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Article type : Editorial

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### **How should we define an indoor surface?**

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**This is the author manuscript accepted for publication and has undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the [Version of Record](#). Please cite this article as [doi: 10.1111/INA.12955](https://doi.org/10.1111/INA.12955)**

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27 An ‘indoor surface’ is a commonly employed term in our community but one for which there is  
28 wide latitude given to its use. Consider the following expressions that all refer to an aspect of  
29 indoor surfaces: surface chemistry, surface-area-to-volume ratio, surface reservoir, surface film,  
30 and partitioning to surfaces. In each case, we understand that ***surfaces represent the elements of***  
31 ***the solid and liquid entities within the indoor environment that interact with indoor air and***  
32 ***aerosol particles*** but the exact nature of the surfaces and the interactions under consideration are  
33 often not defined clearly. Nevertheless, it is implicitly understood that surfaces can influence  
34 contaminant concentrations in indoor air.

35 In the chemistry community, the term surface is sometimes used to describe the (roughly)  
36 molecule-thick interface between the gas phase and a solid or liquid material to which chemicals  
37 can adsorb. For crystalline porous materials, such as zeolites and metal organic frameworks,  
38 internal interfaces represent the majority of the surface area that is responsible for adsorption and  
39 catalysis and is often quantified by a BET surface area. Similarly, the building science  
40 community has long recognized the dynamic interactions that occur between the bulk material  
41 below the interface and the air above it, i.e. these materials act as both sources and sinks for  
42 chemicals in indoor air.<sup>1</sup> Should we also refer to this bulk material, which can be centimeters  
43 thick, and the overlying boundary layer, as a component of the surface? And, what about the  
44 surface area term? It is frequently reported that the surface area-to-volume ratio of an indoor  
45 space has values of a few per meter.<sup>2</sup> But what surface area is being referred to, given that the  
46 surface area at the microscopic scale is much larger than this value and may contain internal void  
47 space which is difficult to quantify? Although these concepts and questions are not new, the  
48 goals of this editorial are to: i) highlight the importance of a common understanding of indoor  
49 surfaces, and ii) place into discussion recommended terms to describe this important component  
50 of the built environment.

51 To start, why does it matter? At the broadest level, loose definitions can lead to ambiguities that  
52 complicate our understanding of the fundamental processes at play. To illustrate, the degree of

53 gas-to-surface partitioning sets the abundance of many chemicals in the air and in surface  
54 materials, thus directly affecting estimates of pollutant inhalation and dermal exposure. Which  
55 partitioning compartments should we use to assess these exposures, and what is the temporal  
56 responsiveness of the chemicals in each surface compartment to environmental perturbations? It  
57 is also important to describe correctly the rates of indoor multiphase chemistry that can both  
58 remove and produce harmful pollutants. As an example, we now know that reactive chemistry  
59 occurs not only on surfaces such as carpets, walls and windows, but also on humans.<sup>3</sup> The  
60 reactive surfaces on humans – their skin, hair and clothing – are chemically and morphologically  
61 complex. A clear description of these complex surfaces is needed to accurately represent such  
62 processes.

63 Consider the interactions that occur between indoor air and indoor surfaces (see Figure 1). First,  
64 a gas-phase molecule that passes through the gas-side boundary layer encounters ***an interface, a***  
65 ***few tenths to a few nm thick, where there is an abrupt transition from a (low) gas-phase***  
66 ***density to a (high) condensed-phase density.*** Depending on the interaction, molecules can  
67 physi- and chemi-sorb to interfaces<sup>4</sup> and, given that the interfacial structure and composition may  
68 differ from the underlying bulk materials, adsorbed species may react in an interface-specific  
69 manner. Interfaces of accessible internal voids present in porous solids, such as wood, may also  
70 exist. As indicated in Figure 1, the movement of molecules can be both from the air to the  
71 surface and vice versa.

72 ***Impermeable surface materials are those that do not experience mass transfer into their***  
73 ***interiors over relevant timescales.*** Examples are silica present in glass windows, quartz in  
74 kitchen countertops, and stainless steel on our appliances. By contrast, ***permeable surface***  
75 ***materials undergo substantial diffusion of molecules beyond their interfaces into the bulk.***  
76 Mass transfer can occur either within the condensed phase, as for liquids such as skin oil or semi-  
77 solids such as resin or polymer coatings, or within the gaseous voids of porous solids, such as  
78 concrete, wood, textiles, carpets, or wall board, into which indoor air gases can diffuse. The  
79 depths to which molecules penetrate determine the extent to which surface materials interact  
80 with air.<sup>5</sup> The effective penetration depth can extend from nanometers to hundreds of microns or  
81 more, with its value controlled by time, and a combination of diffusive and reactive factors.

82 Semi-volatile molecules, such as most organics and water vapor, partition in significant  
83 quantities to indoor condensed-phase materials.<sup>6,7</sup> ***Surface films arise when these molecules***  
84 ***accumulate to thicknesses greater than one monolayer.*** At the microscopic level, these  
85 accumulations may be continuous or exist as islands, be morphologically rough, and contain  
86 deposited particles.<sup>8</sup> They are likely thicker on impermeable materials (e.g. semi-volatile organic  
87 films with thicknesses of nanometers to tens of nanometers develop over weeks or months)<sup>6,9</sup>  
88 than on permeable materials, because of diffusion into the bulk in the latter case. If exposed to  
89 the same levels of semi-volatile species in the gas phase, the film composition of semi-volatile  
90 species may be uniform from room to room when deposited onto an impermeable surface  
91 material over a period of a few weeks.<sup>9</sup>

92 ***The term surface reservoirs collectively refers to all the condensed-phase compartments that***  
93 ***interact with the gas phase and aerosol particles over relevant time scales.***<sup>1,10</sup> These include:  
94 interfaces, surface films, deposited particles, and all the accessible components of bulk materials  
95 that are present in our indoor environment (building materials, furnishings, humans, liquid water,  
96 etc.). The timescales for gas-surface interactions are important to consider. Molecules adsorbed  
97 to interfaces typically undergo very rapid exchange with the gas phase (seconds or less)<sup>4</sup> whereas  
98 the timescale for diffusion of molecules through paint layers to the underlying wallboard is on  
99 the order of hours or more<sup>11</sup> and PCBs can take years to partition into concrete and other  
100 building materials.<sup>12</sup>

101 We note that in this editorial we distinguish between deposited particles and suspended (i.e.  
102 aerosol) particles for two reasons that differentiate their impact on indoor air. One, the residence  
103 time of suspended particles is very much shorter than deposited particles, with removal times  
104 determined by air exchange and settling. Two, the partitioning capacity of aerosol particles is  
105 orders of magnitude smaller than that of immobile surfaces. Nevertheless, important reactive and  
106 partitioning processes occur with aerosol particles, occurring both at the air-particle interface and  
107 via diffusion below the interface.

108 Important quantities such as the indoor surface area-to-volume ratio and partitioning capacity  
109 have clear meanings using these definitions. Specifically, when evaluated over a macroscopic  
110 length scale, the geometric surface area-to-volume ratio is useful to quantify the rates of gas-  
111 surface interactions using a deposition velocity approach. However, to determine the degree to

112 which molecules partition to interfaces via adsorption, a much larger surface area determined at  
113 the molecular length scale is required. The total degree to which partitioning occurs reflects both  
114 the molecules that adsorb and those that absorb, by diffusing beyond the interface into the bulk  
115 of all the surface reservoirs present. The indoor partitioning capacity is so large that even highly  
116 volatile species, including many VOCs and HONO, are predominantly partitioned to indoor  
117 surface reservoirs, with a much smaller abundance in the gas phase.<sup>10</sup> We note that the effective  
118 partitioning capacity is a time-dependent value, increasing as longer times are considered.<sup>11,12</sup> As  
119 an everyday example, third-hand smoke presumably penetrates deeply into surface reservoirs,  
120 like upholstery or paint, in residences with long-term smokers.

121 These definitions are not necessarily new but, with the inherent complexity of indoor surfaces,  
122 the use of these terms remains inconsistent.<sup>13,14,15,16</sup> In particular, it is important to recognize the  
123 wide range of distance and time scales that characterize interactions of indoor air with the  
124 different surface reservoirs. It is hoped that this editorial encourages discussion on this topic as  
125 we continue to evaluate the important roles that surfaces play in the indoor environment.

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#### 127 CONFLICT OF INTEREST STATEMENT

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129 No conflicts of interest declared.

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#### 131 AUTHOR CONTRIBUTION STATEMENT

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133 This article was conceptualized by JA and GM. The original draft was written by JA. The final  
134 draft has been reviewed and edited by all authors.

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#### 136 DATA AVAILABILITY STATEMENT

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138 Not applicable

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140 **Figure 1.** Indoor surface reservoirs, including interfaces with the gas phase (solid blue lines),  
141 films, deposited particles, internal voids, and permeable bulk materials. Via mass transfer  
142 processes (red arrows), gaseous molecules can adsorb to interfaces (a), create surface films (b),  
143 partition to deposited particles (d), and move into surface materials by diffusion within pores (c)  
144 or within the condensed-phase material (e). Multiphase reactions can occur within any surface  
145 reservoir. The red arrows are two-headed, indicating mass transfer both to and from surface  
146 reservoirs. Mass transfer in the gas phase is depicted with a solid red arrow, whereas a dashed  
147 red arrow is used for (slower) diffusion in the condensed phase. Gas-phase mass transfer occurs  
148 via diffusion through a few mm-thick boundary layer adjacent to the interface. Internal voids  
149 may or may not interact with the gas phase depending on condensed-phase mass transfer rates.

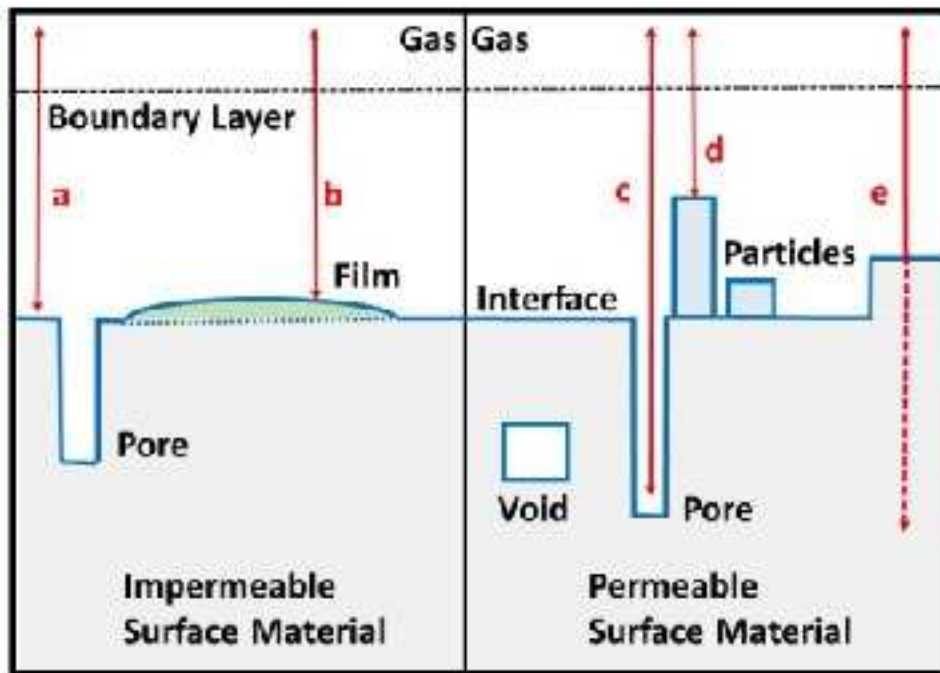
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