale Maggiore Policlinico (Milan, Lat. 45.5°N and Long. 9.2°E, 120 m a.m.s.l.). The library collection of the IRCC Foundation (Istituto di Ricovero e Cura a Carattere Scientifico) represents a national *unicum* in terms of richness and specialisation on medical sciences, with a patrimony estimated in about 100.000 printed volumes among monographies, periodicals and magazines from XV to XX century [53]. The history of the collection dates back to the foundation of the Hospital (1456), but the main core is from the XIX century and it is continuously updated thanks to private donations. The collection is deployed in a three-level wooden shelf on the perimeter of a room at the ground floor (Fig. 2a).

2.1.2. Delfiniana Library (Udine)

The Library of the Archbishop's Palace (Udine, Lat. 46.1°N and Long. 13.2°E, 113 m a.m.s.l.) was named "Delfiniana" after his founder, Dionisio Delfino, which was the patriarch of the diocese of Aquileia from 1699 to 1734 [54]. The foundation of the library dates back to 1708, when the patriarch commissioned the construction of its building, to be donated to eternal public utility. The collection was organised in a two-level wooden shelf deployed on the perimeter of a hall at the second

floor of the patriarchal palace (Fig. 2b). The collection comprises printed volumes, manuscripts (dated from XVI to XIX century) and some illuminated liturgical codices (dated from IX to XIV century). The library is nowadays part of the touristic itinerary of the Diocesan Museum of IIdine

2.1.3. CREA Meteorological Library at the Collegio Romano (Rome)

The Historical Meteorological Library in the monumental complex of Collegio Romano, hereafter called simply Collegio Romano (Rome, Lat. 41.9°N and Long. 12.5°E, 21 m a.m.s.l.), is considered the most important collection in Italy devoted to Atmospheric Sciences, Meteorology and Geophysics of the Modern Age (Fig. 2c). Although its original nucleus, gathered by Jesuits, dates back to the foundation of the building (1584), the current location on its fourth floor was elected in 1879, after the establishment of the first Central Meteorological Office in Italy, so as to become an integral part of it (currently an important sector of CREA, the leading Italian research organization dedicated to the agrifood supply chains). The collection includes several national and international periodicals published by scientific societies, CREA's interesting publications and original books and manuscripts, mostly from the XIX century [55].

The plans of three historic libraries are shown in Fig. 2. Ca' Granda library (Fig. 2a) has the largest volume (approximately $2100 \, \text{m}^3$), with rectangular shape and high ceiling (up to $12 \, \text{m}$). Delfiniana (Fig. 2b) has similar features as Ca' Granda, but smaller dimensions (approximately $1150 \, \text{m}^3$). Collegio Romano (Fig. 2c) has a 4.5 m high ceiling and the smallest volume (approximately $380 \, \text{m}^3$). All the libraries are covered by a wooden roof and have SW-W-facing windows on two orders. The windows in Collegio Romano and Ca' Granda have simple glass panes and wooden shadings which are closed most of time during the year. New air-tight windows with UV-IR-filtered glass were mounted in Delfiniana in May 2017 in place of the previous single panes [54]. During the microclimate monitoring campaigns, Ca' Granda and Delfiniana had no HVAC (Heating, Ventilation and Air-Conditioning) system, whereas

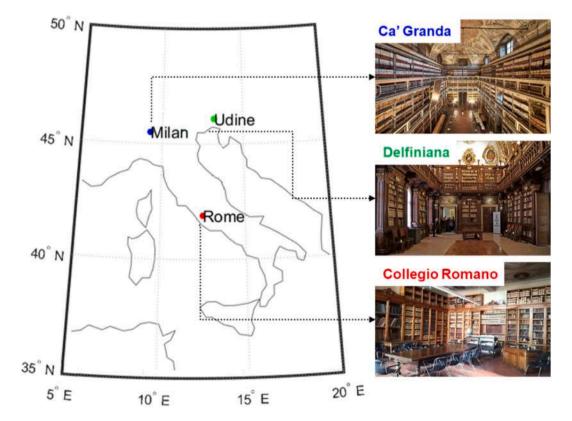
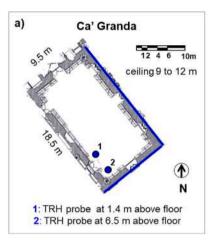
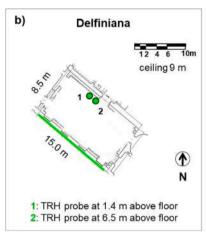


Fig. 1. The geographical position of the three historic libraries (Italy) under study: Ca' Granda in Milan, Delfiniana in Udine and Collegio Romano in Rome.





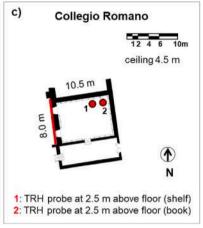


Fig. 2. Plans of the historical libraries with the position of the monitoring system devices (TRH) marked as coloured points. External walls are highlighted with coloured lines.

Collegio Romano was heated by cast iron radiators from November 1st to April 15th (operating from 6 a.m. to 6 p.m. local time) and cooled through fan-coils (switched on when needed). All the libraries were naturally ventilated.

The three library collections are mostly made of mid-19th—mid-20th century paper of Western origin (lignin containing, rosin sized, printed and non-coated), therefore the prevalent composition of the collections was assumed to be acidic. Fig. 3 depicts the percentages of paper types constituting each of the historic collections under study, as estimated on private communication with the library conservators.

2.2. The monitoring campaigns

The indoor climate monitoring campaigns to study the hygrothermal behaviour of the three libraries covered different periods (Table 3), as they were independently planned before this comparative investigation. Two thermo-hygrometers (Fig. 2) were placed inside each library to measure temperature (T) and relative humidity (RH) at local time. In Ca' Granda and Delfiniana, where ceilings are high, TRH probes were deployed at two different heights (1.4 m and 6.5 m above floor) to monitor vertical temperature gradients. In Collegio Romano, one TRH probe was placed on a shelf and the other one inside a mock-up book on the same shelf with the aim to derive the book response time, i.e. the lag between the RH values measured inside the book with respect to those collected in the room. The mock-up book (250 mm \times 180 mm \times 28 mm) was made using modern rag paper sheets, with cardboard covers and leather spine. The metrological features of the indoor T sensors (Pt100 resistance thermometers) and RH sensors (thin film capacitive sensors), reported in Table 3, were in accordance with the current European Standards on the instruments to be used in cultural heritage

conservation [56,57]. Although a higher availability of instruments -also to measure other environmental variables-would have provided a better representativeness of the microclimate behaviour in the libraries, the choice of the instrument number and types was limited due to technical and financial reasons.

2.3. Indoor climate characterisation

The long time series of microclimate data collected in the historic libraries were evaluated for completeness before applying data analysis through the Completeness Index (CoI) and the Continuity Index (CI), two indexes proposed in Ref. [58] and already applied to long-term measurements in other conservation contexts [59,60]. Temperature and relative humidity values collected over the year in the libraries were compared in order to highlight the differences among the sites, if any. Mixing Ratio (MR) was calculated from T and RH values using the formula provided in EN 16242 [57] and used as a proxy to estimate the magnitude of the indoor-outdoor water vapour exchanges through infiltrations and openings [61–63].

The buffering capacity of the building envelopes to smooth out the maximum outdoor temperatures occurring in daylight hours was evaluated using the Normalized Diurnal Range (NDR). NDR is an index defined in Ref. [64] as the ratio between the observed diurnal indoor temperature range (calculated as the difference between the temperatures at 14:00 ($T_{\rm d,14}$) and 22:00 ($T_{\rm d,22}$) of dth calendar day) in the monitored period (MP) with respect to the reference period (RP). T $_{\rm d,14}$ and T $_{\rm d,22}$ were chosen as they are close to the outdoor diurnal maximum and mean temperatures, respectively. Since the NDR value results from the combined effect of building envelope, solar exposure, room ventilation and use [64], this index is not able to distinguish among their

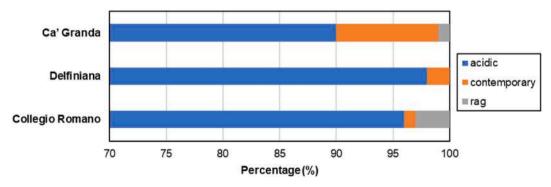


Fig. 3. Paper types constituting the library collections.

Table 3Microclimate monitoring campaigns in the three historic libraries in Italy.

Library (foundation year)	Place	Monitoring period	Instrument (uncertainty)	Sampling frequency	Outdoor climate	Analysed period
Ca' Granda (1456)	Milan	Sep 2011–Jan 2013	Comet®, model R3121 (0.4 $^{\circ}$ C for T, 2.5% for RH)	15 min	Weather station (Lat. 45.5°N and Long. 9.2°E)	1 st Jan 2012–31 st Dec 2012
Delfiniana (1708)	Udine	Oct 2016–Mar 2018	Testo®, model 175H1 (0.4 °C for T, 2% for RH)	15 min	Weather station (Lat. 46.1°N and Long. 13.2°E)	1 st Jan 2017–31 st Dec 2017
Collegio Romano (1584)	Rome	Jun 2019 – ongoing	Rotronic®, model HC2A-S3 (0.3 $^{\circ}$ C for T, 1.5% for RH)	10 min (recorded every 30 min as averaged values)	Weather station (Lat. 41.9° N and Long. 12.5° E)	1 st Jun 2020–31 st May 2021

individual contributions. Nevertheless, although NDR does not entirely depend from the air exchange rate (AER), it can be considered as a proxy of AER to a certain extent. The outdoor temperatures in the 1991–2020 reference period were extracted from the E-OBS v23.1e [65] gridded observational dataset (spatial resolution of $0.1^{\circ} \times 0.25^{\circ}$) via the Climate Data Store (CDS) infrastructure. As E-OBS v23.1e does not include hourly data, the daily maximum temperature $(T_{d,max})$ and the daily mean temperature $(T_{d,mean})$ in the reference period were extracted and used in the place of $T_{d.14}$ and $T_{d.22}$ [64].

The formulation of NDR was here adjusted to enhance its capability to sense the active climate control using heating systems as follows and it was calculated in each site under study:

$$NDR = \frac{\left(T_{d,14} - T_{d,22}\right)_{MP}}{\left(T_{d,max} - T_{d,mean}\right)_{RP}}$$
(1)

where n_{RP} is the total number of days in the reference period (i.e. 30 years). For this reason, if NDR is around zero, the building has a high thermal buffering capacity and low ventilation, while if NDR is close to unity, the indoor and outdoor temperatures are practically identical. The latter condition can be experienced when there is a persistence of sufficient ventilation due, for example, to open windows. Negative NDR can also occur, mainly in summer, when the effect of the highest outdoor temperature values is visible later on the indoor evening temperatures.

In Ca' Granda and Delfiniana libraries, where ceilings are high, the indoor air stability was evaluated by comparing the measured vertical temperature gradient (as the difference between the temperature values measured at 1.4 m and 6.5 m above the floor) with the dry adiabatic gradient of vertical temperature, i.e. $\Gamma_d = \Delta T/\Delta z \cong -0.1~^{\circ}\text{C}/10~\text{m}.$

The possible effect of the moisture exchanges between the hygroscopic collections and the surrounding air on the stabilisation on the indoor RH values was explored. Since RH is not an independent variable, it must be studied together with air temperature or mixing ratio, which both influence its behaviour. The RH variability was assessed based on the following approaches: a) analysis of the concurrent time evolutions of indoor T, RH and MR measurements over the year (Fig. 5); b) synthetic visualisation of the frequency distributions of the indoor and outdoor RH observations (Fig. 8); c) calculation of the Relative Humidity ratio (RH ratio, Table 4); d) assessment of the average absolute differences between simultaneous outdoor and indoor MR values (\Delta MR, Table 4). The RH ratio was defined as the ratio (in percentage) between the indoor (in) and the outdoor (out) seasonal spreads, i.e., the sum over a year of the absolute differences between the i-th centred 90-day moving average of relative humidity (RH_{i,90d}) and the i-th relative humidity observation (RHi):

Table 4 RH ratio, average absolute difference indoor/outdoor MR observations ($\overline{\Delta MR}$) and hygroscopic ratio.

Library	RH ratio	$\overline{\Delta MR}$	Hygroscopic ratio
Ca' Granda	24.1%	1.3 g/kg	11.3%
Delfiniana	12.7%	1.2 g/kg	6.2%
Collegio Romano	9.3%	1.8 g/kg	7.9%

RH ratio =
$$\frac{\left(\sum_{i=1}^{n} |RH_{i,90d} - RH_{i}|\right)_{in}}{\left(\sum_{i=1}^{n} |RH_{i,90d} - RH_{i}|\right)_{out}} \cdot 100$$
 (2)

When the RH ratio is low, the library has limited air exchanges with the outdoors together with high moisture buffering capacity by the hygroscopic materials (if present); when the RH ratio is close to 100%, the indoor and outdoor RH seasonal spread are practically identical.

2.4. Conservation risk assessment

As evaluated by the conservators of the libraries under study, based on their regular and qualitative risk assessment to determine the optimal preservation of their paper collections, the major recognized deterioration risk is cellulose hydrolysis due to the high temperatures occurring in summer. Indeed, since no fungal proliferation was reported to be observed in the case studies before the monitoring campaigns (Dr. Paolo Galimberti for Ca' Granda, Dr. Dania Nobili for Delfiniana and Dr. Luigi Iafrate for Collegio Romano, personal communications), this risk was no further investigated. This investigation was stimulated by the interest of the conservators and managers of the libraries to explore the possible climate-induced deterioration risks by using specific dose-response functions. In fact, it was not possible to ascribe the actual preservation state of the collections over time to the monitored indoor climate, because the previous conditions and arrangement of the collections were not fully known and might have changed through their history. A dose response function for paper was developed in Ref. [9] by modelling k, i. e. the rate of paper degradation per year (at the conditions of dark storage), as a function of both the acidity of paper (pH) and the indoor temperature and relative humidity, as follows:

$$\ln(k) = 36.981 + 36.72 \cdot \left(\frac{\ln(1 - RH)}{1.67 \cdot T - 285.66}\right)^{\frac{1}{2.491 - 0.0127}} + 0.244 \cdot \ln(10^{-pH}) - \frac{14300}{(T + 273.15)}$$
(3)

According to Ref. [9], Equation (3) can be embedded in the Ekenstam's function to obtain a damage function for paper where the expected lifetime, EL (i.e. the time required for objects to become unfit for use), is calculated as a function of the initial degree of polymerisation (DP) and the critical degree of polymerisation, which is typically 300, at which objects are no longer suitable for general access [9]:

$$EL = \left(\frac{1}{300} - \frac{1}{DP}\right) \cdot k^{-1} \tag{4}$$

To account for an average risk of chemical degradation for paper collections due to cellulose hydrolysis, we defined a synthetic index, hereafter named as Time Weighted Expected Lifetime (TWEL). The TWEL index integrates the damage function for paper derived and validated in Ref. [9] with the concept of a time-weighted index to quantitatively compare the chemical risk due to the indoor climate conditions over different time windows (e.g. a year or a season). Inspired

by the well-established TWPI index formulated for the chemical degradation of organic materials [8], the TWEL is calculated based on the running sum of the multiplicative inverses of EL at each time t. Indeed, we examined various formulations for averaging the EL (e.g. the simple arithmetic mean, the inverse of the arithmetic mean, etc.) and found that the proposed calculation of TWEL was the one giving more weight to the lower values of EL, thus considering the risk underlying the worst-case scenario. In this way, the TWEL emphasises the impact of the most adverse climate conditions (mainly temperature) on the overall chemical deterioration risk for paper collections on a yearly and seasonal basis.

Fig. 4 shows the workflow for the calculation of the TWEL, which can be summarised in the following steps:

- I. Calculation of 24-h moving average of T (T_{24h}) and 30-day moving average of RH (RH_{30d});
- II. Calculation of EL from T_{24h} and RH_{30d} according to Equation (4);
- III. Calculation of running sum of the reciprocals of EL at each time t (EL_{runsum}) ;
- IV. Calculation of seasonal/yearly TWEL value as the ratio between the number of observations in the selected period (n) and the ${\rm EL}_{\rm runsum}$ value.

The running average over 24 h for temperature and 30 days for relative humidity, chosen according to Ref. [8], were found to be compatible with the response time of paper books as estimated by using the measurements collected in Collegio Romano according to the formula in Ref. [66]. Based on the prevalent composition of the library collections (Fig. 3), in this study we used DP = 826.3 and pH = 5.2 typical of acidic collections [47]. Since it was not experimentally validated yet, the TWEL value expresses only a weighted average of the

chemical degradation risk based on the EL.

Demographics of collections were used to investigate the combined effect of paper handling and indoor climate conditions over the time required for objects to become unfit for use. It was estimated that each handling event could develop, on average, at least one large missing piece every 100 pages for objects having DP < 300, thus losing their fitness for use [4,9]. The Collection Demography App [17], was used to run simulations for estimating the accumulation of large missing pieces per number of handlings. To calculate the demographic curves, the yearly average temperature and relative humidity and an average access every 2 years in the library were used. This analysis was based on the percentage of paper typologies constituting each library collection (Fig. 3). The effect of deacidification of 10% of the acidic collection was finally explored by recalculating the demographic curves while modelling the deacidified fraction with DP =640 and pH =8.1.

3. Results and discussion

3.1. Indoor climate characterisation

The comparative analysis carried out in this study was based on the periods reported in Table 3. The measurements collected by the TRH probes at 1.4 above floor for Ca' Granda and Delfiniana and TRH probe on the shelf at 2.5 m above floor for Collegio Romano (Fig. 2) were taken into account. The quality of T and RH time series was objectively evaluated before analysing microclimate data in the three libraries and resulted to be complete (CoI = 1) and continuous (CI = 1) for the considered periods.

On a yearly basis, the indoor median temperatures were higher than the outdoor ones (Fig. 5). The outdoor temperatures differed depending on the site, with Udine having cooler average values and larger

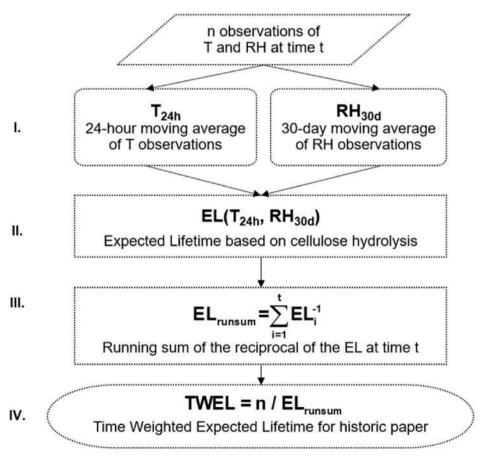


Fig. 4. Workflow of the procedure to calculate the Time Weighted Expected Lifetime (TWEL).

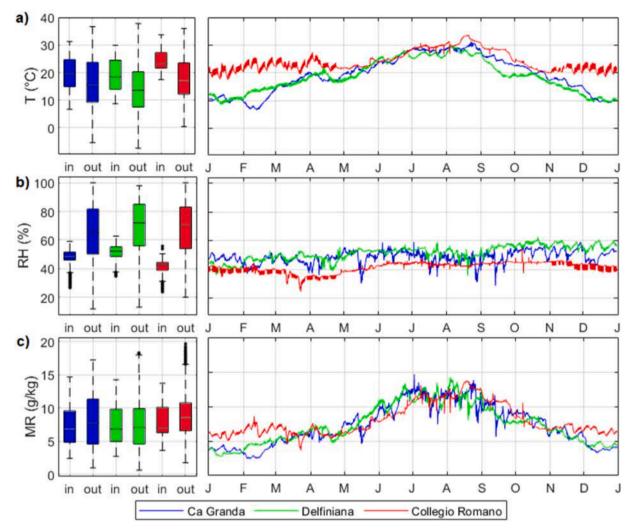


Fig. 5. Left: box-and-whisker plots of indoor (in) and outdoor (out) temperature (a), relative humidity (b) and mixing ratio (c) observations. Right: time plots of indoor T, RH and MR values. In the box-and-whisker plots, the medians are indicated with horizontal lines dividing each box and the outliers, i.e., the values above or below 1.5 × IQR (IQR = interquartile range), with black circles.

variability (from $-7.4\,^{\circ}\text{C}$ to $37.8\,^{\circ}\text{C}$) if compared to Milan (from $-5.4\,^{\circ}\text{C}$ to $36.7\,^{\circ}\text{C}$) and Rome (from $0.4\,^{\circ}\text{C}$ to $36.0\,^{\circ}\text{C}$). The indoor temperatures in the three libraries showed comparable levels and trends from June to September, except for some sporadic T drops in Collegio Romano (red line) occurred in July/August caused by the cooling system. In cold periods, Ca' Granda and Delfiniana behaved similarly, whereas Collegio Romano experienced higher T due to the switching on of the heating system in the working days (from Monday to Friday).

The indoor relative humidity observations were found to be significantly lower than the outdoor ones (Fig. 5), with medians of indoor relative humidity around 50% in Ca' Granda and Delfiniana and around 40% in Collegio Romano. The outdoor RH variability, ranging from 11.8 to 100% in Ca' Granda, from 13 to 98% in Delfiniana and from 20 to 100% in Collegio Romano, was smoothed out inside the libraries. The drops in the RH observations in the libraries were mainly driven by MR; however, in Collegio Romano the RH drops were also associated with T rises caused by the heating system.

No significant differences in the median of the indoor MR levels (Fig. 5) were observed between Ca' Granda, Delfiniana and Collegio Romano libraries (MR in \approx 7.0 g/kg), with outdoor MR medians equal to 7.7 g/kg in Milan, 7.1 g/kg in Udine and 8.6 g/kg in Rome. Although the indoor MR levels in Collegio Romano were only slightly different from those in the other two libraries from spring to autumn, in winter a different MR behaviour was observed, probably due to the moisture

release from hygroscopic materials caused by the heating system.

The Normalized Diurnal Range index is shown in Fig. 6 to compare the thermal buffering capacity of the buildings housing the historic libraries. Ca' Granda had NDR values ranging from -0.1 to +0.1 and the highest frequency of zero values, meaning that the building envelope cut off most of the largest outdoor thermal variability, probably due to massive building envelopes and low air exchange rates [67]. The latter assumption could be evaluated by specific methods (e.g. the decay curves of tracer gas), which however are beyond the scopes of this work. The NDR values obtained in Delfiniana showed a different behaviour in the cold seasons with respect to the warm ones, with NDR values around zero in winter and negative NDR values during summer. The negative NDR values, meaning that $T_{n,21} > T_{n,14}$, were probably due to indoor summer T peaks being delayed after the outdoor T peaks. In Collegio Romano, summer NDR values ranged from -0.1 to +0.1 whereas distinct features characterised the heating period (from November until April), with a higher variability of NDR values. The short-term fluctuations caused by the active climate control system could be limited allowing for more relaxed climate targets in winter (e.g. switching off the radiators or lowering their temperature setpoints), with relevant beneficial effects also in terms of energy consumption [68]. The NDR values in Collegio Romano highlighted a higher heat accumulation (NDR<0) due to both the use of the radiators and the low air exchanges in summer. It was not possible to establish a correspondence between

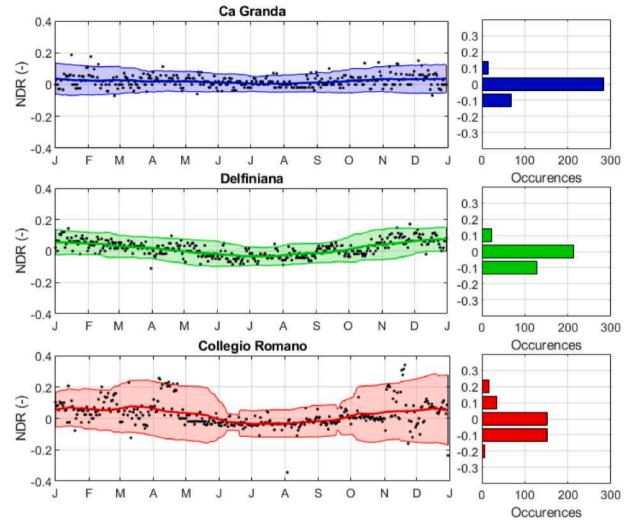


Fig. 6. Normalized Diurnal Range (NDR): time plots (left) and histograms (right). NDR values in time plots (black dots) are shown together with the curves of the 90-day moving average (thick coloured curves) and double moving standard deviation (thin coloured curves).

the anomalous NDR values (i.e. those lying outside the coloured band in Fig. 6) and particular events occurred in the libraries. This latter fact was interpreted as the consequence of the operation of averaging the daily outdoor values over the reference period, that might have smoothed out the real pattern of the outdoor thermal variability occurred in the monitored years.

In Fig. 7 the thermal vertical gradients ($\Delta T/\Delta z$) in Ca' Granda and Delfiniana were compared with the adiabatic gradient of vertical

temperature for dry air ($\Gamma_d=-0.05~^{\circ}C/5~m$). Air instability occurs when $\Delta T/\Delta z > \Gamma_d$. In Delfiniana air instability (i.e. upraising flow of warmer air masses from the floor and descending flow of colder air masses from the ceiling) occurred in winter due to the higher T measured at 1.4 m above floor with respect to those at 6.5 m (Fig. 7). Air instability can affect paper conservation as it could be responsible for transporting pollutants and other airborne particulate matter, thus causing soiling and deposition of dust and fungal spores that are

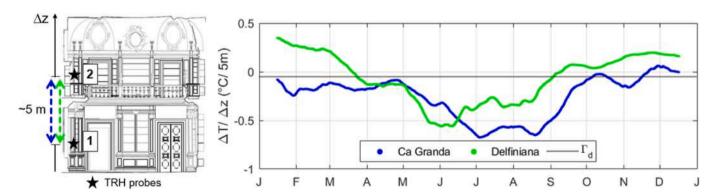


Fig. 7. Thermal vertical gradient ($\Delta T/\Delta z$, where $\Delta T=T1$ -T2), shown as 30-day centred moving averages, together with the adiabatic gradient of vertical temperature for dry air ($\Gamma_d=-0.05~^{\circ}C/5$ m). Thermal vertical gradients exceeding Γ_d indicate vertical instability.

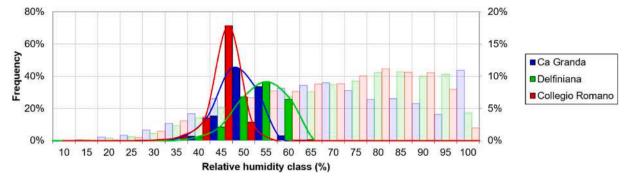


Fig. 8. Frequency distribution plots of the indoor (opaque) and outdoor (transparent) RH observations in a year. The secondary vertical axis refers to the frequency of outdoor RH values in each class.

ubiquitous. The lower temperatures collected near the ceiling were interpreted as the possible consequence of the poorly insulated roof causing heat losses, which were mitigated by the thermal masses in the lower air layers. As expected, during the warm season the air temperatures near the ceiling were higher than those collected at lower levels above the floor. The natural summer negative peaks were observed from mid-May to mid-June in Ca' Granda and from mid-June to August in Delfiniana. Thermal stratification may affect the conservation of the collections closer to the ceiling due to accelerated rates of chemical deterioration.

Fig. 8 shows the frequency distribution plots of the indoor and outdoor RH observations in the three libraries over the selected periods. All the libraries were characterized by high RH stability if compared to the outdoor variability. Indeed, the spread of the indoor RH observations was narrow and almost symmetric around the medians, as opposed to the markedly skewed distributions of the outdoor RH towards the highest values. Ca' Granda had RH observations centred around 48% (median) varying between 44% (7th percentile) and 52% (93rd percentile), Delfiniana RH median = 52% (varying between 48% and 57%) and Collegio Romano RH median = 43% (varying between 39% and 46%). It is worth noticing that, in the case of a Gaussian distribution, 7th and 93rd percentiles correspond to 1.5 standard deviations [24].

The RH ratio results are summarised in Table 4 together with the average absolute differences between the indoor MR observations and the outdoor ones $(\overline{\Delta MR})$ and the estimated hygroscopic ratio, calculated as the ratio between the volume of total hygroscopic materials (i.e. collection and wooden shelves), and the volume of library room. Ca' Granda had the relatively highest RH ratio (i.e., 24.1%), whereas Delfiniana had the relative lowest hygroscopic ratio (i.e., 6.2%). Although the libraries had comparable values of $\overline{\Delta MR}$ (i.e., 1.3 g/kg and 1.2 g/kg, respectively), the RH ratio resulted to be almost doubled in Ca' Granda (RH ratio = 24.1%) with respect to Delfiniana (RH ratio = 12.7%). Collegio Romano showed the relatively highest value of $\overline{\Delta MR}$ (i.e., 1.8 g/kg), together with the lowest value of RH ratio (i.e., 9.3%). Since it was not possible to measure the air exchanges, the MR values were used as a proxy of the indoor/outdoor air masses exchanges [57,61], as already done in literature [62,63]. For this reason, the relatively higher value of ΔMR in Collegio Romano was interpreted as the possible consequence of reduced indoor/outdoor air masses exchanges with the outdoors.

Fig. 8 and Table 4 seem to confirm a key aspect of the buffering behaviour of hygroscopic materials [42,69] in confined environment: when the percentage of storage space occupied by hygroscopic materials (i.e., the hygroscopic ratio) is high and air exchanges are low, the hygroscopic materials actively contribute to the stabilisation of RH (i.e., lower values of the RH ratio). However, even though stable relative humidity values limit the risk of tensile creep of paper [5], it has to be considered that reduced ventilation may favour fungal growth and pollutant accumulation.

3.2. Conservation risk assessment

Fig. 9 shows the temporal difference of the expected lifetime (EL) from the planning horizon of 500 years in the libraries. The conservation risk assessment took into account acidic historic paper, since it was the predominant paper type in the library collections as well as the one needing more attention due to its high sensitivity to indoor climate. From March to December, the EL values resulted to be lower than 500 years, meaning that temperature and relative humidity levels affected the durability of acidic historic paper. In winter, the low temperatures observed in Ca' Granda and Delfiniana (where indoor climate is not controlled) led to a significant increase in the expected lifetime of the collections. In Collegio Romano, in which radiators were active from Monday to Friday during the heating season (from November till April), the paper collection was under risky conditions.

The T and RH observations collected in the libraries are shown in Fig. 10 together with the T and RH ranges recommended by some of the standards in Table 1 (i.e., [18–20,29,31]). The microclimate conditions inside Ca' Granda and Delfiniana fitted the selected specifications for a small amount of the year and substantially never in the case of Collegio Romano.

In Fig. 11 the temperature and relative humidity observations collected in the libraries were plotted over the isochrones for acidic paper collections to evaluate the expected lifetime associated with the indoor climate. The TWEL index was used to average the overall chemical risk on both a yearly and a seasonal basis. The value of TWEL in all the libraries was lower than 350 years in autum, lower than 460 years in spring and only around 135 years in summer. In winter, TWEL was equal to 1540 and 1230 years respectively in Ca' Granda and Delfiniana but only 400 years in Collegio Romano. It is worth noticing that, since the calculation of TWEL does not linearly weight the expected lifetime over time, lower values of EL have a higher influence on the final output, thus considering a worst-case scenario. For this reason, although the winter TWEL values in Ca' Granda and Delfiniana were more than three times higher than in Collegio Romano, the yearly TWEL values in the three libraries were comparable in magnitude (293, 299 and 235 years, respectively). In Ca' Granda, the heat accumulation observed in summer (Fig. 7) caused the yearly TWEL near the ceiling to be reduced of a further 5% with respect to the TWEL at 1.4 m above floor. These results better highlighted not only that winter heating should be avoided whenever possible as a preventive conservation measure, but also that it is necessary to reduce indoor temperatures exceeding 20 °C in order to significantly mitigate the risk of cellulose hydrolysis. Moreover, the comparison among the libraries showed that the natural unconditioned indoor climates within historic buildings are preferable and thus could be suggested as a sustainable and effective preservation strategy for paper collections.

Looking at Fig. 10 in light of the isochrones in Fig. 11, it can be noticed that the T and RH ranges recommended by EN 16893 [29] are advisable for the preservation of acidic paper collections; conversely, the

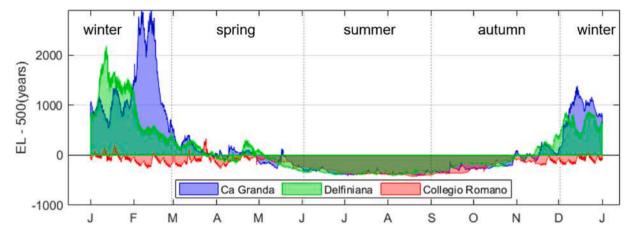


Fig. 9. Expected Lifetime (EL) spread from 500-year planning horizon for acidic paper.

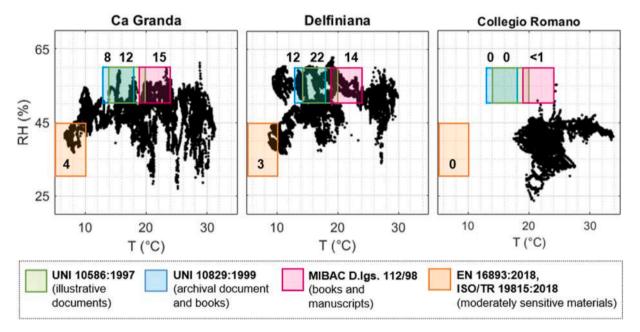


Fig. 10. Temperature and relative humidity observations (black dots) together with the T and RH ranges recommended by some of the standards in Table 1 (coloured boxes). In each box is reported the percentage of observations inside each target.

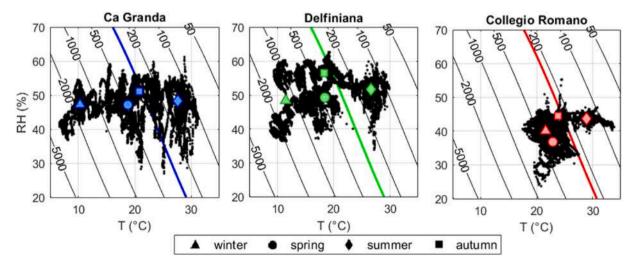


Fig. 11. Temperature and relative humidity observations (black dots) plotted on the isochrones of expected lifetime for acidic paper, together with the yearly (coloured curves) and seasonal (coloured markers) TWEL.

T and RH ranges recommended by Italian standards UNI 10586 [18], UNI 10829 [19] and legislative decree D.lgs. 112/98 [20] may not prevent from the chemical decay of acidic paper collections before the planning horizon of 500 years. It has to be borne in mind that, although the 500-year planning horizon was here used as a reference to guide the evaluation, each cultural institution should seek the expected lifetime being more appropriate for its collection given the peculiar contextual factors (e.g., the mission, the purpose, the building performance and the climate forcing due to its geographic location [10]). In this sense, the use of the TWEL index can be advantageous to inform preventive conservation strategies through an evidence-based criteria.

Fig. 12 shows the current scenario of the time required for the library collections to become unfit for use based on the proportions of paper types in each library (Fig. 3) and the average thermo-hygrometric conditions inside the libraries (Fig. 5). Over the next 200 years, large missing pieces could occur in Ca' Granda and Delfiniana to up to 20% of their collections and in Collegio Romano to up to 40%. If considering the conservation planning horizon of 500 years (vertical dashed black line), Collegio Romano could experience the whole collection being damaged from the combination of handling and chemical decay, while the wear and tear risk in Ca' Granda and Delfiniana could affect more than a half of the collections.

Table 5 summarises the increase in the fraction of collection fit for purpose through the preservation measure scenario of the deacidification of 10% of the acidic collection with respect to the current scenario (Fig. 12). Deacidification is a conventional treatment for the restoration of acidic paper that rises the pH by removing the accumulated acid components as well as introducing an alkaline reserve that protects the material from further acid degradation. In the tested scenario, 10% of the acidic part of collections was replaced by an identical amount of paper having $\mathrm{DP}=640$ and higher $\mathrm{pH}=8.1$. Taking into account the high investments and time required to accomplish such an intervention, the expected benefit after the deacidification is low in terms of the increase in the fit-for-use fraction of the total collection if compared to the current scenario (Table 5). For this reason, if the current indoor climate conditions will be maintained, the deacidification of 10% of the collections might be not an effective preservation measure.

Finally, based on the EL definition (Equation (4)), it was possible to explore the effect of mitigation measures -such as climate control and deacidification on the resulting chemical risk for acidic paper preservation. Fig. 13 illustrates some examples of the changes in the EL values of severely deteriorated acidic paper collections (DP = 600) from the initial conditions (where EL = 68 years, at T = 30 °C and RH = 50%) to the final conditions (highlighted in bold on the y-axes), where temperature (T), relative humidity (RH) or paper acidity (pH) were changed. As shown in the graphs, a RH reduction of 20% (Fig. 13a) and a T decrease of 10 °C (Fig. 13b) are not sufficient to make EL > 255 years; deacidification up to pH = 8 (Fig. 13c) increases the EL value up to 369 years;

Table 5 Fit-for-use fraction of the total collection before (current scenario in Fig. 12) and after the deacidification of 10% of the acidic collection fraction in 200 and 500 years.

Fit-for-use fraction of collection (%) before (left) and after (right) deacidification							
Time horizon	Ca' Granda		Delfiniana		Colle	egio Romano	
200 years	83	87	79	81	64	65	
500 years	35	42	28	34	6	13	

however, only through the concurrent T and RH reduction (Fig. 13d) it is possible to reach the planning horizon of EL=500 years.

4. Conclusions

The study of the historic libraries can be addressed both to the book collections and the building itself. For the first time, the availability of long thermo-hygrometric series collected in three historic libraries located in northern and central Italy allowed us to comparatively study their indoor climate conditions to provide more insight into the preventive conservation of paper. The study provided the indoor climate characterisation of the historic buildings housing the libraries and the climate-induced risk assessment based on the chemical risks for the preservation of the library collections.

The approach developed in our study enables a wide-ranging investigation on historic libraries. Indeed, the Normalized Diurnal Index (NDR) was used to compare the natural thermal inertia of the unconditioned buildings of Ca' Granda and Delfiniana with respect to that of Collegio Romano where winter heating is active. In addition, the proposed Time Weighted Expected Lifetime (TWEL) index proved to be useful to explore the effect on paper conservation of changes in the indoor climate conditions resulting from climate control strategies, retrofitting measures and/or the possible future climate change.

The results of the investigation highlighted that the historic libraries had high thermal inertia and moisture buffering capacity due to the possible combined effect of the massive building envelopes, low air exchanges and large total volumes of hygroscopic materials (i.e., paper collections and wooden shelves). In terms of paper preservation, it was found that the monitored microclimate conditions in all the libraries would lead to the loss of their acidic collections in less than 300 years, mostly because of the high temperatures typically occurring in summer. Besides, the use of winter heating in Collegio Romano further reduced the expected lifetime with respect to that estimated in the free-floating climates of Ca' Granda and Delfiniana in the same season. For this reason, the natural unconditioned indoor climates within historic buildings could be suggested as a sustainable preservation strategy for paper collections in winter. This can be relevant for library conservation, since nowadays the increased fruition, the extensive use of heating

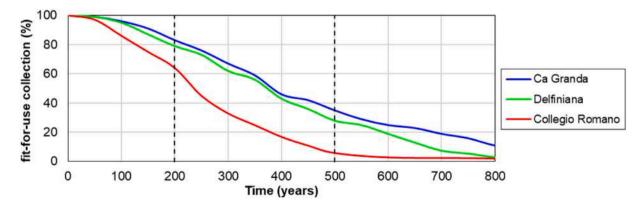


Fig. 12. Scenario of lifetime profiles over time showing the percentage of collection fit-for-use (i.e., items that can be safely handled) if stored at the current average thermo-hygrometric conditions inside the libraries. The vertical dashed lines highlight the current scenario in 200 and 500 years.

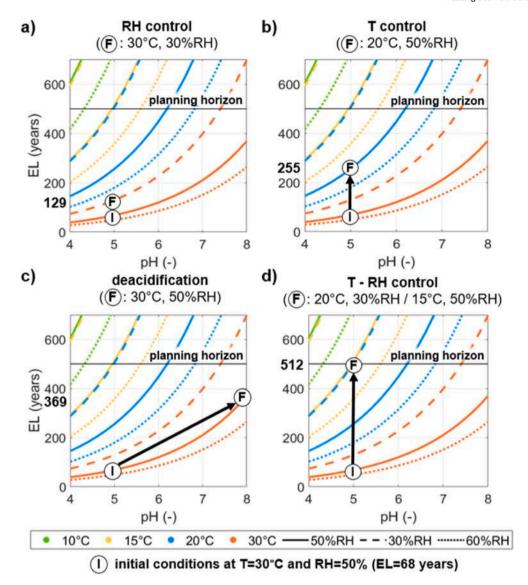


Fig. 13. Examples of mitigation strategies through T/RH reduction (a, b, d) or deacidification (c) by changing temperature (T), relative humidity (RH) or paper acidity (pH): changes in the expected Lifetime (EL) of severely deteriorated acidic paper collections (DP = 600) from the initial condition (I) at T = 30 °C and RH = 50% (EL = 68 years) to the final conditions (F).

systems and the latest climate change scenario [70] are leading the microclimate within historic buildings away from the natural one.

Some considerations about the limitations of our study are here outlined. First, the limited number of thermo-hygrometers per case study made it unfeasible to analyse spatial differences in T and RH values within each library. Moreover, the impact of the solar radiation entering the room and the air exchanges with the outdoors could not be quantitatively assessed because specific instruments for their measurements were not available. Finally, the proposed TWEL has not been experimentally validated yet, hence its value only expresses a weighted average of the chemical degradation risk based on the expected lifetime.

The above reflections stimulate further research on the conservation of historic libraries in view of the climate change predictions.

Funding sources

The APC was funded by CollectionCare project, which has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 814624.

Authors' contribution

All the authors have equally contributed to this study.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this manuscript.

Acknowledgements

Siani A.M., Frasca F. and Verticchio E. thank the CollectionCare project (European Union's Horizon 2020 research and innovation programme under grant agreement No 814624) for the financial support of the APC. Verticchio E., Siani A.M. and Frasca F. are indebted to Dr Luigi Iafrate for the invaluable support in the data acquisition at Collegio Romano. Bertolin C. is indebted to the project Climate for Culture (European Union's Seventh Framework Programme for Research under Grant Agreement No. 22 6973, Prof. Johanna Leissner and Emeritus Prof. Dario Camuffo) for giving financial support in collecting data for

the Ca' Granda (data are available to CfC partners at http://www.monumenten.bwk.tue.nl/CfC/) and to the Diocesi of Udine for having funded NTNU with a project to monitor the Delfiniana Library. Bertolin C. is indebted with Dr. Paolo Galimberti for the availability demonstrated during the monitoring campaign in the Ca' Granda in Milan; to the whole staff of the Museo Diocesano in Udine and in particular to the director Giuseppe Bergamini, the conservators Dr. Dania Nobili and Arch. Giorgio Della Longa for their continuous and kind availability and to Mr. Marco Birri for the support provided in situ during the campaign. The authors thank Prof. Branko Mitrovic for the support in providing architectural history information. The authors also acknowledge the E-OBS dataset from the EU-FP6 project UERRA (https://www.uerra.eu) and the Copernicus Climate Change Service, and the data providers in the ECA&D project (https://www.uerra.eu).

References

- H.A. Taylor, The collective memory: archives and libraries as heritage, Archivaria 15 (15) (1982) 118–130.
- [2] D. Beales, Prosperity and Plunder: European Catholic Monasteries in the Age of Revolution, 1650-1815, Cambridge University Press, 2003.
- [3] S. VanSnick, K. Ntanos, On digitisation as a preservation measure, Stud. Conserv. 63 (sup1) (2018) 282–287, https://doi.org/10.1080/00393630.2018.1504451.
- [4] M. Strlič, et al., Damage function for historic paper. Part II: wear and tear, Herit. Sci. 3 (1) (2015) 1–11, https://doi.org/10.1186/s40494-015-0065-v.
- Sci. 3 (1) (2015) 1–11, https://doi.org/10.1186/s40494-015-0065-y.
 E. Menart, G. De Bruin, M. Strlič, "Dose-response Functions for Historic Paper," Polymer Degradation And Stability, 2011, https://doi.org/10.1016/j. polymdegradstab.2011.09.002.
- [6] M. Strlič, et al., Development and mining of a database of historic European paper properties, Cellulose 27 (2020) 8287–8299, https://doi.org/10.1007/s10570-020-03344-x.
- [7] S. Michalski, Double the life for each five-degree drop, more than double the life for each halving of relative humidity, Thirteen. Trienn. Meet. ICOM-CC 3 (2002) 66–72.
- [8] D.W. Nishimura, Understanding Preservation Metrics, 2011.
- [9] M. Strlič, et al., Damage function for historic paper. Part III: Isochrones, Herit. Sci. (2015), https://doi.org/10.1186/s40494-015-0062-1.
- [10] ", ASHRAE Handbook—HVAC Applications, Chapter 24. Museums, Galleries, Archives, and Libraries, American Society of Heating, Refrigerating and Air-Conditioning Engineers, 2019.
- [11] S. López-Aparicio, et al., Relationship of indoor and outdoor air pollutants in a naturally ventilated historical building envelope, Build. Environ. 46 (7) (2011) 1460–1468, https://doi.org/10.1016/j.buildenv.2011.01.013.
- [12] M. Andretta, F. Coppola, L. Seccia, Investigation on the interaction between the outdoor environment and the indoor microclimate of a historical library, J. Cult. Herit. (2016), https://doi.org/10.1016/j.culher.2015.07.002.
- [13] E. Menart, G. de Bruin, M. Strlić, Effects of NO2 and acetic acid on the stability of historic paper, Cellulose 21 (5) (2014) 3701–3713, https://doi.org/10.1007/ e10570.014.0324.4
- [14] B. Bartl, L. Mašková, H. Paulusová, J. Smolík, L. Bartlová, P. Vodička, The effect of dust particles on cellulose degradation, Stud. Conserv. 61 (4) (2016) 203–208, https://doi.org/10.1179/2047058414Y.0000000158.
- [15] E. Vereecken, S. Roels, Review of mould prediction models and their influence on mould risk evaluation, Build. Environ. (2012), https://doi.org/10.1016/j. buildenv.2011.11.003.
- [16] Jerzy haber Institute of catalysis and surface chemistry polish academy of sciences, HERIe 2.0 (2020) [Online]. Available: https://herie.pl/. Accessed: 10-Aug-2021.
- [17] David ribar in collaboration with the heritage science lab ljubljana and UCL Institute for sustainable heritage, "collections demography app [Online]. Available: https://hsll.shinyapps.io/collections_demography_app/, 2015. (Accessed 26 May 2021). Accessed.
- [18] Uni 10586, Climatic Conditions for Storage Environments of Graphic Documents and Features of the Housings, Ente Italiano di Normazione, 1997.
- [19] UNI 10829, Properties of historical and artistic interest environmental conservation – measurement and analysis, Ente Italiano di Normazione, 1999.
- [20] Dlgs 112/98 Art. 150 Comma 6. Guideline on Technical and Scientific Criteria and Standards of Functioning and Development of Museum, 2001.
- [21] ANSI/NISO Z39.79 Environmental Conditions for Exhibiting Library and Archival Materials, "American National Standards Institute – National Information Standards Organization, 2001.
- [22] ISO 11799, Document Storage Requirements for Archive and Library Materials, International Organization for Standardization, 2003.
- [23] S. Michalski, The ideal climate, risk management, the ASHRAE chapter, proofed fluctuations, and toward a full risk analysis model, Contrib. to Expert. Roundtable Sustain. Clim. Manag. Strateg. held April 2007 (April 2007) (2007) 1–19. Tenerife, Spain.
- [24] EN 15757, Conservation of Cultural Property Specifications for Temperature and Relative Humidity to Limit Climate-Induced Mechanical Damage in Organic Hygroscopic Materials, "European Committee for Standardization, Brussels, 2010.

- [25] PD 5454, Guide for the Storage and Exhibition of Archival Materials, "British Standards Institution, 2012.
- [26] PAS 198, Specification for Managing Environmental Conditions for Cultural Collections, "British Standards Institution, 2012.
- [27] ISO 11799, Information and Documentation Document Storage Requirements for Archive and Library Materials, "International Organization for Standardization, 2015.
- [28] BS 4971, Conservation and Care of Archive and Library Collections, 2017.
- [29] EN 16893, Conservation of Cultural Heritage New Sites and Buildings Intended for the Storage and Use of Collections, "European Committee for Standardization, Brussels, 2018.
- [30] ASHRAE Handbook—HVAC Applications, Chapter 23. Museums, Galleries, Archives, and Libraries, American Society of Heating, Refrigerating and Air-Conditioning Engineers, 2011.
- [31] "ISO/TR 19815, Information and Documentation Management of the Environmental Conditions for Archive and Library Collections, International Organization for Standardization, 2018.
- [32] IFLA Principles for the Care and Handling of Library Materials, International Federation of Library Associations and Institutions, 2016.
- [33] ICCU, "Statistics on library data," Anagrafe delle Biblioteche Italiane. [Online]. Available: https://anagrafe.iccu.sbn.it/it/statistiche/statistiche-al-31-12-2020/. [Accessed: 22-Apr-2020].
- [34] A.E. Bülow, B.J. Colston, D.S. Watt, Preventive conservation of paper-based collections within historic buildings, Stud. Conserv. 47 (2002) 27–31, https://doi. org/10.1179/sic.2002.47.s3.006.
- [35] L. Tronchin, K. Fabbri, Energy and microclimate simulation in a heritage building: further studies on the Malatestiana Library, Energies (2017), https://doi.org/ 10.3390/en10101621.
- [36] E. Schito, L. Dias Pereira, D. Testi, M. Gameiro da Silva, A procedure for identifying chemical and biological risks for books in historic libraries based on microclimate analysis, J. Cult. Herit. (2018), https://doi.org/10.1016/j.culher.2018.10.005.
- [37] F. Drougka, et al., Indoor air quality assessment at the library of the national observatory of Athens, Greece, Aerosol Air Qual. Res. 20 (4) (2020) 889–903, https://doi.org/10.4209/aaqr.2019.07.0360.
- [38] K. Fabbri, M. Pretelli, Heritage Buildings and Historic Microclimate without HVAC Technology: Malatestiana Library in Cesena, Italy, UNESCO Memory of the World, Energy Build., 2014, https://doi.org/10.1016/j.enbuild.2014.02.051.
- [39] C. Balocco, G. Petrone, O. Maggi, G. Pasquariello, R. Albertini, C. Pasquarella, Indoor microclimatic study for Cultural Heritage protection and preventive conservation in the Palatina Library, J. Cult. Herit. 22 (2016) 956–967, https://doi. org/10.1016/j.culher.2016.05.009.
- [40] M. Mamoli, Towards a Theory of Reconstructing Ancient Libraries, 2014. PhD Diss.
- [41] A. Dressen, J. Connors, Biblioteche l'architettura e l'ordinamento del sapere, Rinascim. Ital. e l'Europa 6 (2010) 199–228. Luoghi, Spaz. Archit., vol. 6.
- [42] A. Kupczak, A. Sadłowska-Sałęga, L. Krzemień, J. Sobczyk, J. Radoń, R. Kozłowski, Impact of paper and wooden collections on humidity stability and energy consumption in museums and libraries, Energy Build. 158 (2018) 77–85, https://doi.org/10.1016/j.enbuild.2017.10.005
- [43] E. Verticchio, F. Frasca, F.-J. Garcia-Diego, A.M. Siani, Investigation on the use of passive microclimate frames in view of the climate change scenario, Climate 7 (8) (Aug. 2019) 98, https://doi.org/10.3390/cli7080098.
- [44] M. Steeman, M. De Paepe, A. Janssens, Impact of whole-building hygrothermal modelling on the assessment of indoor climate in a library building, Build. Environ. 45 (7) (2010) 1641–1652, https://doi.org/10.1016/j.buildenv.2010.01.012.
- [45] E. Verticchio, F. Frasca, C. Cornaro, A.M. Siani, Investigation on the use of hygrothermal modelling for paper collections, IOP Conf. Ser. Mater. Sci. Eng. 949 (1) (2020), https://doi.org/10.1088/1757-899X/949/1/012015.
 [46] F. Frasca, E. Verticchio, C. Cornaro, A.M. Siani, Performance assessment of
- [46] F. Frasca, E. Verticchio, C. Cornaro, A.M. Siani, Performance assessment of hygrothermal modelling for diagnostics and conservation in an Italian historical church, Build. Environ. 193 (2021) 107672, https://doi.org/10.1016/j. buildenv.2021.107672.
- [47] M. de la P. Diulio, P. Mercader-Moyano, A.F. Gómez, The influence of the envelope in the preventive conservation of books and paper records. Case study: libraries and archives in La Plata, Argentina, Energy Build. 183 (2019) 727–738, https:// doi.org/10.1016/j.enbuild.2018.11.048.
- [48] C.D. Sahin, T. Coşkun, Z.D. Arsan, G. Gökçen Akkurt, Investigation of indoor microclimate of historic libraries for preventive conservation of manuscripts. Case Study: tire Necip Paşa Library, İzmir-Turkey, Sustain. Cities Soc. (2017), https://doi.org/10.1016/j.scs.2016.11.002.
- [49] D. Thickett, R. Soek-Joo, S. Lambarth, Libraries and Archives in Historic Buildings, Museum Microclim. Natl. Museum Denmark, Copenhagen, 2007, pp. 145–156.
- [50] F. Coppola, N. Brown, F. Amicucci, M. Strlič, A. Modelli, Non-destructive collection survey of the historical Classense Library. Part II: conservation scenarios, Herit. Sci. 8 (1) (2020) 1–10, https://doi.org/10.1186/s40494-020-00430-y.
- [51] B. Świątkowska, Ł. Skoczeń-Rapała, D. Okrągła, M. Jędrychowski, J. Czop, Chemical Degradation and Physical Failure: Risk Analysis for a Paper Collection, vol. 3630, 2018, https://doi.org/10.1080/00393630.2018.1504450.
- [52] H.E. Beck, N.E. Zimmermann, T.R. McVicar, N. Vergopolan, A. Berg, E.F. Wood, Present and future köppen-geiger climate classification maps at 1-km resolution, Sci. Data 5 (2018) 1–12, https://doi.org/10.1038/sdata.2018.214.
- [53] M. Carlessi, A. Kluzer, Il cuore dell'antico Ospedale Maggiore di Milano: i luoghi dell'archivio e la chiesa della B.V. Annunciata, Silvana Ed., Milano, 2011.
- [54] C. Bertolin, Il ruolo del microclima nella conservazione del patrimonio librario e archivistico. Il caso della Biblioteca Delfiniana nel palazzo Arcivescovile di Udine,, in: Proceedings of the International Conference "Clima negli edifici di culto. Esperienze e prospettive, in publication, Venice, 2018.

- [55] M.C. Beltrano, S. Esposito, L. Iafrate, The archive and library of the former Italian central Office for Meteorology and climatology, Adv. Sci. Res. 8 (1) (2012) 59–65, https://doi.org/10.5194/asr-8-59-2012.
- [56] EN 15758:2010. Conservation of Cultural Property Procedures and Instruments for Measuring Temperatures of the Air and the Surfaces of Objects, Brussels, 2010.
- [57] EN 16242. Conservation of Cultural Heritage Procedures and Instruments for Measuring Humidity in the Air and Moisture Exchanges between Air and Cultural Property, European Committee for Standardization, Brussels, 2012.
- [58] F. Frasca, et al., Assessment of indoor climate of Mogila Abbey in Kraków (Poland) and the application of the analogues method to predict microclimate indoor conditions, Environ. Sci. Pollut. Res. 24 (16) (2017) 13895–13907, https://doi.org/10.1007/s11356-016-6504-9.
- [59] F. Frasca, E. Verticchio, A. Caratelli, C. Bertolin, D. Camuffo, A.M. Siani, A comprehensive study of the microclimate-induced conservation risks in hypogeal sites: the mithraeum of the baths of Caracalla (Rome), Sensors (Switzerland) 20 (11) (2020) 1–18, https://doi.org/10.3390/s20113310.
- [60] A.M. Siani, F. Frasca, M. Di Michele, V. Bonacquisti, E. Fazio, Cluster analysis of microclimate data to optimize the number of sensors for the assessment of indoor environment within museums, Environ. Sci. Pollut. Res. (2018), https://doi.org/ 10.1007/s11356-018-2021-3.
- [61] D. Camuffo, Microclimate for Cultural Heritage Conservation and Restoration of Indoor and Outdoor Monuments, Elsevier, Amsterdam, 2019.
- [62] H.E. Huerto-Cardenas, N. Aste, C. Del Pero, S. Della Torre, Effects of Climate Change on the Future of Heritage Buildings: Case Study and Applied Methodology, 2021

- [63] C. Bertolin, D. Camuffo, I. Bighignoli, Past Reconstruction and Future Forecast of Domains of Indoor Relative Humidity Fluctuations Calculated According to EN 15757:2010, Energy Build., 2015, https://doi.org/10.1016/j.enbuild.2015.05.028.
- [64] D. Camuffo, A. della Valle, C. Bertolin, E. Santorelli, Temperature observations in Bologna, Italy, from 1715 to 1815: a comparison with other contemporary series and an overview of three centuries of changing climate, Climatic Change 142 (1–2) (2017) 7–22, https://doi.org/10.1007/s10584-017-1931-2.
- [65] R. Cornes, G. van der Schrier, E.J.M. van den Besselaar, P.D. Jones, An ensemble version of the E-OBS temperature and precipitation datasets, J. Geophys. Res. Atmos. 123 (2018), https://doi.org/10.1029/2017JD028200.
- [66] M.H.J. Martens, "Climate risk assessment in museums: degradation risks determined from temperature and relative humidity data, Build. Environ. 2005 (2012) (2012) 41–44, https://doi.org/10.6100/IR729797.
- [67] F. Cappitelli, et al., "Chemical-physical and microbiological measurements for indoor air quality assessment at the ca' granda historical archive, Milan (Italy)," *Water*, Air. Soil Pollut. 201 (1–4) (2009) 109–120, https://doi.org/10.1007/ s11270-008-9931-5.
- [68] R.P. Kramer, H.L. Schellen, A.W.M. van Schijndel, "Impact of ASHRAE's museum climate classes on energy consumption and indoor climate fluctuations: full-scale measurements in museum Hermitage Amsterdam, Energy Build. 130 (2016) 286–294, https://doi.org/10.1016/j.enbuild.2016.08.016.
- [69] H. Derluyn, H. Janssen, J. Diepens, D. Derome, J. Carmeliet, Hygroscopic behavior of paper and books, J. Build. Phys. 31 (1) (2007) 9–34, https://doi.org/10.1177/ 1744259107079143.
- [70] IPCC, Assessment Report 6 climate change 2021: the physical science basis [Online]. Available: https://www.ipcc.ch/report/ar6/wg1/, 2021.