



Ozone removal on building material surface: A literature review

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ARTICLE INFO

Keywords:

Ozone
Surface removal
Building material
Deposition velocity
Reaction probability
Indoor air quality

ABSTRACT

Ozone is a reactive gas that can have negative health effects on human. Building materials can be significant sinks for indoor ozone, owing to the irreversible heterogeneous reactions between ozone and material surfaces. Therefore, the ozone removal on material surfaces is crucial for evaluating indoor ozone concentrations and human exposure. This paper presents a review of previous investigations on ozone removal on building materials. The reaction probabilities of common indoor building materials range from 10^{-8} to 10^{-4} , and depend on the material chemical compounds and surface characteristics. The surface-treated materials are probably more important than the underlying material substrate in determining ozone deposition velocities. Ozone removal on material surface is also associated with the fluid mechanics near the surface. Reactions between ozone and unsaturated organic compounds that constituting or adsorbed on material surfaces may result in oxidized by-products yields, while inorganic materials usually exhibit negligible by-products yields. Besides, the ozone surface removal on building materials under various conditions, i.e. ozone concentrations, air flow conditions, relative humidity and temperature, are discussed. Ozone removal on building materials after short-term and long-term exposure is presented.

1. Introduction

Ozone is a reactive gas that can have negative health effects on human, including increasing in respiratory-related morbidity, cardiovascular morbidity and premature mortality [1–17]. Weschler [18] presented that the indoor ozone concentration is dependent on the outdoor concentration, the air change rate, indoor ozone sources, and the removal by indoor surfaces and gaseous chemicals. In the absence of indoor ozone emission sources such as laser printers, photocopiers, and ionization/ozonolysis air cleaners [19–25], ambient outdoor ozone entering into buildings is the primary source of indoor ozone.

Buildings and aircrafts are usually the most typical indoor ozone exposure scenarios. Indoor ozone concentrations in buildings are generally below 50 ppb [26], while the concentrations in aircrafts can reach an elevated level of 300 ppb because of the high ambient ozone concentration in the air at typical cruise altitudes [27]. Usually, indoor ozone concentrations are lower than outdoor concentrations owing to the irreversible reactions at indoor materials and human surfaces that consume ozone [28]. The ratios of indoor to outdoor ozone concentrations are mainly in the range of 0.2–0.7 for most buildings according to numerous investigations [18]. However, albeit lower concentrations, indoor ozone exposures in buildings are around 43–76% of total daily ozone exposures [29], due to the fact that people spend average approximately 90% of their time indoors [30–33].

Building materials can be significant sinks for indoor ozone, owing to the irreversible heterogeneous reactions between ozone and material surfaces. The rate of ozone removal at the surfaces of building materials, which is typically quantified by deposition velocity, is governed by the transport of ozone to the material surface and the ozone uptake onto the surface [34,35]. Numerous researches have been conducted to study the ozone removal rate of some widely used building materials with the methods of laboratory experiments and field tests [35–40]. Available building materials studied in literature include glass, metals, ceramic materials, synthetic materials, finished floors, wallpapers, wooden boards, paints, ceiling tiles, concretes, cottons, wools, gypsum boards, carpets, activated carbon materials and bricks.

Reactions between ozone and material surfaces may also result in oxidized by-products yields, including C1–C13 carbonyls, dicarbonyls and hydroxycarbonyls [36–38,41–54], which can adversely affect occupants' health and perceived air quality [18,29,45,55–63]. These by-products are usually produced from reactions between ozone and the unsaturated organic compounds that constituting or adsorbed on material surfaces [29], while inorganic materials usually exhibit negligible by-products yields [37,38,45,64]. Some of these by-products with low vapor pressures can nucleate to new particles or condense on existing particles to form secondary organic aerosols (SOA) [65–68].

Recently, the investigations into ozone removal of passive removal materials (PRMs) have received increasing attention

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[35,37–39,44,45,64,69]. Compared to the conventional energy-consumed active filtration, passive removal materials such as activated carbon cloth, gypsum boards, carpet and clay based materials, can remove considerable indoor ozone through the chemical reactions between ozone and material surfaces without additional building energy consumption, and meanwhile form negligible by-products [35,37–39,44,45]. Therefore, PRMs have been considered to be an efficient and minimal energy-consumed passive control strategy for lowering indoor ozone concentrations and reducing human exposure.

There have been some detailed review articles focused on indoor ozone. Weschler [18,70–73], Salthammer and Bahadir [74], and Fadeyi [26] conducted reviews on previous researches about chemical reactions and products of indoor ozone. Weschler et al. [29,75] and Sundell et al. [76] highlighted the impact of indoor ozone on occupant health. Weschler [77] introduced the indoor ozone removal by human occupant. Darling et al. [64] presented a detailed review of the ozone surface removal on PRMs. However, these studies did not focus specifically on the ozone removal on common widely-used building materials. This paper attempts to provide the detailed reviews of the previous investigations on the surface removal of indoor ozone for a wide range of building materials. The ozone surface removal on building materials under various conditions are discussed.

The scientific literature reviewed in this paper was gathered by searching through ISI Web of Science (1900-present) and ScienceDirect (1823-present). In addition, Google Scholar was used as a supplementary search. As a source of search records, the following keywords were used: indoor ozone; surface removal; deposition velocity; reaction probability; building material; CFD simulation; mortality; morbidity; occupant health; ozone generation source; ozone surface chemistry; passive removal material; secondary emission.

Articles and publications were considered for inclusion based on the following criteria:

- Original research articles in English;
- Articles relevant to the key research questions identified;
- Publications up to December 2017;
- Articles without information on indoor air pollutants were excluded;
- Abstracts and purely descriptive articles without a detailed analytic component were excluded;
- Conference papers were excluded.

2. Mechanism and research methods

2.1. Mechanism

The level of indoor ozone concentration depends on the outdoor ozone concentration, air change rate, indoor emission rate, surface removal rate, and reaction between ozone and other chemicals in air [18]. Grøntoft [78,79] developed a group of multi-parameter models to describe the reaction of ozone on material surfaces, which consider the adsorption of ozone to materials, the desorption of ozone to air, and the diffusion of ozone into the materials. However, the majority of the published literature usually described the reaction of ozone to materials with a single-parameter mass balance model in an assumed well-mixed space, without considering the desorption of ozone to air and the diffusion of ozone into the materials. Based on the single-parameter model, the indoor ozone concentration can be defined by

$$\frac{dC_{in}}{dt} = P\lambda C_{out} + \frac{E}{V} - \lambda C_{in} - \sum k_{o_3,j} C_j C_{in} - \sum \frac{v_{d,b} A_b C_{in}}{V} - \sum \frac{v_{d,h} A_h C_{in}}{V} \quad (1)$$

where C_{in} is the indoor ozone concentration (ppb), P is the ozone penetration factor (0–1), λ is the air change rate (h^{-1}), C_{out} is the outdoor ozone concentration (ppb), E is the emission rate of ozone into the space ($ppb \cdot m^3/h$), V is the volume of the air indoors (m^3), $\sum k_{o_3,j} C_j$ is the

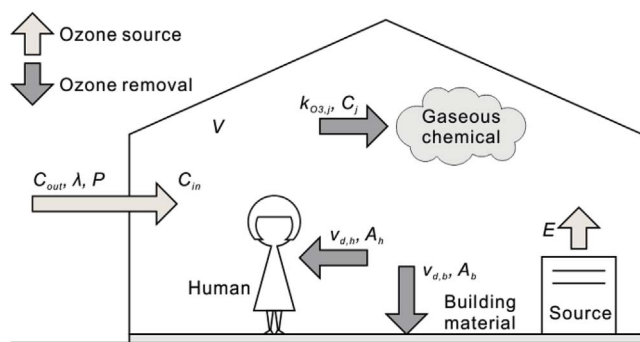


Fig. 1. Schematic illustration of indoor ozone source and sink.

ozone removal rate by gaseous chemicals through chemical reactions (h^{-1}), $k_{o_3,j}$ is the rate constant for the reactions between ozone and gaseous chemical j ($ppb^{-1} \cdot h^{-1}$), C_j is the indoor concentration of gaseous chemicals j (ppb), $\sum v_{d,b} A_b / V$ is the ozone removal rate by indoor building materials (h^{-1}), $v_{d,b}$ is the ozone deposition velocity of the building material (m/h), A_b is the surface area of the building material (m^2), $\sum v_{d,h} A_h / V$ is the rate at which ozone is removed by human surfaces (h^{-1}), $v_{d,h}$ is the ozone deposition velocity of the human surface (m/h), and A_h is the surface area of the human surface (m^2) (see Fig. 1). The surface removal of indoor ozone can be determined by the first-order irreversible surface heterogeneous reaction, which can be quantified by ozone deposition velocity (v_d). Ozone deposition velocity is a mass-transfer coefficient governed by the ozone transport to the surface and uptake by the surface, which is associated with fluid mechanics near the material surface and the chemical reactivity of the material with ozone [34]. The inverse of deposition velocity can be taken as an overall resistance to surface reactions (r_o), and is equal to the sum of transport resistance (r_t) and surface uptake resistance (r_s):

$$\frac{1}{v_d} = r_o = r_t + r_s = \frac{1}{v_t} + \frac{1}{v_s} = \frac{1}{v_t} + \frac{4}{\gamma v} \quad (2)$$

where v_t is the transport-limited deposition velocity (m/h), v_s is the reaction-limited deposition velocity (m/h), γ is reaction probability (–), and $\langle v \rangle$ is Boltzmann velocity for ozone ($\langle v \rangle = 3.60 \times 10^4$ cm/s). The transport-limited deposition velocity depends on the boundary layer fluid mechanics near material surfaces. The reaction-limited deposition velocity can be quantified by reaction probability, which is the ratio of the removal rate to the collision rate of ozone on the surface. The magnitude of the ozone reaction probability is material specific and system-independent, which can be calculated by

$$\gamma = \left[\frac{v}{4} \left(\frac{1}{v_d} - \frac{1}{v_t} \right) \right]^{-1} \quad (3)$$

The values of γ range from as low as 10^{-8} for glass and metal to 10^{-4} for activated carbon and brick. Cano-Ruiz et al. [34] indicated that the surface removal of ozone is transport-limited when γ is larger than 3×10^{-4} , and reaction-limited when γ is smaller than 5×10^{-7} for typical indoor air flow conditions.

The by-products of reaction between ozone and material can be quantified by the molar yield (y_i), which is defined as the ratio of moles of the by-product i emitted from the material to moles of ozone removed by the material:

$$y_i = \frac{\text{moles of } i \text{ emitted}}{\text{moles of ozone removed}} = \frac{\Delta C_{prod,i}}{C_{out} - C_{in}} \quad (4)$$

where C_{in} is the indoor ozone concentration (ppb), C_{out} is the outdoor ozone concentration entering into the building (ppb) and $\Delta C_{prod,i}$ is the increased concentration of the by-product i due to the reaction between ozone and material (ppb).

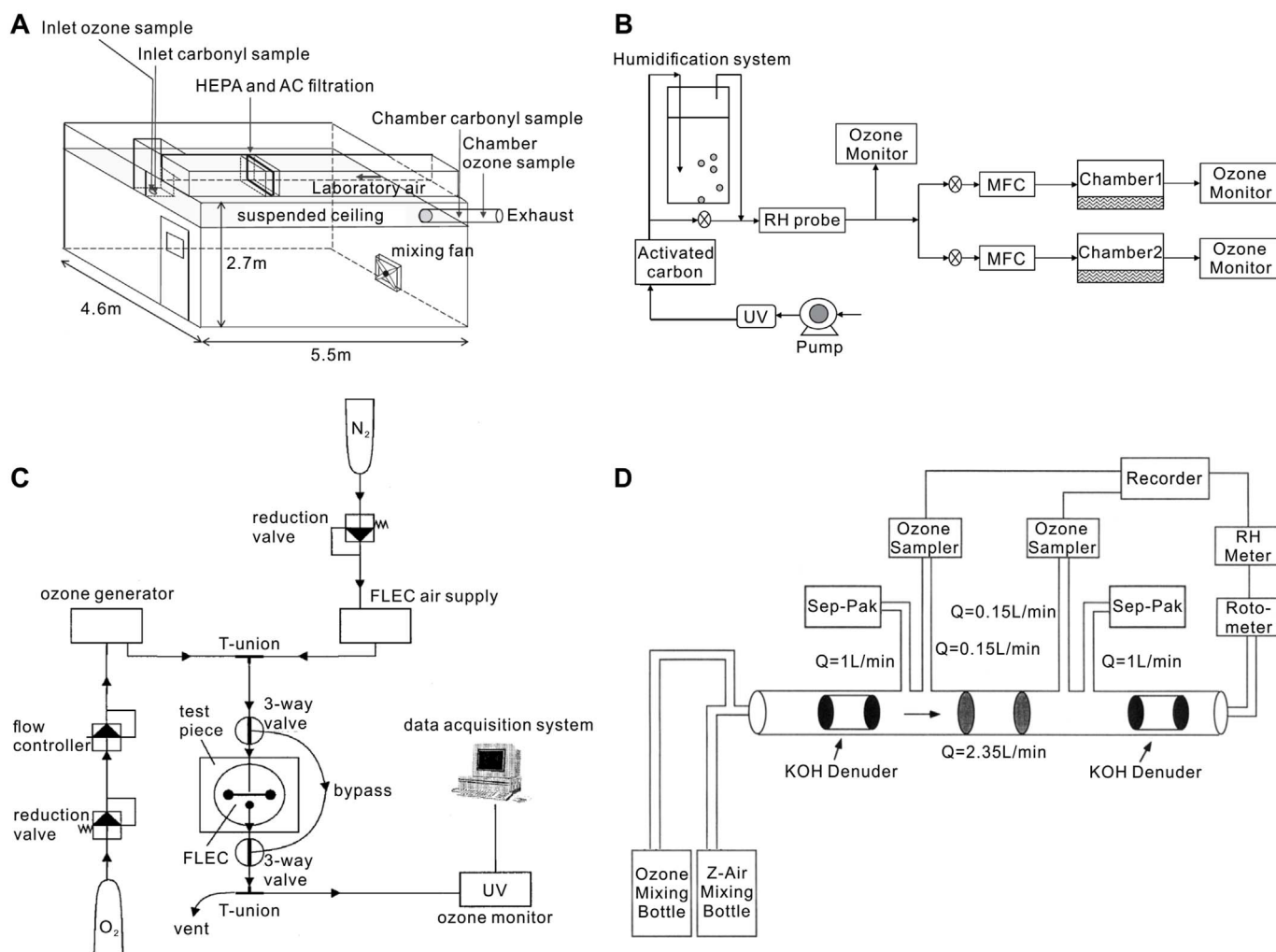


Fig. 2. Typical chamber experimental apparatuses: A. large-scale chamber experimental apparatus [38]; B. small-scale chamber experimental apparatus [35]; C. FLEC experimental apparatus [89]; D. tube flow reactor experimental apparatus [43].

2.2. Research methods

2.2.1. Experimental studies

Previous studies usually conducted field tests and environmental chamber experiments to determine the ozone removal rate of studied materials, covering unused and used materials. The environmental chambers included large-scale (room-scale) chambers, small-scale chambers, field laboratory emission cells (FLEC) and tube flow reactors. Typical experimental apparatuses of different types of chamber experiments are shown in Fig. 2. The experimental conditions in literature are shown in Table 1. Purified air (free of particles, volatile organic compounds, ozone and water) was first pre-conditioned to target temperature, humidity, ozone concentration and flow rate, and then introduced into the experimental chamber. There were two common methods to measure the ozone removal rates of materials. With regard to the experiments in large-scale chambers [38,39,44], ozone was usually introduced into the chamber until its concentration reached a specific level. The ozone generator was then switched off and the ozone concentration in chamber began to decay to the lower level, which was measured by ozone monitors. The ozone removal rate of material could then be computed by curve-fitting the measured ozone concentration decay. Another method was widely used in small-scale chamber experiments, that ozone was continuously injected into the chambers and the chamber ozone concentration levels were continuously sampled. This method could be used to measure both steady and time-dependent

ozone deposition velocities. In the experiments with relatively large volume chambers, additional mixing fans were usually put in the chamber to make the in-chamber air well-mixed [39,40,44,80–82]. The ozone-initiated by-products were then sampled after a period of ozone exposure.

The mass-balance model in Eq. (1) can be simplified to Eq. (5) to determine the ozone deposition velocities in chamber experiments, assuming that there are no ozone source and no gas-phase reaction in the chambers:

$$\frac{dC_{in}}{dt} = \lambda C_{out} - \lambda C_{in} - \frac{v_d A C_{in}}{V} - \frac{v_{d,c} A_c C_{in}}{V} \tag{5}$$

where C_{in} is the ozone concentrations inside the chamber (ppb), C_{out} is the inflow ozone concentrations (ppb), v_d is the deposition velocity of studied material specimen (m/h), A is the area of material specimen (m^2), $v_{d,c}$ is the chamber background deposition velocity (m/h) and A_c is the chamber boundary area uncovered by the material specimen (m^2).

The majority of the materials of environmental chambers are stainless steel (SS) and glass, owing to their inert reactivity with ozone. According to the previous studies, the reaction probabilities of stainless steel and glass are in the order of 10^{-7} or 10^{-8} . In addition, Lucite and Teflon are sometimes used as the chamber materials in the experiments [46,49,50,81,82], also because of their inert reactivity with ozone (5.5×10^{-8} for Lucite and 5.56×10^{-7} for Teflon). Usually, the experiments were conducted with the chamber empty first (no studied

Table 1
Experimental conditions conducted in environmental chambers in literature.

Chamber	V [L] ^a	Material	T [°C] ^a	RH [%] ^a	C _{out} [ppb] ^a	λ [h ⁻¹] ^a	Reference
Large-scale	14200	SS ^b		20–60	150–200	^c	Kunkel et al., 2010 [39]
	30000	SS	23	33	80	1.5 ^c	Darling et al., 2012 [44]
	68000	SS	25.2	25–75	140	1.1 ^c	Gall et al., 2013 [38]
Small-scale	2380	Lucite	22.2	50		^c	Sabersky et al., 1973 [81]
	125	Teflon				^c	Simmons and Colbeck, 1990 [82]
	10.5	SS	23	50	100	5.7	Morrison et al., 1998 [41]
	10.5	SS		50	100	6.9	Morrison and Nazaroff, 2000 [50]
	10.9	Glass	22	50–70	40	11	Grøntoft, 2002 [79]
	10.9	Glass	22	0–90	40	11	Grøntoft and Raychaudhuri, 2004 [90]
	10.9	Glass	22	50–70	40	11	Grøntoft, 2004 [78]
	10.9	Glass	22	0–90	40	11	Grøntoft et al., 2004 [91]
	4.25	Teflon		50	100–150	28.2	Wang and Morrison, 2006 [49]
	6	Glass	20		1000	2400	Ito, 2007 [83]
	17	Glass	23	30–50	0–300	12	Nicolas et al., 2007 [47]
	48	SS	25	50	1.0 × 10 ⁶ –1.2 × 10 ⁶	0.5	Poppendieck et al., 2007 [48]
	10.5	SS	23	10	105–120	17.1–22.9	Coleman et al., 2008 [84]
	48	SS	22–24	39–56	100–150	1	Hoang et al., 2009 [35]
	4.25	Teflon		50	150	28.2	Wang and Morrison, 2010 [46]
	10	SS	25	50	150–200	12.3	Lamble et al., 2011 [45]
	48	SS		50	147	2	Cros et al., 2012 [37]
	10	SS	23.4	51	105	11.7	Gall et al., 2014 [80]
	216	SS	25	50	75	0.5 ^c	Lin and Hsu, 2015 [40]
	10.7	SS	22–28	25–75	60–62	10	Rim et al., 2016 [85]
52	Glass	21	50	120	3	Abbass et al., 2017 [36]	
FLEC	0.107	SS/Glass	21	50	50	504.7	Kleno et al., 2001 [89]
	0.035	Glass		< 2	100	1714.3	Schripp et al., 2012 [92]
Tube flow reactor	0.104–0.208	Glass		0–100	70–100		Reiss et al., 1994 [93]
	0.104	Glass			50–150	1442.3	Reiss et al., 1995 [43]
	0.036	Teflon			100		Morrison and Nazaroff, 2000 [50]

^a V is the volume of the chamber (L), T is the conditional temperature (°C), RH is the conditional relative humidity (%), C_{out} is the inflow ozone concentrations (ppb), λ is the air change rate of the chamber (h⁻¹).

^b SS stands for stainless steel.

^c Using additional mixing fans in chamber.

material specimen) to assess the background ozone deposition velocity ($v_{d,c}$) according to Eq. (5) to compensate the ozone removal contribution by chamber surfaces for ozone deposition of studied material specimen. In some investigations, the ozone removal by chamber surface was neglected as a result of its negligible removal contribution [48,50,83].

The ozone deposition velocities measured in different experiments may vary a lot due to various transport-limited deposition velocities of specific flow conditions. Given the measured deposition velocity v_d , and the transport-limited deposition velocity v_t , the reaction probability (γ) of specific material, which is considered to be chamber-independent, can be estimated based on Eq. (3) in the environmental chamber experiments. The transport-limited deposition velocity can be obtained experimentally by eliminating uptake resistance at the material surface (r_s). In environmental chamber experiments, it can be achieved by coating the material with a highly ozone-reactive component, e.g. potassium iodide [45,48,50,80,82,84], nitrite [35,40], or polybutadiene [85], which is considered to be a perfect sink for ozone. Experiments identical to ones conducted to measure v_d then can be conducted with the material coated with the highly ozone-reactive component to measure v_t . The transport-limited deposition velocity can be determined by applying Eq. (2). In addition to this method, Morrison et al. [86–88] developed the deposition velocity sensor (DeVS) method to evaluate the transport-limited ozone deposition velocity to surfaces by quantifying the conversion by ozone of nitrite to nitrate on a glass fiber filter employing a small quartz crystal microbalance, which was usually used in field tests [86–88].

2.2.2. Numerical studies

Although experimental studies can provide reliable results, they

can't provide the distribution of ozone concentrations in indoor environments, because experimental studies are usually based on the single-parameter mass balance model in assumed well-mixed indoor environments. However, the distribution of ozone concentration in indoor environment, particularly the ozone concentrations in the breathing zone, is crucial to ozone inhalation exposure and thus closely associated with occupant health, which is thus deserved to be investigated.

Hence, a numerical method, i.e. computational fluid dynamics (CFD) simulation, has been attempted to obtain the distribution of ozone and associated by-products in indoor environments considering the ozone removal by building materials. Sørensen and Weschler [94] used CFD simulations to study the chemical reaction between ozone and d-limonene with a hypothetical product and surface deposition of ozone in a 2D computational domain. Ito [83] conducted CFD simulations to analyze ozone distribution of model rooms with different kinds of materials. Russo and Khalifa [95] improved a CFD model to predict the concentration distribution of ozone, d-limonene, and a hypothetical product in a typical office space with optional personal ventilation system considering ozone removal by both building material and human occupant. Ito and Harashima [96] investigated the ozone deposition on building materials and the generation of secondary organic aerosols by chemical reactions of ozone and limonene numerically and experimentally. Rai et al. [97] developed a CFD model to predict the ozone distribution in an aircraft cabin with the ozone removal on building materials and human-related surfaces. Gao and Zhang [98] simulated ozone penetration through the wall assembly considering ozone deposition on the wall materials. The ozone concentration distribution can be solved by the following species transport equation in CFD simulations:

$$\nabla \cdot (\rho \vec{u} C) = \nabla \cdot \left(\left(\rho D_o + \frac{\mu_t}{Sc_t} \right) \nabla C \right) + S_c \tag{6}$$

where ρ is air density (kg/m^3), \vec{u} is air velocity vector (m/s), C is ozone concentration ($-$), D_o is binary diffusion coefficient of ozone in air (m^2/s), μ_t is turbulent viscosity ($\text{kg/m}\cdot\text{s}$), Sc_t is turbulent Schmidt number ($-$), and S_c is ozone source ($\text{kg/m}^3\cdot\text{s}$). The ozone surface removal on material and human surfaces depends on the local ozone concentration adjacent to the surface, and can be modeled as a surface deposition flux as given by Ref. [34].

$$J_s = -\gamma \frac{v}{4} C(\Delta y) \tag{7}$$

where $C(\Delta y)$ (kg/m^3) is the concentration at $\Delta y = 2\lambda/3$, where λ is the mean molecular free path ($6.5 \times 10^{-8} \text{ m}$).

However, with Eq. (7), an extremely fine grid size near the deposition surface is required, compared to the length scales necessary to resolve the flow and concentration fields within the CFD model, which makes the CFD simulations unavailable. To increase the grid size to an available size, Sørensen and Weschler [94] developed a flux model assuming that a boundary layer flow prevails across the majority of the surface area and the velocity goes towards zero for areas $y^+ < 1$, which indicated a laminar diffusion, i.e. a linear concentration profile, in this region. The surface flux can therefore be calculated by

$$J_s = \frac{-\gamma v/4}{1 + (\gamma v/4/D_m)\Delta y_1} C(\Delta y_1) \tag{8}$$

where D_m is the binary diffusion coefficient of the chemical species in air (m^2/s). Thus, knowing the grid height at the wall (Δy_1), the surface material (γ), and the chemical species (v , D_m), the flux is a known function of $C(\Delta y_1)$, which in turn can be known from the CFD simulation. It needs to be emphasized that Eq. (8) is valid only when the distance from surface to the first grid point is small enough ($y^+ < 1$). Table 2 demonstrates the CFD settings of modelling ozone removal on material surfaces.

Eq. (8) was usually used to determine the ozone flux at material surfaces such as the walls, ceilings and floors. However, due to the high reactivity of ozone with human surfaces such as skin, hair, and clothing [99–101], the ozone concentration is expected to be very low at those human surfaces [101]. Therefore, most papers set zero ozone concentration at human surfaces in CFD simulations, rather than using Eq. (8) to simulate the ozone removal flux [97,102].

Because of the probable errors in CFD simulations which may affect the accuracy of the results, reliable experimental data is necessary to validate the simulation results. Therefore, in order to study the distributions of ozone and its by-products in indoor environments, CFD

simulation is a good method, but experimental data are still needed to validate its results.

3. Ozone surface removal on different materials

3.1. Building materials

Numerous papers have investigated ozone surface removal on some widely-used materials. Available building materials studied in literature include glass, metals (stainless steel, aluminum etc.), ceramic materials, synthetic materials (linoleum, rubber, Teflon, nylon, polyethylene materials etc.), finished floors (wooden and bamboo floors), wallpapers (vinyl and paper wallpapers etc.), wooden boards (particle boards, fiberboards, plywood etc.), paints (water-based, oil-based, latex, clay, collagen paints), ceiling tiles (mineral fiber ceilings, perlite-based ceiling tiles, fiberglass ceiling tiles etc.), concretes, cottons, wools, gypsum boards (painted and unpainted gypsum boards), carpets (nylon, wool, polypropylene, polyester carpets etc.), activated carbon materials and bricks. The reaction probabilities of different materials presented in literature [35,40,41,45,48,50,79–85,90,93,103–108], which are believed to be system-independent and material specific, are summarized in Fig. 3. Mean value of the reaction probability of a material is chosen if there are multiple sets of experiments of the same material in one paper. The reaction probabilities of different materials range from 10^{-8} to 10^{-4} . Among these, synthetic materials range several orders of magnitude difference in reaction probabilities, approximately from 10^{-8} to 10^{-5} , owing to that synthetic materials covered various kinds of materials, such as PVC, rubber and nylon, which have quite different chemical components. Similarly, the reaction probabilities of ceiling tiles, gypsums and carpets also range in orders of magnitude difference.

The reaction probabilities of building materials depend on the material chemical compounds and surface characteristics. Generally, the unsaturated organic compound (e.g. cotton, wool, and carpet) are highly reactive with ozone, and may produce oxidized by-products. Some materials containing clays (e.g. brick, gypsum, clay-based paintings and ceiling tiles) consume ozone readily while exhibits negligible by-products yield, probably due to the reaction catalyzed by metals presented in the clay [109].

However, materials composed of clays and other inorganic ingredients are not necessarily good at removing ozone. Factors such as morphology and available surface area of the materials are also important for ozone removal. For example, clean glass has quite low ozone removal rate. However, by contrast, the ceiling tile composed of perlite, an expanded high-surface-area volcanic glass, had a moderate deposition velocity and reaction probability [35]. Besides, ceramic tiles in

Table 2
CFD settings of modelling indoor ozone removal in literature.

Turbulence	Pressure-velocity Algorithm	Discretization Scheme	Δy_1 [mm]	Sc_t	Reference
Low Re k-e	SIMPLE	n/a	0.58	0.9	Sørensen and Weschler, 2002 [94]
Low Re k-e	SIMPLE	Momentum: QUICK Energy: second-order upwind Pressure: second-order	0.006	n/a	Ito, 2007 [83]
Realizable k-e	n/a	Momentum: second-order upwind Energy: second-order upwind Pressure: second-order	1.5	0.9	Russo and Khalifa, 2010 [95]
Low Re k-e	SIMPLE	Momentum: QUICK Energy: second-order upwind Pressure: second-order	n/a	1.0	Ito and Harashima, 2011 [96]
RNG k-e	SIMPLE	Momentum: second-order upwind Energy: second-order upwind Pressure: PRESTO!	2	n/a	Rai et al., 2012 [97]
Laminar	SIMPLE	Momentum: second-order upwind Energy: second-order upwind Pressure: PRESTO!	0.1	0.7	Gao and Zhang, 2012 [98]

n/a: not available in literature.

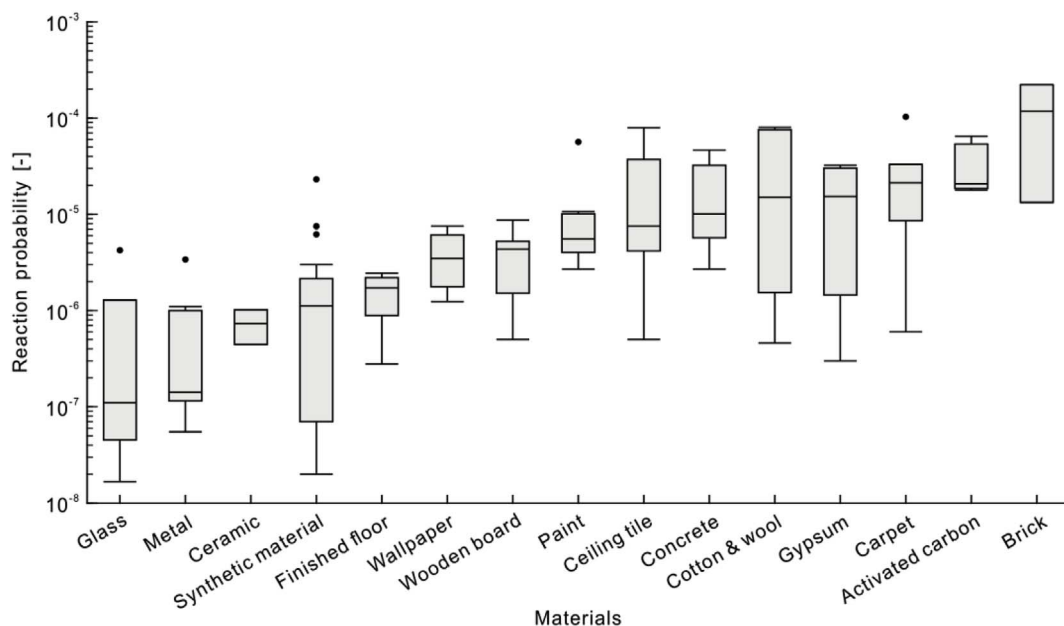


Fig. 3. Reaction probabilities of different materials presented in literature [35,40,41,45,48,50,79–85,90,93,103–108]. Mean value of the reaction probability of a material is chosen if there are multiple sets of experiments of the same material in one paper. In each box, the mid-line shows the median value, the top and bottom of the boxes show the upper and lower quartiles (the 75th and 25th percentiles), and the top and bottom of the whiskers represent the 90th and 10th percentiles. The extreme values farther from the median than 1.25 times the whisker end are drawn with points.

Lamble et al. [45] and in Hoang et al. [35] exhibited low deposition velocities, even though they are composed of clay. Ceramic tiles were with smooth surface and the clay composite would have been modified in the kiln. The clay plaster, with very high ozone deposition velocity, has substantial surface roughness and porosity. Therefore, generally, fleecy and porous materials, e.g. activated carbon cloth and carpet, exhibit higher reaction probabilities than smooth, non-porous surfaces, e.g. glass, metal and ceramic. Lin and Hsu [40] investigated the impact of material physical properties on ozone removal for 8 building materials and proposed that the specific surface area of the building material is more closely related to ozone reaction probability than total pore volume of building materials. It is probably because ozone reactive compounds could have better diffusion in more small pores compared to less big pores when total pore volumes are the same. Gall et al. [80] described the impact of internal reaction and diffusion on ozone removal by Thiele modulus ϕ which relates the rate of reaction to the rate of diffusion through the material substrate and can be described by

$$\phi = \left(\frac{4k''z^2}{d_{p,A}D_e} \right)^{0.5} \tag{9}$$

where k'' is the surface area-normalized reaction rate constant (cm/s), z is the length of a pore (cm), $d_{p,A}$ is the area-weighted average pore diameter (cm), and D_e is the effective diffusion coefficient in a test material of the indicated species (cm²/s). Value of ϕ less than 1 indicates that reaction processes are slow relative to diffusion processes and ozone removal is reaction-limited, while value of ϕ greater than 1 indicates reactions processes are quicker relative to internal diffusions and ozone removal is internal diffusion-limited. When the materials are reaction-limited, the ozone deposition velocities and reaction probabilities increased with increasing in material thickness owing to the internal area throughout the entire ranges of thicknesses contributing to the determined deposition velocity. In an internal transport-limited condition, additional thickness would not increase reactions [110].

Ozone deposition velocity depends on the reaction probability of material and the air flow condition near the material surface. Fig. 4 illustrates the relationship between deposition velocities and reaction probabilities in different air flow conditions in literature. Points indicated the measured results [35,40,41,45,48,50,79,82,84,85], and dot

lines indicated the analytical results [34] in different flow conditions. In the case of lower reaction probabilities, the measured results are concentrated and highly consistent with the analytical results, which indicates that the deposition velocities are reaction-limited and insignificantly affected by the air flow conditions. In the case of higher reaction probabilities, the influence of air flow conditions to ozone deposition velocities become much more significant. The measured results become scattered owing to the varied flow conditions in experiments. This finding is consistent with the conclusion presented by Cano-Ruiz et al. [34] that ozone deposition was transport-limited when $\gamma > \sim 3 \times 10^{-4}$ for typical indoor air flow conditions, and that ozone deposition was reaction-limited when $\gamma < \sim 5 \times 10^{-7}$. According to Figs. 3 and 4, there are few building materials with γ greater than 3×10^{-4} , which indicates that the ozone removals on most indoor materials are reaction-limited or dependent on both reaction and transport.

3.2. Surface treatment

Surface treatment is quite common for indoor building materials, e.g. materials with coverings or paints. The paints or covering materials are probably more important than the underlying material in determining ozone deposition velocities. Hoang et al. [35] observed that materials with coatings or pigments exhibited lower ozone deposition velocities, probably because surface reaction sites were covered by coating or pigment components which were less reactive with ozone. Poppendieck et al. [48] observed that ozone deposition velocities for gypsum wallboard backings were around one to two orders of magnitude greater than those for coated gypsum wallboards. Kunkel et al. [39] also observed approximately 41% higher of ozone deposition velocities for paperless gypsum wallboard compared to the gypsum wall board covered by wallpaper. These results [39,48] suggest that painting and wallpaper can provide a relatively effective barrier to ozone transport and reactions with the underlying gypsum substrate. Grøntoft and Raychaudhuri [90] proposed that the deposition velocity on surface treated woodwork probably depends on the chemical composition and gas permeability of the surface coating, as well as the composition and porosity of the underlying wood substrate. The available values in Grøntoft and Raychaudhuri [90] indicate a ranking of deposition

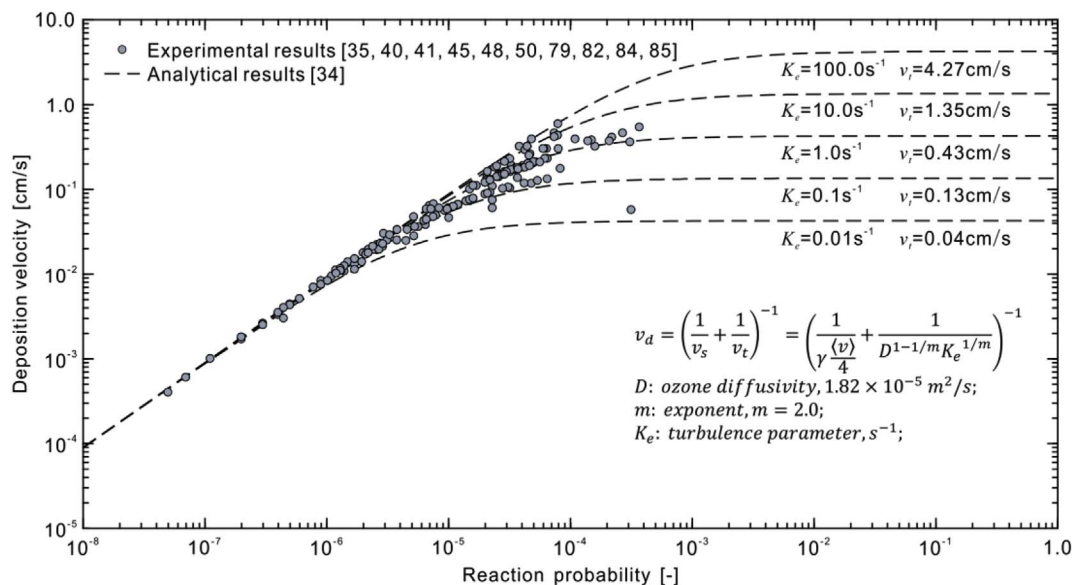


Fig. 4. Relationship between deposition velocities and reaction probabilities in different air flow conditions in literature [35,40,41,45,48,50,79,82,84,85].

velocities with painted > oiled > lacquered wood and with particle and fibreboards and plywood > whole wood board.

3.3. Transport-limited ozone removal rate

Reaction probabilities can reveal the ozone removal abilities of indoor materials. However, specifying materials with high reaction probabilities may not always result in substantial ozone removal due to transport limitations near the material surfaces. Values of transport-limited deposition velocities in rooms can be inferred from Wilson [111] ranged from 0.07 cm/s to 0.20 cm/s, and from Lai and Nazaroff [112] ranged from 0.02 cm/s to 0.23 cm/s. Morrison et al. [86–88] developed the deposition velocity sensor (DeVS) method to evaluate v_t in different kinds of rooms. Fig. 5 shows the range of v_t observed in offices, laboratories and apartments [86–88]. The results demonstrate that v_t are mainly within the range between 0.1 cm/s and 0.7 cm/s. Values of v_t can vary over an order of magnitude in the same room and can be influenced by location within the room, ventilation condition,

occupant movement, and even indoor furnishings which may change the air movement [64]. Areas with stronger air movement in a room tend to exhibit higher v_t . Morrison et al. [88] measured values of v_t between 0.06 cm/s and 0.09 cm/s under cabinets and desks, v_t between 0.12 cm/s and 0.14 cm/s in areas near hoods and operating computers, and v_t around 0.52 cm/s near a window and a supply vent.

Values of v_t in majority of chamber experiments are generally consistent with v_t observed in field tests, approximately between 0.1 cm/s and 0.8 cm/s (the boxplot in Fig. 5) [35,38,40,41,45,48,50,79,80,82,84,85,90]. The markers of the boxplot demonstrate some extreme values of v_t in chamber experiments. For example, Lin and Hsu [40] observed extremely high v_t between 10.94 cm/s to 17.89 cm/s, even more than an order of magnitude than the results of other experiments. Considering that the experiment was conducted in a large-scale chamber with some mixing fans, the highly rigorous airflows might occur near the material surface owing to the blowing of the mixing fans.

3.4. Chemistry products

Reactions between ozone and material surfaces may result in the formation of oxidized by-products, including C1-C13 carbonyls, dicarbonyls and hydroxycarbonyls [36–38,41–54], which can adversely affect occupants' health and perceived air quality [18,29,45,55–63]. These by-products are usually produced from reactions between ozone and the unsaturated organic compounds that constituting or adsorbed on material surfaces [29]. For example, previous studies observed high by-products yields dominated by C9 aldehyde for various kinds of carpets [28,37,38,42,44–47,51]. Human-related organic compounds adsorbed on material surfaces, e.g. adherent human skin oil, may also contribute a lot to ozone-initiated by-products with building materials. Human skin oil is highly reactive with ozone and may result in high yields of oxidized by-products, which mainly consist of acetone, C9-10 aldehydes, 6-methyl-5-hepten-2-one and 4-oxopentanal [52,113]. However, inorganic materials usually exhibit negligible by-products yields [37,38,45,64]. Therefore, some inorganic materials, e.g. bricks, clay-based plasters and perlite-based ceiling tiles, are usually considered as the most promising PRMs, since they can remove substantial ozone while yield negligible by-products [64]. Some of these by-products with low vapor pressures can nucleate to new particles or condense on existing particles to form secondary organic aerosols [65–68].

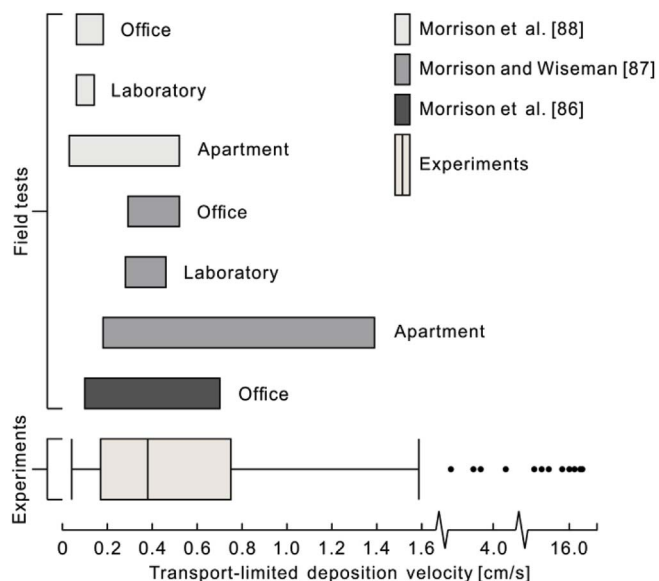


Fig. 5. Transport-limited deposition velocities in field tests [86–88] and experiments [35,38,40,41,45,48,50,79,80,82,84,85,90] in literature.

4. Ozone surface removal under different conditions

4.1. Impacts of ozone concentration and environmental conditions on ozone removal

Ozone concentration and environmental conditions such as air flow, relative humidity and temperature can influence the ozone removal on surfaces to different extents. Understanding the impacts of ozone concentration and environmental conditions on ozone removal can be important for better evaluating the indoor ozone concentrations in different conditions. However, owing to the limited investigations, the impacts of ozone concentration and environmental conditions on ozone removal are still uncertain. Thus, further investigations are warranted.

4.1.1. Impact of ozone concentration

According to the experimental conditions listed in Table 1, the ozone concentrations in most of the publications were less than 300 ppb, approximately within the normal range of ambient ozone concentrations. Within such a range of ozone concentrations, no significant impact of ozone concentrations on ozone surface removal was observed [35,40,45,48,50,79–85,90,104,105,108]. Poppendieck et al. [48] conducted a chamber experiment with the inlet ozone concentrations between 1000 and 1200 ppm (i.e. 1.0×10^6 – 1.2×10^6 ppb) to investigate the ozone deposition in the case of building disinfection, which the ozone concentrations were several orders of magnitude greater than the usual ozone concentrations conducted by other studies (less than 300 ppb). According to their results, initial ozone deposition velocities for most tested materials were similar to those reported in literature with much lower experimental ozone concentrations, but decayed rapidly as reaction sites on material surfaces were consumed. As shown in Fig. 6, the final reaction probabilities of tested materials were much lower (nearly an order of magnitude for some materials) than the reaction probabilities reported in other literature. Therefore, such high ozone concentrations are believed to have rapidly consumed a majority of available reaction sites for most materials, resulting in the declined reaction probabilities. Molar yields of oxidized by-products have been observed to decrease for some materials as the ozone concentration is raised [45,48,64,84].

4.1.2. Impact of air flow in chamber

The air flow in chamber, which depends on the volume and the flow rate of the chamber, can significantly influence the ozone surface removal rate due to that ozone removal depends on the fluid mechanics

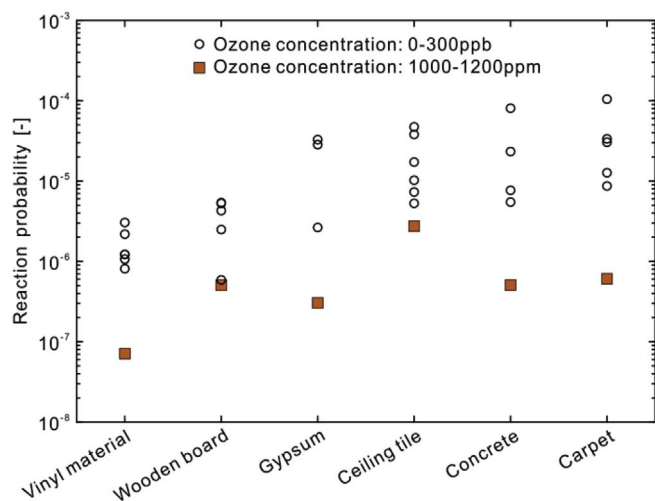


Fig. 6. Reaction probabilities of some materials under normal (0–300 ppb) [35,40,45,48,50,79–85,90,104,105,108] and extremely high (1000–1200 ppm) [48] ozone concentrations.

near the material surface. The environmental chambers in previous investigations included large-scale chambers, small-scale chambers, field laboratory emission cells and tube flow reactors. The experimental conditions in literature are shown in Table 1. According to the previous studies on ozone removal rates of some kinds of materials (Table 3) [35–40,45,47–50,79,80,84,85,89,90], the measured reaction probabilities of the same material in different papers are generally in good agreement even with different flow conditions, which indicates that the air flow in chamber has insignificant impact on the reaction probabilities of materials. For example, the overall reaction probabilities for carpet, painted gypsum board and activated carbon were found to be in the order of 10^{-5} . The reaction probabilities for unpainted gypsum board were in the order of 10^{-6} to 10^{-5} , while for pertile-based ceiling tile, those were in the order of 10^{-6} . The much lower values of reaction probabilities for carpet and painted gypsum board ($\sim 10^{-7}$ – 10^{-6}) observed by Poppendieck et al. [48] were probably associated with the extremely high ozone concentrations (1000–1200 ppm). Such high ozone concentrations were believed to have rapidly consumed a majority of available reaction sites for these materials, which resulted in the lower observed reaction probabilities [48].

However, the deposition velocities of the same materials measured in different papers varied a lot, which might be due to varying degrees of material aging, or differences of air flow in experimental chambers [38]. The results observed in FLEC chamber in Kleno et al. [89] were generally higher than the results in other investigations. Kunkel et al. [39] speculated that this may be due to a boundary layer that was not fully developed above the material surface in such a small volume and with a high air change rate of the FLEC environment, which leads to reduced transport resistance and greater deposition velocity.

4.1.3. Impact of relative humidity

There is no certain conclusion about the impact of relative humidity (RH) on ozone surface removal. According to the previous studies, the impact of humidity on ozone surface removal generally depended on the nature of the material surface. Weschler [18] presented that the higher the relative humidity, the larger the deposition velocities of ozone to material surfaces, albeit the effect is relatively small. He proposed that the magnitude of this effect varies with the nature of the material surface, i.e. the more hydrophilic the surface, the larger this effect, which was in agreement with the results reported by Cox and Penkett [106] for aluminum, Mueller et al. [107] for aluminum, Reiss et al. [93] for latex paint, and Valuntaitė et al. [114] for polyethylene sheet.

Grøntoft et al. [90,91] also presented that, for majority of building materials, increase in RH would lead to higher ozone surface deposition. However, for some materials (e.g. calcareous stone and concrete), they proposed that at low RH range (e.g. from 0% to 50% RH), increase in RH may not necessarily lead to higher ozone surface deposition velocity, while increase in RH at higher RH range would lead to higher deposition velocity, which was in agreement with the results reported by Nicolas et al. [47] for pine wood and Kunkel et al. [39] for activated carbon. According to Grøntoft et al. [91], for this kinds of materials, at low RH range, more adsorbed water on the material surface due to increase in RH would form a monolayer water film on the material surface and prevent direct contact of ozone molecules with material surface. However, as further increase in RH at high RH range, more water would be adsorbed on the material surface, and the bond between the new layer of adsorbed water and the material surface becomes relatively weaker. This phenomenon would cause new layer of adsorbed water to be available for reaction with ozone molecules, thereby causing higher ozone surface deposition at higher RH range.

However, some other investigations observed no significant impact of relative humidity on ozone deposition. Sabersky et al. [81] observed little influence of the RH on the ozone deposition for Lucite. No significant effect of RH on the ozone deposition onto vinyl and paper wallpaper was observed by Reiss et al. [93]. Hoang et al. [35] observed

Table 3
Measured deposition velocities and reaction probabilities of some materials in literature.

Material	v_d [cm/s]	v_t [cm/s]	γ [-]	V [L]	λ [h ⁻¹]	Reference
Carpet	0.04–0.11	0.17	6×10^{-6} – 3×10^{-5}	10.5	6.9	Morrison and Nazaroff, 2000 [50]
	0.05–0.12	0.75 ^a	6×10^{-6} – 2×10^{-5}	10.9	11	Grøntoft and Raychaudhuri, 2004 [90]
	0.01–0.04	0.04–0.11	3×10^{-7} – 7×10^{-6}	48	0.5	Poppendieck et al., 2007 [48]
	0.04–0.08			17	12	Nicolas et al., 2007 [47]
	0.25–0.54	0.41–0.65	5×10^{-5} – 4×10^{-4}	10.5	17.1/22.9	Coleman et al., 2008 [84]
	0.04–0.15			4.25	28.2	Wang and Morrison, 2009 [49]
	0.11–0.14	0.23	2×10^{-5} – 4×10^{-5}	10	12.3	Lamble et al., 2011 [45]
	0.05–0.08			48	2	Cros et al., 2012 [37]
	0.15–0.21	0.35–1.00	3×10^{-5} ^b	68000	1.1 ^c	Gall et al., 2013 [38]
	0.15–0.23	0.35–0.41	3×10^{-5} – 4×10^{-5}	10.7	10	Rim et al., 2016 [85]
	0.06–0.19		52	3	Abbass et al., 2017 [36]	
Unpainted gypsum board	0.8			0.107	504.7	Kleno et al., 2001 [89]
	0.10–0.13	0.75 ^a	1×10^{-5} – 2×10^{-5}	10.9	11	Grøntoft and Raychaudhuri, 2004 [90]
	0.07			17	12	Nicolas et al., 2007 [47]
	0.04–0.08			14200	^c	Kunkel et al., 2010 [39]
	0.02–0.03	13.06–17.61	3×10^{-6} – 4×10^{-6}	216	0.5 ^c	Lin and Hsu, 2015 [40]
	0.15–0.23	0.35–0.41	3×10^{-5} – 7×10^{-5}	10.7	10	Rim et al., 2016 [85]
Painted gypsum board	0.03–0.67			0.107	504.7	Kleno et al., 2001 [89]
	0.00–0.01	0.06	2×10^{-7} – 3×10^{-7}	48	0.5	Poppendieck et al., 2007 [48]
	0.18	0.34	4×10^{-5}	10	12.3	Lamble et al., 2011 [45]
	0.02–0.04			48	2	Cros et al., 2012 [37]
	0.01–0.02			68000	1.1 ^c	Gall et al., 2013 [38]
Pertile-based ceiling tile	0.05	0.09	1×10^{-5}	48	1	Hoang et al., 2009 [35]
	0.06	0.58	7×10^{-6}	10	12.3	Lamble et al., 2011 [45]
	0.06–0.09			48	2	Cros et al., 2012 [37]
	0.06–0.07	0.17–0.70	9×10^{-6} ^b	68000	1.1 ^c	Gall et al., 2013 [38]
Activated carbon cloth	0.14	0.52	2×10^{-5}	10.9	11	Grøntoft, 2002 [79]
	0.13–0.14	0.75	2×10^{-5}	10.9	11	Grøntoft and Raychaudhuri, 2004 [90]
	0.11–0.22			14200	^c	Kunkel et al., 2010 [39]
	0.07–0.11			48	2	Cros et al., 2012 [37]
	0.12–0.43	0.17–1.10	4×10^{-5} – 8×10^{-5}	10	11.7	Gall et al., 2014 [80]

^a The values of v_t are from Grøntoft [79].

^b The reaction probabilities are from Lamble et al. [45].

^c Using additional mixing fans in chamber.

that within the range of tested RH, ozone deposition velocity varied only slightly with variations in RH for tested materials. Similarly, Kunkel et al. [39] reported that ozone deposition velocities for gypsum wallboard were not affected by the varied RH. Lamble et al. [45] didn't report any statistically significant effects of RH on ozone deposition for tested materials. Gall et al. [38] found minimal impacts of RH on ozone deposition velocities for some green building materials. Rim et al. [85] suggested a minimal impact of RH on ozone deposition velocity for the tested samples of three widely used indoor materials. Therefore, further investigations of ozone deposition of materials at a variety of RH are warranted.

4.1.4. Impact of temperature

Some investigations [18,85,114] presented that increase in temperature would increase ozone deposition velocity. However, most investigations considered that the increase in ozone deposition velocities with the increasing temperature for studied materials was so moderate as to be within the range of measurement uncertainty [18,85]. Besides, Lamble et al. [45] didn't observe any statistically significant impacts of temperature on ozone deposition velocity for tested materials. Therefore, the present paper suggests a negligible impact of temperature on ozone deposition velocity.

4.2. Time-dependent ozone surface removal

Most of the previous researches investigated the ozone removal on unused new materials. Few researches have been conducted to study the time-dependent ozone removal. However, the time-dependent

ozone surface removal is important for evaluating the ozone removal ability of materials for a period of ozone exposure, particularly for a long-term period.

4.2.1. Ozone removal during short-term period

Some previous researches [35,41,48,85] conducted chamber experiments to determine the time-dependent deposition velocities of materials using Eq. (5), which can also be solved in a discrete form:

$$v_d(t = t^{ave}) = \frac{(2/\Delta t)[C_{in}^n - C_{in}^{n+1}] + \lambda[C_{out}^{n+1} + C_{out}^n - C_{in}^{n+1} - C_{in}^n]}{(A/V)[C_{in}^{n+1} + C_{in}^n]} \quad (10)$$

where n and $n + 1$ indicate consecutive data points, and t^{ave} is the time midway between the times corresponding to data points at n and $n + 1$. Many investigations have observed the decreasing reactivity of the material surface with increasing cumulative ozone exposure, which can be termed aging effect [34,35,41,43,45,48,50,78,81,84,85]. The ozone deposition velocity of the material is highest during the initial ozone exposure, probably due to the abundant reactive sites on the material surface. Then for most materials there was a rapid decay in deposition velocity with exposure to ozone. Following decay, the deposition velocity became nearly constant, albeit much lower than the initial value. A rapid decay in deposition velocities was likely because of a consumption of surface reactive sites. These surface sites might be associated with the material itself or with composition attached to the material surface, e.g. adherent human skin oil, deposited particles or adsorbed gases.

Aging effect was widely observed in almost all the materials in previous experiments. However, different materials exhibited varying extent of aging effect. Morrison et al. [41] proposed a power-law function to describe the relationship between the reaction probability and the cumulative ozone uptake on duct surfaces. Morrison and Nazaroff [50] found this relationship could also fit reasonably well for carpets. The power-law relationship can be expressed by

$$\gamma(t) = \alpha [U(t)]^\beta \tag{11}$$

where $\gamma(t)$ is the reaction probability at moment t , α and β are the fitted parameters, and $U(t)$ is the cumulative ozone uptake on surface, which can be computed by

$$U(t) = \int_0^t v_d C dt \tag{12}$$

Some investigations observed the regeneration of ozone reactivity after a period without exposure to ozone, followed by a second decay in deposition velocity [35,50,84,85]. Such regeneration was considered not to be due to the deposition of particles or adsorption of reactive gases onto the material surfaces during the period without ozone exposure, since the test materials were contained in the experimental chambers without any possible introduced source of particles or reactive gases. Instead, it is conceivable that reaction sites on material surfaces were consumed during ozone exposure, thus establishing a concentration gradient between reactive molecules in the test material matrix and the material surface. This would induce reactive molecular diffusion from the matrix to the surface of the material, resulting in effective surface replenishment of reaction sites. The differences in abundance of transportable reactive chemicals beneath the surface of the materials may result in different levels of regeneration, e.g. observed regeneration by Rim et al. [85] appeared smaller than that reported by Hoang et al [35].

Regeneration of ozone reactivity is potentially important in realistic indoor environments considering the diurnal variations of ambient outdoor ozone (a major source of indoor ozone) and intermittent usage of ozone-released devices indoors, e.g. using a laser printer during a period of time in a room. Rim et al. [85] observed both surface aging and regeneration during a two-day period with diurnal ozone concentration variations, although the regeneration was not pronounced during the experimental period. Fig. 7 shows a schematic illustration of aging and regeneration effects.

4.2.2. Ozone removal during long-term period

Removal of indoor ozone by building materials has been quantified through several experimental studies. However, most of these studies

were short-term studies (i.e. up to 48 h of ozone-exposure). Long-term ozone exposure on materials in real scenario may change the reactivity of material surfaces, which may influence the ozone removal on surfaces. Besides, the occupant activities indoors may also influence the material reactivity and ozone removal on surfaces. However, there haven't been abundant studies focused on long-term evaluations of ozone removal on building materials.

Rim et al. [85] measured ozone surface deposition in an occupied office for 2 months. After the first month of exposure, the deposition velocity decreased by 22%, 15%, and 16% for painted drywall, carpet, and ceiling tile, respectively. After the second month in the field, deposition velocities increased compared to the first month values for carpet and ceiling tile and were similar to the first month values for painted drywall. The initial decay of deposition velocity was likely due to the consumption of available surface reaction sites. During the second month of exposure, carpet and ceiling tile regained some of their surface ozone reactivity, perhaps owing to soiled by particles or organic components emitted from occupants and their activities [77,102].

Cros et al. [37] conducted a field study in office and houses over a 6-month period (summer 2009 to winter 2010) and intermittently conducted chamber experiments on materials samples retrieved from the field. Ozone deposition velocities for activated carbon and ceiling tile exhibited little variation over time. The deposition velocity for painted gypsum wallboard decreased by 30% for the first 2 months and then converged to a relatively low value. The deposition velocity for carpet was high initially, but decayed significantly during the test period. After 6 months in the field, the deposition velocity for carpet decreased by about 30% than it did initially.

Wang and Morrison [46] conducted field studies in five homes over a period of 1.5 years during three seasons (summer 2005, summer 2006 and winter 2007). A significant decrease was observed in reaction probabilities for three homes from summer 2006 to winter 2007, while a consistent increase in reaction probabilities observed in another home, which may be due to a high rate of surface replenishment with reactive compounds such as cooking oils or skin oils caused by increased occupants. The observed seasonal decline (summer 2006 to winter 2007) of reaction probability was consistent with the observation of decrease in deposition velocities of painted gypsum wallboard and carpet by Cros et al. [37] (summer 2009 to winter 2010). An explanation is that the seasonal decline is probably related to the observation that the ozone deposition velocity tends to increase with increasing relative humidity [93,106,107] since indoor conditions were dryer during winter than summer. However, during the experimental period of Wang and Morrison [46], the relative humidity over the surfaces was maintained at 50%. Cros et al. [37] also observed weak associations between field conditions (including relative humidity and

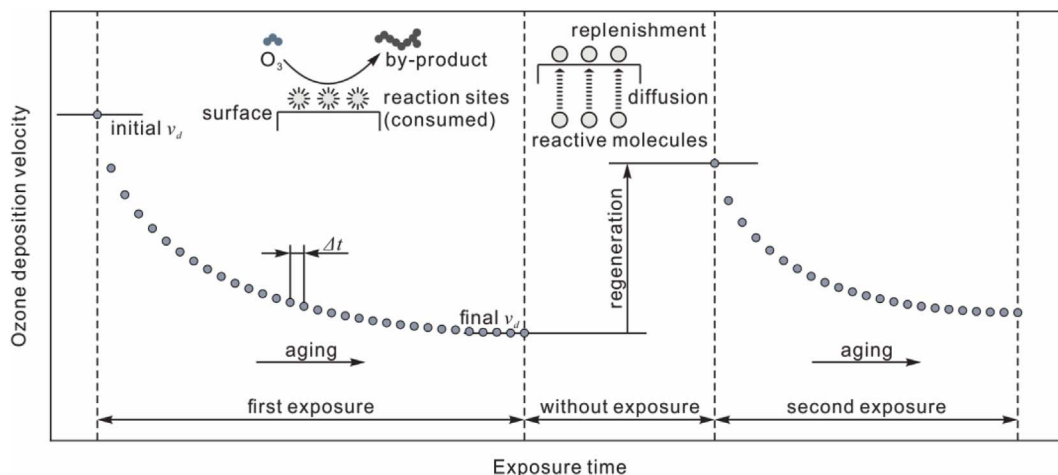


Fig. 7. Schematic illustration of aging and regeneration effects.

temperature) and ozone deposition velocities. Thus relative humidity probably may not be the actual influence factor. Additional researches are warranted to further explore and understand the observation of the variation of reaction probabilities. However, over the whole 1.5-year period, no statistically significant trend of reaction probabilities was observed. The aging effect of ozone on building materials may be too slow to recognize over this period. However, Wang and Morrison [46,49] measured the ozone deposition in different aged homes, ranged from 1 years old to 14 years old. They observed carpet surfaces in older homes were less reactive than in newer homes. But the reaction probabilities on kitchen countertop did not exhibit any significant decline, probably owing to occupant activities such as cooking and cleaning which replenish surface reaction sites.

According to the above discussions, materials may become less reactive after a relative short period of ozone exposure, e.g. several months, which is likely due to the consumption of available surface reaction sites. However, during a longer-term exposure to ozone, some materials in occupied environments may exhibit remained surface reactivity on account of the replenishment of the reactive compounds on material surfaces, which is believed to be closely associated with human occupants or occupant activities. Squalene, which constitutes around 5–15% of skin oil [115], is believed to be responsible for much of such increase in reactivity owing to its abundant unsaturated carbon bonds [116].

5. Conclusions

Building materials can be significant sinks for indoor ozone, because of the irreversible heterogeneous reactions between ozone and material surfaces. Therefore, the ozone removal on material surfaces is crucial for evaluating indoor ozone concentrations and human exposure. The previous investigations observed that the reaction probabilities of common indoor building materials range from 10^{-8} to 10^{-4} [34], and depend on the material chemical compounds and surface characteristics. Generally, unsaturated organic compounds and some materials containing clays have higher reaction probabilities, probably owing to the high reactivity with ozone, and the reaction catalyzed by metal in the clay, respectively [109]. Fleecy and porous materials, e.g. activated carbon cloth and carpet, usually exhibit higher reaction probabilities than smooth, non-porous surfaces, e.g. glass, metal and ceramic [35,45]. For surface treatments, the surface-treated materials such as paints and covering materials are probably more important than the underlying material substrate in determining ozone deposition velocities [39,48]. Ozone removal on material surface is also associated with the fluid mechanics near the surface. The results of field tests in previous investigations demonstrated that v_t observed in offices, laboratories and apartments are mainly within the range between 0.1 cm/s and 0.7 cm/s [86–88]. Values of v_t in majority of chamber experiments are generally consistent with v_t observed in field tests, approximately between 0.1 cm/s and 0.8 cm/s [35,38,40,41,45,48,50,79,80,82,84,85,90]. Reactions between ozone and unsaturated organic compounds that constituting or adsorbed on material surfaces may also result in oxidized by-products yields, while inorganic materials usually exhibit negligible by-products yields [28,37,38,42,44–47].

The air flow in chamber, which depends on the volume and air change rate of the chamber, usually has insignificant impact on the reaction probabilities of materials [35–40,45,47–50,79,80,84,85,89,90]. The ozone concentrations in most studies were less than 300 ppb, approximately within the normal range of ambient ozone concentrations. No significant impact of ozone concentrations on ozone surface removal was observed within such a range of ozone concentrations [35,40,45,48,50,79–85,90,104,105,108]. However, when the ozone concentrations are extremely high, e.g. over 1000 ppm in the case of building disinfection, the reaction probabilities of tested materials were much lower than the reaction probabilities reported within the normal

range of ozone concentrations, because such high ozone concentrations are believed to have rapidly consumed a majority of available reaction sites for most materials [48]. There is no certain conclusion about the impact of relative humidity on ozone surface removal, while the impact of temperature is believed to be negligible [16,18,45]. Many materials exhibited aging effect, decreasing reactivity of the material surface with increasing cumulative ozone exposure during short-term period [34,35,41,43,45,48,50,78,81,84,85]. Some materials exhibited re-generation of ozone reactivity after a period without exposure to ozone [35,50,84,85]. However, during long-term period, no statistically significant trend of reaction probabilities was observed, probably due to occupant activities such as cooking and cleaning which replenish surface reaction sites [46,49].

Acknowledgements

The research was supported financially by the national key project of the Ministry of Science and Technology, China on “Green Buildings and Building Industrialization” through Grant No. 2016YFC0700500. We would like to appreciate Liyan Rui and Zhe Yang for their selfless assistance in literature searching and article writing.

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