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THE DYNAMICS AND CONTROL OF INDOOR AIR POLLUTION IN REPOSITORIES WITHOUT MECHANICAL VENTILATION FOR CULTURAL HERITAGE COLLECTIONS. A LITERATURE REVIEW

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REVIEW PAPER

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Abstract

Airflow distributes contaminants inside buildings. Infiltration through unintentional openings in the building envelope controls the airflow in unoccupied repositories without heating, ventilation and air-conditioning (HVAC) systems. A restricted airflow may lead to the development of thermal stratification and “dead-spaces” where air pollutants emitted from construction materials or the heritage collection accumulates.

Heritage collections can act as both an emission source and a sink for carboxylic acids. Carboxylic acids can cause irreversible heterogeneous reactions with the surface of materials, e.g. tarnishing metals. It is therefore crucial to establish the dynamics of carboxylic acids inside repositories without HVAC systems, in order to determine and control its impact on the long-term preservation of heritage collections.

This paper presents a literature review on air dynamics and control of carboxylic acids inside unoccupied repositories without HVAC systems. It furthermore reviews reported levels of carboxylic acids found inside heritage institutions and sorbents used to remove them. Further research on air dynamics and whether carboxylic acids is removed primarily by deposition onto collections, or by filtration in HVAC systems inside heritage institutions is, however, necessary in order to establish the benefits of air filtration.

1 Introduction

It is crucial to determine the contribution of carboxylic acids emitted from objects in heritage collections and other indoor sources, as well as their distribution and removal on material surfaces inside heritage institutions, in order to assess air pollution’s impact on the preservation of the collections. Carboxylic acids in air causes irreversible heterogeneous reactions with material surfaces, e.g. tarnishing metals[1-5], particularly lead[5-7], formation of white crystalline deposits on glass[8], efflorescence on seashells[2], limestone[9] and ceramic[10] leading to severe pitting and disruption of the surface and hydrolysis of paper leading to loss of physical strength and brittleness[11].

A well-documented example of the contribution from heritage collections to the contamination of air by carboxylic acids inside heritage institutions is the “vinegar syndrome”, where autocatalytic deterioration of cellulose acetate, especially photographic film introduced in the first half of the 20th century, due to hydrolysis of acetate groups in the polymer structure releases acetic acid and thereby causing deterioration of collection items stored in the vicinity[12].

This paper presents a literature review on air dynamics and control inside unoccupied repositories without heating, ventilation and air-conditioning (HVAC) systems. Section 2 lists the principal building design of unoccupied repositories with semi-passive climate control as found in several Danish storage facilities with heritage collections. Information from a state-of-the-art repository in Vejle, Denmark, with semi-passive climate control is used as an example; however, the data is applicable to other countries as well.

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Section 3 describes airflow due to infiltration in repositories without HVAC systems. Section 4 describes outdoor and indoor air contaminants and levels of carboxylic acids previously measured inside heritage institutions. Section 5 list methods to control air contaminants continuing into section 6 with a focus on sorbents used to remove carboxylic acids by passive or active means. The paper draws attention to the lack of research on carboxylic acids dispersion inside unoccupied repositories without mechanical ventilation systems.

2 Unoccupied repositories without mechanical ventilation

A rough estimate predicts that in Europe buildings contribute with 40% of the anthropogenic emission of greenhouse gases and that ventilation including heating and air-conditioning accounts for one third of the energy use in buildings[13]. Controlling climate conditions inside heritage institutions with narrow permissible fluctuations increase the energy consumption in HVAC systems. Research by Mecklenburg and Tumosa[14] based on museum buildings of the Smithsonian Institution show that permitting a fluctuation larger than ± 5% in the relative humidity set point values increases energy savings significantly. A case study by Hong *et al.*[15] in a museum exhibition showed a 40% decrease in energy consumption when increasing the allowable fluctuation from 50 ±2% RH to 50 ±10% RH, and Ascione *et al.*[16] found an energy saving as high as 43% when introducing a seasonal adjustment to the set point of the HVAC system.

Within recent years a growing amount of research, roundtable discussions and conferences in the cultural heritage sector have addressed the need to revise control strategies for a sustainable management of the long-term preservation of collections[15-29]. This has pushed forward the development of unoccupied repositories without HVAC systems using passive or semi-passive climate control to provide acceptable hydrothermal conditions.

The most widespread principle for passive climate control is to use a well-insulated building envelope with high thermal inertia, to buffer against daily fluctuations in the outside climate, placed on an uninsulated concrete floor acting as a cooling source during summer and heating source during winter[30-35]. In regions with considerable temperature differences between seasons, as in Northern Europe, buildings with a high thermal inertia will have an inside temperature below the outdoor in summer and above in winter. The building envelope will buffer seasonal fluctuations in temperature to a lesser extent than daily fluctuations.

Several repositories with semi-passive climate control have been built in Denmark where one of the earliest is a storage facility at The Centre for Preservation of Cultural Heritage in the region of Vejle[36-38] (Figure 1).

The first part of the storage facility in Vejle was constructed in 2003 and it has since then been expanded in 2013 and will expand again within the next couple of years. Due to the insulation and the thermal inertia of the construction, the indoor temperature is extremely stable on a daily basis, but varies seasonally between 8 °C in winter and 16 °C in summer[37] (Figure 2).

Figure 1: A state-of-the-art storage facility in the region Vejle, Denmark, for general museum collections with semi-passive climate control. What is noticeable is how the facility consists of large storage halls with museum collections stored closely packed on mobile shelving with a large loading of collection materials in a small space. Collections made of diverse materials are stored together.

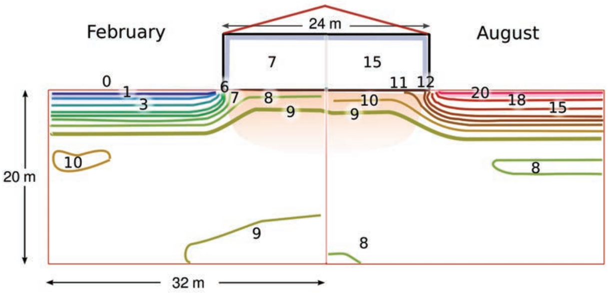

Figure 2: Computer simulation of the temperature in a repository with passive climate control. The building is constructed with a well-insulated building envelope with high thermal inertia placed on an uninsulated concrete floor. Based on outside climate conditions in Denmark the simulation show a winter scenario where the temperature fluctuates from down to 7°C in winter (left side) up to 15°C in summer (right side). (Reproduced with permission by T. Padfield[34] based on a simulation by Bøhm and Ryhl-Svendsen[32] using the COMSOL Multiphysics® Modelling Software).

The relative humidity inside the repository is kept constant by recirculating the air internally through a dehumidifier, consuming only 1.5 kWh annually per cubic metre of storage space[38-39]. Infiltration through unintentional leaks in the building envelope and opening of doors fitted with two-door airlocks is the only factors which affects the exchange of air with ambient. This result in a low air exchange rate of 0.04 to 0.05 h^{-1}.[39]

3 Airflow in buildings

Air dynamics inside museum enclosures, e.g. display cases, have been described by Padfield[40], Thomson[41], Brimblecombe[42] and Michalski[43]. In buildings, airflow can be divided in two categories: ventilation and infiltration (see ASHRAE[44] for general terminology). Ventilation is the intentional process of supplying or removing outside air into a building. The ventilation effectiveness is a measure of the air distribution system's ability to remove contaminants from the indoor environment by replacement with new air. Ventilation is further divided into mechanical ventilation, where fans control the intake, distribution and exhaust of air from outside, and natural ventilation, where air enter the building through intentional openings in the building envelope e.g. windows or ventilation grates.

Infiltration causes an unintentional exchange of air. Kalamees[45] investigated infiltration in residential houses in Finland and found that infiltration typically were through openings such as cracks around doors and window frames, and in the intersection between walls and floor or ceiling. The opposite airflow, from inside to outside, is referred to as exfiltration.

Pressure differences across the building envelope caused by wind and temperature drives infiltration. Wind induces a high-pressure on the windward areas of a building envelope and a low-pressure on downwind areas[46]. The pressure difference will create an airflow from high-pressure towards low-pressure areas horizontally across the building envelope. The pressure depends on wind direction, wind speed, the surrounding terrain and proximity to other buildings[47]. Wind will lose its force and be redirected due to friction and obstacles in the terrain and exposed buildings can therefore be sheltered from wind driven infiltration by planting trees, etc[48].

A higher temperature inside a building than outside will cause air to infiltrate through openings at ground level, rise up and escape through openings at the top of the building, and by this generating a vertical airflow. When the inside air temperature is lower than outside the airflow will be reversed. This phenomenon is called the stack-effect and depends on the building height and the temperature difference across the building envelope[49-50]. Infiltration or the airtightness of a building can be measured using the fan pressurization test[51] or calculated using the European standard 16798-7:2017 (*Calculation Methods for the Determination of Airflow Rates in Buildings Including Infiltration*)[52].

Disregarding the internal airflow resistance the stack pressure difference per meter building height per temperature difference (°C) between inside and outside is around 0.02 Pa. The outside wind pressure is generally 1-2 Pa at low wind speed (below 2.5.m s^{-1}) and 25 Pa or more at higher wind speed (above 10 m s^{-1})[53]. In most situations, a combination of wind and temperature pressure differences persist as the driving force however, as the wind speed decreases, infiltration due to the stack effect starts to dominate[54]. In buildings with a mechanical ventilation system, the unintentional infiltration of outside air can be controlled by enhancing the pressure difference across the building envelope[55].

A disadvantage of infiltration is that the airflow into and distribution within the building is unstable. This can lead to thermal stratification and the development of "dead-spaces" with static conditions where the airflow is insufficient for the dilution of internally generated air pollutants[54]. The largest temperature gradient measured in the storage facility Vejle was only 2.7 °C between the room air at 5.8 m above the floor (0.5 m below the ceiling) and the floor surface[38]. Ryhl-Svendsen[56] showed that inadequate airflow (0.3 h^{-1}) in a room without mechanical ventilation led to a concentration gradient of carboxylic acids across a room with a three times larger concentration close to the emission source compared to an adsorbing clay wall.

3.1 Airflow through large openings

Infiltration will depend on the airflow through doors, as their size, opening time and frequency increases[50]. Single-sided open door airing in historical churches can increase the air exchange rate approximately ten times[57]. European standard 16893:2018 "*Conservation of Cultural Heritage. Specifications for location, construction and modification of buildings or rooms intended for the storage or use of heritage collections*"[58] recommends using two-door airlocks in repositories with heritage collections in order to reduce infiltration from outside and between varying climate sections. The driving force for airflow through large openings inside a building is a temperature difference between sections[57]. Ryhl-Svendsen *et al.*[39] measured the airflow between two sections inside the storage facility in Vejle with semi-passive climate control, and found a two-six times larger internal air exchange rate between one section and the other as compared to the exchange with outside air.

4 Air pollution

Air pollution can originate, from sources outside and inside buildings. Two comprehensive reviews on air pollution in museums are provided by Hatchfield[59], and Tétreault[60]. Outside contaminants, e.g. ozone, sulfur dioxide, nitrogen dioxide, and hydrogen sulfide, are transported into buildings by airflow. The level of outdoor pollution inside heritage institutions have been extensively measured[32,61-85], together with a few studies on air quality in museum exhibitions in connection to human comfort[86-87].

Typical indoor contaminants, e.g. volatile organic compounds (VOC) and carboxylic acids are emitted from indoor construction materials and heritage collections. Cellulose acetate[12], paper[88-101] wood[102-103] and other materials, e.g. adhesives, paints, varnishes and plastics[104] found in cultural heritage collections release formic and acetic acid as secondary emission products during the material's own deterioration. The presence of acetic acid in indoor air has been identified as a marker of the chemical degradation of paper in library or archival collections[89]. The emission of formic acid from wood is generally an order of magnitude lower than the emission of acetic acid, but still not negligible[102].

The outdoor concentration of formic and acetic acid is approximately 0.75 ppb and 0.5 ppb[105], whereas Zhang *et al.*[106] measured a concentration of 8.77 ± 4.67 ppb for formic acid and 23.97 ±16.20 ppb for acetic acid inside residential buildings. A concentration of formic acid and acetic acid up to 361 ppb has been measured inside repositories containing heritage collections[9]. Figure 3 collates reported measurements of formic and acetic acid levels inside exhibitions and repositories as well as some studies on display cases and microclimate frames for comparison[2,8,39,74,76,82,107-120].

Enclosures, e.g. microclimate frames, display cases and repositories containing a high loading of emissive construction materials and heritage collections can result in an accumulation of airborne contaminants to high concentrations. Thickett[84] measured a concentration of acetic acid close to 2000 ppb (converted from $\mu g \cdot m^{-3}$) in a display case inside The British

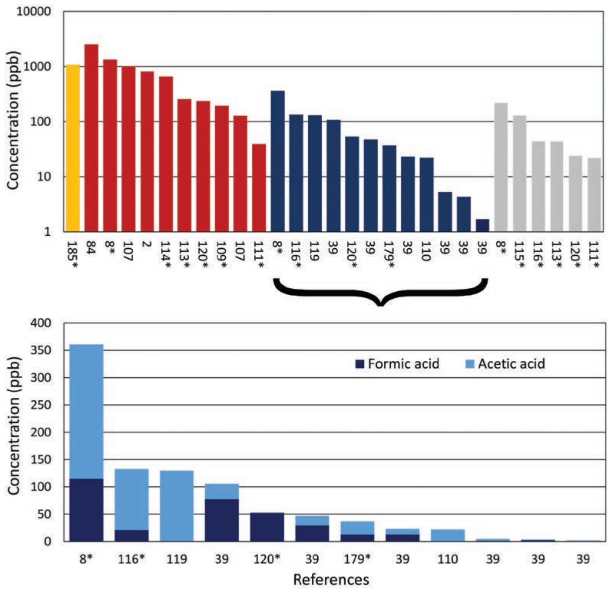

Figure 3: Concentration of carboxylic acids in heritage institutions as reported in literature. At the top diagram and displayed as sum of total carboxylic acids: inside a microclimate frame (yellow), display cases and cupboards (red), repositories (blue), and exhibitions (grey). Please note log scale, which is used due the large span of reported values. The bottom diagram zoom in on the observations from repositories and displays the fractions of formic and acetic acid concentration. Only observations of concentrations above the detection limit are included. In cases where a concentration was given as a range, the highest concentration value is used. Numbers on x-axis gives the literature reference. If (*) the concentration value was given as µg/m^3 in the original reference, but converted to ppb by this author.

Museum in London, Great Britain, whereas Godoi *et al.*[111] detected a concentration down to around 10 ppb (converted from µg·m^{-3}) in a display case inside The Rubens House Museum in Antwerp, Belgium. Ryhl-Svendsen *et al*. found a concentration of formic and acetic acid down below 2 ppb in repositories with semi-passive climate control, including a concentration of 4 ppb inside the Centre for Preservation of Cultural Heritage in the region of Vejle, Denmark[39]. Robinet *et al.* detected a concentration of formic and acetic acid up to 361 ppb in the National Museum of Scotland[8]. A reduction in the air exchange rate with the intention of reducing the ingress of outside contaminants and maintaining stable climate conditions can further contribute to the accumulation of indoor air pollutants.

4.1 Emission

Several transport processes control the emission of carboxylic acids from materials. The two main processes are diffusion within a material and surface emission[121]. Fick´s Second Law describes diffusion through a material over time. The diffusion rate depends on temperature, pressure or concentration gradient and on the molecular weight and size of the compound. The diffusion coefficient for formic and acetic acid at room temperature is $1.53\times10^{-5} \pm 0.02$ m^2 s^{-1} and $1.24\times10^{-5} \pm 0.02$ m^2 s^{-1} respectively[122].

The surface emission rate (evaporation) depend on the concentration gradient across the surface (as described by Fick's First Law) and is influenced by factors such as the surface air velocity[123-124]. A number of studies investigate the emission rate from wood[125-126] including wood used in display cases[103,127]. Risholm-Sundman *et al.*[125] measured a surface specific emission rate (*mass emitted per surface area per hour, SERa*) of VOC including acetic acid from natural wood ranging from below 10 µg m^{-2} h^{-1} from birch to 2800 µg m^{-2} h^{-1} from oak. Ramalho *et al.*[98] determined the mass-specific emission rate (*mass emitted per gram material per hour, SERm*) of acetic acid from a paper produced from pure cotton (887 ± 216 ng g^{-1} h^{-1}) and one from ground-wood pulp (4820 ± 1120 ng g^{-1} h^{-1}) after accelerated ageing. However, the emission rate from heritage collections under climate conditions typically present in unheated repositories are less studied.

4.2 Modelling airflow

Airflow transports contaminants into and around inside buildings. The dynamics of air inside buildings can be predicted and described by mathematical modelling and computational fluid dynamics (CFD) simulations. Nazaroff and Cass developed a general mathematical model to predict the concentration of chemical reactive compounds in indoor air[128]. Evola & Popov[129] showed that the Renormalization Group (RNG) theory model is useful to assess air exchange rates and the distribution of air inside buildings with natural ventilation. Özgür & Abo-Serie[130] assessed the natural airflow in a museum using CFD simulations. They showed that within the building, some areas had restricted air movement, and the authors recommend opening windows and doors to increase the ventilation effectiveness. Grau-Bove *et al.*[131] analysed the penetration, dispersion and deposition of particles in a historical house using CFD simulations, and compared the results with field measurements. Their research showed that while the ingress rate of particles was controlled by wind direction and pressure, the amount of particles that reaches a surface ultimately depended on the operation of the ventilation system.

The Innovative Modelling of Museum Pollution and Conservation Thresholds (IMPACT) model was developed during the European research project by the same name, for the use of estimation of concentration and deposition of outdoor pollutants inside museums[132-134]. The IMPACT model was based on the indoor to outdoor pollution ratio model by Weschler *et al.*[135] originally developed for ozone. A corresponding software model is not available for indoor generated air pollution.

5 Control of air pollution

Michalski[136] and Tétreault[60] presents a framework of five levels of control: *avoid, block, detect, respond and treat* for the protection of heritage collections against various agents of deterioration: one being contaminants. Within the framework, the preservation of a collection is continuously assessed at the five levels of control. This section discusses the first two levels *avoid* and *block*. The last three levels will not be part of this review.

For outdoor air pollution the first control level is to avoid sources. Placing heritage institutions in areas without exaggerated pollution levels, e.g., away from heavy traffic, industry, etc., will reduce the presence of outside pollutants. The importance of this was exem-

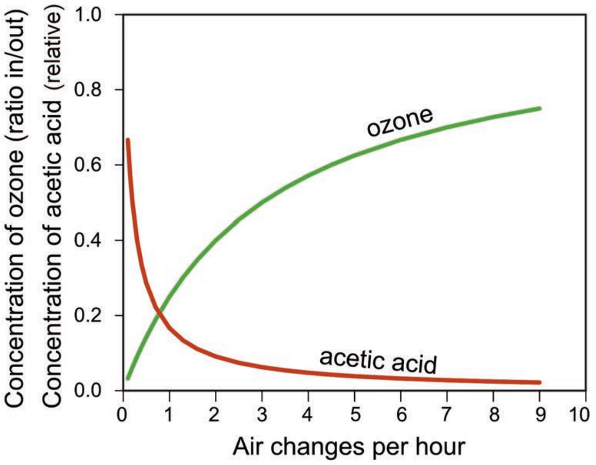

Figure 4. The concentration of outdoor generated ozone and indoor generated acetic acid as a function of the air exchange rate. (Reproduced with permission from Ryhl-Svendsen et al.[33]).

plified in the ENVIRONMENT leather project comparing the impact of air contaminants from outside on two identical sets of vegetable tanned leather books, one set stored at the British Library in central London, and another at the National Library of Wales in Aberystwyth[137-139]. The project illustrated that historically high pollution levels in central London resulted in an increase in the deterioration of book collections compared to the less polluted Aberystwyth.

For indoor pollutants, the first step of control is to avoid construction materials in exhibitions, repositories and display cases, which contribute to the emission of harmful contaminants. However, the contribution of emission products from the collection items cannot be avoided, although emissive and susceptible collection items may be divided into different sections of storage (block action) except for composite items, which at the same time are both emissive and susceptible.

The second step for the control of outdoor contaminants is to block or remove them from the air entering the building, and before it deposit onto the collection. Particle deposition modelling have showed that filtration in mechanical ventilation systems and reducing the air exchange rate are the most effective control strategies to reduce the transport of particles into heritage institutions[140]. A study by Ryhl-Svendsen[141] showed that installing a mechanical ventilation system in a previously naturally ventilated repository increased the concentration of outdoor air pollution despite the use of filtration. Both studies indicate that reducing the airflow will reduce the transport of contaminants into a building. In occupied exhibitions, however, a supply of "fresh air" is necessary to comply with human comfort requirements[142] and at the same time dilute internally generated air pollutants[33]. This is demonstrated in Figure 4, which show how the concentration of an outdoor pollutant such as ozone will increase indoors as the air exchange rate increases (see section 5.1.1). In contrast to this, the concentration of internally generated contaminants, e.g., acetic acid, will decrease with increasing air exchange, and vice-versa other parameters being equal.

5.1 Modelling indoor air pollution

The concentration of indoor generated air pollution can be calculated from the following mass balance, assuming no contribution of pollutants from outside:

$$C_i = G/V\ (n + v_d\ A/V) \qquad (5.1)$$

where C_i is the indoor concentration of air pollutant at steady-state (μg m^{-3}), G is the generation rate of pollutant (μg h^{-1}), V is the air volume (m^3), n is the air exchange rate (h^{-1}), v_d is the deposition velocity (m h^{-1}) (see section 5.1.2.1) and A is the surface area of the enclosure (m^2)[141].

Air contaminants are mainly removed by ventilation, sorption onto surfaces inside the building, and chemical reactions in air[143]. Air pollution measurements performed in cultural heritage institutions have additionally shown a decrease in the concentration of formic and acetic acid from higher summer to lower winter temepratures[39,115] stressing the use of temperature as a mean to control carboxylic acid emission from materials.

5.1.1 Air exchange rate

The air exchange rate is a measure of the air volume in an enclosure (e.g., building, display case) which is replaced with outside air per unit of time:

$$n = Q/V \qquad (5.2)$$

where n is the air exchange rate (h^{-1}), Q is the airflow (m^3 h^{-1}) and V the volume of air (m^3). The ASTM[144] "*Standard Test Method for Determining Air Change in a Single Zone by Means of a Tracer Gas Dilution*" describes three methods to measure the air exchange rate across a building envelope by using a tracer gas: the concentration decay method (based on the measured rate of which a finite amount of tracer gas injected into a room will decrease in concentration over time), the constant injection method (based on the measured steady-state concentration of a tracer gas injected into the room at a constant rate), and the constant concentration method (based on the measured amount of constantly dosed tracer gas necessary to maintain a constant concentration in the air). The latter is especially useful for measuring short-time variations of the air exchange rate.

In residential buildings uncontrolled infiltration of outside air result in an air exchange rate of approximately 1-4 h^{-1} for new and 4-10 h^{-1} for old buildings[46]. Inside repositories containing heritage collections, the air exchange rate can be significantly lower. Thickett *et al.*[83] measured an air exchange rate between 0.28 h^{-1} to 0.93 h^{-1} inside library and archival collection storage facilities in the United Kingdom. Ryhl-Svendsen *et al.*[39] measured an air exchange rate down to 0.03 h^{-1} in repositories with passive and semi-passive climate control. Christoffersen[145] recommended keeping the air exchange rate below 0.1 h^{-1} in order to provide stable climate conditions in repositories with passive climate control.

5.1.2 Sorption

5.1.2.1 Deposition velocity

As outside air infiltrates through the building envelope the concentration of pollution in that air will constantly decrease as contaminants deposit onto surfaces inside the building. The rate of pollutant uptake by surfaces (the sink-effect) is commonly expressed by the deposition velocity (v_d). The deposition velocity depends firstly on the mechanisms that control the flow of pollutant towards the surface, and secondly on the physical or chemical interaction between the pollutant and surface[146]. Nazaroff *et al.*[147] provides a detailed discussion of the deposition velocity concept; in brief the definition is the flux of a pollutant to a surface (F) divided by its concentration in air, and holds the unit of velocity (m h^{-1} or cm s^{-1}).

The deposition velocity of carboxylic acids is only known for a few materials, e.g., 0.005 cm s^{-1} of formic and acetic acid onto silver[148], 0.007 cm s^{-1} and 0.014 cm s^{-1} respectively onto copper[149], and between 0.00089 cm s^{-1} and 0.0095 cm s^{-1} (converted from m s^{-1}) for acetic acid onto canvas[150]. The deposition velocity of carboxylic acids to general interior surfaces has never been described, however, Grøntoft[151] estimated a deposition velocity of 0.0016 cm s^{-1} (converted from m s^{-1}) of acetic acid inside museum display cases based on measurements and modelling. Ryhl-Svendsen[152] suggested a deposition velocity of 0.002 cm s^{-1} formic and acetic acid inside heritage repositories using mass-balance modelling.

5.1.2.2 Surface removal rate

A useful factor used to describe sorption of air contaminants to a surface is the surface removal rate (S), defined as the deposition velocity multiplied by the surface-to-volume ratio of the enclosure:

$$S = v_d A/V \qquad (5.3)$$

where v_d is the deposition velocity (m h^{-1}), A the surface area of the material (m^2) and V the volume (m^3). The surface removal rate has the unit reciprocal time (h^{-1}) and is therefore directly comparable to the air exchange rate.

Several surface mechanisms control the removal of air contaminants by sorption[13]. Air pollution can be adsorbed by two mechanisms: physical or chemical adsorption. Physical adsorption involves a comparatively weak bond between the pollutant and sorbent surface. This process is reversible and desorption can occur due to changes in the concentration gradient between the sorbent and the airflow, or by an increase in temperature. Chemical adsorption involves an irreversible chemical reaction between the pollutant and sorbent.

Thickett[84] found a surface removal rate for organic acids inside display cases made of glass and powder coated metal of approximately 0.0042 h^{-1} to 0.0063 h^{-1} (converted from per day) whereas Ryhl-Svendsen[141] calculated a surface removal rate of 0.26 h^{-1} for organic acids in repositories with brick walls and filled with heritage collection items.

5.1.3 Chemical reactions in air

In cases where chemical reactions in air are fast enough to dominate over the rate of removal by the air exchange or surface reactions they will cause a sink-effect[153-154]. However, no chemical reaction with carboxylic acids will occur at the air exchange rates typically found inside buildings[155].

5.1.4 Temperature

The primary emission of VOC from building products, paints, varnishes, etc., depend on temperature[121,156-157]. For the same reason the level of carboxylic acids inside display cases[151,158] and heritage repositories[39,115] depend on temperature (as well as other environmental parameters). Krupinska *et al.*[115] found a five to six times increase in the concentration of carboxylic acid in The Plantin-Moretus Museum/Print Room Antwerp, Belgium, and Ryhl-Svendsen *et al.*[39] found an up to six times concentration increase in unheated storage buildings in Denmark from winter to summer.

Brimblecombe and Grossi[159] predicted that the in the future increasing temperature due to global warming will lead to a greater impact from carbonyl compounds such as formic and acetic acid. Krupinska *et al.*[115] and Ryhl-Svendsen *et al.*[39] both recommend reducing the temperature in repositories to improve the preservation of cultural heritage collections.

6 Sorbents

Sorbent media are used to remove contaminants actively by passing air through filters in ventilation systems, or passively as reactive interior surfaces inside buildings. Blades *et al.*[160] propose to reduce the air exchange rate and recirculate air through filters inside buildings in order to reduce the ingress of contaminants from outside and the cost of air-conditioning. Ryhl-Svendsen & Clausen[161] showed that recirculating internal air through activated charcoal filters impregnated with potassium permanganate inside a storage room was generally the most effective method to reduce contaminants from outside, as compared to mechanical ventilation with filtration, or passive sorption on active wall materials. Ryhl-Svendsen[56] showed that installing a wall of unfired clay bricks inside a room passively reduced the concentration of organic acids by 71%, due to the sorption of acid onto the clay. The loading of the clay bricks was rather high with a surface-to-volume ratio of 0.46 m^2 m^{-3}. Passive sorption performs better in small repositories compared to larger ones due to a higher ratio of surface-area to volume[161].

Several studies points to activated charcoal as the most efficient sorbent for carboxylic acid removal in heritage institutions[150,162-164]. Carbonaceous materials, such as wood, nutshells, peat, hard coal or lignite is used to produce activated charcoal. A sorbent's removal capacity depends on its surface area, with one gram of activated charcoal having a surface area (depending on the type) of above 1000 m^2.[46] Activated charcoal can furthermore be impregnated, often by an alkali such as KOH, to enhance its removal efficiency towards specific contaminants, i.e. acids. The advantage of impregnated activated charcoal filters is that the contaminants adsorb irreversibly whereas concern about saturation and re-release (desorption) of previously adsorbed contaminants from non-impregnated activated charcoal have been raised[165]. However, research by Thickett and Short-Traxler show no desorption of acetic acid from activated charcoal used to control the concentration of carboxylic acid passively in display cases[166].

According to Brokenhof[165] sorbents impregnated with potassium hydroxide will remove acetic acid more efficiently than sorbents containing calcium carbonate. Schieweck[164] showed that out of 39 sorbents zeolites, activated charcoal, and activated charcoal impregnated with an alkaline substance were the most efficient to remove formic and acetic acid. However, Schieweck[164] recommended using activated charcoal without impregnation due to its lower cost and good adsorption capacity.

European standard EN 16893[167] "*Conservation of cultural heritage – New sites and buildings intended for the storage and use of collections*" recommends using internal recirculation through activated carbon filters to remove air contaminants generated inside repositories with heritage collections, while ASHRAE[168] recommends the application of permanganate impregnated aluminium or activated carbon filters to remove acetic acid from air.

6.1 Justification of the use of air filtration

The increase in energy consumption of a HVAC system necessary to overcome the airflow resistance through filters have traditionally been assumed to account for the largest pressure drop of such installations[169]. In residential and light-commercial buildings, filtration have been shown to account for 21-100% of the total pressure drop[170]. However, new and more energy efficient filters are constantly being developed. Irrespective of the energy use for air filtration in HVAC systems there is a consensus on the benefits associated with filtration in connection to human comfort and productivity with several studies examining energy consumption and efficiency of air filtration[171-176].

Within heritage institutions, there is, however, an ongoing debate on the justification of air filtration for the removal of carboxylic acids inside buildings. Menart *et al.*[177] stated that the effect of acetic acid on the deterioration of paper collections is insignificant and therefore questions whether chemical air filtration in library and archival collections can be justified. Di Pietro *et al.*[178] additionally examined the use of air filtration from a cost-benefit perspective stating that carboxylic acids are expected to cause insignificant deterioration to library and archival collections, and in comparison with the associated expenses air filtration should be discouraged. Mašková *et al.*[179], on the other hand, concluded from outdoor and indoor air pollution measurements in five archives in Czech Republic, that only repositories with air filtration were suitable for long-term storage of archive and library collections.

Further research on air dynamics, and whether carboxylic acids are removed primarily by deposition onto the collections, on interior surfaces, or by filtration in HVAC systems, is however necessary to determine the conservation benefits of filtration for heritage institutions.

7 Conclusion / Summary

Airflow transports and distributes contaminants inside buildings. Infiltration through unintentional openings in the building envelope controls the airflow in unoccupied repositories without HVAC systems. Ryhl-Svendsen *et al.*[39] measured an air exchange rate down to 0.03 h^{-1} in repositories with passive climate control. A restricted airflow can lead to the development of thermal stratification and "dead-spaces" where air pollutants emitted from construction materials or the heritage collection accumulates. As shown by Ryhl-Svendsen[56] a low air exchange rate (0.3 h^{-1}) led to a concentration gradient of carboxylic acids across a room with a three times larger concentration close to the emission source compared to an adsorbing clay wall.

Carboxylic acids can cause irreversible heterogeneous reactions with the surface of materials e.g. tarnishing of metals. It is therefore crucial to establish the route and fate of air pollutants, including the fraction removed by deposition onto heritage collections inside repositories, compared to the fraction removed by recirculation through filters. Thickett[84] estimated a surface removal rate for carboxylic acids inside display cases from 0.0042 h^{-1} to 0.0063 h^{-1} and Ryhl-Svendsen[152] 0.26 h^{-1} in repositories.

The level of carboxylic acids inside repositories can be controlled by using non-emissive construction materials (avoiding the source) and by decreasing the temperature (reducing the emission rate). The use of active and passive sorbents will remove carboxylic acids from air (blocking the pollutant). Several studies points to activated charcoal as the most efficient sorbent for carboxylic acid filtration in heritage institutions[150,162-164]. Passive sorption performs better in small repositories compared to larger ones due to a larger surface-area relative to the air volume[161]. Therefore, passive sorption is probably not efficient at removing carboxylic acids with a high loading of heritage collections stored in a small volume inside repositories with passive climate control only.

No studies on air dynamics inside repositories without mechanical ventilation systems used for the storage of cultural heritage collections were encountered during this literature review. Further research on air dynamics and whether carboxylic acids are removed primarily by deposition onto collections or by filtration in HVAC systems inside collection storage rooms is however necessary to determine the cost and benefits of air filtration.

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