Supporting Information for: "Indoor Surface Chemistry Variability: Microspectroscopic Analysis of Deposited Particles in Dwellings Across the United States"

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I. SUPPLEMENTARY RESULTS

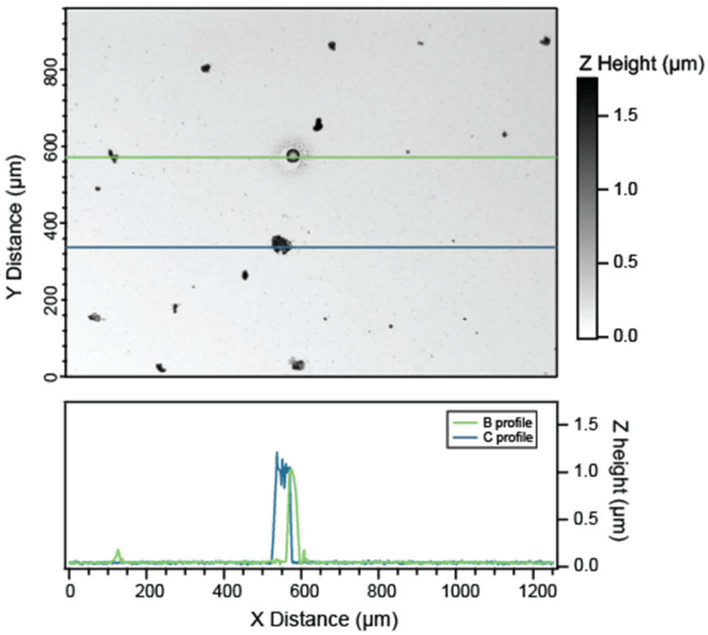

Figure S1. Line profile from a single 3D optical profilometry image of a three-week sample collected at East1. The height of the two largest single particles are over 1 micron in height, and correspond to particles B and C from Figure S2. Image is the same as the 3-D projection given in Figure 3b.

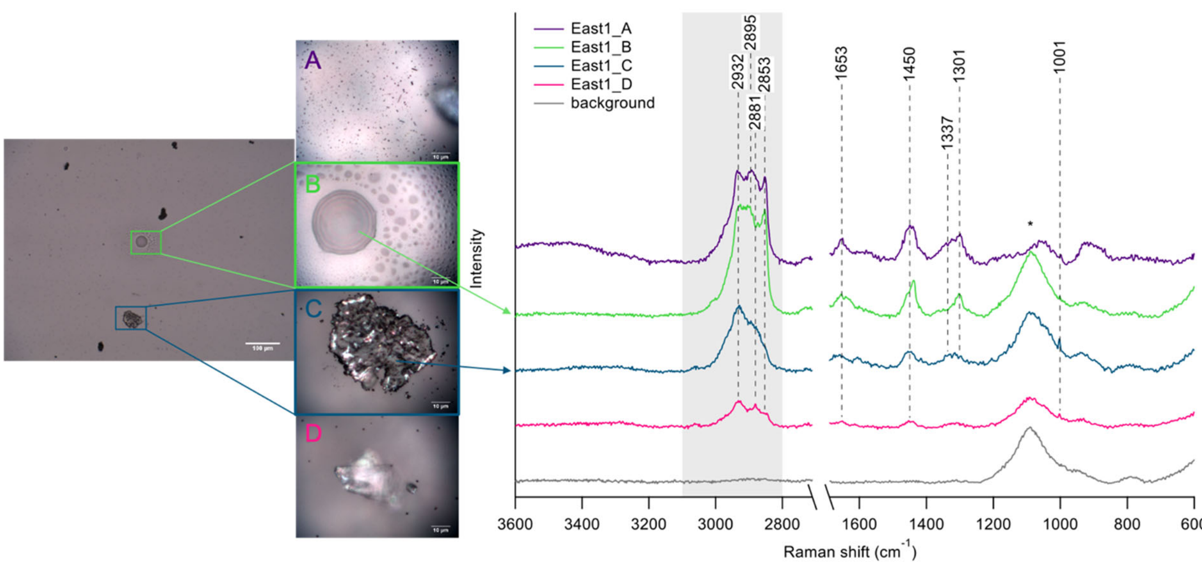

Figure S2. Multiple Raman spectra from East1 show the variability of particle appearance and composition from a single three-week sample. East1_B spectrum and image are the same as given in Figure 4B.

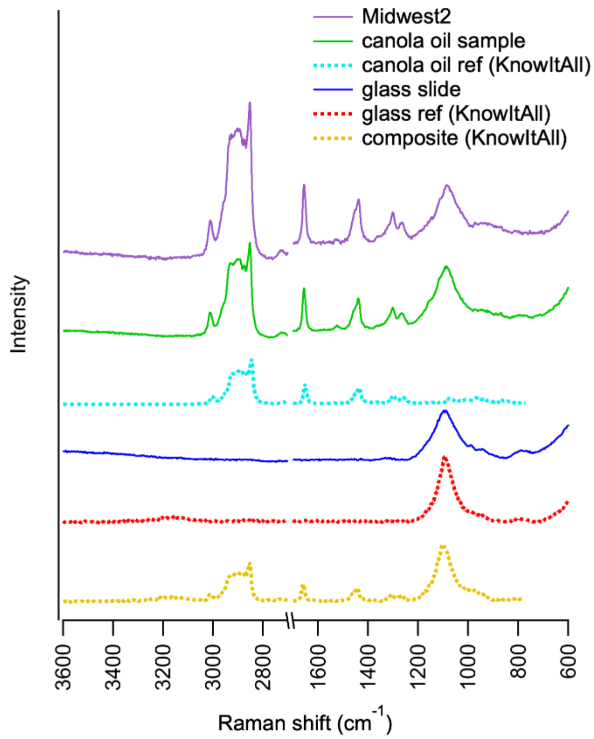

Figure S3. Sample vs reference Raman spectra. Solid lines are spectra captured by Horiba Raman and dashed lines are references or calculations from KnowItAll Spectral Library. Note that the Midwest2 spectrum is the same as shown in Figure 6 for easier comparison.

II. SUPPLEMENTARY CAMPAIGN INFORMATION

Each Stay-at-Home field sampling kit contained the following items:
100 glass microscope slides, 100 cover slips, 50 Falcon tubes, 1 box of 36 Glass jars with metal screw tops, 1 box of 100 Purple slide holders, 1 box Parafilm, 1 box medium binder clips, 1 box small binder clips, 1 bottle ACS grade Methanol, 2 eye dropper bottles, a pack lens cleaning paper sheets, 3 boxes Kimwipes, 1 pair of document gloves, 25 plastic forceps

This campaign did not require Institutional Review Board (IRB) approval, and personal identifying information was kept confidential by separately assigning each participant a unique researcher identifier number unknown to the individual handling the daily logs.

The following Field Manual was followed during the campaign.

Stay-at-HomeChem

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I EXECUTIVE SUMMARY

This text outlines a field study that takes the current stay-at-home orders as the basis for a natural experiment to explore changes in indoor chemistry, air quality and, in particular, the surfaces of the built environment when compared to the 'business as usual' condition.

II MOTIVATION

Americans spend over 90 percent of their time indoors, be it at home, school, or at work.[1] While much of the time we take clean indoor air for granted, research shows that depending on occupancy, indoor air can be more polluted than outdoor air, with links now emerging to human health impacts like chronic fatigue, asthma, and sick building syndrome. Yet, mechanistic insights connecting health outcomes with indoor chemistry remain enigmatic, making fundamental studies on this topic a frontier in field of environmental science and engineering.

Surfaces play an outsized role in the processes driving indoor chemistry, as indoor surface-to-volume ratios are considerably higher than outdoors.[2] Through processes collectively referred to as "sorption", surfaces have long been considered important for regulating physical and chemical transformations that gas-phase species undergo indoors.[3-5] Yet, molecular-level studies on the surface processes of indoor environments are now just beginning to emerge[6-22] and hold the promise of i) providing much needed microscopic and mechanistic insights into chemical reactivity within indoor engineered spaces, and ii) serving as proxies for indoor air chemical composition. These insights are important for disentangling the long-standing problems with indoor air quality on human health through dermal and inhalation exposure.[23-33]

The Covid-19 outbreak in the US has now led to stay-at-home orders in all but five U.S. states. As a result, Americans are currently spending the vast majority of their time in the same location, namely their homes, and, in many cases, with family members who would otherwise live elsewhere. Given that "at least 316 million people in at least 42 states, three counties, nine cities, the District of Columbia, and Puerto Rico are being urged to stay home",[34] there is an opportunity to determine how human emissions (ranging from skin oil constituents to emissions from baking and cooking and cleaning) are changing relative to the "business as usual" condition. How could such a change be quantified in an actual - as opposed to hypothetical - setting? To answer this question, we now hypothesize that the stay-at-home orders can be employed as a "natural experiment" to adress the following science questions:

1) What are the impacts of the stay-at-home orders on indoor air?
2) How do human activities under the stay-at-home orders influence the physical and chemical properties of the built environment?

The project takes place during the current stay-at-home orders in the SurfCIE network of several tens of homes that range from single-family to studio residences and employ low-tech passive samplers for collecting surface films (used as a proxy for air quality as well as for some of the intrinsic air chemistry and physics) over time, with appropriate storage for post-campaign chemical and physical analysis once the stay-at-home orders are lifted. The project represents the largest controlled indoor study of its kind and enable a test of how generalizable single-campaign home chemistry studies are for understanding the chemical reactivity of the built environment.

III. SAFETY

All aspects of the project meet university, state, and local requirements for social distancing with no additional person-to-person contacts, and samples are sent by mail. All methods have been developed.[12,35] SARS-CoV-2 titration studies report the longest lifetime of viable virus is <10 hours on plastic,[36] indicating a negligible health risk from analyzing Stay-at-HomeChem samples once our universities open back up sometime in the Summer or Fall.

IV. APPROACH

The project involves

1) Experimental design (writing of standard operating procedures, selection of materials for sample collection and storage that are widely and readily available);
2) Coordination of sample collection and storage across our network of participating homes while the stay-at-home orders are active;
3) Chemical and physical analysis once our universities reopen; and
4) Controls collected once the stay-at-home orders are lifted under "back to normal" as well as empty home (vacation and public holiday) scenarios.

The approach to be followed during the natural experiment is based on our previously reported method[12,35] for in-home collection of primary and secondary material on passive samplers. We purposefully keep the method at a level of sophistication commensurate with the current situation: the samplers consist of flats and windows appropriate for post-collection chemical and physical analysis (optical microscopy, vibrational and mass spectrometry, scanning probes, etc) placed in a given location (kitchen, living room, etc) in a given home. The samplers are placed upright into a holder and covered from the top by a tent formed from aluminum foil so as to minimize the collection of coarse mode particles, given we are interested in the secondary processes that lead to secondary organic films ranging from molecular to nano to microlayers. The sample collection times are varied from several hours to days to weeks to access different time scales relevant for indoor human occupancy and to meet sensitivity limitations of our broad range of analytical detection methods. Post-event sample analysis will range from high throughput methods (X-ray fluorescence for inorganic content, contact angle goniometry for hydrophobicity, DART-MS and APPI-ESI-MS, ATR-FTIR and Raman spectroscopy for organic functional group assessment, etc, see **Table I**) to more involved and time-consuming molecular probes (scanning probes, vibrational and mass spectromicroscopy, etc).

We employ the broad network of tens of residences to which we already have access through our existing indoor surface chemistry consortium (SurfCIE) across multiple universities and institutions in the US. The homes are coarsely classified into studios with kitchenettes, one- and two-bedroom apartments with separate kitchens, single family homes, etc.

Episodic events (a participant cleans on a given day; another airs out their residence; another prepares dinner; all while sampling at various daily to hourly frequencies) will be instrumental for testing the generalizability of controlled studies of indoor chemistry and inform on in-sample variability and reproducibility of outcomes in different homes via subsequent statistical analysis. The emphasis is less on scripted activities for the participants but instead on appropriately detailed documentation of spontaneous and regular activities by each participant through activity logging in the form of a field intensive diary.

Outcomes from the post-event chemical and physical analysis of the samples will be interpreted using diary entries of indoor behavior (cooking meals vs ordering take-out, working and living and sleeping in the same room vs in different rooms, working out, etc), as well as estimates of how many hours each participant spends in certain parts of their residence under stay-at-home orders relative to “business as usual”.

Complementary activities include the voluntary involvement of undergraduate students currently taking our courses, virtual remote lab rotations of trainees among consortium members, communicating project findings to the general public via peer-edited blog posts, and the proposed synchronized activities for sharing personal experiences and community building during the stay-at-home orders.

1. Table I – Measurement Techniques to be Used Once Labs Open Back Up

Method	# samples hr^{-1}*	Information	Substrate requirement	Can sample be reused after measurement?
DART-MS	High	Chemical composition	n.a.	No
APPI-ESI-MS	TBA	Chemical composition	n.a.	No
Optical microscopy	High	Shape, size, morphology	Reflective/ transparent	Yes
UV-Vis	High	Color, scattering as proxy for amount adsorbed	Reflective/ transparent	Yes
FTIR	High	Chemical bonds	IR transparent	Yes
ATR, PMIRAS		Chemical bonds	n.a.	In part
Raman	Low	Chemical bonds, pH	n.a.	Yes
Contact angle goniometry	Medium	Hydrophobicity	Flat	In part
XRF	High?	Elemental composition	n.a.	Yes
AFM-IR	Low	Chemical bonds, spatially resolved	Flat	Yes
SFG	Low	Chemical bonds	Reflective/ transparent	Yes

*high=>10, medium=~1, low=<<1 hr^{-1}

V METHODS

1. Pre-sampling Activities

Please generate a ***folder in your browser's bookmarks section*** to store your StayAtHomeChem URLs below for easy daily access.

Please note your **RID** in several places. Please contact Franz at f-geiger@northwestern.edu if you have misplaced it and he will send it to you again. You will need your RID for several activities, including sample naming.

Please familiarize yourself with https://www.wunderground.com.
This website is where you will read your meteorological and air quality conditions, plus we will be able to trace detailed met information as the website makes its historic record publicly available.

Please fill out the ***StayAtHomeChem Basic Kit Checklist*** located at
https://docs.google.com/spreadsheets/d/1kW2Zbm2zoThEA_XCnYeBXtTNjghhM1eyUYXOr_MIFAg/edit?usp=sharing

Please fill out the ***StayAtHomeChem Participant Info Form*** that has been emailed to you and submit it.

Please watch the following ***SOP movies***
0. Getting Started:
https://youtu.be/q5hZK0UYQhw
1. Opening shipped items
https://youtu.be/Dw1ZHlQ09Fw
2. Opening glass microscope slide box
https://youtu.be/3WGGSHL8Nbo
3. Cleaning the slides
https://youtu.be/Q4R1TayaF2w
4. Cover slip cleaning and fastening
https://youtu.be/Va_mDFIDpeM
5. Finalizing assembly and placement
https://youtu.be/9Nqg-6AdYj4
6. Partial disassembly and storage
https://youtu.be/5UnS09bywdM
7. More notes on placement
https://youtu.be/y3FPZhpc_pY

Please familiarize yourself with the daily ***activity diary*** example sheet at
https://docs.google.com/spreadsheets/d/1Wnr7YQJN7G5bIi3_1tojkamGVLcqGE3hSKt_pTWv4CQ/edit?usp=sharing

and the ***directions for filling*** it out located at
https://docs.google.com/document/d/188mWlhtf8_RsnSfQMv1InDJoscivOk7bvvxcu8lesYM/edit?usp=sharing for which you will also need to visit https://www.wunderground.com.

2. Sample Handling, Cleaning, and Placement Procedures

Please refer to the links to the YouTube movies above that were distributed by email.
If a sample is tripped, please stand it back up and note that it tripped in your daily log under "Notes".

3. Timing and Timeline: Please see ***pages 6-8 for example experiments***.

a) Timing for Experiments A, B, and C

Given most of us will be least distracted and have the calmest hands in the evenings after dinner, we are asking that you please **place your sample(s)** for a given experiment **in the evening**, once the daily activities are done, i.e. after dinner and doing dishes.

We are also asking that you please **store your samples** in the glass containers **in the evening** once the daily activities are done, i.e. after dinner and doing dishes.

Finally, we are asking that you ***log your activities*** once your samples are stored on the google sheet that was sent out and in hand-written form on paper.

Example: A participant runs Experiment C, page 6. Their dinner ends usually at 8PM. They prep their sample C2 and place it by 9PM. Time zero for sample C2 is then PM on that day.

b). Timing for Experiment D

Up to you!

4. Home Environment

Please refer to the ***StayAtHomeChem Participant Info Form*** that has been emailed to you.

5. Human Activities/Events/Activity Logging/Field Diary

Please refer to the links to the individual googlesheets that were distributed by email.

In addition, please keep a hand-written or electronic daily log of your home activities.

6. Choice of Rooms

Since we are interested in how increased home occupancy during the stay-at-home orders change indoor chemistry, **the rooms to sample should at least be the one of those that are most occupied during the day under the current stay-at-home orders**.

As an example, a home office inside a bedroom makes a lot of sense to sample unlike a bedroom that is used just like before the pandemic.

As another example, a kitchen and perhaps associated dining/living room area are likely to be occupied a lot more now than before, so it will be interesting to sample that area.

7. Sample Storage and Labeling

For consistency, we are asking all participants to store their samples in the glass containers. The Falcon tubes are back-up only in case your glass containers broke during shipment. Please label your samples clearly using a permanent marker on tape that you tape onto the exterior of the sample holder.

Please use the following code:

[Residence ID][experiment label][substrate][room][start datetime– end datetime]

abbreviations:

K = kitchen, B = bedroom, L = living room, D=dining room, T=bathroom, O=office.

MS=microscope slide, F=fabric, PP=polypropylene, etc.

Example: 0+B1+MS+K+1105209PM+1205209PM means that participant number zero ran a glass microscope slide as part of experiment B1 in their kitchen from 9PM on 11 May through 9PM on 12 May.

8. Questions?

Don’t be shy! There will be scenarios/situations that we have not foreseen, so please email your question to Franz at f-geiger@northwestern.edu.

9. Timeline for a given room in a given residence for three weeks
Monday, 11 May – Sunday 31 May 2020

NOTE: Each participant performs Experiment A and B (requiring 30 glass slides and glass jars), and then chooses between C and D, depending on how many glass slides and glass jars they have. You can always supplement your material by ordering from Amazon – you will be reimbursed for materials cost and shipping, but not taxes. Please check with Franz first.

Experiment A: 3-Week sampling per room, in triplicate
Number of samples and glass jars required is 3 per room.

Each participant places three samples next to one another into the room(s) of their choice and keeps them there for the entire duration of the campaign, three weeks.
The samples are named A1, A2, and A3.

Experiment B: daily 24-hour sampling to resolve buildup over each week
Number of samples and glass jars required is 27 per room.

On day 1 of each of the three weeks, each participant stores one field blank (named FB1A, FB1B, FB2A, FB2B, FB3A, and FB3B) and places seven samples next to one another into the room(s) of their choice. The samples are named B1-7 in week 1, B8-14 in week 2, and B15-21 in week 3. Each participant stores one sample on successive days as indicated below.

Example: sample B1 collects for the first 24 hours of week 1, while sample B7 collects for all seven days of week 1.

Note: underscore marks time sampler is in action, storage at end of underscore line

Week 1

Sample Number
B1____
B2__________
B3________________
B4______________________
B5____________________________
B6__________________________________
B7____
FB1A
FB1B

Day 1 2 3 4 5 6 7

Week 2

Sample Number
B8____
B9__________
B10________________
B11______________________
B12____________________________
B13__________________________________
B14____
FB2A
FB2B

Day 8 9 10 11 12 13 14

Week 3

Sample Number
B15____
B16__________
B17________________
B18______________________
B19____________________________
B20__________________________________
B21____
FB3A
FB3B

Day 15 16 17 18 19 20 21

Experiment C: Daily 24 hours sampling to resolve weekly variations

(Day 7 sample not needed as that's already in A7/14/21, field blanks not needed as those are collected by all as part of experiment B)

Number of samples and glass jars required is 18 per room.

Week 1

Sample Number		C1					
			C2				
				C3			
					C4		
						C5	
							C6
Day	**1**	**2**	**3**	**4**	**5**	**6**	**7**

Week 2

Sample Number		C7					
			C8				
				C9			
					C10		
						C11	
							C12
Day	**8**	**9**	**10**	**11**	**12**	**13**	**14**

Week 3

Sample Number		C13					
			C14				
				C15			
					C16		
						C17	
							C18
Day	**15**	**16**	**17**	**18**	**19**	**20**	**21**

Experiment D: 3-Hourly sampling to resolve daily variations on a given day X
Number of samples and glass jars required is 10 per room.
NOTE: This experiment requires a field blank at the beginning and the end of the 24-hour period.

Day X

Sample Number D1 D2 D3 D4 D5 D6 D7 D8

FB4

FB5

Time **00:00 03:00 06:00 09:00 12:00 15:00 18:00 21:00**

Total microscope glass slides used:
Per room, **Experiments A and B** require 3 A samples and 27 B samples, including field blanks, for a total of 30 samples and storage containers for the 3-week intensive.

Per room, **Experiment C** (daily 24-hour sampling) requires 18 samples + the three samples B7, B14, and B21 for all seven days of each of the three weeks for a given room and day. The analysis here only requires the 18 samples collected during the first six days of each week, given that the seventh daily samples are already analyzed as part of Experiment B.
Example: A proposed daily sampling study in three rooms requires 54 samples.

Per room, **Experiment D** (3-hourly sampling) requires 10 samples for a given room and day. **Example:** A proposed Wednesday vs Saturday study in three rooms requires 60 samples.

Bespoke Experiments
Additional experiments can be designed in which participants order materials other than microscope glass slides, like polycarbonate or PET plastic sheets, wood, dry wall, carboard (perhaps from the Amazon deliveries), and fabric (T-shirts). There are plenty of options at Amazon for this purpose.

What if I am running out of slides or glass storage containers?
You have all the resources to carry out experiments A and B. Yet, for Experiment C or D, you may only be able to sample one room, or follow dynamics only on one day. If you wish to add more sampling, please order the necessary parts (largely the glass slides and the glass jars), keeping in mind that the delivery lead times for the jars can be as long as ten days after placing the order. You will be reimbursed for the non-taxable cost (item price+shipping). Please check with Franz first at f-geiger@northwestern.edu.

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