Reviews in Conservation 7, 2006, pp. 27-41.

Indoor air pollution in museums: a review of prediction models and control strategies

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Abstract

The calculation of pollution mass balances is explained with reference to modelling indoor air quality in museums, and the implication of pollution deposition on building interior and museum objects is discussed. A short overview of the museum environments key pollutants and their effect on materials are given. Case studies, which compare buildings with and without air-filtration, old and new buildings, or buildings in urban and rural areas, suggest that pollution mainly infiltrates buildings via free air movement and that passive control measures maybe sufficient to exclude outdoor pollutants. What this means for the level of indoor generated pollutants is not clear. Other studies have shown benefits from the use of active air-filtration, especially in urban areas with high pollution levels. The use of dosimeters rather than concentration measurements is now in focus in conservation research. Different approaches to setting acceptable limits for air pollution are briefly discussed, including the establishment of 'adverse effect levels'.

Introduction

In late 2005 a comprehensive report was published on the condition and preservation needs of cultural heritage collections in the United States [1]. From a survey representing more than 30,000 institutions it was concluded that the most urgent need of US institutions is environmental control, and it was found that 47% of the responding institutions have had 'some' or 'significant' damage to their collections due to air pollution. However, air pollution is not a new museum phenomenon. In 1850 Eastlake, Faraday and Russell described the effect of air pollution on oil paintings in the National Gallery, London [2]. The special problems from indoor generated pollutants, e.g. emission from wooden storage or display materials, was noticed first by Byne in 1899 as he described corrosion damage to sea shells, however, Byne failed to link the damage and the poor storage conditions [3]. However, this was done a few decades later by Scott [4], and Nicholls [5], who described in detail the deterioration of lead medals housed in oak cabinets, and Mollusc shells in oak drawers, respectively [se also 6]. More recently, chapters on air pollution in Thomson's book "The Museum Environment" [7], and texts such as the 1982 paper "Trouble in store" by Padfield *et al* [8] increased the attention to air pollution problems among conservators.

One of the most cited articles on air pollution and museums is the 1990 review by Brimblecombe [9]. In his conclusion Brimblecombe listed the following issues as directions for future work (extract):

- *Monitoring of air in museums:* at the time few field studies, and need for development of small and unobtrusive, yet sensitive methods. Passive sampling seemed promising.
- *Emission from display materials:* corrosive compounds and emission factors not well known.
- *Deposition velocities:* frequently unknown for important pollutants onto common materials.
- *Chemical transformation:* the oxidation of formaldehyde to formic acid and other important reactions had not been investigated.
- *Acceptable concentrations and mechanisms for damage:* little was known of pollution critical levels below which the damage rate to cultural objects falls to acceptable values.

In the present article the main focus will be on recent literature published since the review by Brimblecombe.

Indoor air pollutants

Key pollutants

There seems to be consensus in literature as to which compounds should be considered key pollutants [e.g. 7,10-12]. From outdoor sources these are: sulphur dioxide (of oxidized sulphur gases), nitrogen dioxide (of nitrogen oxides), ozone, and hydrogen sulphide (and other reduced sulphur gases, which may have both outdoor and indoor sources). From indoor sources acetic acid (of carbonyl compounds) is the primary pollutant. Tétreault [12] also includes water vapour as a key pollutant, acknowledging that incorrect relative humidity relates both to chemical, physical and biological material damage, and affects the deterioration processes caused by most other air pollutants. It must be noted that also fine particles are considered a key pollutant. However, to limit the extent of this review only gaseous air pollutants will be discussed.

Effect on materials

In general, pollution compounds, their sources, and effect on materials are known. There are numerous reports in literature on indoor air pollutants and material damage, for example: carbonyls and corrosion of lead [13-16], copper [16], or bronze [17]; carbonyls and the degradation of paper [18]; reduced sulphur compounds and tarnishing of silver [19,20]; sulphur dioxide and/or nitrogen oxides and the degradation of leather [21], paper [22-26], or dyes [27]; peroxides and discolouration of photographs [28,29], ozone and the degradation of rubber [30], or fading of dyes [31-33]; and carbonyls and salt efflorescence on calcareous materials [34-38], or glass [39]. General reviews of air pollution and material damage were previously given by Thomson [40], Baer and Banks [41], Brimblecombe [9] and Graedel and McGill [42]. A thorough description of general air pollution physics and chemistry is available from environmental text books by, for example, Brimblecombe [43], Seinfeld [44], or Wadden and Scheff [45].

Pollution levels
To illustrate the typical range of air pollution in museum buildings table 1 and 2 show examples of ozone and nitrogen dioxide measurements from building studies. The trend for ozone (table 1) is that concentrations typically are below 30 ppb, more than half of the reported ozone concentrations in table 1 are even below 10 ppb. However, a few reported short term incidents had higher concentrations, up to 143 ppb. These high incidents all take place in buildings with a high air exchange rate (e.g. mechanical ventilation) but no chemical air-filtration. The matching indoor/outdoor (I/O) concentration ratios for ozone show the same trend, with the typical I/O ratio below 0.70 (however, a few as high as 0.84), and half of these I/O ratios even below 0.20.

For nitrogen dioxide (table 2) all reported concentrations are below 25 ppb. The typical I/O ratio is in the range of 0.60-0.80. There are exceptions: at sites with a low air exchange rate, or with chemical air-filtration, the I/O ratio can be less than 0.10. At one place the I/O was >1 which hints an indoor generation of nitrogen dioxide.

It should be noted that tables 1 and 2 are not fully comprehensive in respect of all literature reports; however, they show the trends. For example, the summer and winter data from the Correr Museum (table 2) shows that infiltration of outdoor pollutants is high where free air movement is possible (through open windows etc.) and that the indoor level therefore is highly dependent on air exchange rates as well as on the outdoor concentration level. The highest I/O ratios are found in summer, when windows and doors tend to be kept open during museum gallery opening hours.

Compared to the pollutants which infiltrate from outdoors, the indoor generated pollutants can be found in much higher concentrations. This is especially true where there is a strong source (e.g. emission from new building materials), and where the air exchange rate is low. Not surprisingly extremes are found in display cases and other confined spaces. Reported concentrations of carboxylic acids from rooms and display cases are shown in table 3.

Quantification of air pollutants is becoming trivial. Sampling methods for most gaseous pollutants are made available, either by so-called passive sampling (by diffusion) [e.g. 46-52], or by active sampling (pump-driven) as used e.g. by [53-55]. What is left for the practising conservator, after the initial decisions of ‘why’ and ‘how’ to monitor for air pollutants, is to understand the results and be able to act upon them, as discussed e.g. by Blades [56].

Modelling air quality in buildings
The general behaviour of air and its rate of exchange in confined spaces was explained by, for example, Padfield [80], Thomson [81], and Michalski [82], as well as Brimblecombe who explained air pollution behaviour also in confined air volumes [83]. These four papers all related theory to museum environment situations. Prediction models for indoor air pollution covers a wide field, from the simple “100,10,1” rule-of-thumb by Tétreault [12, p.35] (that an outdoor air pollutant will typically be present as 10% indoors, and 1% in display cases, compared to an outdoor concentration of 100%), to other quite detailed models. Studies of air pollution behaviour in buildings, and of the

sorption properties of specific building materials, have resulted in several indoor air quality prediction tools.

Steady-state I/O ratio
With a deposition mass balance Weschler *et al* [84] described the steady-state indoor/outdoor (I/O) relation of ozone in buildings:

$$\frac{C_i}{C_o} = \frac{n}{n + v_d\left(\frac{A}{V}\right)} \qquad (1)$$

Where: C_i = indoor concentration of pollutant [ppb or μgm^{-3}]
C_o = outdoor concentration of pollutant [ppb or μgm^{-3}]
n = air exchange rate [h^{-1}]
v_d = deposition velocity [mh^{-1}]
A = inside surface area of room [m^2]
V = volume of room [m^3]

This much cited model provides, within its limitations, a good approximation of real life situations. It assumes no indoor pollution sources and only takes surface reactions into account, neglecting any reactions in air. Furthermore, it is assumed that the surface reactions irreversibly remove pollutants from air, and finally it is assumed that indoor air and pollutants are perfectly mixed. However, taking this into account the model works well with pollutants such as ozone and sulphur dioxide and to a certain degree also nitrogen dioxide. The mass balance in equation 1 was used as the basis of the IMPACT model, which was developed during the EC project 'Innovative Modelling of Museum Pollution and Conservation Thresholds' (EVK4-CT-2000-00031) [85]. The model estimates the indoor concentration and deposition of pollutants originating from outdoors. It works for one zone (one room) only, and the I/O ratio therefore refers to inside and outside this zone. For a room located in the interior of a building the "outside" will often be the surrounding rooms rather than the outdoors. The model is available on the Internet as a Java Applet at <http://www.ucl.ac.uk/sustainableheritage/impact/> (accessed 1 September 2006).

Deposition velocity and surface removal rates
The deposition velocity is a central factor of Weschler's model above. It is defined as the flux of a pollutant to a surface divided by its concentration in air, and is a mass transfer coefficient with the unit of velocity. A detailed discussion of the deposition velocity concept is given by Nazaroff *et al* [86]. For highly reactive pollutants such as ozone, removal by surface reactions makes a significant part of the total pollution loss indoors, compared to removal by air exchange. It is possible to determine the average deposition velocity of all surfaces in a room from equation 1, as well as the so-called 'surface removal rate': $v_d(A/V)$. This rate is directly comparable to the air exchange rate: if, for example, a room has the surface removal rate of 2 h^{-1}, then pollutants will react with the indoor surfaces in an amount equal to what would be ventilated away at 2 air changes per hour. The deposition velocity of a particular pollutant varies between

different material types and under different conditions such as changing relative humidity. Compiled data on ozone, sulphur dioxide, and nitrogen dioxide to a number of typical materials have been published over the years [87-93]. From building studies the reported values of surface removal rates for ozone in rooms have been in the range of about 1 to 8 h^{-1}, depending on room size and whether the interiors were made from inert surfaces such as glass or steel, or from reactive materials such as textile [94 and references herein]. For nitrogen dioxide a study of museum buildings showed a surface removal rate of 4.5 h^{-1} for a storage room, compared with 0.4 h^{-1} for a large and open gallery [78]. Table 4 shows a selection of surface removal rates calculated for ozone in a range of American museums. For comparison also a few values from domestic or work rooms are shown.

General pollution mass balances

The mass balance in equation 1 is for steady-state only. Generally, a transient-state mass balance can be described for indoor pollutant's concentration change over the time *dt*:

$$V\frac{dC_i(t)}{dt} = G + C_oQ_v - C_i(t)Q_v - C_i(t)Q_s \qquad (2)$$

Where: G = indoor generation of pollutants [$\mu g h^{-1}$]

Q_v = ventilation flow rate (equals $n \times V$) [$m^3 h^{-1}$]

Q_s = sorption flow rate (equals $v_d \times A$) [$m^3 h^{-1}$]

t = time [h]

The general solution to equation 2 is:

$$C_i(t) = C_o e^{-(Q_v+Q_s)\frac{t}{V}} + \frac{C_oQ_v + G}{Q_v + Q_s}(1 - e^{-(Q_v+Q_s)\frac{t}{V}}) \qquad (3)$$

The mass balance from equation 1 is a simplified case of equation 3, derived from steady-state conditions ($t \rightarrow \infty$) with no indoor pollution generation.

Influence of chemical reactions in air

If reactions between compounds in air should make any significant influence on the indoor pollution level, then the rate of reaction must be equal to or faster than the rate of removal by air exchange. For example, for ozone the fastest reaction in air will happen with nitrogen oxide, where the half-life of 50 ppb ozone is as low as half a minute. This reaction produces a new compound of concern; nitrogen dioxide, which itself may react again with ozone producing the nitrate radical, which in the absence of high light levels indoors will accumulate and end up converted into nitric acid [94,95]. Under the right conditions this generation rate of nitrogen dioxide can be high; in a large art gallery the

indoor level of nitrogen dioxide has been reported to exceed that of outdoors (I/O about 1.3) [70]. At the same time surface reactions with moisture will remove nitrogen dioxide from the air, converting it into nitrous and nitric acid. The nitric acid will remain in the surface water film, while the nitrous acid will form an equilibrium between liquid and gas phase [95,96], in which about 98% of the nitrous acid will be in the air [97,98]. This can lead to high indoor concentrations of nitrous acid, which when measured in the air will hint that a similar amount of the damaging nitric acid is deposited on the interior. Inside an urban museum gallery and a church both in Italy, the indoor levels of nitrous acid has been found to be even higher than outdoors [59,69].

The implication of this is that while nitrogen oxide and nitrogen dioxide themselves are not especially harmful to materials, and in fact even act as an ozone sink, they can, when infiltrating buildings, end up as nitric acid on interior surfaces. In a survey of five museums in California, USA, the deposition of inorganic nitrate (from gaseous nitric acid) onto vertical surfaces was measured to be between 0.18-5.82 $ngm^{2}s^{-1}$, with the highest deposition rate in a historic house with a high infiltration rate of outdoor air [99].

The I/O mass balance in equation 1 does not take reactions in air into account, which is the main limitation of that model. For the oxides of nitrogen, and some reactions between ozone and organic compounds, the air chemistry is in fact a significant sink. However, this can be incorporated into the general mass balance from equation 2:

$$V\frac{dC_i(t)}{dt} = G + C_oQ_v - C_i(t)Q_v - C_i(t)Q_s - C_i(t)\sum_i k_iC_{chem-i} \qquad (4)$$

Where:

$C_{chem\text{-}i}$ = concentration of the *i*th (1st, 2nd, 3rd, etc.) chemical in the air, which reacts with pollutant [μgm^{-3} or ppb]

k_i = 2nd order rate constant for the reaction between *i*th chemical and pollutant in the air [$ppb^{-1}h^{-1}$ or $\mu g^{-1}m^{3}h^{-1}$]

This expression is, for example, given by Weschler [94]. A graphical outline of the mass transfer pathways in a museum room is shown in figure 1, including ventilation, emission, air chemistry, and sorption mechanisms.

Other indoor air quality models

Nazaroff and Cass developed a prediction model which took both reactions in air and on surfaces into account, as well as ventilation, filtration, and indoor sources [63]. Both photolytic and kinetic reactions in the air are accounted for. The model was applied to measured data from a museum building, and it was found that reactions in air were an appreciable sink. The Nazaroff and Cass model has since been applied to the study of cultural heritage buildings in the USA [64], and in Greece [100].

The indoor generated pollutants are the most difficult to model, as the source strength is rarely known and is difficult to estimate because the emission will originate from a mixture of many different objects and materials. The conservation field lacks good tools which take these internally generated pollutants into account, the same way the IMPACT model covers outdoor generated pollutants. However, advanced multi-zone modelling software does exist for human comfort studies, some of which, with some adaptation, may be of use also for conservation research. One example is 'CONTAM' of the US National Institute of Standards and Technology (NIST) [101].

Mitigation of air pollution in buildings

Existing guidelines

Several guidelines on pollution control in museum buildings have been published during recent years. Blades *et al* gave an overall introduction to air pollutants, their monitoring and control, with case study examples, in their 2000 guide [10]. In 2002 Hatchfield reviewed air pollutant types and their sources, and gave a comprehensive introduction to construction materials for display and storage, and how to test such materials for the possible emission of harmful vapours [11]. Since 1999 the American Society of Heating Refrigerating and Air-conditioning Engineers (ASHRAE) included a chapter on air quality for museums, libraries and archives in their Applications Handbook, the latest edition (2003) being much updated with regard to pollutant sources and threats to museum objects [102]. At the Canadian Conservation Institute a general framework for the preservation of museum collections was developed. In a chart matrix nine 'deterioration agents' (including air pollutants - 'contaminants') were listed together with five 'stages of control' by which they can be fought: *Avoid, Block, Detect, Respond,* and *Treat.* The general concept was described by Michalski [103], and implemented into Tétreault's book on risk assessment, control strategies, and preservation management for air pollution in museums [12].

Control strategies

The central point in pollution control is to avoid or at least limit the mass flow of air pollutants toward the objects that are being protected. There are different approaches to achieve this, their effectiveness depending of many factors, including the location of the object (exhibition, storage) or the type of building in question. Pollutants in outdoor air can be removed from the air while it enters a building by the use of chemical filtration in a ventilation system. At the same time this provides the possibility of diluting internally generated pollutants, by removal through the ventilation exhaust. Another strategy for reducing the mass flow of outdoor pollutants is to keep the building as air tight as possible, which decreases the air exchange rate and by this the pollution infiltration. In areas with staff or visitor activity, this may conflict with the comfort demands of people; however, for storage areas this strategy may be feasible. The transport of outdoor air pollution to the indoors will be greatly decreased by the blocking by the tight building envelope as well as by removal via sorption mechanisms indoors. However, from studies of display cases it is well known that low air exchanges may provide problems with build-up of high levels of internal generated pollutants. For small display cases the situation is extreme because of a high surface to volume ratio. As this ratio decreases for bigger volumes this problem may be less significant at room or building scale. If pollutants in the ambient environment can be avoided in the first

place, then this is an excellent starting point. In general the pollution level is higher in urban areas than in rural, but local variations must be taken into account; the vicinity of roads, industry, or farms may considerably influence the local air quality.

Ventilation and filtration
The use of mechanical ventilation is one traditional means of air pollution control. By forcing the supply of air through filters and by controlling its pathway through the building, clean air can be directed to rooms or zones as desired. Mechanical ventilation can also create a slight over-pressure indoors, which minimizes uncontrolled infiltration of outdoor air. Studies from urbanized areas with high outdoor pollution levels have shown that a considerable improvement to the indoor level of air pollution is possible with chemical air-filtration, e.g. by the use of activated carbon. This has been exemplified by ozone investigations in various museum buildings in California, USA [31,64], and by pollution measurements in different areas within the same building with or without air-filtration, for example in museums in the UK and Ireland [66-68]. From model and field studies of buildings housing delicate electronics such as telephone switching equipment, it has been predicted that the benefit of improving the air quality, thus decreasing the failure rate of the equipment, was greater than the cost of the increased operation time and maintenance used on the ventilation and filtration systems [104]. This last study focussed on fine particles only; however, analogies can be made for gaseous pollutants. And it would be possible to carry out similar cost/benefit analysis for the balance between the costs of filtration against the future cost of conservation in a museum.

Other studies have not given the same unambiguous results. Cassar *et al* [77] compared the indoor air pollution inside two museum buildings, both located in highly polluted areas of London: The Museum of London, with air-conditioning and air-filtration, and Bethnal Green Museum, a naturally ventilated building with a low air exchange rate. It was concluded that although the Museum of London's air-filtration system did control the indoor air quality better than the performance of the building's structure alone at Bethnal Green, the benefit was not that great. The authors speculated whether sensitive objects, which normally are protected inside display cases would benefit much from air-filtration at room level, as cases will largely reduce external pollutants anyway. This is similar to conclusions made by Salmon *et al* [62]; from a study of five museums in central Cracow, of which one had air-filtration, it was suggested that in order to lower the ozone level in the vicinity of exhibited museum objects, the use of display cases would give sufficient protection. As such cases would be expected to lower the inside ozone concentration 90% compared to the surrounding room, the ozone level would drop below 1 ppb around the objects. In general it was recommended to keep the air exchange rate low in the galleries at all sites, in order to minimize the ozone infiltration from outdoors.

The use of re-circulation of the indoor air through chemical filters is a special case of mechanical ventilation, which has not been treated as a separate issue in conservation literature; however, it is mentioned as one possible strategy among others in a couple of building studies [62,78]. Re-circulation of air within a building may be preferred over the intake of outdoor air, as this will reduce the cost of controlling the temperature and relative humidity indoors. At the same time not only the outdoor pollutants will be

removed by filtration, but also the internal generated pollutants. However, lowering the ventilation rate may conflict with human comfort demands [see also 105, and 106, p. 107-108]. Modern HVAC systems normally give the opportunity of combining re-circulation with a fraction of new air intake.

Sorption on building materials
The studies [62,77] hint that for some situations a more passive control strategy will be feasible. The fact that air pollution will be ab- or adsorbed by interior surfaces and thereby removed from the air can be used deliberately for air pollution control. In order to be effective the air exchange rate must be low, for which reason this control strategy will mainly be successful in building areas with a low daily activity, such as storage rooms.

In a study by Blades *et al* [78] a number of passive and active control strategies for urban air pollution control in museum buildings were investigated. This was done in five museums in the UK, in buildings with or without air-conditioning and air-filtration. It was found that the outdoor pollution level could vary much around a building, depending, for example, on the vicinity of roads. Therefore it was suggested as a general strategy to seal buildings on the most polluted sides, and to ensure that the air intake was from the least polluted side. Another passive control strategy suggested in this study was to exploit the pollution sorption properties of the building fabric. Of active control strategies the use of filters was discussed. Illustrated by the nitrogen dioxide I/O ratios from the museums galleries it was shown that when the air-conditioning system had air-filters installed the I/O ratio was about 0.2 or less. However, by removing the filters from the same systems, or comparing with sites with no air-filtration, the I/O ratio was higher; about 0.5-0.9. It was suggested that the intake of outdoor air should be minimised by re-circulating a larger fraction of the indoor air through filters instead. While the authors do not say so directly, it seems that they would advise passive control strategies before active. Both the capital and the operational cost of air-conditioning were mentioned as weighty expenses, and it was mentioned that in future cost-benefit assessments of pollution control sustainability should also be included as a factor. This is much in line with studies on heat and humidity buffering of museum buildings, where it has been found that for large areas of the temperate zone of the World it is possible to design buildings where the indoor climate can be controlled by passive means. Natural ventilation and intelligent use of the buildings structure and fabric may provide an indoor climate, which from a conservation point of view, is no worse than those in buildings with air-conditioning [107,108].

Within the EC-project ‘Assessment of Environmental Risk Related to Unsound Use of Technologies and Mass Tourism’ (ENV4-CT95-0088), four museums in different parts of Europe with different local climate and pollution conditions were investigated; one modern, and three traditional buildings [70-73,109]. One aim of the study was to describe the effect of different design factors and construction materials on the indoor climate and air quality. It was found in all studies that the traditional buildings performed better both with regard to reducing outdoor pollutants in the indoor environment and to climate control. It was stated that museums in traditional buildings benefit from a long term building tradition and knowledge about the use of structures and materials which naturally stabilize the climate and yield deposition surfaces for air

pollutants. This is not necessarily so in modern buildings, which are often made from relatively inert materials such as glass, metal, and polymers. As it was found that the infiltration of air pollutants was mainly driven by free air movement the authors advocated the use of passive control: insulation, shielding, only backed-up by air-conditioning when absolutely necessary.

How passive air quality control influence the air quality with regard to the indoor generated pollutants is not well documented on room or building scale. During an investigation of about 250 sites within 17 American and seven European museums, carbonyl compounds were never found in concentrations above 100 ppb in the room air of galleries and stores [46,79]. The air exchange rates of the sites were not reported, but presumably none of the buildings were designed to be especially air tight or controlled by passive means.

Variations in ambient pollution levels

Major cultural heritage institutions are often located in urban areas, which is something that must be accepted. However, if less polluted areas for e.g. storage facilities can be chosen, this will be an effective control strategy. This was illustrated in the European ENVIRONMENT Leather project (EV5V-CT94-0514) [21,75,76], where the effect of the environment on book bindings was studied. This was done based on the analysis of two identical sets of leather-bound books; one placed in the British Library in the highly polluted centre of London, and the other in the much cleaner area of Aberystwyth at the National Library of Wales, both since 1932. The dosage of sulphur dioxide and nitrogen dioxide, which the books in the two libraries received over 60 years, was estimated from contemporary measurements, and from historical air pollution data. In London the outdoor sulphur dioxide concentration decreased from around 150 ppb in the early 1930s to below 20 ppb in the early 1990s. Despite similar I/O ratios at the two library buildings, the exposure to the books in Aberystwyth was only 5% of that in London. The preservation state of the books was assessed from measurement of the accumulated sulphate in the leather, its pH, and from visual inspection. In London the book bindings was substantially more deteriorated than in Aberystwyth, where the leather generally was in an acceptable state. From this it was determined that with a preservational aim of no more than 0.5% sulphate accumulation (2.5 $g(SO_4)/m^2$) within one century, the average sulphur dioxide concentration should not exceed 0.06 ppb. Besides the illustration of the decreasing sulphur dioxide levels in urban Europe, the ENVIRONMENT Leather project is one of the few studies which convincingly have estimated acceptable levels of accumulative exposure to air pollutants for cultural heritage objects.

As the outdoor pollution picture has changed over time, the main focus has changed from sulphur compounds originating from the burning of coal (at least in Western cities), toward the pollutants and their secondary compounds originating from car traffic [110]. Likewise within conservation; whereas earlier texts mainly dealt with sulphur dioxide [e.g. 7,66], today nitrogen dioxide and/or ozone are the main compounds of concern. However, as the ENVIRONMENT Leather project showed the accumulation of the past does ad up and remains as damage already done. Finally it must be noted, that not only the ambient conditions influence the indoor environment: Ryhl-Svendsen *et al* analysed the microclimate in two historic buildings in Spain and Denmark [58].

They found that the structure of the room, the objects, and the curatorial decisions in combination influenced the indoor climate and air quality to such a degree that they could vary more within one building than between the investigated buildings located at either end of Europe.

Using mass balance predictions in building studies

The practical use of the pollution mass balances previously described requires that a number of parameters within the space of investigation must be known. In the simplest case these are the concentration of a pollutant inside and outside the space, its volume and interior surface area, the rate of air exchange, as well as the pollutant's deposition velocities. The latter has been made available for a number of pollutants and material types, as mentioned above [e.g. 93], or can, if being the only unknown, be estimated from equation 1 as a room average. By establishing the pollution mass balance of a museum gallery or storage room, the mass budget will reveal where the pollutants end up. The size of the surface loss is the key parameter here, as the pollutant-surface reactions include not only sorption on building parts, but also deterioration processes on museum objects. Mass balance calculations will also show the effect from changes in ventilation rate and other interventions.

This leads to an interesting aspect of this approach. While the deposition of pollutants on museum objects as such is regarded as a deteriorative mechanism, the effect may be effectively minimized by the dilution due to deposition to large surface areas. A case in point, while a few silver objects in a large room may tarnish quickly, that air may be almost harmless to the objects if the same room is completely filled with silver objects (all other things being equal). However, reality is more complex than this, and the strength of the pollution source is particularly important. The above argument should never be used for avoiding air pollution control measures, unless the present case in question is thoroughly analysed. The use of the sorption potential of inner surfaces in museum rooms is an interesting approach to air quality control. Deliberate use of reactive wall materials may prove to be a feasible method for passive pollution control, much similar to passive humidity control [e.g. 111]. More research and field studies on this subject are needed, as until now it has only been investigated in a few studies, none of which focused on museum environments [112-114].

Acceptable pollution levels

Concentration or dose?

The modelling studies and building investigations lead toward a central question in museum air quality studies: while it is concentration which is measured or modelled, how does material damage relate to this? Air pollution monitoring is traditionally expressed in concentration (or average concentration), however, dosage relations are revealed from material studies, e.g. for lead and acetic acid [14]. The use of environmental dosimeters has gained much attention in conservation. Currently most of these are at the prototype level [115-119]; however, commercial products based on corrosion measurement on silver and copper plated sensors are also available [120,121]. The International Organization for Standardization has recently published standards on the evaluation of indoor atmospheres based on metal corrosion measurements [122,123]. The dosimetry approach raises the question on how allowable pollution limits should be defined. The existence of true thresholds may be argued by

thermodynamics; however, calculations show these to be improbably low [124]. Brimblecombe [125] reviewed how standards for air pollution and museum objects must differ from those applicable to human comfort and health, and suggested two approaches: An 'operational threshold'; the concentration where the rate of deterioration from the pollutant becomes less significant than deterioration by other mechanisms, or; 'the concentration where survival time is sufficient'. Although expressed in concentration, in reality this is a practical way of expressing a dosage. In opposition to recommendations based on 'best technology' [e.g. 126], Tétreault [127] argues that allowable limits should evolve from the ongoing collection of empirical and experimental data ('best knowledge'). Tétreault also introduced the 'no' or 'lowest-observed adverse effect level' (NOAEL and LOAEL) approach to conservation, originally a concept used in toxicology.

Adverse effect levels

The 'no-observed adverse effect level' defines the level at which damage is not observed for a specific setup. In principle it has already been applied to material studies for some years; as mentioned previously a level of 0.06 ppb sulphur dioxide over 100 years was considered safe for leather [76], and for silver and hydrogen sulphide the LOAEL (at which tarnish was just visible) was determined to be 0.385 μgm^{-3} (0.272 ppb) over 10 years [20]. The NOAEL approach was included in the 2003 ASHRAE guideline [102], of which the guideline's air pollution targets are reproduced here in table 5. A thorough discussion of the NOAEL and LOAEL approach for indoor museum environments is given by Tétreault [12]. The concept has been widely discussed in recent years [127-129].

In a recent paper Brimblecombe [130] discussed the complexity of establishing standards on air quality for cultural heritage. One example was: the level of formaldehyde in air is not a decisive factor for lead corrosion, but the potential that it will oxidize into formic acid is. However, still the concentration of formaldehyde will be the typical choice of measurement [see also 131]. Tétreault [127] warned that, when establishing and using NOEAL values, premature conclusions may be drawn due to incomplete information on all parameters which determine the damage rate. He listed that a useful syntax for NOAEL would include *NOEAL = Concentration (temperature, relative humidity, time, property measured)* where 'property measured' includes the nature and history of the object. However, such detail on the classification of the aggressiveness of indoor climate is yet to be seen in the studies reported in the conservation literature. To establish dose-response functions may be complicated but by no means impossible. For outdoor environments, both synergy effects and complex dose-response functions have already been determined for the wet and dry acid deposition on metals and stone [132-135].

Conclusion

Returning to Brimblecombe's 1990 list of directions for future work [9], we can see that the use of passive sampling for monitoring is widespread today [e.g. 52]. Deposition velocities for several key pollutants onto common materials are known [87-93], and the problem of emission from display materials is continuously being investigated [e.g. 11]. Some work on chemical transformation has been carried out, indeed on formaldehyde oxidation [131], and on the nitrogen oxide chemistry [59,69,70,94-99]. The latter

deserves continuous attention in research in order to further establish the importance of nitrous and nitric acid formation in museum environments.

From a practical point of view, the simplicity of e.g. the IMPACT model and its Internet based platform makes it useful for predicting a qualified estimate of the indoor air quality of a building [85]. For investigations involving the more complex chemistry between pollutants, a more detailed mass balance must be set up. Mass budget calculations are useful for establishing the pathways for air pollutants in locations where cultural heritage objects are at risk. The use of reactive building surfaces for removal of air pollutants is an interesting option, and should receive more attention in future research.

One part of conservation literature holds the view that passive ventilation provides sufficient protection from outdoor air pollution for most museum building types [70-73,77,78,109]. This is based on measurements in real buildings; however, none of these investigations included indoor-generated pollutants such as carbonyls. Other studies showed that in areas with high outdoor pollution levels, air-filtration does indeed provide an efficient means of pollution control, even for dynamic environments such as exhibition galleries [31,64,66-68,104]. Generally, comparative studies where indoor air pollution models are tested against real cultural heritage buildings are still lacking.

Acceptable pollution levels are still widely debated. The turn from solely focusing on concentration toward dose functions is illustrated by the increasing interest in dosimetry. Although theoretical thresholds may be established using thermodynamics or kinetics, the current approach is toward operational limits based on pollution levels at which no or low but acceptable damage is observed for a certain length of time. However, dose-response functions for materials in indoor environments are an issue which still needs much investigation. Except for the latest ASHRAE guideline [102] we are still waiting for recommendations or standards which are not solely given as concentration values.

Acknowledgements
The author thanks Geo Clausen, International Centre of Indoor Environment and Energy at the Technical University of Denmark, and René Larsen, School of Conservation at the Royal Danish Academy of Fine Arts, for useful discussions throughout the writing of this article.

Glossary of terms
Please note: some of these terms may have a wider definition than given here within other fields, such as human comfort studies.

Air-conditioning: Heating, cooling, humidification, or dehumidification of indoor air, in order to maintain either a comfortable environment, or to meet specific preservation targets.

Air exchange rate: The rate at which ambient air replaces the indoor air in a building or room. Expressed as the number of changes of outside air per unit of time, e.g. per hour (h^{-1}).

Air-filtration (chemical air-filtration): removing pollutant gasses and/or particles from air by forcing it through a porous filter media. Gasses are withheld by sorption.

Ambient: The environment in the vicinity of a building ('outdoors').

Concentration: The volume or mass of a pollutant per unit of volume air. Units may be expressed as ppb ('parts-per-billion') or μgm^{-3}. The relation between the two units is at room temperature: $[ppb]=([\mu gm^{-3}]\times 24.04)/M_w$, where M_w = the molar weight of the pollutant. In this paper all units have been converted to ppb for the ease of comparison.

Deposition velocity: The flux of a pollutant to a surface divided by its concentration in air. Expressed in the unit of velocity [ms^{-1}].

Dosage: The amount of a pollutant deposited on a surface. Defined as concentration × time × deposition velocity. Expressed as mass per unit of surface area, e.g. μgm^{-2}.

Emission: The release of compounds from a material (e.g. organic acids from wood), or during a process such as combustion. Material emission is sometime named off-gassing. Expressed as the mass release pr unit of time, e.g. mgh^{-1}

Exposure: The time which a material is exposed to a pollutant multiplied by the pollution concentration. For example expressed as ppb × years

HVAC: Heating, ventilation, and air-conditioning system

Infiltration: Uncontrolled flow of ambient air entering a building or room, through leaky windows etc.

I/O ratio: The ratio between the concentration of a pollutant inside a room (or building) and in the ambient air. Dimensionless.

Mechanical ventilation: Exchange of air driven by a motorized fan. May or may not include other system components such as air-conditioning and air-filters.

Natural ventilation: Controlled air exchange driven by convection, created by deliberate use of the building structure.

Pollutant: A compound, which is unwanted in the museum environment because of its ability to damage materials. Primary pollutants are directly emitted from a source (see emission) indoors or outdoors. Secondary pollutants are the product between reactions of primary pollutants, e.g. the formation of nitric acid from reactions between nitrogen oxides and ozone.

Sink: A material or process that acts as a temporary storage or absolute removal mechanism for pollutants, for example; fleecy textile surfaces are large ozone sinks as they effectively remove ozone from air by sorption.

Sorption: The taking up of one compound by another. May be either absorption where a compound penetrates into the pores of a solid and are held either chemically or physical, or adsorption where the compound adhere on a solids surface, e.g. in a liquid film. If the conditions are in favour of releasing the compound back into the air, this is called desorption.

Steady-state concentration: The resulting pollution concentration in a room from the balance of all generation and removal mechanisms in that room.

Threshold: Concentration at which a reaction is not occurring, or is happening so slow that it is non-observable within the limits of observation (relies on reaction kinetics and thermodynamics).

Transient-state concentration: Concentration at a given time, under non steady-state (changing) conditions.

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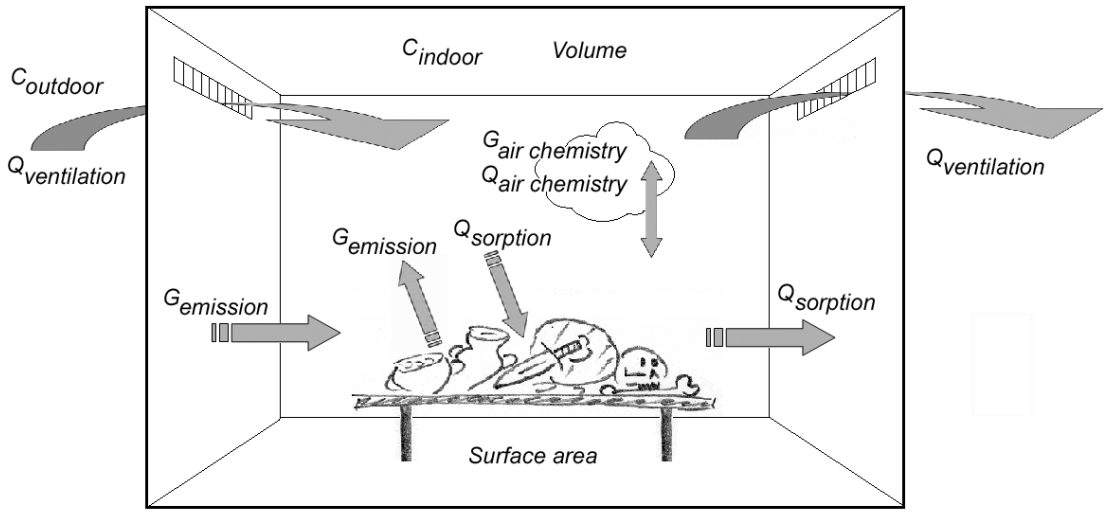

Figure 1. Mass transfer pathways in a museum room. The system depends on an outside concentration (*C*), pollution mass flows (*Q*) by ventilation or air infiltration, sorption, and removal by chemical reactions, as well as pollution generation (*G*) by emission and chemical reactions. This results in an indoor concentration (*C*) balanced by the volume (*V*) of the room.

Building	Type	O_3 Indoor [ppb]	I/O ratio	Chemical air filtration	Reference
Sainsbury Centre for Visual Arts, UK	Gallery and living area	<3-40	0.70	No	[57]
Baxter Art Gallery, USA	Gallery	120	0.60	No	[31]
Huntington Gallery, USA	Gallery	<10	c. 0.10	Yes	[31]
Los Angeles County Museum of Art, USA	Gallery	<10	c. 0.10	Yes	[31]
National Museum of Denmark	Gallery	2 [a]	0.05	No	[58]
Alcazar Castle, Spain	Galleries	8 [a]	0.18	No	[58]
Uffizi Gallery, Italy	Galleries	19-30	c. 0.35 [b]	No	[59]
An archive, Canada	Vault	<2	<0.09	Yes	[60]
Musical Instrument Museum, Belgium	Galleries	<1 [a]	0.03	No	[61]
Plantin-Moretus Museum, Belgium	Galleries	<1 [a]	0.05	No	[61]
Wawel Castle, Poland	Galleries	7 - 8	0.17 – 0.19	No	[62]
Matejko Museum, Poland	Gallery	9	0.43	No	[62]
National Museum Krakow, Poland	Galleries	5 - 6	0.19 – 0.23	Yes	[62]
Cloth Hall Museum, Poland	Gallery	11	0.44	No	[62]
Virginia Steele Scott Gallery, USA	Galleries	14	0.45	No	[63]
Villa Montezuma, USA	Historic house	14-22	0.33-0.49	No	[64]
Southwest Museum, USA	Galleries	90-143	0.69-0.84	No	[64]
Junipero Serra Museum, USA	Galleries	22-34	0.69-0.79	No	[64]
Pasadena Historical Museum, USA	Galleries	19 - 25	0.14-0.16	No	[64]
Lang Gallery, USA	Galleries	17 – 30	0.10-0.20	No	[64]
Virginia Steele Scott Gallery, USA	Galleries	43-65	0.24-0.29	No [c]	[64]
Montgomery Gallery, USA	Galleries	60 - 67	0.39 – 0.40	No [c]	[64]
4 museums with HVAC: Huntington Art Gallery; J. Paul Getty Museum; Southwest Museum Library; LA County Museum of Art, all USA	Galleries and library	Generally <10	Generally <0.10	Yes	[64]
Salarjung Museum, India	Galleries	6 [a,b]	0.58	No	[65]

Values have been rounded up

[a] converted from μgm^{-3} in original paper

[b] value read from graphed data in original paper

[c] Air-conditioning but no air filtration

Table 1. Indoor ozone concentrations and I/O ratios found at various building studies.

Building	Type	NO_2 Indoor [ppb]	I/O ratio	Chemical air filtration	Reference
Tate Gallery, UK	Galleries	15-12	-	No	[66]
Tate Gallery, UK	Storage	2-3	-	No	[66]
National Museum of Denmark	Gallery	12 [a]	0.66	No	[58]
Alcazar Castle, Spain	Galleries	4 [a]	0.72	No	[58]
National Gallery, UK.	Galleries	10-24 [a,b]	0.38-0.80	No	[68]
National Gallery, UK.	Galleries	2-5 [a,b]	0.05-0.19	Yes	[68]
Church of San Luigi Dei Francesi, Italy	Church room	6-21 [a,b]	0.19-0.78 [b]	No	[69]
An archive, Canada	Vault	1-2 [a]	0.05	No	[60]
Sainsbury Centre for Visual Arts, UK	Gallery and living area	9 [c]	0.74 [c]	No	[70,73]
Sainsbury Centre for Visual Arts, UK	Gallery and living area	13 [d]	1.32 [d]	No	[70,73]
Correr Museum, Italy	Galleries	10 [c]	0.43 [c]	No	[71,73]
Correr Museum, Italy	Galleries	11 [d]	0.75 [d]	No [e]	[71,73]
Kunsthistorisches Museum, Austria	Galleries	21 [c]	0.63 [c]	No	[72,73]
Kunsthistorisches Museum, Austria	Galleries	26 [d]	0.64 [d]	No	[72,73]
National Library of Wales	Library store	0.7-5	0.26-0.75	No	[74,75]
British Library	Library store 4th floor	5-13	0.17-0.38	No	[74,75]
Museum of London, UK	Galleries	6	0.19	Yes	[77]
Bethnal Green Museum, UK	Gallery	23	0.84	No	[77]
Manchester Museum, UK	Gallery	14	0.66	No	[78]
Horniman Museum (DSCC), UK	Storage	1	0.09	No	[78]

Values have been rounded up
[a] converted from μgm^{-3} in original paper
[b] value read from graphed data in original paper
[c] Winter
[d] Summer
[e] Windows open daily

Table 2. Indoor nitrogen dioxide concentrations and I/O ratios found at various building studies.

Building	Type	Acetic acid [ppb]	Formic acid [ppb]	Reference
Royal Museum of Scotland	Galleries	98-167 [a]	38-52 [a]	[39]
Royal Museum of Scotland	Storage room	99-246 [a]	43-115 [a]	[39]
Royal Museum of Scotland	Display and storage cases	82-808 [a]	62-520 [a]	[39]
Lower Saxony State Museum, Germany	Art gallery	24 [a]	<5 [a]	[55]
Lower Saxony State Museum, Germany	Storage rooms	<2 [a]	28 – 53 [a]	[55]
Lower Saxony State Museum, Germany	Storage cases	49 – 196 [a]	38 [a]	[55]
Musical Instrument Museum, Belgium	Galleries and storage	33 - 41 [a]	11-27 [a]	[61]
Plantin-Moretus Museum, Belgium	Galleries and storage	43 [a]	13 [a]	[61]
2 Belgium museums (*ibid*)	Display cases	49 – 1310 [a]	8 – 233 [a]	[61]
17 museums, USA	Galleries, storage, and display cases	<0.5-1600 [b]	<0.3-260 [b]	[46]
7 museums, Europe	Enclosures, e.g. display cases [c]	20-753	<0.3- 61	[79]
7 museums, Europe	Galleries and storage [c]	15-39	<0.3-15	[79]

[a] converted from μgm^{-3} in original paper
[b] Only enclosures such as display cases showed concentrations above 100 ppb
[c] At locations with artifact damage

Table 3. Indoor carboxylic acid concentrations found at various building studies.

Building	**Air exchange rate [h^{-1}]**	**Surface removal rate [h^{-1}]**	**Reference**
Virginia Steele Scott Gallery, USA	2	4.3	[63]
Villa Montezuma, USA	1.8 – 2.6 (*2.2*)	3.2	[64]
Southwest Museum, USA	5.5	1.7	[64]
MontgomeryGallery, USA	1.6	2.5	[64]
Pasadena Historical Museum, USA	0.06 – 2.2 (*0.5*)	2.8	[64]
Lang Gallery, USA	<0.99	4.0 [a]	[64]
Virginia Steele Scott Gallery, USA	1.6	4.4	[64]
Non-museum environments			
Bedroom		7.1	[94 and references herein]
Small offices		4.0 – 4.3	[94 and references herein]
Large telephone office		0.8 – 1.0	[94 and references herein]

(*italic*) average or normal air exchange rate used for calculation of surface removal rate
[a] I/O about 0.2 with doors and windows open

Table 4. Surface removal rates for ozone at various building studies. For locations where the air exchange rate or I/O ratio lay within an interval, the typical or average value was estimated before calculating the surface removal rate.

Preservation target µgm^{-3} and (*ppb*)	**100 years**	**10 years**	**1 year**
Acetic acid	100 [a]	100	1000 (*400*)
Hydrogen sulphide	0.01	0.1	1 (*0.7*)
Nitrogen dioxide	0.1	1	10 (*5.2*)
Ozone	0.1	1	10 (*5.0*)
Sulphur dioxide	0.1	1	10 (*3.8*)
Fine particles ($PM_{2.5}$)	0.1	1	10

[a] 'Because most objects have high NOAEL of acetic acid, concentrations below 100 µgm^{-3} are not mandatory'

Table 5. Maximum air pollution performance targets, according to ASHRAE [102, p. 21.9]. The targets are valid for typical museum collections, however, not for hypersensitive materials such as silver, lead, or rubber.