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Supporting Information for "Human skin oil: a major ozone reactant indoors"

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S1 Whole body emission rates of volatile organic compounds (VOCs)

In addition to the whole-body emission rates measured by Wang *et al.*¹ and discussed in Section 2.4 of the primary article, two other studies warrant consideration. Tang *et al.*² continuously measured time-resolved concentrations of VOCs in room air and supply air of a university classroom during occupied and unoccupied periods. Based on these measurements, they calculated human occupant emission factors (μ g h⁻¹ per person) for selected chemicals. (See Figure 3 of cited article.) They also reported an average whole body emission rate of 6250 μ g h⁻¹ per person for the classroom occupants. This value is larger than the values of 2180 ± 620 μ g h⁻¹ per person (no O₃) and 4600 ± 500 μ g h⁻¹ per person (with 37 ppb O₃) reported by Wang *et al.*¹ The larger value for the classroom measurements of Tang *et al.*² is expected, since much of the measured emissions originated from personal care products. By contrast, in the Wang *et al.* study, participants were instructed to avoid the use personal care products and wore clothing freshly laundered with scent-free detergent and then aired.³ Note that Tang *et al.* reported emission factors for 6-MHO (99 μ g h⁻¹ per person) and 4-OPA (37 μ g h⁻¹ per person), indicative of ozone/skin oil chemistry in the classroom.²

Recently, Zou and Yang⁴ reported whole body emission rates measured for young adults under controlled conditions in a specially designed single-occupant chamber. The measured values ranged from 164 to 518 μ g h⁻¹ per person. Starting three days before the measurements, the subjects were not permitted to use personal care products and were instructed to use only fragrance-free soap when washing. "On the day of the experiment (before testing), subjects were instructed to wash their hair and bathe with clean water, and wear clean attire. ... During the experiment, subjects were required to wear clean experimental clothes, including short-sleeved cotton T-shirts, cotton shorts, and thin cotton socks." During these measurements, the ozone concentration in the chamber was less than 5 ppb. The range of whole-body emission rates measured by Zou and Yang⁴ is much smaller than those measured by Wang *et al.*¹ in the absence of ozone. A potential explanation is the different analytical methods used in the two studies. Whereas Wang *et al.*¹ used a highly sensitive PTR-ToF-MS for their measurements, Zou and Yang⁴ collected their samples on Tenax-TA and DNPH sorbent tubes. The Tenax tubes were

subsequently thermally desorbed into a GC-MS and the DNPH tubes were eluted with solvent, which was subsequently injected into an HPLC. PTR-ToF-MS detects a broader range of compounds and is a more sensitive analytical method than sorbent sampling.

S2 Airborne transport of squalene to indoor surfaces

The vapor pressure of squalene at 25 °C, as determined using correlation gas chromatography, is $(3.7 \pm 1.3) \times 10^{-7}$ Pa,⁵ corresponding to a saturation vapor concentration of 62 ng m⁻³. At this level, and assuming a deposition velocity for squalene to indoor surfaces of 3 m h⁻¹, the corresponding upper bound estimate for squalene flux from the gas phase to indoor surfaces would be 0.2 μ g m⁻² h⁻¹ or 5 \times 10⁻⁴ μ mole m⁻² h⁻¹. It is informative to compare this upper limit to ozone's flux to indoor surfaces. A typical ozone concentration in indoor air is about 5 ppb = 0.20µmole m^{-3.6} With a nominal deposition velocity for ozone of 1 m h⁻¹, the ozone flux to indoor surfaces would be 0.20 µmole m⁻² h⁻¹, much larger than the upper bound estimate of squalene flux. Note that this comparison uses the squalene vapor pressure at 25 °C. Various indoor surfaces are occasionally heated, e.g., during cooking, heating, bathing, or via sunshine. The resulting increases in surface temperature and vapor pressure of surface-accumulated skin oil constituents might promote the redistribution of squalene to other indoor surfaces. It is also possible that skin oils nucleating from heated surfaces or from the body envelope become ultrafine particles (UFP). Such particles would deposit without preference on surfaces of any orientation. However, calculations using what we know about such processes suggest that the flux of skin-oil-rich UFP to indoor surfaces would be too small for the particle constituents to serve as a major sink for ozone through surface reactions (see S3).

S3 Flux of ultrafine particles that are rich in skin oils to indoor surfaces

Might ultrafine particles (UFP) rich in unsaturated skin oil oxidation products ^{7,8} soil indoor surfaces fast enough to constitute a major surface-reaction sink for indoor ozone? To answer this question, we can make an upper-bound estimate of the flux of double bonds in UFP to room surfaces. We begin by assuming that a reasonable upper bound for the time-averaged indoor particle number concentration of UFP is 20,000 per cm^{3,9,10} For these bounding calculations, we consider three simplified cases, in each case assuming that all particles have the same diameter: Case 1, 10 nm; Case 2, 30 nm, and Case 3, 100 nm. We further assume the UFP have unit density, so the mass of a single particle is as follows: Case 1, 5.2×10^{-19} g; Case 2, 1.4×10^{-17} g; and Case 3, 5.2×10^{-16} g. We estimate a deposition loss-rate coefficient (k) for UFP of these three diameters using the relationship presented for "fan-on condition" in Rim et al.¹¹: $k_{10 \text{ nm}} = 1.8 \text{ h}^{-1}$; $k_{30 \text{ nm}} = 0.70 \text{ h}^{-1}$; and $k_{100 \text{ nm}} = 0.26 \text{ h}^{-1}$. Consistent with this being an upper bound estimate, these loss-rate values are relatively high compared to model predictions.¹² We assume that the surface-to-volume ratio is 4 m²/m³, so that the average deposition velocities for the three cases are: $v_{d_{10} \text{ nm}} = 0.44 \text{ m h}^{-1}$; $v_{d_{30} \text{ nm}} = 0.17 \text{ m h}^{-1}$; $v_{d_{100} \text{ nm}} = 0.06 \text{ m h}^{-1}$. The resulting flux of UFP to indoor surfaces for the three cases are as follows: Case 1, $4.6 \times 10^{-3} \mu g$ $m^{-2} h^{-1}$; Case 2, 5.0 × 10⁻² µg $m^{-2} h^{-1}$; Case 3, 0.67 µg $m^{-2} h^{-1}$. We now make the extreme bounding assumption that the UFP consists entirely of skin oil constituents. Using 280 g/mol for the average molecular weight of constituents coupled with an average of 0.9 double bonds per molecule in skin oil,¹³ the upper bound flux of UFP double bonds to indoor surfaces would be as follows: Case 1, $1.5\times10^{\text{-5}}$ µmole m^2 h^1; Case 2, $1.6\times10^{\text{-4}}$ µmole m^2 h^1; Case 3, $2.1\times10^{\text{-3}}$ µmole m^2 h^1.

To put these upper bound estimates in context, compare the flux of double bonds via UFP to surfaces with the ozone flux to indoor surfaces. Assuming a typical indoor ozone concentration of 5 ppb and a nominal deposition velocity of ozone of 1 m h⁻¹, the ozone flux would be 0.2 μ mol m⁻² h⁻¹ to indoor surfaces. Comparing the upper bound estimates of the flux of double bonds delivered to surfaces by UFP to the flux of ozone to surfaces under typical indoor conditions, one sees that transport via UFP is much too small to constitute a major surface-reaction sink for indoor ozone. Ozone is lost on indoor surfaces at a rate that is orders of magnitude higher than can be accounted for by the deposition of skin-oil constituents in ultrafine particles.

S4 Contribution of skin oil to ozone removal in an occupied home

Liu et al.¹⁴ found that during the initial hours of vacancy, off-body skin oil was responsible for a 6-MHO production rate that was 80% of its production rate during occupancy. More specifically, the concentration of 6-MHO in the living space steadily declined from 350 ppt during occupancy to 90 ppt after a six-day unoccupied period. In principle, the slowly declining concentration of 6-MHO during the unoccupied period could reflect changing net emissions from two processes: i) 6-MHO generated by O_3 reactions with squalene and selected products (e.g., 4,8,13,17,21-tetramethyl-octadeca-4,8,12,16,20-pentaenal (TOP; five double bonds); 4,9,13,17-tetramethyl-octadeca-4,8,12,16-tetraenal (TOT; four double bonds); 5,9,13-trimethyltetradeca-4,8,12-trienal (TTT; three double bonds); and the corresponding C27-pentaenoic acid, C22-tetraenoic acid, C17-trienoic acid; as well as OH-geranyl acetone, and geranyl acetone) on inanimate indoor surfaces and ii) desorption of 6-MHO from indoor surface reservoirs. The small intercept (0.007 ppb/h) in a plot of the source strength of $C_8H_{15}O^+$ (an ion characteristic of 6-MHO) vs. indoor O₃ concentration (see Figure 3A in the primary article), indicates that the contribution from surface reservoirs is small, an inference also supported by the relatively low value of the octanol-air partition coefficient (K_{oa}) for 6-MHO. Hence, we infer that the 6-MHO emitted during the initial 130 h of vacancy reflects chiefly the O₃ oxidation of squalene and squalene-derived products on inanimate indoor surfaces, without replenishment. When there were one or two occupants, the source strength of $C_8H_{15}O^+$ averaged 0.056 ± 0.14 ppb/h. During the period when the home was unoccupied, the source strength of $C_8H_{15}O^+$ decreased from 0.045 ± 0.016 ppb/h during the initial unoccupied hours to 0.023 ± 0.003 ppb/h after 130 h of vacancy (see Figure 3C). To summarize, evidence suggests that during the initial hours of vacancy, off-body skin oil was responsible for a 6-MHO production rate that was 80% of its production rate during occupancy (0.045/0.056 = 0.80).

S5 Ozone reactions with skin oil on off-body surfaces in an occupied office

In a regularly occupied office, from 14 February to 18 March 2019, Wu *et al.*¹⁵ made continuous, real-time measurements that included concentrations of indoor and outdoor ozone and volatile organic compounds (using PTR-ToF-MS), as well as the fraction of recirculated air in the HVAC system. We noted in the primary article that, in analyzing their data, the investigators miscalculated the ozone loss rate to indoor office surfaces. Page 16483 states:

"We estimated the maximum production rates of SOOPs [skin oil oxidation products] via heterogeneous reactions using O_3 loss rates and the assumed maximum yields for indoor surfaces. For example, the O_3 loss rate to indoor surfaces during occupied periods was 6.7 µmol h⁻¹ on February 27."

Based on their reported data, this O₃ loss rate is incorrect. On February 27, the average indoor O₃ concentration was 10.9 ppb (Table S2 of Wu *et al.*¹⁵), corresponding to 21.8 µg m⁻³ or 0.45 µmoles m⁻³. The reported mean O₃ deposition velocity was 0.045 cm s⁻¹ = 1.6 m h⁻¹ (Table S1 in the cited article). Hence, the O₃ flux to indoor surfaces would have been 0.45 µmoles m⁻³ × 1.6 m h⁻¹ = 0.73 µmoles m⁻² h⁻¹. The office has a reported indoor surface area of 705 m² (p. 16481). Combining these values, the O₃ loss rate to indoor surfaces on February 27 would have been approximately 520 µmol h⁻¹ (0.73 µmoles m⁻² h⁻¹ × 705 m²), about two orders of magnitude larger than the value reported by Wu *et al.*¹⁵

A consequence of this correction is that a large fraction of what Wu *et al.*¹⁵ ascribed to desorption of 6-MHO, 4-OPA, and decanal from surfaces is more probably attributable to ozone reactions with surfaces. We can use the data from Table S2 in the cited article to further examine this point in the case of decanal, a primary product whose resulting indoor concentration is not influenced by secondary reactions with ozone. When the indoor surface emission rate of decanal is plotted against the indoor O₃ concentration as measured on days between 14 February and 15 March, the relationship is well-described as linear with a strong goodness of fit, $R^2 = 0.81$. Such a relationship indicates that the surface emission rate of decanal scales with the indoor ozone concentration, consistent with the hypothesis that ozone-initiated surface chemistry is the main driver as opposed desorption from a preexisting surface reservoir of decanal. It is also important to note that decanal from ozone/surface chemistry arises almost exclusively from reactions with skin oil constituents. Consequently, the experimental evidence reported in Wu *et al.*¹⁵ is strongly consistent with the hypothesis that skin oil on indoor surfaces is a large contributor to indoor ozone reactivity.

S6 Further discussion of the entries in Table 4 of the primary article

Table 4 of the primary article presents estimates of decanal yields (moles of measured products emitted per mole of ozone consumed) resulting from ozone reactions with on-body and offbody surfaces as reported in different studies. This section presents additional details regarding the cited studies.

Hair, skin surfaces, skin oil on glass wool. Pandrangi and Morrison¹³ used a tubular flow reactor to investigate the reaction of ozone with human hair samples. Samples from volunteers of four ethnicities were collected before and after washing and at different distances from the scalp. Samples from different volunteers varied in their reactivity. Unwashed hair had a higher ozone uptake and reaction probability than freshly washed hair. The reaction products were collected on Tenax-TA sorbent tubes and analyzed using a thermal desorber interfaced to a GC-MS. Yields were measured for various aldehydes. The mean yield of decanal and its standard deviation for unwashed hair samples from six volunteers was 0.072 ± 0.029 .

Wisthaler and Weschler¹⁶ passed air containing ozone through a flow reactor affixed to a volunteer's forearm or forehead. The initial measured decanal yield was 0.051. When silanized glass wool was "rubbed between the fingers and across the forehead and nose-bridge of human volunteers" and immediately placed in a Teflon tube through which air containing ozone passed, the initial yield of decanal was 0.060.

Morrison *et al.*¹⁷ have probed the person-to-person variability in the products derived from ozone-initiated chemistry with human skin. In their study, air at ~60% RH containing ~110 ppb O_3 flowed through an enclosure affixed to the arm of a volunteer into a measurement instrument. Eighteen females and three males, 20 to 61 years of age, participated as subjects. The net yield of gaseous products among the 21 volunteers ranged from 0.33 to 0.93. The evolution of product emission rates over time varied among the volunteers, suggesting variability among individuals in the abundance of skin surface lipids. Variability might also reflect different ratios of fresh to partially oxidized lipids on the skin of the volunteers at the time of measurement. The average decanal yield was 0.016 (interquartile range: 0.012-0.020).

In addition to differences associated with different parts of the body sampled, yields in these three studies may reflect different analytical methods including different instrumentation and different approaches to calibrating for decanal. Pandrangi and Morrison¹³ used sorbent sampling followed by GC-MS analysis to quantify decanal resulting from hair ozonolysis. Their calibration standard for decanal was a pure compound (Sigma-Aldrich). Wisthaler and Weschler¹⁶ measured decanal in real time using proton-transfer-reaction mass spectrometry. The "instrument was calibrated using dynamically diluted gas standards containing ~1 ppm (±5% accuracy) of saturated linear C2-C10 aldehydes." Morrison et al.¹⁷ measured decanal in real time using an advanced proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS) that was "calibrated every two hours with a 16-component VOC standard mixture containing around 1 ppm of each compound." From Figure S5 of that paper, it appears that this mixture included acetone and butanal, but did not include higher molecular weight aldehydes, which are more prone to fragmentation. The standard mixture specifically did not contain decanal. Fragmentation was reported to be higher for 6-MHO and geranyl acetone than found in other studies. In summary, decanal quantification involved the use of authentic standards in Pandrangi and Morrison¹³ and Wisthaler and Weschler,¹⁶ whereas instrument sensitivity to decanal was estimated in Morrison et al.¹⁷ It also appears that decanal fragmentation may have influenced the findings in the latter study.

Occupied chambers. In a chamber configured as a simulated aircraft with seats, carpeting, and fittings from an actual aircraft, and containing 16 "passengers," investigators measured decanal yields to be between 0.016 and 0.036, with a mean value of 0.025.¹⁸ This series of four experiments was conducted with two different groups of volunteers and at two air-change rates. In another chamber, configured as a simulated office with sparse furnishings and containing two occupants, Wisthaler and Weschler¹⁶ measured a decanal yield of 0.063. In chamber experiments with four occupants,¹ the net yield of decanal from reactions of ozone with occupants was found to be sensitive to relative humidity, consistent with the findings of Arata *et al.*,¹⁹ and relatively insensitive to temperature. In the case of decanal, yields measured

at 30% and 70% RH were 0.012 and 0.029, respectively. These measurements would have been influenced by ozone reactive compounds other than skin oil on the occupants' clothing.

Isolated room surfaces. Wang and Morrison²⁰ assessed the emission rates of various aldehydes when ozone reacted with room surfaces in four residences. The emission rates were measured using a "Teflon-coated field emission chamber" that was open on one side and could be sealed to a surface. Humidified air containing ozone, initially at 100-150 ppb, passed through the chamber. Samples collected on Tenax-TA sorbent tubes were analyzed for the target aldehydes by GC-FID. Measurements were made on the living room carpet or rug, kitchen floor, and kitchen countertop in three single-family homes and one apartment during summer months. Measurements were also made on a living room wall. The decanal yields listed in Table 4 of the main article were calculated as the decanal emission rate from a tested surface divided by the net ozone loss rate to that surface.

Wang and Morrison²¹ extended the results reported previously²⁰ using the same sampling and analytical procedures. Decanal was among the targeted aldehydes. Altogether, measurements of decanal yields from ozone reacting with different indoor surfaces were made in three single-family homes and two apartments during summer 2005, summer 2006 and winter 2007. "There were no significant temporal trends in reaction probabilities for any surfaces ... over the entire 1.5-year period". It is noteworthy that Table 2 of Wang and Morrison²¹ shows deposition velocities for different horizontal indoor surfaces that span narrower ranges in the four studied homes (H1: 0.9–11.7 m/h; H2: 1.6–12.8 m/h; H3: 1.9–2.9 m/h; H4: 0.6–4.5 m/h) than the ranges observed in chamber experiments conducted on common indoor materials.²² The limited range of deposition velocities compared with expectations for diverse surface materials (carpets, vinyl flooring, kitchen countertops) may reflect commonalities resulting from soiling by skin oils.

All room surfaces. At the beginning of the unoccupied period in the Oakland home, Liu *et al.*¹⁴ measured a production rate of ~0.065 ppb/h (see Figure 3F in the main article) for $C_{10}H_{21}O^+$, an ion characteristic of decanal. The ozone loss rate during this period was ~3.6 ppb/h (3 ppb $O_3 \times 1.2 h^{-1}$). The ratio of this production rate to the ozone loss rate suggests a decanal yield of at least 0.018, given the possible presence of other decanal fragment ions.

Wu *et al.*¹⁵ made continuous time-resolved measurements of 6-MHO, 4-OPA, and decanal at a university office building in Indiana. Table S2 of the cited article reports daily mean ozone concentrations and decanal surface emission rates for eleven days with different fractions of recirculated air. The decanal yield for the eleven days ranges from 0.0033 to 0.0114 with a mean \pm standard deviation of 0.0071 \pm 0.0023.

S7 Indoor decanal concentrations

Country (reference)	N	% quantified	AM (μg/m³)
Canada (Zhu <i>et al.,</i> 2013) ²³	3857	82%	1.45
Canada (Li <i>et al.,</i> 2019) ²⁴	3524	97%	2.17
Japan, winter (Azuma <i>et al.</i> , 2016) ²⁵	602	97%	1.4
Japan, summer (Azuma <i>et al.</i> , 2016) ²⁵	602	98%	3.7
Germany (Birmili <i>et al.,</i> 2022) ²⁶	639	58%	3.0

Table S1 Decanal concentrations measured in large-scale residential VOC surveys.

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