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1 Mass accretion and ozone reactivity of idealized indoor surfaces in mechanically or naturally
2 ventilated indoor environments

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11 **Abstract**

12 In indoor environments, accretion of mass to materials may provide sites for surface chemistry that differ
13 from those of the original material. Since indoor surfaces are a major sink of oxidant gases, surface mass
14 accretion may impact indoor O₃ chemistry. In this study, the effect of surface mass accretion on O₃
15 surface deposition was tested by deploying cleaned borosilicate glass plates in two types of indoor
16 environments: a mechanically ventilated (MV) office and a naturally ventilated (NV) residence located in
17 Singapore. In each environment, seven replicate glass plates and one field blank were deployed for
18 between 7-56 days and examined in a laboratory chamber for O₃ deposition rate and surface reaction
19 probability. Average mass accretion to plates, deployed in a horizontal position and including deposited
20 particles, was 10.6 mg/(m² d) in the MV office vs. 18.5 mg/(m² d) in the NV residence and the
21 comparison is at the threshold of statistical significance ($p = 0.054$). Ozone reactivity to the plates
22 increased in magnitude and persistence with longer plate deployment. Ozone reaction probabilities to
23 cleaned plates prior to deployment ranged $[0.06-0.74] \times 10^{-6}$ for two hours of observable removal whereas
24 plates deployed for 56 days ranged $[0.15-1.2] \times 10^{-6}$ for four hours of observable removal. Regressions of
25 cumulative O₃ removed during chamber tests vs. mass accreted show removal of 4.3 nmol O₃/mg for the
26 NV residence and 2.4 nmol O₃/mg for the MV office. These results imply that accretion of mass to
27 surfaces may alter indoor O₃ transport and transformation pathways.

28

29 **1 Introduction**

30 Exposure to elevated O₃ is associated with cardiovascular effects, asthma, and increases in daily mortality
31 [1–3]. Humans are exposed to O₃ primarily in the small fraction of the atmosphere contained inside built
32 environments. The predominant source of indoor O₃ is from outdoor air where O₃ is formed due to
33 photochemical reactions that involve sunlight, volatile organic compounds (VOCs), NO_x, and carbon
34 monoxide [4]. Indoor O₃ levels are 20-70% that of outdoor levels mainly due to chemical reactions with
35 interior surfaces that remove O₃ from air [5]. In spite of lower indoor O₃ levels, total indoor O₃ exposures
36 typically exceed outdoor exposures because of the time spent in indoors [6–8]. Epidemiological studies
37 suggest a safe level of exposure to O₃ is below 10 ppb [9], a level which indoor spaces routinely exceed

38 [5]. Certain surface O₃ reaction pathways may generate byproducts, effectively trading indoor O₃ for
39 other harmful or irritating byproducts [10]. There is interest in developing beneficial indoor surface O₃
40 pathways to passively reduce human exposure to O₃, if removal results in no byproducts or only benign
41 byproducts [11,12].

42 In a typical indoor space, many exposed surfaces can react with O₃. A seminal study of indoor O₃
43 dynamics reported ozone decay constants for homes and decomposition rates for specific indoor surfaces
44 [13]. Subsequent studies have expanded our knowledge of indoor O₃ dynamics by characterizing reaction
45 rates with a wide variety of building materials [11,12,14–22], as well as identifying byproduct formed
46 from material-O₃ reactions [10,23–26]. Outcomes of studies of material-O₃ interactions generally report
47 material reactivity as a deposition velocity, v_d [27], or a reaction probability, γ [28]. Such
48 parameterizations enable material balance models of indoor O₃ that describe fate, transport, and
49 transformation of O₃ and O₃ reaction byproducts [16,29–31].

50 As materials react with O₃, so-called “aging” phenomena are observed where ozone reactivity decreases
51 with time. Over short time-scales (< 10 h), this has been observed in chamber tests where materials are
52 exposed to elevated O₃ that is alternated with air free of O₃ on a time scale of hours [13]. Under such
53 conditions, materials generally exhibit a reduction in material-O₃ reactivity with increasing exposures or
54 exposure periods [14,18,21]. This behavior is ascribed to exhaustion of reaction sites on the surface after
55 initial O₃ exposure, with subsequent O₃ removal rate-limited by diffusion of new reaction sites to the
56 surface [32].

57 Over longer time-scales, e.g., months of deployment in real buildings, prior studies demonstrate
58 variability in impacts on magnitudes of material-O₃ reactivity. In a study where four materials were
59 placed in field sites over a period of ~6 months, material-O₃ v_d increased modestly for ceiling tile,
60 decreased for carpet, and was largely unchanged for activated carbon and painted drywall [25]. Another
61 study of material-O₃ reaction showed that for three building materials deployed in an occupied office
62 environment, v_d decreased for painted drywall after 1 and 2 months of deployment while v_d for carpet and

63 ceiling tile decreased after 1 month, subsequently increasing to higher than the “fresh” reactivity after 2
64 months of placement in the field [18]. These studies posit that deposition processes occurring in the
65 indoor environment provide a mechanism by which gases and particles may associate with the surface, in
66 some cases replenishing O₃ surface reaction sites.

67 Accretion of low-volatility gases, particles, water, inorganic species and elemental carbon create a surface
68 film on indoor surfaces [33]. Previous studies show that sorption of low-volatility compounds and
69 deposition of particles impact the chemical composition of building and indoor surfaces. Studies of indoor
70 and exterior window surfaces demonstrate that window surfaces act as a reservoir for alkanes, polycyclic
71 aromatic hydrocarbons, polychlorinated biphenols, and pesticides [34,35]. Another study investigated
72 surface film formation by heating glass and aluminum materials that were initially clean and then exposed
73 to successively longer periods of an indoor air matrix [36]. Time exposed to indoor air was a strong
74 determinant of ultrafine particle formation, due to condensation of SVOCs that were volatilized from the
75 material during a controlled heating process. Settling of indoor dust may also alter surface chemical
76 composition. A study of the O₃ reactivity of indoor dust shows high rates of O₃ removal to dust, with
77 increases of C₇-C₉ aldehydes and other reaction byproducts produced from O₃-dust reactions [32].

78 Given the importance of indoor surfaces in governing indoor O₃ and O₃ reaction byproduct
79 concentrations, the objective of the present study is to investigate dynamics of surface mass accretion and
80 the resulting reactivity of indoor surfaces. This study addresses this knowledge gap by measuring how
81 surface film accretion impacts ozone deposition and surface reaction probability. While other studies have
82 explored this phenomena, this work uses glass plates as an idealized surface that reduces confounders
83 such as initial material composition, material aging, and material cleanliness in subsequent studies of
84 accreted mass. However, glass is also a common indoor material such that mass accretion can be taken as
85 representative of what can occur on certain surfaces in real indoor environments. The study results will
86 provide baseline data for how mass is accreted to initially inert, clean materials and subsequent ozone-

87 surface reaction dynamics in two typical indoor environments: 1) a naturally ventilated residence and 2) a
88 mechanically ventilated office.

89 **2 Methods**

90 **2.1 Glass plates**

91 Borosilicate glass plates were selected as surfaces to facilitate investigations of the dynamics of the mass
92 accumulated to indoor surfaces (i.e., a surface film of deposited particles and/or sorbed gases from indoor
93 air) and resulting ozone reactivity of the glass plate/surface film. Borosilicate glass plates were selected
94 due to the inertness and smoothness of glass and the known low uptake of O₃ to clean glass [19,30]. Glass
95 provides a low baseline from which subsequent signals of O₃ uptake to accreted masses can be more
96 readily differentiated. Borosilicate glass plates (McMaster-Carr) with horizontal projected surface area of
97 232 cm² (15.24 cm × 15.24 cm, thickness = 0.3175 cm) were used in this investigation.

98 Prior to deployment in the field, sixteen glass plates were cleaned by washing with laboratory-grade soap
99 and a soft sponge. All glass plates were then rinsed in water three times to ensure no soap film remained.
100 Glass plates were then cleaned with a lint-free wipe (Kimtech Kimwipes, Kimberly-Clark Professional)
101 dipped in high purity isopropyl alcohol (Levinson Scientific and Chemicals, IPA Technical Grade >99%)
102 and allowed to air dry in a fume hood in a vertical position. After drying, all plates were heated with a
103 heat gun for fifteen minutes to vaporize remaining solvent or other sorbed volatile species. All plates were
104 then placed in a chamber apparatus (details in Section 2.4) and were passivated of ozone reaction sites by
105 supplying approximately 500 ppb of ozone to the chamber for 8 h. All plates were then immediately
106 weighed (see section 2.3) to obtain the initial mass of the plate and placed in polypropylene storage bags
107 for ~1 week until deployment into the field environments. Tests of O₃ reactivity were made for two
108 randomly selected plates to establish a baseline ozone deposition velocity (v_d) to a clean plate.

109 **2.2 Deployment and collection of glass plates**

110 Seven test plates and one field blank plate were randomly selected from the set of sixteen plates for
111 concurrent deployment to two indoor environments: a naturally ventilated (NV) residential dwelling and a

112 mechanically ventilated (MV) office building. In both environments, the seven plates were placed on wire
113 racks such that the plates were horizontal to the ground (see Fig. S1 for an image of the deployment). A
114 field blank, a glass plate prepared as described in Section 2.1, but deployed to the field environment in the
115 polypropylene bag, was deployed to each of the field locations. In both locations, the temperature (°C),
116 relative humidity (%), and light (lux) levels were recorded in 5-minute interval (HOBO U12-012, Onset
117 Computer Corporation), shown in Fig. S2. In both the MV and NV location, plates were deployed for a
118 total of 56 days. Test glass plates were collected at approximately weekly intervals while a field blank
119 was collected after deployment for three weeks (NV residence) and eight weeks (MV office).

120 Racks used to support glass plates were placed in unobtrusive locations that were representative of typical
121 tabletop height surfaces in each environment. Racks were used to facilitate testing of ozone removal to
122 the downward facing side of the plate subsequent to the upward face. However, no observable O₃ removal
123 could be discerned to the downward facing plate. Results presented here are tests where only the upward
124 facing side of the plate was exposed to O₃ inside the chamber. In the MV office building, the wire racks
125 with plates were placed on a desk height table top, typical of an office environment where computer-
126 related work tasks are conducted. The office building is served by a demand-controlled ventilation system
127 that introduces outdoor air proportional to the amount of carbon dioxide in the space, and includes
128 mechanical systems for filtration (MERV 6-8 rated filters), cooling, and dehumidification. The ventilation
129 rate was not measured during the period of the deployment. The plates were placed on an L-shaped desk,
130 approximately 1 m in distance from the workstation where an office worker was present during normal
131 working hours. The zone was typically occupied by 5-15 office workers.

132 In the NV residential environment, the racks with the seven test plates and one field blank were placed on
133 a television stand in a living room at approximately 1 m in height. The residential environment was
134 occupied by two individuals, typically during non-working hours. The zone the plates were placed in was
135 served only by fan-assisted natural ventilation; ventilation and cooling were provided to the space by
136 window opening and fans that were operated to draw outdoor air into the room based on the preference of

137 the occupants. The typical schedule was windows closed with fans off during working hours, windows
138 open with fans on during evenings and weekends, and windows closed with fans off during sleeping
139 hours. The ventilation rate was not measured during the period of the deployment.

140 The two sites investigated were located within 1 km of each other in vicinity of the National University of
141 Singapore and we expect the outdoor conditions to be similar. We lack accompanying air quality data at
142 the field sites; in general, the Singapore National Environment Association reported Pollutant Standard
143 Index levels of 30-50 across the period of this study, indicating “good” to “moderate” outdoor air quality.
144 In such conditions, indoor particle and ozone concentrations are likely higher for the naturally ventilated
145 residence than the mechanically ventilated office because of no filtration with frequently open windows in
146 the residence. The MV and NV location were approximately a 5 min and 15 min walk, respectively, from
147 the laboratory where analyses occurred. The protocol developed for transporting plates for analysis
148 consisted of wearing new nitrile gloves to carefully place plates into polypropylene bags sized such that
149 there was no contact between the top surface of the plate and the bag. The plate was then placed in a
150 container to be immediately transported, in the same orientation as when placed in the field, to the
151 laboratory for weighing and analysis of ozone removal. It took less than two hours until the samples were
152 analyzed in the laboratory.

153 **2.3 Measurement of mass accretion to glass plates**

154 Initial plate mass was measured with an analytical balance (ME204, Mettler Toledo), with an absolute
155 uncertainty of ± 0.0001 g. While plates were nominally of uniform dimension and material, each plate was
156 tracked specifically as small variations in initial plate mass were noted ($\mu = 170.1751$ g, $\sigma = 0.44$ g). After
157 deployment to the field and subsequent return to the laboratory, each plate was measured by placing the
158 plate on a clean sheet of weighing paper and allowing the mass reading to stabilize over a period of
159 approximately 1 min and the change in mass from its initial “clean” measurement was noted. The
160 environmental conditions of the laboratory were different from that of both the MV office environment
161 and the NV residence. The 1 min duration was chosen to allow the reading to stabilize from the

162 disturbance of placement of the plate onto the scale, but to preclude longer-duration phenomena such as
163 the equilibration of sorbed water with the laboratory environment.

164 Average mass accretion rates to the plates deployed in the field were calculated according to eq. 1:

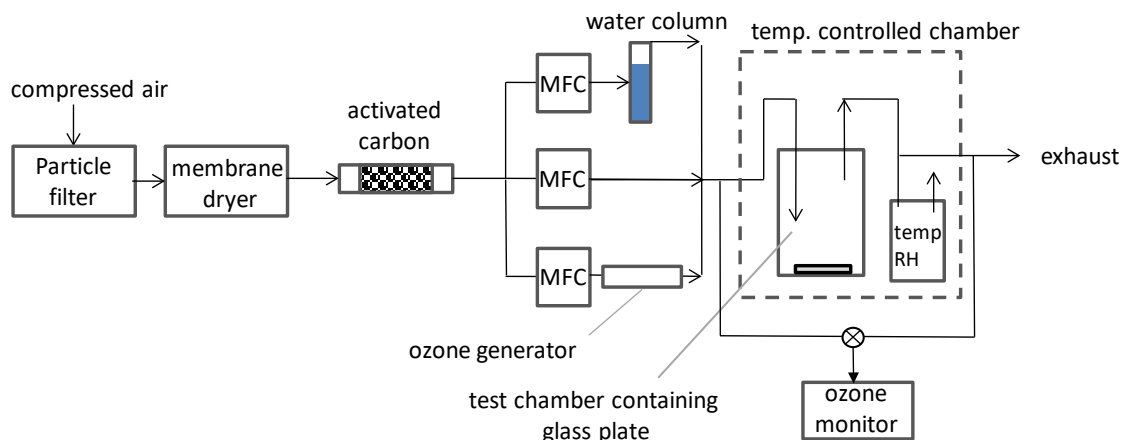
$\dot{m} = \frac{(m_f - m_i)}{t_d A}$	(1)
---------------------------------------	-----

165
166 where \dot{m} is the mass accretion rate to a test plate ($\text{g}/(\text{m}^2 \text{d})$), m_f is the final mass accreted to a test plate
167 after a field deployment (g), m_i is the initial mass of a cleaned plate prior to deployment (g), t_d is the time
168 duration the plate spent deployed in the field (d), and A is the exposed area of the top surface of the plate
169 (m^2). Since the bottom surface was also exposed to the environment, the normalization introduces a small
170 error as some mass may have accumulated on the bottom surface. The implications of this assumption are
171 discussed in Section 3.1.

172 **2.4 Test apparatus**

173 Ozone deposition velocities were measured in a laboratory chamber apparatus; a schematic of the
174 apparatus is shown in Fig. 1. Compressed laboratory air passed through a membrane dryer that included a
175 0.1 micron pre-filter (Laman MD-15LS and Laman SAM350-E, Air Parts Center Pte Ltd) to remove
176 particles and water vapor present in the compressed air. Dried, particle-free air was then passed through a
177 packed bed of activated carbon (BPL 6×16, Calgon Carbon) to remove ozone and volatile organic
178 compounds. The air flow was then split into three streams with flow rates set and maintained using mass
179 flow controllers (Omega FMA5500, Omega Singapore). One stream was used to humidify the airflow to
180 the desired setpoint based on the depth of deionized water in the water column and the flowrate through
181 the column. The second stream was an unmodified flow of dry, clean air that was mixed with the
182 humidified stream to maintain the desired relative humidity (RH) set-point. The third stream passed
183 through a stable ozone generator (U04/97-0066-02, UVP LLC). These three flow streams were re-
184 combined prior to entering a temperature controlled enclosure (KBE3.1, Binder GmbH) that was used to

185 maintain the air temperature and humidity to constant setpoints. Inside the temperature controlled
 186 enclosure was an 11.4 L electropolished stainless steel chamber (CTH-24, Eagle Stainless) in which glass
 187 plates were placed. A sensor was placed in an outlet stream from the test chamber to measure the chamber
 188 temperature and RH (U12-012, Onset Computer Corporation).



189
 190 **Fig. 1.** Schematic of test apparatus for measuring ozone deposition occurring on clean and field deployed
 191 glass surfaces. MFC = mass flow controller, RH = relative humidity.

192 2.5 Experimental protocol

193 Prior to a test of a glass plate, a test of background O₃ removal of the empty stainless steel chamber was
 194 conducted. Ozone was injected into the empty chamber at a stable until steady-state conditions, defined as
 195 changing less than 2 ppb in a 20 minute time period [37]. After conducting a background test of an empty
 196 chamber, a cleaned (week 0), field-environment deployed plate (weeks 1-7), or field blank glass plate was
 197 placed in the chamber and the O₃ injection was repeated. Ozone injections when glass plates were present
 198 were conducted for a period of four hours; longer test durations were explored but resulted in deposition
 199 velocities that were indistinguishable from background removal. A switching valve was used to
 200 periodically alternate between inlet and outlet concentrations of ozone (Model 202, 2BTech). Across all
 201 experiments, the average (mean ± 1 std. dev.) temperature and RH was 25.2 ± 0.057 °C and 49.3 ± 1.8%.
 202 Experiments were conducted with an average chamber flowrate of 2.2 ± 0.1 L/min (air exchange rate =
 203 11.5 /h). Inlet ozone concentrations to the test chamber were an average 96.8 ± 2.0 ppb.

204 2.6 Data analysis

205 2.6.1 Determination of v_d , v_b , γ

206 The background loss rate L_{bg} (h^{-1}) of ozone was calculated from steady-state data according to eq. 2:

$$\frac{dC_{outlet}}{dt} = 0 = \lambda C_{inlet} - \lambda C_{outlet} - L_{bg} C_{outlet} \quad (2)$$

207

208 where C_{inlet} and C_{outlet} are the inlet and outlet concentrations of ozone (ppb), t is time (h), λ is the air
209 exchange rate (h^{-1}) and L_{bg} is the background loss rate (h^{-1}) to stainless steel chamber surfaces. Ozone v_d
210 to background chamber stainless steel surfaces were calculated using the exposed area of the stainless
211 steel chamber surfaces (1810 cm^2) and the chamber volume ($11,400 \text{ cm}^3$). Ozone v_d to stainless steel
212 surfaces were, on average, $\sim 0.002 \text{ cm s}^{-1}$ and was determined in a background test prior to each
213 experiment. Ozone deposition velocity (v_d) to glass plates was determined using a transient mass balance
214 relationship that was solved numerically for the deposition velocity in 5-min interval, as shown in eq. 3:

$$v_d^t = \frac{V}{A_S} \cdot \frac{1}{C_{outlet}^t} \left[\lambda (C_{inlet}^t - C_{outlet}^t) - L_{bg} \left(\frac{A_{BG} - A_{S,p}}{A_{BG}} \right) C_{outlet}^t - \frac{C_{outlet}^t - C_{outlet}^{t+1}}{\Delta t} \right] \quad (3)$$

215

216 where v_d^t is the ozone deposition velocity at time t to the glass plate (cm s^{-1}), V is the volume of the
217 chamber (cm^3), A_S is the area of the top and sides of the glass plate (cm^2), C_{inlet}^t is the ozone concentration
218 entering the chamber at time t (ppb), C_{outlet}^t is the ozone concentration exiting the chamber at time t
219 (ppb), A_{BG} is the area of background chamber surface (cm^2), $A_{S,p}$ is the projected area of the glass plate
220 (cm^2), and all other terms as defined previously.

221 The total molar uptake of ozone to a test glass plate, Φ (mol), was determined as shown in eq. 4:

$$\Phi = \sum_{t=1}^n v_d^t \times \bar{C}_{\Delta t} \times A_S \times \Delta t \quad (4)$$

222

223 where $\bar{C}_{\Delta t}$ is the average ozone concentration (mol m^{-3}) in the chamber during the five-minute period, Δt ,
224 in which v_d^t is calculated.

225 Ozone deposition velocities were further parameterized according to resistance-uptake theory, where v_d
226 can be expressed in terms of a transport limited deposition velocity, v_t (cm s^{-1}) and a reaction probability,
227 γ (-), or the fraction of ozone-surface collisions that result in a reaction [14,28,38], shown in eq. 5:

$\frac{1}{v_d} = \frac{1}{v_t} + \frac{4}{\gamma \langle v \rangle}$	(5)
--	-----

228

229 where $\langle v \rangle$ is the Boltzmann velocity (cm s^{-1}).

230 The v_t was determined from an experiment in which cleaned glass was coated in a solution of 20 g of
231 potassium iodide (>99% purity, Sigma-Aldrich) dissolved in 25 mL of deionized water. The solution was
232 applied to a cleaned borosilicate glass dish with a small lip. The dish was then dried in the chamber with
233 cleaned, ozone-free air until a solid KI film coated the surface of the dish. An ozone deposition
234 experiment was conducted and the deposition velocity was determined using eq. 2. This condition
235 assumed negligible reaction resistance (i.e., $\frac{4}{\gamma \langle v \rangle} \ll \frac{1}{v_t}$) such that $v_d = v_t$. Since fluid dynamic conditions in
236 the chamber were held constant across all experiments, this allowed determination of γ in subsequent
237 experiments. The experimentally determined v_t ($\mu \pm \sigma = 0.57 \pm 0.028 \text{ cm/s}$) is shown in Fig. S3 and is in
238 general agreement with previously determined values in laboratory [18] and field environments [39].

239 To enable statistical comparison across dynamic estimates of ozone reactivity, we employed a metric that
240 characterized the time-variant reaction probability to enable regression analysis. First-order kinetics were
241 applied to the transient values of the reaction probability as shown in equation 6:

$\frac{d\gamma(t)}{dt} = -\beta \times \gamma(t)$	(6)
---	-----

242

243 where $\gamma(t)$ is the time-varying reaction probability (-), t is the time (min), and β (min^{-1}) is the first-order
244 decay constant of the reaction probability.

245 With the initial condition of $t = 0$, $\gamma(t) = \gamma(t = 0)$, the solution to eq. 6 takes the form $y = mx$ where y is
246 $-\ln\left(\frac{\gamma(t)}{\gamma(t=0)}\right)$, m is β , and x is the time during an experiment where reaction probability is being
247 measured. Higher values of β indicate a faster decay of the reaction probability than lower values of β .

248 2.6.2 Quality assurance

249 Experimental uncertainty in estimates of deposition velocities were determined using a propagation of
250 errors analysis for eq. 3, based on the manufacturer's specifications of instrument error. An uncertainty of
251 the greater of 2 ppb or 2% was used for the ozone monitor for inlet and outlet ozone measurements, 1.5%
252 of the reading for the mass flow controller for uncertainty on air exchange, while the error introduced due
253 to area and volume measurements of plates was assumed to be 1%. Uncertainty in calculated reaction
254 probabilities was taken as the sum of the experimental uncertainty for v_d and v_t in quadrature. Reported
255 uncertainties on absolute mass measurements are given as the repeatability of the balance summed in
256 quadrature for two measurements, or ± 0.14 mg. Uncertainties reported for cumulative ozone uptake are
257 reported from a propagation of errors analysis on eq. 4 considering uncertainty on v_d^t and the chamber
258 ozone concentration. The total uncertainty on molar uptake was determined by summing uncertainty at
259 each time step in quadrature.

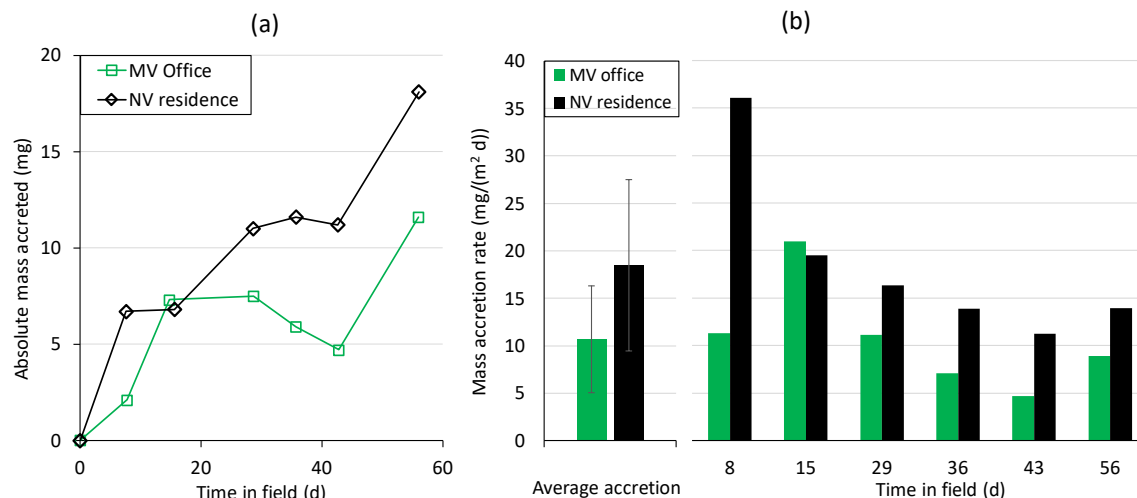
260 3. Results and Discussion

261 3.1 Rates of mass accretion to glass plates

262 Measurements of total mass accretion to glass plates as a function of time deployed are shown in Fig. 2.
263 The figure shows the absolute mass accreted and indicates a general trend of increasing mass with more
264 time spent in the field. Rates of mass accretion were normalized to the area of the upward facing top of
265 the plate. Note that plates were placed on wire racks to avoid direct contact with actual indoor surfaces
266 and enable testing of O_3 removal both upper and lower surfaces. This protocol resulted in exposure of

267 both the top and bottom of glass plates to air (see Figure S1). In the results presented here, only the top
268 surface and sides were exposed to the ozone deposition test chamber. Given that plates were deployed in a
269 horizontal orientation, and were not processed to remove settled particles, we expect the surface film to be
270 comprised of both settled particles and sorbed organic compounds. However, as will be discussed
271 subsequently, mass accretion rates were normalized to the top area of plates because measured
272 magnitudes are in the range that appears predominantly due to deposition of coarse-mode particles and
273 rates of dustfall [34,36].

274 Weschler and Nazaroff [33] summarize the range of magnitudes of mass accretion of particles and
275 SVOCs to surfaces in models and in previous empirical studies. Their work reports a range of mass
276 accretion rates due to adsorption of gas-phase organics in the range of 30-300 $\mu\text{g}/(\text{m}^2 \text{ d})$ [33]. This
277 reported rate is approximately 1-2 orders of magnitude lower than the accretion rates observed in this
278 study. Mass accretion rates observed here are in general agreement with the upper limit of the range
279 reported by Weschler and Nazaroff [33] to include deposition of coarse mode particles of 20-9000 $\mu\text{g}/(\text{m}^2$
280 $\text{d})$. For example, after 56 days in the field, glass plates in the MV office and NV residence accreted ~0.50-
281 0.78 g/m^2 of additional mass, or 9000-14,000 $\mu\text{g}/(\text{m}^2 \text{ d})$. The average mass accretion rate across all
282 collected plates is 10,600 $\mu\text{g}/(\text{m}^2 \text{ d})$ in the MV office vs. 18,500 $\mu\text{g}/(\text{m}^2 \text{ d})$ in the NV residence, and a one-
283 tailed t-test comparing higher accretion rates in the NV residences vs. MV office is at the threshold of
284 statistical significance ($p = 0.054$). The range of mass accretion shown in Figure 2 is also in reasonable
285 agreement with experimental results of dustfall rates in the literature. Edwards et al. [40] reported mean
286 values of 2200 and 3700 $\mu\text{g}/(\text{m}^2 \text{ d})$ in the summer and winter, respectively, in a study of four homes in
287 New Jersey, USA. In a study of 559 Canadian homes meeting a specified cleaning protocol, the reported
288 median dustfall rate was 10,000 $\mu\text{g}/(\text{m}^2 \text{ d})$ [41]. Thus, the exposure of the bottom face of the plate likely
289 introduced a small, acceptable error (<3%) to the mass accretion rates determined here.



290
 291 **Fig. 2.** Summary of mass accretion to test plates in two field environments: mechanically ventilated (MV)
 292 office and naturally ventilated (NV) residence. Panel a) shows absolute mass accretion while panel b)
 293 shows accretion rates. The average mass accretion rate ($\text{mg}/(\text{m}^2 \text{d})$) was calculated as the arithmetic mean
 294 of seven mass accretion rates measured over seven weeks. Error bars on average accretion rate are the
 295 standard deviation of the mean mass accretion rate. Uncertainty on each estimate of absolute mass
 296 accreted is ± 0.14 mg as described in the text.

297 While the mean mass accretion rate in the MV and NV locations are on the threshold of statistical
 298 significance in their difference, the magnitude of the difference in all but one sequential measurement of
 299 mass accreted to plates in the NV residence is consistently larger than that accreted to the MV office, and
 300 greater than the propagated uncertainty in the mass measurement from the analytical balance (i.e.,
 301 measurement > 0.19 mg). We speculate the consistent difference between the MV and NV plates is due to
 302 lack of particle filtration in the NV residence which led to higher indoor particle concentrations and
 303 greater deposition flux of particles to the sample surface. Previous studies in Singapore of MV vs. NV
 304 residences indicate NV environments have a higher proportion of outdoor particulate matter present
 305 indoors than MV environments [42,43].

306 3.2 Ozone reactivity of plates with accreted mass

307 3.2.1 Parameterizing transient O_3 removal to surfaces

308 The deposition of O_3 to indoor surfaces is typically parameterized according to equation 5, where the
 309 resistance uptake model ascribes resistances to removal due to transport from a well-mixed bulk core
 310 through a concentration boundary layer and subsequent reaction with the surface [27]. Across ozone

311 deposition experiments, chamber conditions were held constant with the exception of changes in the
312 accreted mass on the glass plate. Therefore, observed changes in v_d can be attributed to the accreted mass
313 present on the glass plate. An illustrative result showing the data collected during an experiment is
314 provided in Figure S4.

315 As discussed previously in Section 3.1, it appears that the majority of the accreted mass to the glass plates
316 deployed to field environments is derived from deposited particles. Thus, a recent study of ozone
317 reactions occurring on various indoor dust samples [32] provides a relevant basis for comparison for
318 ozone sink strengths. Deposition velocities reported in the first 90 min of each experiment, when
319 observable values are reported, are generally in the range of 0.002-0.025 cm/s across all plates with
320 accreted mass (see Figure S5). These values are roughly consistent with steady-state v_d converted from
321 O_3 -dust reaction rates in Vibenholt et al. [32] who report a range of values from 0.008-0.29 $cm\ s^{-1}$. We
322 speculate that higher dust loadings in those experiments (1 g vs. ~0.01 g here) explains the generally
323 higher deposition and observation of removal over >12 h time-scales not observed in this study. It is
324 worth noting that v_d reported in both studies are under reaction-limited conditions as the Vibenholt et al.
325 study was conducted in a FLEC cell operated at very high air exchange rate (AER) while for the
326 conditions of this study surface resistance contributes >99% of total resistance for all experiments.

327 Ozone removal was highest initially and decayed for the duration of the experiment until a steady-state
328 removal indistinguishable from that of background removal was reached. Reaction probabilities for each
329 plate are reported in Table 1; values are arithmetic averages across each hour of the experiment for which
330 γ could be calculated from removal statistically higher than that of the background chamber surfaces. In
331 general, with additional time deployed to the field, reported reaction probabilities increased and O_3
332 removal persisted for longer periods of testing in the laboratory chamber.

333

334 **Table 1.** Summary of regression parameters and time-averaged reaction probabilities across the first four
 335 hours of each experiment of ozone reactivity with deployed plates.

	MV office							NV residence						
	Regression%			Rxn. Probability [#] , $\gamma \times 10^{-6}$				Regression			Rxn. probability, $\gamma \times 10^{-6}$			
	β (min^{-1})	SE $\times 10^{-3}$	r^2	0-1 h	1-2 h	2-3 h	3-4 h	β (min^{-1})	SE $\times 10^{-3}$	r^2	0-1 h	1-2 h	2-3 h	3-4 h
0 d	0.045	5.2	0.54	0.74	0.06	-	-	0.045	5.2	0.54	0.74	0.06	-	-
8 d	0.026*	2.9	0.62	0.70	0.01	-	-	0.024*	2.1	0.29	1.0	0.34	0.23	-
15 d	0.020*	1.4	0.62	1.3	0.41	-	-	0.031*	1.9	0.33	1.6	0.43	0.01	-
29 d	0.013*	0.68	0.70	1.1	0.55	-	-	0.0093 *	0.46	0.51	1.6	0.76	0.55	0.57
36 d	0.019*	2.0	0.09	1.2	0.38	0.16	-	0.025*	0.61	0.47	1.8	0.82	0.60	0.46
42 d	0.019	1.7	0.35	1.2	0.24	0.06	-	0.013*	0.65	0.67	1.5	0.72	0.46	0.18
56 d	0.018	2.0	0.23	1.2	0.29	0.17	0.15	0.013	0.61	0.75	1.8	0.83	0.48	0.19
Field Blank	0.046	4.7	0.53	1.3	0.07	-	-	0.051	2.5	0.61	1.5	0.08	-	-

336 [%]Reported regression parameters are the resulting slope, standard error (SE) of the slope, and correlation coefficient
 337 from the $y = mx$ linear regression of natural logarithm transformed reaction probabilities.

338 [#]Reaction probabilities are the arithmetic average across the indicated period of the experiment. Dashes indicate the
 339 reaction probabilities were indistinguishable from background removal for that period.

340 ^{*}Indicated values of β are statistically significantly different than the estimate of the preceding plate. Note that
 341 slopes for all plates > 0 d are statistically significantly different from the 0 d plate.

342

343 In general, the parameterization of the time-variant reaction probability with equation 6 resulted in r^2

344 values of the regression that were typically > 0.5 (see Figure S6 for examples of the regression). Several

345 experiments had low r^2 values (< 0.4) but are included in subsequent analyses for completion and to

346 facilitate the goals of the regression analysis to enable statistical comparison across experiments. The

347 analysis of transient decay data also enables determination of the dynamics of ozone reactions to accreted

348 masses that would be obscured by solely comparing the cumulative O_3 removed as in Section 3.2.2. For

349 example, the data reported in Table 1 demonstrate that as larger masses accrete to surfaces, increasing

350 periods of time are required to increase cumulative O_3 removal.

351 Across plates deployed in both the MV office and the NV residence, values of β decreased as plates were

352 deployed for longer periods. This trend continued until approximately 42 days of deployment (Table 1).

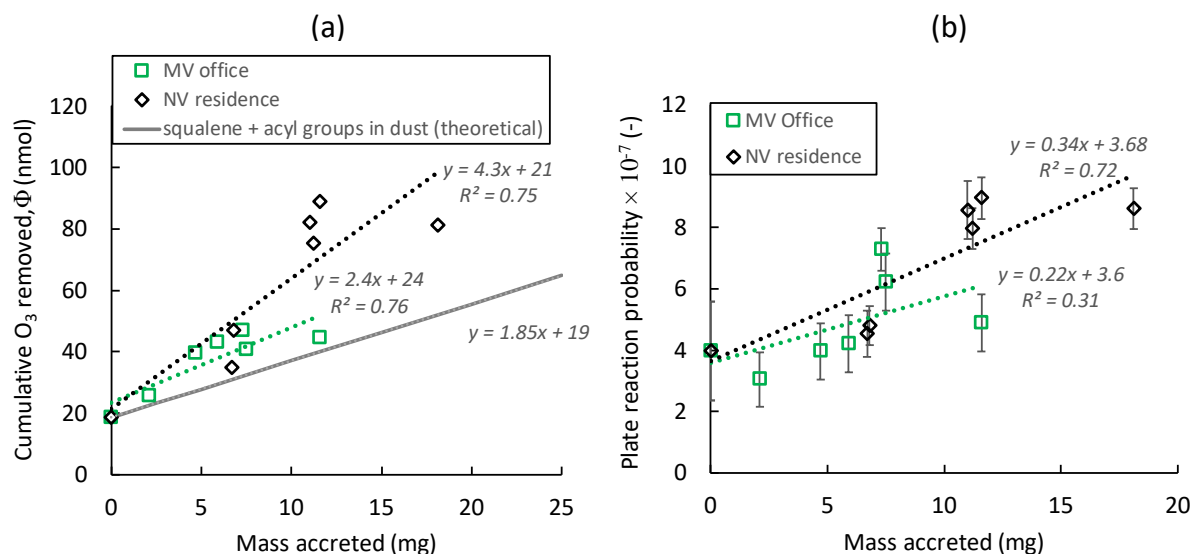
353 From 42-56 days, slopes of the regression appear to stabilize at approximately $\beta = 0.013$ - 0.019 . Note that

354 higher values of β indicate the magnitude of γ is "decaying" more rapidly than for lower values of β . This

355 observation is also observed in that measurable values of γ persisted into 3-4 hours of the experimental
356 testing only for glass plates deployed for 56 d (MV office) or 29 days or greater (NV residence).
357 Statistical significance of differences in β were determined by calculating z-scores from the difference in
358 slopes divided by the difference in the standard error of the slopes of the regression. The detailed
359 procedure is described in the Supporting Information. A summary of p -values for comparisons between
360 each plate and the preceding plate, as well as each plate compared to the 0 day plate, are provided in
361 Tables S1 and S2 of the Supporting Information. It appears that surface mass accretion results in
362 significant differences in O_3 reactivity, especially in the first month of deployment. Differences in β are
363 not statistically distinguishable when comparing longer deployment periods for plates deployed in MV
364 offices or NV residences, e.g., the plates deployed for 36 d and 42 d are statistically similar, as are plates
365 deployed for 42 d and 56 d, for the MV office. For the NV residence, the plate deployed for 42 d is
366 similar to the plate deployed for 56 d.

367 *3.2.2 Parameterizing cumulative ozone removal to surfaces with accreted mass*

368 It appears that for glass plates, accretion of mass to the surface of the plate results in a general trend of
369 increasing O_3 reactivity. The relationship between mass accretion and cumulative O_3 removed is explored
370 in Fig. 3, which presents cumulative O_3 removed as a function of mass accreted (panel a) and the average
371 reaction probability as a function of total mass accreted (panel b). The period of 15-200 minutes was
372 selected to calculate the cumulative O_3 removal and the average γ . Cumulative O_3 uptake to the accreted
373 mass is in the range of 20-90 nmol (Figure 3a), with a general increasing trend with greater accreted mass
374 to the plate for both MV and NV environments. Variance in the observed cumulative O_3 removed was
375 similarly and relatively well-explained by the mass accreted to the plate for both MV and NV
376 environments ($r^2 = 0.75-0.76$).



377

378 **Fig. 3.** Panel a) Cumulative O₃ a function of mass accreted to field deployed glass plates. Cumulative
 379 uptake is determined from the period 15-200 minutes following the injection of ozone into the chamber.
 380 The theoretical uptake from squalene and acyl groups assumes accretion of mass with reaction sites from
 381 human-derived constituents onto a surface with the same initial reactivity (that is, 0 mg of accreted mass)
 382 as the clean glass plates used in the NV and MV environments. Panel b) Reaction probability as a
 383 function of accreted mass.

384 The slopes shown in Figure 3a provide a proxy for the total reactivity of the accreted surface mass. The
 385 intercept is indicative of the reactivity of a clean plate, that is, the plate with no accreted mass from
 386 deployment to a field environment. The cumulative O₃ removal to a clean glass plate was 19 nmol. The
 387 results shown in Figure 3a indicate that 4.3 nmol O₃ was removed per mg of accreted mass for the NV
 388 residence vs. 2.4 nmol O₃ per mg of accreted mass for the MV office. The *p*-values for slopes of the
 389 regressions in Figure 3a were statistically significant for both the MV and NV environments (*p* = 0.011
 390 and 0.011, respectively), which together with relatively high *r*² values implies mass accreted is a
 391 meaningful predictor of observed changes in cumulative O₃ removal. The data reported in Figure 3a
 392 indicate that the mass accreted in the NV residence results in nearly twice the cumulative surface O₃
 393 removal compared to the MV office. However, a comparison of the standard error of the difference
 394 between the slopes shown in Figure 3a is calculated and is not statistically significant (*p* = 0.13).

395 Two pathways are typically considered as mechanisms of O₃ reaction with indoor surfaces. Ozone may
 396 react with the accreted mass via catalytic degradation [32] or via the Criegee mechanism at carbon double

397 bonds present in organic compounds in the accreted mass [5,32,44]. Previous studies, summarized by
398 Weschler [45], show that the presence of humans in indoor spaces results in transfer of skin flakes
399 (desquamation) and skin oils to indoor surfaces. These human-derived constituents contain compounds,
400 namely squalene [46] and acyl groups [47], that may act as O₃ surface reaction sites. The subsequent
401 analysis quantitatively evaluates the extent to which human-derived skin flakes and skin oils may explain
402 the observed removal to accreted mass on glass plates deployed to the MV and NV test environments.

403 Weschler et al. [48] estimate that dust contains approximately 60 µg of squalene per gram. This is
404 equivalent to 150 nmol of squalene present in each gram of accreted dust. There are six available double
405 bonds in a squalene molecule with which O₃ may react [49]. In addition to squalene, there are other co-
406 occurring compounds present in human skin oil that also contribute unsaturated sites where ozone
407 chemistry can occur. Pandrangi and Morrison [47] estimate that the molar fraction of unsaturations are
408 split between acyl groups (0.48) and squalene (0.44). Combining squalene and acyl groups, there are
409 available unsaturated reaction sites to remove 12.5 nmol of O₃ per nmol of squalene present in dust.
410 Normalizing by the previously noted mass of squalene in dust yields a ratio of ~1.85 nmol of O₃ removal
411 due to squalene and acyl groups for every mg of accreted dust.

412 The previous estimate of the reactivity of squalene and acyl groups in particles calculated from the
413 literature, 1.85 nmol O₃ removal/mg of accreted mass, can be compared with the reactivity of the accreted
414 mass calculated using equation 4. The theoretical O₃ removal due to squalene and acyl groups in dust is
415 shown in Figure 3a, with the same intercept as a cleaned glass plate. Comparing the slope of the
416 theoretical squalene and acyl groups line with slopes from plates deployed in the two field environments
417 shows that 42% (NV residence) and 75% (MV office) of O₃ may be attributed to these components on
418 human skin oil. Note again that this analysis assumes, as discussed in Section 3.1, that particles or dust
419 are the predominant contributor to the accreted mass observed here.

420 The calculation of 42-75% of the total removed O₃ as due to skin oil constituents implies other O₃
421 reactive constituents are also present in the accreting mass. A recent study of heterogeneous oxidation of

422 squalene with O₃ reported reactive uptake coefficients of $[4.0-4.3] \times 10^{-4}$ [50]. Values of plate reaction
423 probability reported here are much lower, with average values between $[2-9] \times 10^{-7}$ (Figure 3b). We
424 speculate this difference is a result of other compounds present in the accreting mass, as well as a
425 relatively low total accreted mass and potential for oxidation of highly reactive compounds like squalene
426 to occur while aging in the field. A study of the chemical composition of house dust reports total carbon
427 content on the order of 23-33% by mass [51,52]. Metals are shown to be a contributor to the indoor dust
428 composition, e.g., calcium levels of between 5-7% by mass are reported in two studies of the composition
429 of house dust [41,52]. While the form of calcium in dust is not reported, there is precedence in the
430 literature that ozone reactions with, for example, calcium carbonate, will not proceed via irreversible
431 deposition, but rather reversible adsorption [53]. Given that the composition of indoor dust is expected to
432 be highly variable [41], this discussion is speculative and intended to be illustrative of the potential for
433 sources of lower reactivity compounds in masses accreting to indoor surfaces. The need for further study
434 of oxidation on indoor accreted mass paired with chemical composition of the accreted mass is noted.

435 *3.2.3 Study limitations and future directions*

436 It is worth noting that plates were deployed in real field environments with low, but non-zero O₃
437 concentrations; the O₃ reactivity reported from testing in the laboratory does not account for oxidation
438 reactions that are likely to have occurred while the plates were deployed. Continuous interactions of
439 ozone with indoor surface films while deployed can reduce concentrations of surface unsaturated sites by
440 oxidation. Therefore, the estimates of the reactivity of the surface film reported here are likely
441 underestimating the true reactivity of the accreted mass. It is possible that the indoor concentration of
442 oxidants, rather than a difference in the amount or chemical makeup of the accreted mass, may be the
443 driver of the observed differences in reactivity between the NV residence (no filtration, frequently open
444 windows) and MV office (filters present in HVAC system, windows never open). We parameterized the
445 changing reaction probability using first-order kinetics to enable regression analysis, effectively treating
446 the changing reaction probability as a proxy for the changing density of surface reaction sites. It is

447 possible that these dynamics could be more mechanistically explained if surface concentrations of
448 unsaturated sites in the accreted mass were known.

449 We tested field blanks where cleaned glass plates were deployed inside sealed polypropylene bags for the
450 duration of their deployment to the field. However, we speculate that the mass observed on the field blank
451 plates was unacceptably influenced by water vapor condensing on the plate that penetrated the consumer
452 grade polypropylene storage bag during the deployment. There are limited points of comparison in the
453 literature to either inform or compare the quality assurance measures taken here. In a study of surface
454 films on windows, Liu et al.[34] show that field blank Kimwipes, which were waved in the indoor air of
455 the field environment but were not in contact with the target window, contained 24% of the average
456 sample value of n-alkanes and 1% of the sample concentrations of polar compounds. Wallace et al. [36],
457 who conducted a study using collection methods similar to those described here, investigate the potential
458 for bare hand contact to contaminate surfaces, but do not isolate surfaces to consider as a field blank.
459 Further effort to standardize field sampling methods that use inert surfaces as media for experiments of
460 aging and surface film accretion in indoor spaces is warranted.

461 The findings of this study compel further quantitative research into the study of the reactivity of accreted
462 masses on surfaces as a function of building operation, building type, and indoor air pollution levels.
463 Characterization of surface-bound compounds pre- and post-ozonation would be beneficial in elucidating
464 the specific contributors to O₃ reaction sites in mass accreted to indoor surfaces. There is precedence in
465 the literature for indoor and outdoor derived aerosols to differ in terms of chemical composition [54,55],
466 biological content [56], and water content [54]. Future studies should examine how these factors influence
467 the reactivity of the surface in addition to amount of mass accreted.

468 **4. Conclusions**

469 Accretion of mass to surfaces may alter reaction pathways by providing distinct reaction sites from those
470 of the original material. Previous studies of O₃ removal to materials have used aged building materials in
471 studies, most notably carpet, drywall, and ceiling tile samples [14,18,25]. This study employed an initially

472 inert, cleaned glass material to provide a baseline that allows isolation of the effect of the accreted mass
473 on surface O₃ reactions. The results show that the accretion of mass to the glass plates resulted in as
474 much as a factor of 4.5 increase in cumulative O₃ surface removal. Ozone reaction probabilities increased
475 with time spent in the field and mass accreted. Notable differences in the cumulative O₃ removed per
476 mass accreted to the surface were observed for plates deployed to MV vs. NV environments, implying
477 building operation may affect how materials interact with indoor O₃. In the range of ~40-70% of total O₃
478 removal may result from human skin and skin oil constituents present in the accreted mass. Given the
479 large extent of available surface area in indoor environments, further study of aging, mass accretion,
480 oxidation and oxidant cycling, byproduct formation and the integrated effect on indoor air quality and
481 human exposure is warranted.

482

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489

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